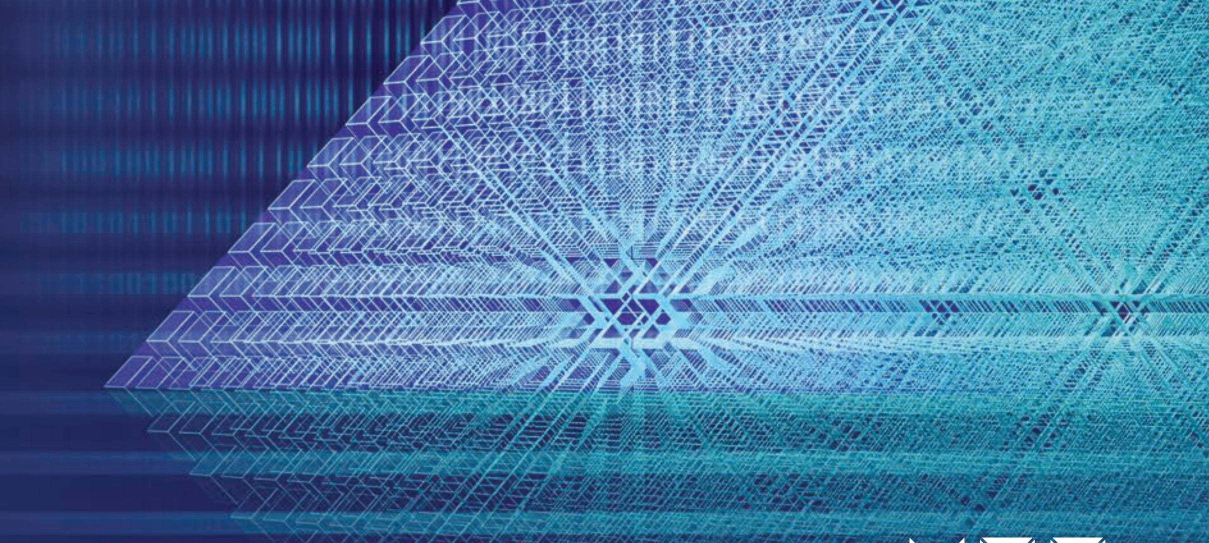

TECHNICAL REPORTS SERIES NO. 488

The Environmental Behaviour of Uranium

F.P. Carvalho, S. Fesenko, A.R. Harbottle,
T. Lavrova, N.G. Mitchell, T.E. Payne, A. Rigol,
M.C. Thorne, A. Ulanowski, M. Vidal,
O. Voitsekhovych, J.M. West, T. Yankovich

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THE ENVIRONMENTAL BEHAVIOUR OF URANIUM

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J.M. WEST, T. YANKOVICH

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Marketing and Sales Unit, Publishing Section
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PO Box 100
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fax: +43 1 26007 22529
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FOREWORD

The IAEA prioritizes the dissemination of information that can assist Member States with the development and implementation of activities intended to support various nuclear applications, including the legacy of past practices and accidents. To support radiological environmental impact assessments and the implementation of radiation safety standards for the protection of the environment, the IAEA has prepared a series of publications covering all aspects of the environmental behaviour and impacts of radionuclides of the uranium and thorium decay series, such as radon, radium, polonium and thorium.

Residues from activities involving radionuclides of the uranium and thorium series have recently received considerable interest. One of the main reasons is the significant contamination of vast areas around the world by natural radionuclides, mainly because of insufficient environmental protection during uranium mining activities between 1940 and 1960. Many such areas are classified as radiation legacy sites, where implementation of extensive monitoring and remediation programmes is recognized as an issue of very high priority. Another reason is that these radionuclides are mainly responsible for the environmental impact associated with the nuclear fuel cycle.

This publication outlines the behaviour of uranium in different environments, as well as its transfer to, and metabolism in, humans. It also provides concepts, models and data required for the radiological assessment of the impacts of uranium on non-human species. Assessing the environmental and health significance of uranium poses specific challenges because of the combination of different types of hazard and potential exposures. Therefore, both the radiotoxicity and chemical toxicity of uranium are considered.

The IAEA wishes to express its gratitude to M.C. Thorne for editorial support and to the other experts who contributed to the development and review of this publication. The IAEA officers responsible for this publication were S. Fesenko and A.R. Harbottle of the IAEA Environment Laboratories.

EDITORIAL NOTE

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CONTENTS

Chapter 1

INTRODUCTION

1.1. BACKGROUND

Uranium is a naturally occurring element that is present at low concentrations in all environmental media. Elevated concentrations of uranium can be found in some minerals, such as uraninite, in uranium rich ores. Over the past several decades, the assessment of the environmental impact of radionuclides of the uranium and thorium series has become increasingly important in many countries. Uranium production is growing and the demand for uranium may be expected to continue to rise for the next few decades [1.1]. The expansion of uranium mining activities often includes involvement of countries that have not previously hosted uranium mining and have limited expertise in the associated environmental protection requirements (see IAEA Safety Standards Series No. SSG-23, The Safety Case and Safety Assessment for the Disposal of Radioactive Waste [1.2]). There are also vast areas around the world contaminated with radioactive residues from former mining activities, giving rise to radiation exposure of humans and the environment [1.3]. Such cases include abandoned mining areas in Central Asian countries [1.4], as well as similar highly contaminated areas in many other countries, which require both assessment and remediation [1.5].

All uranium isotopes are radioactive. The three natural uranium isotopes generally found in the environment, ^{234}U, ^{235}U and ^{238}U, undergo radioactive decay by emission of an alpha particle accompanied by weak gamma radiation. The dominant isotope, ^{238}U, forms a long series of decay products that includes the key radionuclides ^{226}Ra and ^{222}Rn. The decay process continues until a stable, non-radioactive decay product is formed.

The IAEA attaches high importance to assisting its Member States with assessments of the environmental consequences of the nuclear fuel cycle and nuclear applications, including the legacy of past practices and accidents. In keeping with this, the IAEA has initiated the preparation of publications in the Technical Reports Series covering the environmental behaviour and impacts of radionuclides of the uranium and thorium series, including The Environmental Behaviour of Radium: Revised Edition [1.6] and The Environmental Behaviour of Polonium [1.7].

The IAEA has published a number of technical documents on radionuclide transfers in terrestrial, freshwater and marine environments [1.8–1.10]. These publications provide information on key transfer processes, concepts and models important for radiological assessments for all radionuclides, including radioisotopes of uranium, radium, thorium and polonium.

In a series of IAEA projects aimed at improving environmental assessment and remediation, the environmental behaviour of uranium was considered mainly in the context of contaminated site characterization [1.11–1.13] and environmental remediation [1.14], including the remediation of dispersed contamination [1.15] and uranium mill tailings [1.16], the decontamination of buildings and roads, the characterization of decommissioned sites, radiation protection and management of radioactive waste in the oil and gas industry [1.17], and radiological protection issues in the phosphate industry [1.18].

Assessments of the impact of uranium on humans and other biota are a specific challenge because of the combination of different types of hazard and potential exposures [1.19]. Furthermore, for the range of occupational and public exposure situations that can occur, either radiotoxicity or chemical toxicity may be the limiting factor, depending on the circumstances of the exposure situation.

In assessment studies, it is important to recognize that ^{238}U has progeny, including radionuclides such as ^{226}Ra, ^{222}Rn, ^{210}Pb and ^{210}Po, that represent important sources of exposure of humans and other types of biota. For both natural uranium and depleted uranium (which contains smaller proportions of ^{234}U and ^{235}U than are present in natural uranium), chemical toxicity is often of greater significance than radiotoxicity. However, if progeny of ^{238}U and ^{235}U are present to a significant degree in natural uranium, then the radiotoxicity per unit mass of uranium will be substantially increased, whereas the chemical toxicity will be essentially unaltered. The radiotoxicity of uranium relative to its chemical toxicity will also be substantially increased if it contains enhanced concentrations of ^{235}U and ^{234}U relative to natural uranium (as in enriched uranium). Enriched uranium is produced for use as fuel in nuclear reactors and for military applications. Depleted uranium arises from the enrichment process as a by-product that may be used in various applications or treated as a waste product requiring storage and disposal.

In this publication, issues relating to both the radiotoxicity and chemical toxicity of uranium are discussed. Specifically, the effects of the degree of enrichment or depletion, the presence or absence of radioactive progeny (decay products) and their chemical and physical form are all considered.

As this publication is primarily directed to the environmental behaviour and impacts of uranium, the main emphasis is on exposures of members of the public (with exposures of non-human biota also addressed). Exposures of members of the public are typically mainly by ingestion of contaminated water and food products, with inhalation (e.g. through intake of resuspended particles) a secondary consideration. In contrast, for occupational exposures, which are mentioned only briefly in this publication, inhalation is typically the dominant exposure route, with ingestion of only secondary significance. Other routes of exposure (e.g. uptake through wounds or across intact skin) will generally be of negligible importance in comparison to ingestion and inhalation.

1.2. OBJECTIVE

This publication is intended to provide information on the environmental behaviour of uranium for use in environmental impact assessments of routine discharges and accidental releases, for uranium impact assessments in different contamination scenarios, and for remediation planning of sites contaminated with uranium. Some of this information may also be useful in the context of the interpretation of uranium occurrence and isotopic distributions in environmental applications. Guidance provided here, describing good practices, represents expert opinion but does not constitute recommendations made on the basis of a consensus of Member States.

1.3. SCOPE

This publication covers the behaviour of uranium in the atmosphere, as well as in terrestrial, freshwater and marine environments. The primary focus of the publication is the environmental behaviour of uranium; the environmental behaviour of uranium progeny, such as radioisotopes of radium, radon, polonium and thorium, is considered in other publications. The information presented here is relevant to the environmental transfer of uranium to both humans and non-human biota. The publication is intended to provide an overview of the behaviour of uranium in natural environments. This will facilitate the use and updating of the following IAEA Safety Standards series publications related to the assessment of the radiological impact of radioactive discharges:

- The Safety Case and Safety Assessment for the Disposal of Radioactive Waste (IAEA Safety Standards Series No. SSG-23) [1.2];
- Safety Assessment for Facilities and Activities (IAEA Safety Standards Series No. GSR Part 4 (Rev. 1)) [1.20];
- Remediation Process for Areas Affected by Past Activities and Accidents (IAEA Safety Standards Series No. WS-G-3.1) [1.21];
- Regulatory Control of Radioactive Discharges to the Environment (IAEA Safety Standards Series No. GSG-9) [1.22];
- Management of Radioactive Waste from the Mining and Milling of Ores (IAEA Safety Standards Series No. WS-G-1.2) [1.23].

Other related information has been published in the Safety Reports series (see Refs [1.24, 1.25]). Guidance provided here, describing good practices, represents expert opinion but does not constitute recommendations made on the basis of a consensus of Member States.

1.4. STRUCTURE

Chapter 2 introduces the history of the discovery and application of uranium and of radioactive materials in general, and different applications of uranium in the present day. The physical and chemical properties of uranium are presented in Chapter 3. Chapter 4 addresses the presence of uranium in the environment, including data on representative uranium concentrations in parent materials, soils, water bodies, terrestrial plants and animals, and marine ecosystems. Chapters 5–7 provide key information on the radiological and toxicological significance of environmental uranium, including data for parameterization of environmental transfer processes and for the assessment of radiological and chemical impacts. Chapter 8 addresses the environmental impacts of uranium that can arise in various industries, the mitigation of those impacts and the remediation of the sites at and around which such impacts can occur. Chapter 9 summarizes different case studies related to the environmental behaviour of uranium, shedding further light on environmental impacts and aspects of mitigation and remediation.

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Chapter 2

HISTORY AND USES OF URANIUM

N.G. MITCHELL
Eden Nuclear and Environment Ltd, United Kingdom

Radioactive substances are part of our natural environment. Owing to their very long half-lives, natural radionuclides, and their shorter lived decay products (progeny), are found throughout the Earth's crust. These naturally occurring radioactive elements (so-called 'primordial radionuclides') include ^{40}K and the uranium and thorium decay series. Uranium, a metal in the actinide series, with the chemical symbol U and atomic number 92, is radioactive and all of its isotopes are unstable. Uranium occurs naturally in very small amounts in rocks, soils, water bodies, plants and animals. Natural uranium comprises ^{238}U (99.274 5% by mass), ^{235}U (0.72%) and ^{234}U (0.005 5%). The history and use of uranium reflect advances in mining, chemistry and physics, as well as in our understanding of radioactive substances and their potential applications.

2.1. EARLY HARD ROCK MINING

Metal ores have been extracted from the Krušné Hory Mountains (Erzgebirge or Ore Mountains) on the border of Germany and the Czech Republic since the Bronze Age. Silver exploitation in the sixteenth and seventeenth centuries gave rise to the development of two, now famous, mining areas: — Jáchymov, Czech Republic (then known as St. Joachimsthal, Bohemia) and Johanngeorgenstadt, Saxony — that both contained a black mineral that became known as 'pitchblende', after the German 'Pechblende' for 'pitch' (or 'bad luck') and 'mineral'.

Silver ores occur in vein type deposits. These tend to have a well defined zone of mineralization that is usually sloping and narrow compared with the length and depth of the deposit. These deposits occur within faults, fissures or shear zones. It is, therefore, common to find localized surface occurrences leading underground and for the vein to be deposited along with gangue minerals, mainly quartz and/or calcite, in a vein system. A vein system comprises a group of discrete veins that have similar characteristics and are usually related to the same rock structure. The surface occurrences were mined first and deeper mining progressed with technological advances.

The history of Jáchymov is closely linked to the success of local mining activities, and the town has seen several periods of mining related booms followed by economic decline [2.1]. Silver mining at Jáchymov started around 1516, when the first rich vein was found near the centre of the town. Jáchymov then quickly became the second largest town in Bohemia and, by 1534, had a population of about 18 000. The depletion of readily exploited silver saw a rapid decline in population to 2 177 by 1601. A long period of cobalt ore mining for use in enamels started around 1611 and later records show nickel extraction. The first state mining school was established at Jáchymov in 1716. Although there were periods of revival when mining practices changed or improved (e.g. a resurgence of silver extraction between 1755 and 1810), mining activities declined until the discovery of uranium. Johanngeorgenstadt was founded in 1654, and by 1680 there were around a hundred mines in the town and surrounding area. The presence of pitchblende in silver lodes had been recorded since 1750 [2.1].

2.2. DISCOVERY OF THE ELEMENT

The discovery of uranium in pitchblende from the mine of Georg Wagsfort (opened in 1670) at Johanngeorgenstadt was made in 1789 by Martin Klaproth, who named the element ‘uranite’ after the newly discovered planet Uranus. Pitchblende from Johanngeorgenstadt (Fig. 2.1) was described as greyish black, exhibiting various degradations, from the glittering to the dull or dim, and was found between strata of schistose mica [2.2]. Klaproth’s chemical analysis of

FIG. 2.1. Uraninite (pitchblende) with torbernite from the collection of Martin Klaproth (reproduced with permission courtesy of Museum für Naturkunde, Berlin, Inv. No. MFN_MIN_1985_006, photo: © Hwa Ja Götz, MfN).

pitchblende, a mineral now named ‘uraninite’, had isolated the oxide of the metal. An extract from his original paper to the Royal Academy of Sciences at Berlin in 1789 is provided in Ref. [2.3] and describes some of the tests undertaken to identify the new metal and the synthesis of the black material with metallic properties. Klaproth published two volumes of analytical essays [2.4] in which his work on uranium and other elements is described. His work included an investigation into the colour that uraninite would give to glass and enamels.

Further work on the oxides of uranium was undertaken by Johan August Arfwedson in 1823, who reduced the green oxide of uranium (believed then to be the lowest oxide) with hydrogen to produce a brown powder which he took to be the metal, but which is now known to be uranous oxide, or uranium dioxide (UO_2) [2.2]. The metal was first prepared by Eugène Peligot in 1841 by heating uranium tetrachloride and potassium.

2.3. USE OF URANIUM AS A COLOURANT

Early uranium production was largely a by-product from mines in Saxony, Bohemia and Cornwall in the first part of the nineteenth century [2.5]. The main use of uranium prior to the discovery of radioactivity was to colour ceramics and glass. It is not clear when uranium was first used to deliberately colour glass or enamels, and in Ref. [2.6] this lack of clarity is attributed to the secretive nature of early glass manufacturers. A historical survey of Cornwall, published in 1817, mentions combining local uraninite with glass in varying proportions to produce different colours [2.7], and the winner of the 1831 Prague International Industrial Glass Exhibition was an exhibit of uranium glass from the Harrachov glassworks, Czech Republic. One of the earliest surviving artefacts of uranium glass is a cut and engraved finger bowl manufactured in 1837 for Queen Victoria of the United Kingdom (Fig. 2.2)[1]. Examples of uranium glass produced by Joseph Riedel dating from about 1840 are more common.

Scavenging mine waste dumps for pitchblende started in the early part of the nineteenth century and continued through to the 1860s, and the development of commercial interests in uranium colouring resulted in the first mining for pitchblende as early as the 1830s [2.1]. Sodium and ammonium diuranates provide a yellow glaze and, by varying the concentration, produce cream, orange, brown, green or black glazes [2.8, 2.9]. In glass, typical uranium concentrations in the range of 0.1–2% by mass are used to produce a fluorescent yellow or light green glow under ultraviolet light. Glass containing uranium continued to be produced until the middle of the twentieth century. Various names have been

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[1] Exhibit No. C.110-1992; see https://www.vam.ac.uk/articles/a-z-of-glass

FIG. 2.2. Uranium coloured glass finger bowl (Davenport & Co., 1837, England; reproduced with permission courtesy of the Victoria and Albert Museum, London).

given to uranium glass-ware; for example, German names include ‘Annagelb’ for yellow glass and ‘Annagrün’ for green glass, while in the United Kingdom and the United States of America (USA) it is usually called ‘vaseline glass’.

The first uranium colour production factory was opened at Jáchymov in 1855 by Adolph Patera. The factory was in a former metallurgical plant for silver extraction and was called The Imperial and Royal Factory for production of uranium yellow colour (k.k. Urangelbfabrik). It produced eight types of yellow colour, adding orange colour in 1858.

2.4. DISCOVERY OF RADIOACTIVITY

Ionizing radiation was discovered in 1895 by Wilhelm Röntgen, who was examining the external effects from cathode ray tubes. He identified a continuous penetrating type of ray that was emitted from a Crookes tube. He named these rays ‘X rays’, although they also became known as ‘Röntgen’ rays [2.10]. Building on this work in early 1896, while studying the phosphorescence of uranium compounds, Henri Becquerel found that uranium salts caused a photographic plate to darken without being exposed to sunlight. He later concluded that these penetrating rays came from the uranium, whose nucleus was excited without the use of an external source of energy, and called them ‘uranium rays’.

An electrometer invented by Pierre Curie and his brother Jacques Curie allowed measurements of air ionization and was used by Marie Curie in her research on uranium rays that started in 1897 [2.11]. In the first of her three papers published in 1898, she considered uranium and thorium, and examined the differences in ionization between various sources of uranium (e.g. various oxides, pitchblende from three mining areas, including Jáchymov and Johanngeorgenstadt, other minerals). She concluded [2.10] that the strength of the rays was proportional to the amount of material present and noted that the uranium minerals also contained an element more active than uranium. The term ‘radio-active’ (in French) was first used in her second paper (July 1898), published with her husband Pierre Curie [2.12], to describe a new substance contained in pitchblende that they named ‘polonium’. The last paper, published in December 1898, identified and named a further strongly radioactive element as ‘radium’ [2.13]. Jáchymov was the source of the pitchblende that was used. This was initially from mine waste and later from the uranium colour production factory. A facility for radium production was subsequently added to the Jáchymov factory in 1904.

The early work of Ernest Rutherford is summarized in Ref. [2.14]. Their early studies looked at the radioactive decay of a gas, then unknown and termed ‘ThX’ (^{220}Rn), and they concluded that radioactivity is a spontaneous event emitting an alpha or beta particle from the atom, creating a different element. Rutherford went on to develop a better understanding of atoms and, in 1911, he interpreted the Geiger–Marsden experiment (1909) on alpha particle scattering by gold foil as indicating the presence of an atomic nucleus [2.15]. The stability of the atomic nucleus was subsequently argued for by Neils Bohr [2.16] in his studies that led to the development of early quantum theory (1913–1915). Then, in 1919, by firing alpha particles from a radium source into nitrogen, Rutherford demonstrated the composite nature of the nucleus by showing that nuclear rearrangement was occurring, with the formation of oxygen [2.17].

2.5. EXPLOITATION OF URANIUM BODIES FOR RADIUM

The early history of uranium mining is closely linked to the exploitation of other metals and it was common for mine tailings to be reworked, as illustrated by the early activities at Jáchymov. In the late nineteenth century, commercial mining of uranium ores was limited to a few locations [2.18]. The other two sites commonly referenced for their commercial production of uranium ores are the South Terras mine in Cornwall and several mines near Central City, USA. It is difficult to obtain estimates of the quantities produced at these and other smaller sites.

The mines at Jáchymov produced 620 t of high grade uranium ore between 1850 and 1898, and produced about 10 000 t of uranium before they were closed in 1968 [2.5]. The South Terras mine started uranium production in 1873 and exploited a rich vein of pitchblende found in 1889. It is estimated that about 275 t of uranium were produced before 1900 [2.5]. The South Terras mine closed in 1930. The first reports of uranium in the USA were from the gold mines at Central City and date from 1872; production is estimated at several hundred tonnes over the period 1872–1916 [2.19].

The first two decades of the twentieth century saw commercial uranium mining development spurred on by the high price of radium. Mines producing uranium ores were established near Osh, Tyuya-Muyun pass, Kyrgyzstan in 1908, at Stora Stolan, Billingen, Sweden in 1909, at Radium Hill in 1906 and later at Mount Painter in 1910, both in Australia. Uranium was exported from the south-eastern area of Utah, USA on the Colorado Plateau from 1902 to 1904 to develop a source of radium, followed by small scale production, beginning in 1911, from several deposits in the Moab and Monticello districts in Utah [2.20]. Uranium (in euxenite) was discovered in Madagascar in 1907, and further prospecting in 1908–1909 identified uranium deposits near Betafo, leading to shipments to France from 1911 [2.21]. The first uranium mining concessions in central Portugal were assigned in 1907 and led to development of the Urgeiriça mining area, with exports to France from about 1911, and then from the Sabugal region in 1913. The USA dominated the world radium market from 1912 to 1922 until high grade ore from the then Belgian Congo entered the market. The Shinkolobwe deposit in Katanga, Belgian Congo (now Shaba Province, Democratic Republic of the Congo) was discovered in 1915. It was exploited by the Union Minière du Haut Katanga from 1921 and was used to produce radium after transport of ore to Olen, Belgium [2.22].

Statistics on worldwide mineral production are available from the Imperial Mineral Resources Bureau, United Kingdom, which published annual estimates of uranium production from 1913 [2.23]. In that year, the total production of uranium ore was 3343 long tons (1 long ton = 1.016 metric tonnes), with most uranium ore produced in the USA (2026 long tons) followed by Portugal (1206 long tons), the United Kingdom (95 long tons), Austria–Hungary (11 long tons) and Madagascar (5 long tons). The report stated that uranium was also produced in Australia, Germany and India but provided no quantities.

The next major uranium ore discovery was in 1930 at Great Bear Lake in north-west Canada. The site at Labine Point was developed by the Eldorado Gold Mining Company to mine pitchblende and, although operations started in 1932, they had stopped by 1940, when European radium markets were disrupted. The mining company had constructed a radium refining plant at Port Hope on Lake Ontario, in south-east Canada, with operations starting in 1933. Based on the early production figures for radium at Port Hope, at least 400 t of pitchblende had been transported from the mine to Port Hope by 1937. When the mine reopened in 1942 for uranium extraction, it was known as Port Radium and continued uranium extraction until 1960 [2.24].

2.6. DISCOVERY OF FISSION

The discovery and explanation of fission, at the end of 1938, is the beginning of a period in which aspects of science fiction (e.g. Ref. [2.25]) became a reality at the start of the atomic age. Some of the many notable scientists involved in the studies that supported this discovery were awarded Nobel prizes in physics and chemistry in the first decades of the twentieth century. Their work highlights the scientific landmarks of this period (as demonstrated by the wording of the Nobel Prize citations listed below) and the gradual changes in scientific understanding that supported the discovery of fission:

- Wilhelm Conrad Röntgen (1901) “in recognition of the extraordinary services he has rendered by the discovery of the remarkable rays subsequently named after him”;
- Antoine Henri Becquerel (1903) “in recognition of the extraordinary services he has rendered by his discovery of spontaneous radioactivity”;
- Pierre Curie and Marie Curie (1903) “in recognition of the extraordinary services they have rendered by their joint researches on the radiation phenomena discovered by Professor Henri Becquerel”;
- Ernest Rutherford (1908) “for his investigations into the disintegration of the elements, and the chemistry of radioactive substances”;
- Marie Curie (1911) “in recognition of her services to the advancement of chemistry by the discovery of the elements radium and polonium, by the isolation of radium and the study of the nature and compounds of this remarkable element”;
- Max Karl Ernst Ludwig Planck (1918) “in recognition of the services he rendered to the advancement of Physics by his discovery of energy quanta”;
- Frederick Soddy (1921) “for his contributions to our knowledge of the chemistry of radioactive substances, and his investigations into the origin and nature of isotopes”;
- Albert Einstein (1921) “for his services to Theoretical Physics, and especially for his discovery of the law of the photoelectric effect”;
- Niels Henrik David Bohr (1922) “for his services in the investigation of the structure of atoms and of the radiation emanating from them”;
- James Franck and Gustav Ludwig Hertz (1925) “for their discovery of the laws governing the impact of an electron upon an atom”;

— Arthur Holly Compton (1927) “for his discovery of the effect named after him”;
— James Chadwick (1935) “for the discovery of the neutron”;
— Frédéric Joliot and Irène Joliot-Curie (1935) “in recognition of their synthesis of new radioactive elements”;
— Enrico Fermi (1938) “for his demonstrations of the existence of new radioactive elements produced by neutron irradiation, and for his related discovery of nuclear reactions brought about by slow neutrons”.

Otto Hahn published his recollections on the discovery of fission in Scientific American in 1958 [2.26] and it is his account that is summarized here. Hahn recounts how James Chadwick’s discovery of the neutron in 1932 [2.27] gave physicists a new method of producing artificial elements. It allowed Enrico Fermi and co-workers (Amaldi, D’Agostino, Rasetti and Segrè) to bombard elements of the periodic table with neutrons and to produce a great number of artificial, radioactive isotopes [2.28]. Fermi’s group found that uranium also gave rise to new rapidly decaying substances and they suggested that neutron bombardment may have produced elements with a higher atomic number than uranium — elements at that time unknown [2.29]. Hahn recalls that he subjected the resulting substances to the same chemical procedures he had used to identify protactinium and the results appeared to confirm Fermi’s assertion that he had detected ‘transuranic elements’. In October 1934, Fermi reported that slowing the neutrons using large paraffin blocks enhanced the activation of the target material by factors ranging from several tens to several hundreds depending on the material [2.28, 2.30].

In a paper published in 1937, Irène Curie and Paul Savitch [2.31] described their production of a transuranic element with properties very different from those of the other known transuranic elements. This prompted Otto Hahn and Fritz Strassmann to look further into their results, and they found that after the transuranic elements had been precipitated and removed, the solution still contained some radioactive products. Their subsequent experiments produced four radioactive isotopes, and the precipitates were either barium or radium. At that time, there was no evidence suggesting that barium could possibly be produced from the irradiation of uranium with neutrons. They called the products radium I, II, III and IV with half-lives of <1 min (an estimate), 14 min, 86 min and approximately 300 h, respectively. Further analysis using carrier and tracer techniques with natural radium showed that these were not isotopes of radium, but were in fact barium and other elements. They shared their findings with Lise Meitner, a physicist they had worked with until she moved to the Netherlands in July 1938 and later to Sweden.

In January 1939, Hahn and Strassmann published their observations stating that the “experiments are at variance with all previous experiences in nuclear physics” [2.32] and a month later, Lise Meitner and Otto Frisch published their historic letter to Nature, ‘Disintegration of uranium by neutrons: A new type of nuclear reaction’ [2.33]. This interpreted the results of Hahn and Strassmann using Bohr’s model of the atom [2.16] to explain the cleavage of a heavy nucleus into two nuclei of medium size — an event that Meitner and Frisch named ‘fission’ — and estimating the energy release [2.34]. The fact that fission was accompanied by the release of enormous amounts of energy was soon confirmed by Frisch [2.35], and within weeks by Frédéric Joliot, Hans von Halban and Lew Kowarski [2.36], who showed that the process liberated sufficient neutrons to produce a chain reaction. The last key account of 1939 was that of Niels Bohr and John A. Wheeler who demonstrated that a chain reaction based on fast neutrons could not be sustained with natural uranium and that ^{235}U would need to be enriched relative to ^{238}U for this to happen [2.37]. The Frisch–Peierls Memorandum of March 1940 to the United Kingdom Government first suggested how this enrichment could be achieved and estimated the critical mass required [2.38]. In 1944, Hahn was awarded the Nobel Prize in chemistry “for his discovery of the fission of heavy nuclei”.

2.7. USE OF URANIUM IN NUCLEAR APPLICATIONS

There are many detailed accounts of the next period in the history of uranium, but it was some years before the Manhattan Project and the development of controlled nuclear chain reactions and nuclear explosive devices became public knowledge, owing to the secrecy of these events from 1940 to 1945. The first controlled and self-sustaining nuclear chain reaction was achieved by Fermi and co-workers at 15:25 on 2 December 1942 with Chicago Pile 1 using natural uranium [2.39]. At completion, the pile contained 5.4 t of uranium metal, 45 t of uranium oxide and 360 t of graphite. The pile was constructed using very pure graphite as a moderator to slow neutrons, and the nuclear reaction was controlled with wooden rods covered in cadmium foil. The reactor operated at about 0.5 W(th) on that first occasion and never exceeded 200 W, having no mechanism to remove the heat generated by the reaction [2.40]. The Manhattan Project saw the development of facilities for large scale enrichment of uranium (Oak Ridge, USA) and separation of plutonium from irradiated fuel (neutron capture by ^{238}U produces ^{239}U; this decays to ^{239}Np and then to ^{239}Pu) (Hanford, USA). Nuclear reactors were built at both sites:

(a) The X-10 Graphite Reactor (4 MW) at Oak Ridge: a natural uranium, graphite moderated and air cooled reactor used for research and development (November 1943).
(b) The B Reactor (250 MW) at Hanford: the first in a series of water cooled graphite moderated natural uranium reactors (from September 1944) used to produce plutonium

The first explosive device was detonated at the Trinity test site, located at the Alamogordo Air Base, USA at 05:29 on 16 July 1945 [2.41]. The Trinity test used a plutonium implosion device (assembled at Los Alamos, USA) and was soon followed by the bombing of Hiroshima, Japan at 08:15 on 6 August 1945 using a device constructed from ^{235}U. The Prefectural Industrial Promotion Hall in Hiroshima was close to the epicentre (Fig. 2.3)[2], and what remained of that building was preserved and is now a UNESCO World Heritage site:

> "The Hiroshima Peace Memorial (Genbaku Dome) was the only structure left standing in the area where the first atomic bomb exploded on 6 August 1945. Through the efforts of many people, including those of the city of Hiroshima, it has been preserved in the same state as immediately after the bombing. Not only is it a stark and powerful symbol of the most destructive force ever created by humankind; it also expresses the hope for world peace and the ultimate elimination of all nuclear weapons."[3]

In December 1953, President Dwight D. Eisenhower of the USA addressed the United Nations with his 'Atoms for Peace' speech and proposed:

> "the…responsibility of this [international] atomic energy agency would be to devise methods whereby this fissionable material would be allocated to serve the peaceful pursuits of mankind. Experts would be mobilized to apply atomic energy to the needs of agriculture, medicine and other peaceful activities. A special purpose would be to provide abundant electrical energy in the power-starved areas of the world."[4]

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[2] For the 1933 and 1945 photographs, all rights reserved by the Hiroshima Peace Memorial Museum.

[3] See https://whc.unesco.org/en/list/775

[4] See https://www.iaea.org/about/history/atoms-for-peace-speech

Following this statement, a programme was developed in the USA for the generation of electricity from nuclear power [2.42, 2.43].

The use of heat from nuclear reactors to generate electricity was first achieved at Oak Ridge (September 1948) and then at Idaho Falls, USA (December 1951), the latter providing an iconic photograph of four ordinary 200 W light bulbs powered by steam generated from a nuclear reactor (Fig. 2.4).

FIG. 2.3. The Hiroshima Peace Memorial in different years (from left to right): 1933 (photograph taken by M. Okuno, reproduced with permission courtesy of the Hiroshima Municipal Archives); 20 August 1945 (photograph taken by O. Masami, reproduced with permission courtesy of the Hiroshima Peace Memorial Museum); 18 July 2009 (reproduced with permission courtesy of C. Zeballos).

FIG. 2.4. Light bulbs at Idaho Falls, USA powered by steam generated by a nuclear reactor (reproduced with permission courtesy of the Idaho National Laboratory, USA).

These were followed by reactors supplying larger amounts of electricity under commercial operation, including the following:

- Obninsk, former Soviet Union (now Russian Federation) in December 1954: 5 MW(e) enriched uranium, pressurized water cooling and graphite moderator.
- Calder Hall, United Kingdom in October 1956: 49 MW(e) natural uranium, gas cooled and graphite moderator.
- Shippingport, USA in May 1958: 60 MW(e) pressurized water reactor containing a highly enriched uranium core and a natural uranium blanket.
- Marcoule, France in April 1959: 39 MW(e) natural uranium, gas cooled and graphite moderator.

It was recognized at an early stage that nuclear propulsion would allow naval vessels to operate almost continuously. The USA developed the pressurized water reactor, and the first nuclear powered submarine, USS Nautilus, was launched in 1954. Nuclear powered surface vessels were commissioned by both the USA and the former Soviet Union in 1959.

These developments resulted in the rapid growth of large scale uranium prospecting and mining activities during the 1950s and associated facilities providing uranium extraction from ore, uranium enrichment, fuel fabrication and hydrometallurgical treatment of irradiated uranium. The recovery of uranium from sources other than uranium deposits — such as fly ash [2.44], sea water [2.45], phosphate deposits [2.46] and sands (e.g. monazite) — was also considered.

2.8. IMPACT OF URANIUM PRODUCTION

2.8.1. Occupational impact of uranium mining

Although the occupational health risks associated with mining have been apparent for a long time, it is only in the last 50–60 years that controls have been implemented [2.47]. The impact on miners working in proximity to uranium deposits was first observed in cobalt miners in Schneeberg, Saxony in the sixteenth century [2.48] and was firmly linked to lung cancer in 1879 [2.49], when it was confirmed as causing the death of 23% of miners between 1869 and 1877. Further study confirmed primary carcinoma of the lung [2.50], and excess cases of lung cancer were also identified at Jáchymov in 1932 [2.51], as reported in a review by the United States National Research Council [2.52]. The link between radioactive substances and lung cancers was established [2.53] using epidemiological statistics and clinical observations, although others considered dusts and arsenic as causal agents. However, it was not until 1951 that the causal effect of alpha radiation emitted by radon progeny was identified by William F. Bale [2.54] in a memorandum to the United States Atomic Energy Commission. The United States Public Health Service started studies on the health of uranium miners in 1950, and in 1957 guidelines began to develop concerning worker limits for radon exposure [2.55]. In 1958, the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) noted that exposure of miners to radon was related to a high incidence of lung cancer; the available evidence was reviewed in 1964 but risk estimates could not be determined. In 1977, UNSCEAR again considered the high doses to the lung caused by exposure to radon and its progeny (producing dose to exposure quotients) and in 1987 the International Agency for Research on Cancer produced a monograph on radon as a carcinogen [2.56].

2.8.2. Environmental impact of uranium production

Once uranium ore has been mined/extracted, the next step in uranium production involves concentrating the uranium, and this is usually undertaken near the mine to reduce transportation costs. The method adopted depends on the nature of the ore and, before in-situ leaching became a widely used process, involved crushing and screening, calcining in some cases, followed by acid or alkaline leaching, solvent extraction or ion exchange and precipitation. In the case of in-situ leaching, the uranium dissolved in recovered leachate is mainly treated using ion exchange, stripping and precipitation; small distributed ion exchange facilities may feed a central plant where the ion exchange resin or polymer is stripped and the uranium precipitated. The dried precipitate is often referred to as ‘yellow cake’ (ammonium diuranate ($(NH_4)_2U_2O_7$)) owing to the colour of the products from the early production facilities, although it is now commonly oxidized to produce uranium oxide (U_3O_8 or UO_3) for shipment and varies in colour from brown to black. The material produced by the concentration stage typically contains 40–70% uranium by weight (but sometimes up to 90%) and is, generally, transported to a processing plant to be further refined. Sites involved in the production of yellow cake have large volumes of mill tailings associated with them, comprised of fine sands containing uranium progeny and other metals associated with the ore [2.57].

A refining stage then produces the uranium metal. This involves dissolving yellow cake in nitric acid and then selectively extracting uranium from the acid feed using tributyl phosphate diluted with kerosene (or another suitable hydrocarbon mixture). Finally, uranium is stripped from the tributyl phosphate extract into acidified water to yield a highly purified uranyl nitrate ($UO_2(NO_3)_2$).

The last part of the process begins with calcining the nitrate to UO_3 and then reducing the trioxide with hydrogen to UO_2. Powdered UO_2 is treated with gaseous hydrogen fluoride (HF) at 550°C to produce uranium tetrafluoride (UF_4). Conversion to uranium metal is accomplished through the Ames process, in which UF_4 is reduced with magnesium at temperatures exceeding 1300°C, producing metallic uranium and a slag of magnesium fluoride (MgF_2). Uranium tetrafluoride can also be fluorinated at 350°C with fluorine gas to volatile uranium hexafluoride (UF_6) used as a feedstock for isotopic enrichment.

Several methods, such as gaseous diffusion, gas centrifugation or liquid thermal diffusion, are available to concentrate ^{235}U. The degree of enrichment varies from 2–3% ^{235}U, typically used in light water reactors, to 97–99%, used in nuclear weapons. After enrichment, UF_6 is reacted with water vapour to produce hydrated uranyl fluoride ($UO_2F_2 \cdot H_2O$), which is subjected to hydrogen reduction to produce powdered UO_2. The enriched UO_2 can be used to produce ceramic fuel or uranium metal [2.57].

Concerns have developed over the environmental impact of uranium mining and production activities, in many cases leading to cleanup of sites that may have once operated with less stringent controls than those that are now enforced. Examples of site cleanup presented below are taken from mines and facilities mentioned earlier in this chapter and illustrate the large areas and volumes of material that may be involved. The environmental issues associated with former mines and areas investigated for their mining potential that may need to be dealt with include the following:

- Spoil heaps produced from discarded overburden and ore that had uneconomic levels of uranium content or high levels of contaminants;
- Uranium mill tailings;
- Waste from heap leaching operations;
- Water treatment sludges;
- Acid mine drainage leading to surface water and groundwater contamination;
- Contamination of the local environment (e.g. soils, homes);
- Mine shafts and adits, processing equipment and structural components.

The borehole drilling operations for in situ leaching produce relatively little solid waste but the recovery of uranium generates a range of other waste types, including the following:

- Solids and sludge from the neutralization of solutions;
- Spent ion exchange resins;
- Salt residues and filters from reverse osmosis plants;

— Scales from pipework, pumps, valves and filters;
— Residues from evaporation ponds.

Cleanup of the Port Radium mine, Canada was conducted on three occasions, each time to the standards of the day [2.58]. The first cleanup was in 1962, when the underground mines were decommissioned and major equipment removed. Subsequently, in 1982, most of the site structures were demolished, other equipment removed, shafts and adits secured, and some areas with elevated levels of gamma radiation were covered with waste rock. The final phase started on site in 2007 following an options study and consultation with the local community to define a site end state and included post-remedial short and long term monitoring. The approach, common to many cleanup projects, used to assess, plan and execute site remediation involved the following eight stages:

(1) Investigate existing or potential concerns through site assessment;
(2) Develop remedial options, evaluate preliminary engineering plans and costs, and present the preferred action plan;
(3) Develop cost estimates based on the preferred remedial option;
(4) Prepare final remedial engineering and tender documents;
(5) Commence the bidding process, evaluate the submitted bids and award the work;
(6) Undertake the remedial works;
(7) Prepare ‘closure’ documentation and presentation of findings to stakeholders at completion of the works;
(8) Implement post-remedial short and long term monitoring and assessment.

At Central City, USA, a 103 600 ha watershed has been designated as a Superfund site with multiple mine waste piles, tailings impoundments, draining mine adits and impacted groundwater resources[5]. Cleanup is ongoing in this area and includes areas around the Wood Mine near Quartz Hill where uranium was found in 1871. The work includes treatment of various discharges, sediment control by capping or removal of waste piles and construction of an on-site repository. The Quartz Hill tailings pile is being re-graded to a manageable slope for runoff from rain or snow melt and vegetative cover is being reinstated to prevent sediment erosion [2.47].

Uranium mining in Colorado along the Uravan Belt has resulted in over 1200 historical mines that were abandoned once the ore veins were exhausted.

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[5] See https://cumulis.epa.gov/supercpad/SiteProfiles/index.cfm?fuseaction=second.cleanup&id=0800257

The tailings were left unprotected and wastewater from processing facilities was discarded as surface water. Indeed, tailings were used as fill for several purposes including roads, cement mixing and home construction. This resulted in contamination of over 4000 residential and commercial properties in Grand Junction, USA that eventually needed remediation. These problems led to the Uranium Mill Tailings Radiation Control Act (UMTRCA) in 1978 that provided for the regulation of tailings and cleanup of tailings at inactive and/or abandoned mines, and set standards for active processing facilities in the USA. Mill tailings contain the progeny radionuclides and heavy metal contaminants indigenous to the ore, plus the acids or strong bases and organic compounds used to leach the uranium from the rock. Unstabilized tailings material can be spread by wind and water erosion to adjacent land and communities.

Mill tailings ponds constructed prior to the UMTRCA are unlined and have been a source of extensive localized groundwater contamination. One of many site cleanup examples from the USA is the Cotter Uranium Mill, located near Canon City, USA and referred to as the Lincoln Park site [2.59]. This 1050 ha site included an inactive mill, ore stockpiles and tailings ponds (older unlined ponds and lined ponds constructed since 1978). The site history shows numerous interventions to reduce groundwater contamination and is a good example of the public health assessments undertaken to determine the efficacy of cleanup activities. The ongoing cleanup of the uranium processing facility near Moab, USA will remove 16 million m^3 of uranium mill tailings from an area of 52.6 ha immediately next to the Colorado River and place them in a permanent disposal facility (Fig. 2.5). The operation was 52% complete in October 2016 and is expected to continue until 2032 [2.60].

The most recent project to clean up the Port Hope locality, Canada started removing waste for disposal in 2015. Previously, about 100 000 t of uranium contaminated soil had been removed for disposal at the Chalk River Laboratories but this cleanup stopped in 1981. Waste from various sites in Port Hope is being removed and transported to a local, long term, low level radioactive waste management facility for disposal and monitoring for several hundred years. The origins and predicted volumes of waste material are as follows[6]:

(a) Soil contaminated by refinery wastes (620 000 m^3);
(b) Sand and silt soils from harbour dredging and remediation sites (572 000 m^3);
(c) Industrial waste contaminated sites (51 250 m^3);
(d) Decommissioning waste materials (150 000 m^3).

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[6] https://www.phai.ca/wp-content/uploads/EA_ExecutiveSummary_PH.pdf

Examples of legacy sites in Central Asia are included in Section 9.1. Reference [2.61] includes a case study of cleanup at Olen, Belgium, where the uranium ore from Katanga, Belgian Congo was processed.

2.8.3. Environmental impact of used nuclear fuel

After producing energy in a reactor, the amount of ^{235}U in the fuel will have declined and the spent fuel contains new nuclides such as fission products (e.g. radioisotopes of caesium, iodine and strontium), minor actinides (neptunium, americium and curium) and plutonium. The largest proportion of the spent fuel is still comprised of uranium. After removal from the reactor core, spent fuel spends time in water filled cooling ponds close to the reactor, the water providing both shielding and coolant. The period of storage in the ponds varies between facilities and it remains there until accepted for the following:

- Reprocessing (reducing waste, producing plutonium and recycling uranium);
- Geological disposal;
- Interim storage (wet or dry) at facilities on the reactor site or in centralized facilities away from the reactor, pending onward shipment for reprocessing or disposal.

FIG. 2.5. Ongoing cleanup of a uranium processing facility near Moab, USA (photograph looking north-east, April 2014) (photo courtesy of Olympus Aerial Surveys and the United States Department of Energy).

The main options for spent fuel storage have been reviewed recently [2.62] and can be grouped into the following five types:

(1) Pond: A facility that stores spent fuel in water, supported in racks, baskets and/or containers that also contain water.
(2) Vault: A reinforced concrete building containing arrays of storage cavities suitable for containment of one or more spent fuel units.
(3) Metal cask: A container with a bolted lid, designed either for storage only or for storage and transport.
(4) Concrete cask: A thick, welded, steel canister inserted into a concrete overpack with convection cooling.
(5) Silo: Similar to a vertical concrete cask, except that there is no cooling flow inside the monolithic structure.

Although spent fuel storage facilities at reactors were intended to be used for a short period, their use was extended owing to the long period of time necessary to develop disposal facilities and the limited reprocessing facilities available (see IAEA Safety Standards Series No. SSG-15 (Rev. 1), Storage of Spent Nuclear Fuel [2.63]). This change in management of spent fuel also needs to be considered alongside other developments, for example increased ^{235}U enrichment, increased burnup and the use of advanced fuel design and mixed oxide fuel.

The Hanford site reactors, USA shut down in 1987 and plutonium production stopped in 1989. The Hanford site represents one of the largest and most complex cleanup projects in the USA. Work has progressed with cleanup of parts of the site contaminated with radionuclides and chemical waste. A tripartite agreement between the Washington State Department of Ecology, the United States Environmental Protection Agency (EPA) and the United States Department of Energy concerning site cleanup highlights the scale of issues associated with the site [2.64] as follows:

> “An estimated five billion cubic yards of solid and dilute liquid wastes, which include hazardous substances, mixed waste, and hazardous waste and constituents have been disposed of at the Hanford Site. Significant above-background concentrations of hazardous substances, including chromium, strontium-90, tritium, iodine-129, uranium, cyanide, carbon tetrachloride, nitrates, and technetium-99 have been detected in the groundwater (unconfined aquifer) at the Hanford Site. These materials have toxic, carcinogenic, mutagenic, or teratogenic effects on humans and other life forms.”

2.9. CURRENT SITUATION

Over the period 1913–2013, mineral production reports list about 40 countries as having produced uranium ore at some time, with the number of countries and tonnage growing rapidly from the mid-1950s. In 2013, the major producers were Kazakhstan (22 567 t), Canada (9 331 t) and Australia (6 370 t) based on uranium metal content. The uranium produced in Kazakhstan and Australia is all exported to other countries, and in Canada 85% of production is exported. The trade in uranium is subject to IAEA nuclear safeguards under the Treaty on the Non-Proliferation of Nuclear Weapons (NPT) [2.65]. Nuclear materials used in civil nuclear programmes are subject to inspection by the IAEA but this does not include uranium ore and ore concentrates (yellow cake), which are considered source material and are traded as a commodity. The import and export of source materials is reported to the IAEA under the NPT.

The demand, production and reserves of uranium have been reported periodically since 1965 in a joint publication of the OECD Nuclear Energy Agency and the IAEA (Red Book) [2.66]. These joint reports are now produced every two years. Uranium resources are listed by country, showing both identified deposits and "reasonably assured resources", which are characterized in different bands related to the cost of recovery (ranging from <US $40/t to <US $260/t) and by method of production. The global distribution of identified resources (<US $130/kg as of 1 January 2019) shows that Australia (28%), Kazakhstan (15%), the Russian Federation (8%), Canada (9%), Niger (4%), South Africa (5%), Namibia (7%), Brazil (5%) and China (4%) between them hold about 85% of the world's identified uranium reserves. Historical summaries of exploration and production as well as plans for future mine production are also presented in the Red Book [2.66]. Country reports detail recent developments in uranium exploration and production, updates on environmental activities, regulatory requirements and information on relevant national uranium policies.

Uranium mining has progressed from subterranean and open pit excavation of ore and now also involves in situ leaching of unconsolidated sandstone deposits. In Kazakhstan and the USA, almost all extraction is now by in situ leaching, while in Canada uranium ore is still predominantly mined; in Australia about 45% of uranium is mined from open pits, about 6% by in situ leaching and the remainder is produced with, or as a by-product of, mining for other minerals.

The mined uranium is predominantly used for power generation. By 2018, there were 451 operating nuclear power reactors, with a total net installed capacity of 396 911 MW(e), a further 55 nuclear power reactors were under construction and about 172 nuclear power reactors permanently shut down [2.43]. Nuclear reactors were operating in 30 countries at the end of 2018 and a further four countries had reactors under construction. The IAEA carries out regular inspections of nuclear facilities and other locations in 57 non-nuclear-weapon States. In these inspections, State records and the nuclear material are audited. The aim of IAEA safeguards is to deter the diversion of nuclear material from peaceful uses.

2.9.1. Non-nuclear applications

Over the period of enriched uranium production, considerable stocks of depleted uranium have accumulated. The two feedstocks for depleted uranium are enrichment of natural uranium and as a by-product from the recovery of ^{235}U during spent fuel reprocessing. The OECD Nuclear Energy Agency estimates that depleted uranium stockpiles worldwide amounted to 1 600 000 t at the end of 2005 [2.67].

Reference [2.68] discusses the applications of depleted and natural uranium, including its use for colouring glass-ware and in dentistry until the middle of the twentieth century. Reference [2.68] also notes that jewellery pieces are still being made with an enamel powder containing uranium for enamel plates, pendants and rings. The use of natural and depleted uranium for dental porcelains to obtain a natural colour and fluorescence of dentures, and the superficial part of crowns stopped in the early 1980s. The use of uranium as a catalyst has been reviewed in Ref. [2.69] and it is reported that nickel–uranium catalysts are used widely in the oil and gas industry [2.70]. The four other major uses are radiation shielding, counterbalance weights and military armour and ammunition (kinetic energy penetrators) [2.71].

Vessels and equipment such as boats and satellites require high density ballast and uranium has been used for counterbalance weights in civil aircraft, military aerospace and boat building [2.70]. Tungsten has replaced depleted uranium in counterbalance weights in new civil aircraft and the number of aircraft that contain depleted uranium is, therefore, declining as they come to the end of their service life. Military applications of depleted uranium include kinetic energy penetrators and tank armour. These developments started in the early 1970s [2.72] and depleted uranium was selected over tungsten owing to its lower price and high pyrophoricity.

The density of depleted uranium makes it effective at shielding gamma radiation. It has been used extensively in the medical, research and transport sectors as radiation beam collimators and in containers for the transport of radioactive sources [2.73]. Depleted uranium is used for vitrified high level waste packages [2.74] and depleted uranium silicate glasses are used as backfill for spent nuclear fuel waste [2.73]. There have also been investigations into the use of depleted uranium in the construction of casks to hold spent nuclear fuels [2.75].

2.9.2. Uranium present in naturally occurring radioactive material

Naturally occurring radioactive material (NORM) is defined in the IAEA Nuclear Safety and Security Glossary as follows [2.76]:

> "*Radioactive material containing no significant amounts of radionuclides other than naturally occurring radionuclides.*
>
> "Material in which the *activity concentrations* of the *naturally occurring radionuclides* have been changed by a *process* is included in *naturally occurring radioactive material (NORM).*"

There are about 12 industrial activities that produce volumes and radioactivity levels that may need to be considered when controlling exposure to solid, liquid and gaseous NORM waste. As well as uranium derived from mining and processing of uranium ore, uranium can be found in the waste or products from the following industries:

- Mining of ores other than uranium ore;
- Extraction of rare earth elements (REEs);
- Extraction of china clay;
- Production and use of thorium and its compounds, and of niobium and ferroniobium;
- Production of oil and gas;
- Production of aluminium, copper, iron and steel, lead, tin and zinc;
- Manufacture of titanium dioxide pigments;
- Phosphate industry;
- Zircon and zirconia industries;
- Combustion of coal;
- Water treatment.

The IAEA has issued publications in the Safety Reports Series on oil and gas production [2.77], work involving minerals and raw materials [2.78], zircon and zirconia industries [2.79], production of rare earths from thorium-containing minerals [2.80], residues from titanium dioxide processing and related industries [2.81] and residues from phosphate processing [2.82]. The publications provide overviews of each industry and consider radiation protection principles in relation to materials management and waste disposal.

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Chapter 3

PHYSICAL AND CHEMICAL PROPERTIES OF URANIUM

T.E. PAYNE
Australian Nuclear Science and Technology Organisation, Australia

J.M. WEST
West Consult, United Kingdom

3.1. PHYSICAL PROPERTIES

Uranium (atomic number 92) is a silver-white, ductile and slightly paramagnetic metal that has a very high density, being approximately twice as dense as lead. It was the first element that was found to possess the property of radioactivity (by Henri Becquerel in 1896). The melting point of the metal is 1132°C and its density is about 19 g/cm^3 [3.1]. Uranium is ubiquitous in the Earth's crust (with an average concentration of around 2.7 ppm) and is also present in sea water (at around 3.3 ppb) [3.2]. Uranium and its decay products, most of which are also radioactive, contribute to the levels of natural background radiation in the environment.

3.1.1. Isotopes of uranium

There are three main isotopes present in natural uranium: ^{234}U, ^{235}U and ^{238}U (Table 3.1 [3.3]). In addition, traces of ^{236}U are found in nature, with the highest values created by nuclear reactions in rich uranium deposits where ^{236}U:^{238}U mass ratios of about 10^{-10} have been reported [3.4].

The fissile isotope ^{235}U provides the energy source for nuclear power reactors and weapons. For these purposes, the relative proportion of ^{235}U must generally be increased through a process known as 'uranium enrichment' (although some nuclear reactors can utilize natural uranium as fuel). Most nuclear power reactors require uranium in which the ^{235}U content has been enriched to about 3–4%. Typically, enrichment also increases the content of ^{234}U, as most enrichment processes make use of the mass differences between the various isotopes. The uranium remaining after removal of the enriched fraction is referred to as depleted uranium, which typically contains about 99.8% (by mass)

TABLE 3.1. NUCLEAR PROPERTIES OF NATURAL URANIUM [3.3]

Characteristic	^{234}U	^{235}U	^{238}U
Nuclide half-life (a)	2.46×10^5	7.04×10^8	4.47×10^9
Natural abundance (wt%)	0.005 5	0.720	99.274 5
Principal mode of emission	Alpha	Alpha	Alpha
Main alpha energies (MeV)	4.776 4.725	4.395 4.365	4.197 4.147

of ^{238}U, 0.2% of ^{235}U and 0.000 6% of ^{234}U. The depleted uranium contains, on a mass basis, about 60% of the radioactivity of the natural uranium [3.5].

The ratio ^{235}U:^{238}U is generally uniform in natural samples obtained from different environments, although this ratio is gradually decreasing with geological time, owing to the shorter half-life of ^{235}U. In contrast, the ratio ^{234}U:^{238}U can be quite variable in nature, particularly in groundwater samples. Uranium-234 is a decay product in the ^{238}U chain. In a sample that has been undisturbed over long geological timescales, the radioactivity of the parent (^{238}U) and its progeny (^{234}U) in the solid phase will be at equilibrium (i.e. the activity ratio ^{234}U:^{238}U is about 1.0; see Section 3.1.2)). However, ^{234}U is often preferentially released to the groundwater phase, so the progeny to parent (^{234}U:^{238}U) activity ratio in natural waters is often out of secular radioactive equilibrium [3.6]. The major reason for this disequilibrium is related to the energetic alpha decay of ^{238}U and differential release of ^{234}U relative to ^{238}U. This disequilibrium originates from: (i) preferential release of more loosely bound ^{234}U from damaged mineral lattice sites or (ii) direct recoil of ^{234}Th into the surrounding medium from nearby mineral surface boundaries, but it is unclear which of the two mechanisms is more important in nature [3.7]. As a result, ^{234}U:^{238}U activity ratios in nature can vary across a large range with values in groundwater often exceeding ten [3.8]. In one study, ^{234}U:^{238}U ratios of as high as 410 were reported in specific mineral phases that were chemically extracted from a uranium ore sample [3.9]. In some cases, values of ^{234}U:^{238}U significantly below unity have been reported in environmental samples, including groundwaters [3.10].

Whereas the ratio between the ^{235}U and ^{238}U isotopes is generally almost uniform in nature, a significant deviation from the usual ratio has been reported in samples from the Oklo uranium deposits in Africa. In this location, spontaneous nuclear reactions occurred, which have resulted in significant differences in isotopic abundance from typical known values. The Oklo phenomenon has created intense scientific interest and has been studied for several decades. The nuclear reactions at Oklo generated many nuclides not usually present in nature, which has provided an opportunity to study the geochemical mobility of those nuclides [3.11]. In addition to the well known Oklo isotopic anomalies, more recently, evidence has emerged of widespread slight natural fractionation between ^{235}U and ^{238}U [3.12], which is only measurable by high precision analytical techniques.

Human activities have resulted in several additional uranium isotopes being released into the environment (e.g. ^{233}U, ^{236}U) as well as perturbations in natural uranium isotope ratios. For example, depleted uranium (i.e. having low ^{235}U:^{238}U ratios) has been dispersed as a consequence of being used in armour piercing munitions and has also been released into the environment from other sources, such as aircraft crashes, as it is often a component of airplane stabilizers [3.5]. In nuclear fuel, ^{236}U builds up owing to neutron capture by ^{235}U. Consequently, nuclear waste disposal or accidental releases from power plants can lead to variations from the natural ^{236}U abundance in environmental samples [3.9]. However, the most significant source of ^{236}U in the environment is the fast neutron reaction involving ^{238}U when thermonuclear weapons are detonated.

3.1.2. Decay series of ^{238}U and ^{235}U

Uranium-238 is the parent radionuclide in a decay chain (known as the ^{238}U series) that includes 14 radionuclides (Fig. 3.1) of which eight are alpha emitters and six beta emitters. The ^{238}U decay series contains radioisotopes of various elements exhibiting a range of chemical behaviour (some with half-lives >10 a). The parent, ^{238}U ($T_{1/2} = 4.47 \times 10^9$ a), decays by alpha emission to ^{234}Th, which in turn decays to ^{234}Pa and then through a series of other radionuclides to stable ^{206}Pb. The longest lived members of this chain include ^{234}U, ^{230}Th and ^{226}Ra. The ^{238}U decay chain includes the short lived isotope ^{222}Rn, which is a noble gas.

As with ^{238}U, the less abundant isotope ^{235}U decays through a chain to another isotope of lead, ^{207}Pb (Fig. 3.2). In this decay chain, the longest lived isotope is ^{231}Pa, with a half-life of 3.3×10^4 a.

From a public health viewpoint, the ^{238}U series contains two intermediates of special interest, ^{226}Ra ($T_{1/2} = 1600$ a) and its immediate decay product, ^{222}Rn ($T_{1/2} = 3.82$ d). Radium-226 and its progeny are responsible for a major fraction of the radiation dose from uranium series radionuclides received via the inhalation and ingestion pathways [3.13]. The average annual effective dose for this decay series has been estimated as 1.34 mSv, a combination of an external exposure of 0.10 mSv and a larger internal component of 1.24 mSv [3.14].

Inhalation of ^{222}Rn is considered to be a major health issue internationally, since radon can build up to high levels within modern buildings, leading to a

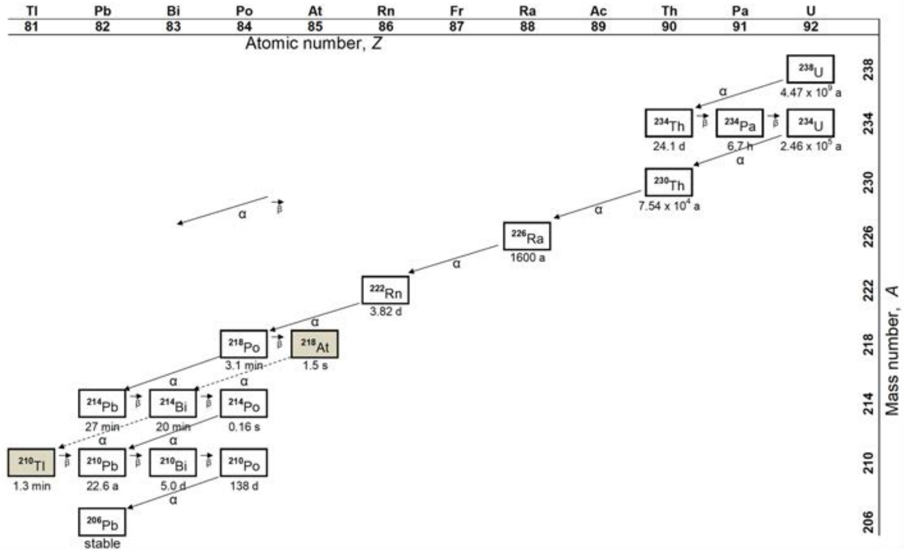

FIG. 3.1. The ^{238}U decay series, showing half-lives and decay modes of the intermediate radionuclides leading to the final product, ^{208}Pb, which is stable. Less than 0.1% of decays involve either ^{218}At or ^{210}Tl [3.13]. These radionuclides are shown in grey boxes.

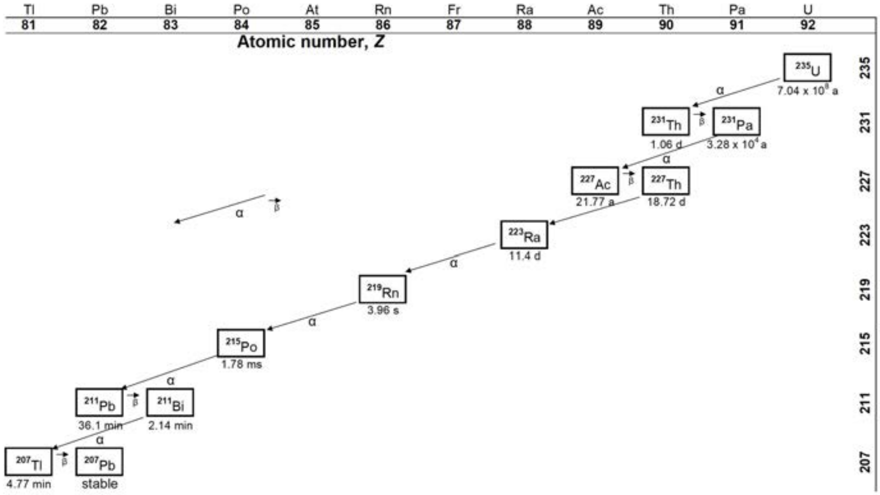

FIG. 3.2. The ^{235}U decay series, showing the intermediate radionuclides leading to the final product, ^{207}Pb, which is stable.

substantial potential dose through inhalation [3.13]. Prolonged exposure to high levels of radon is linked with illnesses such as lung cancer and bronchial tissue damage [3.15]. Radon monitoring at locations with elevated concentrations, such as underground mines or caves, is important to assess the radiological hazards to workers and tourists [3.16, 3.17]. A study of the health impacts of radionuclides in groundwaters in the Guarani aquifer, Brazil [3.18], concluded that the main contributor to the human dose was ^{226}Ra, although ^{222}Rn was also considered to be of concern because of its possible presence inside spa facilities that are common in the region.

Each decay series includes radionuclides having a wide range of half-lives and involves several chemical elements that have substantially different characteristics. The concentration of each radionuclide is related to that of its parent and also depends on the time since fractionation between the radionuclide and its parent has occurred. For samples that have been isolated over long geological timescales, the activity of each radionuclide (the decay rate) is the same as that of its parent; this is referred to as 'secular equilibrium'. For example, for the ^{238}U decay series in a stable system, the following applies:

$$(^{238}\mathrm{U}) = (^{234}\mathrm{Th}) = (^{234}\mathrm{Pa}) = (^{234}\mathrm{U}) = \ldots \tag{3.1}$$

using the standard notation where a radionuclide in parentheses denotes the (radio)activity of that radionuclide.

There are two important points to note [3.19]:

(a) In secular equilibrium, the distribution of a daughter radionuclide is controlled by that of the parent, and so, ultimately, all isotopes in the series are controlled by the long lived ancestor (in this case, ^{238}U);

(b) Whereas the activities are equal, the molar abundances are inversely proportional to the decay constants, so that the mass concentrations of very short lived nuclides are extremely low.

As many of the radionuclides in the ^{238}U decay chain have long half-lives, not all radionuclides in the chain will be in secular equilibrium. However, the radionuclides in part of the series may achieve secular equilibrium rapidly if the system is kept sealed (or isolated). For example, if a sample of gas containing ^{222}Rn is kept in a sealed container, the activities of several short lived progeny radionuclides (i.e. ^{218}Po, ^{214}Pb, ^{214}Bi and ^{214}Po) will quickly build up to equal that of ^{222}Rn (although all isotopes will decay eventually owing to the short half-life of ^{222}Rn). It is important to note that as the progeny are not gaseous, they will behave differently from ^{222}Rn. Therefore, while the total activity of ^{214}Pb may quickly match that of ^{222}Rn, it will tend to be associated with suspended particles or, possibly, the walls of the container, to a much greater extent. Similarly, in the Earth's atmosphere, the decay products, which are more strongly associated with aerosol particles, become subject to gravitational settling and scavenging by rainfall [3.20], again causing separation from their parent. Such differences in chemical form are important considerations both in measurements of radionuclide activities and in assessing doses.

As noted above, in a closed system, the radioactivity of all radionuclides in these decay series tends to approach an equilibrium value, in which the rate of supply of a radionuclide from the decay of its parent is matched by the rate of its decay to subsequent radionuclides in the decay chain. However, the relative abundances of the various radionuclides in a decay series can be highly variable in natural systems if there are processes that lead to the depletion of some radionuclides relative to the other radionuclides. A typical example of this is the escape of radon isotopes, which are present in the decay chains of both ^{238}U and ^{235}U, and, being gaseous, can diffuse out of various environmental matrices. These factors, together with the disruptive nature of the alpha decay process (owing to recoil of the residual nucleus after alpha emission), can result in fractionation between different radionuclides in the chain. As the ^{235}U and ^{238}U series result in different stable isotopes of lead (^{207}Pb and ^{206}Pb, respectively), these decay series provide the basis of methods for determining the provenance of geological samples based on the isotopic ratios of their lead content. These characteristics of the uranium decay chains can be exploited to determine the rates and mechanisms of various environmental processes, including age dating and groundwater mixing processes [3.21].

3.1.3. Research applications of isotopic ratios in the uranium decay series

As mentioned in Section 3.1.2, disequilibria between the members of the uranium series decay chains can be accurately measured and the results can be applied in a wide variety of contexts including geochronology, erosion, sedimentation, exploration geology and assessment of groundwater resources [3.21]. Specifically, uranium series disequilibria can be used to examine the rates and timescales of any dynamic process that induces isotopic fractionation. In many cases, the measurement of disequilibrium provides a powerful means of tracing specific processes.

These phenomena have a wide range of applications in the geosciences and environmental sciences. A major review is provided in Ref. [3.21], which discusses numerous specific research topics, including the following:

- Geochronology: The sequence and timing of events in the Earth’s history.
- Dating: Archaeological applications, including dating of fossil bones and teeth.
- Groundwater and surface hydrology: Sources and mixing proportions of aquifers, dating and resource estimation.
- Geological processes: Processes, timescales and elemental fractionation during formation and alteration of igneous rocks; rock–water interactions; and geothermal systems.
- Weathering, erosion, soil formation and landscape evolution.
- Sediment transport and deposition, including lake sediments and marine sediments.
- Uranium exploration and resource estimation.
- Oceanic studies: Chronometry of marine processes, including aerial deposition, particle settling and ocean circulation.
- Paleoclimatology and ancient sea levels.
- Long term behaviour of radionuclides in the geosphere (which is relevant to radioactive waste disposal studies).

It is valuable to date environmental, geological and archaeological materials for two main reasons [3.22]. First, a precise age for the formation of a sample (be it a bone, environmental media, such as groundwater, or a mineral) establishes its archaeological or geological context. Second, the timespan between samples that were formed under different conditions makes it possible to calculate the rate of change and provides information on the process that caused the change. Uranium series measurements have also been applied recently to help to predict the potential leakage of geologically stored carbon dioxide [3.23, 3.24].

In the groundwater environment, isotopic disequilibrium can be used to identify mixing of water bodies, particularly if several isotopes in the uranium series decay chains can be used. In paleoclimatology, a range of environmental samples may be analysed to give information on climate history. Stalagmites are often used in these types of study, because successive layers of calcite are neatly deposited on top of each other, corresponding to a well defined drip point at the tip of a stalactite. Therefore, they form a long archive of environmental conditions prevailing at the time of deposition. Stalagmites tend to initially be low in ^{230}Th and frequently show evidence of a complicated growth history caused by changes in hydrological conditions because of climate variations such as dry periods or episodes of glaciation [3.22].

3.2. NATURAL DISTRIBUTION AND MINERALOGY

3.2.1. Uranium distribution

Uranium is widely distributed throughout the Earth, with economic deposits occurring in many countries in a range of geological settings in igneous, metamorphic and sedimentary environments. Although uranium can exist in oxidation states from +3 to +6 in aqueous solutions [3.25], it is commonly found in its reduced state, U^{4+}, which is generally highly immobile, or in its more soluble and mobile state, U^{6+}. Rare instances of U^{5+} have also been reported [3.26]. Oxidation states may also be indicated by Roman numerals; for example, U(IV) and U(VI).

Oxidation and chemical alteration of the primary uranium ores can often lead to the formation of U^{6+} minerals, including vanadates, phosphates and silicates [3.2]. Interactions with groundwater and weathering processes are primary factors leading to the migration of uranium from deposits to the biosphere. Sandstone and permeable sedimentary rocks are most susceptible to leaching by groundwater. Bedding planes and structural features (fractures and faults) in less permeable rocks also serve as conduits for groundwater transport [3.27].

Away from such mineralized deposits, uranium is widely dispersed throughout the environment. The uranium content in common rock types is in the range of 0.5–4.7 mg/kg, with an average crustal abundance of about 2.7 mg/kg [3.2]. Lower concentrations of uranium are found in basic rocks, whereas acidic rocks contain higher uranium concentrations. It is also found in phosphate rock, lignite and monazite sands. In soils, uranium can be found in adsorbed, organically bound or precipitated, forms. Retention of uranium by the soil may be due to adsorption, chemisorption, ion exchange or a combination of mechanisms (discussed in Section 3.2.2).

3.2.2. Mineralogy and geochemical behaviour

Although the details of the mineralogy and geological behaviour of uranium are beyond the scope of the present book, a general introduction is provided here, since the principles controlling the geological distribution and geochemistry of uranium provide a useful background to the environmental chemistry of uranium. Furthermore, the formation of uranium minerals can control the groundwater concentrations of uranium in some contaminated soils (see Section 3.3.2) [3.28].

Uranium minerals have a considerable diversity of structure and chemistry, which reflects the variety of conditions under which they formed (Table 3.2). Many major deposits of uranium comprise accumulations of U^{4+} oxides, such as uraninite and pitchblende (UO_2 to U_3O_8), as well as silicates ($USiO_4$). Although many other minerals containing U^{4+} exist, these two types of mineral are the most abundant and of greatest economic importance.

In oxidizing, aqueous environments, uranium mineral alteration and mobilization typically involves oxidation of minerals containing reduced uranium (U^{4+}) to its more mobile form, U^{6+}. Alteration mechanisms may include complete dissolution (e.g. when aqueous carbonate or sulphate complexes can form) or replacement (e.g. by uranyl silicates).

In aqueous solutions, U^{6+} is not present as individual atoms, but is always bound strongly to two oxygen atoms, forming the nearly linear uranyl ion (UO_2^{2+}). This is a highly stable species and forms a building block for numerous aqueous uranyl species and uranyl minerals (i.e. containing U^{6+} as UO_2^{2+}). Some of the main factors influencing the formation of uranyl minerals are the groundwater chemistry (specifically, the abundance of uranyl complexing species, such as carbonate, silica and phosphate), the abundance of other metals, such as copper, calcium and magnesium (which are constituents of many uranyl minerals), the relative mineral solubilities and the stability of relevant uranyl solution complexes. These relationships are summarized in Fig. 3.3, which provides an overview of the behaviour of the UO_2^{2+} ion in near surface groundwaters.

TABLE 3.2. EXAMPLES OF SOME OF THE MAIN CLASSES OF URANIUM MINERALS

Class of mineral	Examples	Simplified chemical formula	Notes
Oxide containing U(IV)	Uraninite, pitchblende	UO_{2+x} (e.g. U_3O_8)[a]	Always partially oxidized and containing other elements
U(IV) silicate	Coffinite	$USiO_4 \cdot nH_2O$[b]	Apart from uraninite, this is the most abundant uranium ore mineral
Uranyl oxyhydroxide	Schoepite	$(UO_2)_8O_2(OH)_{12}(H_2O)_{12}$	
	Becquerelite	$Ca(UO_2)_6O_4(OH)_6(H_2O)_8$	Named after Henri Becquerel

TABLE 3.2. EXAMPLES OF SOME OF THE MAIN CLASSES OF URANIUM MINERALS (cont.)

Class of mineral	Examples	Simplified chemical formula	Notes
Uranyl carbonate	Rutherfordine	UO_2CO_3	Named after Ernest Rutherford
	Bayleyite	$Mg_2(UO_2)(CO_3)_3(H_2O)_{18}$	
Uranyl silicate	Soddyite	$(UO_2)_2SiO_4(H_2O)_2$	Named after Frederick Soddy
	Uranophane-α	$Ca(UO_2)_2(SiO_3OH)_2(H_2O)_5$	Most common uranyl mineral
Uranyl phosphate	Autunite	$Ca[(UO_2PO_4)]_2(H_2O)_{10–12}$	Uranyl phosphates and arsenates constitute a diverse and widely distributed group of uranium minerals
	Saleeite	$Mg[(UO_2PO_4)]_2(H_2O)_{10}$	
	Metatorbernite	$Cu[(UO_2PO_4)]_2(H_2O)_{6–8}$	
Uranyl arsenate	Heinrichite	$Ba[(UO_2)(AsO_4)]_2(H_2O)_{10–12}$	
Uranyl vanadate	Carnotite	$K_2(UO_2)_2(V_2O_8)(H_2O)_3$	
Uranyl sulphate	Johannite	$Cu(UO_2)_2(SO_4)_2(OH)_2(H_2O)_{6–8}$	
Lead-bearing minerals	Uraninite ('old')	$(U,Pb)O_2$	Up to ~20 wt% PbO
	Parsonsite	$Pb_2(UO_2)(PO_4)_2(H_2O)_2$	

[a] x has a numerical value of typically $\ll 1.0$.

[b] n, which is an integer, is the number of water molecules incorporated into the mineral structure.

Table 3.2 shows examples of several of the main classes of uranium minerals, including the main U^{4+} minerals found in nature, namely uraninite and coffinite (a U(IV) silicate). It can be observed that major classes of uranium minerals are often characterized by the anionic species present, commonly oxygen-containing species such as arsenate, carbonate, phosphate and vanadate. Other uranium mineral types include molybdates, selenites, tellurites and tungstates. Additionally, many uranium minerals contain other metals as major components (such as barium, calcium, copper, lead and magnesium).

The minerals in Table 3.2 have a variety of crystal structures that can be determined by techniques such as X ray diffraction. An interesting feature of the geochemistry of uranium is that because it is radioactive and ultimately decays to lead, the uranium content of the original mineral slowly decreases. However, the crystal chemistries of lead and uranium are quite different, which means that the structures of uranium minerals can be destabilized if they are old enough to contain substantial radiogenic lead. It is possible under reducing conditions, if both lead and uranium are immobile, that the uraninite crystal structure becomes strained and potentially fractured. Under oxidizing conditions, lead and UO_2^{2+} can combine and together form a new lead–uranyl mineral. Many of the uranyl mineral types shown in Table 3.2 have related mixed lead–uranyl minerals. Owing to the different mobility of lead and uranium in many groundwaters, lead-bearing uranyl minerals dissolve incongruently. Often, uranyl is released, but lead remains behind and can accumulate, leading to the formation of minerals having higher lead contents.

The Koongarra ore body, northern Australia, demonstrates many of the natural processes that are relevant to the mobilization of uranium [3.29]. The primary ore body formed when uranium was precipitated after a sequence of adsorption and redox reactions in a series of uraninite (UO_2) lenses, adjacent to a graphitic unit that forms a reducing and confining layer (Fig. 3.4). There is evidence of in situ oxidation and alteration to uranyl silicate minerals on the upgradient side of the primary ore body. The conversion of the relatively insoluble U^{4+} to the more mobile U^{6+} form led to movement of uranium away from the primary ore body and transported oxidized U(VI) in the groundwater in the form of uranyl carbonate complexes. Phosphate is present in many groundwater samples and some geological layers contain abundant chlorite, a magnesium-containing mineral of approximate composition $Mg_5Al_2Si_3O_{10}(OH)_8$, which produces dissolved magnesium and silica through weathering reactions [3.10]. The re-deposition of uranium from the groundwater in the surrounding rocks has led to the formation of a secondary ore body that contains U(VI) phosphate minerals such as saleeite ($Mg(UO_2PO_4)_2 \cdot 10H_2O$) and metatorbernite ($Cu(UO_2)_2(PO_4)_2 \cdot (H_2O)_8$). The strong scavenging of dissolved UO_2^{2+}, copper and phosphate by abundant iron oxides has favoured the formation of the metatorbernite at Koongarra [3.30]. Farther away from the ore body, dissolved uranium concentrations are lower, and adsorption of U(VI) on iron oxides and clays appears to play a major role in reducing uranium mobility. This natural system displays many of the processes that are summarized in diagrammatic form in Fig. 3.3.

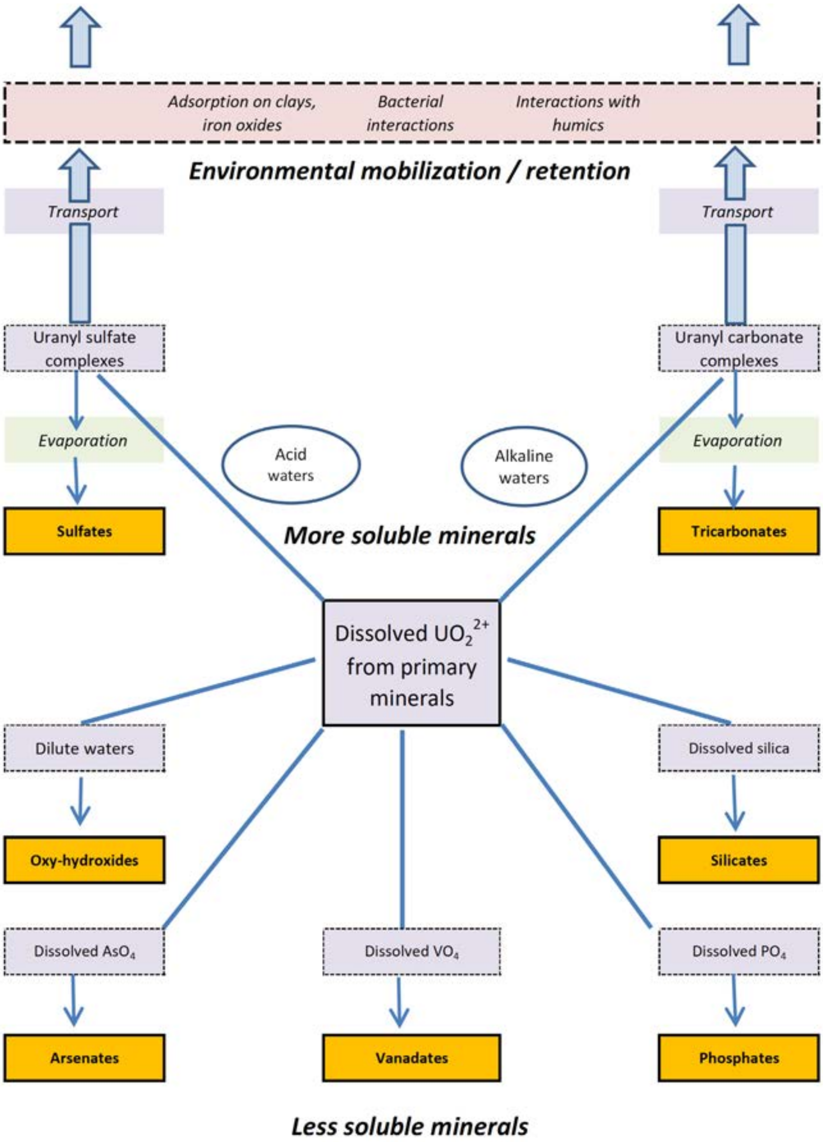

FIG. 3.3. Schematic representation of the formation of several types of uranyl mineral (shown in orange) from dissolved UO_2^{2+} and of the processes leading to environmental mobility of uranium (at the top of the figure). Lines indicate interactions with dissolved species. Downward arrows indicate precipitation.

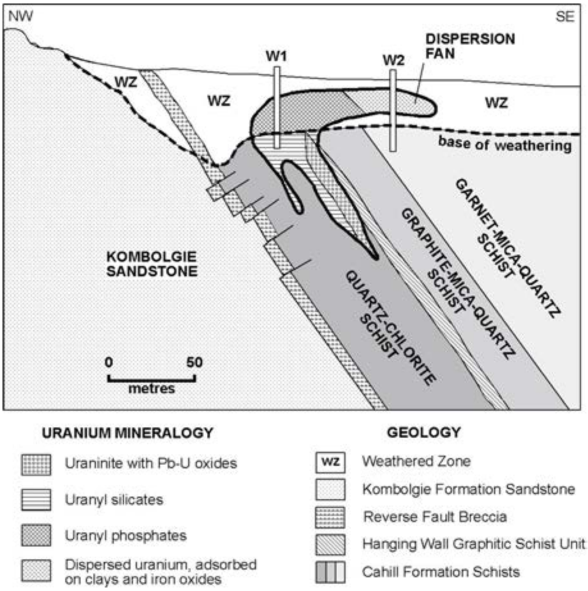

FIG. 3.4. Cross-section of the Koongarra deposit in northern Australia. Both U(IV) and U(VI) minerals are present, as well as a region of dispersed uranium adsorbed on clays on iron oxides, known as the 'dispersion fan' [3.29]. Borehole W1 penetrates the secondary ore zone (rich in uranyl phosphates) as well as the deeper uranyl silicate zone. Borehole W2 intersects the dispersion fan, where uranium is predominantly present in adsorbed forms (reprinted from Ref. [3.29], Copyright (2006), with permission from Elsevier).

3.3. ENVIRONMENTAL CHEMISTRY

3.3.1. Aqueous speciation and oxidation state

Uranium in shallow or surface natural waters is typically in the oxidized form (U^{6+}) and its aquatic chemistry involves the soluble uranyl ion (UO_2^{2+}) and its aqueous complexes. This linear dioxo-cation can be represented as $[O=U=O]^{2+}$. Complexes are aqueous species in which the uranyl ion is combined with other dissolved ligands that may also be present in the water (e.g. $UO_2(CO_3)_3^{4-}$ is one of several uranyl carbonate complexes). As with other actinides, UO_2^{2+} generally forms strong complexes with oxygen-containing ligands (oxides, hydroxides, phosphates and carbonates). Uranium speciation is also affected by inorganic ligands, such as fluoride; organic ligands, such as citrate; and complex naturally occurring organic ligands, including humic and fulvic materials.

Complexation reactions are extremely important from a geochemical viewpoint for the following reasons [3.2, 3.31]:

- Complexation of a dissolved species tends to increase its solubility;
- Some elements are predominantly present in solution as complexes rather than free ions;
- Adsorption may be dramatically increased or decreased by complexation;
- The toxicity and bioavailability of metals depend on their aqueous speciation or complexation.

At low pH values, uranium exists in solution, predominantly as UO_2^{2+}, but in pure water, uranyl usually forms a series of hydrolysis species at higher pH values such as UO_2OH^+, $(UO_2)_3(OH)_5^+$ and $UO_2(OH)_2^0$. Uranyl has a strong tendency to form carbonate complexes [3.32], particularly with increasing pH, and carbonate is commonly present in soil and groundwater environments. Thus, in the pH range of most soils, U^{6+} often forms soluble uranyl hydroxyl carbonate complexes, including $(UO_2)_2CO_3(OH)_3^-$, $UO_2CO_3^0$, $UO_2(CO_3)_2^{2-}$ and $UO_2(CO_3)_3^{4-}$ (Fig. 3.5). In recent years, it has been established that in some natural waters, particularly those containing a large number of Ca^{2+} ions, the mixed $Ca_2UO_2(CO_3)_3$ species may be dominant [3.33]. This complex is believed to be the dominant uranium-containing species in sea water [3.34], which is significant because it is a neutral species (in contrast to the series of negatively charged complexes which dominate at high pH in calcium free conditions).

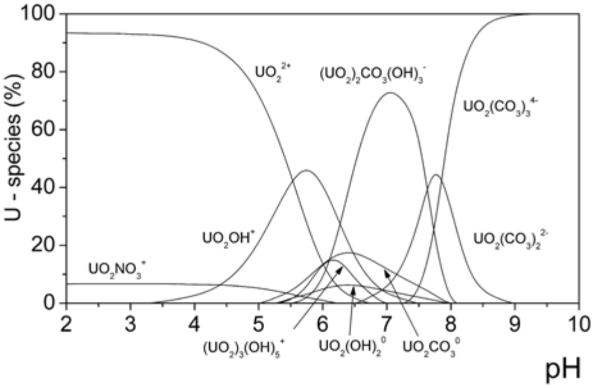

FIG. 3.5. Speciation of uranium (total uranium concentration of 1 μmol/L in 0.1M sodium nitrate) as a function of pH, when equilibrated with atmospheric air containing carbon dioxide. The importance of negatively charged uranyl carbonate species at high pH values should be noted.

Carbonate is, in general, the most significant uranium ligand in natural waters, and the greater solubility of the UO_2^{2+} ion is partly due to its tendency to form carbonate complexes that tend to be highly stable in solution. The environmental mobility of these complexes is, in part, determined by their negative charge (Fig. 3.5), which decreases their tendency to bind to the surfaces of negatively charged clay minerals. Thus, uranium can have significant mobility, particularly in systems with high pH and in the presence of high partial pressures of CO_2. Carbonate tends to strongly decrease the affinity of uranium for the surfaces of solid phases, such as iron oxides, in the environment [3.32, 3.35].

Organic complexes may also be important for uranium aqueous chemistry, affecting its solubility and mobility in soil. For example, when citrate is present, there may be a wide pH range across which the negatively charged uranyl citrate complex is dominant [3.2]. Dissolved humic substances can be strong complexing agents for many trace metals in the environment, forming stable complexes with radionuclides. It has been found that the presence of humic acid on iron oxide surfaces can increase the uptake of uranium by the solid phase, particularly at acidic pH values [3.36, 3.37], although the effect can be reversed at higher pH values. In general, there are many examples of organic complexants increasing the mobility of uranium.

The preceding discussion has mainly focused on the chemistry of uranium in the near surface environment, where the U^{6+} oxidation state often dominates. In less oxidizing conditions, the reduced U^{4+} oxidation state becomes more dominant. In general, the mobility of uranium is largely determined by its redox state. This behaviour can be summarized in a diagram known as a Pourbaix or Eh–pH diagram (Fig. 3.6).

This depicts the dominant forms of uranium as a function of the acidity (pH) and redox status (Eh or pE). The acidity depicted in Fig. 3.6 ranges from 0 to 12 but most natural waters are in the pH range between about 4 and 9. It can also be seen that in the upper part of Fig. 3.6 (more oxidizing conditions) the uranyl carbonate complexes dominate. However, in the lower part of the figure (more reducing conditions), reduced forms of uranium are dominant. The stability field of crystalline UO_2 is also indicated in Fig. 3.6. In this region, it would be expected that the solid phase would be stable and that levels of dissolved uranium would be very low. In general, reduced uranium species (i.e. U^{4+}) would be expected to be dominant at low Eh values and, under these conditions, the solubility of uranium is much lower.

Whereas uranium in natural fresh waters usually occurs as U^{6+} species, dissolved U^{4+} can be present in reducing groundwaters. As has been discussed, U^{4+} is the main oxidation state of uranium in primary ore minerals (such as pitchblende) and, owing to the low solubility of these minerals, the uranium concentrations in such groundwaters are typically below 10^{-8}M (or ~2 ppb) [3.31].

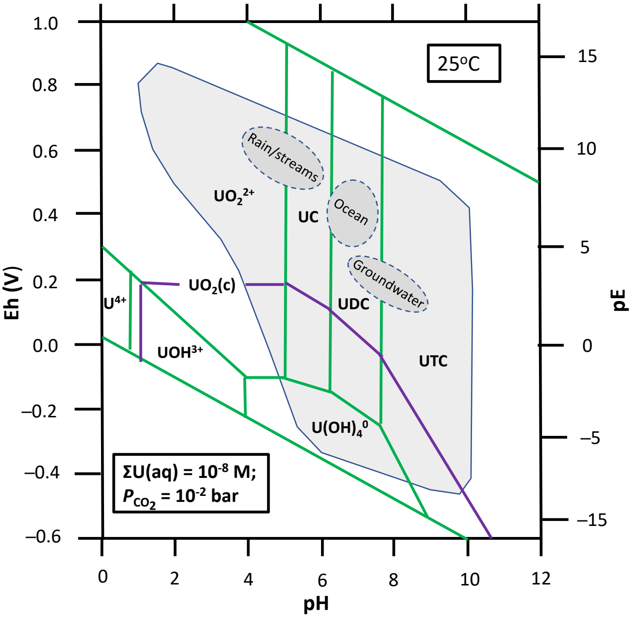

FIG. 3.6. Eh–pH diagram for aqueous species in the U–O_2–CO_2–H_2O system in pure water at 25°C and a total pressure of 100 kPa for a total number of U moles per litre of solution of 10^{-8}M and a CO_2 groundwater pressure of 1 kPa [3.31, 3.38, 3.39]. UC, UDC and UTC represent the aqueous complexes $UO_2CO_3^0$, $UO_2(CO_3)_2^{2-}$ and $UO_2(CO_3)_3^{4-}$, respectively. The grey area represents a range of conditions found in natural waters (adapted from Ref. [3.39]).

Reduced uranium has a strong tendency to hydrolyse, easily forming colloids, especially when environmental conditions change. In most environmental systems, it is likely that the reduced uranium species will bind to carrier colloids that primarily consist of other components such as silicic acid, colloidal oxides and clay particles [3.40].

The primary abiotic and biological processes that transform uranium in soil involve redox reactions that convert U^{6+} to U^{4+} and vice versa. There is a more detailed discussion of abiotic and biological processes that can transform uranium in the environment in Section 3.4.

3.3.2. Solubility and precipitation

The solubility of minerals generally puts an upper limit on the concentration of dissolved species in solution. The saturation index (SI) of a mineral in an aqueous system gives an indication of the potential for it to dissolve or precipitate. SI is defined as follows:

$$\mathrm{SI} = \lg(\mathrm{IAP}/K) \tag{3.2}$$

where IAP is the ion activity product for a specific water sample and K is the solubility product for a particular mineral. For example, for a natural water containing Ca^{2+} and SO_4^{2-} ions, the IAP for gypsum ($CaSO_4$) is calculated as the product of the ion activities in the water sample as follows:

$$\mathrm{IAP}_{\mathrm{gypsum}} = [\mathrm{Ca}^{2+}] \cdot [\mathrm{SO}_4^{2-}] \tag{3.3}$$

Here, the square brackets denote chemical activities, which are calculated from measured ion concentrations by applying a correction factor known as the activity coefficient [3.41]. In dilute solutions, this correction factor is close to unity. In such cases, it can be considered that, as a first approximation, the IAP is the product of the concentrations of the dissolved ions.

If $SI = 0$, the mineral is at saturation in the water sample. Values of SI above or below zero indicate a tendency to precipitate or dissolve, respectively [3.31]. Although the supersaturation of a system with a particular phase ($SI > 0$) does not guarantee that it will precipitate or control solubility, it gives a useful indication of the likely direction of system evolution. In practice, the full computation of the chemical state of a specific groundwater requires a complex iterative calculation in which the formation of all likely solid phases is considered. In general, the associated computations are complex and require the application of a specialist computer code (e.g. PHREEQC [3.42]).

The solubility product, K, has been measured for numerous uranium minerals, and relevant data can be found in various sources (e.g. Ref. [3.25]). Although these compilations provide useful indications of the solubility of uranium minerals, the data are not necessarily relevant to the real behaviour of trace uranium, because they focus on the solubility of well defined crystalline phases. The amounts of uranium in many environmental systems are often not sufficient to induce precipitation. Furthermore, many of the phases found in nature contain impurities that also affect their thermodynamic properties. Another complexity may derive from the fact that amorphous phases (such as freshly formed precipitates) can have significantly greater solubilities than pure mineral phases. These amorphous minerals form rapidly but eventually transform towards more stable crystalline phases. In the early stages of the process, the system can be supersaturated with respect to the more stable minerals, but they may not play a role in limiting the aqueous uranium concentration.

A related process that influences uranium transport and bioavailability in near surface systems is the co-precipitation of uranium with oxide minerals or other naturally occurring mineral phases. This process can occur in uranium contaminated soils and subsurface materials. Although uranium is not thought to be incorporated into the iron oxide structure because of differences in ionic radius, many field and experimental studies have reported the uptake of uranium during the formation of crystalline and amorphous iron oxides [3.30, 3.43–3.45]. The formation and dissolution of iron minerals can occur cyclically as redox conditions vary. The sorption of uranium on these newly formed minerals and incorporation of uranium into their iron oxide structures may occur during such cycles. The co-precipitation of uranium with other minerals can be considered as an intermediate process between surface adsorption and precipitation. However, a key distinction is that co-precipitation can remove uranium from solution when its concentration is lower than would be required to induce precipitation. As such, it is an important environmental process. In addition, it can be exploited as a method for concentrating radionuclides from natural waters for isotopic analysis [3.46].

It has proved extremely useful to study the formation of uranium-containing minerals in sites into which large amounts of uranium have been released, such as contaminated sediments obtained from the Hanford site, USA [3.47]. Uranium in Hanford sediments was found to be present as discrete precipitates within the interiors of sediment grains, particularly within microfractures of variable width and length. The precipitates were small crystals (generally, <3 μm in diameter), occurring as uranyl silicate microprecipitates in fractures, cleavages and cavities within sediment grains. Uranium dissolution was studied in various experiments. It increased with increasing carbonate concentration and decreased with time as solubility equilibrium was attained. The experimental uranium dissolution data were considered to be consistent with the presence of Na-boltwoodite ($Na[UO_2(SiO_3OH)](H_2O)_{1.5}$) and/or uranophane ($Ca[UO_2(SiO_3OH)]_2(H_2O)_5$), which exhibited similar solubility under the experimental conditions.

3.3.3. Adsorption and desorption

As noted in the previous section, the solubility of uranium minerals indicates the likelihood of them forming by precipitation from a given solution. However, in many environmental systems, the concentration of dissolved radionuclide is maintained at much lower values owing to the phenomenon of surface adsorption, which is an interaction with mineral surfaces. These interactions of dissolved uranium with the surfaces of naturally occurring substances can include processes such as adsorption on iron oxide surfaces, ion exchange in the interlayers of clay minerals or binding by humic substances. Adsorption is the uptake of a chemical species onto the surface of a mineral, with the corresponding release of a species from the surface into the aqueous phase being known as desorption. Together these processes are described as 'sorption'.

The aqueous concentration of many trace elements is controlled by sorption processes, which usually result in the dissolved concentration of a radionuclide being much lower than would be expected based on solubility alone. Adsorption of radionuclides onto mineral surfaces can be quantified and modelled in a variety of ways. A simple, widely used approach is to compute the experimental distribution coefficient (K_d value). K_d is the ratio of the radionuclide concentration adsorbed by the solid divided by the concentration in the liquid. Thus, a high K_d value indicates a strong retention on the surface, implying a significant retardation of migration. The K_d parameter enables comparison of experimental datasets obtained under various experimental conditions, which has led to several compilations and discussions of K_d values [3.48, 3.49]. Many more complex models of sorption have been developed, such as the Langmuir and Freundlich isotherms, and various types of surface complexation model [3.50]. However, the simpler approaches, such as use of a K_d value, remain in widespread use in environmental radionuclide transport modelling.

The adsorption of uranium is strongly dependent on the aqueous chemistry, including the pH, ionic strength, presence of complexing ligands such as carbonate, and the presence of organic matter such as humic acids. Typically, the effect of the presence of natural organic matter is to enhance uranium uptake on solids at low pH values and slightly reduce it at more alkaline pH values [3.37, 3.51].

Figure 3.7 shows some of the significant characteristics of K_d for uranium under some specific experimental conditions. There is a vast range of K_d values ranging over several orders of magnitude, with a strong pH dependence exhibited in all datasets. It can be observed in Fig. 3.7(a) that there are significantly different K_d values for different minerals. The effect of the presence of various ligands is demonstrated in Fig. 3.7(b). In this example, the diverse effects of the different ligands are shown, with humic acid and phosphate strongly increasing uranium K_d values across a wide pH range, whereas citrate significantly reduces uranium sorption. Interactions in these systems are complex, with many possible solution phase and sorption reactions occurring.

3.4. MICROBIAL BIOGEOCHEMISTRY

It has been recognized in recent decades that microorganisms play an important role in the biogeochemical cycling of metals, including uranium, in the environment [3.39, 3.52–3.60]. Metal microbe redox interactions are particularly

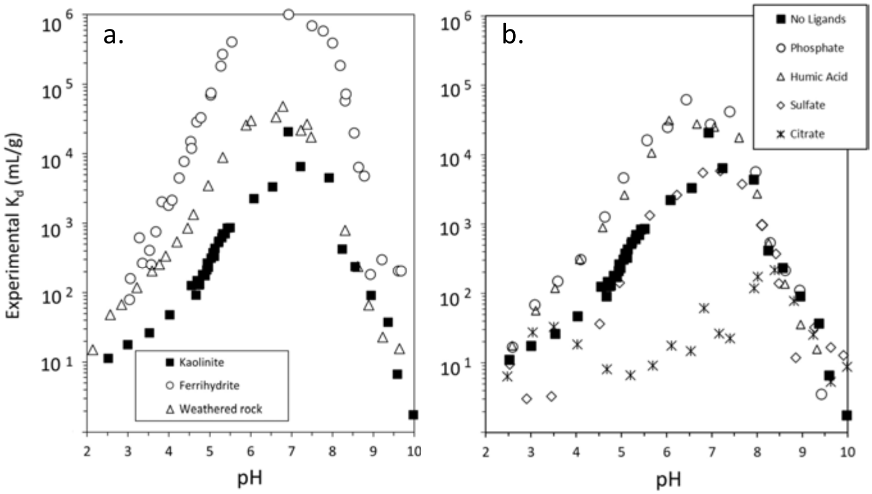

FIG. 3.7. Influence of mineralogy and complexing ligands on uranium sorption. (a) Uranium K_d *values on various minerals: iron oxide (ferrihydrite), clay (kaolinite) and natural soil material (weathered rock) under similar conditions. (b) Uranium* K_d *values on kaolinite in the presence of phosphate (100 µmol/L), humic acid (9 mg/L), sulphate (0.01 mol/L) and citrate (100 µmol/L). Experimental conditions: total uranium of 1 µmol/L in 0.1M* $NaNO_3$ *equilibrated with air (adapted from Ref. [3.2] with permission courtesy of John Wiley & Sons).*

significant when considering ore formation and recovery of metals from ores (e.g. Ref. [3.61]). Certain aerobic organisms can catalyse the oxidation of reduced metals in bioleaching processes (e.g. Ref. [3.62]). Conversely, anaerobic organisms can catalyse metal reduction, often to a less mobile species. This often occurs in anaerobic sediments [3.57].

As already discussed, the mobility of uranium depends on its speciation and redox state (Fig. 3.6), with reduced uranium, U(IV), being very insoluble and commonly present in uranium-containing ores. Oxidized uranium, U(VI), is relatively soluble and is, therefore, more mobile in the environment. Biogeochemical processes and interactions play a crucial role in controlling uranium speciation and mobility through direct microbial metabolic processes, such as respiration, or indirectly by microbial alteration of redox and pH in some specific environments (e.g. Refs [3.60, 3.63, 3.64]). These processes, once understood, can be manipulated to, for example, accelerate bioleaching or to remediate environments contaminated with uranium.

3.4.1. Aerobic microbial interactions with uranium

Microbially enhanced oxidation of U(IV) to U(VI) is often used to recover uranium, especially from low grade ores (e.g. Refs [3.57, 3.62, 3.65, 3.66]). Low grade ores (0.04–0.4% uranium) are often chemically extracted using an aerobic, acid leaching process that is enhanced by the presence of Fe(II) and S(0) oxidizing bacteria [3.57]. Iron(III) acts as an oxidant for U(IV) and, under acidic conditions, solubilizes uranium as U(VI) in the leachate. Iron(III) is reduced during uranium oxidation but can be oxidized by iron oxidizing bacteria such as the acidophile *Acidithiobacillus* (*Thiobacillus*) *ferrooxidans* [3.62, 3.65]. Uranium ore often contains pyrite that can also be acted on by acid tolerant Fe(II) and sulphide oxidizing bacteria, again generating Fe(III) and also sulphuric acid.

Metal oxidizing bacteria can also oxidize U(IV) to U(VI), directly deriving energy for metabolic activity [3.57]. However, U(VI) is toxic to many organisms, inhibiting Fe(II) oxidation in unadapted *Acidithiobacillus* spp. Nevertheless, cultures can be adapted to tolerate high U(VI) concentrations [3.65, 3.67].

Metal and sulphur oxidizing bacteria are ubiquitous in the environment, particularly where reduced minerals are in contact with atmospheric oxygen. Consequently, their action on uranium-containing ores in surface environments will result in uranium mobilization and, potentially, contamination of surface waters and groundwaters. This is of concern in areas where past uranium operations have left tailings where low level leaching of uranium within the piles provides a continuing source of U(VI) to aquifers (e.g. Refs [3.68, 3.69]).

3.4.2. Anaerobic microbial interactions with uranium

Decades of nuclear activities related to uranium mining, processing and use have left a legacy of environmental contamination in many countries. Anaerobic microbial processes have the potential to remove U(VI) from contaminated groundwater by precipitating aqueous uranium into insoluble minerals in situ. Reference [3.39] suggests the following four main mechanisms for microbe–uranium interactions, which could be used for such remediation:

(1) Bioreduction: this involves stimulating an environment (e.g. by introducing an electron donor for microbial use) to promote uranium reduction (see Refs [3.52, 3.57, 3.70]). A wide range of organisms can reduce uranium and much work is currently being undertaken to understand the microbial mechanisms and processes involved (see Refs [3.39, 3.60, 3.71]).

(2) Biomineralization: uranium is precipitated with microbially generated ligands such as sulphide and phosphate, or as hydroxides or carbonates on cell surfaces under alkaline conditions (see Refs [3.72, 3.73]).

(3) Bioaccumulation: in this process, uranium is actively taken up by microbial cells (see Ref. [3.63]). However, uranium has no known biological function and there is little evidence currently that this would be a viable technique [3.74].

(4) Biosorption: this is the passive uptake of uranium onto living or dead cell surfaces. This is a complex process that has not yet been fully evaluated [3.75].

3.4.3. Field studies of bioreduction and biomineralization

In situ uranium bioreduction has been demonstrated successfully at various sites, including the United States Department of Energy sites at Oak Ridge, Hanford and Rifle as well as at Sellafield in the United Kingdom [3.76]. However, the potential for the reoxidation of U(IV) needs to be carefully considered if bioreduction is to be a viable strategy for long term remediation of any site. Factors to be considered include the following:

- Overall stability of the environment, especially with regard to exposure to oxygen and/or nitrate;
- Presence and supply of suitable electron donors;
- Competition for use of electron donors;
- Microbial population composition;
- Stability of minerals formed;
- Radiotoxicity of the environment.

Consequently, any contaminated site needs to be carefully characterized and evaluated for its suitability prior to the start of a bioreduction programme.

Uranium phosphate mineralization, involving uranium sequestration as insoluble uranyl U(VI) phosphate biominerals, has been investigated for sites where bioreduction may not be appropriate. Considerable laboratory work has been undertaken to understand the microbiological processes involved [3.72, 3.77–3.79]. The technique has been investigated for potential use at the Oak Ridge site, where bioreduction may be inhibited by the high nitrate concentrations. The technique appears promising although the limited commercial availability of some proposed carbon and phosphorus sources may limit its use [3.39].

3.5. MEASUREMENT OF URANIUM

3.5.1. Survey of techniques in common usage

Selecting appropriate analytical methods is a critical part of environmental analysis and can involve a complex series of decisions. Methods must have sufficiently low detection levels, suitable precision and analyte recovery, and acceptable selectivity for the specific purpose, and must meet minimum criteria for data quality objectives (see also Section 3.5.3). To study the distribution of uranium in the environment, samples of water, such as drinking water, fresh water and wastewater, are often required to be analysed. For non-aqueous samples, such as soil, tailings and vegetation, the first step is often to bring the sample into solution by an appropriate dissolution process. Therefore, reference techniques that involve analysis of aqueous samples are particularly useful in environmental research.

Several techniques for uranium determination in water samples have found widespread acceptance in the scientific community and are considered to be reference techniques. In many cases, these techniques have also been approved by regulatory agencies. For example, the EPA has made available a list of analytical methods that have been approved for drinking water compliance of radionuclides [3.80].

A useful source of available methods is the National Environment Methods Index (NEMI) database[7]. This is a joint initiative of three United States Government agencies, including the EPA. The NEMI is a searchable database of environmental methods, protocols, and statistical and analytical methods that enables users to find and compare methods for environmental monitoring applications. Since its release, approximately ten years ago, the NEMI has been updated several times, reflecting improvements in technology and developments in analytical methods.

Table 3.3 provides a summary and categorization of the techniques available in the NEMI database. The most common technique is inductively coupled plasma mass spectrometry (ICP-MS), for which several standard methods have been developed. In general, the NEMI recommends widely used techniques for common situations rather than techniques appropriate to research applications. For example, the NEMI does not list ICP-MS for isotopic measurements of uranium because ^{234}U is not as readily measured by this technique as are the longer lived isotopes that are more abundant by mass (^{235}U and ^{238}U). Thus, most of the ICP-MS methods listed in Table 3.3 are considered as suitable for measuring ^{238}U or total uranium, rather than individual isotopes. Of the

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[7] See www.nemi.gov/home

techniques given in Table 3.3, alpha spectrometry is the method of choice when information on individual isotopes is required. Several techniques employing alpha scintillation are listed for determining total uranium alpha activity. Finally, the technique of kinetic phosphorescence analysis (KPA) provides an alternative that is particularly suited for routine analysis but, unlike ICP-MS, can only be used for uranium.

Several analytical techniques were mentioned in a survey of the Canadian literature of the past several decades [3.27]. In addition to those techniques in popular current usage mentioned in Table 3.3, the list included methods such as laser induced fluorescence, fluorometry (after sample fusion), spectrophotometry (with arsenazo III) and reactor based neutron activation analysis (NAA). The survey of techniques included some older techniques that are falling out of favour, as well as some more advanced techniques requiring facilities such as nuclear reactors (e.g. NAA). While such techniques may not be available to a widespread community (or endorsed for routine usage by organizations such as the EPA), they have applications in research and specialized contexts. For example, the NAA technique has been routinely used for assaying uranium ores. Similarly, accelerator mass spectrometry (AMS) has been applied for identifying traces of nuclear materials in the environment.

While the techniques listed in Table 3.3 do not comprise a comprehensive list of specialized techniques for measuring uranium in all types of environmental sample, they constitute a set of widely utilized and accepted techniques that are considered to be reliable and applicable to a range of situations. These techniques are discussed in more detail in Section 3.5.2.

3.5.2. Major analytical techniques for uranium determination

3.5.2.1. Inductively coupled plasma mass spectrometry and other mass spectrometry techniques

Various mass spectrometry techniques have been developed and utilized for measuring total uranium concentrations and, particularly, uranium isotopic ratios. A commonly available technique is ICP-MS, in which ions for the measurements are produced by an inductively coupled plasma, and a mass spectrometer is utilized for separating and detecting the ions. The key feature of ICP-MS and related mass spectrometry techniques is that they measure the mass of a nuclide present, rather than its radioactivity [3.81]. These techniques are, therefore, generally most useful for longer lived radionuclides, and most uranium isotopes of environmental interest have been successfully detected. The applications of ICP-MS for radionuclide measurements in environmental samples have been reviewed in Ref. [3.82].

TABLE 3.3. ANALYTICAL METHODS FOR MEASURING TOTAL URANIUM AND URANIUM ISOTOPE CONCENTRATIONS

Technique	Number of methods	Source	Method ID[a]	U_{tot}, U_{nat}	^{238}U	^{235}U	^{234}U	Total U_{alpha}	Other analytes[b]	EPA method[c]
Inductively coupled plasma mass spectrometry	6	EPA	200.8	Yes	Yes	n.a.[d]	n.a.[d]	n.a.[d]	Metals, trace elements	Yes
		EPA	200.10	Yes	Yes	n.a.[d]	n.a.[d]	n.a.[d]		n.a.[d]
		DOE	MM800	Yes	Yes	Yes	n.a.[d]	n.a.[d]		n.a.[d]
		ASTM[e]	D5673	Yes	Yes	n.a.[d]	n.a.[d]	n.a.[d]		Yes
		Standard methods[f]	3125	Yes	Yes	n.a.[d]	n.a.[d]	n.a.[d]		Yes
		USGS	I-4472-97	Yes	Yes	n.a.[d]	n.a.[d]	n.a.[d]		n.a.[d]
Alpha spectrometry	5	DOE	U-02	Yes	Yes	Yes	Yes	Yes	n.a.[d]	Yes
		DOE	Se-03	Yes	Yes	Yes	Yes	Yes	Am, Pu	n.a.[d]
		DOE	TP-A.20	Yes	Yes	Yes	Yes	Yes	Actinides	n.a.[d]

TABLE 3.3. ANALYTICAL METHODS FOR MEASURING TOTAL URANIUM AND URANIUM ISOTOPE CONCENTRATIONS (cont.)

Technique	Number of methods	Source	Method ID[a]	U_{tot}, U_{nat}	^{238}U	^{235}U	^{234}U	Total U_{alpha}	Other analytes[b]	EPA method[c]
		Standard methods[f]	7500-U C	Yes	Yes	Yes	Yes	Yes	n.a.[d]	Yes
		ASTM[e]	D3972	Yes	Yes	Yes	Yes	Yes	n.a.[d]	Yes
Alpha scintillation	3	ASTM[e]	D6239	n.a.[d]	n.a.[d]	n.a.[d]	n.a.[d]	Yes	n.a.[d]	Yes
		Standard methods[f]	7500-U B	n.a.[d]	n.a.[d]	n.a.[d]	n.a.[d]	Yes	n.a.[d]	Yes
		EPA	908.0	n.a.[d]	n.a.[d]	n.a.[d]	n.a.[d]	Yes	n.a.[d]	Yes
Kinetic phosphorescence analysis	1	ASTM[e]	D5174	Yes	n.a.[d]	n.a.[d]	n.a.[d]	n.a.[d]	n.a.[d]	Yes

TABLE 3.3. ANALYTICAL METHODS FOR MEASURING TOTAL URANIUM AND URANIUM ISOTOPE CONCENTRATIONS (cont.)

Technique	Number of methods	Source	Method ID[a]	U_{tot}, U_{nat}	^{238}U	^{235}U	^{234}U	Total U_{alpha}	Other analytes[b]	EPA method[c]

Note: The data in this table are taken from the National Environmental Methods Index (NEMI) database for uranium measurements in environmental water samples, but with methods approved by the United States Environmental Protection Agency (EPA) [3.80] for drinking waters also shown. ASTM: ASTM International; DOE: United States Department of Energy; USGS: United States Geological Survey.

[a] Unique method identifier. For more information, the reader is referred to the NEMI web site or source organization.

[b] Other analytes are only listed where they are analysed as part of the specific method referenced. Suitable variations on the techniques would permit the determination of a range of other analytes.

[c] Listed in Ref. [3.80].

[d] n.a.: not applicable.

[e] Standard technique of ASTM International.

[f] Standard Methods for the Examination of Water and Wastewater. See https://www.standardmethods.org/

Of the radionuclide measurements performed by ICP-MS, those of long lived actinides have been the most successful, with ^{238}U and ^{232}Th being by far the most common radioisotopes mentioned, representing over 60% of all published reports according to Ref. [3.82]. This can be explained by several factors, including the long half-life of these isotopes, their high elemental abundance in the environment and the low instrumental background typically associated with their measurement. Such interference as does occur can be caused by the generation of isobaric ions created by the plasma gas and/or aqueous solvent commonly used in ICP-MS analysis. According to Ref. [3.82], ^{234}U and ^{235}U are also discussed in a significant fraction of the research publications involving ICP-MS, although measurements of these isotopes are less prevalent than those of ^{238}U because of their lower isotopic abundances. Although mass spectrometry responds to the mass of the atom, this technique has not been routinely applied to measure ^{234}U in common environmental applications and can be usually considered a technique for measuring total uranium (the mass of uranium in environmental samples being dominated by the ^{238}U isotope).

There have been several variations and refinements of mass spectrometry based techniques, including thermal ionization mass spectrometry, AMS and selective laser ionization mass spectrometry. These methods are becoming increasingly applied for the determination of isotopic ratios. For example, the measurement of ^{236}U:^{238}U ratios by AMS can be a valuable forensic tool to determine the origin of uranium in environmental samples. The long lived uranium isotope ^{236}U is produced by ^{235}U neutron capture and builds up to elevated levels (0.5%) in nuclear fuel. It has been distributed in the environment because of nuclear activities, including nuclear explosions, accidents at nuclear plants, dumping of nuclear waste and releases from nuclear facilities. Thus, ^{236}U is a potentially useful tracer of irradiated uranium for nuclear safeguards or other applications [3.4].

ICP-MS usually requires a liquid sample and, therefore, solid samples must be digested and dissolved prior to sample analysis. A variant on ICP-MS measurement in a dissolved sample is laser ablation ICP-MS (LA-ICP-MS), in which a solid sample is sputtered by a laser beam and the released ions are counted in the ICP-MS. As it requires little or no sample preparation, LA-ICP-MS is advantageous for isotopic determination in solid samples and is useful to assess the spatial variation of uranium in a matrix. However, quantitative analysis is difficult because reference materials may differ from the sample matrix. Furthermore, solids are typically inhomogeneous, and the precision achieved by LA-ICP-MS is normally poorer than for analyses based on digested samples. Although it is the most sensitive of the solid state mass spectrometry methods, LA-ICP-MS is not nearly as sensitive as liquid sample introduction ICP-MS. It should be kept in mind that mass spectrometry often requires considerable operator skill to obtain reliable results. In conclusion, ICP-MS is a fast multi-element technique with high sensitivity for nuclides with a long half-life and has many advantages for the determination of actinides in the environment [3.83].

3.5.2.2. Alpha particle spectrometry

Alpha particle spectrometry has been used for more than 30 years to determine the natural composition of uranium isotopes and is well established and very reliable. This method requires a carefully prepared, thin, solid source, typically either a fine precipitate or an electrodeposited layer on a metal planchette (disc). Owing to the number of preparation steps, full recovery of the uranium from the sample cannot always be achieved. Therefore, a yield determinant (tracer) is added at the beginning of the radiochemical procedure. For uranium measurements, ^{232}U or ^{236}U tracers are often utilized. An example of a uranium spectrum with both ^{236}U and ^{232}U tracers is given in Fig. 3.8. Excellent resolution of the different isotopes can be seen. In contrast to mass spectrometry techniques, the ^{234}U and ^{238}U peaks are of similar intensity, and the spectrum gives a direct measure of the relative activity of the two isotopes.

The choice of tracer depends on several factors. The use of ^{232}U can lead to contamination of the detectors with ^{228}Th and ^{224}Ra (progeny of ^{232}U). To minimize this, a self-cleaning decontamination solution of the ^{232}U tracer containing barium sulphate can be used [3.2]. In this solution, all of the radioactive progeny of ^{232}U are continuously and automatically removed without

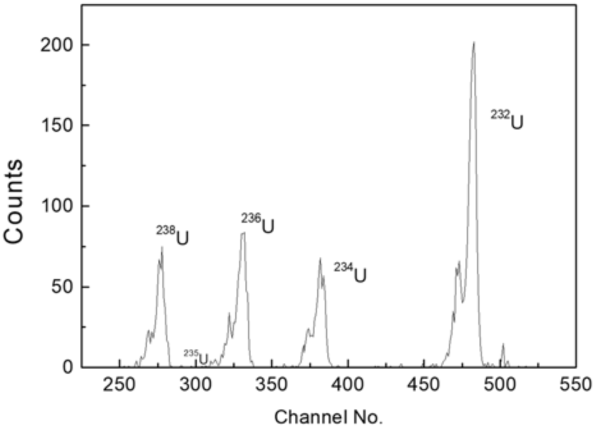

FIG. 3.8. An alpha spectrum showing natural uranium peaks (^{238}U and ^{234}U) as well as tracer peaks (^{236}U and ^{232}U). In this example, the ^{235}U peak is obscured by the peak of the ^{236}U tracer.

changing the ^{232}U concentration. A ^{236}U tracer may also be used for alpha particle spectrometry. This avoids the contamination of the detectors with progeny of ^{232}U. However, the ^{236}U peak overlies the ^{235}U peak (see Fig. 3.8), which means that a correction for the presence of the ^{235}U peak is required, based on an assumed natural isotopic content ratio of ^{235}U:^{238}U. Deviations from the ^{235}U:^{238}U isotope ratio in the sample cannot be detected. Furthermore, if the measurement of ^{236}U is of interest, then the ^{236}U tracer cannot be used.

Although alpha particle spectrometry can be carried out by several methods, only two are commonly used for environmental analyses: gridded ion chambers and semiconductor detectors. The ion grid chamber has a lower resolution but higher counting efficiency, which makes it useful for low activity samples. The ion grid chamber can accommodate sources up to 10 cm in diameter so that for low activity samples, the source thickness can be reduced by effectively spreading the activity over a large surface. Silicon surface barrier detectors have been most used for alpha particle spectrometry, mainly because of their excellent energy resolution, compact size, simple power requirement, low gamma ray sensitivities and their cost. Nowadays, ion implanted silicon detectors, showing even better characteristics, have supplanted surface barrier detectors and are commonly used in routine measurements. The major shortcomings of alpha particle spectrometry for uranium (as well as thorium isotopes) can be long counting times (days to weeks) and limited precision due to the counting statistics, in addition to the complexity and time required for the sample preparations.

3.5.2.3. Liquid scintillation counting

Liquid scintillation counting (LSC) has been widely used in the measurement of beta emitting radionuclides in water samples. The basic principle is that light formation is induced by the incoming particle and this is transformed into electric pulses. The liquid to be analysed is mixed with an LSC ‘cocktail’ consisting of a scintillation agent dissolved in an organic solvent. The scintillator molecules are excited by the transfer of kinetic energy from alpha or beta particles, which causes light photons to be emitted [3.84]. The principal disadvantage of LSC for alpha measurements is its poor resolution, around 200 keV, which is around ten times worse than that of alpha spectrometry using semiconductor detectors, which results in the loss of isotopic information. Therefore, LSC is mainly used for gross alpha (total alpha) measurements and in cases where high resolution is not required. This is reflected in the NEMI recommendations summarized in Table 3.3.

The determination of activities of uranium (as well as of other actinides — thorium, plutonium, americium and curium) at very low levels has been performed by employing advanced types of liquid scintillation system, such as the photon–electron rejecting alpha liquid scintillation (PERALS) system [3.85]. The principal advantage of this method compared with alpha spectrometry is that the preparation of the samples is much simpler. Procedures such as precipitations, evaporations, elutions (elemental separations) and filtrations are often not required. The limit of detection for many radionuclides is often lower than that obtained by other methods such as ICP-MS, time resolved laser induced spectrofluorometry and alpha spectrometry; thus, the PERALS system is promising for the determination of uranium at very low concentrations. However, its energy resolution is inferior to that obtained by alpha spectrometry. For some complex media, the extraction recoveries are not quantitative, and it is necessary to determine the recovery yields by labelling with a spike of ^{232}U [3.85].

The PERALS system has been applied to the analyses of a variety of non-routine environmental samples and has been found to be flexible and robust [3.83]. Compared with alpha spectrometry, it eliminates many of the complex chemical separation steps and does not require highly trained personnel. Another important advantage of the method is a better sensitivity (by about an order of magnitude) for the determination of low activities of the major alpha emitters compared with alpha spectrometry. The main drawback of the method is its poor energy resolution. In most real samples, a deconvolution of the spectrum is required to measure individual isotopes. This approach enables measurements of the overlapping peaks but increases the uncertainty of the measurements.

3.5.2.4. Kinetic phosphorescence analysis

KPA enables rapid determination of uranium concentrations to be made in aqueous samples with good sensitivity and accuracy [3.86], particularly for routine applications. In KPA, a laser pulse is used to electronically excite uranyl in the presence of a proprietary phosphate based complexing solution. KPA measures the time dependent decay of light intensity (lifetime: approximately 250 μs) and, by extrapolation to time zero (i.e. the end of the laser pulse), calculates the initial intensity, which is proportional to the uranium concentration. For good precision, several hundred laser pulses are used for each measurement and the results are averaged. The time resolved technique significantly reduces problems associated with interferences from other species and quenching effects. KPA has a detection limit of around 1 ng/L although the possible residual effects of matrix quenching and interferences should be considered and controlled for optimal results [3.87].

3.5.2.5. Other techniques

NAA is a technique that relies on the irradiation of samples in a nuclear reactor [3.88]. Irradiation results in the following capture reaction on ^{238}U:

$$^{238}\text{U}(\text{n}, \gamma)^{239}\text{U}\ (T_{1/2} = 23.5\ \text{min}) \rightarrow {}^{239}\text{Np}\ (T_{1/2} = 2.35\ \text{d}) \rightarrow \ldots \tag{3.4}$$

In principle, it is possible to quantify either ^{239}U or ^{239}Np [3.89], with the latter technique being originally proposed in Ref. [3.88] based on the longer half-life of ^{239}Np. The ^{239}U technique is, however, normally preferred because it is rapid and more sensitive [3.89]. In this procedure, the short lived radionuclide ^{239}U is separated from other activation product nuclides immediately following the irradiation and purified using solvent extraction (with tri-n-butylphosphate in toluene). The gamma peak of ^{239}U, at 74.7 keV, is used for the determination of the ^{238}U concentration. A uranium carrier is added to each sample prior to separation to evaluate chemical recovery using the 185.7 keV gamma emission of ^{235}U. Gamma ray spectrometry is generally performed in a high purity germanium well type detector.

3.5.3. Issues in technique selection for environmental samples

Several considerations arise prior to the commencement of analysis, including the following:

- Sample type: water, vegetation, soil and sludge.
- Objective of analysis: regulatory compliance, routine monitoring, research and process control.
- Type of information required: such as total uranium concentration, total alpha activity, full radiochemical analysis and other trace metals.
- Analytical questions: such as detection limit, accuracy and reproducibility.
- Practical considerations: available instruments, budget and speed of sample turnaround.

In an interesting comparison [3.90], laser photometry, LSC, gamma ray spectrometry and alpha particle spectrometry were used to estimate uranium concentrations in various water samples. The study concluded that laser photometry is a straightforward and accurate method capable of estimating low uranium concentrations but is not applicable for higher concentrations, difficult water matrices and isotope determinations. Alpha particle spectrometry provided precise isotope determination but with the disadvantage of difficult chemical preparation and long measurement times. An intermediate method is LSC, with the problem of isotopic concentration estimation due to poor energy resolution.

In many environmental situations, the uranium concentration (or isotopic activity) in water samples is a key consideration. Some types of water sample can be analysed with limited sample preparation in some techniques, but even where sample preparation is simple, interferences may need to be considered. However, some analytical techniques involve numerous separation and purification steps. For example, in alpha spectrometry, an extensive series of chemical separations and purification steps is required to provide a source suitable for analysis. Where the sample matrix comprises non-aqueous materials, such as soils, vegetation or rock, the components must usually be brought into solution by sample dissolution before most types of analysis are attempted. However, there are some types of analytical technique that can analyse solid samples (NAA, gamma ray spectrometry, although these might not provide adequate information for the required purpose.

The geochronology and geochemistry communities often seek to obtain high precision isotope ratios (^{234}U:^{238}U, ^{230}Th:^{234}U), whereas the health physics interest is often focused on concentrations or activities. In the case of nuclear forensic groups, the most important requirement is often high precision ^{235}U:^{238}U and ^{236}U:^{238}U ratios [3.81]. These objectives will strongly influence the choice of technique.

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Chapter 4

URANIUM DISTRIBUTION IN THE ENVIRONMENT

F.P. CARVALHO
University of Lisbon, Portugal

S. FESENKO
Russian Institute of Agricultural Radiology and AgroEcology, Russian Federation

N.G. MITCHELL
Eden Nuclear and Environment Ltd, United Kingdom

T.E. PAYNE
Australian Nuclear Science and Technology Organisation, Australia

M.C. THORNE
Mike Thorne and Associates Ltd, United Kingdom

T. YANKOVICH, A.R. HARBOTTLE
International Atomic Energy Agency

4.1. CYCLING OF URANIUM IN THE ENVIRONMENT

The purpose of this chapter is to provide an overview of the cycling and the occurrence of uranium in the environment. The data are presented specifically to demonstrate differences of orders of magnitude in uranium concentrations in different environments (and sample types) as well as the relative significance of various pathways. However, the discussions provided related to the data do not give details about the mechanisms and pathways involved. Environmental media and pathways are discussed, including further data on transfer parameters, in Chapter 6.

In this publication, both radiological and chemical impacts of uranium are considered. This chapter provides data for uranium both in mass concentrations (ppm or mg/g, or sometimes mg/kg) and activity concentrations (Bq/kg). The specific activity value 12.3 Bq/mg was applied to mass concentration data of natural uranium to obtain the corresponding ^{238}U activity concentration values [4.1].

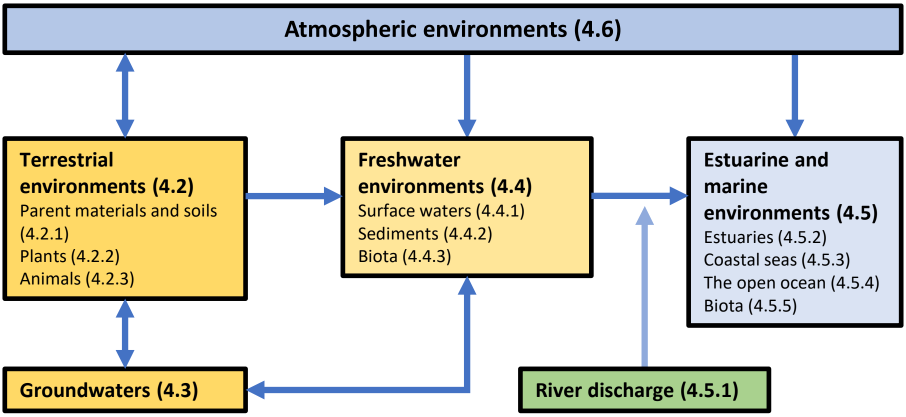

FIG. 4.1. Major components of the environment discussed in the sections of this chapter (indicated in parentheses).

In broad terms, the environment is considered in terms of its atmospheric, terrestrial, freshwater, estuarine and marine components. In addition, consideration is given to groundwaters, since uranium transport in groundwaters can impact other components of the environment, and specific consideration is given to the uptake of uranium in the biota present in each of the components of the environment. The sections of this chapter in which these components of the environment are addressed are illustrated in Fig. 4.1.

Naturally occurring uranium is ubiquitously present in the environment at a wide range of concentrations. Typical concentrations in different types of environmental media and in biota are given in the following sections. In addition, uranium concentrations in the environment and the bioavailability of uranium have sometimes been altered by human activities (e.g. mining and milling ores containing uranium). Therefore, specific attention is also given to contexts in which uranium concentrations and/or bioavailability have been modified by human activities.

4.2. URANIUM IN TERRESTRIAL ENVIRONMENTS

At the largest spatial and temporal scales, the general movement of uranium in the environment is from terrestrial uplands to the surface drainage network, then downstream to the oceans. Movement is both by particle transport and in dissolved form, with the uranium liberated for transport by erosive processes. The oceans act as an accumulating sink for uranium, but this is eventually

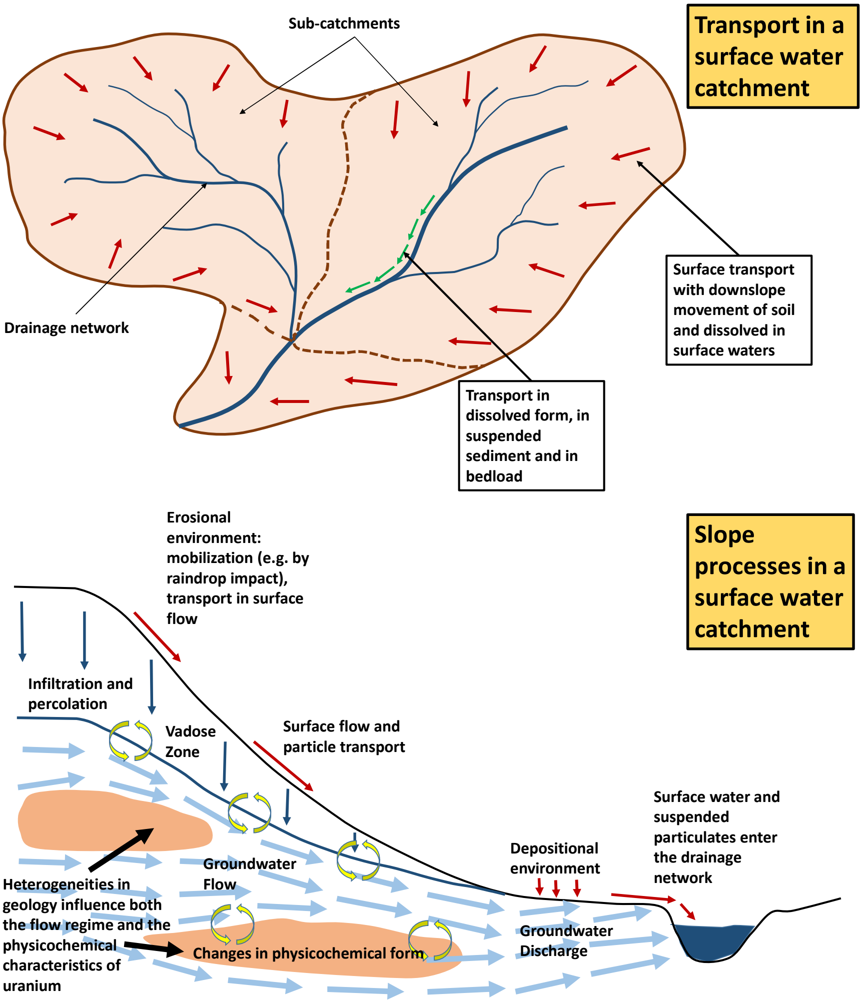

FIG. 4.2. *Processes that transport uranium in a surface water catchment.*

incorporated in bottom sediments and recycled to the mantle by subduction processes [4.2]. However, in the context of this review, the local transport of uranium from natural or anthropogenic sources is of greater relevance, since it is this local transport that must be considered when assessing potential impacts on human health and on the surrounding environment.

The transport of uranium in terrestrial environments is shown schematically in Fig. 4.2. This illustrates the processes operating at the scale of a surface water catchment. The top panel of Fig. 4.2 shows the catchment in plan view and the lower panel shows an illustrative vertical cross-section of a hill slope associated with one of the streams of the surface drainage network. At the catchment scale, particle transport is generally downslope from the interfluves (ridges) towards the stream channels. Once particulate material enters the streams, it is carried downstream either as suspended sediment or with a generalized movement of bed sediments (bedload transport), which includes siltation. However, uranium can also be carried downslope dissolved in water. This transport may be in surface flows but can also be in subsurface flow in the unsaturated or vadose zone (interflow) or in groundwater flows at and below the water table.

Uranium can be present as a variety of chemical species under different environmental conditions and these species exhibit different affinities for attaching to solid materials. Thus, transport of the element is affected by biogeochemical conditions, and spatial variations in those conditions can lead to accumulation of uranium, for example in regions over which the redox potential (Eh) changes markedly, as is the case in the capillary fringe at the water table or where deposits of organic materials are embedded in primarily mineral deposits. Temporal changes in physicochemical conditions, for example, at different times of the year can result in seasonal changes in uranium speciation as well.

Where uranium mining operations are ongoing or have occurred, a wide variety of sources of uranium may be imposed on the natural environment. These include engineered facilities, open cast pits, shafts and adits, waste heaps and tailings ponds [4.3, 4.4]. Such features can give rise to groundwater plumes of uranium, downslope movement of particulate material, ranging from fine-grained sediment to coarse aggregate, resulting in debris fans or talus deposits. In addition, where uranium enters the surface water drainage network, infiltration into the groundwater table, stream banks and overbank flooding events can give rise to contamination of the adjacent floodplain (Fig. 4.3).

4.2.1. Parent materials and soils

Isotopes of uranium are widely distributed in the Earth's crust. Uranium mass concentrations in parent rocks vary from 0.1 to 1250 ppm [4.5], with a typical range of 2.0–6.1 ppm or activity concentrations of 25–75 Bq/kg ^{238}U [4.6] (Table 4.1). The estimated average ^{238}U activity concentration in the continental crust is 32.9 Bq/kg [4.7].

Relatively high uranium concentrations are observed in black shale and phosphate rocks. The high uranium concentrations in shale are likely due to associations of clay rich material of organic origin, whereas phosphate rocks of sedimentary origin are composed of minerals rich in uranium. Silicic rocks, granites and basalts tend to have similar uranium concentrations. Concentrations

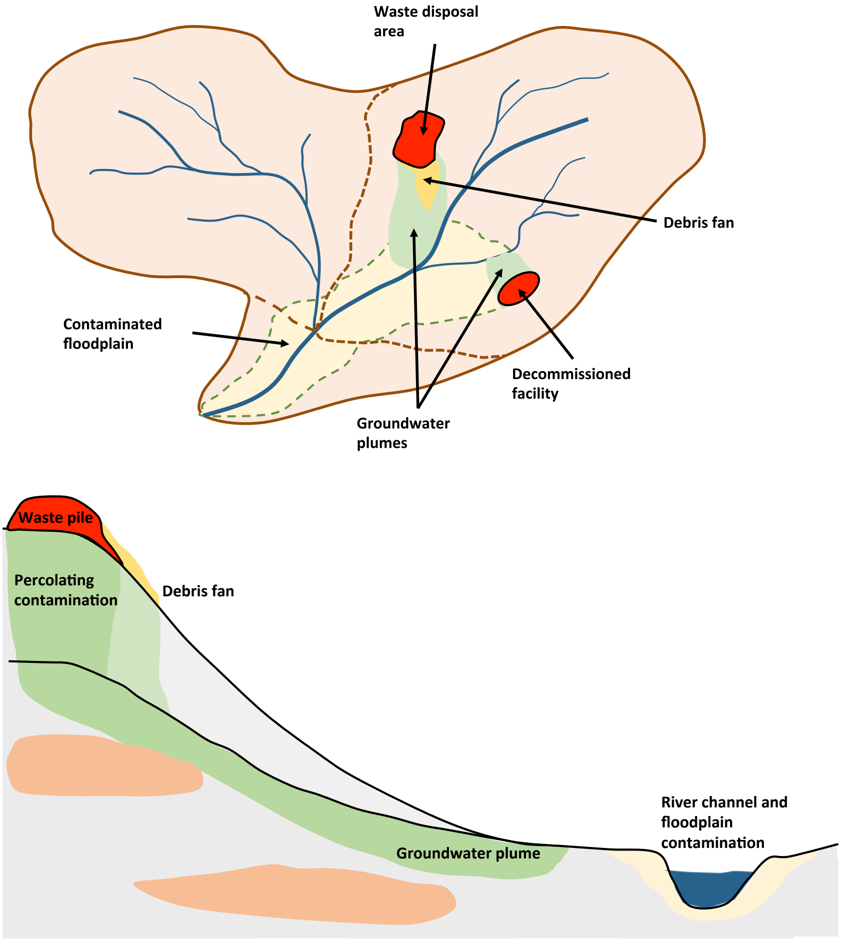

FIG. 4.3. Dispersal of uranium from mining activities.

of uranium in the soil depend, to a large extent, on its concentration in the parent rock. Parent rocks are affected by many environmental factors, resulting in soil formation. In particular, because of uranium weathering from the parent rocks, uranium can be transferred and deposited as loess, silt placers and tertiary soils (Table 4.2 [4.8–4.13]).

Text cont. on p. 79.

TABLE 4.1. TYPICAL URANIUM CONCENTRATION IN DIFFERENT ROCK TYPES

Rock type	Uranium mass concentration (mg/kg)	^{238}U activity concentration (Bq/kg)
Silicic (granite–dacite)	2.2–6.1	27–75
Basalt	0.1–1	1–12
Gneiss	2	25
Schist	2–5	25–62
Limestone	2	25
Black shale	3–1 250	37–15 000
Phosphat	50–300	620–3 700

Source: Adapted from Ref. [4.5].

TABLE 4.2. NATURAL URANIUM CONCENTRATION IN DIFFERENT SOIL TYPES

Landscape	Soil type	Texture	Uranium mass concentration (mg/kg)	Reference
Tundra	Soddy-podzolic[a]	Loam	2.5 ± 0.6	[4.8]
Taiga	Soddy-podzolic[a]	Loam	20.1 ± 2.5	[4.9]
Taiga	Soddy-podzolic	Loam	4.7 ± 2.9	[4.10]
Mixed forest	Typical chernozem	Clay	2.7 ± 0.2	[4.11]
Deciduous forest	Grey forest	Loam	1.2 ± 0.1	[4.11]
Meadows, steppe	Chernozem, brown earth[b]	Clay, heavy loam	3.0 ± 0.7	[4.11]
	Chernozem	Clay	2.6 ± 0.2	[4.12]
	Light brown	Clay, heavy loam	2.9 ± 0.7	[4.12]

TABLE 4.2. NATURAL URANIUM CONCENTRATION IN DIFFERENT SOIL TYPES (cont.)

Landscape	Soil type	Texture	Uranium mass concentration (mg/kg)	Reference
Dry subtropics	Red earth	Heavy loam	5.1 ± 0.3	[4.8]
Desert	Light brown desert		5.3 ± 0.5	[4.8]
Atlantic maritime	Podzolic	Sandy loam	1.9	[4.13]
Mixed wood plain	Grey brown luvisol and related gleysol phase	Loam	1.7	[4.13]
Boreal shield	Podzolic	Sandy loam	1.5	[4.13]
Mixed wood plain	Calcareous brunisol	Sandy loam	2.1	[4.13]
Boreal shield	Heavy clay grey luvisol	Clay loam	2.6	[4.13]
Boreal shield	Acidic brunisol (some marginal podzol)	Sand	0.4	[4.13]

[a] Area with elevated natural background, Komi Republic, Russian Federation.

[b] Samples taken from arable soil.

TABLE 4.3. CONCENTRATION OF ^{238}U IN SOIL BY COUNTRY

Region	Country	^{238}U activity concentration (Bq/kg)		^{238}U mass concentration (mg/kg)	
		Mean	Range	Mean	Range
Africa	Algeria	30	2–110	2.4	0.2–8.8
	Egypt	37	6–120	3.0	0.5–9.6
North America	Costa Rica	46	11–130	3.7	0.9–10.5

TABLE 4.3. CONCENTRATION OF ^{238}U IN SOIL BY COUNTRY (cont.)

Region	Country	^{238}U activity concentration (Bq/kg)		^{238}U mass concentration (mg/kg)	
		Mean	Range	Mean	Range
	USA	35	4–140	2.8	0.3–11.3
East Asia	China	33	2–690	2.7	0.2–55.5
	India	29	7–81	2.3	0.6–6.5
	Japan	29	2–59	2.3	0.2–4.7
	Kazakhstan	37	12–120	3.0	1.0–9.6
	Malaysia	66	49–86	5.3	3.9–6.9
	Thailand	114	3–370	9.2	0.2–29.7
West Asia	Armenia	46	20–78	3.7	1.6–6.3
	Syrian Arab Republic	23	10–64	1.8	0.8–5.1
Northern Europe	Lithuania	16	3–30	1.3	0.2–2.4
	Norway	50	—[a]	4.0	—[a]
Western Europe	Ireland	37	8–120	3.0	0.6–9.6
	Switzerland	40	10–150	3.2	0.8–12.1
Eastern Europe	Bulgaria	40	8–190	3.2	0.6–15.3
	Hungary	29	12–66	2.3	1.0–5.3
	Poland	26	5–120	2.1	0.4–9.6

TABLE 4.3. CONCENTRATION OF ^{238}U IN SOIL BY COUNTRY (cont.)

Region	Country	^{238}U activity concentration (Bq/kg)		^{238}U mass concentration (mg/kg)	
		Mean	Range	Mean	Range
	Romania	32	8–60	2.6	0.6–4.8
	Russian Federation	19	1–67	1.5	0.1–5.4
	Slovakia	32	15–130	2.6	1.2–10.5
Southern Europe	Albania	23	—[a]	1.8	—[a]
	Croatia	110	21–77	8.8	1.7–6.2
	Greece	25	1–240	2.0	0.1–19.3
	Portugal	49	26–82	3.9	2.1–6.6
Median		35	17–60	2.8	1.4–4.8

Source: Adapted from Ref. [4.14].

[a] —: data not available.

The data in Table 4.2 show that U mass concentrations in soils in areas of normal natural radiation background vary from 0.4 to 20 mg/kg. The highest mean values of 20.1 ± 2.5 mg/kg were found in soddy-podzolic loam soils of the taiga.

Available data on uranium measurements in soils of different countries are reported by UNSCEAR [4.14]. Table 4.3 shows that ^{238}U concentrations in soil vary over a wider range than that reported in Table 4.2, even within the same country, most likely because of the wider range of soil types investigated, inhomogeneity of the soils studied, a wider range of uranium concentrations in the parent rocks and other properties (in some cases localized) favouring uranium accumulation in the top soil. For example, this is the case for China (0.2–55.5 mg/kg) and Thailand (0.2–29.7 mg/kg).

The median value for the ^{238}U concentration in soil, calculated based on these data, is 35 Bq/kg or 2.8 mg/kg. However, there are some areas in the world

TABLE 4.4. EXAMPLES OF AREAS OF HIGH ^{238}U ACTIVITY CONCENTRATION IN SOIL

Country	Site	^{238}U activity concentration (Bq/kg)	Reference
Azerbaijan	Phosphate area	100–7 000	[4.14]
Brazil	Phosphate area	38–300	[4.14]
Czech Republic	Central Bohemia, Pluton middle area	68–220	[4.14]
Nigeria	Bisichi	8 700 ± 500	[4.15]
Nigeria	Bisichi (tin tailing)	22 000 ± 2 000	[4.15]
Russian Federation	Komi Republic	259–7 100	[4.16]

where the normal range of variation of naturally occurring radionuclides in soils or waters is greatly exceeded. These high radiation background regions are known as enhanced natural radiation areas. Some of the data available for these areas are given in Table 4.4 [4.14–4.16].

Maximum values were found at some Azerbaijan sites, at Ramsar, Iran and at Komi Republic, Russian Federation, where ^{238}U activity concentrations were two to three orders of magnitude higher than the average worldwide value of 32 Bq/kg reported in Ref. [4.14].

4.2.2. Terrestrial plants

The major source of uranium for terrestrial plants is root uptake from soil. Contaminated water used for irrigation can also affect plants through both soil contamination and direct translocation of uranium deposited on the leaves. Secondary external contamination, due to resuspension or rain splash of soil particles, can also contribute to external surface contamination on plants and plant parts close to the soil surface. For some contamination scenarios, soil particles retained on plant surfaces can be a major contributor to overall plant contamination [4.17]. The relative importance of the root and foliar uptake pathways depends on the concentration in the soil, the soil to plant transfer factor value and the uranium concentration in the water used for irrigation.

Plants demonstrate a high variability in terms of accumulation of radionuclides from the soil. However, the most striking point, relevant to all crops and areas, is that a large degree of variability in the accumulation of uranium (such as that observed for ^{137}Cs) by plants has not been observed (Table 4.5 [4.18–4.34]). Concentrations of uranium in wild species (such as mosses and wild grasses) are generally higher than those in agricultural plants. The highest concentrations in mosses (1.7–25.1 Bq/kg) were measured in Serbia [4.25].

Another general point is that uranium is distributed within plants very non-homogeneously. Thus, for trees, the highest uranium concentrations were found to be in roots, followed by the leaves (or needles) and branches, and finally the sapwood and hardwood [4.35, 4.36] (see Table 4.6 [4.35–4.45]). Specifically, the fine roots were shown to be very efficient in accumulating uranium [4.36].

This observation agrees with uranium accumulation in roots of woody plants reported in Refs [4.35, 4.46]. The uranium content in bulk root samples was consistently higher for fine roots with a diameter of <2 mm than for bigger roots. In addition, newly formed needles (aged <1 year) accumulated three times less uranium than did needles aged 2–4 years [4.36]. A similar effect was observed for other types of plant such as cereals and wild grasses [4.29]. Ratios of radionuclide concentrations in roots, compared with those in above ground biomass, may vary across a wide range and depend on the plant species [4.39]. Concentrations of uranium in the green parts of plants (leaves, stems) tend to be about one order of magnitude higher than in the fruits [4.33, 4.37]. The accumulation of uranium in plants greatly depends on the soil properties. This dependence is rather complex, both because of the presence of several uranium species and the variety of the processes affecting the bioavailability of this element in the soil [4.47]. Overall, plant accumulation of uranium from sandy soils is relatively high, which may be attributed to its low sorption (weak binding) in sandy soils. This is due to the low abundance of fine grained components that can strongly sorb uranium in such soils, whereas these components are more abundant in finer textured soils [4.17, 4.48]. Although uranium binding in sandy soils is relatively weak, leading to easier uptake by plants, these types of soil tend to have lower uranium content. Organic soils are known to give rise to limited plant accumulation of uranium (low concentration ratios), due to their strong retention of bound uranium.

Concentrations of uranium in plants or tree leaves vary seasonally. Uranium concentrations in vegetation samples taken in summer are normally two to three times higher than those taken in autumn [4.34]. Overall, mature plants accumulate more uranium compared with younger plants. Similar evidence was

Text cont. on p. 91.

TABLE 4.5. ACTIVITY CONCENTRATION OF ^{238}U IN PLANTS (UNDISTURBED AREAS)

Country	Plant	^{238}U activity concentration (Bq/kg, dry weight)		Reference
		Mean	Range	
Belarus	Forest berries (7 sites)	—[a]	(0.038 ± 0.007)–(0.39 ± 0.007)	[4.18]
	Wild grass (10 sites)	—[a]	(0.24 ± 0.04)–(3.9 ± 0.30)	[4.18]
Brazil	Black beans	0.012	<(0.004–0.024)	[4.19]
	Lettuce	0.70	0.30–1.37	[4.19]
	Carrots	0.05	<(0.04–1.32)	[4.19]
Canada	Barley (grain)	0.039	0.018–0.068	[4.10]
	Blueberry (leaves)	0.30	0.12–0.62	[4.10]
	Corn (grain)	0.0067	0.003–0.011	[4.10]
	Potato (tuber)	0.33	0.015–0.60	[4.10]
	Wild rice (grain)	0.010	0.004–0.017	[4.10]
India	Cereals (grain)	0.020 or 0.050	(0.016–0.030) or (0.030–0.060)	[4.20]

TABLE 4.5. ACTIVITY CONCENTRATION OF ^{238}U IN PLANTS (UNDISTURBED AREAS) (cont.)

Country	Plant	^{238}U activity concentration (Bq/kg, dry weight)		Reference
		Mean	Range	
	Sunflower	0.024 ± 0.001	—[a]	[4.21]
	Vegetables	—[a]	0.002 0 ± 0.000 3–0.064 ± 0.002	[4.21]
Japan	Brown rice	—[a]	0.002 5	[4.22]
	Polished rice	—[a]	0.006 2	[4.22]
Morocco	Medicinal plants (roots)	—[a]	(6.5 ± 0.5)–(12.3 ± 1.0)	[4.23]
	Medicinal plants (stems)	—[a]	(3.9 ± 0.3)–(7.3 ± 0.6)	[4.23]
	Medicinal plants (leaves)	—[a]	(4.6 ± 0.4)–(8.8 ± 0.7)	[4.23]
	Medicinal plants (fruits)	—[a]	(3.2 ± 0.3)–(4.8 ± 0.4)	[4.23]
	Medicinal plants (seeds)	—[a]	(3.4 ± 0.3)–(5.2 ± 0.4)	[4.23]
Poland	Moss	—[a]	(1.4 ± 0.1)–(3.3 ± 0.1)	[4.24]
Serbia	Moss	—[a]	(1.7 ± 0.2)–(25.1 ± 2.4)	[4.25]

TABLE 4.5. ACTIVITY CONCENTRATION OF ^{238}U IN PLANTS (UNDISTURBED AREAS) (cont.)

Country	Plant	^{238}U activity concentration (Bq/kg, dry weight)		Reference
		Mean	Range	
Slovenia	Pooled data (50 plants)	—[a]	(0.6 ± 0.2)–(1.0 ± 0.2)	[4.26]
Sweden	Cereals (grain)	0.085 0.096	0.05–0.14 0.03–0.20	[4.27]
	Grass	0.19	—[a]	[4.27]
	Rape (seeds)	0.095	0.043–0.192	[4.27]
	Sugar beet (tops)	3.2	0.8–5.6	[4.27]
	Turnip rape (seeds)	0.010	0.004–0.015	[4.27]
	Pooled data (40 crops)	0.17	0.003–1.32	[4.28]
Russian Federation	Wheat (leaves, roots)	—[a]	0.070–0.20	[4.29]
United Kingdom	Wild grass	0.36	>0.02–2.3	[4.30]
	Pine tree	0.007 3	0.000 8–0.026 0	[4.30]
USA	Beans	—[a]	0.030 ± 0.006	[4.31]

TABLE 4.5. ACTIVITY CONCENTRATION OF ^{238}U IN PLANTS (UNDISTURBED AREAS) (cont.)

Country	Plant	^{238}U activity concentration (Bq/kg, dry weight)		Reference
		Mean	Range	
	Corn	—[a]	0.012 ± 0.002	[4.31]
	Squash	—[a]	0.079 ± 0.007	[4.31]
	Wild grass	4.4 ± 0.7	—[a]	[4.32]
	Pinon pine (shoots)	—[a]	(0.40 ± 0.15)–(0.58 ± 0.22)	[4.33]
	Pinon pine (nuts)	0.016 ± 0.003	—[a]	[4.33]
	Boxelder tree (leaves)	—[a]	0.07–1.25	[4.34]

[a] —: data not available.

TABLE 4.6. ACTIVITY CONCENTRATION OF ^{238}U IN PLANTS (AREAS PERTURBED BY HUMAN ACTIVITIES AND AREAS WITH HIGH NATURAL BACKGROUND)

Country and source	Plant	^{238}U activity concentration (Bq/kg, dry weight)		Reference
		Mean	Range	
Canada (former uranium mining site)	Mixed grass	—[a]	10–1024	[4.37]
	Willow	—[a]	10–30	[4.37]
	Black spruce	10	—[a]	[4.37]
	Jack pine	30	—[a]	[4.37]
Canada (waste disposal site)	Scots pine (shoots)	6.2	4.1–12	[4.35]
	Scots pine (roots)	740	120–1700	[4.35]
Germany (former uranium mining site)	Scots pine (needles, 1 year)	1.6	—[a]	[4.36]
	Scots pine (needles, >1 year)	4.1		
	Scots pine (sapwood)	0.025	—[a]	[4.36]
	Scots pine (heartwood)	0.012	—[a]	[4.36]
	Scots pine (roots)	140.0	—[a]	[4.36]
	Grass (roots)	420 ± 110	—[a]	[4.37]
	Grass (shoots)	200 ± 50	—[a]	[4.37]

TABLE 4.6. ACTIVITY CONCENTRATION OF ^{238}U IN PLANTS (AREAS PERTURBED BY HUMAN ACTIVITIES AND AREAS WITH HIGH NATURAL BACKGROUND) (cont.)

Country and source	Plant	^{238}U activity concentration (Bq/kg, dry weight)		Reference
		Mean	Range	
India (uranium mining area, 0–10 km)	Vegetables	—[a]	(0.56 ± 0.08)–(1.2 ± 0.52)	[4.38]
	Fruit	—[a]	(0.68 ± 0.31)–(0.86 ± 0.69)	[4.38]
India (uranium mining area, 10–20 km)	Vegetables	—[a]	(0.46 ± 0.14)–(1.4 ± 0.82)	[4.38]
	Fruit	—[a]	(0.34 ± 0.08)–(0.91 ± 0.35)	[4.38]
Jordan (uranium deposit site)	Watermelon (pulp)	0.010	—[a]	[4.39]
	Zucchini	0.025	—[a]	[4.39]
Nigeria (area with high radiation background)	Red beans	9.7 ± 2.9	—[a]	[4.40]
	Rice	2.8 ± 1.2	—[a]	[4.40]
	Cassava roots	—[a]	(15 ± 9)–(40 ± 10)	[4.40]
	Cereals	6.1	—[a]	[4.41]

TABLE 4.6. ACTIVITY CONCENTRATION OF ^{238}U IN PLANTS (AREAS PERTURBED BY HUMAN ACTIVITIES AND AREAS WITH HIGH NATURAL BACKGROUND) (cont.)

Country and source	Plant	^{238}U activity concentration (Bq/kg, dry weight)		Reference
		Mean	Range	
	Tubers	5.6	—[a]	[4.41]
Nigeria (tin mining site)	Vegetables	—[a]	(0.032 ± 0.003)–(1.9 ± 0.19)	[4.15]
	Beans	—[a]	(0.034 ± 0.004)–(0.46 ± 0.05)	[4.15]
	Cereals	—[a]	(0.021 ± 0.002)–(0.050 ± 0.008)	[4.15]
	Tubers	—[a]	(0.25 ± 0.03)–(0.47 ± 0.04)	[4.15]
Portugal (near uranium mine)	Lettuce[b]	—[a]	(2.4 ± 1.0)–(22 ± 3)	[4.42]
	Lettuce[b]	—[a]	(15 ± 7)–(66 ± 11)	[4.42]
Serbia (uranium mining site)	Leafy vegetables	—[a]	2 000–14 000	[4.43]
	Potato	—[a]	5 000–13 000	[4.43]
	Bean	—[a]	5 300–6 500	[4.43]
	Sunflower	—[a]	1 700–5 900	[4.43]

TABLE 4.6. ACTIVITY CONCENTRATION OF ^{238}U IN PLANTS (AREAS PERTURBED BY HUMAN ACTIVITIES AND AREAS WITH HIGH NATURAL BACKGROUND) (cont.)

Country and source	Plant	^{238}U activity concentration (Bq/kg, dry weight)		Reference
		Mean	Range	
	Maize	—[a]	360 ± 5 700	[4.43]
Slovenia (uranium mining site)	Pooled data: 50 plants	—[a]	(1.9 ± 0.3)–(7.8 ± 0.6)	[4.26]
Spain	Tomato (fruit)[b]	0.090 ± 0.020	—[a]	[4.44]
United Kingdom (Dartmoor area)	Heather	1.7	0.5–3.1	[4.45]
	Gorse	1.6	0.6–3.9	[4.45]
	Rowan	0.9	0.6–1.5	[4.45]
	Fir (*Abies alba*)	1.5	0.9–2.1	[4.45]
	Spruce (*Picea abies*)	2.2	1.2–3.1	[4.45]
USA (former uranium mining site)	Mixed grass[c]	5.6 ± 0.5	—[a]	[4.32]
	Mixed grass	—[a]	(9.6 ± 0.7)–(407 ± 85)	[4.32]
USA (Los Alamos National Laboratory area)	Pinon pine (shoots)	—[a]	1.3 ± 1.4	[4.33]

TABLE 4.6. ACTIVITY CONCENTRATION OF ^{238}U IN PLANTS (AREAS PERTURBED BY HUMAN ACTIVITIES AND AREAS WITH HIGH NATURAL BACKGROUND) (cont.)

Country and source	Plant	^{238}U activity concentration (Bq/kg, dry weight)		Reference
		Mean	Range	
USA (Los Alamos National Laboratory area)	Pinon pine (nuts)	—[a]	0.016 ± 0.006	[4.33]
	Beans	—[a]	0.012 ± 0.006	[4.31]
	Corn	—[a]	0.025 0 ± 0.002 5	[4.31]
	Squash	—[a]	0.034 ± 0.010	[4.31]

[a] —: data not available.

[b] Irrigated with contaminated water.

[c] Remediated area.

provided in Ref. [4.43], where it was reported that leaves accumulated more uranium than storage organs (bulbs, tubers, cobs, pods and seeds) and that older leaves accumulated more uranium than younger leaves.

The uranium content of phosphate fertilizers can be significantly elevated owing to the naturally high uranium content of many source phosphate rocks used to make fertilizer [4.14]. However, no significant differences were found in uranium activity concentrations in vegetables from conventional farming systems (based on the application of mineral fertilizers) and organic ones [4.19]. This is possibly because the amount of uranium supplied by the fertilization is relatively low compared with the total concentration of uranium in soil.

The accumulation of uranium in plants depends on their overall ability to take up micro- and macro-elements. In Ref. [4.49], it is reported that uranium accumulation in plants decreases from maize to lentil to chickpea to rice to wheat. In Ref. [4.50], data are reported for 34 species of vegetation. A significant difference was found in the accumulation of uranium by different plant species. The highest values were found in sunflower, beetroot and Indian mustard, and the lowest values in wheat grain.

As previously discussed for uranium in the soil, activity concentrations of radionuclides in plants in areas of high natural background or areas perturbed by human activities, such as mining and waste disposal sites, were typically found to be two to four orders of magnitude higher than concentrations from areas with a normal radiation background.

In sandy soils, plants can take more uranium through fine roots located mainly in the top-soil layers (shallow); thus, the higher presence of roots in that horizon can lead to enhanced transfer of uranium to the above ground biomass [4.51]. However, as noted previously, total uptake may be limited by the low uranium concentrations often found in sandy soils.

Plants grown in uranium enriched soils demonstrated significant increases in concentrations of uranium in roots, whereas concentrations in the above ground biomass remained rather low. Limited transfers of uranium to plants in uranium enriched soils may be due to the sublinearity of accumulation of uranium in plant tissues with increasing soil concentration observed in some studies in controlled conditions [4.52].

4.2.3. Terrestrial animals

4.2.3.1. Domesticated animals

The main source of uranium (^{238}U) in terrestrial animal products is intake of contaminated feed, uranium adhering to soil particles on the surface of ingested vegetation for herbivores and omnivores, and drinking water. A review of gastrointestinal fractional absorption values for domestic ruminants [4.53] found only two similar values for cows $(1.1 \pm 0.14) \times 10^{-2}$ [4.54, 4.55]. These values are similar to those recommended for humans in Ref. [4.56] and by the International Commission on Radiological Protection (ICRP) [4.57] of 2.0×10^{-2}. Thus, the low degree of fractional absorption limits bioaccumulation in tissues, as discussed further in Chapter 6.

Collated information on ^{238}U activity concentrations in milk and meat products, such as beef, poultry, pork and mutton, of domestic and non-domesticated animals is given here, as well as some data for liver and kidney, which also constitute components of the human diet. The ^{238}U activity concentrations are given for typical ‘undisturbed’ areas of land with a normal range of natural uranium concentration (Tables 4.7 and 4.8 [4.58–4.72]) and for areas with a high natural background or ‘perturbed’ sites such as uranium and coal mining, and nuclear weapon testing sites (Tables 4.9 and 4.10 [4.73–4.82]). For each type of area, the data are subdivided into animal products from domesticated and non-domesticated animals.

4.2.3.2. Undisturbed areas

For undisturbed areas, most of the data come from market and food basket studies conducted in different countries. Such food basket data can comprise milk and/or a combination of milk products. Meat activity concentrations have been reported for unspecified ‘meat’ values as well as for beef, pork, chicken, poultry, mutton, lamb and venison. Data values have not been included when they refer to a mixture of animal products and additional non-animal items. Quality control of potential data inputs also led to the exclusion of some outlying activity concentration values.

Although some values are the same as those in Ref. [4.83], other values in Tables 4.7 and 4.8 are not directly comparable. Several vales of minimum detectable amounts reported in the literature are considerably higher than the values reported in Tables 4.7 and 4.8, so the application of these values in providing cautious estimates of intakes would be likely to result in excessive overestimation.

Collated milk ^{238}U activity concentrations in undisturbed areas from nine countries range from 0.6×10^{-4} to 8.0×10^{-1} Bq/kg. An arithmetic mean of 7.0×10^{-2} Bq/kg, fresh weight (FW) can be derived from Table 4.7 (using the midpoint for ranges), which is substantially higher than the UNSCEAR recommended value of 1.0×10^{-3} Bq/kg [4.84].

Collated domestic animal meat ^{238}U activity concentrations in undisturbed areas from nine countries range from 0.8×10^{-3} to 4.6×10^{-1} Bq/kg. An arithmetic mean of 4.9×10^{-2} Bq/kg, FW can be derived from Table 4.7 (using the midpoint

TABLE 4.7. ACTIVITY CONCENTRATION OF ^{238}U IN PRODUCTS FROM DOMESTICATED ANIMALS (UNDISTURBED AREAS)

Country	Animal product	^{238}U activity concentration (Bq/kg, fresh weight)		N^{a}	Reference
		Mean	Range		
Brazil	Milk	$(1.2 \pm 0.74) \times 10^{-3}$	—[b]	—[b]	[4.58]
	Milk	6.2×10^{-3}	—[b]	3	[4.59]
Denmark	Milk	—[b]	$(0.6\text{–}1.0) \times 10^{-4}$	2	[4.60]
Finland	Milk	—[b]	$(2.0\text{–}7.0) \times 10^{-4}$	4	[4.60]
Germany	Milk (condensed and powdered)	—[b]	$(2.0\text{–}6.0) \times 10^{-2}$	6	[4.61]
India	Milk products	—[b]	$(1.6\text{–}1.7) \times 10^{-2}$	2	[4.62]
Poland	Milk	—[b]	$(3.0 \pm 0.8) \times 10^{-3}$	—[b]	[4.63]
	Milk (all milk products except butter)	—[b]	$(2.6 \pm 0.3) \times 10^{-3}$	—[b]	[4.64]
Serbia	Cow milk	—[b]	$(6.0\text{–}8.0) \times 10^{-1}$	10	[4.65]

TABLE 4.7. ACTIVITY CONCENTRATION OF ^{238}U IN PRODUCTS FROM DOMESTICATED ANIMALS (UNDISTURBED AREAS) (cont.)

Country	Animal product	^{238}U activity concentration (Bq/kg, fresh weight)		*N*[a]	Reference
		Mean	Range		
United Kingdom	Milk	1.2×10^{-4}	—[b]	6	[4.66]
USA	Milk products	7.3×10^{-4}	—[b]	—[b]	[4.67]
Brazil	Meat	1.2×10^{-2}	—[b]	4	[4.59]
	Meat	$(2.3 \pm 0.01) \times 10^{-2}$ (beef)	—[b]	—[b]	[4.58]
		$(5.0 \pm 0.3) \times 10^{-3}$ (chicken)		—[b]	
Bosnia and Herzegovina	Meat	—[b]	$(1.9–3.5) \times 10^{-3}$	10	[4.68]
Germany	Meat	1.1×10^{-1} (beef)	—[b]	6	[4.61]
		6.6×10^{-2} (pork)	—[b]	—[b]	
		1.0×10^{-1} (chicken)	—[b]	—[b]	
		1.2×10^{-1} (mutton)	—[b]	—[b]	
Poland	Meat	$(3.8 \pm 0.6) \times 10^{-3}$ (beef)	—[b]	—[b]	[4.63]
		$(2.3 \pm 0.5) \times 10^{-3}$ (pork)	—[b]	—[b]	
		$(2.6 \pm 0.2) \times 10^{-3}$ (poultry)	—[b]	—[b]	

TABLE 4.7. ACTIVITY CONCENTRATION OF ^{238}U IN PRODUCTS FROM DOMESTICATED ANIMALS (UNDISTURBED AREAS) (cont.)

Country	Animal product	^{238}U activity concentration (Bq/kg, fresh weight)		N[a]	Reference
		Mean	Range		
	Meat	$(5.6 \pm 0.1) \times 10^{-3}$ (beef) $(1.6 \pm 0.5) \times 10^{-3}$ (pork) $(1.7 \pm 0.3) \times 10^{-3}$ (poultry)	—[b] —[b] —[b]	—[b] —[b] —[b]	[4.64]
United Kingdom	Meat	4.9×10^{-3} (beef, pork, lamb)	—[b]	14	[4.66]
USA	Beef	$(2.6 \pm 0.7) \times 10^{-2}$	—[b]	5	[4.69]
	Meat, poultry	0.8×10^{-3} (poultry) 2.3×10^{-3} (meat)	—[b] —[b]	—[b] —[b]	[4.67]
Former Yugoslavia	Beef	2.6×10^{-2}	—[b]	6	[4.70]
	Venison	4.6×10^{-1}	—[b]	—[b]	[4.71]

[a] N: Number of individual data entries.

[b] —: data not available.

TABLE 4.8. ACTIVITY CONCENTRATION OF ^{238}U IN THE FLESH OF WILD ANIMALS (UNDISTURBED AREAS) [4.72]

Country	Animal product	^{238}U activity concentration (Bq/kg, fresh weight)	No. of individual data entries
Australia	Bandicoot	4.5×10^{-2}	1
	Buffalo	9.4×10^{-3}	4
	Goanna	3.5×10^{-2}	1
	Magpie goose	1.6×10^{-2}	5
	Snake	3.4×10^{-2}	1
	Wild boar	1.4×10^{-2}	3

for ranges), which is more than an order of magnitude higher than the UNSCEAR recommended value of 2.0×10^{-3} Bq/kg [4.84].

The small amount of data for food products from non-domesticated animals in undisturbed areas are reported in Table 4.8. Collated non-domesticated animal product ^{238}U activity concentrations in undisturbed areas from Australia range from 9.4×10^{-3} Bq/kg for buffalo meat to 4.5×10^{-2} Bq/kg for bandicoot flesh [4.72]. All the flesh ^{238}U activity concentrations exceed the value for meat cited by UNSCEAR of 2×10^{-3} Bq/kg [4.84].

4.2.3.3. Perturbed areas

The ^{238}U activity concentrations in domesticated animal products in areas perturbed by humans or with high natural background are largely based on data from uranium mining sites (see Table 4.9). In moose and cattle from uranium mining areas in Saskatchewan, Canada, uranium concentrations in bone were about one order of magnitude higher than concentrations in soft tissues, all of which exhibited similar concentrations [4.78].

The few data for ^{238}U activity concentrations in milk in perturbed areas from two countries in Table 4.9 range from 1.4×10^{-2} Bq/kg for a coal mine in Bosnia and Herzegovina to 3.3×10^{-2} Bq/kg for a proposed uranium mine site in India.

TABLE 4.9. ACTIVITY CONCENTRATION OF ^{238}U IN PRODUCTS FROM DOMESTICATED ANIMALS (AREAS PERTURBED BY HUMAN ACTIVITIES OR WITH A HIGH NATURAL BACKGROUND)

Country and source	Animal product	^{238}U activity concentration (Bq/kg, fresh weight)		*N*[a]	Reference
		Mean	Range		
Bosnia and Herzegovina (Brown coal mine)	Milk	1.4×10^{-2} (beef) 3.0×10^{-2} (mutton)	—[b] —[b]	2 —[b]	[4.73]
India (proposed uranium mine area)	Milk	2.1×10^{-2}	$(1.5–3.3) \times 10^{-2}$	60	[4.74]
	Milk	—[b]	$(1.9–3.3) \times 10^{-2}$	12	[4.75]
Slovenia (former uranium mine)	Milk, powdered milk and infant diet samples	1.4×10^{-2}	—[b]	5	[4.76]
Bosnia and Herzegovina (Brown coal mine)	Meat	1.5×10^{-2} (mutton) 1.6 (chicken)	—[b] —[b]	2 —[b]	[4.73]
Brazil (high natural background)	Meat	$(1.1 \pm 0.8) \times 10^{-2}$ (beef) $(3.4 \pm 0.6) \times 10^{-3}$ (pork) $(1.2 \pm 0.6) \times 10^{-2}$ (poultry)	—[b] —[b] —[b] —[b]	3 —[b] —[b] —[b]	[4.77]

TABLE 4.9. ACTIVITY CONCENTRATION OF ^{238}U IN PRODUCTS FROM DOMESTICATED ANIMALS (AREAS PERTURBED BY HUMAN ACTIVITIES OR WITH A HIGH NATURAL BACKGROUND) (cont.)

Country and source	Animal product	^{238}U activity concentration (Bq/kg, fresh weight)		N^a	Reference
		Mean	Range		
Canada (uranium mine near Saskatchewan)	Beef	1.9×10^{-2}	—^{b}	4	[4.78]
India (proposed uranium mine area)	Beef	$(300 \pm 9) \times 10^{-3}$	—^{b}	4	[4.79]
	Chicken	—^{b}	$(1.9–7.5) \times 10^{-2}$	8	[4.75]
Serbia (coal fired power station)	Beef	—^{b}	2.1 ± 0.2	—^{b}	[4.80]
USA (Nevada test site)	Beef	3.0×10^{-3}	—^{b}	—^{b}	[4.81]
USA (New Mexico uranium mining; Ambrosia Lake flooded by effluent from a uranium mine)	Beef	$(3.0 \pm 0.7) \times 10^{-2}$	—^{b}	5	[4.69]

TABLE 4.9. ACTIVITY CONCENTRATION OF ^{238}U IN PRODUCTS FROM DOMESTICATED ANIMALS (AREAS PERTURBED BY HUMAN ACTIVITIES OR WITH A HIGH NATURAL BACKGROUND) (cont.)

Country and source	Animal product	^{238}U activity concentration (Bq/kg, fresh weight)		N^a	Reference
		Mean	Range		
USA (New Mexico uranium mining; Ambrosia Lake access to surface impoundments)	Beef	$(1.9 \pm 0.7) \times 10^{-2}$	—[b]	5	[4.69]

[a] Number of individual data points.

[b] —: data not available.

The data for domestic animal meat ^{238}U activity concentrations in perturbed areas from five countries range from 3.0×10^{-3} Bq/kg for beef at the Nevada test site, USA up to 1.6 Bq/kg for chicken from a coal mining area in Bosnia and Herzegovina. The chicken value exceeds the value for meat in normal background areas cited by UNSCEAR by three orders of magnitude and was attributed to the foraging habits of the chickens [4.73].

As might be expected, all the milk and meat activity concentration values collated in perturbed or high natural background areas exceeded the value cited by UNSCEAR for normal background areas.

The ^{238}U activity concentrations for non-domesticated animal products from perturbed areas are reported in Table 4.10. The non-domesticated animal products in Table 4.10 are meat from caribou from Canada, and wallaby, bandicoot and wild boar, which are consumed by Aboriginal communities in Australia. The data are for areas located close to uranium mines in Canada and for perturbed areas of the Ranger uranium mine in Australia.

Reference [4.85] also reported uranium concentrations in various small mammals (redbacked, meadow and heather voles; masked shrews; deer

TABLE 4.10. ACTIVITY CONCENTRATION OF ^{238}U IN THE FLESH OF WILD ANIMALS (AREAS PERTURBED BY HUMAN ACTIVITIES OR WITH A HIGH NATURAL BACKGROUND)

Country and source	Animal species	^{238}U activity concentration (Bq/kg, fresh weight)	No. of individual data entries	Reference
Australia (Ranger uranium mine)	Wallaby	6.1×10^{-2}	1	[4.72]
Canada (near uranium mine)	Caribou	4.0×10^{-2}	18	[4.82]
Woolaston Lake area	Caribou	3.8×10^{-2}	—[a]	[4.78]
Key Lake area	Caribou	7.2×10^{-2}	—[a]	[4.78]
Uranium City	Caribou	4.1×10^{-2}	1	[4.78]
Meadow Cake area	Caribou	1.6×10^{-2}	—[a]	[4.78]

[a] —: data not available.

mice and meadow jumping mice) from the Key Lake mine and mill complex in north-central Saskatchewan. At the three locations studied, arithmetic mean concentrations in the whole body on an FW basis were 24.5, 48.1 and 28.2 ppb (μg/kg). These correspond to 0.30, 0.59 and 0.35 Bq/kg; that is, somewhat higher than concentrations in caribou from the same site (see Table 4.10). Uranium concentrations in the tissues of the small mammals were about a factor of a hundred lower than concentrations in associated vegetation (i.e. uranium was bioexcluded rather than bioaccumulated) [4.86].

Collated non-domesticated animal ^{238}U activity concentrations in meat from Australia and Canada have a narrow range of the order of 10^{-2} Bq/kg.

Extensive data on uranium concentrations in wildlife are available for the Russian Federation (Table 4.11 [4.87]). There are no substantial differences between concentrations for different species or between different types of organism. Specifically, there is no evidence of strong bioaccumulation or bioexclusion at higher trophic levels of animals. Bioaccumulation or bioexclusion relative to plants is not addressed by these data. Limited data from areas contaminated with natural and depleted uranium at the Los Alamos National Laboratory, USA showed strong bioexclusion in two species of small mammals that had tissue:soil concentration ratios of 10^{-3} and 10^{-4} [4.88] (see also Ref. [4.89]).

Liver and kidney ^{238}U activity concentrations have been reported for cattle in both undisturbed and perturbed sites. For undisturbed areas, values are reported from the Nevada test site ranging from 2.0×10^{-3} to 1.5×10^{-2} Bq/kg for liver and 5.0×10^{-2} Bq/kg for kidney [4.81]. Reported ^{238}U activity concentrations in kidney for perturbed areas range from 4.4×10^{-4} to 4.1×10^{-1} Bq/kg and for liver from 2.2×10^{-3} to 1.9×10^{-1} Bq/kg.

In Australia, ^{238}U activity concentration for bandicoot living in undisturbed areas is higher in kidney than liver and muscle (Fig. 4.4). However, for wallaby living in perturbed regions, liver activity concentration is the highest [4.72]. In Canada, caribou ^{238}U activity concentrations at three sites were higher for liver than for muscle and kidney, but at a fourth site, those for kidney were higher [4.78].

UNSCEAR reported that ^{238}U concentrations in bone are normally higher than in other tissues in humans and that bone contains most of the body burden. The ^{238}U data for femurs reported in Ref. [4.81] show higher concentrations than for liver and kidney for cattle located in both a control site and contaminated areas in New Mexico, USA. Caribou bone ^{238}U activity concentrations were an order of magnitude higher than those for liver and kidney at a uranium mine site in Canada [4.78].

Text cont. on p. 108.

TABLE 4.11. ACTIVITY CONCENTRATION OF ^{238}U IN SOME WILDLIFE SPECIES IN AREAS IN THE RUSSIAN FEDERATION WITH DIFFERENT RADIATION BACKGROUND LEVELS [4.87]

Biota species	Areas of normal range of radiation background		Areas of enhanced radiation background	
	No. of samples	^{238}U activity concentration (Bq/kg, fresh weight)	No. of samples	^{238}U activity concentration (Bq/kg, fresh weight)
Small rodents	862		309	
Mole (*Talpa europaea*)	36	0.005	49	0.123
Shrew (*Sorex* L.)	94	0.03	56	0.21
Tundra vole (*Microtus oeconomus* Pall.)	491	0.039	38	0.22
Dark vole (*Microtus agrestis*)	68	0.037	7	0.17
Redhead vole (*Myodes glareolus*)	46	0.031	74	0.18
Water vole (*Arvicola terrestris*)	39	0.022	11	0.08
Red vole (*Clethrionomys rutilus* Pall.)	49	0.028	63	0.17
Bank vole (*Myodes glareolus*)	39	0.02	11	0.08
Small herbivorous animals	45		55	
Squirrel (*Sciurus*)	15	0.011	36	0.022
Chipmunk (*Tamias*)	18	0.015	5	0.027
Hare (*Lepus timidus*)	12	0.012	14	0.026

TABLE 4.11. ACTIVITY CONCENTRATION OF ^{238}U IN SOME WILDLIFE SPECIES IN AREAS IN THE RUSSIAN FEDERATION WITH DIFFERENT RADIATION BACKGROUND LEVELS [4.87] (cont.)

Biota species	Areas of normal range of radiation background		Areas of enhanced radiation background	
	No. of samples	^{238}U activity concentration (Bq/kg, fresh weight)	No. of samples	^{238}U activity concentration (Bq/kg, fresh weight)
Small carnivorous animals	44		44	
Marten (*Martes*)	12	0.018	8	0.022
Ermine (*Mustela erminea*)	13	0.022	9	0.12
Weasel (*Mustela nivalis*)	5	0.018	10	0.69
European mink (*Mustela lutreola*)	5	0.015	5	0.022
Otter (*Lutra lutra*)	6	0.018	9	0.033
Fox (*Vulpes vulpes*)	3	0.011	3	0.015
Amphibians and reptiles	88		19	
Viviparous lizard (*Zootoca vivipara*)	57	0.043	—[a]	—[a]
Moor frog (*Rana arvalis*)	31	0.015	19	0.49
Herbivorous and insectivorous birds	264		834	
Western capercaillie (*Tetrao urogallus*)	17	0.044	205	0.154
Black grouse (*Lyrurus tetrix*)	15	0.05	184	0.19
Hazel grouse (*Tetrastes bonasia*)	25	0.033	208	0.12

TABLE 4.11. ACTIVITY CONCENTRATION OF ^{238}U IN SOME WILDLIFE SPECIES IN AREAS IN THE RUSSIAN FEDERATION WITH DIFFERENT RADIATION BACKGROUND LEVELS [4.87] (cont.)

Biota species	Areas of normal range of radiation background		Areas of enhanced radiation background	
	No. of samples	^{238}U activity concentration (Bq/kg, fresh weight)	No. of samples	^{238}U activity concentration (Bq/kg, fresh weight)
Willow grouse (*Lagopus lagopus*)	15	0.041	61	0.11
Eurasian teal (*Anas crecca*)	7	0.07	5	0.11
Eurasian wigeon (*Anas penelope*)	3	0.053	—[a]	—[a]
Mergus merganser (*Mergus merganser*)	4	0.047	4	0.12
Siberian jay (*Perisoreus infaustus*)	28	0.011	41	0.12
Common crossbill (*Loxia curvirostra*)	30	0.006	12	0.11
Parrot crossbill (*Loxia pytyopsittacus*)	18	0.042	41	0.13
Great tit (*Parus major*)	15	0.15	19	0.3
White wagtail (*Motacilla alba*)	12	0.19	21	0.3
Bullfinch (*Pyrrhula pyrrhula*)	6	0.15	17	0.2
Greater spotted woodpecker (*Dendrocopos major*)	12	0.007	6	0.11
Woodpecker (*Dryocopus martius*)	57	0.37	10	4.4

TABLE 4.11. ACTIVITY CONCENTRATION OF ^{238}U IN SOME WILDLIFE SPECIES IN AREAS IN THE RUSSIAN FEDERATION WITH DIFFERENT RADIATION BACKGROUND LEVELS [4.87] (cont.)

Biota species	Areas of normal range of radiation background		Areas of enhanced radiation background	
	No. of samples	^{238}U activity concentration (Bq/kg, fresh weight)	No. of samples	^{238}U activity concentration (Bq/kg, fresh weight)
Carnivorous birds	11		13	
Goshawk (*Accipiter gentilis*)	2	0.01	3	0.043
Sparrow-hawk (*Accipiter castanilius*)	3	0.011	4	0.037
Eurasian eagle-owl (*Bubo bubo*)	2	0.06	3	0.12
Ural owl *(Strix uralensis)*	4	1.07	3	11.5

Note: Data for ^{238}U activity concentration are given for the whole body.

[a] —: data not available.

TABLE 4.12. URANIUM DISTRIBUTION IN MUTTON TISSUES AND ORGANS FROM PERTURBED AND UNDISTURBED SITES

Mutton organ/tissue	% of total body weight	Weight of organs and tissues (g)	Uranium content (μg) in the whole organ or tissue[a]					
			Perturbed sites (low organic, shale soils)			Undisturbed sites (black soils and podzols)		
			Kok Moinok, Kyrgyzstan	Ton, Kyrgyzstan	Cholpan-Ata, Kyrgyzstan	Tula, Russian Federation	Moscow, Russian Federation	Kaluga, Russian Federation
Muscle	39	135 000	159	97.0	87.7	33.6	24.3	28.3
Alimentary canal	11.4	4 000	128	81.6	76	18.4	13.6	15.2
Liver	2.2	740	5.89	4.77	3.90	2.06	1.88	1.77
Lung	2.4	800	7.55	6.24	5.28	1.62	1.10	1.44
Heart	0.7	240	2.54	1.89	1.70	0.60	0.48	0.50
Kidney	0.35	120	2.30	1.58	1.56	0.40	0.28	0.30
Spleen	0.6	90	2.25	1.20	1.15	0.30	0.2	0.20
Brain	0.35	120	1.20	0.70	0.60	0.20	0.26	0.20
Testes	1	340	3.10	2.75	2.58	2.20	1.96	2.00

TABLE 4.12. URANIUM DISTRIBUTION IN MUTTON TISSUES AND ORGANS FROM PERTURBED AND UNDISTURBED SITES (cont.)

Mutton organ/tissue	% of total body weight	Weight of organs and tissues (g)	Uranium content (μg) in the whole organ or tissue[a]					
			Perturbed sites (low organic, shale soils)			Undisturbed sites (black soils and podzols)		
			Kok Moinok, Kyrgyzstan	Ton, Kyrgyzstan	Cholpan-Ata, Kyrgyzstan	Tula, Russian Federation	Moscow, Russian Federation	Kaluga, Russian Federation
Blood	5.7	2 000	14.7	10.8	7.74	4.18	3.92	3.80
Bone	18	6 200	948	595	515	144	118	130
Skin	6	2 100	378	191	172	25.2	12.6	11.7
Wool	4.7	1 600	334	157	144	22.4	9.92	9.6
Whole organism	100		1 986	1 511	1 019	251	188	205

Source: adapted from Ref. [4.90].

[a] Organs and tissues taken from four to eight animals; uranium content for the whole organism was calculated from the sum of uranium in different organs.

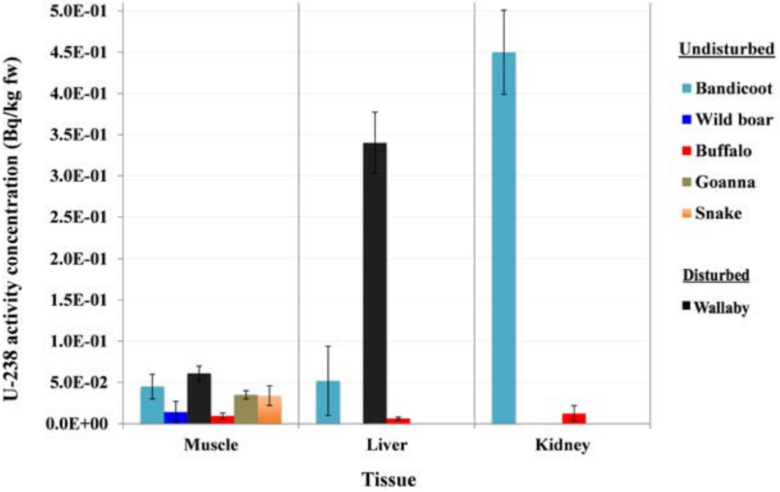

FIG. 4.4. Activity concentration of ^{238}*U (individual data and/or arithmetic means, with their measurement uncertainty or standard deviation) for selected wildlife tissues and organisms in undisturbed and perturbed areas of Australia [4.72]. fw: fresh weight.*

Data reported in Ref. [4.90] give uranium contents in various mutton tissues, with uranium in bone ranging from 19 to 153 µg/kg for six different sites and accounting for most of the body burden (Table 4.12). Uranium contents in muscle ranged from 0.18 to 0.72 µg/kg and these contents were higher than the liver or kidney values. In Australia, uranium concentrations in bone were 4.7 µg/kg for bandicoot and 1 600 µg/kg (fresh weight) for wild boar in undisturbed areas and ranged from 610 to 1 700 µg/kg for wild boar in perturbed areas [4.72].

4.3. URANIUM IN GROUNDWATER

4.3.1. Ranges of uranium concentration in groundwater

Trace element concentrations in groundwaters have been analysed for samples collected between 1992 and 2003 from aquifers across the USA as part of the United States Geological Survey National Water-Quality Assessment study. The trace elements include uranium, and this dataset [4.91] is a valuable major national survey of uranium concentrations. Data from 5183 monitoring and drinking water wells, representing more than 40 principal and other aquifers in humid and dry regions and in various land use settings were reported. The data were assessed in terms of concentration above analytical reporting levels and by comparing concentrations with human health benchmarks (HHBs), also described as maximum contaminant levels. The data collected in this major study represent various types of aquifer, ranging from unconsolidated sand and gravel aquifers through to crystalline rock aquifers, which are briefly described in Table 4.13.

TABLE 4.13. BRIEF DESCRIPTIONS OF AQUIFER TYPES IN THE UNITED STATES GEOLOGICAL SURVEY NATIONAL WATER QUALITY ASSESSMENT STUDY [4.91]

Aquifer type	Description
Unconsolidated sand and gravel	Includes basin fill (valley fill) aquifers, and sand and gravel aquifers, as well as stream valley aquifers. These aquifers are characterized by intergranular porosity, and all contain water primarily under unconfined or water table conditions in different hydrogeological settings. Examples include the High Plains aquifer, the most intensively pumped aquifer in the USA.
Glacial unconsolidated sand and gravel	Sediments deposited during cycles of continental glaciation. These sedimentsare unconsolidated sand and gravel characterized by intergranular porosity, contain water primarily under unconfined or water table conditions in different hydrogeological settings.
Semi-consolidated sand	Sediments that primarily consist of semi-consolidated sand, silt and clay, interbedded with some carbonate rocks, underlie the coastal plains that border the Atlantic Ocean and the Gulf of Mexico. Depositional environments include fluvial, deltaic and shallow marine.
Sandstone	Aquifers in sandstone are more widespread than those in all other kinds of consolidated rock. Sandstone aquifers commonly grade laterally into fine grained, low permeability rocks such as shale or siltstone. Many sandstone aquifers are parts of complex interbedded sequences of sedimentary rocks.
Sandstone and carbonate rock	These aquifers consist of interbedded sandstone and carbonate rocks.
Carbonate rock	Most of the carbonate rock aquifers consist of limestone. Dolomite and marble are also possible local sources of water.

TABLE 4.13. BRIEF DESCRIPTIONS OF AQUIFER TYPES IN THE UNITED STATES GEOLOGICAL SURVEY NATIONAL WATER QUALITY ASSESSMENT STUDY [4.91] (cont.)

Aquifer type	Description
Basalt and volcanic rock	Aquifers in volcanic rocks having a wide range of chemical, mineralogical, structural and hydraulic properties. The variability of these properties is related largely to rock type and the way in which the rock was ejected and deposited.
Crystalline rock	The igneous and metamorphic rocks are permeable where fractured, and generally yield only small amounts of water to wells. However, in many places, they are the only reliable source of water supply.

The numerical results for uranium concentrations tabulated in Ref. [4.91] are presented in Fig. 4.5; the vertical bars represent the maximum, 99th percentile, 95th percentile, 90th percentile and median for each set of samples. It should be noted that the percentiles represent the percentage of samples that have a concentration below the stated value. For example, 5% of samples exceed the 95th percentile. The figure also has two horizontal dashed lines — the lower one is a typical limit of the quantification value (also referred to in this study as a ‘censor’ value) below which most laboratories did not report uranium concentrations. The second, higher line is at a value of 30 μg [U]/L, which, in this study, was taken as an HHB.

An examination of Fig. 4.5 shows that for all types of aquifer, median values were well below the HHB; indeed, for most types of aquifer, the median value of the uranium concentration was below the quantification limit. In most types of aquifer, the 99th percentile of the dataset was below the HHB, with the exceptions being the unconsolidated sand and gravel aquifers and the crystalline rock aquifers. In the unconsolidated sand and gravel aquifers, nearly 9% of samples exceeded the HHB (Table 4.14). However, the maximum measured value exceeded the HHB in almost every type of aquifer. This extensive dataset permits the following conclusions to be drawn:

- Patterns of uranium concentration in groundwaters are related to the host rock and type of aquifer;
- In almost all aquifer types, there is a possibility that individual samples exceed the HHB;
- Unconsolidated sand and gravel aquifers and crystalline rock aquifers are most likely to yield samples exceeding HHB values.

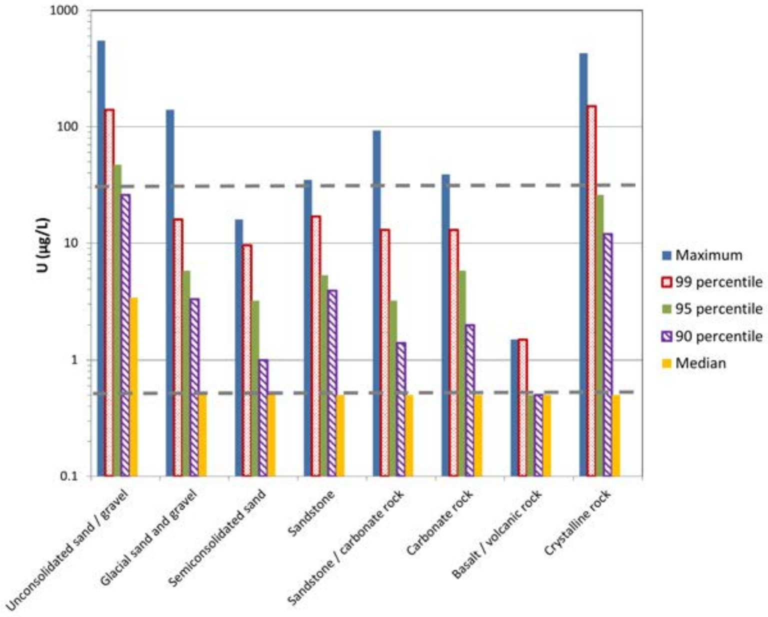

FIG. 4.5. Graphical representation of uranium data for groundwater in the USA (the data are from Ref. [4.91]). The vertical bars represent the percentiles and median values of the data as shown in the legend. The lower horizontal dashed line is the quantification limit. The higher dashed line is the human health benchmark value of 30 µg [U]/L.

The high percentage of unconsolidated sand and gravel aquifer samples exceeding the HHB was discussed in some detail in Ref. [4.91]. The water chemistry in this aquifer group was predominantly of the calcium–magnesium bicarbonate type, with a median pH of 7.3. The majority of the groundwaters were described as oxic, which is favourable for uranium mobility. Concentrations of uranium as well as of other trace elements (arsenic, boron, lithium, molybdenum, selenium and vanadium) were generally greater in the unconsolidated sand and gravel aquifers than in any other aquifer groups. About 9% of water samples in this aquifer group had uranium concentrations above the EPA drinking water standard, set to an HHB value of 30 µg [U]/L (see Table 4.14). This is the greatest rate among all the aquifer groups for uranium and accounts for about 95% of all uranium HHB exceedances in the dataset. Many of these samples had relatively high groundwater pH, and, in many cases, several oxyanionic and ion complexes, which can enhance the solubility of trace elements, were present.

Although about 4% of uranium concentrations in all groundwater samples exceeded the HHB, there were even greater levels of exceedances for some trace

TABLE 4.14. PERCENTAGE EXCEEDANCES OF HUMAN HEALTH BENCHMARK VALUES FOR URANIUM IN AQUIFERS IN THE USA [4.91]

Aquifer type	No. of samples	Percentage exceeding the EPA human health benchmark of 30 µg [U]/L
Unconsolidated sand and gravel	1409	8.7
Glacial unconsolidated sand and gravel	640	0.5
Semi-consolidated sand	339	0
Sandstone	336	0.3
Sandstone and carbonate rock	187	0.5
Carbonate rock	318	0.3
Basalt and volcanic rock	33	0
Crystalline rock	279	3.9
Total	3541	3.9

Note: EPA: United States Environmental Protection Agency.

elements, including manganese (12% above HHB), arsenic (7% above HHB) and strontium (4.3% above HHB). In fact, groundwater in about 19% of all wells (962 of 5097) exceeded an HHB for at least one trace element. The largest number of exceedances were for the radioactive gas radon, which occurred at concentrations greater than the EPA proposed maximum contaminant level of 11 Bq/L in 65% of samples. This major study of groundwaters in the USA highlights the major impact of host aquifer type and groundwater chemistry in determining groundwater uranium concentrations, as well as those of other trace elements.

In Canada, naturally occurring uranium concentrations in groundwaters are generally low (typically <1 µg/L) but may vary considerably, with much higher levels in some private and community wells [4.92]. Uranium concentrations for groundwater samples collected in various locations in British Columbia ranged from 0.30 to 0.50 µg/L, with an overall average of 0.38 µg/L. Groundwater collected within the vicinity of the Blizzard uranium deposit contained from 0.65 to 85 µg [U]/L, with a mean value of 18.79 µg [U]/L. Groundwater was evaluated for uranium isotopes in a region of Nova Scotia with naturally elevated concentrations of uranium in the bedrock. Concentrations were typically under 1.0 µg [U]/L (85%) and ranged from 0.024 to 41 µg [U]/L. Uranium concentrations in groundwater collected around the Prairie Flats surficial uranium deposit ranged from 9 to 3961 µg [U]/L. Data for total uranium in numerous water samples from groundwater supply wells in Ontario ranged from 0.005 to 11.5 µg [U]/L, with an overall mean of 0.8 µg [U]/L. Uranium concentrations in groundwater collected in a regional study area in Ontario ranged from <0.2 to 73.0 µg [U]/L. In general, the Canadian survey showed that uranium concentrations <1 µg [U]/L are commonly encountered in drinking water but drinking water concentrations from regions with known uranium deposits or generally elevated background uranium concentrations can often be orders of magnitude higher [4.92]. The overall conclusions from the Canadian study are similar to those of the more detailed United States study [4.91].

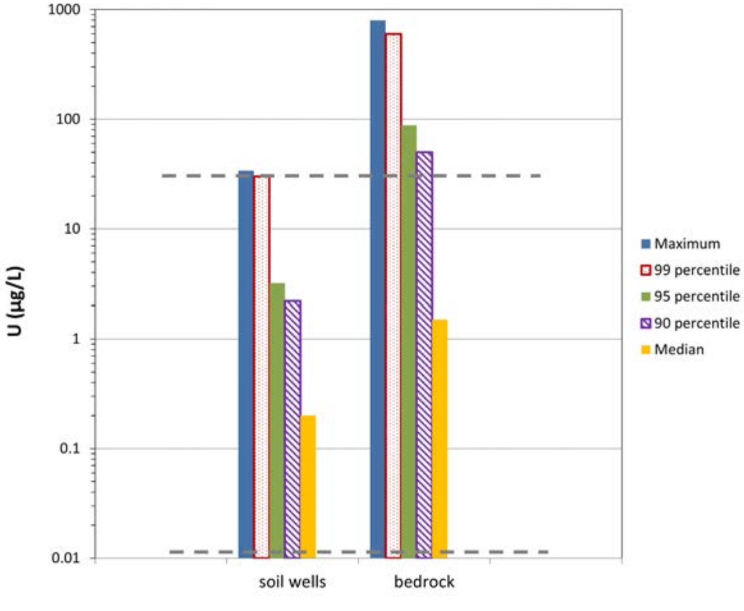

FIG. 4.6. Graphical representation of uranium data for groundwater from soil wells and bedrock wells in Finland (the data are from Ref. [4.93]). The vertical bars represent the percentiles and median values of the data as shown in the legend. The lower horizontal dashed line is the quantification limit. The higher dashed line is the EPA human health benchmark value of 30 µg [U]/L [4.91].

A survey of radioactivity in drinking water from private wells in Finland [4.93] has shown that the uranium content of waters from wells drilled in bedrock was much higher than from wells dug in the soil. The data are presented in Fig. 4.6. As can be seen, more than 10% of samples from wells drilled in bedrock exceeded the EPA HHB value for drinking water of 30 μg [U]/L. Although the ^{238}U levels are of some concern, the bulk of the effective dose from the groundwaters arose from ^{222}Rn. The range of results for the Finnish bedrock samples is similar to (but slightly higher than) the range for samples described as ‘crystalline rock samples’ in the United States study [4.91]. In the Finnish study, more than 10% of samples exceeded the HHB values.

4.3.2. Variations in aquifer characteristics due to uranium anomalies

Given the discussion in the previous sections, some caution needs to be expressed in assessing whether a reported uranium value is anomalous — typically, there is a wide range of natural concentrations. In some contexts, a concentration is significant if a stated concentration limit is exceeded, for example the World Health Organization (WHO) provisional guideline value for uranium in drinking water of 30 μg/L (see Section 7.4 for a discussion). Anomalous uranium concentrations cannot always be assumed to be due to anthropogenic contamination.

A typical reason for a natural uranium anomaly is the presence of an accumulation of uranium, which may lead to local impacts (within a few tens of metres) but, in some cases, can cause a much more widespread groundwater signature, depending on the geochemical environment. One example is the Koongarra uranium deposit in northern Australia (see Chapter 3). Figure 4.7 [4.94] shows uranium concentrations in groundwater across the site. The direction of groundwater movement is from left to right. There is a substantial elevation of uranium concentrations when waters intersect the deposit, which is hosted in a zone of weathered schists (Cahill formation), adjacent to a fault bounding the local sandstone (Kombolgie formation). As can be observed, there is a significant elevation of groundwater uranium concentrations within and immediately downgradient of the ore zone, but uranium concentrations return to background levels within a few hundred metres of the uranium deposit [4.95].

4.3.3. Fractionation of uranium isotopes in groundwaters

Given sufficient time in a closed system, the activity of ^{234}U in groundwater matches that of its parent ^{238}U, a state which is known as ‘secular equilibrium’. However, it is common for natural groundwaters to have ^{234}U:^{238}U isotope ratios exceeding unity [4.5, 4.96]. There are several processes that may lead to excesses

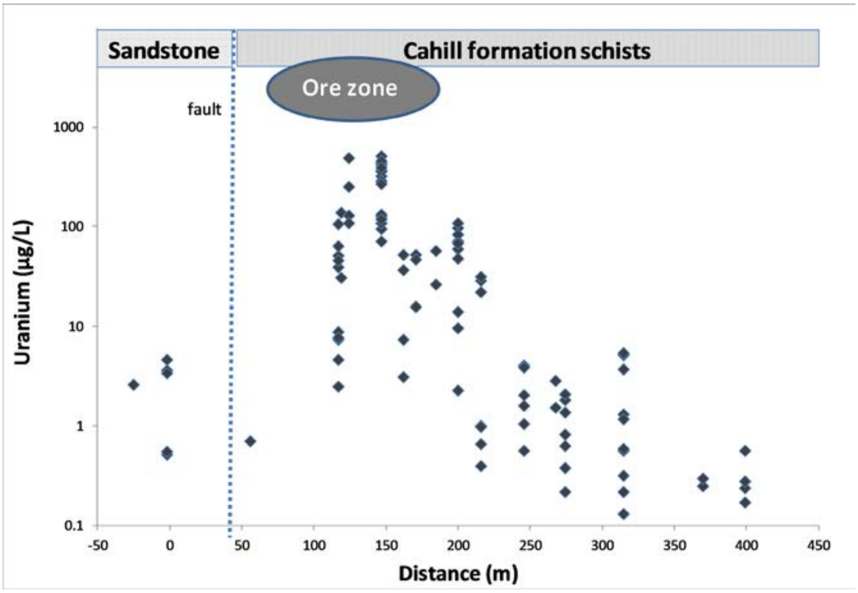

FIG. 4.7. Uranium concentration in groundwater sampled across a transect through the Koongarra uranium deposit, Australia (the data are from Ref. [4.94]).

of ^{234}U relative to its parent, ^{238}U, in groundwater. These mechanisms have been categorized as follows [4.97]:

(a) Lattice damage caused by radioactive decay;
(b) Location of daughter atoms in weakly bound or interstitial sites;
(c) Oxidation state change from +4 to +6 induced by decay and leading to greater solubility of the progeny;
(d) Direct alpha recoil from the solid into the aqueous phase.

The process of alpha recoil is caused by the sudden movement of the progeny nucleus when an energetic alpha particle is emitted. This displacement is estimated to be approximately 20 nm [4.98]. Thus, if the parent ^{238}U atom is close to the mineral surface, the progeny ^{234}Th can recoil directly into the surrounding aqueous phase. Upon decay to ^{234}U, the progeny would remain in surface or solution phases to a greater extent than their ancestor ^{238}U. Although ^{234}U:^{238}U ratios above unity are commonly found in groundwaters, there are a few examples of lower ^{234}U:^{238}U ratios. One example is groundwaters from the weathered zone of the Koongarra deposit in Australia [4.94, 4.99]. It is illustrative to compare data for ^{234}U:^{238}U ratios in groundwater for a range of geological environments. Such a comparison is made in Fig. 4.8, which shows the uranium concentrations and ^{234}U:^{238}U ratios for numerous samples from different regions.

The first set of data shown is for the Blumau gravel aquifer, Austria [4.100]. These samples (indicated by blue diamonds in Fig. 4.8) fall into a distinct field of relatively low uranium content and elevated ^{234}U:^{238}U ratios. A contrasting set of data is numerous samples from the vicinity of the Koongarra uranium deposit in Australia [4.94]. A number of these samples (indicated by green triangles in Fig. 4.8) have very high dissolved uranium contents. Overall, this dataset exhibits ^{234}U:^{238}U ratios quite close to unity, as would be expected given the proximity of the uranium deposit, containing abundant relatively soluble uranium minerals. However, as noted above, many of the groundwater samples from this deposit have ^{234}U:^{238}U ratios slightly below unity and this is the only dataset on the diagram containing samples with ratios below the horizontal line indicating a ^{234}U:^{238}U ratio of one.

A very interesting dataset in Fig. 4.8 shows the samples from the Tatsunokuchi hot springs in Japan [4.101]. Here the uranium concentrations are relatively low, but extremely high isotope ratios of up to around 50 were reported (indicated by orange circles in Fig. 4.8). This confirms the general trend of higher ^{234}U ratios with lower uranium concentrations. An interesting feature of this dataset is that all the points fall on a similar trend line, which was interpreted

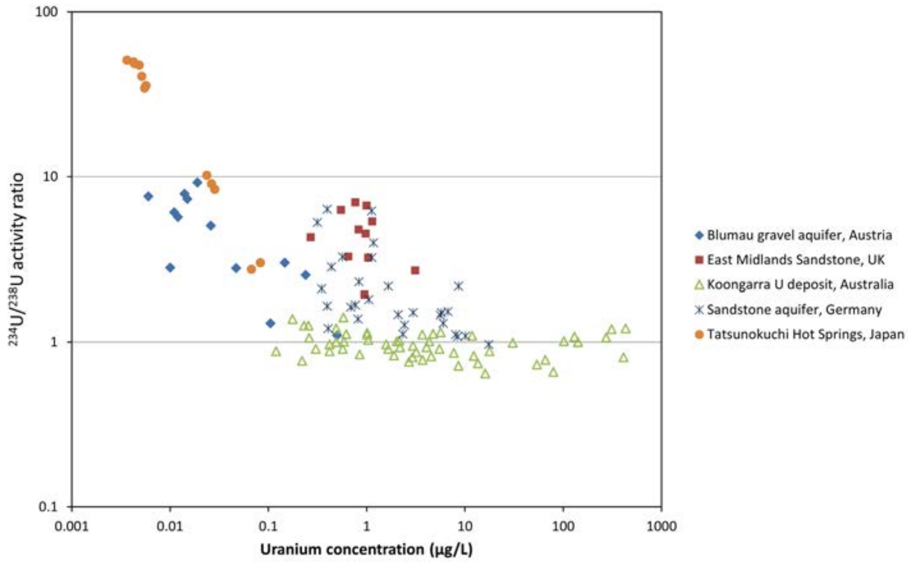

FIG. 4.8. Uranium isotope ratios for a diverse range of groundwaters from different geological environments (the data are from Refs [4.94, 4.100–4.102]).

by the authors as reflecting mixing between two water sources, one having a very low ^{234}U:^{238}U ratio and a lower uranium content. This explains the strong correlation seen in this dataset.

The data for the East Midlands sandstone aquifer in the United Kingdom [4.100] fall in a relatively narrow field of moderately low uranium content and somewhat elevated ^{234}U:^{238}U ratios (indicated by red squares in Fig. 4.8). This is typical of a drinking water aquifer, and the uranium content probably reflects the host geology and geochemistry, where the water may be slightly oxidizing but nevertheless the sandstone is not enriched to a significant extent in uranium

Finally, the sandstone aquifer in Germany [4.102] shows a range of uranium concentrations and ^{234}U:^{238}U isotope ratios (indicated by blue crosses in Fig. 4.8). The elevated uranium concentrations in some samples may reflect the fact that the sample region contains some uranium rich geology. As with the other datasets, samples with lower uranium content tend to exhibit higher ^{234}U:^{238}U ratios.

Thus, the datasets reflect distinct geological environments, and it can be seen that each set of samples occupies an identifiable field on the diagram. The overall trend in Fig. 4.8 is for samples with higher uranium content, particularly those influenced by the presence of uranium deposits, to exhibit uranium isotopic ratios near equilibrium. This reflects the results of uranium mineral dissolution processes. It is also common for samples from aquifers of lower uranium content to have higher ^{234}U:^{238}U ratios, due to enrichment of daughter isotopes in the solution phase caused by processes such as alpha recoil and mineral lattice damage. Thus, a general trend of increasing ^{234}U:^{238}U ratios with decreasing uranium concentration is evident in Fig. 4.8.

Uranium in groundwater can arise from natural sources because of the release of natural uranium from uranium-bearing materials such as rocks, soil, ore bodies and other source materials. The concentration of uranium (and its decay products) in percolating groundwater is influenced by the amount and availability of the nuclides in the geological materials in contact with the groundwater, as well as by the groundwater chemistry. Parameters such as the groundwater acidity (pH), the presence of ligands (such as carbonate) and the prevailing redox conditions have a major influence on the mobility of uranium (as discussed in Section 3.3). These parameters are highly variable and, therefore, natural groundwater ^{238}U concentrations vary across a wide range. However, there are some geological regions where natural groundwater ^{238}U concentrations are consistently elevated, which is usually due to available uranium in the formation rocks, often in combination with specific types of groundwater chemistry.

Uranium radioisotopes and other naturally occurring radionuclides can also be released through the exploitation of radioactive minerals (e.g. those containing uranium and thorium) as part of the nuclear fuel cycle. The mining and processing of various other minerals of commercial importance (unrelated to the nuclear fuel cycle) can also result in elevated concentrations of uranium and its decay products in local groundwaters, owing to the presence of uranium in the minerals. Typical examples include phosphate minerals (such as apatite), copper, gold, coal and many other types of ore, with which uranium has a geochemical association.

In situ uranium leaching processes may result in the development of contaminated water plumes that extend beyond the boundary of producing well fields [4.103]. During in situ mining, well patterns are intended to create a controlled flow regime for the lixiviant, thus minimizing dispersion losses and the contamination of surrounding aquifers. Nevertheless, various examples of contaminated groundwater have been reported near in situ uranium mining operations [4.104]. In some cases, other contaminants (apart from uranium) are also present, including arsenic, manganese, molybdenum, nitrate, radium, selenium and sulphate, which may be derived from natural sources in rocks or chemicals added in the mining process.

Other sources of natural and anthropogenic uranium contamination often encountered in groundwater studies include, but are not limited to, natural geothermal springs, geothermal energy production, waste disposal, water treatment sludges and tailings dams.

4.4. URANIUM IN FRESHWATER ENVIRONMENTS

4.4.1. Surface waters and rainfall

Uranium in fresh water enters surface waters primarily through dissolution and natural processes of erosion of rock and soil [4.105]. In areas with naturally elevated uranium concentrations — for example, in areas with ore deposits — anthropogenic activities, such as mining, may be undertaken that can lead to releases of uranium to freshwater environments. Uranium can enter surface waters through discharge of aquatic effluents and stack emissions from uranium processing facilities, as well as through releases from waste rock, uranium mill tailings and other mining by-products [4.106]. Dust particles originating from unconfined tailings in the terrestrial environment and from stack emissions can be wind blown and, subsequently, deposited or washed into water bodies, potentially contributing uranium. In addition, treated aquatic effluents can be discharged directly into surface waters from uranium mills. Soluble and insoluble forms of uranium can also be released to the atmosphere from uranium refining and conversion plants and discharged to aquatic environments as effluents through municipal sanitary sewer systems [4.107]. Such pathways can lead to increased concentrations of uranium in aquatic systems relative

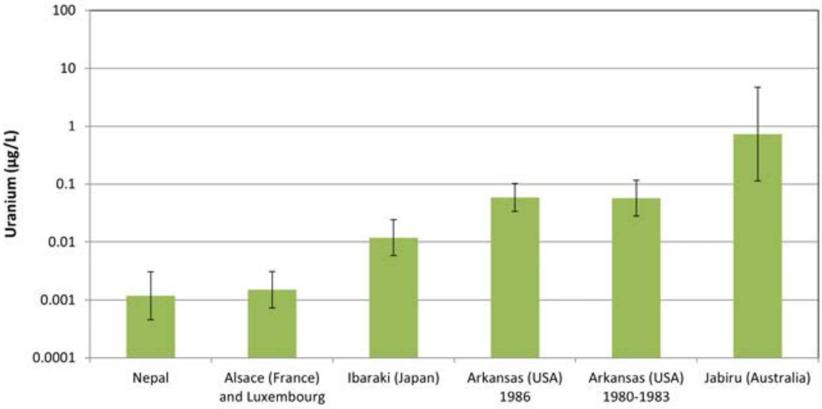

FIG. 4.9. Uranium concentrations in uncontaminated and contaminated rainfall for different sites (the data are from Refs [4.108–4.113]). The period of measurements (years) is given for Arkansas, USA.

to baseline values. In general, rainfall contains only a low concentration of uranium, although higher concentrations can occur, notably due to the capture of dust particles with significant contents of uranium. Typical and enhanced concentrations are illustrated in Fig. 4.9.

The data for Nepal [4.108] and France (Alsace) [4.109] illustrate concentrations in uncontaminated rainfall from remote rural and industrial contexts, respectively. The data from Ibaraki, Japan were interpreted by the authors of the study as showing no impact from a (then) recent nuclear accident in the former Soviet Union [4.110]. The data from Arkansas, USA [4.111, 4.112] demonstrate effects from the 18 May 1980 eruption of Mount St. Helens, USA and the 28 March 1982 eruption of El Chichón, Mexico. These volcanic materials seem to have stayed airborne for several years and have profoundly affected the global atmospheric inventories of thorium and uranium isotopes. For Jabiru, Australia, the measured activity concentrations of ^{238}U and ^{234}U (as well as other radionuclides) were due to below cloud washout of dust transported from the nearby Ranger uranium mine [4.113].

In summary, uncontaminated rainfall appears to have a uranium concentration of around 1–10 ng/L (about two orders of magnitude lower than the concentration in most surface waters), although even this small amount may be largely contributed by entrained dust. Slightly higher values have been attributed to contamination from volcanic dust. Even so, uranium concentrations in rainwater are clearly among the lowest in any environmental medium. In the cases of rainfall affected by serious storms entraining dust from a nearby major uranium mine, much higher values were obtained.

The data in Tables 4.15 and 4.16 [4.114–4.160] provide mean concentrations of uranium in fresh surface waters in background areas and mining areas, respectively. Uranium concentrations in fresh waters can range from 0.081 to 6200 µg/L in different geographical areas of the world where mining is undertaken, as indicated by the data included in Table 4.16. Peak uranium concentrations were measured at mining sites in Brazil (3000 ± 2200 µg/L), with similar values reported in mining waters from the Czech Republic (8800 ± 4100 µg/L). Receiving waters in mining areas showed lower uranium concentrations compared with those at mine sites. By comparison, baseline uranium concentrations in fresh waters not impacted by anthropogenic activities can range from 0.008 1 to 45 µg/L globally, as indicated by the data considered (see Table 4.15).

4.4.2. Sediments

The distribution and behaviour of uranium in sediments are influenced by many factors, including the physicochemical conditions in the water, basin bathymetry and water flow patterns, sediment size distributions, the mineral composition of the sediments, and the types of uranium ion complex present (i.e. uranium speciation). The presence of organic substances also plays an important role in uranium accumulation in sediments [4.161–4.163] and, within the sediments, biotic processes, such as microbial activity, can affect environmental fate [4.106]. Such factors can result in relatively large variability in uranium concentrations in sediments, as well as affect its distribution in freshwater ecosystems.

Uranium has a strong tendency to partition into sediments, as indicated by its relatively high sediment:water partition coefficient, K_d [4.164, 4.165]. Such partitioning is dictated by precipitation and sorption reactions, both of which affect the proportion of uranium that is bioavailable, through its removal from the aqueous phase [4.106]. Since chemical reactions in aquatic systems primarily occur at the sediment–water interface, sorption can have a significant influence on the fate of uranium in freshwater environments [4.166]. For example, uranium sorption onto sediment particles can influence surface reactivity and subsequent rates of precipitation [4.166].

The cation exchange capacity of different types of sediment allows reversible binding to occur at surface exchange sites [4.167]. At low pH, hydrogen ions can compete for surface exchange sites, thereby reducing

Text cont. on p. 129.

TABLE 4.15. CONCENTRATION OF URANIUM IN FRESH SURFACE WATERS IN REFERENCE AREAS NOT INFLUENCED BY URANIUM SITES

Region and country	Description	*N*[a]	Mean uranium concentration ± standard deviation (minimum–maximum) (μg/L)[b]	Reference
Africa				
Democratic Republic of the Congo	River	1	0.080	[4.114]
Mozambique	River	1	0.26	[4.115]
South Africa	Rivers	2	1.8 ± 1.4 (0.79–2.8)	[4.115]
Asia				
Bangladesh	Rivers	11	1.5 ± 1.3 (0.10–4.9)	[4.108]
China	Rivers	2	4.3 ± 4.5 (1.1–7.5)	[4.108]
India	Rivers	7	1.2 ± 1.5 (0.25–4.5)	[4.116, 4.117]
Kazakhstan	River	1	7.8 ± 0.4	[4.118]
Korea, Rep. of	Rivers	42	0.23 ± 0.14 (0.020–0.47)	[4.119]
Kyrgyzstan	Reservoir	1	1.9	[4.120]

TABLE 4.15. CONCENTRATION OF URANIUM IN FRESH SURFACE WATERS IN REFERENCE AREAS NOT INFLUENCED BY URANIUM SITES (cont.)

Region and country	Description	N^a	Mean uranium concentration ± standard deviation (minimum–maximum) (μg/L)[b]	Reference
Kyrgyzstan	Lake, rivers	16	4.7 ± 2.4 (1.9–9.3)	[4.120–4.122]
Nepal	Rainwater	10	2.4 ± 3.8 (0.60–13)	[4.108]
Nepal and Bangladesh	Rivers, streams	62	3.6 ± 4.4 (0.030–18.5)	[4.108]
Pakistan	River	1	4.9	[4.108]
Tajikistan	Rivers Syr-Darya river	6 3	6.6 ± 3.1 (3.4–9.7) 20.1 (±5.2)	[4.118, 4.123, 4.124]
Tajikistan	Lake, river	2	33 ± 14 (17–45)	[4.118]
Uzbekistan	Rivers	10	2.5–7.8	[4.123, 4.125]
Australia				
Australia	Billabong (filtered water)	7–11	0.017[c]	[4.126]
Australia	Billabong (unfiltered water)	7–11	0.022[c]	[4.126]

TABLE 4.15. CONCENTRATION OF URANIUM IN FRESH SURFACE WATERS IN REFERENCE AREAS NOT INFLUENCED BY URANIUM SITES (cont.)

Region and country	Description	N^a	Mean uranium concentration ± standard deviation (minimum–maximum) (µg/L)^b	Reference
Europe				
Czech Republic	River, stream	2	13 ± 5.9 (8.6–17)	[4.127, 4.128]
France	Rivers	4	0.87 ± 0.76 (0.37–2.0)	[4.108, 4.128]
Germany	Surface waters	4	0.90 ± 1.1 (0.20–2.5)	[4.129–4.132]
Poland	River	1	0.72	[4.133]
Portugal	Reference rivers	28	0.65 ± 0.43 (0.096–1.9)	[4.134]
Spain	Reference river	1	0.75	[4.135]
Sweden	Reference river	1	0.17	[4.117, 4.133, 4.136–4.138]
Ukraine	Rivers	15	1.4 ± 0.9 (0.8–2.0)	[4.139–4.141]
United Kingdom	Reference rivers	4	0.30 ± 0.41 (0.040–0.92)	[4.142, 4.143]
North America				

TABLE 4.15. CONCENTRATION OF URANIUM IN FRESH SURFACE WATERS IN REFERENCE AREAS NOT INFLUENCED BY URANIUM SITES (cont.)

Region and country	Description	*N*[a]	Mean uranium concentration ± standard deviation (minimum–maximum) (µg/L)[b]	Reference
Canada	Rivers	320	0.19 ± 0.18 (0.008 1–1.3)	[4.108, 4.144]
USA	Rivers, streams	7	0.43 ± 0.61 (0.022–1.6)	[4.117, 4.137, 4.145, 4.146]
South America				
Brazil	Reference river	1	0.040	[4.146]

[a] Number of fresh surface water sites.

[b] The calculator in Ref. [4.147] was used to correct for unequal sample sizes.

[c] Uranium was estimated from ^{238}U by applying a specific activity value of 12.3 Bq/mg (from Ref. [4.1]).

TABLE 4.16. CONCENTRATION OF URANIUM IN FRESH SURFACE WATERS IN AREAS INFLUENCED BY URANIUM SITES

Region and country	Description	*N*[a]	Mean U concentration ± standard deviation (minimum–maximum) (µg/L)[b]	Reference
Africa	—[c]			
Asia				
Kazakhstan	Lakes and rivers in mining areas	25	150 ± 440 (7.9–1 900)	[4.148]
Kyrgyzstan	Rivers in mining areas	4	5.1 ± 1.5 (3.6–7.1)	[4.120]
Kyrgyzstan	Rivers in mining areas	4	9.9 ± 6.6 (2.8–24.2)	[4.120, 4.149, 4.150]
Kyrgyzstan	Mining waters inlet to the Minkush river	5	103 ± 48 (48–337)	[4.149]
Kyrgyzstan	Drainages from tailings inlet to the Mailuu-Suu river	4	5 427 ± 3 409 (1 715–8 417)	[4.121, 4.150]
Tajikistan	Lakes and rivers in tailing areas Digmai	6	8.8 ± 8.6 (2.0–22.6)	[4.150, 4.151]

TABLE 4.16. CONCENTRATION OF URANIUM IN FRESH SURFACE WATERS IN AREAS INFLUENCED BY URANIUM SITES (cont.)

Region and country	Description	*N*[a]	Mean U concentration ± standard deviation (minimum–maximum) (µg/L)[b]	Reference
Tajikistan	Lakes and rivers in mining areas Taboshar	7	1 700 ± 490 (1 100–2 400)	[4.149]
Tajikistan	Creek in tailing areas Taboshar	3	(5 500–68 600)	[4.149]
Uzbekistan	Rivers in mining areas	2	27.7 ± 5.1 (24.1–31.3)	[4.149]
Uzbekistan	Rivers in mining and tailing areas	2	256 ± 82 (167–326)	[4.149]
Australia				
	Billabongs (filtered water)	21–33	0.28 ± 0.42 (0.037–0.76)[d]	[4.126]
	Billabongs (unfiltered water)	21–33	0.92 ± 1.4 (0.081–2.5)[d]	[4.126]
Europe				

TABLE 4.16. CONCENTRATION OF URANIUM IN FRESH SURFACE WATERS IN AREAS INFLUENCED BY URANIUM SITES (cont.)

Region and country	Description	*N*[a]	Mean U concentration ± standard deviation (minimum–maximum) (μg/L)[b]	Reference
Czech Republic	Mine waters after flooding	2	8 800 ± 4 100 (5 900–12 000)	[4.127]
Czech Republic	Rivers and streams in mining area	10	83 ± 110 (4.3–330)	[4.127]
Germany	Dump water	2	25 000 ± 7 100 (20 000–30 000)	[4.152, 4.153]
Germany	Mine water	7	2 300 ± 2 700 (11–6 400)	[4.153–4.155]
Germany	Surface waters in mining areas	2	92 ± 170 (6.9–600)	[4.129, 4.130,4.132, 4.152, 4.153]
Spain	Rivers near potash mining	82	5.8 ± 4.2 (1.6–21)[d]	[4.135]
Ukraine	Surface waters in mining areas	10	28 ± 17 (16–61)	[4.139, 4.156, 4.157]
Ukraine	Dump water	75	2 000 ± 2 500 (220–72 500)	[4.149, 4.158]
North America				

TABLE 4.16. CONCENTRATION OF URANIUM IN FRESH SURFACE WATERS IN AREAS INFLUENCED BY URANIUM SITES (cont.)

Region and country	Description	*N*[a]	Mean U concentration ± standard deviation (minimum–maximum) (μg/L)[b]	Reference
USA	Stream near uranium mine (unfiltered water)	6	13 000 ± 19 000 (3300–52 000)	[4.145]
South America				
Brazil	Lakes in mining area	38	3 000 ± 2 200 (41–6 200)[d]	[4.159, 4.160]

[a] Number of individual replicate data points.

[b] The calculator in Ref. [4.147] was used to correct for unequal sample sizes.

[c] —: no data available.

[d] Uranium was estimated from ^{238}U by applying a specific activity value of 12.3 Bq/mg (from Ref. [4.1]).

sorption of uranyl ions (UO_2^{2+}) [4.106, 4.168]. At high pH, sorption also tends to decrease, although there is more variability in this relationship than at the lower end of the pH scale [4.169, 4.170]. Changes in ionic strength can also affect sorption of uranyl ions to mineral surfaces owing to competition with cations for exchange sites [4.170].

Owing to the formation of ternary complexes at the junctions between a mineral surface, an organic ligand (e.g. humic or fulvic acid) and the metal ion, the presence of organic ligands can result in increased adsorption of the uranyl ion to mineral substrates (e.g. haematite) at low pH and some degree of decreased adsorption at high pH [4.169]. Such trends are strong for some minerals, such as haematite, but can vary for other types of mineral.

The highest concentrations of uranium have been reported to occur in sedimentary rocks containing organic substances, owing to adsorption through the process of ion exchange or formation of chemical compounds [4.171].

Although information is available on the processes of sorption and precipitation, relatively less is known about uranium release from contaminated sediments, and detection limits can be an issue in quantification of releases from sediments to the water column (see Ref. [4.172]).

Tables 4.17 and 4.18 provide mean concentrations of uranium in freshwater sediments in background areas and mining areas, respectively. Uranium concentrations in freshwater sediments in different geographical areas of the world where mining is undertaken can range from 0.15 to 3 024 mg/kg, dry weight (DW), according to the data included in Table 4.18. High uranium concentrations in sediment (mean values) were measured in mining areas of the following countries:

- Germany (373 ± 470 mg/kg, DW);
- Kyrgyzstan (181 ± 39 mg/kg, DW);
- Portugal (176 ± 405 mg/kg, DW).

The lowest concentrations were reported in sediments collected in mining sites in the USA, with a mean of 8.6 ± 1.7 mg/kg, DW. Intermediate uranium concentrations have been reported in areas of uranium mining in the following countries:

- Czech Republic (122 ± 95 mg/kg, DW);
- Kazakhstan (56 ± 105 mg/kg, DW);
- Slovenia (52 ± 85 mg/kg, DW).

By comparison, baseline uranium concentrations in freshwater sediments not impacted by anthropogenic activities range from 0.5 to 45 mg/kg, DW, globally according to the data in Table 4.17.

4.4.3. Freshwater biota

Uranium bioavailability can be defined as its ability to bind to (adsorb) or cross the cell surface of an organism [4.184]. For example, available literature indicates that uranium can adhere to the external surface of fish gills and can be taken up internally across the gills [4.185, 4.186]. Uranium can also be absorbed across the gastrointestinal tract [4.187]. Uranium absorption across the gastrointestinal tract is influenced by the solubility of the compound and, generally, increases with increasing solubility. In this way, diet and/or sediment pathways represent possible routes of uptake, which may vary with feeding strategy. Nevertheless, assimilation efficiencies across the gastrointestinal tract are likely low, even for relatively soluble uranium compounds, resulting in low trophic transfer [4.188–4.190].

The physicochemical form of uranium (or its speciation) affects its bioavailability, and a variety of physicochemical forms of uranium of varying sizes (molecular masses) and charge properties (each representing a different species of uranium) can occur in freshwater systems [4.191]. These include the free metal ion (U^{4+} or UO_2^{2+}), complexes with inorganic ligands (e.g. uranyl phosphate) and complexes with humic substances (e.g. uranyl fulvate or humate), which can occur in dissolved, colloidal and/or particulate forms. Low molecular mass forms are generally considered to be mobile and bioavailable, whereas colloids or high molecular mass forms are less mobile [4.191]. Available information suggests that the major forms of bioavailable uranium are UO_2^{2+} and UO_2OH^+ [4.106, 4.184]. Nevertheless, for assessment purposes, potential impacts from uranium are typically assessed based on total uranium concentration, representing a conservative approach, as it is assumed that all uranium is present in a bioavailable form.

Factors such as the concentration of major cations (calcium and magnesium), phosphate and organic ligands, can limit accumulation of uranium by freshwater primary producers. In addition, depending on the plant species, adsorption onto cell surfaces may represent the dominant mechanism of accumulation by freshwater primary producers [4.192]. Such processes are driven by the form in which the uranium is found, the physicochemical conditions in the water body and the surface area:volume ratio of the plant species under consideration. Examples of uranium concentration data (mg/kg, FW) in fresh water and wetland vegetation

Text cont. on p. 140.

TABLE 4.17. CONCENTRATION OF URANIUM IN FRESHWATER SEDIMENTS IN REFERENCE AREAS NOT INFLUENCED BY URANIUM MINING SITES

Region and country	Sample type	*N*[a]	Mean ^{238}U activity concentration ± standard deviation (minimum–maximum) (Bq/kg, dry weight)[b]	Mean uranium mass concentration ± standard deviation (minimum–maximum) (mg/kg, dry weight)[c]	Reference
Asia					[4.120]
Kyrgyzstan	Sediment[d]	1	(3–383)	275	[4.120]
Kyrgyzstan	Sediment	—[e]	(65–120)	(5–10)	[4.173]
Tajikistan	Sediment	8	42 ± 8 (28–50)	3.4 ± 0.6 (2.3–4.0)	—[f]
Australia					
Australia	Suspended particulates	20	43 ± 22 (7.9–107)	3.5 ± 1.8 (0.64–8.7)	[4.174]
Europe					
Czech Republic	Sediment	1	555	45	[4.127]
Portugal	Sediment	1	231	19	[4.134]
Portugal	Soil	1	230	19	[4.175]

TABLE 4.17. CONCENTRATION OF URANIUM IN FRESHWATER SEDIMENTS IN REFERENCE AREAS NOT INFLUENCED BY URANIUM MINING SITES (cont.)

Region and country	Sample type	*N*[a]	Mean ^{238}U activity concentration ± standard deviation (minimum–maximum) (Bq/kg, dry weight)[b]	Mean uranium mass concentration ± standard deviation (minimum–maximum) (mg/kg, dry weight)[c]	Reference
Slovenia	Sediment	1	35	2.8	[4.176]
United Kingdom	Sediment	8	28 ± 8.3 (16–39)	2.3 ± 0.7 (1.3–3.2)	[4.177]
North America					
USA	Sediment	22	36 ± 34 (5.9–152)	3.0 ± 2.7 (0.5–12)	[4.145, 4.178]
USA	Sediment leachate	3	12 ± 0.25 (11.9–12.4)	1.0 ± 0.02 (0.97–1.0)	[4.145]

[a] Number of individual replicate data points.

[b] The calculator in Ref. [4.147] was used to correct for unequal sample sizes.

[c] Uranium was estimated from ^{238}U by applying a specific activity value of 12.3 Bq/mg from Ref. [4.1]).

[d] The high sediment concentrations suggest the possibility of contamination.

[e] —: no data available.

[f] —: Data from IAEA regional project RER/9/086, Safe Management of Residues from Former Mining and Milling Activities in Central Asia (2005–2009).

TABLE 4.18. CONCENTRATION OF URANIUM IN FRESHWATER SEDIMENTS IN AREAS INFLUENCED BY URANIUM MINING SITES

Region and country	Sample type	*N*[a]	Mean ^{238}U activity concentration ± standard deviation (minimum–maximum) (Bq/kg, dry weight)[b]	Mean uranium mass concentration ± standard deviation (minimum–maximum) (mg/kg, dry weight)[c]	Reference
Asia					
Kazakhstan	Sediment	17	683 ± 1 287 (28–4 080)	56 ± 105 (2.28–332)	[4.179, 4.180]
Kyrgyzstan	Sediment	4	2 226 ± 482 (1 538–2 558)	181 ± 39 (125–208)	[4.120]
Kyrgyzstan	Sediment	—[d]	100–400 (maximum: 2 200)[e]	8–32	[4.173]
Tajikistan	Sediment	1	2 500	203	
Tajikistan	Sediment	10	926 ± 866 (267–3 120)	75 ± 70 (22–251)	—[f]
Tajikistan	Sediment (tailings material)	3	25 880 ± 10 240 (18 000–37 500)	2 087 ± 826 (1 452–3 024)	—[f]
Europe					
Czech Republic	Sediment	5	1 496 ± 1 166 (62–2 854)	122 ± 95 (5.0–232)	[4.127]

TABLE 4.18. CONCENTRATION OF URANIUM IN FRESHWATER SEDIMENTS IN AREAS INFLUENCED BY URANIUM MINING SITES (cont.)

Region and country	Sample type	*N*[a]	Mean ^{238}U activity concentration ± standard deviation (minimum–maximum) (Bq/kg, dry weight)[b]	Mean uranium mass concentration ± standard deviation (minimum–maximum) (mg/kg, dry weight)[c]	Reference
Czech Republic	Sediment (slightly exposed)	1	881	72	[4.127]
Germany	Sediment	21	4 588 ± 5 781 (197–24 698)	373 ± 470 (16–2 008)	[4.132]
Germany	Tailings	8	1 133 ± 1 368 (0.49–4 178)	92 ± 111 (0.04–340)	[4.132]
Portugal	Sediment	33	2 166 ± 4 979 (129–18 100)	176 ± 405 (10–1 472)	[4.175, 4.181]
Portugal	Tailings	1	2 530	206	[4.175, 4.181]
Portugal	Waste rock	1	38 316	3 115	[4.175, 4.181]
Portugal	Waste treatment plant sludge	1	41 598	3 382	[4.175, 4.181]
Slovenia	Sediment	5	641 ± 1 048 (65–2 500)	52 ± 85 (5.3–203)	[4.176]
Slovenia	Soil	1	150	12	[4.176]

TABLE 4.18. CONCENTRATION OF URANIUM IN FRESHWATER SEDIMENTS IN AREAS INFLUENCED BY URANIUM MINING SITES (cont.)

Region and country	Sample type	*N*[a]	Mean ^{238}U activity concentration ± standard deviation (minimum–maximum) (Bq/kg, dry weight)[b]	Mean uranium mass concentration ± standard deviation (minimum–maximum) (mg/kg, dry weight)[c]	Reference
Slovenia	Tailings	1	850	69	[4.176]
Ukraine	Tailings	100	257 000 ± 223 200 (9 500–560 000)	20 725 ± 18 000 (772–45 450)	[4.182]
Ukraine	Spillage (yellow cake residues)	50	36 200 ± 26 600 (5 500–3 100 000)	2 920 ± 2 145 (444–250 000)	[4.182]
Ukraine	Uranium raw materials	45	53 300 ± 37 600 (16 000–132 400)	4 300 ± 3 032 (1 290–10 677)	[4.182]
Ukraine	Soil mixed with uranium raw materials	100	8 340 ± 4 600 (150–15 600)	835 ± 272 (12–1 260)	[4.182]
North America					
USA	Sediment leachate	7	182 ± 65 (82–272)	15 ± 5.3 (6.6–22)	[4.145]

TABLE 4.18. CONCENTRATION OF URANIUM IN FRESHWATER SEDIMENTS IN AREAS INFLUENCED BY URANIUM MINING SITES (cont.)

Region and country	Sample type	*N*[a]	Mean ^{238}U activity concentration ± standard deviation (minimum–maximum) (Bq/kg, dry weight)[b]	Mean uranium mass concentration ± standard deviation (minimum–maximum) (mg/kg, dry weight)[c]	Reference
USA	Sediment	35	106 ± 21 (1.9–614)	8.6 ± 1.7 (0.15–50)	[4.183]

[a] Number of individual replicate data points.

[b] The calculator in Ref. [4.147] was used to correct for unequal sample sizes.

[c] Uranium was estimated from ^{238}U by applying a specific activity value of 12.3 Bq/mg (from Ref. [4.1]).

[d] —: no data available.

[e] The maximum activity concentrations of uranium in the bottom sediments of Issyk-Kul Lake were identified in deep sedimentation areas where coal and ash from the Kadji-Say uranium processing facilities were accumulated and became mixed with silt sediments of the lake.

[f] —: Data from IAEA regional project RER/9/086, Safe Management of Residues from Former Mining and Milling Activities in Central Asia (2005–2009).

TABLE 4.19. MASS CONCENTRATION OF URANIUM IN VEGETATION COLLECTED AT REFERENCE SITES AND AT EXPOSURE SITES IMPACTED BY URANIUM MINING

		Reference site					Exposure site					
Country	Species category	*N*[a]	Arithmetic mean (mg/kg, fresh weight)	Standard deviation	Min.	Max.	*N*[a]	Arithmetic mean (mg/kg, fresh weight)	Standard deviation	Min.	Max.	Reference
Fresh water (flowing)												
Canada	Grasses[b]	—[c]	—[c]	—[c]	—[c]	—[c]	6	0.93	0.55	0.4	1.6	[4.144]
Canada	Herbs[d]	3	0.16	0.21	0.005	0.404	—[c]	—[c]	—[c]	—[c]	—[c]	[4.144]
Canada	Vascular plants[e]	—[c]	—[c]	—[c]	—[c]	—[c]	3	0.013	0.0058	0.01	0.02	[4.144]
Ukraine[f]	Perennial grasses[g]	—[c]	—[c]	—[c]	—[c]	—[c]	12	—[c]	—[c]	0.29	0.86	[4.193]
Ukraine[f]	Submerged aquatic vegetation[h]	—[c]	—[c]	—[c]	—[c]	—[c]	10	—[c]	—[c]	0.82	4.07	[4.193]

TABLE 4.19. MASS CONCENTRATION OF URANIUM IN VEGETATION COLLECTED AT REFERENCE SITES AND AT EXPOSURE SITES IMPACTED BY URANIUM MINING (cont.)

		Reference site					Exposure site					
Country	Species category	*N*[a]	Arithmetic mean (mg/kg, fresh weight)	Standard deviation	Min.	Max.	*N*[a]	Arithmetic mean (mg/kg, fresh weight)	Standard deviation	Min.	Max.	Reference
Fresh water (lake)												
Canada	Grasses[i]	92	0.04	0.005	0.01	0.3	89	26	16	0.01	1200	[4.144]
Canada	Herbs[j]	—[c]	—[c]	—[c]	—[c]	—[c]	15	22	12	0.004	138	[4.144]
Canada	Vascular plants[k]	14	0.05	0.042	0.02	0.14	36	21	58	0.02	260	[4.144]
Ukraine[f]	Perennial grasses[g]	15	—[c]	—[c]	0.09	0.31	—[c]	—[c]	—[c]	—[c]	—[c]	[4.193]
Ukraine[f]	Submerged aquatic vegetation[h]	18	—[c]	—[c]	0.42	1.72	—[c]	—[c]	—[c]	—[c]	—[c]	[4.193]
Wetland												
Canada	Grasses[l]	—[c]	—[c]	—[c]	—[c]	—[c]	6	1.1	0.60	0.057	3.07	[4.144]
Canada	Herbs[m]	—[c]	—[c]	—[c]	—[c]	—[c]	18	33	68	2.07	297	[4.144]
Canada	Shrubs[n]	79	0.12	0.19	0.002	1.4	267	1.6	4.1	0.002	35.6	[4.144]

TABLE 4.19. MASS CONCENTRATION OF URANIUM IN VEGETATION COLLECTED AT REFERENCE SITES AND AT EXPOSURE SITES IMPACTED BY URANIUM MINING (cont.)

		Reference site					Exposure site					
Country	Species category	N^a	Arithmetic mean (mg/kg, fresh weight)	Standard deviation	Min.	Max.	N^a	Arithmetic mean (mg/kg, fresh weight)	Standard deviation	Min.	Max.	Reference

Note: Max.: maximum; Min.: minimum.

[a] Number of individual replicate data points.

[b] *Carex* spp. (exposure site).

[c] —: data not available.

[d] *Typha latifolia* (reference site).

[e] *Nuphar lutea* (exposure site).

[f] Data from the Dnieper reservoir (reference site) and Konoplyanka river in the proximity of the Prydniprovskyy Chemical Plant (historically referred to as Pridneprovsky Chemical Plant), Ukraine (exposed uranium mill site).

[g] Vegetation species included: *Phragmites australis* (Cav.) Trin., *Typha angustifolia* L., *Carex acuta* L.

[h] Vegetation species included: *Potamogeton perfoliatus* L., *Pectinatus* L., *Najas marina* L., *Myriophyllum spicatum* L., *Ceratophyllum demersum* L.

[i] *Carex* spp. (reference site and exposure site).

[j] *Typha latifolia* (exposure site).

[k] *Nuphar lutea* and *Potamogeton* spp. (reference site and exposure site).

[l] *Hordeum jubatum* and *Puccinellia nuttalliana* (exposure site).

[m] *Typha* spp. (exposure site).

[n] *Cladinia* spp., *Ledum* spp. and *Vaccinium* spp. (reference site and exposure site).

(e.g. grasses, shrubs, herbs), collected at reference sites and at exposure sites impacted by uranium mining, are presented in Table 4.19 [4.144, 4.193].

Once absorbed, uranium has a strong tendency to complex with proteins and lipids within primary producers, showing limited reversibility of absorption due to the influence of the intracellular pH [4.194]. Based on relatively limited data available for cattails (*Typha* spp.) collected under field conditions, within aquatic plants, uranium concentrations in root tissues can be elevated compared with vegetative shoots [4.195].

In temperate systems, aquatic primary producers typically undergo annual senescence (or seasonal die-off), depositing contaminants into surficial sediments through the process of cellular lysis (or breakdown of organic plant material) [4.106, 4.196].

Such contaminants, including uranium, that have been accumulated by freshwater primary producers, can be transferred to higher levels of aquatic food chains through consumption by herbivorous and omnivorous animals. Nevertheless, as already discussed, in general, uranium tends to show a relatively low assimilation efficiency and does not biomagnify (i.e. it does not successively increase in concentration at higher levels of the food chain) [4.165, 4.197, 4.198]. For example, trophic transfer rates of 1–13% have been reported in Ref. [4.199], and lower trophic level organisms have typically shown higher concentrations of uranium than those present at higher levels of the food chain [4.106]. This is consistent with results reported in Ref. [4.190], which indicate that uranium concentrations in the material passing through the gastrointestinal tract of the fish species *Danio rerio* are higher than those in the body. In addition, Ref. [4.190] reported uranium trophic transfer rates of 0.52% in *Danio rerio*, resulting in some accumulation in the digestive organs (liver and digestive tract) following dietary exposure. Therefore, based on available literature, uranium is expected to show a very low rate of uptake through the gut of many species, indicating that dietary exposure is likely not the primary route of uptake [4.106]. Instead, water borne exposures and subsequent uptake via the gills may be more important [4.198]. Thus, in long term exposure situations, it is possible that uranium may be accumulated from water, as well as from contaminated food [4.106].

Ingestion of freshwater sediments can also represent a potential route of uranium exposure, although some studies indicate that accumulation by aquatic invertebrates (e.g. *Hyalella azteca*) is primarily from the dissolved phase, as opposed to being from sediment particulates [4.200]. Desorption from sediments into the dissolved phase and subsequent speciation are influenced by the physicochemical conditions in the overlying water column and, in particular, pH. In contrast to results in Ref. [4.200], the evaluation of uranium concentration data for tissues of benthic and piscivorous fish species collected at reference sites in Canada shows significantly higher uranium concentrations in benthic relative to

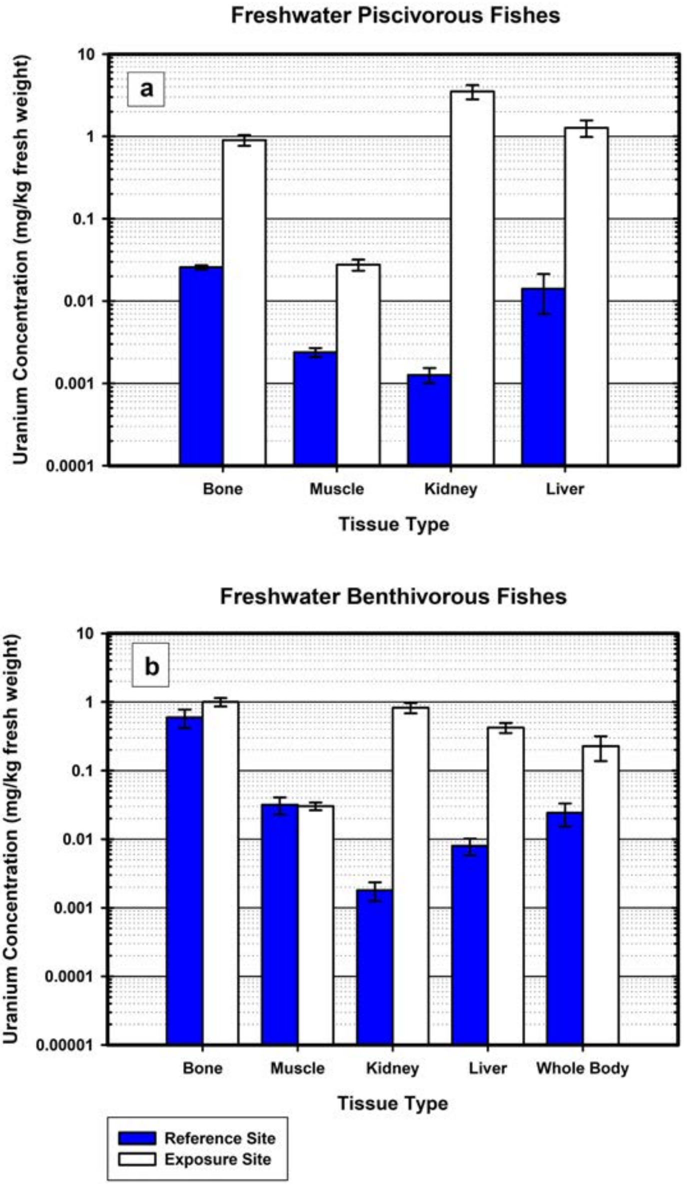

FIG. 4.10. Comparison of uranium concentrations (arithmetic mean ± standard error) in tissues of freshwater piscivorous (a) and benthivorous (b) fish collected at reference sites and at exposure sites impacted by uranium mining (based on data from Ref. [4.144]).

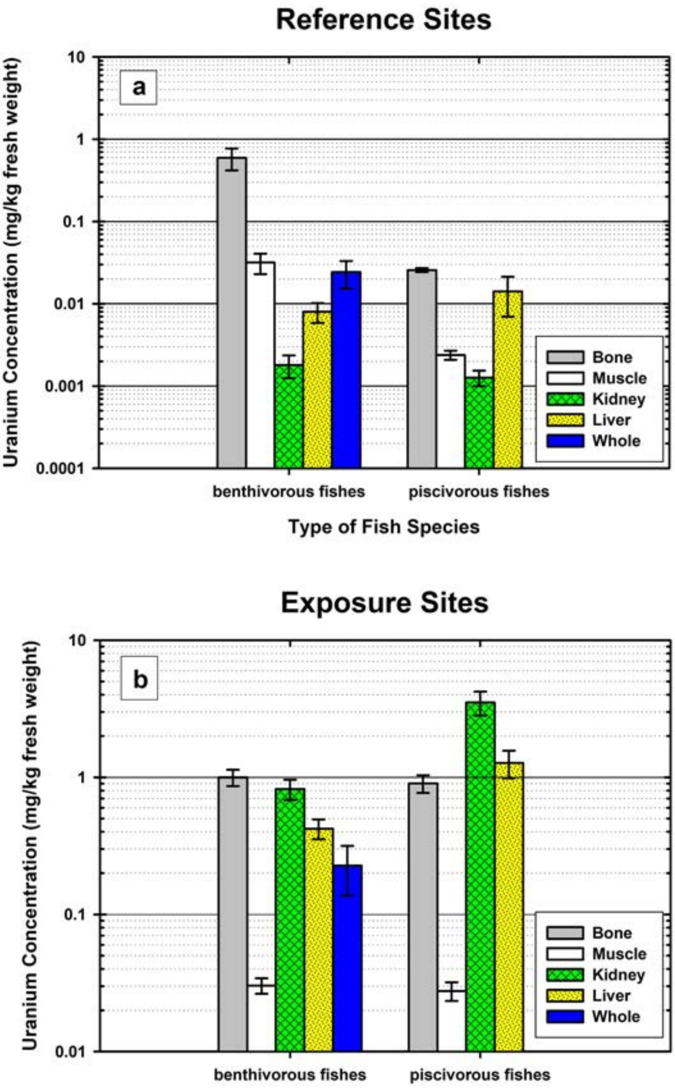

FIG. 4.11. Comparison of uranium concentrations (arithmetic mean ± standard error) in tissues of freshwater fish collected at reference sites (a) and at exposure sites impacted by uranium mining (b), organized by type of fish species (based on data from Ref. [4.144]).

piscivorous species, with similar values measured in liver and kidney of the two fish types (Figs 4.10 and 4.11). It is possible that this indicates the relative importance of uranium accumulation through ingestion of sediments and dietary routes compared with uptake via the gills at reference sites. Although not the primary route of uptake, some uranium is known to bioaccumulate via the gastrointestinal tract of fish [4.187, 4.190] and, in water bodies with relatively low dissolved uranium in the surface waters, it is possible that dietary uptake predominates. Concentrations of ^{238}U (mBq/g, FW) in different fish species are given in Table 4.20 for areas of the Komi Republic, Russian Federation, with normal and enhanced radiation background levels. Higher ^{238}U values for Atlantic salmon, *Salmo salar*, are shown in young (<4 years) fish compared with adult specimens in both radiation exposure situations.

In comparison, at Canadian exposure sites, uranium concentrations in piscivorous species were significantly higher in liver and kidney than in benthic species, with similar concentrations in bone and muscle between these different fish types. As already discussed, accumulation via the gills reportedly represents the dominant accumulation pathway. In addition, the physicochemical form of uranium can have a significant influence on uranium bioaccumulation, where low molecular mass forms of uranium tend to show enhanced mobility and bioavailability [4.191]. Such forms can then traverse the gills, accumulating in the body [4.185, 4.186].

TABLE 4.20. ACTIVITY CONCENTRATION OF ^{238}U IN FISH SPECIES IN AREAS OF THE KOMI REPUBLIC, RUSSIAN FEDERATION WITH DIFFERENT RADIATION BACKGROUND LEVELS [4.16, 4.87]

Biota species	Areas of normal radiation background		Areas of enhanced radiation background	
	No. of samples	^{238}U activity concentration (mBq/g, fresh weight)	No. of samples	^{238}U activity concentration (mBq/g, fresh weight)
Grayling (*Thymallus thymallus*)	15	0.003	18	0.036
Burbot (*Lota lota*)	5	0.017	5	0.074
Atlantic salmon (*Salmo salar*, adult)	15	0.001	15	0.012
Atlantic salmon (*Salmo salar*, age <4 years)	10	0.003	15	0.1
Whitefish (*Coregonus*)	3	0.002	5	0.032

In comparison, high molecular mass forms of uranium tend to be less mobile. It is, therefore, possible that gill uptake may represent the dominant pathway for piscivorous fish, which tend to be found in the water column, and that owing to the physiochemical conditions, less bioavailable forms of uranium may be present in the sediments where benthivorous fish feed.

Once ingested, concentrations of uranium tend to vary in different tissues of freshwater animals and can also differ between material in the gastrointestinal tract and in internal tissues. For example, concentrations of uranium in the gut contents of fish, including whitefish (*Coregonus clupeaformis* and *Prosopium cylindraceum*), rainbow trout (*Oncorhynchus mykiss*), lake trout (*Salvelinus namaycush*) and northern pike (*Esox lucius*), are generally higher than those present in internal fish tissues [4.195, 4.201–4.204]. This is not surprising, since, as noted above, uranium tends to show a very low degree of uptake through the gut of many species.

Within the body itself, uranium concentrations differ between tissues, with higher concentrations observed in bones (Table 4.21). As already noted, uranium is accumulated in the kidney, liver and bone but does not biomagnify. For example, concentrations reported in bone and gut tissues of lake trout (piscivorous) and whitefish (benthivorous) collected in uranium mining areas are significantly higher than those found in muscle [4.201]. Similar trends have been observed for piscivorous and benthivorous fish species collected in both reference and exposure areas of northern Saskatchewan, Canada, with elevated uranium concentrations occurring in the kidney, liver, bone and whole body, and relatively lower levels in muscle tissues (see Fig. 4.10). The elevated concentrations in the kidney may be related to its role as the primary site of accumulation for food-borne exposure under long term exposure conditions [4.106]. In addition, uranium partitioning into liver tissue may be related to its function in detoxification within the body. These results are consistent with those reported in Ref. [4.190], where it was concluded that uranium accumulation in fish (*Danio rerio*) occurred in the mineralized tissues (bone and scales) but also in the intestine, liver and kidney. In the latter case, relatively high uranium concentrations were found in the gonads of *Danio rerio* and, specifically, in the eggs of females (>20% relative burden), indicating maternal transfer of uranium [4.190]. Elevated uranium concentrations have also been reported to occur in fish gonadal tissues under certain conditions [4.202]. Reference [4.188] refers to studies that report that uranium tends to accumulate in mineralized tissue (e.g. bone, scales), with lower concentrations occurring in the kidney and measurable partitioning into the liver, gills, skin and muscle [4.202, 4.204].

As with fish, following exposure to elevated uranium concentrations, freshwater bivalves (e.g. *Corbicula fluminea*) have shown relatively higher concentrations of uranium in the gills and visceral mass than in foot tissue

TABLE 4.21. MASS CONCENTRATION OF URANIUM IN TISSUES OF FRESHWATER FISH COLLECTED AT REFERENCE SITES AND AT EXPOSURE SITES IMPACTED BY URANIUM MINING

			Reference site					Exposure site					
Country	Species category	Tissue type	N^a	Arithmetic mean (mg/kg, fresh weight)	Standard deviation	Min.	Max.	N^a	Arithmetic mean (mg/kg, fresh weight)	Standard deviation	Min.	Max.	Reference
Fresh water (flowing)													
Canada	Forage fish[b]	Muscle	5	0.014	0.0055	0.01	0.02	10	0.022	0.012	0.01	0.04	[4.144, 4.205]
Canada	Piscivorous fish[c]	Bone	85	0.021	0.0044	0.02	0.04	108	0.027	0.020	0.01	0.114	[4.144, 4.205]
		Muscle	85	0.0013	0.00085	0.01	0.004	135	0.0012	0.00078	0.001	0.007	[4.144, 4.205]
		Whole body	5	0.010	0	0.01	0.01	—[d]	—[d]	—[d]	—[d]	—[d]	
Canada	Benthic fish[e]	Bone	87	0.034	0.021	0.02	0.1	101	0.033	0.051	0.01	0.31	[4.144, 4.205]

TABLE 4.21. MASS CONCENTRATION OF URANIUM IN TISSUES OF FRESHWATER FISH COLLECTED AT REFERENCE SITES AND AT EXPOSURE SITES IMPACTED BY URANIUM MINING (cont.)

			Reference site					Exposure site					
Country	Species category	Tissue type	*N*[a]	Arithmetic mean (mg/kg, fresh weight)	Standard deviation	Min.	Max.	*N*[a]	Arithmetic mean (mg/kg, fresh weight)	Standard deviation	Min.	Max.	Reference
Canada	Benthic fish[e]	Muscle	87	0.0018	0.0017	0.001	0.01	146	0.0019	0.0017	0.001	0.009	[4.144, 4.205]
Fresh water (lake)													
Canada	Forage fish[f]	Whole body	—[d]	—[d]	—[d]	—[d]	—[d]	11	0.045	0.059	0.001	0.18	[4.144, 4.205]
Canada	Piscivorous fish[g]	Bone	151	0.026	0.018	0.01	0.1	516	0.90	3.0	0.001	19.9	[4.144, 4.205]
		Muscle	151	0.0024	0.0037	0.001	0.03	516	0.028	0.097	0.001	0.84	[4.144, 4.205]
		Kidney	15	0.0013	0.0010	0.001	0.005	80	3.5	6.2	0.006	23.8	[4.144, 4.205]

TABLE 4.21. MASS CONCENTRATION OF URANIUM IN TISSUES OF FRESHWATER FISH COLLECTED AT REFERENCE SITES AND AT EXPOSURE SITES IMPACTED BY URANIUM MINING (cont.)

			Reference site					Exposure site					
Country	Species category	Tissue type	*N*[a]	Arithmetic mean (mg/kg, fresh weight)	Standard deviation	Min.	Max.	*N*[a]	Arithmetic mean (mg/kg, fresh weight)	Standard deviation	Min.	Max.	Reference
Canada	Piscivorous fish[g]	Liver	15	0.014	0.028	0.001	0.11	95	1.3	2.8	0.001	15	[4.144, 4.205]
Canada	Benthic fish[h]	Bone	115	0.59	1.9	0.004	10.9	440	1.0	2.9	0.001	30	[4.144, 4.205]
		Muscle	115	0.032	0.095	0.001	0.539	440	0.030	0.082	0.001	0.67	[4.144, 4.205]
		Kidney	15	0.0018	0.0021	0.001	0.009	79	0.82	1.2	0.01	7.2	[4.144, 4.205]
		Liver	15	0.0080	0.0084	0.001	0.026	63	0.42	0.55	0.003	3.4	[4.144, 4.205]
		Whole body	84	0.024	0.082	0.01	0.546	6	0.23	0.22	0.002 8	0.495	[4.144, 4.205]

TABLE 4.21. MASS CONCENTRATION OF URANIUM IN TISSUES OF FRESHWATER FISH COLLECTED AT REFERENCE SITES AND AT EXPOSURE SITES IMPACTED BY URANIUM MINING (cont.)

			Reference site					Exposure site					
Country	Species category	Tissue type	*N*[a]	Arithmetic mean (mg/kg, fresh weight)	Standard deviation	Min.	Max.	*N*[a]	Arithmetic mean (mg/kg, fresh weight)	Standard deviation	Min.	Max.	Reference
Tajikistan	Benthic fish[i]	Bone	2	—[d]	—[d]	0.021	0.037	13	13	5.0	—[d]	—[d]	[4.118]
		Muscle	2	—[d]	—[d]	<0.01		13	0.30	0.10	—[d]	—[d]	[4.118]
		Kidney	2	—[d]	—[d]	0.018	0.020	13	13	10	—[d]	—[d]	[4.118]
		Liver	2	—[d]	—[d]	0.022	0.023	13	14	29	—[d]	—[d]	[4.118]
		Gills	2	—[d]	—[d]	0.026	0.027	13	8.9	2.3	—[d]	—[d]	[4.118]
Kazakhstan	Piscivorous fish[j]	Muscle	—[d]	—[d]	—[d]	—[d]	—[d]	12	0.14	—[d]	—[d]	—[d]	[4.117, 4.179]
		Liver	—[d]	—[d]	—[d]	—[d]	—[d]	20	3.1	3.4	—[d]	—[d]	[4.117, 4.179]

TABLE 4.21. MASS CONCENTRATION OF URANIUM IN TISSUES OF FRESHWATER FISH COLLECTED AT REFERENCE SITES AND AT EXPOSURE SITES IMPACTED BY URANIUM MINING (cont.)

			Reference site					Exposure site					
Country	Species category	Tissue type	*N*[a]	Arithmetic mean (mg/kg, fresh weight)	Standard deviation	Min.	Max.	*N*[a]	Arithmetic mean (mg/kg, fresh weight)	Standard deviation	Min.	Max.	Reference
Kazakhstan	Piscivorous fish[j]	Gills	—[d]	—[d]	—[d]	—[d]	—[d]	23	3.5	1.1	—[d]	—[d]	[4.117, 4.179]

Note: Max.: maximum; Min.: minimum.

[a] Number of individual replicate data points.

[b] *Notropis hudsonius* (reference site and exposure site).

[c] *Esox lucius* and *Lota lota* (reference site), and *Esox lucius* (exposure site).

[d] —: data not available.

[e] *Catostomus catostomus*, *Catostomus commersonii* and *Coregonus clupeaformis* (reference site); *Catostomus commersonii* and *Coregonus clupeaformis* (exposure site).

[f] *Couesius plumbeus*, *Notropis hudsonius*, *Percopsis omiscomaycus*, *Rutilus rutilus* (exposure site).

[g] *Esox lucius* and *Salvelinus namaycush* (reference site and exposure site).

[h] *Catostomus catostomus*, *Catostomus commersonii*, *Coregonus clupeaformis*, *Couesius plumbeus* and *Percopsis omiscomaycus* (reference site and exposure site).

[i] Fish species: *Carrasius. auratus* from Taboshar uranium pit lake, Tajikistan (exposure site) and *Cyprinus carpio* from the Kairrakum reservoir, Tajikistan (reference site).

[j] Fish species: *Perca schrenkii* from Kurday uranium pit lake, Kazakhstan (exposure site).

[4.206, 4.207]. In addition, uranium has been reported to accumulate primarily in the stomach and, in particular, in the digestive gland of crayfish (*Orconectes limosus*) [4.199], whereas it was primarily found in exuvia (i.e. the pupal skin) of chironomids (*Chironomus tentans*), resulting in elimination during moulting [4.208].

4.5. URANIUM IN THE MARINE ENVIRONMENT

As with many other elements, uranium dissolved in sea water originates in erosion from the lithosphere and river discharges into coastal seas. Uranium is carried by rivers both in dissolved and particulate fractions. An additional component is wind transported dust deposited at the ocean surface, with important contributions from desert areas, such as dust from the Sahara that is deposited in the North Atlantic Ocean [4.209, 4.210]. There is also a contribution from anthropogenic uranium inflow to rivers and then to the ocean [4.211, 4.212]. Ocean water circulation in sediments and rocks in the seabed and deep-sea hydrothermal sources are additional uranium sources and sinks [4.213].

4.5.1. River water discharges

Uranium is weathered from rocks and soils, and in oxygenated river water it is mostly present as dissolved uranyl carbonate species $UO_2(CO_3)_3^{4-}$ [4.214]. The average concentration of uranium in rivers has been estimated as 0.3 µg/L and, more recently, as 0.6 µg/L, but higher concentrations have been reported of up to 7 µg/L, and these seem likely to have a natural origin and not to have resulted from contamination such as the usage of phosphate fertilizer in the watershed. Concentrations of uranium in river waters have been consistently found to be significantly correlated with total dissolved solids and, thus, river transport of uranium is controlled by chemical weathering in watersheds [4.135].

As uranium easily dissolves in aerated water, most uranium in river water is associated with the soluble phase, and lower amounts are carried in the suspended particulate phase. For example, in the Tejo river, Brazil, in the upper catchment, uranium in suspended particulate matter could be around 43% of total uranium; but in the lower catchment, the contribution from particulate matter decreased to 0.3%, and this decrease was associated with the increased dissolution of particulate uranium. Similar trends in uranium dissolution from riverbed sediments and suspended particulate matter were observed in rivers with sources at granitic mountains such as the Mondego river, Portugal [4.175, 4.215].

The naturally occurring uranium isotopes ^{238}U, ^{235}U, and ^{234}U are all present in river water and in the lithosphere. Generally, in the lithosphere, the

^{234}U:^{238}U activity ratios are around one. However, owing to recoil effects, ^{234}U seems more easily dissolved than ^{238}U, and the ^{234}U:^{238}U activity ratios in river waters have been commonly reported to be above unity, often around 1.2–1.3 [4.108]. Uranium in river water has been measured on several continents and attempts have been made to calculate the present total rate of input of uranium with fresh waters into the world's oceans. This input is estimated at around 1×10^{10} g [U]/a [4.211, 4.216].

4.5.2. Estuaries

In estuaries, river water mixes with sea water and enters the sea. The behaviour of dissolved uranium has been investigated in estuaries around the world and, in some cases, it was described as conservative; that is, the uranium from river discharge exhibited a gradient consistent with the salinity gradient across the estuary and, thus, the variation in uranium concentration was similar to the salinity gradient generated by fresh water and sea water mixing, which basically means that the flux of uranium delivered by the river is transferred through the estuary and delivered unmodified into the coastal sea [4.217, 4.218]. In other estuaries, uranium behaviour has been described as non-conservative and, among these, there are estuaries that are a sink of uranium, whereas others are a source of additional uranium. In both cases, the flux of uranium from rivers is modified in the estuary before delivery into the sea [4.219, 4.220]. These studies have mostly been carried out through measurements of uranium in surface waters, but the analysis of uranium in sediment pore water clarified that uranium release from sediments occurred in association with dissolution/precipitation of iron and manganese oxides [4.221–4.223]. Uranium behaviour in estuaries and mudflats is closely related to the redox chemistry of the sediments, and it is likely that river discharges of uranium into the world's oceans, rather than being constant at a given location, may fluctuate over time with formation and destruction of estuaries, lagoons and salt marshes, and, thus, with organic matter loads in such transitional environments. This fluctuation might be enhanced in the next few decades due to various influences of climate change.

4.5.3. Coastal seas

In coastal seas, uranium concentrations in sea water seem more variable than those in the open ocean. Uranium concentrations in coastal sea water have been reported to be either below or much above the ocean average, and the reasons for this are not yet fully understood. It has been suggested that in some cases, this could be related to phosphate residue discharges and phosphate fertilizer usage in watersheds [4.212]. Submarine water discharges may be another source of unusual concentrations of dissolved uranium in coastal waters, but these discharges remain unstudied in respect of their uranium input [4.223].

4.5.4. The open ocean

Reported uranium concentrations in ocean water range from 1 to 5 µg/L, but there is general agreement among reports that the average is around 3.3 µg/L and that large variations, especially in older measurements, are related to analytical difficulties. Indeed, more recent analyses have narrowed the uncertainty in the uranium concentration in ocean water and in ^{234}U:^{238}U activity concentration ratios. For example, analyses of a large number of samples from the world's oceans agreed on reported activity concentration ratios of 1.14 ± 0.03 and 1.13 ± 0.04 for the Atlantic Ocean, measured by alpha spectrometry, and 1.144 ± 0.002 for the Atlantic and Pacific oceans, determined by inductively coupled plasma mass spectrometry [4.224–4.226]. It has been suggested [4.227] that the ^{234}U:^{238}U ratio of sea water has remained within about 20% of the modern sea water value over the past 1–1.4 million years. By far the largest input of uranium into the oceans is the world river discharge, estimated at around 1×10^{7} kg [U]/a. Dust transported by atmospheric mechanisms and deposited on the ocean surface gives an input that for the north-eastern Atlantic Ocean was estimated at 1.3 $\mu g \cdot m^{-2} \cdot a^{-1}$. If this is assumed as representative of the global input of atmospheric dust, then dust deposition would account for about 2% of the uranium in world river discharges [4.228].

The total uranium content in ocean water is calculated as 4.5×10^{12} kg and is assumed to be mostly in the dissolved phase. Analyses of uranium in the particulate matter of a 400 L subsurface sample of sea water confirmed that only 0.02% of uranium was associated with suspended particulate matter [4.229].

Considering the ocean volume and the average uranium concentration in ocean water, the uranium residence time was calculated as 4.5×10^{5} a [4.230]. This is sufficiently long to ensure a very uniform distribution of this element in the world's oceans. Differences in uranium concentrations in sea water do exist in the world's oceans both geographically and in the vertical water column at the same location, and both seem correlated with differences in salinity [4.230]. In the vertical profiles of uranium concentrations in north-eastern Atlantic Ocean waters, uranium concentrations varied from 2.1 to 3.7 µg/L, along with salinity, and corresponding to water layers from different origins in the global ocean circulation. Data for several oceans indicate similar values (Table 4.22 [4.229, 4.231–4.237]).

Uranium in ocean sea water is removed by the biogenic flux of particulate material (particularly in siliceous and calcareous debris), sorption onto bottom sediments, incorporation into corals and shells, and by contact with reducing

TABLE 4.22. ACTIVITY CONCENTRATIONS OF URANIUM RADIOISOTOPES IN THE WORLD'S OCEANS AND SEAS

Ocean or sea	No. of samples	^{238}U activity concentration (mBq/L)[a]	^{234}U activity concentration (mBq/L)[a]	Salinity (‰)[a]	Reference
North Pacific Ocean (Hawaii)	—[b]	35.5 ± 2.3	37.3 ± 2.6	—[b]	[4.231]
North-eastern Atlantic Ocean	24	37 ± 4	42 ± 5	36	[4.229]
Equatorial Atlantic Ocean (Bahamas)	12	42.3–44.3	—[b]	35.4–37.2	[4.232]
North Subtropical Atlantic Ocean (Canary Islands)	—[b]	47.4 ± 3.5	51.0 ± 3.8	—[b]	[4.231]
Indian Ocean	—[b]	40.2–40.5	—[b]	34.7 ($n = 19$)	[4.233]
Open Mediterranean Sea (Adriatic and Ionian Seas)	—[b]	43.7–46.3	—[b]	37.7–38.9	[4.234]
Baltic Sea					
Gdańsk Deep	11	74.9 ± 0.64	85.5 ± 0.64	—[b]	[4.213]
Supsk Narrow	13	45.3 ± 0.86	51.3 ± 0.80	—[b]	[4.213]
Bornholm Deep	12	48.6 ± 0.76	54.7 ± 0.91	—[b]	[4.213]
Arctic Ocean (Canada)					
Under ice surface water	3	28–36.3	—[b]	24.5–32.1	[4.235]
Ice melt	5	0.1–5.1	—[b]	0.1–4.7	[4.235]

TABLE 4.22. ACTIVITY CONCENTRATIONS OF URANIUM RADIOISOTOPES IN THE WORLD’S OCEANS AND SEAS (cont.)

Ocean or sea	No. of samples	^{238}U activity concentration (mBq/L)[a]	^{234}U activity concentration (mBq/L)[a]	Salinity (‰)[a]	Reference
Subtropical Atlantic Ocean (Bermuda)	54	28.7–41.9	—[b]	34.9–36.6	[4.236]
South Atlantic Ocean	23	35.6–41.3	—[b]	38.7–34.7	[4.236]
North Tropical Atlantic Ocean	17	38.8–42.8	—[b]	34.8–37.1	[4.236]
Arabian Sea	—[b]	34.3–44.7	32.0–41.7	34.9–36.5	[4.237]
Irish Sea	—[b]	30.1 ± 2.6	32.8 ± 2.8	—[b]	[4.231]
Black Sea (Romanian shore)	—[b]	22.8 ± 1.3	26.5 ± 1.7	—[b]	[4.231]

[a] Numbers are given as average ± one standard deviation, or as a range of values.

[b] —: data not available.

environments, such as hydrothermal vents in the deep-sea floor, and weathering of marine basalts [4.230]. Uranium removed to these sinks is considered to balance the inputs into the oceans and, therefore, the uranium concentration in sea water is considered to represent a steady state balance of supply and removal processes [4.230].

4.5.5. Uptake in marine biota

4.5.5.1. Biokinetics of uranium accumulation and elimination

Uranium is a non-essential element and there are no known biomolecules in physiological processes using uranium atoms. However, the uranyl ion, the principal form of uranium found in freshwater and marine environments, is a very large divalent positively charged ionic species, with a size similar to that of potassium, calcium and caesium ions [4.238]. Therefore, in marine biota, its transfer across cell membranes is likely to occur in branchial organs through ion transport processes, as is the case for calcium and potassium ions, although probably in very limited amounts.

There have been few experimental studies in the laboratory on the kinetics of uranium uptake by marine biota, and rates of water–organism exchanges are not known. Experiments with phytoplankton showed that concentration of uranium from sea water takes place and accumulation occurs through adsorption onto external structures, as well as through intake into the cytoplasm, reaching volume concentration factors between 20 and 600, much lower than for ^{228}Th and ^{210}Pb [4.239]. Equilibrium between the concentration of uranium in cells and in sea water was reached in no more than about 24 h.

4.5.5.2. Accumulation in marine organisms

Measurements on natural phytoplankton from the sea surface at the outer continental shelf gave an activity concentration of uranium of around 3.4 Bq/kg, fresh weight. Coastal filter feeding organisms, such as mussels (*Mytilus galloprovincialis*), concentrate uranium of up to 660 mBq/kg (fresh weight) in soft tissues. Uranium incorporated into mussels may originate from dissolved uranium in sea water, but also from ingestion of suspended matter, namely phytoplankton cells and sediment particles.

Table 4.23 shows uranium isotope concentrations for several marine organisms organized according to food chains and trophic levels in marine ecosystems. The data available are limited but are sufficient to derive a general pattern. Phytoplankton concentrates the dissolved uranium from sea water up to concentration levels that are not exceeded by any other organisms belonging to the main groups of marine fauna.

In coastal seas, a typical pelagic food chain, such as phytoplankton to planktonivorous fish (sardine) to carnivorous fish (tuna), displays a clear decrease in uranium activity concentrations when going up the food chain to higher trophic levels.

Decreases in the concentrations of uranium at higher trophic levels were also observed in organisms in other marine food chains, such as demersal food chains in the North Atlantic Ocean, deep-sea fish from the continental slope (rat tail fish), and pelagic food chains in the open ocean, such as those involving squid and the sperm whale. Even organisms from special environments, such as deep-sea hydrothermal vents, show a decrease in uranium concentrations going up the food chain. Therefore, the accumulation of uranium in marine species does not imply any biomagnification of uranium concentrations in marine food chains.

As uranium concentrations in sea water are remarkably constant, at around 3.3 µg/L, which corresponds to about 38 mBq/L of ^{238}U, marine organisms with a similar physiology should concentrate dissolved uranium up to similar levels. Differences related to geography, such as water temperature and salinity acting, for example, on metabolic rates would play a minor role in the uranium accumulation process.

However, significant differences in uranium concentrations among species of the same group, such as teleost fish, all living in sea water with similar concentrations of dissolved uranium, have been observed. These could be attributed to the additional effect of uranium absorption from food and, in some cases, to contributions from sediment particles ingested by demersal species with benthic prey. However, this effect of food chain transfers in relation to similar concentrations of uranium dissolved in sea water cannot satisfactorily explain the departure of activity concentrations from a linear relationship with trophic level.

Data on activity concentrations of uranium in fish and other large organisms (see Table 4.23 [4.240–4.250]) indicate that the uranium concentration decreases as a power function of body size. This fits well with an exponential decrease of metabolic rates with increasing body size (i.e. an allometric relationship). At least in bivalves, this suggests that the main source of uranium is the ingested water through the drinking water reflex to compensate ion losses and maintain the osmotic balance with external media [4.251]. The uranium source would be the dissolved uranium in sea water, not through branchial uptake (divalent ion transport across epithelia), but rather through water ingestion and uranium absorption through the gut wall. This process has also been observed with other non-essential and high atomic mass elements in fish, namely transuranium elements such as plutonium and americium [4.252, 4.253].

TABLE 4.23. ACTIVITY CONCENTRATIONS OF URANIUM RADIOISOTOPES IN MARINE ORGANISMS

Organism	^{238}U activity concentration (Bq/kg)[a]	^{234}U activity concentration (Bq/kg)[a]	Reference
Phytoplankton (FW)	0.4–1.9	—[b]	[4.240]
Macroalgae			
Brown macroalgae	3.8–11	—[b]	[4.241]
Macroalgae (USA)	1.7-8.5	—[b]	[4.242]
Zooplankton			
Copepods	12.6	—[b]	[4.240]
Copepods (*Anomalocera patersoni*)	1.7–4.7	—[b]	[4.243]
Crab larvae	0.75	—[b]	[4.243]
Euphausiid	0.78	—[b]	[4.244]
Mixed copepods	2.3–12.5	—[b]	[4.243]
Salps			
Salps	12.5	—[b]	[4.240]
Salps (*Salpa maxima*)	4.3	—[b]	[4.243]
Salps (*Thallia democratica*)	15–22	—[b]	[4.243]
Crustaceans			
Shrimps carapace (*Callichirus laura*)	11 ± 16	—[b]	[4.245]
Shrimps hepatopancreas (*Callichirus*	14.3 ± 9.0	—[b]	[4.245]
laura)	2.8 ± 19	—[b]	[4.245]
Shrimps muscles (*Callichirus laura*)	9.9	—[b]	[4.241]
Barnacles soft parts			
Jellyfish			
Rhopilema nomadica	0.8–1.8	—[b]	[4.246]
Aurelia aurita	0.4	—[b]	[4.246]
Aequorea forskalea	0.2	0.9–2.0	[4.246]
Molluscs		0.4–0.5	
Abalone (USA)	13	0.2	[4.242]
Littorina littorea (United Kingdom)	1.36–18.9	—[b]	[4.247]
Molluscs	1.4	—[b]	[4.241]
Mussels (United Kingdom)	1.97–4.18	—[b]	[4.242]
Mytilus edulis (United Kingdom)	1.01–37.1	—[b]	[4.247]
Patinopecten yessoensis (Japan)	4.6	—[b]	[4.242]

TABLE 4.23. ACTIVITY CONCENTRATIONS OF URANIUM RADIOISOTOPES IN MARINE ORGANISMS (cont.)

Organism	^{238}U activity concentration (Bq/kg)[a]	^{234}U activity concentration (Bq/kg)[a]	Reference
Fish			
Coastal elasmobranch (FW)	0.14	—[b]	[4.248]
Demersal coastal teleost (FW)	0.06–0.84	—[b]	[4.248]
Fish muscle (FW)	0.003–1.1	—[b]	[4.240]
Cod (*Gadus morhua*, FW)	2.6–6.0	2.4–5.6	[4.249]
Herring (*Clupea harengus*, FW)	2.6–3.6	2.4–3.3	[4.249]
Sprat (*Sprattus sprattus*, FW)	5.4	5.1	[4.249]
Flounder (*Platichthys flesus*, FW)	2.7	2.5	[4.249]
Garfish (*Belone belone*, FW)	3.7–5.2	3.5–4.8	[4.249]
Greater sand eel (*Hyperoplus lanceolatus*,	5.4	5.1	[4.249]
FW whole fish)	32.1	29.9	[4.249]
Stickleback (*Gasterosteus aculeatus*,			
FW whole fish)			
Greenland halibut (*Reinhardtius*	12.7 ± 0.5	16.4 ± 0.6	[4.250]
hippoglossoides, FW)	16.4 ± 0.6	4.3 ± 0.2	[4.250]
Plaice (*Hippoglossoides platessoides*, FW)	8.1 ± 0.3	8.5 ± 0.3	[4.250]
Red fish (*Sebastes mentella*, FW)	6.1 ± 0.2	3.4 ± 0.1	[4.250]
Ray (*Raja centa*, FW)	24.1 ± 1.0	30.9 ± 1.2	[4.250]
Roundnose grenadier (*Coryphaenoides*	210 ± 9	193 ± 9	[4.250]
rupestris, FW)	26.1 ± 1.2	34.6 ± 1.6	[4.250]
Red hake (*Urophycis chuss*, FW)	12.0 ± 0.5	9.0 ± 0.4	[4.250]
Kite fin shark (*Dalatias licha*, FW)			
Red fish (*Sebastes mentella*, FW)			

Note: The activity concentration is for dry weight unless otherwise indicated as fresh weight (FW).

[a] Numbers are given as average ± one standard deviation, or as a range of values. Information on the number of samples is not available.

[b] —: data not available.

There are limited data on concentrations of uranium in marine birds. Reference [4.254] reported that uranium concentrations in marine birds from the Baltic are low, but higher than in marine fish. Whole body concentrations ranged from 2.33 mg/kg, FW in razorbill (*Alca tarda*) to 18.08 mg/kg, FW in tufted duck (*Aythya fuligula*). The highest concentrations were observed in viscera (up to 75.23 mg/kg, FW in *A. fuligula*) and feathers (up to 23.97 mg/kg, FW in the great cormorant, *Phalacrocorax carbo*). The lowest concentrations were observed in muscle (0.21–11.77 mg/kg, FW in various species). Moulting was found to be a significant contributor to loss of uranium from marine birds. Detailed data for tissues and organs of individual species are given in Ref. [4.255]. A multi-element analysis of the composition of individuals from a colony of great cormorants (*P. carbo*) from near Olkiluoto on the Baltic coast of Finland [4.256] gave whole body uranium concentrations of 1.3 μg/kg (adults) and 4.1 μg/kg (juveniles). The mean concentration in eggs was 0.6 μg/kg, and in guano it was much higher, at 550 μg/kg. About 15–25% of the uranium was found in the bones of the birds, with the remainder being widely distributed among the other tissues of the body. Both marine and terrestrial birds were included in a wider ranging study [4.257]. This addressed the accumulation of naturally occurring and artificial radionuclides in birds from Ethiopia, Mongolia, Poland, the Russian Federation, Ukraine and Viet Nam. Whole body concentrations of ^{238}U, expressed as ppm (mg/kg) on a DW basis, were mainly <0.2, but ranged up to about 1.5 mg/kg. On a FW basis, the concentrations would have been about a factor of four lower, which is substantially less than those in marine birds from the Baltic [4.254], but generally higher than those in great cormorants from near Olkiluoto.

4.6. URANIUM IN THE ATMOSPHERE

The natural sources of uranium are soils and crustal rocks. The uranium present in air is dependent on the mass of suspended particles, the uranium concentrations in the parent material and contributions from local industrial sources [4.258]. Typical uranium concentrations in the Earth's crust range from 2.0 to 6.1 ppm (see Section 4.2.1), but local concentrations can be much higher, reaching 200 000 ppm in ore bodies currently being mined [4.259]. Measurements of the natural uranium isotopes, ^{234}U, ^{235}U and ^{238}U, and sometimes also of the artificial isotope, ^{236}U, in the atmosphere are reported for several locations.

Uranium concentrations in air are frequently reported either as a mass (e.g. ng/m^3, pg/m^3) or an activity (e.g. μBq/m^3). For consistency, the tables in this section are presented with common units and original data are converted when necessary. Measurements of uranium concentrations in remote areas give an indication of background levels of uranium in air. Reference [4.260] provided information on uranium measurements from areas of low anthropogenic input during an ocean traverse from Belgium to Antarctica (Table 4.24 [4.260–4.263]). The air concentration of uranium in the northern hemisphere was 4.1 pg/m^3, whereas in the southern hemisphere, uranium was not detected in most of the samples. A mean concentration of 3 pg/m^3 was obtained for air sampled at the Belgian Roi Baudouin base in Antarctica (closed in 1967). Similar air concentrations were observed in northern Norway [4.261, 4.262].

TABLE 4.24. CONCENTRATION OF URANIUM IN AIR IN REMOTE AREAS

Location	Uranium concentration (pg/m^3)	Notes	Reference
North Atlantic Ocean	4.1 ± 1.2		[4.260]
South Atlantic Ocean	2.2 ± 0.5	Mean for 5 samples, no uranium detected in 4 other samples	[4.260]
Antarctica, pack ice	1.2 ± 0.3	Mean for 3 samples, no uranium detected in 15 other samples	[4.260]
Antarctica, Roi Baudouin base	3 ± 1	Mean for 18 samples, no uranium detected in 2 other samples	[4.260]
Skibotn, Norway	2.7 ± 1.3		[4.261]
Vardö, Norway	5.5 ± 0.9		[4.262]
Central Russian Federation	11.3 ± 6.4	$n = 27$	[4.263]

Note: Several of the mean values are based only on those samples in which uranium was detected. It is difficult to decide how best to treat samples with results below the limit of detection and several alternatives have been proposed. In Ref. [4.260], it was opted to exclude them from the averaging process.

The ambient particle load in air results from several mechanisms in rural and urban areas, and includes both natural (e.g. by wind) and anthropogenic (e.g. from passing vehicles) suspension processes. This increases the atmospheric particle load, thereby elevating uranium concentrations in air. Measurements presented in Table 4.25 [4.260, 4.261, 4.264–4.274] include locations reported to be influenced by local burning of coal (e.g. data from Ref. [4.261]) releasing fly ash into the atmosphere. Among the natural processes, wind suspension of soils at local or regional scales is a continuous but time varying process. An occasional feature in Europe is the transport of desert dust from the Sahara [4.275], which is also a significant source for the oceans (see Section 4.5). Suspension can also occur because of burning biomass; this can be a wildfire or be planned under controlled conditions, and may involve managed or natural forest, moorland or agricultural stubbles. Anthropogenic emissions into the atmosphere include those due to agricultural practices, road traffic and industrial facilities.

It could be argued that some of the sites in Table 4.25 should be included in later tables of recorded air concentrations close to uranium mining or processing facilities; however, sites that show little difference from typical ambient uranium air concentrations are included in Table 4.25 (e.g. Los Alamos National Laboratory in the USA, Yokosuka in Japan).

TABLE 4.25. CONCENTRATION OF URANIUM IN AIR IN RURAL AND URBAN AREAS

Location	Uranium concentration (pg/m^3)	Notes	Reference
Vienna, Austria	102 ± 43	Mean of samples from 1974 to 1978	[4.264]
Mol, Belgium	115 ± 95		[4.265]
Hinschu, China	40		[4.266]
Berlin, Germany	133 ± 20		[4.261]
Braunschweig, Germany	85 ± 40		[4.261]
Kamisaibara, Japan	236	Samples over a ten year period	[4.267]
Odawara, Japan	130		[4.268]
Tokyo, Japan	24 ± 15		[4.269]
Tsukuba, Japan	14 ± 10		[4.269]
Yokohama, Japan	110 ± 50	Mean of five samples	[4.268]
Yokosuka (south), Japan	140 ± 80	Mean of ten samples	[4.268]
Sutton, United Kingdom	62 ± 78		[4.260]

TABLE 4.25. CONCENTRATION OF URANIUM IN AIR IN RURAL AND URBAN AREAS (cont.)

Location	Uranium concentration (pg/m^3)	Notes	Reference
Hanford, USA	60	Midpoint from graph of ten year monitoring	[4.270]
Los Alamos National Laboratory, USA	26	East gate	[4.271]
Los Alamos National Laboratory, USA	37	Area G	[4.271]
Santa Fe, USA	42	Off-site from the Los Alamos National Laboratory	[4.271]
USA	103	1978–1993 mean of 25 EPA monitoring stations	[4.272]
USA	50	2013 mean of 142 EPA monitoring stations	[4.273]
Global average	81.5		[4.274]

UNSCEAR [4.276] notes that a particle concentration in air of about 50 $\mu g/m^3$ is often assumed for assessment of the inhalation pathway. Based on this figure and an activity concentration of 25–50 Bq/kg for ^{238}U in soil, the uranium concentration in air can be estimated to be 100–200 pg/m^3.

Particles are removed from the atmosphere mainly by wet deposition processes and, to a lesser extent, by dry deposition. The lowest uranium air concentrations near Malvesi, France were obtained when it rained during the sampling period [4.277]. Seasonal trends have also been reported. In Ref. [4.267], it was found that ^{238}U air concentrations were greater in summer samples than in those collected during the winter. In Ref. [4.272], it was reported that measurements at the Argonne National Laboratory, USA showed peaks in

April and November and a low in August, noting that the ground is often covered in snow during winter months.

Monitoring of uranium concentrations in air is undertaken near uranium mining and milling facilities (Table 4.26). The suspension of particles because of mining activities is a recognized environmental issue. Processes associated with mining and milling activities that can release particles into the air include crushing and grinding of ores, transport of ore, ore roasting, drying and packaging of the final product. Releases can also occur from ore piles, tailings piles and roads. The uranium concentration in air declines rapidly with distance from the source [4.278]. Reference [4.279] presented an interesting analysis of estimated air and soil concentrations from a uranium milling facility at Uravan, USA. The town is situated in a complex terrain and the analysis was undertaken to estimate historical doses. Predicted concentrations in air and soil were compared to measurements from continuous air samplers from 1979 to 1986 and to soil profile sampling performed in 2006. The geometric mean of the predicted:observed ratio for annual average air concentrations was 1.25, with a geometric standard deviation of 1.8. The data are presented in a graphical format indicating peak observed uranium air concentrations of about 163 500 pg/m^3 in 1980.

Assessments of uranium concentrations in air should differentiate local background levels from the releases that originate from a facility. At Port Hope,

TABLE 4.26. CONCENTRATION OF URANIUM IN AIR IN URANIUM MINING AREAS

Location	Uranium concentration (pg/m^3)	Notes	Reference
Caetité, Brazil	66 000	On-site average	[4.160]
Caetité, Brazil	4 900	Off-site average	[4.160]
Key Lake, Saskatchewan, Canada	1 300	Open mine	[4.86]
Tarkwa, Ghana	400 ± 356	Average near gold mine	[4.280]
Russian Federation	209 ± 641	*n* = 34, unnamed contaminated site	[4.263]
Bluewater, USA	33 200 ± 5 700	Peak at Anaconda uranium mine	[4.281]

Canada, measurements in the early 1980s showed enhanced air concentrations within 2 km of the facility, which released uranium mainly from buildings associated with UO_3 and UF_6 production [4.282]. In this case, the background levels were low (0.5%) relative to those due to releases from the facility. Elevated air concentrations were also found close to the uranium (UF_4) processing facility at Malvesi, France. Two periods of plant shutdown for servicing allowed an estimate of the contribution from stack releases to be distinguished from that from local soil and waste storage ponds. This showed that the emission stack contributed about 80% of air concentrations recorded over the study period (October 2009 to January 2011). In Ref. [4.277], it was noted that uranium isotope concentrations did not show any isotopic enrichment or depletion and, on this basis, the activity concentration of ^{238}U is used to estimate the total uranium load in air (pg/m^3) in Table 4.27.

In some studies, the analysis of uranium concentrations has focused on a specific particle size or range. A recent study of ambient air quality close to the Tummalapalle uranium mining site, Andhra Pradesh, India looked at atmospheric particulate matter with a median diameter of 10 µm [4.284]. The uranium concentration for these particles was found to be in the range of 5000–30 000 pg/m^3. In Ref. [4.268], it was found that 70% of uranium was associated with particles >2 µm and 40–50% was associated with particles >7 µm. Uranium distribution across five particle size fractions was presented in

TABLE 4.27. CONCENTRATION OF URANIUM IN AIR AT URANIUM PROCESSING SITES

Location	Uranium concentration (pg/m^3)	Notes	Reference
Olen, Belgium	501 ± 463		[4.265]
Port Hope, Canada	20 000	Background levels were 100 pg/m^3	[4.282]
Port Hope, Canada	35 600	Peak measurement	[4.283]
Malvesi, France	2 750	Average from October 2009 to January 2011	[4.277]
Andhra Pradesh, India	30 000	Peak measurement of PM_{10}	[4.284]

Ref. [4.285]; it was found that about 28% of uranium associated with each of the >7 µm and 3.3–7 µm particle size fractions.

Depleted uranium is used in a wide range of peaceful applications, such as the provision of radiation shielding for medical sources and as counterweights in aeroplanes. This metal is also used for armoured vehicles and, owing to its high density and high melting point, is used for anti-tank munitions and missiles. At sites in Montenegro and Serbia, where munitions containing depleted uranium were used, activity concentrations in the air varied between 162 and 3430 pg/m^3 [4.286]. The highest value was measured at a site where soils contain a high concentration of natural uranium. However, most of the air samples tested positive for depleted uranium and this agreed well with the widespread contamination detected in biological samples. Routine measurements of uranium in air from seven air samplers at the Pohakuloa training area, USA show a peak of 240 pg/m^3, with most results being <40 pg/m^3 [4.287].

Stratospheric fallout of ^{236}U, following the detonation of nuclear weapons, was measured from 1957 to 1963 [4.288]. The air concentration of ^{236}U ranged from 0.17 to 23 nBq/m^3 over this period, with the peak occurring in 1963.

4.7. SUMMARY OF URANIUM CONCENTRATIONS IN ENVIRONMENTAL MEDIA

As described in earlier sections of this chapter, uranium is ubiquitously present in the environment. In parent materials, soils and sediments, typical concentrations are 2–6 mg [U]/kg, but concentrations of >1000 mg [U]/kg occur in some contexts, notably black shales and phosphate rocks. In the atmosphere, uranium concentrations are determined by the suspended mineral particle load, together with the uranium concentrations in the soils and sediments from which that particle load derives. Typical values are in the range of 100–200 pg/m^3, but much higher values are recorded in uranium mining and milling areas. In general, uranium is bioexcluded from plants and animals, rather than being bioaccumulated. Thus, for example, in plants growing in normal background areas, concentrations of <1 Bq/kg, DW, corresponding to less than about 0.1 mg [U]/kg, DW, are typical. In uranium mining areas, concentrations in plants are often one or two orders of magnitude higher, but this is not always the case and some studies have recorded plant concentrations similar to those in normal background areas. In general, concentrations in terrestrial animals are similar to, or lower than, those in plants.

Uranium concentrations in terrestrial surface waters vary over about six orders of magnitude (Fig. 4.12). The lowest concentrations, about 0.001 µg/L, occur in rainwater, whereas concentrations in river waters are typically

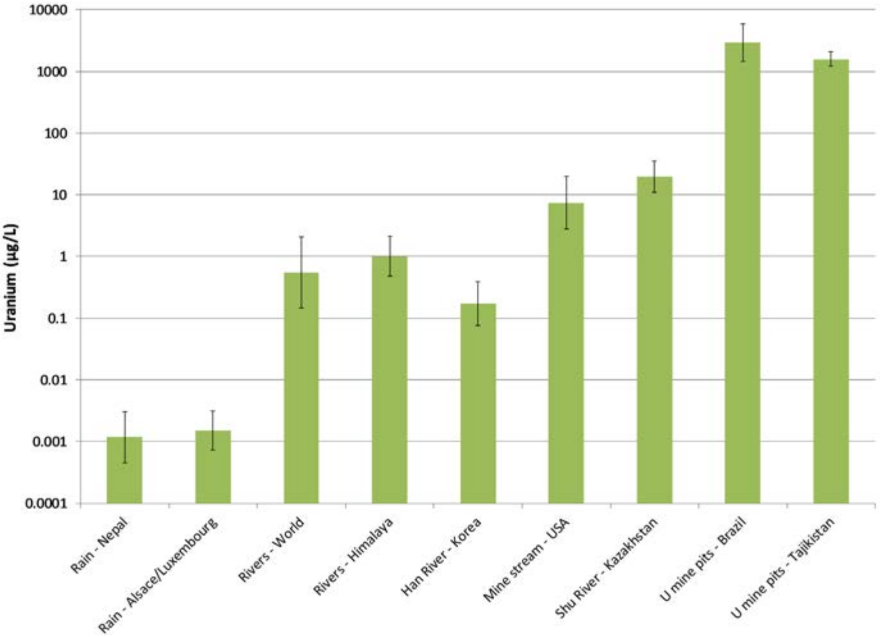

FIG. 4.12. Uranium concentrations in rainfall and some illustrative surface waters (the data are from Refs [4.108, 4.109, 4.118, 4.119, 4.145, 4.148, 4.160]).

0.1–1.0 µg/L. These higher river water concentrations arise because of rock–water interactions that increase uranium concentrations as meteoric waters percolate through soils, sediments and underlying parent materials. The final concentrations in surface waters are then determined, both by the concentrations in groundwaters discharging into those surface waters and by the contribution from recent meteoric waters that enter those surface waters either as runoff or after only limited percolation through the ground.

Where groundwaters encounter uraniferous deposits, the resulting uranium concentrations in surface waters can be substantially higher than those observed elsewhere. The concentrations in enclosed water bodies, such as mine pits, can range up to >1000 µg/L. In the case of rivers and streams in mining areas, concentrations of the order of 10 µg/L are more characteristic, because there is an opportunity for greater dilution by recent meteoric water.

In the case of aquifers, similar considerations apply. Some aquifers (e.g. near surface sand and gravel systems) are characterized by a rapid turnover of recent meteoric water, whereas other types are characterized by long groundwater residence times that permit rock–water interactions to develop more fully. Thus, uranium concentrations in aquifers range from <0.1 up to >100 μg/L (see Section 4.3.1).

As with terrestrial plants and animals, uranium is excluded from freshwater biota rather than being bioaccumulated, and observed concentrations are typically <1 mg [U]/kg, FW, except at some exposure sites impacted by uranium mining.

As uranium behaves rather conservatively in the oceans, having a mean residence time of >100 000 a, it becomes well mixed and exhibits a relatively uniform concentration of about 3.3 μg/L. As with freshwater organisms, uranium tends to be bioexcluded from marine organisms. However, higher concentrations can occur with some types of organism, such as phytoplankton, owing to surface adsorption, and in others, such as filter feeders, because of uptake of suspended matter such a phytoplankton cells and mineral particles.

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Chapter 5

PRINCIPLES OF RADIOLOGICAL AND TOXICOLOGICAL ASSESSMENT OF URANIUM

M.C. THORNE
Mike Thorne and Associates Ltd, United Kingdom

T. YANKOVICH
International Atomic Energy Agency

This chapter and the following two chapters relate to the requirements for, and approaches to, assessments of the radiological and toxicological significance of uranium present in the environment. To assess the impacts of uranium on human health and on the environment, it is necessary to consider the source (or sources) of uranium, its transport pathways through the environment and the receptors (both humans and wildlife) that may be exposed. Such source–pathway–receptor analyses are standardly undertaken for both chemical and radioactive pollutants. Figure 5.1 illustrates relevant considerations appropriate to uranium.

Uranium entering the environment is transported both in dissolved form and bound to, or incorporated in, solid particles. As it is a redox sensitive element, the proportions present in the solid and liquid phases, and the chemical speciation in the liquid phase, vary depending on the environmental context, as has been described in Chapters 2 and 3. At the largest scales, uranium is eroded from parent materials and is transported downslope and downstream until it reaches the ocean environment, where it accumulates in sea water, and is also lost to bottom sediments, which are eventually recycled into the mantle by subduction. However, it is transport at a local scale of tens of metres to a few kilometres that is mainly of relevance in assessing impacts, whether from naturally occurring anomalies or from human activities involving the processing of uranium-containing minerals. Transport also includes uptake by plants and animals, in terrestrial, freshwater, estuarine and marine environments, leading to concentrations of the element in a variety of environmental media, including biota. Mathematical models are generally used in assessment studies to represent both transport in the solid and liquid phases, and uptake and retention in biota. The parameterization of such models and the selection of parameter values appropriate to uranium are the subject of Chapter 6.

Once uranium concentrations in environmental media have been determined, impacts on human health and on the environment can be evaluated. These impacts arise both from the radioactivity of uranium and from its chemical

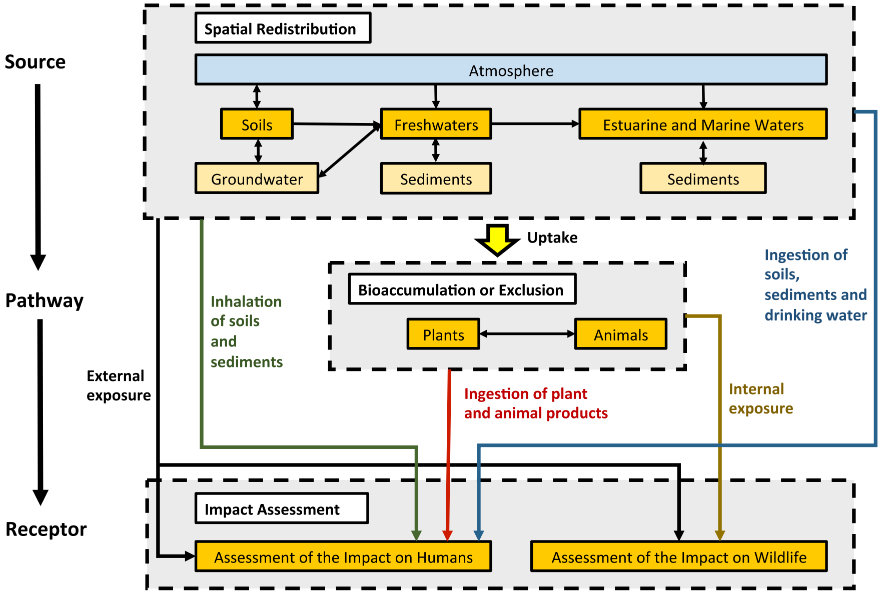

FIG. 5.1. Source–pathway–receptor analysis appropriate to uranium.

toxicity. In evaluating the radiological impacts of uranium, consideration must be given not only to the element itself, but also to its radioactive progeny, to the extent that they are present. The evaluation of these impacts is the subject of Chapter 7, which also provides relevant parameter values for use in quantitative assessments. The impacts on humans arise from external exposure to radiation emitted by uranium and its progeny, ingestion and inhalation of soils and sediments, ingestion of drinking water, and ingestion of contaminated biota, as well as from several additional, less significant pathways. Similar routes of exposure apply to wildlife.

5.1. PROTECTION STANDARDS FOR THE CHEMICAL AND RADIOLOGICAL TOXICITY OF URANIUM

As mentioned in Chapter 1, because uranium is both chemically toxic and radioactive, both concerns must be taken into account when evaluating its effects on human health and the environment. As is shown in Chapter 7, either the radiological or chemical toxicity can be the more important consideration, depending on factors such as the route of exposure, degree of enrichment or depletion, and the presence or absence of radioactive progeny. In general, criteria for controlling exposures to uranium are set by considering its chemical toxicity and radiotoxicity separately. Limited consideration has been given to potential synergistic interactions between its chemical toxicity and its radiotoxicity, but such interactions are implicitly considered when standards are set that are based directly on observed adverse effects on human health or on the environment. In respect of human health and chemical toxicity, the main considerations are rates of intake of the element by ingestion and inhalation. Protection standards are expressed in terms of mass concentrations in inhaled air (μg [U]/m^3) and mass intake rates per unit body mass per day (μg [U] · kg^{-1} · d^{-1}). Different standards apply for exposures of short or long duration, and for various chemical forms of the element, but no distinctions are made between depleted, natural and enriched uranium [5.1]. Furthermore, because the standards are formulated in terms of air concentrations or rates of intake per unit body mass, no distinctions are made based on age [5.1]. These standards are discussed further in Chapter 7.

With respect to human health and radiotoxicity, protection standards are expressed in terms of effective dose for external irradiation and committed effective dose for internal irradiation arising from radionuclides incorporated into body tissues and organs [5.2]. Effective dose rates from uranium and its progeny present in the environment can be calculated using standard radiation transport codes, and compilations of dose rates for idealized geometries (such as planar, slab and semi-infinite volume sources) have been published (e.g. Ref. [5.3]). A compilation of committed effective doses arising from unit intakes of uranium and its progeny has been published by the ICRP [5.4]. The derivation and application of these values is discussed further in Chapter 7.

With regard to chemical toxicity, protection of the environment is achieved by the specification of environmental quality standards (EQSs) defined in terms of mass concentrations of uranium in various environmental media (e.g. mg [U]/L in fresh water, mg [U]/kg in soil; see also Section 7.6) and based on observations of either concentrations at which no adverse effects occur or concentrations at which adverse effects occur at low frequency or are of minimal severity. However, in respect of radiotoxicity, a rather different approach is taken. Although the intent is to protect the environment at various ecological levels and scales (e.g. habitat, community, population), the approach taken is to compute dose rates to representative reference animals and plants (RAPs), and to compare those dose rates with generic screening or consideration values [5.5]. The underlying assumption is that if adverse effects on individual organisms are adequately limited in severity and/or frequency, then adverse effects at higher ecological levels and on biodiversity will also be adequately limited. Currently, alternative sets of RAPs have been proposed by the ICRP [5.5] and the IAEA [5.6], as well as by other organizations, and further work is being undertaken to provide a generally agreed set of RAPs and to define uptake and impact assessment parameter values for use with that agreed set.

5.2. ENVIRONMENTAL MEDIA AND ORGANISMS OF RELEVANCE IN ASSESSMENTS

Whether chemical toxicity or radiotoxicity is the primary consideration, there is a common requirement to determine the concentrations of uranium and its progeny in the environmental media to which the receptors (humans or other species) will be exposed. In the case of existing situations, these concentrations may be established by measurement, but for projected or ongoing releases, or for assessing future conditions in a changing environment, quantitative mathematical modelling will be required. In broad terms, as discussed above and illustrated in Fig. 5.1, assessments of impacts of uranium require the identification of the source of uranium, its transport pathways through the environment from the source to receptor organisms, and the impacts on those receptors evaluated in terms of either intake or radiation dose.

It is convenient to have a well defined set of environmental media for which concentrations are defined. In respect of human foods, the IAEA used a plant classification system [5.7] based on 14 plant groups. Hence, all plants are categorized as either cereals, maize, rice, leafy vegetables, non-leafy vegetables, leguminous vegetables, root crops, tubers, fruits, grasses (cultivated species), fodder leguminous (cultivated species), pasture (species mixture — natural or cultivated), herbs and other crops. This system has been proposed as a basis for estimating transfer of radionuclides to plant foodstuffs in the framework of the assessment of exposures to humans through ingestion.

The plant tissues were also subdivided into ten components: berries, buds, fruits, grain, heads, leaves, roots, seeds and pods, stems and shoots, and tubers. Not all of these ten components are assigned to each plant group. Specifically, an assignment is made only where the portion represents an edible part of a specific plant.

The transfer of radionuclides to plants strongly depends on soil properties. Existing international soil classification systems are intended to provide details that are important in terms of plant cultivation. In the soil classification system of the Food and Agriculture Organization of the United Nations and the United Nations Educational, Scientific and Cultural Organization, there are 28 units (or categories) of soil and 125 subunits [5.8]. Transfer factor values are not available for subunits defined on such a detailed basis; differences between these units in terms of radionuclide transfer are, in general, not substantial.

Therefore, a much simpler classification system based on texture and organic matter content was suggested [5.9], while ensuring that a reasonable amount of data are available for each category. Four soil groups, ‘sand’, ‘loam’, ‘clay’ and ‘organic soil’ were defined for radiological assessments. These classes are mostly based on grain size distributions, with clays typically being ≤0.004 mm, sands ≥0.06–2 mm (gravels above this) and loams intermediate in size. For this scheme, the soils were grouped according to the percentage of sand and clay minerals and the organic matter content in the soil. For the mineral soils, three groups were created according to the sand and clay percentages as follows:

(1) Sand group: Sand fraction ≥65%, clay fraction <18%;
(2) Clay group: Clay fraction ≥35%;
(3) Loam group: All other cases.

Soils were included in the ‘organic group’ if the organic matter content was ≥20%. Finally, an ‘unspecified soil group’ was created for soils without characterization data or for mineral soils with unknown sand and clay contents [5.9].

Only five categories of animals/birds, namely cattle, sheep, goats, pigs and poultry, and three groups of product comprising meat, milk and eggs are considered in defining transfer coefficients from daily radionuclide intakes by animals to animal products that are consumed by humans [5.9]. It is assumed that such a classification system, applied to different environments, is adequate for the assessment of the transfer of uranium to humans through the intake of food, for example implying that specific consideration does not need to be given to the consumption of animal organs such as liver and kidney.

More information can be found in IAEA publications [5.7, 5.9], which have a detailed discussion of the data and concepts that were used to define these categories of soils, plants and animals.

With respect to exposure of wildlife, a classification based on groups of species (or so-called ‘reference organisms’) is applied. Such groups need to be selected based on their representativeness for the environments of interest. This should also allow realistic assessments, illustrating possible exposure pathways in the selected environments. One such ‘reference organism’ approach was introduced during the Environmental Risk from Ionising Contaminants: Assessment and Management (ERICA) project for developing a framework for radiation protection of the environment [5.10]. In this context, ‘reference organisms’ are defined as a “series of entities that provides a basis for the estimation of the radiation dose rate to a range of organisms that are typical, or representative, of a contaminated environment.”

A similar concept has been applied by the ICRP [5.2], which introduced a system of discrete and clearly defined RAPs for assessing radiation effects on non-human organisms. Under this scheme, an RAP was defined as:

> “a hypothetical entity, with the assumed basic characteristics of a specific type of animal or plant, as described to the generality of the taxonomic level of Family, with defined anatomical, physiological, and life-history properties, that can be used for the purposes of relating exposure to dose, and dose to effects, for that type of living organism.”

The ICRP approach is based on the consideration of 12 more or less globally representative RAPs, covering different life stages (e.g. fish egg, adult fish). This is the basis for systematically relating radionuclide exposure to radiation dose, and then dose (or dose rate) to different types of effect, for several organisms that are characteristic of different types of natural environment [5.2].

In 2009, the IAEA, within the international programme Environmental Modelling for Radiation Safety (EMRAS II), initiated a working group to develop an international handbook on estimating transfers of radionuclides to wildlife, similar to that developed for estimating transfers to human foodstuffs [5.7]. The approach adopted for classification of non-human biota was based on a reference organism concept [5.11] and was consistent with the ICRP approach to defining RAPs [5.12]. However, it was applied more generally, defining broader wildlife groups (e.g. soil invertebrate, predatory fish, terrestrial mammal). In some cases, a consideration of specific subcategories was included, for example molluscs are distinguished into bivalves, cephalopods and gastropods. This handbook has been published in 2014 [5.6].

5.3. TRANSFER PARAMETERS

In general, assessment models adopt equilibrium transfer factors appropriate to chronic exposure situations. For plants, it is usual to adopt plant:soil concentration ratios defined as the ratio of the concentration of an element or radionuclide in plants to the concentration in soil. These are typically defined in terms of FW plant and DW soil, but some authors use a DW plant and DW soil basis. The term ‘soil to plant transfer factor’ is also sometimes used. This is defined similarly to the concentration ratio, but with a standardized soil volume. Unfortunately, the term ‘transfer factor’ is also used to denote the ratio between the concentration of an element or radionuclide in an animal food product and the daily rate of intake of that element or radionuclide by the animal in diet plus drinking water. An animal transfer factor typically has units of d/kg

(FW) or d/L for milk. Recently, there has been an increasing tendency to also use concentration ratio values for animals, with the ratio being between the concentration of an element or radionuclide in an animal tissue, organ or food product and the concentration of that element or radionuclide either in the diet or in an environmental medium such as soil.

5.4. ASSESSMENT MODELS

Models used to simulate the transport of uranium and its progeny in the environment for assessment purposes may be very simple; for example, they may involve no more than a one dimensional vertical representation of advection and dispersion in the soil column, with uptake by plants estimated using an empirically derived ratio between the concentration in plants and the concentration in the underlying soil [5.13, 5.14]. However, much more complex models may also be used, for example three dimensional coupled representations of water flow, sediment movement and contaminant transport in a surface water catchment (e.g. Ref. [5.15]). It is emphasized that the level of complexity of the assessment model should be matched to the significance of the environmental issue being addressed (e.g. spatial extent and degree to which uranium concentrations are increased above typical background or baseline values) and the need for assurance that the key factors controlling the transport of uranium in the environment are included in the model, so that the robustness of the results obtained can be determined. In some situations, specific aspects may need to be investigated through detailed modelling, so that a simplified assessment approach can be derived and/or justified.

Additionally, it is often appropriate to use a tiered approach to assessment modelling. Initially, a simple screening model using very cautious, generic assumptions may be adopted. If the results from the application of such a screening model demonstrate that human health and the environment are adequately protected, then no refinement of the assessment is required. However, if adequate protection is not demonstrated by this screening approach, then more detailed modelling can be undertaken, in which the cautious, generic assumptions are replaced by more realistic and, generally, more situation specific assumptions. Successive refinement of the assessment approach corresponds to moving to higher tiers of the assessment modelling hierarchy, and also often involves a move from simple to more complex models.

In a hierarchical approach to assessing impacts on both humans and the environment, generic and cautious exposure scenarios are normally used in the lower tiers, but these are replaced by more realistic scenarios in the higher tiers. The more realistic scenarios will generally require the acquisition of additional site specific information on human habits and on the ecological characteristics on and near the site.

5.5. DATA SELECTION

Assessment models are often empirical in nature (i.e. transfers between different environmental media such as soils and plants) and are calculated based on observed relationships between concentrations in those media, but without reference to the processes controlling those transfers (e.g. Refs [5.6, 5.7]). Furthermore, these empirical data often exhibit wide distributions owing to a combination of inherent variability and a lack of knowledge of key factors influencing the observed values. In selecting data for use in a specific model, consideration should be given to the degree to which the assessment needs to be cautious in nature. Where data are provided for different environmental conditions, the dataset selected should be matched to the environmental situation being addressed.

In the past, assessments were typically deterministic, in which only point, best estimate values of input parameters were used. However, probabilistic assessments are also used, particularly in the context of solid radioactive waste management, in which multiple selections from probability distributions of input parameter values are used to generate a probability distribution of assessment results. Data distributions discussed and presented in this publication have generally been selected with a view to their potential use in both deterministic and probabilistic assessments. Probabilistic approaches are particularly relevant where there are non-linear relationships between input and output parameter values, because, in those circumstances, the best estimate of the output value will not be the same as the output value calculated using best estimates of the input parameter values.

Additionally, in probabilistic assessments, it may be appropriate to distinguish between epistemic uncertainties (i.e. those relating to a lack of knowledge) and aleatory uncertainties (i.e. those intrinsic to the situation, such as variations in habits between individuals in the exposed population).

5.6. DOSE IMPLICATIONS AND COMPARISONS

As established in Chapter 4, uranium and its progeny are ubiquitously present in the environment. As a consequence, humans and other biota are chronically exposed to external and internal radiation from uranium series radionuclides, as well as exhibiting low tissue concentrations of uranium throughout their lifetimes. For humans, the relevant exposure pathways include external exposure, largely from soils and sediments, ingestion of plant and animal products, ingestion of drinking water, inhalation of aerosols arising from soils and sediments, and inhalation of ^{222}Rn and ^{220}Rn and their progeny. Annual doses from these pathways are addressed in reports from UNSCEAR and are not discussed in detail here. However, it is important to recognize that naturally occurring uranium and thorium series radionuclides contribute a large fraction of the total worldwide per capita annual effective dose from natural background radiation (Table 5.1).

TABLE 5.1. AVERAGE WORLDWIDE EXPOSURES TO NATURAL RADIATION SOURCES

Source of exposure	Annual average effective dose (mSv)
Cosmic radiation	
Directly ionizing and photon component	0.28
Neutron component	0.10
Cosmogenic radionuclides	0.01
Total cosmic and cosmogenic	0.39
External terrestrial radiation	
Outdoors	0.07
Indoors	0.41
Total external terrestrial radiation	0.48
Inhalation exposure	
Uranium and thorium series	0.006
^{222}Rn	1.15
^{220}Rn	0.10

TABLE 5.1. AVERAGE WORLDWIDE EXPOSURES TO NATURAL RADIATION SOURCES (cont.)

Source of exposure	Annual average effective dose (mSv)
Total inhalation exposure	1.26
Ingestion exposure	
^{40}K	0.17
Uranium and thorium series	0.12
Total ingestion exposure	0.29
Total	2.4

Source: Based on table 31 of Ref. [5.16].

Much of the external terrestrial radiation originates from uranium and thorium series radionuclides, and both ^{222}Rn and ^{220}Rn are progeny of uranium and thorium [5.16]. For wildlife, typical background dose rates are also given by UNSCEAR [5.16]. These are 0.02–0.7, 0.01–0.44 and 0.022–0.18 µGy/h for terrestrial and aquatic plants, terrestrial animals and freshwater organisms, respectively. In Ref. [5.17], background weighted dose rates are deduced for terrestrial animals and plants in the range of 0.069–0.61 µGy/h. In Ref. [5.18], weighted dose rates were calculated in the range of 0.37–1.9 µGy/h for aquatic reference organisms.

5.7. COMPARISON OF ENHANCED CONCENTRATIONS WITH TYPICAL VALUES

A further implication of the ubiquitous presence of uranium in the environment is that anomalies, whether arising from natural processes or from human activities, must be identified and characterized, and their significance evaluated, in the context of this background. Where a new activity or change of activity involving uranium is contemplated, a baseline survey may be undertaken to determine the pre-existing situation, so that perturbations to the environmental distribution or form of uranium due to the new activity or change of activity can be characterized. It is emphasized that a baseline survey determines the pre-existing local situation and does not necessarily represent a measurement of natural background. Furthermore, even if the pre-existing environment has not been perturbed by human activities, what is measured is the local background, and this cannot necessarily be taken as representative for other locations or at a larger spatial scale, since the distribution of uranium in the environment is highly heterogeneous.

In undertaking a baseline survey, one interest may be in determining the extent of a local anomaly in the distribution of uranium relative to the typical distribution defined at a larger spatial scale. Although the total concentration of uranium in environmental media, such as soils, sediments, groundwaters and surface waters, will often be a useful measure of the extent and nature of the anomaly, it is not the only potentially useful measure. Specifically, measurements of isotopic ratios may be helpful where the anomaly originates from a source of depleted or enriched uranium, and measurements of the bioavailable fraction of uranium (e.g. by use of serial extraction procedures) or of specific chemical species may identify changes in the vicinity of the anomaly, even though total concentrations of uranium are not much altered.

Where changes in the environmental distribution of uranium are anticipated (e.g. owing to development of a groundwater plume due to leaching of uranium from waste material or parent rock), it is likely to be necessary to track those changes in a sequence of surveys. However, it may not be appropriate for those surveys to be undertaken over the same spatial domain or at the same level of detail as the original baseline survey. Therefore, one aim of the baseline survey should be to help to define critical locations and analyses that could be used to provide time series of results after the baseline survey to demonstrate that the situation remains adequately understood and controlled. The results from such time series could be used as an input to decisions on when and if new, more extensive surveys or interventions are required.

In interpreting the results from baseline and subsequent surveys, and in defining the time-series monitoring to be undertaken after such surveys, there is likely to be a need to use mathematical models to project the likely future changes in the situation that will occur. For example, a groundwater flow and contaminant transport model might be used to project the future development of a plume of increased uranium concentrations in groundwater with a view to defining the locations of monitoring boreholes to track the development of that plume, to estimate the environmental impacts that the plume would have when it reached a discharge zone, and/or to evaluate the potential value of mitigation measures designed to avert those environmental impacts. Spatially extended surveys, time series monitoring and mathematical modelling of future developments will generally need to be used interactively and iteratively in the management of larger scale uranium anomalies due to human activities, for example those arising from the mining and milling of uranium or from the phosphate industry.

5.8. CRITERIA ADOPTED FOR PROTECTING HUMAN HEALTH AND THE ENVIRONMENT

Quantitative information on standards and criteria adopted for protecting human health and the environment are given in Chapter 7. These standards and criteria are not necessarily comparable. Some are limiting values set at the boundary between potentially acceptable and unacceptable impacts. The occupational air quality standards promulgated by the United Kingdom Health and Safety Executive [5.19] and the Soil Quality Guidelines of the Canadian Council of Ministers of the Environment [5.20] (see Chapter 7) are of this type. In contrast, others are screening or threshold values that are appropriately considered as targets to be achieved rather than limits to be met. The minimal risk levels (MRLs) promulgated by the United States Agency for Toxic Substances and Disease Registry [5.1] (see Chapter 7) and the derived consideration reference levels of radiation dose recommended by the ICRP [5.21] are of this type. Exceedance of these values signals a need to pay attention to the situation but does not necessarily imply unacceptable adverse effects on either human health or on the environment. Indeed, such screening or threshold values are often set at a small fraction of no observed adverse effect levels (NOAELs) or lowest observed adverse effect levels (LOAELs), so exceedance of them may not imply any adverse effects on either human health or on the environment. Other standards and criteria, such as radiation dose constraints, may be intermediate in nature, for example adopted as bounds on optimization for a single source, facility or process where there is the potential for exposure of the receptor to several such sources, facilities or processes. With such standards and criteria, a specified fraction of a limiting value is defined, and the associated requirement is that the standard or criterion should not be exceeded because of activities related to the single source, facility or process.

In addition to quantitative standards and criteria, protection of human health and the environment from activities involved in the handling, processing and transport of uranium-containing materials include a requirement for optimization. This can be expressed in various ways, for example a need for radiation doses, risks or environmental impacts to be as low as reasonably achievable or a requirement to use best practicable means or best available technology. However, the general principle is in all cases similar, which is that all reasonable means should be taken to reduce adverse impacts below the levels implied by just complying with the relevant limiting standard (or constraint, where a source related standard is applied). However, in most contexts in which uranium is handled, processed or transported, other radioactive substances and/or toxic chemicals will also be handled, processed or transported. Optimization applies to the whole process and needs to consider all of the adverse impacts together with the benefits. Thus, the implications of exposures to uranium cannot be addressed in isolation but should always be considered in this wider context.

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Chapter 6

QUANTIFICATION OF ENVIRONMENTAL TRANSFER PROCESSES OF URANIUM

S. FESENKO, T. YANKOVICH, A.R. HARBOTTLE
International Atomic Energy Agency

O. VOITSEKHOVYCH
Ukrainian Hydrometeorological Institute, Ukraine

T.E. PAYNE
Australian Nuclear Science and Technology Organisation, Australia

N.G. MITCHELL
Eden Nuclear and Environment Ltd, United Kingdom

M.C. THORNE
Mike Thorne and Associates Ltd, United Kingdom

M. VIDAL, A. RIGOL
University of Barcelona, Spain

F.P. CARVALHO
University of Lisbon, Portugal

6.1. GENERAL PRINCIPLES

Uranium released to the environment is transported in and between different environmental media, such as air, water, soil and sediments, by various physical, chemical and biological processes. In the terrestrial environment, the most important medium in which transport occurs is soil, while in aquatic environments, there are different components, such as streams, rivers, lakes, coastal waters, and marine and riverbed sediments. Radionuclides are transferred to terrestrial (plants, invertebrates and vertebrates) and aquatic organisms (plants, phytoplankton, zooplankton, macroinvertebrates, fish, amphibians, crustaceans, mammals and birds) through the food chain.

The ICRP [6.1] developed a set of 12 reference organisms, representative of all biotic species, to be used in environmental assessments and adapted for the conditions of specific site investigations (e.g. caribou in northern Canada).

Ecological risk assessment is based on a conceptual model of the studied site, in which its specific components are defined, such as media and biological environment, radiation sources and exposure pathways for the relevant representative organisms. Habitats and feeding habits of an organism are key elements in selecting the reference organisms, as these are directly related to the potential exposure to radionuclides.

To understand the effects of radiation on non-human biota, it is important to understand the relationship between the radionuclide activity concentration levels in various environmental media from the organism's habitat and the dose rates to the organism or to the organism's tissue or organ. All internal and external exposure pathways, such as inhalation, ingestion, or uptake through skin or membranes, are to be considered for the dose rate estimations. Radionuclide activity concentrations in the organism are commonly determined from data on concentrations in environmental media in which the organism resides, using empirical data of concentration ratios [6.2]. The conversion of radionuclide activity concentrations in environmental media to dose rates to organisms is achieved through dosimetric models, using specific dose conversion coefficients for selected non-human biota (see Chapter 7).

This chapter presents data for the evaluation of the transport and distribution of uranium in the environment, following, in general, the classification schemes suggested in Ref. [6.3] for agricultural plants and animals, and in Ref. [6.4] for wild plants and animals (see Ref. [6.5] for further information).

6.2. TERRESTRIAL ENVIRONMENT

6.2.1. Transport of uranium in groundwaters

General concepts related to contaminant transport in groundwater systems can be found in Refs [6.6, 6.7]. Groundwater movement follows the so-called hydraulic gradient, and it can be described by the simplified equation of Darcy's law for the specific discharge, q, as follows:

$$q = -K\frac{\mathrm{d}h}{\mathrm{d}l} \tag{6.1}$$

where

K is the hydraulic conductivity constant, which is a function of the aquifer material and can vary over a large range (10^{-12}–0.1 m/s);
dh represents the change in hydraulic head;

and dl is the length.

The average groundwater velocity, $\bar{v}$, or seepage velocity, is calculated by dividing the specific discharge by the porosity, n, of the medium:

$$\bar{v} = -\frac{K}{n}\frac{\mathrm{d}h}{\mathrm{d}l} \tag{6.2}$$

The water moves only through the pore openings rather than through the entire rock (soil) volume [6.7]; therefore, the actual groundwater velocity can be much faster than the Darcy velocity. Water flow cannot be simply described by Darcy's equation for specific geological environments such as fractured rocks or the vose zone (particularly important in radionuclide transport studies). Reference [6.8] states:

> "The movement of a contaminant is affected by a number of processes that must be considered in addition to the rate of water movement.
>
> "*Advection* is the movement of solutes that are carried along with the flowing groundwater. The one-dimensional mass flux is estimated by multiplying the quantity of water per unit time by the concentration of dissolved solids.
>
> "*Molecular diffusion* is the movement of a solute from an area of greater concentration to lower concentration. This can occur even in the absence of groundwater movement and can be considered a consequence of random molecular motions. In one dimension, it is described by Fick's law…
>
> "*Mechanical dispersion* is a process by which a solute in the groundwater becomes spread out through a larger volume. This is because groundwater actually travels at various rates in a real aquifer, both faster and slower than the average linear velocity. The flow is faster in the centre of pores compared to the edges, and also faster in larger pores (fractures). Some flow paths will be longer (more tortuous) than others. As a result, the solute becomes dispersed."

As discussed in more detail in Chapter 3, uranium mobility in groundwater is impacted by several aspects of its biogeochemistry, including the following:

- Oxidation state: Uranium mainly occurs in either its reduced state (U(IV)), which is generally highly immobile, or its more soluble and mobile (U(VI)) state.
- Solubility: The solubility of uranium minerals generally puts an upper limit on their concentration in solution.
- Complexation: The association of dissolved uranium with organic and inorganic ligands to form various species in solution has a significant impact on its environmental mobility.
- Sorption: The processes by which dissolved uranium is bound on the surfaces of naturally occurring substances are often collectively referred to as sorption; the nature of this binding can include processes such as adsorption on iron oxide surfaces, ion exchange in the interlayers of clay minerals, or binding by humic substances.
- Colloidal transport: In some circumstances, radionuclides may form colloids (very small particles) that may be mobile in groundwater and, in some situations, may substantially increase the mobility of a radionuclide; this is typically more important for radionuclides that are relatively insoluble (such as Th(IV)) but could influence the mobility of uranium, particularly in its reduced oxidation state (U(IV)).
- Geomicrobiology: The behaviour of uranium can be significantly influenced by interactions with ubiquitous microorganisms.

The above basic principles of uranium chemistry, together with the general principles of groundwater movement and solute transport, are the main determining factors in impacting uranium mobility in groundwaters.

6.2.2. Soil–uranium interactions

6.2.2.1. Sorption processes: The solid–liquid distribution coefficient concept

Dissolved radionuclide ions can bind to solid surfaces by several processes often classified under the broad term of sorption, which affects radionuclide transport in the soil profile, radionuclide accumulation in surface soils and subsequent radionuclide transfer to other environmental compartments[8]. Sorption is element and soil type dependent. On the one hand, sorption is controlled by the radionuclide chemical form and speciation. On the other hand, sorption is also

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[8] This section is based on Ref. [6.2].

affected by soil constituents (e.g. soil mineralogy, iron oxides and hydroxides, organic matter) and soil geochemistry (e.g. pH, redox potential, presence of colloids, presence of competitive species and counterions).

The sorption process is usually quantified by determining the solid–liquid distribution coefficient, that is, the ratio of the concentration of the radionuclide in the soil solid phase to the concentration in the (soil) solution, K_d (L/kg), by applying given experimental and operational procedures, both in laboratory and field conditions. The K_d coefficient is, thus, an operationally defined parameter and the method used for its quantification may significantly contribute to its variability.

The K_d approach is based on the equilibrium concept. It relies on the hypothesis that the radionuclide in the solid phase is in equilibrium with the radionuclide in solution and can, thus, exchange with it. However, the nature of the sorption process and the specific interactions of the target radionuclide in soils mean that the elapsed time since the addition of the radionuclide may affect the value of K_d. Thus, for example, a fraction of the added radionuclide may become fixed by the solid phase, even in a short time, owing to quasi-instantaneous irreversible sorption, co-precipitation on soil components and/or sorption dynamics [6.9], whereas exchange reactions may take hours or longer to approach equilibrium. Most laboratory experiments based on batch tests with recently added radionuclide are, in principle, designed to obtain the so-called exchangeable K_d ($K_{d,exch}$). However, to consider a K_d value deduced from a laboratory test as $K_{d,exch}$ is merely hypothetical (since it may include a rapid, but irreversible, component and may not represent equilibration of a longer term exchangeable component), although this is often accepted by the scientific community.

K_d values can also be straightforwardly quantified from the radionuclide concentration in the solid phase divided by the concentration of the radionuclide in the soil solution obtained from the contaminated soil, which is an approach often used for indigenous elements [6.10, 6.11]. This approach may lead to higher K_d values than those resulting from batch tests with recently added radionuclides, because the radionuclide quantified in the solid phase of the contaminated soil may include sorbed or incorporated activity not readily available for exchange with the soil solution. However, in some approaches, the concentration in the solid phase used for the K_d calculations is not the total radionuclide concentration, but that resulting from extraction with a specific extracting reagent (e.g. ionic exchange reagent or chelating agent) or a sequence of such reagents [6.12]. This can effectively exclude the irreversibly bound radionuclide from consideration, i.e. the pool of radionuclides considered in the solid phase is smaller and, thus, the resulting K_d value is lower.

Overall, the large number of experimental approaches used to quantify K_d values, and the contrasting experimental conditions applied in each case, may contribute to the large variability in K_d values that has been obtained for similar soil and radionuclide combinations.

6.2.2.2. Effect of soil properties on uranium sorption

Soil properties, such as pH, soil texture, specific surface area (SSA), organic matter content, cation exchange capacity, content of amorphous iron oxides, and carbonate and phosphorus status, are known to affect uranium sorption [6.13, 6.14]. The EPA [6.13] performed an extensive review of K_d(U) values for soils, crushed rock material and single mineral phases, which indicated that pH and dissolved carbonate concentrations were the two most important factors influencing the sorption behaviour of U(VI), which is the dominant uranium species in top soils. At pH values <5, U(VI) is present as the uranyl ion, UO_2^{2+}. At a higher pH, the uranyl ion hydrolyses, forming several aqueous hydroxide complexes, which dominate U(VI) speciation in the absence of dissolved inorganic ligands (carbonate, fluoride, sulphate and phosphate). In the pH range of 6–10, highly soluble carbonate complexes dominate (the acids biphosphate, bicarbonate and tricarbonate) [6.15]. Since uranium speciation is related to pH, the K_d(U) values show a specific trend in relation to the pH. In general, the sorption of uranium by soils is low at pH values <3, increases rapidly with increasing pH from 3 to 5, reaches a maximum in the pH range of 5–7 and then decreases with increasing pH at pH values >7 [6.13].

The effect of soil characteristics on uranium sorption was explored in Ref. [6.16]. No significant effect of clay or organic matter content was found. However, a significant relation between soil K_d(U) and pH was deduced. For soils in the pH range of 5.5–8.8, a linear relationship for a reduced soil dataset was obtained as follows:

$$\lg K_d = (-1.3 \times \text{pH}) + 11; \quad R^2 = 0.76 \tag{6.3}$$

The effect of soil pH on uranium availability for spiked soils was examined, and the following similar pattern was observed for soils with pH ≥ 6:

$$\lg K_d = (-1.2 \times \text{pH}) + 10.8; \quad R^2 = 0.65 \tag{6.4}$$

which was explained by the increased amount of soluble uranyl–carbonate complexes at high pH. In Ref. [6.17], the following correlation was found for soils in the pH range of 5.5–8.8:

$$\lg K_d = (-1.1 \times \text{pH}) + 9.8; \quad R^2 = 0.41 \tag{6.5}$$

The relative low percentage of explained variance by the K_d(U)–pH correlations when considering soils and data from varying sources and heterogeneous datasets indicates that K_d(U) cannot be uniquely predicted from pH variation and that other soil properties should be considered. Significant sources of variability in the relationship between K_d(U) and pH, besides the methodology used for their determination, are postulated to be the heterogeneity in soil mineralogy (e.g. soils containing larger percentages of iron oxide minerals and mineral coatings, clay minerals exhibiting higher sorption than soils dominated by quartz and feldspar minerals) and the organic matter content. Updated K_d versus pH and clay correlations have been suggested [6.18] on the basis of a dataset enriched with a significant contribution of field data with indigenous uranium, as follows:

$$\text{pH} < 5.5\text{: } \lg K_d(\text{U}) = 1.75 + (0.0145 \times [\text{clay}] \times \text{pH}) \tag{6.6}$$

$$\text{pH} \geq 5.5\text{: } \lg K_d(\text{U}) = 9.05 - (0.989 \times \text{pH} + 0.0029 \times [\text{clay}] \times \text{pH}) \tag{6.7}$$

where [clay] is the percentage of clay present.

Many studies also highlight the importance of iron oxides and hydroxides for the sorption of uranium [6.19–6.22]. The positively charged uranium species are sorbed to the negatively charged surfaces of the sesquioxides, or uranium species become structurally incorporated into the iron oxides (coatings) over many dissolution–precipitation cycles of these amorphous or poorly crystalline materials [6.23]. Organic matter and clay minerals provide exchange sites that are expected to increase sorption of UO_2^{2+} and other positively charged forms of uranium. The influence of organic matter on uranium mobility is twofold: (i) a decreased mobility through sorption by exchange; and (ii) an increased mobility following formation of soluble organic complexes for those samples having significant dissolved organic matter content and colloids. In the absence of large amounts of organic matter and clay, and of counterions with which uranium may form insoluble compounds (such as phosphates), uranium is relatively mobile. The role of humic substances in the mobility of U(VI) was investigated and uranium was found to be strongly retained by the soil solid phase, mainly owing to organic aggregates and organic coatings on quartz minerals [6.24], whereas a large fraction of U(VI) was found to be associated with humic colloids in soil, thus forming a potential mobile uranium phase. For soils with pH < 6, very significant correlations for a controlled, reduced soil dataset between K_d(U) and the fraction of organic matter were reported [6.25]:

$$K_d\,(\mathrm{U}) = (-1963 \times [\text{organic matter}]) - 5432; \quad R^2 = 0.41 \tag{6.8}$$

The same was true for amorphous iron content:

$$K_d(\mathrm{U}) = (1.02 \times [\text{amorphous Fe}] + 1691; \quad R^2 = 0.88 \tag{6.9}$$

Considering the whole pH domain, these relationships remained significant:

$$K_d(\mathrm{U}) = (1591 \times [\text{organic matter}]) - 3362; \quad R^2 = 0.70 \tag{6.10}$$

$$K_d(\mathrm{U}) = (1.08 \times [\text{amorphous Fe}]) + 2783; \quad R^2 = 0.88 \tag{6.11}$$

SSA is also thought to play a significant role in uranium sorption, as has been shown for pure mineral phases [6.14]. K_d(U) comparisons among materials may improve when values are normalized with respect to the SSA. Constraints on the use of SSA to reduce K_d(U) variability and enhance K_d(U) comparison among geological materials arise because SSA values are usually not reported, and specific sites, rather than SSA, may have a dominant role in sorption.

Uranium is adsorbed on a range of amphoteric oxide minerals (such as iron, aluminium and titanium oxides) [6.20, 6.26]. Thus, a surface area normalized distribution coefficient (K_a value) was introduced for uranium sorption, determined by the ratio K_d:SSA [6.13, 6.14, 6.27]. The units of K_a are millilitres per square metre (differing from K_d, which is expressed in mass normalized units, such as litres per kilogram or millilitres per gram).

A detailed experimental study [6.14] of the relationship between K_d and K_a, performed with uranium and a range of minerals under standardized conditions, showed that K_a curves for uranium sorption by different minerals across the pH range from 3 to 7 conform reasonably well (Fig. 6.1). K_a showed a similar strong dependency as K_d on pH for the experimental conditions studied. However, when the K_a data are plotted as a function of pH and compared to the corresponding K_d curves, significantly less scatter is observed (Fig. 6.1). The K_a parameter was useful in representing U(VI) sorption data obtained across a range of conditions on ferrihydrite, kaolinite, montmorillonite and weathered rock materials. The data show that the variation between different soils and minerals was partially explained by differences in SSA between samples. When this dependency on surface area was removed, the results (expressed as K_a) were in much closer agreement.

These findings imply that K_d values for U(VI) sorption on soils can be estimated from surface area measurements of comparable materials and in similar chemical conditions. Additional information on the variation of K_a in the presence of ligands can also be useful in constraining the range of K_d values.

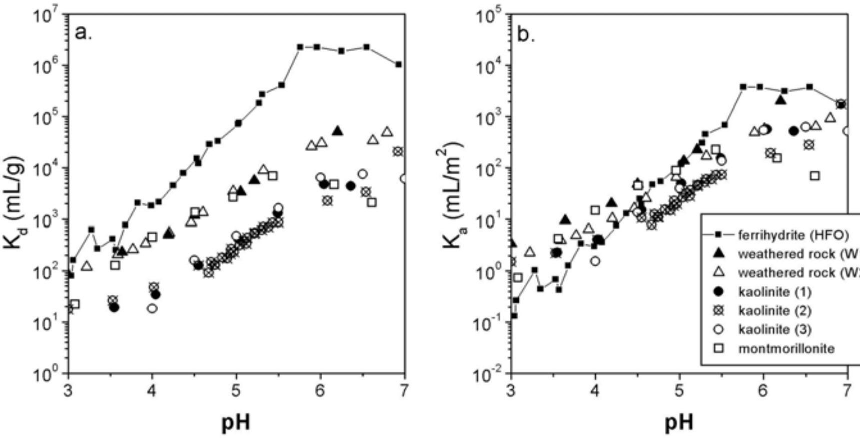

FIG. 6.1. The sorption of U(VI) by different minerals across the pH range from 3 to 7 expressed as: (a) distribution coefficient (K_d) and (b) surface area normalized distribution coefficient (K_a) (the ratio K_d:SSA). It should be noted that there is a much smaller spread of experimental data in the computed K_a compared to the range of K_d for these minerals. The experimental conditions are comparable for each set of experiments and are given in Ref. [6.14] (reprinted from Ref. [6.14], Copyright (2011) with permission from Elsevier).

6.2.2.3. Best estimates for K_d*(U) values*

Many studies have aimed at suggesting best estimates for K_d values. The K_d estimates should be suitable for use in the assessment of radionuclide fate in soils based on models that require specific K_d values as input data, even when specific information on soil properties is not available. A suitable approach to do this is to use probabilistic models to derive most likely (often the 50th percentile of the distribution) K_d values, based on statistical functions describing the overall variability of K_d values, and to derive confidence intervals at desired significance levels. As log-normal distributions of the K_d values are often assumed, best estimates can also be derived from a geometric mean (GM), when probabilistic models cannot be constructed.

Although generic data may not be appropriate for assessing site specific impacts, they may be useful for preliminary or screening assessments. Owing to the strong dependence of K_d(U) on soil properties and uranium speciation, the use of a single K_d(U) value for all soils cannot be supported scientifically and should be generally advised against, as the minimum and maximum values may range over up to five orders of magnitude. A first approach to deriving more appropriate K_d(U) best estimates with a lower inherent variability may be based on the use of parametric equations with those key soil variables governing uranium sorption in soils, such as the previously described K_d versus pH, K_d versus [amorphous Fe] or K_d versus [organic matter] equations. However, when dealing with large datasets with data from different sources and obtained in diverse experimental conditions, univariant linear correlations often fail to describe K_d variability and do not predict K_d values appropriately; they are not applicable within the whole range of the target soil property (as is the case of the U-shaped dependency of K_d(U) on pH) and there is a general lack of soil data to permit constructing multivariant non-linear correlations.

Another strategy followed in the past focuses on reducing K_d variability by suggesting derived best estimates for soils grouped according to the range of value of a key soil variable affecting uranium sorption. This approach can be adopted to the extent that a minimum of soil characterization information is available, which often is not the case. For instance, data on carbonate, SSA or amorphous Fe concentration, which are known to affect uranium sorption, are seldom available. Thus, other soil properties must be chosen as surrogates of the soil properties governing uranium interaction.

A first approach to suggesting K_d(U) best estimates with a reduced variability was based on the use of texture and organic matter content, as the values of these two variables are often available [6.9, 6.17, 6.28]. Although the best estimates derived from these compilations were not the same, owing to a number of differences in the criteria for data acceptance in the respective datasets (e.g. geological materials accepted or not in the dataset, pH restrictions), the K_d(U) best estimates from these compilations indicated that sorption in organic soils is around one order of magnitude higher than that in mineral soils, whereas differences among mineral soils according to their texture are predicted to be of minor significance.

A better soil variable used in suggesting K_d(U) best estimate values is the pH at which the sorption takes place, which strongly affects the uranium species present in the soil solution and, thus, participating in the sorption process. This approach was followed in the data collection in Ref. [6.9], which served as a basis for the K_d(U) best estimates included in Refs. [6.2, 6.3]. In this case, it was determined that the K_d(U) best estimate for the pH range of 5–7 was one order of magnitude higher than for lower or higher pH ranges.

The dataset in Ref. [6.9] has been recently updated using Ref. [6.29] and subsequent works. In the updated dataset, data from pure soil phases (e.g. Fe/Mn oxides, clay minerals) and marine and freshwater sediments were excluded. Reference values (e.g. from previous compilations) and data derived from parametric equations or mass-transport experiments were not considered either. Furthermore, data originating from sets of tests in which the effect of the variation of a given operational variable on the K_d quantification was examined (e.g. contact time in batch experiments) were pooled to become a single entry. In parallel, data from similar geological materials that could be significant in specific terrestrial environments, such as subsoils and till samples, were also included in the general dataset, as differences among the derived K_d values were not statistically significant. Finally, data originating from long term experiments, mainly experiments including indigenous uranium, were also accepted [6.12].

An early examination of the K_d(U) values indicated that the overall K_d(U) for the whole dataset was around 345 L/kg, in agreement with previous reported values [6.2, 6.9, 6.17], although related to a variability of nearly five orders of magnitude. Thus, it is recommended not to use this value, unless no additional soil characterization data, such as pH or organic matter content, are available.

Another preliminary examination focused on the effect of the experimental approach used to derive the K_d(U) values and of the time elapsed since contamination on the K_d(U) values. Two cases were distinguished: (i) a short term scenario, including data from batch sorption and desorption experiments with recently added uranium; and (ii) a long term scenario, originating mostly from data derived from indigenous uranium. The statistical analyses performed showed that the possible effect of the experimental methodology and interaction dynamics was not unequivocally proven and that it was much lower than that of soil properties governing uranium sorption.

6.2.2.4. K_d*(U) best estimates based on texture and organic matter content criteria*

Partial datasets of K_d(U) values were created based on texture (sand, loam and clay soils) and organic matter content (organic soils), following the criteria previously agreed [6.3]. In short, a soil was included in the organic group if its organic matter content was >20%. For mineral soils, three groups (sand, loam and clay) were created according to the criteria given in Section 5.2.

A better description and modelling of data variability was achieved by fitting data to a probability function (tested as log-normal for all of the overall and partial datasets), instead of merely giving minimum and maximum values of the datasets. Thus, the best estimate value (quantified as 50th percentile) the geometric standard deviation (GSD), and the 5th and 95th percentiles of the cumulative density function based on a log-normal distribution were also calculated.

Table 6.1 summarizes the updated K_d(U) best estimates using the organic matter criterion, whereas Fig. 6.2 shows the cumulative distribution functions fitted to the mineral and organic soils. Associated characteristics of those distributions (GM, GSD, and 5th and 95th percentiles) are given in Table 6.1. The K_d(U) best estimate values significantly differed between mineral and organic soils. This confirmed the role of organic matter in the sorption of uranium in soils

TABLE 6.1. K_d(U) (L/kg) GROUPED BY THE ORGANIC MATTER CRITERION

Soil type	N[a]	Best estimate value	Geometric standard deviation	5th percentile	95th percentile
Mineral	171	260	8.8	7	9 250
Organic	30	1 700	6.0	90	31 975

[a] N: Number of individual data entries.

as observed in previous compilations, as the best estimate of the K_d(U) values for organic soils was six times higher than that of the mineral soils.

The 5th–95th percentile range of values showed that 90% of the K_d(U) values ranged over two to three orders of magnitude. Thus, the initial K_d(U) variability was decreased owing to the grouping using the mineral or organic nature of the soils, although the resulting ranges were still quite wide. Additional analyses showed that K_d(U) best estimates were not statistically different among soil texture groups.

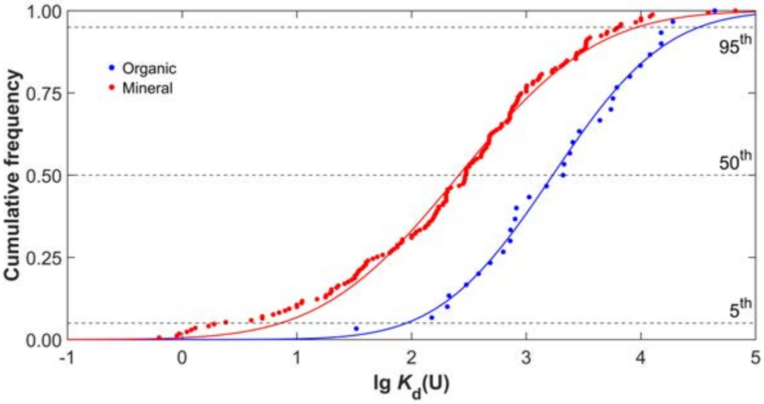

FIG. 6.2. Cumulative distribution functions of K_d(U) for mineral and organic groups.

TABLE 6.2. K_d(U) (L/kg) GROUPED BY THE pH CRITERION

pH	N^a	Best estimate value	Geometric standard deviation	5th percentile	95th percentile
pH < 5	82	285	7.5	10	7 805
5 ≤ pH < 7	94	1 410	4.7	110	17 950
pH ≥ 7	111	100	11	2	4 925

[a] *N*: Number of individual data entries.

6.2.2.5. K*$_d$(U) best estimates based on pH*

The K_d(U) values were also grouped based on the pH at which sorption takes place. Table 6.2 presents the K_d(U) values grouped using three pH categories defined by consideration of the uranium speciation [6.9, 6.16, 6.30], whereas Fig. 6.3 illustrates the cumulative distribution functions obtained for the three pH categories.

A significant increase in K_d(U) was observed with increasing soil pH, reaching a maximum within the pH range of 5–7, in agreement with a previous analysis [6.9] and the expected competition of the uranyl cation with protonated sites at low pH values. At higher pH, K_d(U) would decrease, owing to the

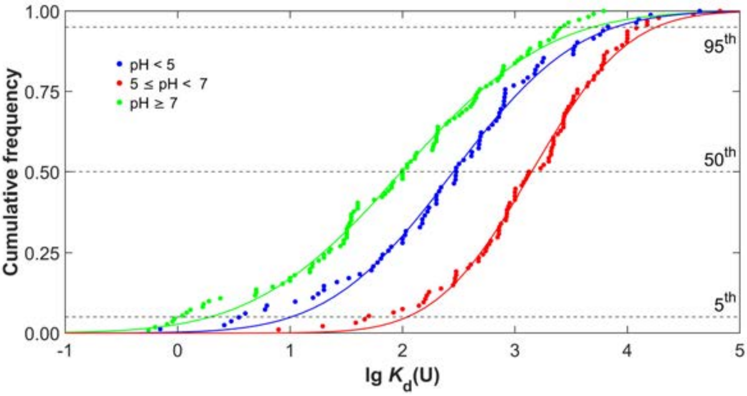

FIG. 6.3. Cumulative distribution functions of K_d(U) for soil pH groups.

formation of stable, weakly sorbing uranyl carbonate complexes [6.14, 6.31]. Variability was also decreased by two to three orders of magnitude based on this grouping criterion.

6.2.2.6. K_d*(U) best estimates based on a combination of factors*

If information on organic matter content and pH is available, it is recommended to use a K_d(U) value based on both factors, as summarized in Table 6.3.

The simultaneous use of the two criteria confirmed the main trends observed with the single criterion, but the variability was much reduced. For a given pH group, organic soils always had larger K_d(U) values than the respective mineral group, whereas K_d(U) values for the intermediate pH group were always the highest. Further variability reduction for mineral samples using texture information failed owing to the low number of entries, especially of clay samples, and differences among textural groups were not statistically significant. However, K_d(U) for sandy samples was systematically lower than that for samples of clay and loam together.

TABLE 6.3. K_d(U) VALUES (L/kg) grouped by pH AND ORGANIC MATTER criteria

pH	Soil type	*N*[a]	Best estimate value	Geometric standard deviation	5th percentile	95th percentile
pH < 5	Mineral	67	195	8.1	6	6 085
	Organic	12	695	3.4	91	5 315
5 ≤ pH < 7	Mineral	74	1 015	4.6	82	12 560
	Organic	15	4 200	2.5	919	19 210
pH ≥ 7	Mineral	93	75	8.8	2	2 610
	Organic	5	500	7.0	n.a.[b]	n.a.[b]

[a] *N*: Number of individual data entries.

[b] n.a.: not applicable.

6.2.2.7. Conclusions

A single K_d(U) value cannot be suggested as an input to risk assessment models for all the possible scenarios to be examined. Instead, modellers, end users and related stakeholders should try to gather all relevant available information about the scenario (including soil properties) in which the risk is to be assessed to use the most suitable K_d(U) best estimate and distribution, with an associated lower variability. It may be critical to select the appropriate K_d(U) best estimate and distribution according to soil properties, as large differences were evidenced with variations among K_d(U) best estimates of up to two orders of magnitude depending on differences in the pH and organic matter content.

Although K_d(U) datasets are continually updated, there are still evident gaps of values of K_d(U) for specific scenarios, such as the case of organic soils at high pH and of clay soils. However, these samples are of minor environmental relevance. Although data from other geological materials, such as subsoils and till, may be used to enhance soil datasets, these data must be statistically evaluated and critically reviewed prior to being considered as analogues to enrich the datasets.

6.2.3. Uranium soil to plant transfer

In general, uranium concentrations in plants are several orders of magnitude lower than in soils. There is no evidence that uranium is essential or beneficial to plant growth, but many plants will absorb uranium and translocation does occur within plants.

There are many studies of uranium uptake by plants near mining or processing facilities that have been performed in support of environmental risk assessments. The uranium levels at these sites can be enhanced and consideration has been given to the potential effect of uranium toxicity on plant growth at contaminated sites [6.32]. A study of Scots pine grown on soil from a uranium contaminated waste site indicated that the plant growth was unaffected. Plant yield data indicated that the phytotoxic concentration of uranium was >100 μg [U]/g soil. In a later study, a predicted no-effect concentration for chemical toxicity of uranium to terrestrial plants of 250 mg [U]/kg dry soil was suggested [6.33]. Although few data exist, potential phytotoxic effects were reported, indicating that plant responses appear to vary between species; for example, the yield of lettuce grown on soil contaminated with uranium at levels ranging from 39 to 252 mg/kg, was lower at the higher soil concentration [6.34]. In a further study [6.33], no consistent differences between concentration ratios measured at contaminated sites and those at control sites with natural background levels of uranium were found, although the discussion by the authors cautioned that fewer data were available from uncontaminated sites because natural levels were not detectable in either soils and/or plants.

The IAEA handbook of parameter values [6.3] describes the plant uptake of a radionuclide from soil using the transfer factor, defined as the ratio of the DW concentration in the plants to the DW concentration in a specified soil layer (to a depth of 10 cm for pasture crops and a depth of 20 cm for other crop types). The DW concentration is used for all plants to reduce potential uncertainty. Soil to plant transfer factors for uranium presented in Ref. [6.3] were reviewed and supporting data published by the IAEA [6.2]; values reported in the literature on a fresh plant weight basis were converted to a DW basis using the FW to DW conversion factors in table 2 of Ref. [6.2]. Reference [6.3] presents uranium data for plant uptake in agricultural systems located in temperate regions, while Ref. [6.35] presents revised data for tropical regions and for transfer to rice. It also discusses radionuclide transfer in subtropical regions, fruit growing in temperate regions, transfer in forests (trees, mushrooms and berries) and transfers in polar and alpine ecosystems; however, these sections do not include information on uranium. There is also a lack of information on the accumulation of uranium deposited onto above ground plant surfaces. Studies have recognized that uranium associated with soil and dust deposited on plant surfaces can influence the transfer factor [6.36] but information on foliar absorption of uranium is lacking.

Data for temperate environments were further discussed in Refs [6.37, 6.38]. Earlier reviews include Refs [6.36, 6.39–6.41]. Plant uptake of uranium has received greater attention over the past two decades, with increasing interest in naturally occurring radionuclides and the cleanup of uranium contaminated sites. For example, the IAEA [6.2] review for uranium transfer factors in temperate environments reported about 910 values referencing pre-2008 publications. An earlier review [6.40] referenced pre-1998 material and reported about a hundred uranium soil to plant transfer factors.

Transfer factors are influenced by aspects such as the physical and chemical form of the uranium when it enters the environment, time after release, soil properties and their management, type of crop, stage of plant growth and agricultural practice [6.2, 6.3]. The occurrence of uranium in different matrices (e.g. type of mineral rock, soil particles, tailings, fuel particles) is of interest in the measurement and reporting of uranium transfers. The transfer factors reported in Ref. [6.3] consider soluble forms of uranium. However, at sites exhibiting uranium contamination, the extraction and subsequent treatment of uranium leads to a broad range of waste types and sources that have different physicochemical characteristics, ranging from uranium incorporated into the mineral phase to releases of soluble forms. This affects both the observed transfers and the selection of appropriate values for use in risk assessments. For example, soil contaminated with a dust derived from crushed uranium ores is likely to produce a lower transfer factor than that derived from soil receiving contaminated waters [6.34].

The inadequacies of the transfer factor concept are widely recognized, not least because of the inherent variability of this quantity. Indeed, uranium transfer factors presented in Ref. [6.3] vary by over five orders of magnitude. However, there is currently no alternative available that covers the broad range of radionuclides considered in radiological assessments. It is now common for transfer factors to be reported as a GM. This is based on several studies showing that data follow a log-normal distribution more closely than a normal distribution [6.42, 6.43].

6.2.3.1. Processes influencing the uptake of uranium by plants from soil

The extensive work on uranium solubility in soils is not reflected in the information that is available on the uptake and translocation of uranium in plants as affected by soil properties [6.2]. The knowledge of the physiological response of plants to, and the bioavailability of, uranium at the soil–root interface is poor. It is suggested that uranium binds strongly to cell walls and that this limits absorption by roots and translocation within the plant (e.g. Ref. [6.36]). Uranium accumulation and translocation in oilseed rape, sunflower and wheat were investigated [6.44], and it was shown that roots exposed to UO_2^{2+} adsorb and accumulate by precipitation on cell walls, and that there is little translocation to shoots. Reference [6.44] also reported that organic complexation (carbonate or citrate) reduces retention in roots and increases translocation to the shoots, and that complexation with phosphate reduces accumulation in all plant tissues by retaining uranium on root epidermal cells. In Ref. [6.45], the relationship between micronutrient deficiency (iron) and uranium uptake and tolerance in native plant species growing at a former uranium mining site was considered. The observations suggested that U(VI) was reduced to U(IV) at the root surface. Uranium(IV) is known to form strong complexes with naturally occurring organic materials but it was not clear whether this was caused by plant reductase activity or soil microbes. More recently, it was noted [6.46] that it is not yet fully established whether fungi are capable of U(IV) reductive immobilization.

Observations of uranium transfer to plants at sites with varying soil uranium concentrations [6.39] show that concentrations are positively correlated, but that the relationship is not linear. This was in contrast to a linear response in gardens at Port Hope, Canada that had been reported earlier [6.47]. A decline in concentration ratio with increasing substrate concentration was reported for crops of pea, brassicas, maize, grass and clover [6.48]; for a marshland plant, *Spartina densiflora* [6.49]; and in a study of Mediterranean flora [6.50]. Uranium transfers to plants at a contaminated mine site were observed and compared with transfers at a control area unaffected by mining activities [6.51]. Contradictory evidence based on uranium soil concentrations was found; while the linearity assumption was valid over the complete range, it was less valid when zones of high and low concentration were examined separately.

The uptake of different chemical forms of uranium was studied [6.52] and it was found that the free uranyl cation (UO_2^{2+}) was the form of uranium most readily accumulated by plant roots. Work in Ref. [6.53] showed that weak organic acids enhanced root uptake. A feature of this and other studies [6.54, 6.55] is the use of geochemical speciation codes to interpret results and assist with experimental design. This approach was used in Ref. [6.56] but the authors were cautious about its application in predicting plant uptake, and it has been noted by others that speciation following absorption is plant specific [6.55]. It has been observed that UO_2^{2+}, uranyl carbonate complexes and $UO_2PO_4^-$ appear to be the uranium species that are preferentially taken up by the roots and transferred to the shoots, but no relationship was found between soil solution uranium concentration and plant uptake [6.57]. The pH dependency of uranium uptake is not straightforward, with observations showing enhanced root uptake at lower pH [6.53], but enhanced translocation at higher pH [6.57].

Recent land remediation studies have considered the extraction of uranium from soil by different plant species [6.53, 6.58–6.62]. Reference [6.63] lists over 70 plant species that have been considered for use in the phytoremediation of uranium and provides an assessment of their bioaccumulation potential. Studies have also considered the use of soil amendments to enhance plant uptake. For example, Ref. [6.64] described tests using willow and sunflower that show enhanced accumulation by a factor of 22 when citric acid is applied to soil and compared this with other studies citing factors of up to 200. In Ref. [6.60], a range of chemicals were applied to soil and it was found that S,S-ethylenediamine disuccinic acid caused the largest increase (by a factor of 18) in shoot uranium concentration of Indian mustard. Phosphates have sometimes been shown to reduce root uptake and translocation [6.59] by a factor of about 25, but other studies have shown no effect [6.25, 6.57, 6.65].

6.2.3.2. Estimation of transfer factor values

The IAEA handbook [6.3] recommendations for transfer factors in a temperate environment are presented in Table 6.4 and provides data for combinations of soil and plant group/plant parts.

Reference [6.37] reported the highest uranium transfer factors for fodder, pasture and herbs ($(2.3–6.5) \times 10^{-2}$), and the lowest values for legumes, tubers and cereals ($(2.2–6.5) \times 10^{-3}$), with significant differences ($p < 0.05$) between them (Fig. 6.4). A range difference of between two or three orders of magnitude

TABLE 6.4. SOIL TO PLANT TRANSFER FACTORS (kg/kg, DRY WEIGHT) FOR URANIUM IN TEMPERATE ENVIRONMENTS

Plant group	Plant compartment	Soil group	*N*[a]	Mean/ value[b]	Geometric standard deviation	Minimum	Maximum
All	n.a.[c]	All	781	2.3×10^{-2}	9.1	3.0×10^{-5}	1.3×10^{1}
Cereals	Grain	All	59	6.2×10^{-3}	7.7	1.6×10^{-4}	8.2×10^{-1}
		Sand	6	8.9×10^{-3}	11.0	1.9×10^{-4}	6.2×10^{-2}
		Loam	20	7.7×10^{-3}	5.1	1.6×10^{-4}	6.2×10^{-2}
		Clay	11	3.8×10^{-3}	4.0	7.6×10^{-4}	5.0×10^{-2}
	Stems, shoots	All	55	2.7×10^{-2}	7.5	3.0×10^{-5}	3.5
		Sand	6	3.4×10^{-2}	6.0	2.1×10^{-3}	1.7×10^{-1}
		Loam	25	5.4×10^{-2}	6.30	7.4×10^{-4}	3.5
		Clay	8	1.0×10^{-2}	3.6	2.8×10^{-3}	9.8×10^{-2}
Maize	Grain	All	9	1.5×10^{-2}	12.0	5.0×10^{-4}	7.1×10^{-1}

TABLE 6.4. SOIL TO PLANT TRANSFER FACTORS (kg/kg, DRY WEIGHT) FOR URANIUM IN TEMPERATE ENVIRONMENTS (cont.)

Plant group	Plant compartment	Soil group	*N*[a]	Mean/ value[b]	Geometric standard deviation	Minimum	Maximum
	Stems, shoots	All	11	7.8×10^{-3}	14.0	1.6×10^{-4}	9.6×10^{-1}
Leafy vegetables	Leaves	All	108	2.0×10^{-2}	7.3	7.8×10^{-5}	8.8
		Sand	7	1.7×10^{-1}	15.0	1.5×10^{-3}	8.8
		Loam	14	4.3×10^{-2}	3.9	7.7×10^{-3}	2.7×10^{-1}
		Clay	9	3.6×10^{-3}	4.2	7.6×10^{-4}	4.3×10^{-2}
		Organic	6	1.8×10^{-1}	9.7	7.9×10^{-3}	8.0
Non-leafy vegetables	Fruits, heads, berries, buds	All	38	1.5×10^{-2}	4.2	5.2×10^{-4}	2.0×10^{-1}
		Sand	7	1.9×10^{-2}	5.5	1.3×10^{-3}	1.6×10^{-1}
		Loam	4	2.3×10^{-2}	2.2	7.6×10^{-3}	4.7×10^{-2}
		Clay	7	1.8×10^{-2}	4.2	5.0×10^{-3}	2.0×10^{-1}

TABLE 6.4. SOIL TO PLANT TRANSFER FACTORS (kg/kg, DRY WEIGHT) FOR URANIUM IN TEMPERATE ENVIRONMENTS (cont.)

Plant group	Plant compartment	Soil group	N^a	Mean/ value[b]	Geometric standard deviation	Minimum	Maximum
	Stems, shoots	All	6	5.3×10^{-2}	9.9	4.3×10^{-3}	7.1×10^{-1}
Leguminous vegetables	Seeds, pods	All	19	2.2×10^{-3}	12.0	5.4×10^{-5}	1.5×10^{-1}
		Loam	4	3.0×10^{-3}	18.0	5.4×10^{-5}	4.7×10^{-2}
		Clay	7	5.5×10^{-4}	4.7	5.7×10^{-5}	5.0×10^{-3}
	Stems, shoots	All	21	6.4×10^{-2}	14.0	7.4×10^{-4}	8.7
		Sand	6	2.8×10^{-1}	20.0	5.3×10^{-3}	8.7
		Loam	6	1.2×10^{-2}	6.2	7.4×10^{-4}	1.4×10^{-1}
Root crops	Roots	All	46	8.4×10^{-3}	6.2	4.9×10^{-4}	2.6×10^{-1}
		Sand	9	7.8×10^{-3}	5.9	9.9×10^{-4}	2.3×10^{-1}
		Loam	10	2.5×10^{-2}	3.2	2.6×10^{-3}	1.2×10^{-1}

TABLE 6.4. SOIL TO PLANT TRANSFER FACTORS (kg/kg, DRY WEIGHT) FOR URANIUM IN TEMPERATE ENVIRONMENTS (cont.)

Plant group	Plant compartment	Soil group	*N*[a]	Mean/ value[b]	Geometric standard deviation	Minimum	Maximum
		Clay	5	6.8×10^{-3}	6.2	7.9×10^{-4}	9.2×10^{-2}
	Stems, shoots	All	37	2.8×10^{-2}	5.4	2.0×10^{-3}	7.0×10^{-1}
		Sand	9	2.5×10^{-2}	5.6	2.0×10^{-3}	2.4×10^{-1}
		Loam	11	5.0×10^{-2}	3.0	1.3×10^{-2}	3.2×10^{-1}
		Clay	5	1.1×10^{-2}	4.3	2.0×10^{-3}	5.8×10^{-2}
Tubers	Tubers	All	28	5.0×10^{-3}	6.4	1.8×10^{-4}	8.0×10^{-2}
		Sand	4	1.9×10^{-2}	3.8	4.3×10^{-3}	7.8×10^{-2}
		Loam	3	2.8×10^{-2}	3.2	8.2×10^{-3}	8.0×10^{-2}
		Clay	6	9.2×10^{-4}	3.0	1.9×10^{-4}	4.8×10^{-3}
	Stem, shoots	All	1	1.9×10^{-1}	n.a.[c]	n.a.[c]	n.a.[c]

TABLE 6.4. SOIL TO PLANT TRANSFER FACTORS (kg/kg, DRY WEIGHT) FOR URANIUM IN TEMPERATE ENVIRONMENTS (cont.)

Plant group	Plant compartment	Soil group	*N*[a]	Mean/ value[b]	Geometric standard deviation	Minimum	Maximum
Herbs	n.a.[c]	All	9	3.6×10^{-2}	4.9	8.6×10^{-3}	4.1×10^{-1}
Other crops	Leaves (sunflower)	All	39	7.1×10^{-2}	3.9	8.9×10^{-3}	7.8
		Sand	5	4.1×10^{-1}	5.3	1.6×10^{-1}	7.8
		Loam	22	7.1×10^{-2}	2.9	1.0×10^{-2}	6.4×10^{-1}
		Clay	11	2.7×10^{-2}	2.1	8.9×10^{-3}	1.0×10^{-1}
	Grain (sunflower)	All	2	1.5×10^{-2}	n.a.[c]	8.2×10^{-3}	2.9×10^{-2}
Grasses	Stems, shoots	All	147	1.7×10^{-2}	9.4	2.0×10^{-4}	5.5
		Sand	19	1.6×10^{-2}	17.0	5.5×10^{-4}	1.8
		Loam	34	9.8×10^{-3}	8.4	3.1×10^{-4}	4.6×10^{-1}
Pasture	Stems, shoots	All	53	4.6×10^{-2}	5.3	1.3×10^{-3}	1.4×10^{1}

TABLE 6.4. SOIL TO PLANT TRANSFER FACTORS (kg/kg, DRY WEIGHT) FOR URANIUM IN TEMPERATE ENVIRONMENTS (cont.)

Plant group	Plant compartment	Soil group	*N*[a]	Mean/ value[b]	Geometric standard deviation	Minimum	Maximum
Leguminous fodder	Stems, shoots	Sand	3	2.7×10^{-3}	1.8	1.3×10^{-3}	3.9×10^{-3}
		Loam	7	7.2×10^{-2}	33.0	1.8×10^{-3}	1.4×10^{1}
		All	15	1.5×10^{-2}	4.2	2.0×10^{-3}	1.6
		Sand	12	1.0×10^{-2}	2.0	2.0×10^{-3}	2.1×10^{-2}

Source: Adapted from Ref. [6.3].

[a] *N*: Number of data entries used for analysis.

[b] Depending on the data available, the mean value refers to the geometric mean (if $N \geq 3$), the arithmetic mean (if $N = 2$) or a single value.

[c] n.a.: not applicable.

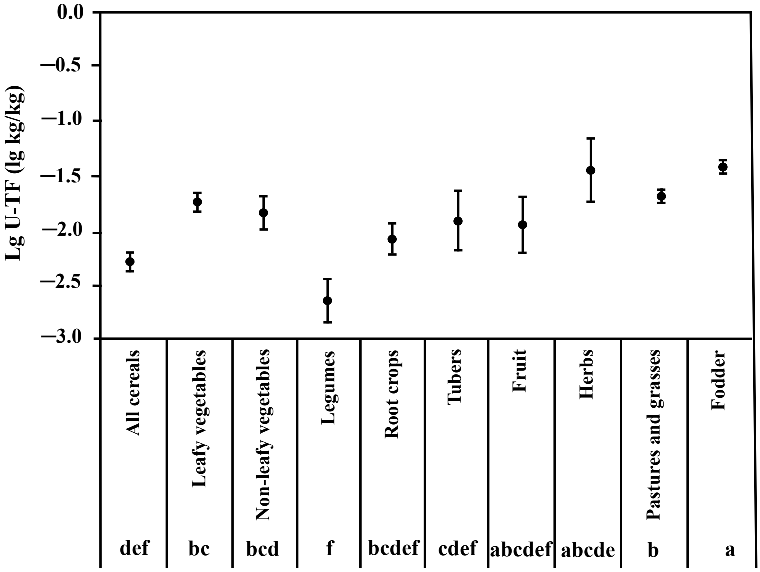

FIG. 6.4. Logarithm of uranium soil to plant transfer factor (TF) values (kg/kg, dry weight) for different crop groups (adapted from Ref. [6.37]). The error bars denote residual standard error after analysis of variance accounting for the effect of plant type. Transfer factors for crop groups assigned the same letter are not significantly different ($p < 0.05$).

was found within crop groups, and it was concluded that transfer factors were not significantly affected by experimental conditions, climate, soil parameters and contamination event.

The impact of soil group, based on texture and organic matter content, on transfer factors was considered in Ref. [6.37] and it was found that uranium transfer factors decline from sandy, through loamy to clay soils (Table 6.5 [6.9]), confirming earlier observations [6.36, 6.66]. There is also a suggestion that uranium transfer is enhanced for plants growing on organic soils (noting that the sample size is relatively small), contradicting earlier views on the effect of soil organic matter [6.36, 6.66, 6.67]. Analysis of soil effects within plant groups showed significant differences between soils for leafy vegetables, tubers and sunflowers only. In the case of tubers, the lowest transfer factor occurs on sandy soil.

The transfer of uranium to plants in tropical and arid regions is considered separately because of the way soils evolve in these environments. Organic materials that reach the soil surface in tropical regions decompose rapidly and the accumulation of organic matter in surface soil is minimal [6.68], which,

TABLE 6.5. TRANSFER FACTORS (kg/kg, DRY WEIGHT) FOR URANIUM BY SOIL GROUP [6.9]

Soil group	*N*[a]	Geometric mean	Geometric standard deviation	Arithmetic mean	Standard deviation	Minimum	Maximum
Sand (b)	105	1.90×10^{-2}	18.0	3.84×10^{-1}	1.47	1.94×10^{-4}	8.82
Loam (ab)	173	2.46×10^{-2}	7.05	2.03×10^{-1}	1.13	5.40×10^{-5}	1.37×10^{1}
Clay (c)	79	5.65×10^{-3}	5.59	1.84×10^{-2}	3.16×10^{-2}	5.68×10^{-5}	2.03×10^{-1}
Organic (a)	14	9.68×10^{-2}	9.23	7.56×10^{-1}	2.10	2.30×10^{-3}	8.02

Note: Transfer factor values for soil groups assigned the same letter are not significantly different.

[a] *N*: Number of data entries used for analysis.

TABLE 6.6. SOIL TO PLANT TRANSFER FACTORS (kg/kg, DRY WEIGHT) FOR URANIUM IN TROPICAL ENVIRONMENTS

Plant group	Plant compartment	Soil group	*N*[a]	Mean value[b]	Geometric standard deviation	Minimum	Maximum
Cereals	Grain	Loam	6	1.9×10^{-3}	1.3	n.a.[d]	n.a.[d]
Maize	Grain	All	4	6.2×10^{-1}	1.2	4.7×10^{-1}	8.0×10^{-1}

TABLE 6.6. SOIL TO PLANT TRANSFER FACTORS (kg/kg, DRY WEIGHT) FOR URANIUM IN TROPICAL ENVIRONMENTS (cont.)

Plant group	Plant compartment	Soil group	*N*[a]	Mean value[b]	Geometric standard deviation	Minimum	Maximum
		Sand	2	7.6×10^{-1}	n.a.[d]	7.2×10^{-1}	8.0×10^{-1}
		Unspecified[c]	2	5.2×10^{-1}	n.a.[d]	4.7×10^{-1}	5.7×10^{-1}
Rice	Grains	Unspecified[c]	15	8.1×10^{-2}	1.7	4.0×10^{-2}	2.0×10^{-1}
	Stems, shoots	Unspecified[c]	10	3.7×10^{-2}	1.7	4.0×10^{-2}	4.5×10^{-2}
Leafy vegetables	Leaves	All	9	3.8×10^{-1}	2.1	2.1×10^{-2}	1.3
		Sand	5	7.1×10^{-1}	1.4	4.0×10^{-1}	1.3
		Unspecified[c]	4	7.8×10^{-2}	3.2	2.1×10^{-2}	5.6×10^{-1}
Non-leafy vegetables	Fruits, heads, berries, buds	Unspecified[c]	33	5.9×10^{-3}	3.4	6.0×10^{-4}	3.1×10^{-2}
Root crops	Roots	All	34	3.1×10^{-1}	2.5	1.9×10^{-2}	2.7
		Loam	8	1.0	1.3	6.1×10^{-1}	1.3

TABLE 6.6. SOIL TO PLANT TRANSFER FACTORS (kg/kg, DRY WEIGHT) FOR URANIUM IN TROPICAL ENVIRONMENTS (cont.)

Plant group	Plant compartment	Soil group	*N*[a]	Mean value[b]	Geometric standard deviation	Minimum	Maximum
		Sand	3	8.1×10^{-1}	1.0	7.8×10^{-1}	8.6×10^{-1}
		Unspecified[c]	23	1.0×10^{-1}	3.7	1.9×10^{-2}	2.7
Tubers	Tubers	All	49	1.0×10^{-1}	2.9	1.6×10^{-3}	1.5
		Loam	8	3.4×10^{-1}	1.3	2.4×10^{-1}	5.9×10^{-1}
		Unspecified[c]	41	7.1×10^{-2}	3.4	1.6×10^{-3}	1.5
Fruits – all	Fruits, heads, berries and buds	All	134	3.7×10^{-3}	5.0	1.2×10^{-5}	3.5×10^{-1}
		Sand	9	1.4×10^{-1}	1.7	7.2×10^{-2}	3.5×10^{-1}
		Unspecified[c]	125	1.0×10^{-3}	3.9	1.2×10^{-5}	5.0×10^{-2}
Fruits – herbaceous plants	Fruits, heads, berries and buds	Unspecified[c]	15	1.2×10^{-3}	3.0	1.2×10^{-5}	1.0×10^{-2}

TABLE 6.6. SOIL TO PLANT TRANSFER FACTORS (kg/kg, DRY WEIGHT) FOR URANIUM IN TROPICAL ENVIRONMENTS (cont.)

Plant group	Plant compartment	Soil group	N[a]	Mean value[b]	Geometric standard deviation	Minimum	Maximum
Fruits – shurbs	Fruits, heads, berries and buds	Unspecified[c]	12	1.0×10^{-3}	3.3	1.4×10^{-4}	1.3×10^{-2}
Fruits – woody trees	Fruits, heads, berries and buds	Unspecified[c]	84	1.1×10^{-3}	3.2	3.7×10^{-5}	2.2×10^{-2}
Fruits – non-woody trees	Fruits, heads, berries and buds	All	23	3.9×10^{-2}	2.9	1.4×10^{-4}	3.5×10^{-1}
	Fruits, heads, berries and buds	Sand	9	1.4×10^{-1}	1.7	7.2×10^{-2}	3.5×10^{-1}
	Fruits, heads, berries and buds	Unspecified[c]	14	2.0×10^{-3}	3.9	1.4×10^{-4}	5.0×10^{-2}
Grasses	Stems and shoots	Unspecified[c]	2	3.1×10^{-1}	n.a.[d]	2.6×10^{-1}	3.5×10^{-1}
Herbs	Leaves	Unspecified[c]	2	2.7×10^{-3}	n.a.[d]	n.a.[d]	n.a.[d]
	Roots	Unspecified[c]	3	2.0×10^{-1}	1.1	n.a.[d]	n.a.[d]

TABLE 6.6. SOIL TO PLANT TRANSFER FACTORS (kg/kg, DRY WEIGHT) FOR URANIUM IN TROPICAL ENVIRONMENTS (cont.)

Plant group	Plant compartment	Soil group	N^a	Mean value[b]	Geometric standard deviation	Minimum	Maximum
	Stems, shoots	Unspecified[c]	2	5.0×10^{-3}	n.a.[d]	n.a.[d]	n.a.[d]
	All plant	Unspecified[c]	3	1.1×10^{-1}	1.2	n.a.[d]	n.a.[d]
Medicinal plants – shurbs	Leaves	Unspecified[c]	5	5.3×10^{-3}	1.5	n.a.[d]	n.a.[d]
Medicinal plants – woody trees	Leaves	Unspecified[c]	10	9.0×10^{-3}	1.4	5.5×10^{-3}	1.4×10^{-2}
Medicinal plants – non-woody trees	Leaves	Unspecified[c]	10	2.8×10^{-3}	1.3	1.4×10^{-3}	4.5×10^{-3}
Other crops	Peanuts	Unspecified[c]	3	3.0×10^{-1}	2.8	1.0×10^{-2}	1.5

Source: Adapted from Ref. [6.35].

[a] N: Number of data entries used for analysis.

[b] Depending on the data available, the mean value refers to the geometric mean (if $N \geq 3$), the arithmetic mean (if $N = 2$) or a single value.

[c] Information for this type of soil (including mineral and organic matter content and soil texture) is not available in the source references.

[d] n.a.: not applicable, because the source values are not available or the number of samples is <3.

coupled with low exchange capacity of the predominant clays, leads to enhanced plant uptake. Data for tropical regions are presented in Table 6.6 [6.35]. The number of soil-to-plant transfer factor values available for tropical environments are significantly enhanced in Ref. [6.35] compared with Ref. [6.3]. Reference [6.35] concluded that for most crop groups grown in tropical environments, transfer factors have higher values than those grown in both temperate and arid environments, however, more relevant conclusions can be drown only for significant sample sizes with known soil characteristics.

Rice, *Oryza sativa* L., is a staple food type consumed in humid tropical regions that is grown under flooded conditions (5–10 cm layer of water). Following the approach used in Ref. [6.35] for tropical regions, soil-to-rice transfer factor data are presented separately in Table 6.7.

A study in Thailand [6.69] provides additional data in a tropical region for rice and other local fruits and vegetables. Soil to plant transfer factors of ^{238}U decreased in the order vegetables to rice to fruit, and values ranged from $<4.7 \times 10^{-4}$ to 6.8×10^{-2}, with a mean of 1.6×10^{-2}. The fruits studied included mango and pineapple. Soil to rice transfer factors are also reported in Ref. [6.70] for two sites in Malaysia, with values ranging from 0.04 to 0.2.

Recent data are available for a subtropical region [6.71] in the West Bank, Palestine. This study considered transfer factors for grass, reporting a mean of 1.12, with a range of 0.36–1.62. While these values appear high, they are within the range observed for temperate systems and represent one of the few subtropical datasets available. Reference [6.50] also provides concentration ratios for a subtropical region of Croatia, with data for various grasses, herbs, shrubs and fruits from trees. This dataset has a GM of 9.6×10^{-3} and a GSD of 2.5 (range: 2.8×10^{-3}–5.4×10^{-2}).

Soil to plant transfer data in arid regions are presented in Table 6.8. Only a limited amount of data is available for arid regions in Ref. [6.35] owing to the scarcity of published data for arid environments compared to those for temperate or tropical regions. The types of soil and vegetation typically found in arid areas and the soil to plant transfer pattern are defined by the specific climatic characteristics, such as scarce precipitation, low humidity and excessive heat.

Recent data for uranium in forest systems are available. Reference [6.72] contains data for four boreal forest species with GMs ranging from 3×10^{-3} (GSD = 3.54) for Rowan to 8×10^{-3} (GSD = 2.71) for May lily (false lily of the valley) in Finland. Transfer to moss, lichen, spruce and pine was investigated in Norway [6.73], with ^{238}U transfer factors of 6.25×10^{-2}, 2.59×10^{-2}, 4.1×10^{-4} and 1.9×10^{-3}, respectively. The uranium uptake by wild berries and grasses in natural ecosystems in Belarus is reported in Ref. [6.74], where forest, marsh and meadow locations were considered (Table 6.9). The highest transfer factors (DW basis) for mixed meadow grasses were observed with moor peats and sandy soils,

TABLE 6.7. SOIL TO RICE TRANSFER FACTORS (kg/kg, DRY WEIGHT) FOR URANIUM

Plant group	Plant compartment	Soil group	*N*[a]	Geometric mean	Geometric standard deviation	Minimum	Maximum
Rice	Grains	Unspecified[b]	15	8.1×10^{-2}	1.7	4.0×10^{-2}	2.0×10^{-1}
	Stems, shoots	Unspecified[b]	10	3.7×10^{-2}	1.7	4.0×10^{-2}	4.5×10^{-2}

Source: Adapted from Ref. [6.35].

[a] *N*: Number of data entries used for the analysis.

[b] Information for this type of soil (including mineral and organic matter content, soil texture) is not available in the source references.

TABLE 6.8. SOIL TO PLANT TRANSFER FACTORS (kg/kg, DRY WEIGHT) FOR URANIUM IN ARID ENVIRONMENTS

Plant group	Plant compartment	Soil group	*N*[a]	Geometric mean	Geometric standard deviation	Minimum	Maximum
Cereals	Stems, shoots	Clay	4	1.1×10^{-2}	1.3	8.0×10^{-3}	1.5×10^{-2}
Fruits – woody trees	Fruits, heads, berries and buds	Clay	6	1.3×10^{-2}	3.4	4.0	1.7×10^{-2}
	Leaves	Clay	3	3.4×10^{-2}	1.0	1.8×10^{-2}	4.8×10^{-2}

TABLE 6.8. SOIL TO PLANT TRANSFER FACTORS (kg/kg, DRY WEIGHT) FOR URANIUM IN ARID ENVIRONMENTS (cont.)

Plant group	Plant compartment	Soil group	*N*[a]	Geometric mean	Geometric standard deviation	Minimum	Maximum
Grasses	Stems, shoots	Clay	10	n.a.[c]	n.a.[b]	6.2×10^{-1}	2.6
	Stems, shoots	Sand	10	5.0×10^{-3}	12	1.9×10^{-4}	2.4×10^{-1}
Leafy vegetables	Leaves	Clay	9	6.3×10^{-3}	n.a.[b]	4.0×10^{-3}	1.1×10^{-1}
Leguminous vegetables	Seeds, pods	Clay	5	1.6×10^{-2}	3.9	3.0×10^{-3}	6.4×10^{-2}
Non-leafy vegetables	Fruits, heads, berries and buds	Clay	7	1.9×10^{-2}	3.0	4.0×10^{-3}	5.7×10^{-2}

Source: Adapted from Ref. [6.35].

[a] *N*: Number of data entries used for analysis.

[b] n.a.: not applicable, because the source values are not available or the number of samples is <3.

TABLE 6.9. FOREST SOIL TO PLANT TRANSFER FACTORS (kg/kg, DRY WEIGHT) FOR URANIUM [6.74]

Plant group	Plant compartment	Isotope	Geometric mean	Geometric standard deviation	Minimum	Maximum
Meadow grasses	Unspecified	^{234}U	0.13	2.4	0.036	0.42
		^{238}U	0.15	2.4	1.93×10^{-3}	1.26×10^{-2}
Wild berries	Bilberry, cranberry, blueberry	^{234}U	0.003 4	1.8	0.001 8	0.008
		^{238}U	0.003 5	1.8	0.001 8	0.008

with the lowest values found for loamy and peat-gley soils. For berries, values for sites with sandy soils exceeded those for sites with loamy soils by factors of three to four.

Transfer to plants in a rain forest of southern India was studied and concentration ratios (DW) for the leaves of woody plants ranging from 0.25 to 0.35 were recorded [6.75]. Transfers within a marsh habitat contaminated by mill tailings was investigated in Ref. [6.76], with uptake by Marsh marigold, *Caltha palustris* L., soft rush, *Juncus effuses* L., and Tall Moor grass, *Molinia arundinacea* L., being studied. Transfer factors were greatest for soft rush (0.02) and lower for the other plant species (both 0.008).

6.2.3.3. Uptake and distribution in plants

Studies on the distribution of uranium in plants show that transport within plants is limited, with a higher uranium content in the roots than in other plant parts. A study of watermelon and zucchini fruits [6.77] found that roots, leaves and stems accumulate uranium to a greater degree than other plant parts by a factor of a hundred. A similar pattern was found using tomato plants [6.78]. Reference [6.54] reports higher transfer and accumulation in tomato plant roots with lower accumulation in stems, but substantially greater than that in leaves. The root content was substantially greater than that in the leaves of rye [6.79], wheat [6.79, 6.80], tobacco [6.81] and other plant types, including pea, brassicas, maize, grass and clover [6.48]. It was also noted [6.80] that the lowest concentration was found in grain. In a study of Mediterranean trees [6.82], the uranium transfer to fruit was always less than that to leaves. The combined data summarized in Tables 6.4–6.9 do not clearly indicate a distinction between plant parts, owing also to the scarce available data for a range of plant compartments.

Contradictory evidence is available on whether the accumulation of uranium increases with the age of the plant or plant part. In Ref. [6.83], it was reported that older plant organs show the greatest accumulation (annual crops), while in Ref. [6.81], it was found that storage was mainly in the apices and young tobacco leaves. In Ref. [6.36], it is reported that after an initial increase in tree twigs over two years, subsequent years showed a decline.

Reference [6.84] highlighted the potential for soil containing uranium to adhere to plant surfaces and influence transfer factors. The impact of soil particles remaining on subterranean surfaces is recognized and steps are usually taken to reduce this by washing or peeling roots or root storage organs. It was suggested [6.67] that soil load could contribute more than root uptake to the uranium associated with both root and leafy crops, leading to the conclusion that soil load should be given careful attention in the derivation of transfer factors [6.36]. For example, tests were undertaken [6.51] for soil associated with plant samples through analysis of scandium and titanium to show that soil had been removed. Reference [6.85] showed that peeled potato had a lower uranium concentration ratio than whole potato (the transfer factor of peeled potato was 0.24–0.74 of that for whole potato), and the outer leaves of cabbage had higher concentration ratios than the inner leaves (the transfer factor of inner leaves was 0.48 of that for outer leaves).

6.3. URANIUM TRANSFER TO ANIMALS

6.3.1. Behaviour of uranium in terrestrial animals

For many purposes, it is convenient to express the behaviour of uranium in terrestrial animals in terms of equilibrium transfer coefficients (F_m) or, alternatively, concentration ratios. A transfer coefficient is the ratio of the concentration of an element or radionuclide in a fluid, tissue or organ, or in the whole body, usually expressed on an FW basis, to the rate of intake of that element or radionuclide in the diet and drinking water. It is usually given in units of days per kilogram or days per litre for milk. A concentration ratio (dimensionless) for a terrestrial animal is the ratio of the concentration of an element or radionuclide in a tissue or organ, or in the whole body, usually expressed on an FW basis, to the concentration of that element or radionuclide in the feed, usually expressed on a DW basis.

However, in some contexts (e.g. in relation to short term releases), the kinetics of uptake and retention may be of importance. In these circumstances, it is appropriate to apply a biokinetic model for uptake and retention in terrestrial animals, as is done for humans (see Chapter 7).

In the following, a summary is first provided of observed transfer coefficient and concentration ratio values for uranium in terrestrial animals. Then, the biokinetics of uranium in such animals is described. Finally, an analysis is provided to demonstrate that the observed transfer coefficient and concentration ratio values are consistent with what is known of the biokinetics of uranium.

6.3.2. Transfer coefficients and concentration ratios

The IAEA [6.3] has summarized transfer coefficientss for uranium applicable to human food products. For cow’s milk, three values were considered. These had a mean of 1.8×10^{-3} d/L, with minimum and maximum values of 5.0×10^{-4} and 6.1×10^{-3} d/L, respectively. For goat’s milk, only a single value of 1.4×10^{-3} d/L was cited. For beef, three values were again considered; these had a mean of 3.9×10^{-4} d/kg, with minimum and maximum values of 2.5×10^{-4}

and 6.3 × 10^{-4} d/kg, respectively. For pork, two values were given (2.6 × 10^{-2} and 6.2 × 10^{-2} d/kg). For poultry meat, two values were given (0.3 and 1.2 d/kg); two values were also given for egg contents (0.92 and 1.2 d/kg).

Transfer coefficients for uranium are reviewed in Ref. [6.86]. For cow's milk, it was identified that Ref. [6.87] gave a single estimate of 7.30 × 10^{-5} d/L based on unpublished data obtained at a Ploughshare cratering event. Based on ^{238}U concentrations in milk and associated forage, a value of 6.10 × 10^{-4} d/L was estimated (see also Ref. [6.88]).

Transfers to sheep milk and cheese were studied at the Boco mine, Portugal [6.89], which is an unremediated uranium mining legacy site. Uranium concentrations in grass, expressed on a FW basis, were about one order of magnitude higher than concentrations in milk, but concentrations in cheese were similar to, or slightly higher than, those in grass. This reflects the consideration that milk comprises about 10% milk solids. The concentration in milk relative to diet may be interpreted in terms of a transfer coefficient by noting that sheep consume about 7 kg/d of fresh forage. Thus, the transfer coefficient is about 1.4 × 10^{-2} d/L. In contrast, in samples of pasture, sheep milk and cheese from the geographical area of the old uranium mine and milling facilities of Urgeiriça, Portugal, the concentration of uranium in milk was about half that in pasture, but the concentration in cheese was about one order of magnitude lower than that in pasture [6.90]. These differences may simply reflect the limited extent of the data available.

For meat, Ref. [6.91] gave values of 6.2 × 10^{-2} d/kg for pork and 1.2 d/kg for the flesh of Russian White hens. For eggs of Russian White hens, values of 0.99 d/kg for contents and 1.3 d/kg for whole eggs were given.. Examination of Ref. [6.88] showed that the pigs used were killed at two months of age and that, although the paper did not state the period of exposure explicitly, the nature of the experiment suggested that the animals were fed contaminated feed from birth [6.86]. In Ref. [6.86], it was considered that, in view of the known high permeability of the gastrointestinal tract to actinides in early life and the possibility that the uranium burden of these very young animals may have been determined, in part, by uptake in utero or from their mothers' milk, the recorded transfer coefficient for pork should be regarded with a degree of caution. For hens, Ref. [6.89] gives results for skeleton and muscle, and the shell, yolk and white of eggs from fully mature birds. More recently, 7–70 day old broilers were used to demonstrate that the administration of phytase to growing fowls improves the availability of uranium and that the rate of uranium accumulation in bone was slower than the rate in increase in bone mass, leading to a growth dilution effect [6.92]. Reference [6.4] provides concentration ratio values for wildlife groups in terrestrial ecosystems (see Section 7.5).

6.3.3. Biokinetics

The biokinetic characteristics of uranium have been studied largely in mammals. A comprehensive account of the early animal experiments is given in Ref. [6.93] and of human exposures in Ref. [6.94]. More recent studies are discussed in Refs [6.95–6.97]. To a large degree, these data have been considered in the biokinetic model for uranium in humans described in Chapter 7. A similar model can be used for other mammals, as required.

A key factor governing the relationship between the intake rate of uranium by ingestion and the content in mammalian tissues and organs is the fractional gastrointestinal absorption, f_1. Based on a review of the older literature, the ICRP [6.98] recommended reference values of f_1 of 0.05 for water soluble inorganic compounds (hexavalent uranium) and 0.002 for relatively insoluble compounds, such as UF_4, UO_2 and U_3O_8, in which the uranium is usually tetravalent. As discussed in Section 7.3.2, overall, the f_1 value can range from <0.1% to 6% depending on the solubility of the uranium compound [6.97], but is typically about 0.02 for dietary uranium [6.99–6.101]. For adult ruminants, the IAEA [6.3] reported two measured values of f_1 of 0.01 and 0.012, but no new data were reported for monogastric species.

Uranium taken up from the gastrointestinal tract enters the blood. Data on laboratory animals indicate that a substantial fraction of uranium leaving the blood initially distributes throughout soft tissues, but that a few days after absorption or intravenous injection, most of the systemic content (typically about 90%) is found in the kidneys and skeleton [6.86].

In long term chronic exposures, distinctions between general soft tissue, liver and kidney concentrations are limited. In broad terms, when uranium enters the blood, 100% is initially associated with fluids in soft tissues. However, by 24 h after entry, about 50% of the uranium has been lost by urinary excretion and about 12–25% of the initial activity is associated with the kidneys. Some deposition to mineral bone will also have occurred by this stage. The content of soft tissues other than the kidneys at 24 h will be around 30% of the uranium that initially entered the blood. At 2.5 d, the other soft tissue content has dropped to around 6% of the initial activity, dropping to 4% at 18 d and 0.3% at 566 d [6.86]. Based on these values, it was estimated [6.86] that retention of uranium in soft tissues other than the kidneys after its entry into the blood is well represented by:

$$R(t) = 0.937e^{-1.4t} + 0.06e^{-0.03t} + 0.003 \tag{6.12}$$

The time integral of this function is shown in Fig. 6.5.

The transfer coefficient for soft tissues is given by the following equation:

$$F_{\mathrm{m}} = \frac{f_1 \times \int R(t)\,\mathrm{d}t}{M} \tag{6.13}$$

where the time integral is over the period of exposure and M (kg) is the mass of soft tissues other than kidneys.

Taking the f_1 value to be in the range of 0.005 to 0.05, as is thought to be appropriate for dietary uranium, and $\int R(t)\,\mathrm{d}t$ to be in the range of 3–6 d, as is suitable for long term exposure:

$$F_{\mathrm{m}} = \frac{(0.015 \text{ to } 0.3)}{M}\,(\mathrm{d/kg}) \tag{6.14}$$

The transfer coefficient varies inversely with the mass of other soft tissue. For cattle, this would typically be 200–1000 kg [6.102], implying that transfer coefficients ranging from 1.5×10^{-5} to 1×10^{-3} d/kg would be expected. This range is broadly consistent with observed values. For mature pigs, the soft tissue mass at maturity would typically be around 100 kg [6.102], giving transfer coefficients in the range 1.5×10^{-4}–3×10^{-3} d/kg. The observed values are about one order of magnitude higher than the top end of this range, but these are for two month old animals. Although $\int R(t)\,\mathrm{d}t$ might be only about 2.5 d for such animals, the f_1 value would probably be at the top end of the observed range, and the other soft tissue mass would be considerably less than in a mature animal. A reasonable estimate for pigs at two months of age is about 10 kg [6.102], giving a transfer coefficient of about 3×10^{-2} d/kg, as observed. For Russian

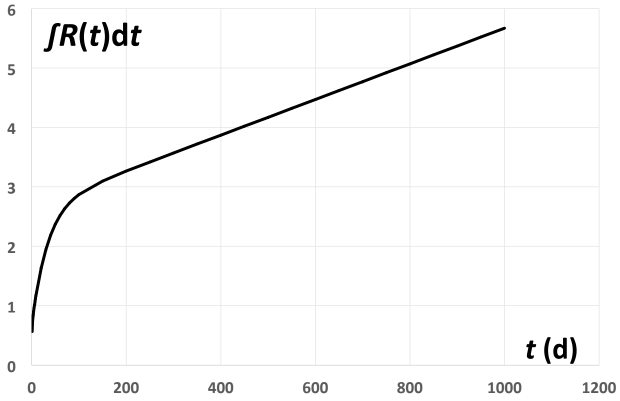

FIG. 6.5. Time integral of the soft tissue retention function.

White hens, the mature body mass is about 2.0–2.1 kg, so the total soft tissue mass will be somewhat less than 2 kg, giving transfer coefficients in the range of 0.01–0.2 d/kg. Observed values are somewhat larger than this, at about 1.0 d/kg. This suggests that gastrointestinal absorption may be significantly enhanced in poultry relative to mammals or that a larger proportion of the uranium absorbed into the blood may exhibit long term retention in soft tissues. However, there are no data available to determine which, if either, of these hypotheses is valid.

6.3.4. Biomarkers

There has been some interest in the use of earthworms as biomarkers for uranium. In this context, the interest is both in the concentrations of uranium that can accumulate in the organisms and the adverse effects that may be induced. The most detailed study of this topic [6.103] studied the accumulation of both natural and depleted uranium in the earthworm, *Eisenia fetida*, using natural and amended soils with uranium concentrations ranging from 1.86 to 600 mg [U]/kg. No effects were observed in terms of mortality or weight reduction, but cytotoxic and genetic effects were observed at quite low concentrations. Uranium concentrations in the earthworms were proportional to the uranium concentrations in the soil to which they were exposed. Concentration ratio values after 7 d of exposure were 0.11 ± 0.06 for depleted uranium and 0.074 ± 0.033 for natural uranium. After 28 d of exposure, the corresponding values were 0.023 ± 0.010 and 0.021 ± 0.009 for depleted and natural uranium, respectively. As the decrease in concentration ratio values between 7 and 28 d was most noticeable at high uranium concentrations in soil (see fig. 2 of Ref. [6.103]), this suggests adaptation of the earthworms to high uranium concentrations in soil by a process of enhanced bioexclusion. This is consistent with the observation that bioaccumulation is greater at lower uranium concentrations in soil [6.104]. Juvenile earthworms were found to accumulate more uranium than adults [6.104]. More recently, a maximum bioaccumulation factor of 0.35 for earthworms in 28 d spiked soil studies has been reported [6.105] and distinctions in uptake between various chemical forms of uranium have been shown [6.106].

In another study [6.107], exposures to contaminated soils containing uranium and radioisotopes of other elements and various metals gave rise to multiple effects, including inhibition of reproduction, growth reduction, DNA damage and cytotoxicity. Histological changes in the body wall and gastrointestinal tract were observed and it has been suggested that such histopathological changes could be a useful biomarker of soil quality [6.108].

Isotopic ratios in earthworms and porewater were studied [6.109] at a United Kingdom weapons' testing range where depleted uranium had been utilized and showed that depleted uranium released into the environment during weapons test firing operations was more labile and bioavailable than naturally occurring uranium in soils at the location studied. This built upon earlier work that had shown that isotopic ratios in earthworms could be useful for detecting localized contamination with depleted uranium [6.110]. Studies of soil dwelling insects in a uranium mining area in south-east Siberia, Russian Federation demonstrated only slight increases in uranium concentrations over the background even at highly contaminated sites (compare tables 1 and 2 of Ref. [6.111]). There is some evidence that terrestrial arthropods may accumulate uranium at contaminated sites [6.112], but the data are insufficient to permit quantitative analysis.

6.4. FRESH WATER

6.4.1. Water–sediment interactions

Freshwater sediments often serve as a sink for uranium, resulting in uranium concentrations that can be several orders of magnitude higher in sediments and suspended solids than in the surrounding water [6.113, 6.114]. ‘Suspended sediments’ are fine texture particles (mainly characterized by a particle size between colloid and silt) suspended in the water column and transported with the water flow. They constitute an important element of aquatic food chains by regulating the exchange between the water column and bed sediments through erosion and deposition processes [6.115, 6.116]. Deposited sediments are characterized by a major coarse fraction and, generally, lower organic matter contents in comparison to suspended sediments. They can accumulate and store metals, including uranium, over the long term, and slowly release them back to the environment when appropriate conditions are met, playing a role of temporary or permanent reservoirs in metal cycles [6.117].

Uranium partitioning between its dissolved and particulate forms, and its corresponding mobility, can be driven by a number of factors, such as pH, mass:volume ratio, alkalinity (bicarbonate), redox potential, dissolved organic matter in the overlying water, phosphorus in surface waters, the properties of complexing agents or ligands, sediment particle size, mineral composition, and the nature and characteristics of sorbing materials (e.g. sediments, suspended particulates) in a given surface water [6.97, 6.113, 6.118–6.124]. This partitioning occurs through complex physical, chemical and biological processes, including sorption, adsorption, desorption, complexation, surface precipitation and co-precipitation [6.125–6.127], but is also controlled by environmental conditions and elemental speciation [6.128, 6.129]. The solid–liquid distribution coefficient, K_d, is determined to assess the partitioning of elements in the environment under the assumptions of equilibrium, reversibility and trace conditions (see also Section 6.2.2).

Uranium can become mobile in freshwater systems following the formation of complexes between uranium and inorganic or organic ligands, where the stability of such complexes is pH dependent [6.97, 6.121]. In oxygenated alkaline systems, uranium tends to form a soluble complex with carbonate, whereas in acidic waters ($pH < 6$) containing low concentrations of inorganic ions and high concentrations of dissolved organic matter, soluble organic complexes tend to be formed [6.113, 6.121].

Redox potential plays an important role in controlling uranium mobility in freshwater systems [6.97]. Under anoxic conditions, U(VI) is reduced to U(IV), which can result in the conversion of soluble compounds into insoluble ones. Owing to its insolubility, U(IV) is then deposited into the sediments in the form of U(IV) salts [6.118, 6.121, 6.130, 6.131]. Transfer of uranium from water to sediment is dependent on sediment type, where transfer affinities decrease in the order: organic sediment, clay, sand [6.132].

Uranium can also be removed from solution through physical adsorption processes, for example, following sorption onto iron or manganese oxides that occur as coatings on the sediment particles, or on naturally occurring organic matter, to form stable complexes, which can serve as uranium sinks [6.19, 6.20, 6.133–6.136]. In surface waters of low ionic strength and low concentrations of U(VI), concentrations of dissolved uranyl ions tend to be controlled by cation exchange and adsorption processes [6.13, 6.137]. Under such conditions, uranyl ions and their complexes can adsorb to surfaces of clays, organics and oxides [6.19, 6.20, 6.133, 6.138–6.141]. As the ionic strength increases within an oxidized solution, ions such as Ca^{2+}, Mg^{2+} and K^{+} can displace uranyl ions from exchange sites on particulate surfaces, resulting in partitioning into solution. Once released from surfaces, uranyl ions can then form strong, soluble complexes with carbonate ions, resulting in further increases of uranium in solution [6.142]. As a result, uranyl ions become particularly mobile in high ionic strength solutions. Similarly, in the presence of organic complexants, sorption to oxide minerals, clays and other particulates can be substantially reduced or greatly inhibited [6.19, 6.137].

The pH of surface water can significantly affect U(VI) sorption to particulates owing to the influence of pH on uranium speciation and the decrease in the number of exchange sites on variably charged particles, such as iron oxides, aluminium oxides and natural organic matter, at low pH [6.13]. At pH values between 6.5 and 10, uranium species with a lower tendency to adsorb tend to be present.

Therefore, based on the available literature, many factors can influence uranium concentrations in suspended and deposited sediments relative to those in fresh waters. Such site specific factors contribute to the large variability of K_d coefficients in freshwater systems, typically in a range between 10^2 to 10^6 L/kg. High K_d values suggest the formation of stable complexes with the particulate phase, whereas elements with low K_d values tend to remain in the dissolved form, thus being characterized by high mobility [6.143, 6.144].

A freshwater K_d database, initially compiled in Ref. [6.3], was updated under Working Group 4 of the IAEA programme MODARIA (Modelling and Data for Radiological Impact Assessments; 2012–2015)[9] and new K_d values for uranium were included. These new K_d values were grouped into three solid–liquid exchange conditions (adsorption, desorption and field) for two environmental components (suspended and deposited sediments) and log-normal distributions were derived for each radionuclide–component–condition association when a minimum of ten K_d values were available [6.145].

Table 6.10 provides the GM, GSD, maximum and minimum values, the 5th and 95th percentiles, the size of the dataset and the number of references for uranium, according to the updated dataset, relevant for anthropogenic releases (suspended sediments) and anthropogenic releases with a risk of overestimation (deposited sediments) [6.145]. As the number of K_d values for uranium was greater than ten, it was assumed that the dataset followed a log-normal distribution.

Within the updated freshwater database, the K_d values for suspended sediments are significantly higher than for deposited sediments. Furthermore, the variability of K_d distributions is larger for deposited sediments than for suspended sediments.

Conditional statistical K_d distributions for in situ suspended matter as a function of suspended load, dissolved organic carbon and pH reduce the global variability of K_d values by several orders of magnitude, thus supporting the reduction of uncertainties in transfer models involving the K_d parameter (see Ref. [6.115] for more details). Within the factors listed above, the suspended load was found to have a major influence (>50%) on the variability of K_d datasets of uranium for suspended sediments in field conditions [6.145].

6.4.2. Transfer to freshwater biota

As defined in Ref. [6.120], "bioaccumulation is the process of a chemical moving from the external environment into the organism from all possible exposure routes (water, sediment, soil, air, or diet) and is expressed as a bioaccumulation factor (BAF)" [6.4, 6.146, 6.147]. Numerous factors can influence the bioaccumulation of uranium in freshwater systems [6.122]. These

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[9] MODARIA programme (Modelling and Data for Radiological Impact Assessments), https://www-ns.iaea.org/projects/modaria/default.asp?s=8&l=81

TABLE 6.10 URANIUM K_d DISTRIBUTIONS AS A FUNCTION OF ENVIRONMENTAL COMPONENTS (SUSPENDED AND DEPOSITED SEDIMENTS)

Component	*N*[a]	Geometric mean (L/kg)	Geometric standard deviation	Minimum (L/kg)	Maximum (L/kg)	5th percentile (L/kg)	95th percentile (L/kg)	No. of references
Deposited sediment	14	3.50×10^3	40.10	9.10×10^1	8.04×10^4	8.07	1.51×10^6	5
Suspended sediment	38	1.19×10^4	5.63	3.05×10^2	1.27×10^5	6.94×10^2	2.04×10^5	6

Note: Geometric mean and geometric standard deviation values were obtained by fitting the empirical cumulative distribution function with the log-normal cumulative distribution function. The representativeness of this adjustment was assessed using the Kolmogorov–Smirnov statistical test.

[a] *N*: Number of data entries used for the analysis.

include the water chemistry, physicochemical form of uranium, seasonal changes in uranium speciation and concentration in surface waters, water temperature, redox potential, organism size and age, and physicochemical attributes of the ecosystem (e.g. total suspended solids, dissolved solids, geology, pH, hardness), as described in Section 6.4.1.

Uranium uptake is proportional to the concentration of bioavailable free uranyl ions present in the water; however, uptake is also influenced by pH through both competitive inhibition (where protons compete with uranyl ions for binding sites where uptake occurs) and non-competitive inhibition (where protons decrease the uptake rate of uranyl ions) [6.120].

Uranium uptake may also be influenced by the concentrations of major cations calcium and magnesium in the water. Specifically, uptake is reduced in the presence of calcium and magnesium, owing to competition with uranium for uptake sites. In addition, at high pH, there is some evidence for the formation of carbonato-complexes, resulting in uranium complexation by carbonates and reduced bioavailability [6.148]. Similarly, dissolved organic matter can bind uranyl ions, thereby reducing uranium bioavailability in freshwater systems.

The concentration of phosphorus in surface waters can also affect uranium speciation and its corresponding bioavailability [6.120]. At concentrations of the order of 50 μg [P]/L (which can occur in eutrophic lakes), uranium will precipitate as $UO_2HPO_4(s)$ [6.120, 6.149].

Therefore, such factors can affect uranium partitioning between different environmental compartments, including water, sediments and dietary items, with which aquatic biota interact and from which they can accumulate uranium [6.150, 6.151]. Uranium can also be accumulated by aquatic plants through direct deposition or resuspension, or it can adhere to the outer membrane of a plant’s root system.

Depending on the environmental conditions, uranium may be taken up through direct contact with the aqueous phase via the gills and/or through ingestion of contaminated dietary items or particulates via the gastrointestinal tract [6.152, 6.153]. Uptake via the gills can occur through passive diffusion of dissolved uranium from the aqueous phase into the organism [6.120, 6.154]. For invertebrates, uranium is transported across the gill membrane into the haemolymph via intracellular high affinity protein carriers that are present in the gills [6.153, 6.155, 6.156]. Available literature suggests that uranium is primarily taken up from the water via the gills (e.g. Ref. [6.120]).

Freshwater biota may also accumulate uranium via ingestion of food and/or ingested sediment particles [6.157], although the assimilation efficiency within the gastrointestinal tract is thought to be low [6.120]. For example, trophic transfer efficiencies of approximately 1–13% have been reported for crayfish feeding on bivalves [6.158]. In addition, comparison of uranium concentrations in food relative to those in fish tissue has shown that concentrations were higher in the food passing through the gastrointestinal tract [6.159–6.161].

Once uranium has been taken up in the body, it may be transported, distributed and/or sequestered within the organism, which may lead to elimination from the animal [6.155]. For example, to avoid potentially toxic effects of uranium, an organism may be able to excrete it or store it in a soluble or insoluble form that is not able to cause adverse effects on the organism [6.152, 6.153, 6.162]. The uranium remaining in the organism following elimination will then partition between different tissue types, which have varying kinetics. The balance between uptake and excretion of uranium and the kinetics of these processes will determine uranium bioaccumulation within an organism. Factors such as organism growth and body size can also contribute to variability in total uranium bioaccumulation estimates owing to their influence on uranium uptake kinetics, bioaccumulation and elimination from the organism [6.152, 6.157].

In general, the available literature suggests that there are two pools in which uranium can be present in the body, one with relatively rapid kinetics (thereby allowing uranium to be shed quickly from the body) and a second with relatively slow kinetics (which tends to maintain the uranium load in the body) [6.120]. Based on available information, liver and gill tissues are expected to show rapid kinetics, with the potential to depurate uranium if concentrations in surrounding waters decline, whereas brain and muscle are expected to show slow kinetics, with no significant depuration [6.163]. No kinetic data are available for kidney.

For some types of aquatic organism, such as those with an exoskeleton (e.g. invertebrates), uranium may adsorb to the outer surface of the body, contributing to the concentration measured in the whole organism [6.155, 6.164].

The size of a given compartment within the body and the concentration of uranium in each compartment can be evaluated to quantify the load of uranium in different compartments, which can be monitored under changing conditions to determine the rate of kinetics for different compartments in the body. Reference [6.2] provides information on the mean biomasses of different tissues relative to the whole body mass for fish and amphibians. Table 6.11 provides a summary of mean to reference tissue concentration ratios, which allow the uranium load in the whole body to be estimated, even if data on only a specific tissue type are available [6.3, 6.165–6.168]. Using a mass balance approach, accounting for the concentration of uranium in each tissue relative to a reference tissue, it is possible to estimate concentrations in different compartments within an organism.

Although many factors can influence uranium dynamics within organisms and in the surrounding environment, for the purposes of screening during the assessment process, in many cases, it may be pragmatic to estimate concentrations of uranium in biota using characterization or monitoring data for water (or, in some cases, sediments), along with surface water to biota (and, in some cases, sediment to biota) concentration ratios (Tables 6.12 and 6.13 [6.2–6.4, 6.169–6.186]). In cases where there is reason to believe that uranium may be contributing to significant adverse effects, additional work may be required to make site specific measurements of uranium in biota, taking account of the site specific conditions with respect to the factors potentially influencing uranium bioaccumulation.

TABLE 6.11. SUMMARY OF MEAN TO REFERENCE TISSUE CONCENTRATION RATIOS FOR FRESHWATER BIOTA [6.3]

Type of organism	Reference tissue[a]	Tissue(i)	N[b]	Arithmetic mean[c] $C_{\text{tissue}(i)}$:$C_{\text{reference tissue}}$	Standard deviation	Range
Amphibians	Muscle or carcass	Bone	2	33	38	6.3–60
		Liver	4	21	20	5.0–50
		Muscle (Ref.)	*4*	*1*	*n.a.*[d]	*n.a.*[d]
Freshwater fish	Muscle	Bone	12	56	72	2–210
		Liver	5	2.8	1.1	4
		Muscle (Ref.)	*12*	*1*	*n.a.*[d]	*n.a.*[d]
		Skin	7	8.1	9.8	30
		Skin and muscle	3	3.4	0.38	3.8
		Whole body	7	14	22	61

Note: Reference tissues (Ref.) are indicated in italics.

[a] The reference tissue chosen depends on the data available in the source publication.

[b] *N*: Number of data entries used for analysis.

[c] Represents the ratio of the uranium concentration in a given tissue *i* relative to the concentration in the reference tissue for the type of organism under consideration [6.165–6.168].

[d] n.a.: not applicable.

TABLE 6.12. WATER TO BIOTA CONCENTRATION RATIOS FOR FRESHWATER BIOTA [6.2–6.4]

Type of biota	Tissue type	*N*[a]	Mean value[b] (L/kg, fresh weight)	Range	Reference
Algae	Whole	1	160	n.a.[d]	[6.2]
Algae	Whole	—[c]	1 576	—[c]	[6.169]
Phytoplankton	Whole	40	59	40–180	[6.4, 6.170]
Vascular plants	Shoots	3	230	81–520	[6.2, 6.3]
Vascular plants	Whole	386	130	29–2 700	[6.4, 6.171, 6.172]
Bacteria	Whole	—[c]	—[c]	2 794–354 000	[6.173]
Plankton	Whole	—[c]	459	—[c]	[6.169]
Freshwater invertebrates	Whole body	9	170	3.6–60 000	[6.2, 6.3]
Crustaceans	Whole body	5	110	—[c]	[6.4, 6.174]
Bivalve molluscs	Whole body	3	540	—[c]	[6.4, 6.171]
Fish (generic)	Whole body	2	2.4	1.5–3.3	[6.2, 6.3]
Fish (generic)	Whole body	1 294	9.1	0.05–760	[6.4, 6.171, 6.172, 6.174–6.185]

TABLE 6.12. WATER TO BIOTA CONCENTRATION RATIOS FOR FRESHWATER BIOTA [6.2–6.4] (cont.)

Type of biota	Tissue type	N[a]	Mean value[b] (L/kg, fresh weight)	Range	Reference
Benthic fish	Whole body	99	26	0.6–760	[6.4, 6.174, 6.177–6.185]
Piscivorous fish	Whole body	84	11	0.51–170	[6.4, 6.174, 6.176, 6.179–6.185]
Fish (generic)	Muscle	12	0.96	0.02–20	[6.2, 6.3]
Reptiles	Whole body	8	90	45–190	[6.4, 6.186]

[a] N: Number of data entries used for analysis.

[b] Depending on the data available, the mean value refers to the geometric mean (if $N \geq 3$), the arithmetic mean (if $N = 2$) or a single value.

[c] —: data not available.

[d] n.a.: not applicable.

TABLE 6.13. SEDIMENT TO BIOTA CONCENTRATION RATIOS FOR FRESHWATER BIOTA [6.2, 6.3]

Type of biota	Tissue type	N[a]	Geometric mean value (L/kg, fresh weight)	Range	Reference
Freshwater invertebrates	Whole body	6	0.017	0.002 9–0.064	[6.2, 6.3]

[a] N: Number of data entries used for analysis.

6.5. MARINE ENVIRONMENT

6.5.1. Transport through estuaries and the coastal environment

There are different transport pathways and modalities for the transport of uranium through estuaries and the coastal environment: (i) directly through groundwater or in freshwater discharges; (ii) directly through surface and subsurface runoff from the terrestrial environment (e.g. in springs or seeps); and (iii) indirectly through smaller rivers and streams, which are not directly connected to the marine environment.

Both estuarine and coastal environments are described in detail in Ref. [6.187] and their characteristics have been summarized in Ref. [6.188]. Estuaries are described as complex systems which include channels, bare areas and/or vegetational stabilized sediment and salt marshes. They have a transient estuarine configuration with changes over a large timescale (from that of the tidal cycle to channel alteration over hundreds of years) as a result of different co-factors such as sea level increase, sediment supply and human activities (e.g. dredging). Lagoons are distinct coastal systems having a continuous barrier between them and the sea, which facilitate the water exchange.

In general, estuaries tend to be net sediment deposition systems, with fluvially transported sediments interacting with incoming tidal waters to give rise to deposition either within the estuary or in a submarine delta beyond its mouth. However, although estuaries are generally accreting systems, this should not be considered a unidirectional process. Sediment banks and water channels will change continually so that deposited sediments may be eroded by the migration of the channels and deposited elsewhere (either within the estuary or beyond its mouth). Unusually high water levels may occur under storm conditions, causing inundation of salt marshes and remobilizing much of the deposited sediment. Indeed, salt marshes can be removed down to the underlying bedrock surface by severe winter storms. Thus, both waters and sediments in estuaries are likely to be highly mobile and this tends to limit radionuclide residence times, even for elements that are much more strongly particle reactive than uranium.

The coastal environment is associated with the cliff line or line of highest tide, and it comprises the beach (if it exists) and the foreshore. This region is in permanent contact with the sea, with a frequency ranging from every tidal cycle down to a few times per year (when inundations occur), and is significantly influenced by wave action. Consequently, the transport of shallow bottom sediments in the near shore coastal region is also influenced by waves. Sediments transported by the rivers seawards generally deposit within estuaries or may build up deltas at the coast.

In the coastal environment, the sediments available are typically sand sized or larger. Radionuclide sorption on these sediments will, in general, be less than would occur on finer materials owing to both the lower surface area:volume ratio of larger particles and because the mineralogy of finer clays and silts has an intrinsically greater sorptive capacity than the mineralogy (typically quartz) associated with coarser fractions. Furthermore, the sediments are subject to relatively rapid transport both onshore and offshore, and along the coastline. Water movement through much of the sediment is likely to be rapid, with inundation and draining at each tidal cycle or, at least, several times per year. Thus, again, radionuclide residence times are likely to be limited.

In near shore and offshore environments, radionuclides may exhibit long term accumulation in bottom sediments. These environments will also be much less labile than estuarine or coastal environments. Detailed modelling of estuarine and coastal evolution is possible at a site specific level, but requires a large historical database coupled with sophisticated analytical techniques. The marine environment is conveniently distinguished into a near shore component, immediately seaward of the lowest low water mark and extending out to a depth of 10–20 m, where the effects of wave action on bottom sediments become negligible, and an offshore component that extends from this boundary farther out to sea.

6.5.2. Water–sediment interactions

Uranium K_d data for marine sediments are limited, however marine K_d values are generally considered to be less variable than those for freshwater. The reference values provided by the IAEA are based on exchange with carbonates in sediments [6.189] and given as the ratio of the uranium activity concentration in sediment (Bq/kg) to the uranium activity concentration in sea water (Bq/kg). A more common approach is the use of the ratio of the uranium activity concentration in sediment (Bq/kg) to the volumetric activity concentration of uranium in sea water (Bq/L). When site specific K_d data are not available for radiological environmental impact assessments, the recommended uranium K_d value in Ref. [6.189] of 500 L/kg for open ocean and 1000 L/kg for ocean margin can be used. The approach used in Ref. [6.189] for K_d determination does not adequately reflect the in situ behaviour of radionuclides, and the values recommended in Ref. [6.189] should be used with caution.

6.5.3. Transfers to marine biota: Uranium concentration ratios

The average uranium concentration value in sea water from coastal environments as well as in the deep sea is about 3.3 μg/L (corresponding to 38 mBq/L of ^{238}U), considering that the concentration of dissolved uranium in sea water is relatively similar across marine systems. At present, unusual uranium accumulation or cases of uranium buildup in the food chain have not been reported, allowing a reasonable confidence estimation of concentration ratios when direct measurements are not available. Concentration ratio values for various groups of marine biota have been reviewed and compiled by the IAEA [6.189], and a summary of these is given in Table 6.14.

TABLE 6.14. RECOMMENDED CONCENTRATION RATIOS FOR URANIUM IN MARINE BIOTA [6.189]

Biota	Recommended concentration ratio	Comments
Fish	1.0	The recommended concentration ratio was increased from the previous value of 0.1 [6.190] to 1.0 to allow for the possible inclusion of some bone in the edible fraction
Crustaceans	10	Given in Ref. [6.190]
Molluscs (excluding cephalopods)	30	A mean ^{238}U concentration of 1.2 Bq/kg (fresh weight) was used to estimate the recommended concentration ratio and it was derived from four different lamellibranch molluscs [6.191]
Macroalgae	100	Previously, a concentration ratio value of 10 was recommended [6.190], but this was increased to 100 by the IAEA [6.189], as much higher values were reported in brackish water [6.192, 6.193]
Zooplankton	30	Derived using data from Ref. [6.194]
Phytoplankton	20	Derived using data from Ref. [6.195]

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Chapter 7

ASSESSMENT OF THE RADIOLOGICAL AND CHEMICAL IMPACTS OF URANIUM

M.C. THORNE
Mike Thorne and Associates Ltd, United Kingdom

T. YANKOVICH, A. ULANOWSKI
International Atomic Energy Agency

7.1. URANIUM IN FOOD AND DRINKING WATER

Background radiation levels, climatic conditions and agriculture practices are among the major factors that contribute to the wide range of uranium concentrations in foods and drinking water [7.1]. Representative concentrations of uranium in various food products and in drinking water are listed in Tables 7.1 and 7.2. It should be noted that the values in Table 7.1 are taken from table 15 of Ref. [7.1] and are in units of millibecquerels per kilogram of ^{238}U. Reference values are given in Table 7.2 in mass terms, considering the specific activity of natural uranium of 12.3 Bq of ^{238}U per milligram of uranium [7.2]. The highest concentrations in drinking water listed in Table 7.1 (up to 150 Bq/kg) correspond to mass concentrations of up to 12 mg [U]/L, which would not be acceptable for human consumption on the grounds of chemical toxicity (discussed in Section 7.4).

Based on the data shown in Table 7.1, UNSCEAR [7.1] estimated the annual intake of uranium in diet to vary from 2.9 Bq of ^{238}U in India to 57 Bq of ^{238}U in China, primarily owing to differences in drinking water concentrations. The reference value was given as 5.7 Bq of ^{238}U, corresponding to 460 µg. Based on detailed analyses of 116 human foodstuffs, Ref. [7.3] estimates a change in the daily intake of uranium by adults in Germany after reunification from 2.0 to 3.0 µg [U]/d for men and from 2.2 to 2.6 µg [U]/d for women, presumably because of different averaging of food sources. An intake of 3.0 µg [U]/d corresponds to 13.4 Bq of ^{238}U per year, which is in line with estimates for other countries.

TABLE 7.1. ACTIVITY CONCENTRATION OF ^{238}U IN FOODS AND DRINKING WATER

Country	^{238}U activity concentration or range of values (mBq/kg)						
	Milk products	Meat products	Grain products	Leafy vegetables	Root vegetables and fruits	Fish products	Drinking water
China	13	10	9.8	16	13	12	0.1–700
Finland	—[a]	—[a]	—[a]	—[a]	—[a]	—[a]	0.5–150 000
France	—[a]	—[a]	—[a]	—[a]	—[a]	—[a]	4.4–930
Germany	—[a]	1–20	20–400	6–2 200	10–2 900	—[a]	0.4–600
India	17	—[a]	7.4–67	61–72	0.4–77	—[a]	0.09–1.5
Italy	—[a]	—[a]	—[a]	—[a]	—[a]	—[a]	0.5–130
Japan	0.55	13	1.2	—[a]	26	—[a]	—[a]
Poland	2.6	1.6–5.6	4.7–11	14–15	0.9–10	—[a]	7.3
Romania	—[a]	—[a]	6.1–85	—[a]	6–120	—[a]	0.4–37
Spain	—[a]	—[a]	—[a]	—[a]	—[a]	—[a]	3.7–4.4
Switzerland	—[a]	—[a]	—[a]	—[a]	—[a]	—[a]	0–1 000
United Kingdom	0.1–4.9	4.9	6.2–35	9.8–400	6	2.5	—[a]
USA	0.7	0.8–2.3	3–23	24	0.9–7.7	13–1 900	0.3–77
Reference value	1	2	20	20	3	30	1

Source: Based on table 15 of Ref. [7.1].
[a] —: data not available.

TABLE 7.2. TYPICAL CONCENTRATION OF URANIUM IN FOODS AND DRINKING WATER

Food	Activity concentration of ^{238}U (mBq/kg)	Mass concentration of uranium (μg/kg)
Milk products	1	0.081
Meat products	2	0.163
Grain products	20	1.63
Leafy vegetables	20	1.63
Root vegetables and fruits	3	0.244
Fish products	30	2.44
Drinking water	1	0.081

Note: Data derived from Table 7.1.

7.2. DISTRIBUTION OF URANIUM IN HUMAN ORGANS AND TISSUES AND LOSSES BY EXCRETION

The total body content of uranium in Reference Man has been estimated to be 90 μg, with about 31 μg in soft tissues and 59 μg in the skeleton [7.4]. Annual intakes for Reference Man [7.4] were estimated to be 694 μg by ingestion and 2.6 μg by inhalation. Typical excretory losses were estimated at 0.05–0.5 μg/d (18–182μg/a) in urine, 1.4–1.8 μg/d (511–657 μg/a) in faeces and 0.02 μg/d (7.3 μg/a) in hair. The predominance of faecal excretion is consistent with the low fractional degree of absorption of uranium from the gastrointestinal tract (see Section 7.3.2). Much higher rates of urinary excretion of up to about 10 μg/d were reported for some areas (e.g. in Ohio, USA). More recent data for the US population (tables 6–8 of Ref. [7.5]) give a much lower geometric mean of urinary excretion rate of around 0.01 μg/d (based on a urinary excretion rate of 1.4 L/d), with a 95th percentile rate of about 0.06 μg/d. In that study, excretion rates were not strongly dependent on age, sex or ethnicity. In the Czech Republic, urinary excretion rates have been estimated to be 0.009 5–0.016 1 μg/d [7.6] (i.e. similar to those for the US population).

The mean worldwide uranium concentration in whole blood was estimated at 0.58 µg/kg [7.5]. Uranium concentration values in the lungs, liver, kidneys and bones (vertebrae, ribs and skeleton) of different age groups of people from the USA were reported as 0.5–1.17, 0.12–0.33, 0.39–1.00 and 0.25–1.9 µg/kg, respectively [7.5]. No specific accumulation of uranium in skeletal tissues was found in this study.

The amounts of uranium in tissues and organs were measured for four Asian populations (China, India, Republic of Korea, Philippines) [7.7] and the results compared with the amounts derived for the Japanese population based on earlier studies and those given for Reference Man [7.4]. These data are summarized in Table 7.3.

In Ref. [7.7], it is noted that although the median value for uranium in kidneys for the Asian population is about a factor of 40 lower than that reported for Reference Man [7.4], it compares closely with the value of 0.13 ± 0.08 µg reported for the adult US population [7.8]. The Reference Man data are based on analyses from the 1960s and are likely to be less reliable than the more recent data. For liver, it is noted in Ref. [7.7] that the value for the Asian population is within the global range of 0.1–0.5 µg reported in Ref. [7.9] and comparable with the value of 0.36 ± 0.56 µg reported for the adult US population [7.8]. For skeleton, the median value for Asian residents of 5.2 µg is an order of magnitude lower than the value for Reference Man but is comparable with the value of 6.6 ± 5.8 µg reported for the adult US population [7.8]. For the lungs, the Asian value is higher than the US value of 0.5 ± 0.39 µg reported in Ref. [7.8].

TABLE 7.3. URANIUM CONTENT IN TISSUES AND ORGANS OF FIVE ASIAN POPULATIONS AND REFERENCE MAN [7.7]

Tissue or organ	Uranium content (µg)			
	China, India, Republic of Korea and Philippines		Japan	Reference Man
	Range	Median		
Kidney	0.14–0.26	0.19	0.11	7.0
Liver	0.08–0.77	0.20	0.38	0.45
Skeleton	2.85–12.58	5.23	6.3	59.0
Lung	0.36–1.92	1.09	2.0	1.00

It has been suggested [7.7] that the higher Asian value may be due to higher concentrations of dust in air in tropical Asian regions.

7.3. MODELS AND DATA FOR ESTIMATING INTERNAL EXPOSURES TO HUMANS

7.3.1. Biokinetic modelling

Biokinetic models for humans establish relationships between time dependent intakes of a substance and time dependent concentrations of that substance or substances derived from it in tissues and organs and in excreta. In the case of uranium, it is convenient to discuss biokinetic modelling in terms of uptake from the gastrointestinal tract, retention in, and uptake from the respiratory system, and retention in tissues and organs, together with subsequent excretion.

7.3.2. Gastrointestinal transport and uptake

Uranium is taken up from the gastrointestinal tract to only a limited degree. For soluble forms of the element, the fractional absorption is seldom >0.05 and can be <0.01. For insoluble forms, such as UO_2, fractional gastrointestinal absorption may be considerably lower than this. Overall, the fractional gastrointestinal absorption of uranium can vary from <0.1% to 6%, depending on the solubility of the uranium compound [7.5]. Studies in volunteers indicate that approximately 0.02 of the uranium from drinking water and dietary sources is absorbed in humans [7.10–7.12]. The ICRP [7.13] undertook a comprehensive review of the gastrointestinal absorption of various chemical forms of uranium and recommended that the fractional gastrointestinal absorption should be taken as 0.002 for insoluble compounds and 0.02 for soluble hexavalent compounds.

7.3.3. Retention in, and uptake from, the respiratory tract

Deposition in the respiratory system, subsequent retention and redistribution within that system, and transfers to the gastrointestinal tract and systemic circulation can be represented using the ICRP respiratory tract model [7.14], but with parameter values that have subsequently been updated [7.15]. This approach is in almost universal use for modelling the deposition and retention of radioactive aerosols in the respiratory tract, but it is also used by the US Agency for Toxic Substances and Disease Registry (ATSDR) [7.5] in the context of evaluations of the chemical toxicity of uranium.

The size of dust particles is relevant for the deposition of inhalable uranium in different locations of the lungs. Size is generally given in terms of the aerodynamic diameter of the particles rather than their physical diameter. The 'aerodynamic' diameter is the diameter of a particle with unit density (1×10^3 kg/m^3) that has the same terminal settling velocity in air as the actual particle. This factor is significant for uranium, as compounds of the element typically have a density of around 1×10^4 kg/m^3, implying that the aerodynamic diameter is about a factor of three larger than the physical diameter.

Aerosols are also typically log-normally distributed in size. Their total deposition in the respiratory tract and the distribution of that deposition between the different anatomical regions is determined primarily by the median aerodynamic diameter and only secondarily by the associated geometric standard deviation. Thus, it is usual to report deposition and transport in the respiratory system in terms of either the mass median aerodynamic diameter (MMAD) or the activity median aerodynamic diameter (AMAD) of the aerosol. Typically, the MMAD and AMAD will be similar in magnitude, but this will not always be the case for aerosols of heterogeneous composition. In the context of radiological protection, values of committed effective dose per unit intake by inhalation are typically given as functions of both age at intake and AMAD, and for materials of different degrees of solubility in the respiratory tract [7.13].

Small particles can reach the alveolar region and subsequently transfer into the blood if the uranium compound is soluble, while particles with high MMAD or AMAD (>10 μm) have a high probability of being transported out of the extra-thoracic and tracheobronchial regions and swallowed [7.5, 7.14]. Fast soluble uranium compounds (e.g. uranium hexafluoride, uranyl fluoride, uranium tetrachloride, uranyl nitrate hexahydrate), likely to be absorbed into the blood within days, are defined by the ICRP [7.13] as inhalation type F (fast dissolution). Less soluble compounds (e.g. uranium tetrafluoride, uranium trioxide, triuranium octaoxide), with a residence time in the lung tissue of weeks, are classified as type M (medium dissolution), while insoluble compounds (e.g. uranium dioxide) are designated type S (slow dissolution) [7.5, 7.13]. In practice, compounds of uranium can exhibit characteristics that are intermediate between those of type F, M and S aerosols, so it may sometimes be appropriate to adjust the parameter values of the ICRP model to take account of this (e.g. based on urine monitoring data [7.15] or using results from dissolution of aerosol material in simulated lung fluids [7.16]).

In the ICRP respiratory tract model [7.14], mechanical clearance of aerosol particles by mucocilliary action and through macrophage involvement competes with solubilization. Thus, some of the deposited material is transported to the gastrointestinal tract and some is transferred to the systemic circulation.

7.3.4. Systemic retention

Uranium entering the systemic circulation either by uptake from the gastrointestinal tract or directly from the respiratory tract is widely distributed among soft tissues, but is primarily deposited and retained in mineral bone, owing to chemical similarities between uranium and calcium. Thus, it initially deposits on bone surfaces, but then becomes redistributed throughout the mineral matrix both in trabecular (spongy) and cortical (compact) bone. Uranium is also preferentially transferred to, and retained in, the kidneys, which is why its nephrotoxic effects are an important consideration in limiting exposures to the element. For modelling purposes, uranium contents in tissues are usually normalized to the liver concentrations, although the liver is not a major repository for uranium. A value of about 90 μg is given for the normal adult body burden of uranium, with a distribution of about 66% of this total in bone, 16% in the liver, 8% in the kidneys and 10% in other tissues [7.13, 7.17, 7.18]. A comprehensive, multicompartmental model for the distribution and retention of uranium in the human body has been developed by the ICRP [7.13, 7.18] and is illustrated in Fig. 7.1.

Parameter values for use with this model differ with age, but not with the physical or chemical form ingested or inhaled. These age dependent data are tabulated by the ICRP [7.18] and are not reproduced here.

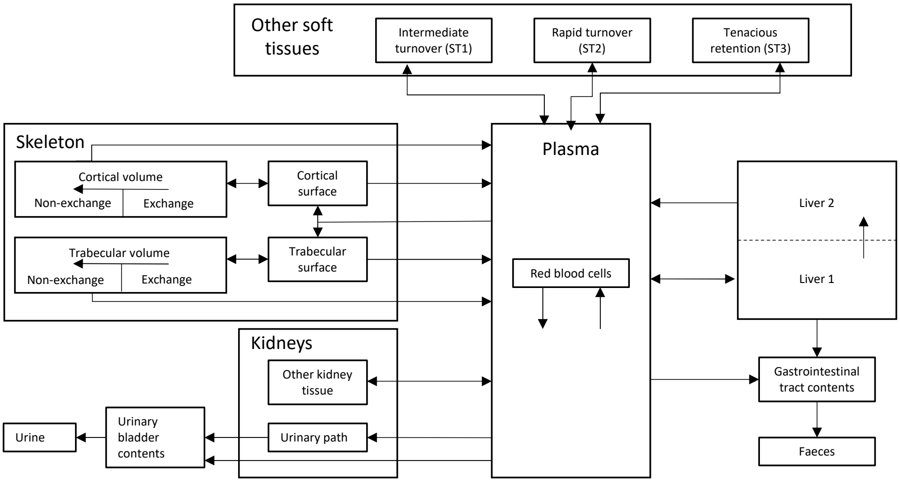

FIG. 7.1. Biokinetic model for uranium after entry to the systemic circulation (adapted from Ref. [7.18] with permission from the International Commission on Radiological Protection).

7.3.5. Data on the radiotoxicity of uranium

The radiotoxicity of uranium is expressed in terms of the committed effective dose received per unit intake. Conventionally, the intake is measured in terms of activity and the units of committed effective dose per unit intake are sieverts per becquerel. Values of committed effective dose per unit intake expressed in these terms are given for occupational and public exposure by the ICRP [7.19]. Appropriate dose coefficients for members of the public are listed in Table 7.4, which includes both uranium radioisotopes and their progeny, since the data for progeny are required when estimating the radiotoxicity per unit mass of uranium, allowing for ingrowth of those progeny, as discussed below.

To make comparisons between the chemical toxicity and radiotoxicity of uranium, it is often useful to give values of committed effective dose per unit mass of ingested or inhaled uranium. In this context, consideration must be given as to whether the uranium is depleted, natural or enriched, as well as to the degree to which shorter lived progeny are present together with the isotopes of uranium. In addition, the physical and chemical form of the uranium are to be considered.

TABLE 7.4. COMMITTED EFFECTIVE DOSE PER UNIT INTAKE FOR ADULT MEMBERS OF THE PUBLIC

Radionuclide	Branching ratio	Effective dose per unit intake for adult members of the public (Sv/Bq)			
		Inhalation of type F	Inhalation of type M	Inhalation of type S	Ingestion
^{238}U	1.00	5.00×10^{-7}	2.90×10^{-6}	8.00×10^{-6}	4.50×10^{-8}
^{234}Th	1.00	2.50×10^{-9}	6.60×10^{-9}	7.70×10^{-9}	3.40×10^{-9}
^{234}Pa	1.00	—[a]	3.80×10^{-10}	4.00×10^{-10}	5.10×10^{-10}
^{234}U	1.00	5.60×10^{-7}	3.50×10^{-6}	9.40×10^{-6}	4.90×10^{-8}
^{230}Th	1.00	1.00×10^{-4}	4.30×10^{-5}	1.40×10^{-5}	2.10×10^{-7}
^{226}Ra	1.00	3.60×10^{-7}	3.50×10^{-6}	9.50×10^{-6}	2.80×10^{-7}
^{214}Pb	1.00	2.80×10^{-9}	1.40×10^{-8}	1.50×10^{-8}	1.40×10^{-10}
^{214}Bi	1.00	7.10×10^{-9}	1.40×10^{-8}	—[a]	1.10×10^{-10}

TABLE 7.4. COMMITTED EFFECTIVE DOSE PER UNIT INTAKE FOR ADULT MEMBERS OF THE PUBLIC (cont.)

Radionuclide	Branching ratio	Effective dose per unit intake for adult members of the public (Sv/Bq)			
		Inhalation of type F	Inhalation of type M	Inhalation of type S	Ingestion
^{210}Pb	1.00	9.00×10^{-7}	1.10×10^{-6}	5.60×10^{-6}	6.90×10^{-7}
^{210}Bi	1.00	1.10×10^{-9}	9.30×10^{-8}	—[a]	1.30×10^{-9}
^{210}Po	1.00	6.10×10^{-7}	3.30×10^{-6}	4.30×10^{-6}	1.20×10^{-6}
^{235}U	1.00	5.20×10^{-7}	3.10×10^{-6}	8.50×10^{-6}	4.70×10^{-8}
^{231}Th	1.00	7.80×10^{-11}	3.10×10^{-10}	3.30×10^{-10}	3.40×10^{-10}
^{231}Pa	1.00	—[a]	1.40×10^{-4}	3.40×10^{-5}	7.10×10^{-7}
^{227}Ac	1.00	5.50×10^{-4}	2.20×10^{-4}	7.20×10^{-5}	1.10×10^{-6}
^{227}Th	0.99	6.70×10^{-7}	8.50×10^{-6}	1.00×10^{-5}	8.80×10^{-9}
^{223}Fr	0.01	8.90×10^{-10}	—[a]	—[a]	2.40×10^{-9}
^{223}Ra	1.00	1.20×10^{-7}	7.40×10^{-6}	8.70×10^{-6}	1.00×10^{-7}
^{211}Pb	1.00	3.90×10^{-9}	1.10×10^{-8}	1.20×10^{-8}	1.80×10^{-10}

Note: Types F (fast), M (medium) and S (slow) relate to different rates of solubilization of aerosols in the respiratory system (see Section 7.3.3 for details).

[a] —: data not available.

All of these factors are addressed in Ref. [7.2], where a tabulation is provided of values of committed effective dose per unit mass intake (Sv/mg) for natural uranium (with and without progeny in equilibrium), depleted uranium and enriched uranium (1–90% enriched). These values are listed in Table 7.5 for both occupationally exposed persons and adult members of the public.

TABLE 7.5. COMMITTED EFFECTIVE DOSE PER UNIT MASS INTAKE OF VARIOUS FORMS OF URANIUM [7.2]

Material	Committed effective dose per unit mass intake of various forms of uranium (Sv/mg)							
	Occupationally exposed persons				Adult members of the public			
	Inhalation of type F	Inhalation of type M	Inhalation of type S	Ingestion	Inhalation of type F	Inhalation of type M	Inhalation of type S	Ingestion
Natural: no progeny	1.54×10^{-5}	4.66×10^{-5}	1.57×10^{-4}	1.22×10^{-6}	1.34×10^{-5}	8.06×10^{-5}	2.19×10^{-4}	1.23×10^{-6}
Natural: progeny in equilibrium	2.04×10^{-3}	6.00×10^{-4}	4.62×10^{-4}	1.97×10^{-5}	1.66×10^{-3}	9.24×10^{-4}	7.03×10^{-4}	3.16×10^{-5}
Depleted uranium	1.53×10^{-5}	4.62×10^{-5}	1.56×10^{-4}	1.21×10^{-6}	1.33×10^{-5}	7.99×10^{-5}	2.17×10^{-4}	1.22×10^{-6}
1% enriched: no ^{234}U enrichment	1.55×10^{-5}	4.70×10^{-5}	1.59×10^{-4}	1.23×10^{-6}	1.35×10^{-5}	8.13×10^{-5}	2.21×10^{-4}	1.24×10^{-6}
1% enriched: ^{234}U enrichment	1.96×10^{-5}	6.05×10^{-5}	2.02×10^{-4}	1.54×10^{-6}	1.71×10^{-5}	1.04×10^{-4}	2.81×10^{-4}	1.56×10^{-6}
2% enriched: no ^{234}U enrichment	1.59×10^{-5}	4.81×10^{-5}	1.62×10^{-4}	1.26×10^{-6}	1.38×10^{-5}	8.31×10^{-5}	2.26×10^{-4}	1.27×10^{-6}
2% enriched: ^{234}U enrichment	3.47×10^{-5}	1.10×10^{-4}	3.62×10^{-4}	2.70×10^{-6}	3.03×10^{-5}	1.84×10^{-4}	5.02×10^{-4}	2.71×10^{-6}

TABLE 7.5. COMMITTED EFFECTIVE DOSE PER UNIT MASS INTAKE OF VARIOUS FORMS OF URANIUM [7.2] (cont.)

Material	Committed effective dose per unit mass intake of various forms of uranium (Sv/mg)							
	Occupationally exposed persons				Adult members of the public			
	Inhalation of type F	Inhalation of type M	Inhalation of type S	Ingestion	Inhalation of type F	Inhalation of type M	Inhalation of type S	Ingestion
3% enriched: no ^{234}U enrichment	1.62×10^{-5}	4.92×10^{-5}	1.66×10^{-4}	1.28×10^{-6}	1.41×10^{-5}	8.50×10^{-5}	2.31×10^{-4}	1.30×10^{-6}
3% enriched: ^{234}U enrichment	4.97×10^{-5}	1.59×10^{-4}	5.22×10^{-4}	3.85×10^{-6}	4.34×10^{-5}	2.68×10^{-4}	7.23×10^{-4}	3.86×10^{-6}
4% enriched: no ^{234}U enrichment	1.65×10^{-5}	4.99×10^{-5}	1.68×10^{-4}	1.30×10^{-6}	1.43×10^{-5}	8.62×10^{-5}	2.34×10^{-4}	1.32×10^{-6}
4% enriched: ^{234}U enrichment	6.47×10^{-5}	2.08×10^{-4}	6.81×10^{-4}	5.00×10^{-6}	5.65×10^{-5}	3.50×10^{-4}	9.43×10^{-4}	5.01×10^{-6}
5% enriched: no ^{234}U enrichment	1.68×10^{-5}	5.09×10^{-5}	1.72×10^{-4}	1.33×10^{-6}	1.46×10^{-5}	8.80×10^{-5}	2.39×10^{-4}	1.34×10^{-6}
5% enriched: ^{234}U enrichment	7.97×10^{-5}	2.57×10^{-4}	8.40×10^{-4}	6.14×10^{-6}	6.96×10^{-5}	4.32×10^{-4}	1.16×10^{-3}	6.16×10^{-6}

TABLE 7.5. COMMITTED EFFECTIVE DOSE PER UNIT MASS INTAKE OF VARIOUS FORMS OF URANIUM [7.2] (cont.)

Material	Committed effective dose per unit mass intake of various forms of uranium (Sv/mg)							
	Occupationally exposed persons				Adult members of the public			
	Inhalation of type F	Inhalation of type M	Inhalation of type S	Ingestion	Inhalation of type F	Inhalation of type M	Inhalation of type S	Ingestion
20% enriched: ^{234}U enrichment	2.21×10^{-4}	7.21×10^{-4}	2.34×10^{-3}	1.70×10^{-5}	1.94×10^{-4}	1.21×10^{-3}	3.24×10^{-3}	1.70×10^{-5}
90% enriched: ^{234}U enrichment	1.52×10^{-3}	4.98×10^{-3}	1.62×10^{-2}	1.17×10^{-4}	1.33×10^{-3}	8.31×10^{-3}	2.23×10^{-2}	1.17×10^{-4}

Note: Types F (fast), M (medium) and S (slow) relate to different rates of solubilization of aerosols in the respiratory system (see Section 7.3.3 for details). For depleted uranium, the calculations assume that ^{234}U is in secular equilibrium with ^{238}U. If ^{234}U is depleted to any significant degree, the values of committed effective dose per unit mass intake will be somewhat decreased.

7.4. MODELS AND DATA FOR ESTIMATING CHEMICAL TOXICITY TO HUMANS

In terms of chemical toxicity to humans, all but the most insoluble forms of the element are primarily of concern because of their adverse effects on the kidneys [7.5, 7.20]. Taking this into account, two broad approaches to evaluating toxicological significance and limiting intakes can be adopted. The first is that adopted by the ATSDR, in which human and animal exposures (expressed as air concentrations for inhalation and intake rates for ingestion) are evaluated in terms of their adverse health impacts. A first approach is the establishment of NOAELs or LOAELs for inhalation and ingestion exposures. MRLs for exposure and associated uncertainty factors (Table 7.6) are set by deriving estimates of daily human exposure to a hazardous substance that are unlikely to be linked to an appreciable risk of adverse, non-cancer health effects in specific exposure conditions. Table 7.6 summarizes the MRLs recommended by the ATSDR [7.5] and gives the basis for their derivation (see also Ref. [7.21]).

In Table 7.6, the benchmark dose lower bound (BMDL) is defined by the ATSDR [7.5] to be a concentration or intake rate that produces a predetermined change in the response rate of an adverse effect in an organism compared with the background. Thus, $BMDL_{10}$ represents a 10% change in response rate. Acute, intermediate and chronic exposure durations are <14 d, 14–365 d and >365 d, respectively. It should be noted that MRL values for ingestion are expressed per unit of body mass. Thus, they imply different limits on total intake rates for infants, children and adults.

In a second approach, the maximum acceptable kidney concentration (that is unlikely to be associated with an appreciable risk of adverse non-cancer effects) [7.20, 7.21] is used to set a limit — which relates to an intake rate by ingestion or inhalation — by application of a biokinetic model. Animal studies conducted in the early 1940s suggest that up to 2–3 μg [U]/g kidney might not induce serious effects; however, no defined toxicity threshold was given. Data from the 1950s from workers exposed to high concentrations of uranium in air and data from experiments on patients injected with uranium reveal no serious effects on humans from concentrations of 2–6 μg [U]/g kidney. In 1959, the ICRP set a concentration limit of 3 μg [U]/g kidney.

Following the work in Ref. [7.22], which raises concerns of mild renal injuries at concentrations of 0.1–0.4 μg [U]/g kidney, it was concluded [7.20] that the toxic action of uranium in kidneys may depend on the level and pattern of exposure, and repeated exposures may bring on the development of tolerance to uranium.

The kidney may also develop a kind of acquired tolerance to uranium after repeated exposures, but a kidney that has developed such tolerance is not normal.

TABLE 7.6. MINIMAL RISK LEVELS FOR URANIUM

Route	Form	Duration	Minimal risk level	Basis	Uncertainty factor
Inhaled	Soluble	Intermediate	1×10^{-4} mg [U]/m^3	LOAEL of 0.15 mg [U]/m^3 for minimal renal lesions in dogs. Adjusted for intermittent exposure, resulting in an adjusted LOAEL of 0.032 mg [U]/m^3.	300
		Chronic	4×10^{-5} mg [U]/m^3	One year dog study involving exposure to UCl_4. A $BMCL_{10}$ of 0.019 mg [U]/m^3 was selected. This $BMCL_{10}$ was adjusted for intermittent exposure, resulting in an adjusted $BMCL_{10}$ of 0.003 7 mg [U]/m^3.	100
	Insoluble	Intermediate	2×10^{-3} mg [U]/m^3	A NOAEL of 1.1 mg [U]/m^3 in dogs was used (based on renal effects). This value was adjusted for intermittent exposure, resulting in an adjusted NOAEL of 0.24 mg [U]/m^3.	100

TABLE 7.6. MINIMAL RISK LEVELS FOR URANIUM (cont.)

Route	Form	Duration	Minimal risk level	Basis	Uncertainty factor
		Chronic	8×10^{-4} mg [U]/m^3	A LOAEL of 5.1 mg [U]/m^3 for lung fibrosis and metaplasia in rhesus monkeys was selected. This LOAEL was adjusted for intermittent exposure, resulting in an adjusted LOAEL of 0.82 mg [U]/m^3.	1000
Ingested	Soluble	Acute	2×10^{-3} mg [U]·kg^{-1}·d^{-1}	A BMDL$_{05}$ value of 0.20 mg [U] · kg^{-1} · d^{-1} for cleft palate in mice was selected; this is lower than the BMDL$_{05}$ values for other foetal effects in mice, and is approximately ten times lower than the LOAEL for maternal and foetal body weight effects. Thus, it is likely to be protective of the other effects.	100
		Intermediate	2×10^{-4} mg [U]·kg^{-1}·d^{-1}	Statistically significant increases in renal lesions in rats with a LOAEL value of 0.06 mg [U] · kg^{-1} · d^{-1}.	300

TABLE 7.6. MINIMAL RISK LEVELS FOR URANIUM (cont.)

Route	Form	Duration	Minimal risk level	Basis	Uncertainty factor
		Chronic	2×10^{-4} mg [U]·kg^{-1}·d^{-1}	Given the ability of the kidney to repair renal damage at low exposure levels, the intermediate-duration oral minimal risk level was taken as protective for chronic exposures.	Not applicable

Source: Based on Ref. [7.5].

Note: BMCL: benchmark concentration lower bound; BMCL_{10}: BMCL associated with a benchmark response of 10%; BMDL: benchmark dose lower bound; BMDL_{05}: BMDL associated with a benchmark response of 5%; LOAEL: lowest observed adverse effect level; NOAEL: no observed adverse effect level.

Tolerant animals have high urine volumes, a diminished glomerular filtration rate and a loss of concentrating capacity by the kidney. According to Ref. [7.20], the use of the concentration of 3 μg [U]/g kidney applied for many years as a guidance level for limiting occupational exposures was based on tests of chemical toxicity that were less sensitive, and on definitions of chemical toxicity that were less stringent, than those developed subsequently. Moreover, in the underpinning human studies, the subjects may have been exposed to kidney concentrations of the order of 3 μg [U]/g kidney only for very brief periods. Reference [7.20] concludes that it might be prudent to lower this long standing guidance level by roughly an order of magnitude.

More recently, WHO [7.23] has set the tolerable daily intake (TDI) of uranium to 60 μg, based on an epidemiological study [7.24] that was used to define a no-effect group. The value of the 95th percentile of the uranium exposure distribution for this group was estimated to be 1094 μg/d (the 95% confidence interval on the 95th percentile was 637–1646 μg/d). The TDI was taken to be 10% of the lower bound of the 95% confidence interval on the 95th percentile of the exposure distribution for this group. For comparison, the ATSDR [7.5] recommends an MRL value for chronic exposure to uranium by ingestion of $0.2\ \mu g \cdot kg^{-1} \cdot d^{-1}$. For a reference adult with a mass of 70 kg [7.4], this corresponds to 14 μg/d. Given the very different bases of derivation, the relatively small difference between the TDI and MRL values suggests that a robust basis exists for limiting intakes of uranium by ingestion.

To interpret the TDI of WHO in terms of the equivalent kidney concentration, it is necessary to establish the relationship between the chronic intake rate of uranium and the maintained kidney concentration that is achieved at equilibrium. Reference [7.21] shows that an ingestion intake rate of 1 μg [U]/d by adults corresponds to a maintained kidney concentration of 2.2×10^{-4} μg [U]/g kidney. Thus, the TDI corresponds to 0.013 μg [U]/g kidney. This is well below the value of 0.3 μg [U]/g kidney that Ref. [7.20] considered prudent.

For inhalation, Ref. [7.21] shows that the maintained kidney concentrations in adult members of the public chronically exposed to uranium by inhalation at an intake rate of 1 μg [U]/d are 2.7×10^{-3}, 1.1×10^{-3} and 2.1×10^{-4} μg [U]/g kidney for type F, M and S aerosols, respectively, with the default aerosol size characteristics adopted by the ICRP [7.13, 7.14]. Thus, considering soluble forms of uranium to be type F or M and to have an MRL value of 4×10^{-2} μg [U]/m^3, and adopting an inhalation rate of 22 m^3/d [7.4], chronic intake at the MRL would result in a maintained kidney concentration of $(1.0–2.4) \times 10^{-3}$ μg [U]/g kidney. Again, this is well below the value of 0.3 μg [U]/g kidney that Ref. [7.20] considered prudent.

In contrast, for occupational exposure, the United Kingdom Health and Safety Executive [7.25] adopts a long term exposure limit for uranium of

200 μg [U]/m^3. For occupational situations, Ref. [7.21] gives maintained kidney concentrations for chronic exposure to uranium by inhalation at an intake rate of 1 μg [U]/d of 3.2×10^{-3}, 7.7×10^{-4} and 1.7×10^{-4} μg [U]/g kidney for type F, M and S aerosols, respectively. In this case, the inhalation rate is typically 1.2 m^3/h for 8 h/d. Thus, for prolonged exposure at the exposure limit, the maintained kidney concentrations would be 6, 1.5 and 0.3 μg [U]/g kidney for type F, M and S aerosols, respectively. These values are comparable to the threshold adopted by the ICRP in 1959, but only for type S aerosols is the value sufficiently low to be consistent with the recommendations of Ref. [7.20].

As already noted, WHO [7.23] set the TDI for uranium as 60 μg. Based on a water consumption rate of 2 L/d, WHO [7.23] set a provisional guideline value for uranium in drinking water of 30 μg [U]/L. This may be compared to the value of 20 μg [U]/L adopted in Canada [7.26] and Australia [7.27], and that of 30 μg [U]/L in the USA [7.28]. To date, no European statutory limits have been imposed for uranium in drinking water. However, European Union Council Directive 98/83/EC [7.29] stipulates that waters in which alpha emitter concentrations exceed 0.1 Bq/L should be investigated to determine what corrective action, if any, is required. For natural uranium present without any progeny, 0.1 Bq/L corresponds to 4 μg [U]/L, which is a more stringent restriction than the values recommended to prevent chemotoxic effects.

7.5. DOSE CONVERSION FACTORS FOR ESTIMATING EXTERNAL AND INTERNAL DOSES TO BIOTA

Fundamentally, the approach to estimating external and internal dose rates to humans and other types of biota are identical. Geometric models are established both for the environment and for the exposed individual, a distribution of contamination is specified either in the environment, for external exposure, or within the individual, for internal exposure, and radiation transport calculations are undertaken to determine the pattern of energy deposition within the exposed individual. However, whereas for humans the geometric models are typically complex anthropomorphic phantoms that include detailed representations of internal tissues and organs, for other organisms the phantoms are typically simple ellipsoids. Until recently, only average whole body dose rates had been estimated for organisms other than humans, so these organisms were represented by a single ellipsoid. However, somewhat more complex geometric representations are now being developed and applied.

Owing to the wide range of different types of biota that must be considered, it is not possible to address them all at the species level. Therefore, sets of reference organisms have been defined as specified in Section 5.2 [7.30].

Slightly different terms and definitions are used by various groups [7.31], but the approaches adopted are generally similar. The selection of reference organisms may consider the need to encompass protected species, and different trophic levels and exposure pathways. Reference organisms have tended to be defined at a broad wildlife group level (e.g. soil invertebrate, predatory fish, terrestrial mammal). In some cases, a consideration of individual species has been included [7.32].

The ICRP has established a group of 12 standardized reference organisms (RAPs) to relate exposure to dose and dose to effects within its framework. Information on the ecological characteristics, dosimetry and radiation induced effects relevant to these RAPs is presented in Ref. [7.33]. A revised and extended ICRP dosimetric framework for non-human biota is presented in Ref. [7.34], in which the dose coefficients for external exposure of terrestrial biota have been substantially revised. These dose coefficient values supersede those in annex C of Ref. [7.33].

Dose coefficients for non-human biota exposed to uranium radioisotopes are shown in Table 7.7. These are presented as dose rates (μGy/d) per unit activity concentration in the given media: becquerel per kilogram for internal exposure and external exposure of terrestrial organisms; becquerel per litre for external exposure of aquatic organisms; and becquerel per cubic metre for external exposure of terrestrial organisms immersed in radioactively contaminated air above ground.

The dose coefficients for external exposure of aquatic organisms are computed with the BiotaDC 1.5 tool (a web based complement to Ref. [7.34]) for an infinite water medium. These dose coefficients can also be applied to assess external exposure from radioactivity in bed sediments, provided that the density and chemical composition of the sediments are close to those of water. The dose coefficients for terrestrial organisms exposed on the soil surface are for the infinitely deep uniform source in soil.

Energy emitted by radioactive progeny is added to the dose coefficient according to the progeny's relative average activity per becquerel of parent radionuclide. Averaging is performed for decay chain members over a one year period or the organism's lifetime, whichever is smaller.

The organisms listed in Table 7.7 are those from the terrestrial environment followed by aquatic organisms. Each group is sorted by increasing mass of the organism. The dose coefficients for internal exposure are followed by a set of fractions representing contributions of various radiation types to internal dose: f_0 represents the contribution of fission fragments and alpha recoil nuclei; f_1 alpha particles; f_2 low energy beta and gamma radiation (E < 10 keV); and f_3 other beta and gamma radiations (E > 10 keV). The fractions can be used to modify absorbed dose rates by radiation weighting factors, which account for the different radiobiological effectiveness of various radiation types.

Given the concentrations of ^{238}U, ^{235}U, ^{234}U and their progeny in environmental media, such as soils and water bodies, and/or in various types of organism, external and internal dose rates to a wide variety of types of reference organism can be calculated using the ERICA tool (see Ref. [7.35]).

As discussed in Chapter 6, radionuclide transport models or measurements are used to either estimate or determine concentrations of uranium series radionuclides in environmental media. Measurements may also be used to determine the concentrations of uranium series radionuclides in the tissues and organs of the biota for which dose rate estimates are required. However, in many contexts, only concentrations in environmental media may be available, or concentrations may be available only for a subset of the types of organism for which dose rates need to be estimated. Thus, data are needed to relate concentrations in environmental media to concentrations in organisms utilizing those media. Ratios between tissue and organ concentrations and concentrations in environmental media are typically used for this purpose (as is the case in the ERICA tool). The reference concentration ratios used for radioisotopes of uranium and its progeny for the RAPs adopted by the ICRP are given in Table 7.8 [7.36]. These are based, in part, on information in the primary literature, but also on assessments from applying that information by analogy or extension to the RAPs.

Where additional information is available or where an alternative set of reference organisms is adopted, these concentration ratios should be modified to conform to the requirements of the situation. For example, there may be specific types of organism present and requiring protection that are not adequately represented by one of the RAPs, or measured concentrations in one or a few types of biota may need to be used to inform estimates of concentrations in other types of biota. In these circumstances, a wide variety of factors may need to be considered in making such modifications. These will include similarities and differences in habitat, diet and physiology between the types of organism of interest and the RAPs. The IAEA has provided a compilation of data that may be used for this purpose [7.32]. Relevant data for uranium are given in Table 7.9 and are included in Version 1.2 of the ERICA tool. The dose conversion factors may also require modification, but these are only weak functions of body size and composition, so such modifications are more straightforward to make and justify, and this can be done using the BiotaDC 1.5 tool discussed for the organisms listed in Table 7.7.

Text cont. on p. 300.

TABLE 7.7. DOSE CONVERSION COEFFICIENTS FOR NON-HUMAN BIOTA EXPOSED TO RADIOACTIVE ISOTOPES OF URANIUM

	Internal exposure					External exposure				
Organism	Dose coefficients for internal exposure (μGy·h^{-1}·Bq^{-1}·kg)	Fractions representing contributions of various radiation types to internal dose				Aquatic environment (μGy·h^{-1}·Bq^{-1}·L)	Terrestrial environment			
		f_0	f_1	f_2	f_3		In soil (μGy·h^{-1}·Bq^{-1}·kg)	On ground (μGy·h^{-1}·Bq^{-1}·kg)	Above ground (μGy·h^{-1}·Bq^{-1}·kg)	Submersion in air (μGy·h^{-1}·Bq^{-1}·m^3)
^{227}U (progeny included: ^{223}Th, ^{219}Ra, ^{215}Rn, ^{211}Po)										
Bee	2.2×10^{-2}	0.018	0.976	0.000	0.005	n.a.^{a}	n.a.^{a}	7.1×10^{-5}	7.0×10^{-5}	8.0×10^{-5}
Wild grass (spike)	2.2×10^{-2}	0.018	0.976	0.000	0.005	n.a.^{a}	n.a.^{a}	7.1×10^{-5}	n.a.^{a}	7.8×10^{-5}
Earthworm	2.2×10^{-2}	0.018	0.976	0.000	0.005	n.a.^{a}	1.5×10^{-4}	7.1×10^{-5}	n.a.^{a}	7.7×10^{-5}
Frog	2.2×10^{-2}	0.018	0.976	0.000	0.006	2.0×10^{-4}	1.5×10^{-4}	7.1×10^{-5}	n.a.^{a}	7.6×10^{-5}
Rat	2.2×10^{-2}	0.018	0.975	0.000	0.006	n.a.^{a}	1.4×10^{-4}	6.9×10^{-5}	n.a.^{a}	7.4×10^{-5}
Duck	2.2×10^{-2}	0.018	0.975	0.000	0.007	1.8×10^{-4}	n.a.^{a}	6.7×10^{-5}	6.2×10^{-5}	7.9×10^{-5}
Deer	2.3×10^{-2}	0.018	0.970	0.000	0.011	n.a.^{a}	n.a.^{a}	3.9×10^{-5}	n.a.^{a}	4.1×10^{-5}
Pine tree (trunk)	2.3×10^{-2}	0.018	0.970	0.000	0.011	n.a.^{a}.	n.a.^{a}	5.0×10^{-5}	n.a.^{a}	5.7×10^{-5}
Brown seaweed	2.2×10^{-2}	0.018	0.975	0.000	0.006	1.9×10^{-4}	n.a.^{a}	n.a.^{a}	n.a.^{a}	n.a.^{a}
Crab	2.2×10^{-2}	0.018	0.975	0.000	0.007	1.8×10^{-4}	n.a.^{a}	n.a.^{a}	n.a.^{a}	n.a.^{a}
Trout	2.2×10^{-2}	0.018	0.975	0.000	0.007	1.8×10^{-4}	n.a.^{a}	n.a.^{a}	n.a.^{a}	n.a.^{a}
Flatfish	2.2×10^{-2}	0.018	0.975	0.000	0.006	1.9×10^{-4}	n.a.^{a}	n.a.^{a}	n.a.^{a}	n.a.^{a}
^{228}U (progeny included: ^{224}Th, ^{220}Ra, ^{216}Rn, ^{212}Po)										
Bee	2.2×10^{-2}	0.018	0.981	0.000	0.001	n.a.^{a}	n.a.^{a}	7.1×10^{-6}	7.0×10^{-6}	7.9×10^{-6}
Wild grass (spike)	2.2×10^{-2}	0.018	0.981	0.000	0.001	n.a.^{a}	n.a.^{a}	7.0×10^{-6}	n.a.^{a}	7.6×10^{-6}

Earthworm	2.2×10^{-2}	0.018	0.981	0.000	0.001	n.a.[a]	1.3×10^{-5}	7.1×10^{-6}	n.a.[a]	7.5×10^{-6}
Frog	2.2×10^{-2}	0.018	0.981	0.000	0.001	1.7×10^{-5}	1.3×10^{-5}	7.0×10^{-6}	n.a.[a]	7.3×10^{-6}
Rat	2.2×10^{-2}	0.018	0.981	0.000	0.001	n.a.[a]	1.3×10^{-5}	6.8×10^{-6}	n.a.[a]	7.1×10^{-6}
Duck	2.2×10^{-2}	0.018	0.981	0.000	0.001	1.5×10^{-5}	n.a.[a]	6.7×10^{-6}	6.1×10^{-6}	7.6×10^{-6}
Deer	2.2×10^{-2}	0.018	0.980	0.000	0.001	n.a.[a]	n.a.[a]	3.9×10^{-6}	n.a.[a]	3.9×10^{-6}
Pine tree (trunk)	2.2×10^{-2}	0.018	0.980	0.000	0.001	n.a.[a]	n.a.[a]	5.0×10^{-6}	n.a.[a]	5.4×10^{-6}
Brown seaweed	2.2×10^{-2}	0.018	0.981	0.000	0.001	1.6×10^{-5}	n.a.[a]	n.a.[a]	n.a.[a]	n.a.[a]
Crab	2.2×10^{-2}	0.018	0.981	0.000	0.001	1.6×10^{-5}	n.a.[a]	n.a.[a]	n.a.[a]	n.a.[a]
Trout	2.2×10^{-2}	0.018	0.981	0.000	0.001	1.5×10^{-5}	n.a.[a]	n.a.[a]	n.a.[a]	n.a.[a]
Flatfish	2.2×10^{-2}	0.018	0.981	0.000	0.001	1.6×10^{-5}	n.a.[a]	n.a.[a]	n.a.[a]	n.a.[a]
^{230}U (progeny included: ^{226}Th, ^{222}Ra, ^{218}Rn, ^{214}Po, ^{210}Pb, ^{210}Bi, ^{206}Hg, ^{210}Po, ^{206}Tl)										
Bee	2.0×10^{-2}	0.018	0.981	0.000	0.001	n.a.[a]	n.a.[a]	4.9×10^{-6}	4.8×10^{-6}	5.7×10^{-6}
Wild grass (spike)	2.0×10^{-2}	0.018	0.980	0.000	0.002	n.a.[a]	n.a.[a]	4.9×10^{-6}	n.a.[a]	5.4×10^{-6}
Earthworm	2.0×10^{-2}	0.018	0.980	0.000	0.002	n.a.[a]	8.3×10^{-6}	4.9×10^{-6}	n.a.[a]	5.2×10^{-6}
Frog	2.0×10^{-2}	0.018	0.980	0.000	0.002	1.1×10^{-5}	8.2×10^{-6}	4.8×10^{-6}	n.a.[a]	5.1×10^{-6}
Rat	2.0×10^{-2}	0.018	0.980	0.000	0.002	n.a.[a]	7.8×10^{-6}	4.7×10^{-6}	n.a.[a]	4.8×10^{-6}
Duck	2.0×10^{-2}	0.018	0.980	0.000	0.002	9.4×10^{-6}	n.a.[a]	4.6×10^{-6}	4.2×10^{-6}	5.1×10^{-6}
Deer	2.0×10^{-2}	0.018	0.980	0.000	0.002	n.a.[a]	n.a.[a]	2.7×10^{-6}	n.a.[a]	2.6×10^{-6}
Pine tree (trunk)	2.0×10^{-2}	0.018	0.980	0.000	0.002	n.a.[a]	n.a.[a]	3.4×10^{-6}	n.a.[a]	3.7×10^{-6}
Brown seaweed	2.0×10^{-2}	0.018	0.980	0.000	0.002	1.0×10^{-5}	n.a.[a]	n.a.[a]	n.a.[a]	n.a.[a]
Crab	2.0×10^{-2}	0.018	0.980	0.000	0.002	9.8×10^{-6}	n.a.[a]	n.a.[a]	n.a.[a]	n.a.[a]
Trout	2.0×10^{-2}	0.018	0.980	0.000	0.002	9.7×10^{-6}	n.a.[a]	n.a.[a]	n.a.[a]	n.a.[a]
Flatfish	2.0×10^{-2}	0.018	0.980	0.000	0.002	1.0×10^{-5}	n.a.[a]	n.a.[a]	n.a.[a]	n.a.[a]
Earthworm	5.6×10^{-5}	0.000	0.017	0.272	0.711	n.a.[a]	2.1×10^{-5}	1.1×10^{-5}	n.a.[a]	1.7×10^{-5}

TABLE 7.7. DOSE CONVERSION COEFFICIENTS FOR NON-HUMAN BIOTA EXPOSED TO RADIOACTIVE ISOTOPES OF URANIUM (cont.)

Organism	Internal exposure					External exposure				
	Dose coefficients for internal exposure ($\mu Gy \cdot h^{-1} \cdot Bq^{-1} \cdot kg$)	Fractions representing contributions of various radiation types to internal dose				Aquatic environment ($\mu Gy \cdot h^{-1} \cdot Bq^{-1} \cdot L$)	Terrestrial environment			
		f_0	f_1	f_2	f_3		In soil ($\mu Gy \cdot h^{-1} \cdot Bq^{-1} \cdot kg$)	On ground ($\mu Gy \cdot h^{-1} \cdot Bq^{-1} \cdot kg$)	Above ground ($\mu Gy \cdot h^{-1} \cdot Bq^{-1} \cdot kg$)	Submersion in air ($\mu Gy \cdot h^{-1} \cdot Bq^{-1} \cdot m^3$)
Frog	6.0×10^{-5}	0.000	0.016	0.257	0.727	4.1×10^{-5}	2.1×10^{-5}	1.0×10^{-5}	n.a.[a]	1.6×10^{-5}
Rat	6.4×10^{-5}	0.000	0.015	0.241	0.744	n.a.[a]	1.9×10^{-5}	9.9×10^{-6}	n.a.[a]	1.4×10^{-5}
Duck	6.8×10^{-5}	0.000	0.014	0.228	0.758	3.4×10^{-5}	n.a.[a]	9.6×10^{-6}	8.7×10^{-6}	1.6×10^{-5}
Deer	9.0×10^{-5}	0.000	0.010	0.171	0.818	n.a.[a]	n.a.[a]	5.0×10^{-6}	n.a.[a]	6.6×10^{-6}
Pine tree (trunk)	9.1×10^{-5}	0.000	0.010	0.170	0.820	n.a.[a]	n.a.[a]	6.7×10^{-6}	n.a.[a]	9.9×10^{-6}
Brown seaweed	6.3×10^{-5}	0.000	0.015	0.243	0.742	3.8×10^{-5}	n.a.[a]	n.a.[a]	n.a.[a]	n.a.[a]
Crab	6.6×10^{-5}	0.000	0.014	0.232	0.753	3.5×10^{-5}	n.a.[a]	n.a.[a]	n.a.[a]	n.a.[a]
Trout	6.7×10^{-5}	0.000	0.014	0.230	0.755	3.5×10^{-5}	n.a.[a]	n.a.[a]	n.a.[a]	n.a.[a]
Flatfish	6.5×10^{-5}	0.000	0.014	0.239	0.747	3.7×10^{-5}	n.a.[a]	n.a.[a]	n.a.[a]	n.a.[a]
^{232}U (progeny included: ^{228}Th, ^{224}Ra, ^{220}Rn, ^{216}Po, ^{212}Pb, ^{212}Bi, ^{212}Po, ^{208}Tl)										
Bee	4.0×10^{-3}	0.017	0.976	0.000	0.007	n.a.[a]	n.a.[a]	1.5×10^{-5}	1.4×10^{-5}	1.4×10^{-5}
Wild grass (spike)	6.2×10^{-3}	0.018	0.969	0.000	0.013	n.a.[a]	n.a.[a]	6.0×10^{-5}	n.a.[a]	5.4×10^{-5}
Earthworm	6.2×10^{-3}	0.018	0.969	0.000	0.013	n.a.[a]	1.2×10^{-4}	6.3×10^{-5}	n.a.[a]	5.6×10^{-5}
Frog	6.2×10^{-3}	0.017	0.968	0.000	0.015	1.4×10^{-4}	1.1×10^{-4}	6.7×10^{-5}	n.a.[a]	6.0×10^{-5}
Rat	6.2×10^{-3}	0.017	0.966	0.000	0.016	n.a.[a]	1.1×10^{-4}	6.9×10^{-5}	n.a.[a]	6.1×10^{-5}
Duck	6.2×10^{-3}	0.017	0.965	0.000	0.017	1.2×10^{-4}	n.a.[a]	6.9×10^{-5}	6.4×10^{-5}	6.5×10^{-5}
Deer	6.2×10^{-3}	0.017	0.956	0.000	0.026	n.a.[a]	n.a.[a]	5.0×10^{-5}	n.a.[a]	4.4×10^{-5}

Pine tree (trunk)	6.2×10^{-3}	0.017	0.958	0.000	0.024	n.a.[a]	n.a.[a]	5.8×10^{-5}	n.a.[a]	5.5×10^{-5}
Brown seaweed	6.2×10^{-3}	0.017	0.967	0.000	0.016	1.3×10^{-4}	n.a.[a]	n.a.[a]	n.a.[a]	n.a.[a]
Crab	6.2×10^{-3}	0.017	0.966	0.000	0.017	1.2×10^{-4}	n.a.[a]	n.a.[a]	n.a.[a]	n.a.[a]
Trout	6.2×10^{-3}	0.017	0.965	0.000	0.017	1.2×10^{-4}	n.a.[a]	n.a.[a]	n.a.[a]	n.a.[a]
Flatfish	6.2×10^{-3}	0.017	0.966	0.000	0.016	1.3×10^{-4}	n.a.[a]	n.a.[a]	n.a.[a]	n.a.[a]
^{233}U (progeny included: ^{229}Th, ^{225}Ra, ^{225}Ac, ^{221}Fr, ^{217}At, ^{213}Bi, ^{213}Po, ^{209}Tl, ^{209}Pb)										
Bee	2.8×10^{-3}	0.017	0.982	0.000	0.001	n.a.[a]	n.a.[a]	9.2×10^{-8}	7.8×10^{-8}	2.8×10^{-7}
Wild grass (spike)	2.8×10^{-3}	0.017	0.982	0.000	0.001	n.a.[a]	n.a.[a]	8.2×10^{-8}	n.a.[a]	2.2×10^{-7}
Earthworm	2.8×10^{-3}	0.017	0.982	0.000	0.001	n.a.[a]	1.7×10^{-7}	8.2×10^{-8}	n.a.[a]	1.9×10^{-7}
Frog	2.8×10^{-3}	0.017	0.982	0.000	0.001	3.0×10^{-7}	1.7×10^{-7}	7.2×10^{-8}	n.a.[a]	1.4×10^{-7}
Rat	2.8×10^{-3}	0.017	0.981	0.000	0.001	n.a.[a]	1.5×10^{-7}	6.1×10^{-8}	n.a.[a]	1.0×10^{-7}
Duck	2.8×10^{-3}	0.017	0.981	0.000	0.001	1.7×10^{-7}	n.a.[a]	5.6×10^{-8}	4.6×10^{-8}	1.1×10^{-7}
Deer	2.8×10^{-3}	0.017	0.981	0.000	0.001	n.a.[a]	n.a.[a]	2.8×10^{-8}	n.a.[a]	3.5×10^{-8}
Pine tree (trunk)	2.8×10^{-3}	0.017	0.981	0.000	0.001	n.a.[a]	n.a.[a]	3.6×10^{-8}	n.a.[a]	5.5×10^{-8}
Brown seaweed	2.8×10^{-3}	0.017	0.982	0.000	0.001	2.3×10^{-7}	n.a.[a]	n.a.[a]	n.a.[a]	n.a.[a]
Crab	2.8×10^{-3}	0.017	0.981	0.000	0.001	1.9×10^{-7}	n.a.[a]	n.a.[a]	n.a.[a]	n.a.[a]
Trout	2.8×10^{-3}	0.017	0.981	0.000	0.001	1.8×10^{-7}	n.a.[a]	n.a.[a]	n.a.[a]	n.a.[a]
Flatfish	2.8×10^{-3}	0.017	0.981	0.000	0.001	2.1×10^{-7}	n.a.[a]	n.a.[a]	n.a.[a]	n.a.[a]
^{234}U (progeny included: ^{230}Th, ^{226}Ra, ^{222}Rn, ^{218}Po, ^{214}Pb, ^{218}At, ^{214}Bi, ^{218}Rn, ^{210}Tl, ^{214}Po)										
Bee	2.8×10^{-3}	0.017	0.980	0.000	0.003	n.a.[a]	n.a.[a]	9.7×10^{-8}	7.4×10^{-8}	4.4×10^{-7}
Wild grass (spike)	2.8×10^{-3}	0.017	0.980	0.000	0.003	n.a.[a]	n.a.[a]	7.4×10^{-8}	n.a.[a]	3.2×10^{-7}
Earthworm	2.8×10^{-3}	0.017	0.980	0.000	0.003	n.a.[a]	1.7×10^{-7}	7.5×10^{-8}	n.a.[a]	2.6×10^{-7}
Frog	2.8×10^{-3}	0.017	0.980	0.000	0.003	3.7×10^{-7}	1.7×10^{-7}	5.6×10^{-8}	n.a.[a]	1.9×10^{-7}

TABLE 7.7. DOSE CONVERSION COEFFICIENTS FOR NON-HUMAN BIOTA EXPOSED TO RADIOACTIVE ISOTOPES OF URANIUM (cont.)

Organism	Internal exposure					External exposure				
	Dose coefficients for internal exposure (μGy·h^{-1}·Bq^{-1}·kg)	Fractions representing contributions of various radiation types to internal dose				Aquatic environment (μGy·h^{-1}·Bq^{-1}·L)	Terrestrial environment			
		f_0	f_1	f_2	f_3		In soil (μGy·h^{-1}·Bq^{-1}·kg)	On ground (μGy·h^{-1}·Bq^{-1}·kg)	Above ground (μGy·h^{-1}·Bq^{-1}·kg)	Submersion in air (μGy·h^{-1}·Bq^{-1}·m^3)
Trout	2.8×10^{-3}	0.017	0.980	0.000	0.003	1.6×10^{-7}	n.a.[a]	n.a.[a]	n.a.[a]	n.a.[a]
Flatfish	2.8×10^{-3}	0.017	0.980	0.000	0.003	2.0×10^{-7}	n.a.[a]	n.a.[a]	n.a.[a]	n.a.[a]
^{235}U (progeny included: ^{231}Th, ^{231}Pa, ^{227}Ac, ^{227}Th, ^{223}Fr, ^{223}Ra, ^{219}At, ^{219}Rn, ^{215}Bi, ^{215}Po, ^{211}Pb, ^{211}Bi, ^{207}Tl, ^{211}Po)										
Bee	2.7×10^{-3}	0.016	0.937	0.006	0.041	n.a.[a]	n.a.[a]	3.7×10^{-5}	3.6×10^{-5}	4.4×10^{-5}
Wild grass (spike)	2.7×10^{-3}	0.016	0.936	0.006	0.042	n.a.[a]	n.a.[a]	3.6×10^{-5}	n.a.[a]	4.3×10^{-5}
Earthworm	2.7×10^{-3}	0.016	0.935	0.006	0.042	n.a.[a]	7.1×10^{-5}	3.6×10^{-5}	n.a.[a]	4.2×10^{-5}
Frog	2.7×10^{-3}	0.016	0.934	0.006	0.044	1.0×10^{-4}	7.0×10^{-5}	3.6×10^{-5}	n.a.[a]	4.1×10^{-5}
Rat	2.7×10^{-3}	0.016	0.931	0.006	0.046	n.a.[a]	6.6×10^{-5}	3.5×10^{-5}	n.a.[a]	3.9×10^{-5}
Duck	2.7×10^{-3}	0.016	0.929	0.006	0.049	8.6×10^{-5}	n.a.[a]	3.4×10^{-5}	3.1×10^{-5}	4.1×10^{-5}
Deer	2.8×10^{-3}	0.016	0.911	0.006	0.067	n.a.[a]	n.a.[a]	1.9×10^{-5}	n.a.[a]	2.0×10^{-5}
Pine tree (trunk)	2.8×10^{-3}	0.016	0.911	0.006	0.068	n.a.[a]	n.a.[a]	2.5×10^{-5}	n.a.[a]	2.9×10^{-5}
Brown seaweed	2.7×10^{-3}	0.016	0.932	0.006	0.046	9.4×10^{-5}	n.a.[a]	n.a.[a]	n.a.[a]	n.a.[a]
Crab	2.7×10^{-3}	0.016	0.930	0.006	0.048	8.8×10^{-5}	n.a.[a]	n.a.[a]	n.a.[a]	n.a.[a]
Trout	2.7×10^{-3}	0.016	0.929	0.006	0.048	8.7×10^{-5}	n.a.[a]	n.a.[a]	n.a.[a]	n.a.[a]
Flatfish	2.7×10^{-3}	0.016	0.931	0.006	0.047	9.2×10^{-5}	n.a.[a]	n.a.[a]	n.a.[a]	n.a.[a]
^{235m}U (progeny included: ^{235}U, ^{231}Th, ^{231}Pa, ^{227}Ac, ^{227}Th, ^{223}Fr, ^{223}Ra, ^{219}At, ^{219}Rn, ^{215}Bi, ^{215}Po, ^{211}Pb, ^{211}Bi, ^{207}Tl, ^{211}Po)										
Bee	4.4×10^{-8}	0.000	0.000	1.000	0.000	n.a.[a]	n.a.[a]	9.9×10^{-15}	9.6×10^{-15}	1.2×10^{-14}

Wild grass (spike)	4.4×10^{-8}	0.000	0.000	1.000	0.000	n.a.[a]	n.a.[a]	3.6×10^{-14}	n.a.[a]	4.2×10^{-14}
Earthworm	4.4×10^{-8}	0.000	0.000	1.000	0.000	n.a.[a]	6.9×10^{-14}	3.6×10^{-14}	n.a.[a]	4.1×10^{-14}
Frog	4.4×10^{-8}	0.000	0.000	1.000	0.000	9.8×10^{-14}	6.8×10^{-14}	3.5×10^{-14}	n.a.[a]	4.0×10^{-14}
Rat	4.4×10^{-8}	0.000	0.000	1.000	0.000	n.a.[a]	6.5×10^{-14}	3.4×10^{-14}	n.a.[a]	3.8×10^{-14}
Duck	4.4×10^{-8}	0.000	0.000	1.000	0.000	8.4×10^{-14}	n.a.[a]	3.3×10^{-14}	3.1×10^{-14}	4.1×10^{-14}
Deer	4.4×10^{-8}	0.000	0.000	1.000	0.000	n.a.[a]	n.a.[a]	1.9×10^{-14}	n.a.[a]	2.0×10^{-14}
Pine tree (trunk)	4.4×10^{-8}	0.000	0.000	1.000	0.000	n.a.[a]	n.a.[a]	2.5×10^{-14}	n.a.[a]	2.8×10^{-14}
Brown seaweed	4.4×10^{-8}	0.000	0.000	1.000	0.000	9.2×10^{-14}	n.a.[a]	n.a.[a]	n.a.[a]	n.a.[a]
Crab	4.4×10^{-8}	0.000	0.000	1.000	0.000	8.7×10^{-14}	n.a.[a]	n.a.[a]	n.a.[a]	n.a.[a]
Trout	4.4×10^{-8}	0.000	0.000	1.000	0.000	8.6×10^{-14}	n.a.[a]	n.a.[a]	n.a.[a]	n.a.[a]
Flatfish	4.4×10^{-8}	0.000	0.000	1.000	0.000	9.1×10^{-14}	n.a.[a]	n.a.[a]	n.a.[a]	n.a.[a]
^{236}U (progeny included: none)										
Bee	2.6×10^{-3}	0.017	0.980	0.000	0.002	n.a.[a]	n.a.[a]	8.0×10^{-8}	6.0×10^{-8}	3.9×10^{-7}
Wild grass (spike)	2.6×10^{-3}	0.017	0.980	0.000	0.002	n.a.[a]	n.a.[a]	6.0×10^{-8}	n.a.[a]	2.8×10^{-7}
Earthworm	2.6×10^{-3}	0.017	0.980	0.000	0.002	n.a.[a]	1.4×10^{-7}	6.0×10^{-8}	n.a.[a]	2.3×10^{-7}
Frog	2.6×10^{-3}	0.017	0.980	0.000	0.002	3.0×10^{-7}	1.4×10^{-7}	4.3×10^{-8}	n.a.[a]	1.6×10^{-7}
Rat	2.6×10^{-3}	0.017	0.980	0.000	0.002	n.a.[a]	1.2×10^{-7}	2.6×10^{-8}	n.a.[a]	8.9×10^{-8}
Duck	2.6×10^{-3}	0.017	0.980	0.000	0.002	1.2×10^{-7}	n.a.[a]	1.9×10^{-8}	9.7×10^{-9}	9.7×10^{-8}
Deer	2.6×10^{-3}	0.017	0.980	0.000	0.002	n.a.[a]	n.a.[a]	5.2×10^{-9}	n.a.[a]	1.5×10^{-8}
Pine tree (trunk)	2.6×10^{-3}	0.017	0.980	0.000	0.002	n.a.[a]	n.a.[a]	6.4×10^{-9}	n.a.[a]	3.0×10^{-8}
Flatfish	2.6×10^{-3}	0.017	0.980	0.000	0.002	1.6×10^{-7}	n.a.[a]	n.a.[a]	n.a.[a]	n.a.[a]
^{237}U (progeny included: ^{237}Np, ^{233}Pa, ^{233}U)										
Bee	1.2×10^{-4}	0.000	0.000	0.083	0.917	n.a.[a]	n.a.[a]	2.8×10^{-5}	2.7×10^{-5}	3.5×10^{-5}

TABLE 7.7. DOSE CONVERSION COEFFICIENTS FOR NON-HUMAN BIOTA EXPOSED TO RADIOACTIVE ISOTOPES OF URANIUM (cont.)

Organism	Internal exposure					External exposure				
	Dose coefficients for internal exposure ($\mu Gy \cdot h^{-1} \cdot Bq^{-1} \cdot kg$)	Fractions representing contributions of various radiation types to internal dose				Aquatic environment ($\mu Gy \cdot h^{-1} \cdot Bq^{-1} \cdot L$)	Terrestrial environment			
		f_0	f_1	f_2	f_3		In soil ($\mu Gy \cdot h^{-1} \cdot Bq^{-1} \cdot kg$)	On ground ($\mu Gy \cdot h^{-1} \cdot Bq^{-1} \cdot kg$)	Above ground ($\mu Gy \cdot h^{-1} \cdot Bq^{-1} \cdot kg$)	Submersion in air ($\mu Gy \cdot h^{-1} \cdot Bq^{-1} \cdot m^3$)
Rat	1.3×10^{-4}	0.000	0.000	0.076	0.923	n.a.[a]	4.4×10^{-5}	2.6×10^{-5}	n.a.[a]	3.1×10^{-5}
Duck	1.3×10^{-4}	0.000	0.000	0.073	0.927	6.4×10^{-5}	n.a.[a]	2.6×10^{-5}	2.3×10^{-5}	3.3×10^{-5}
Deer	1.7×10^{-4}	0.000	0.000	0.056	0.944	n.a.[a]	n.a.[a]	1.4×10^{-5}	n.a.[a]	1.6×10^{-5}
Pine tree (trunk)	1.8×10^{-4}	0.000	0.000	0.056	0.944	n.a.[a]	n.a.[a]	1.9×10^{-5}	n.a.[a]	2.3×10^{-5}
Brown seaweed	1.3×10^{-4}	0.000	0.000	0.077	0.923	7.1×10^{-5}	n.a.[a]	n.a.[a]	n.a.[a]	n.a.[a]
Crab	1.3×10^{-4}	0.000	0.000	0.074	0.926	6.6×10^{-5}	n.a.[a]	n.a.[a]	n.a.[a]	n.a.[a]
Trout	1.3×10^{-4}	0.000	0.000	0.074	0.926	6.6×10^{-5}	n.a.[a]	n.a.[a]	n.a.[a]	n.a.[a]
Flatfish	1.3×10^{-4}	0.000	0.000	0.076	0.924	6.9×10^{-5}	n.a.[a]	n.a.[a]	n.a.[a]	n.a.[a]
^{238}U (progeny included: ^{234}Th, ^{234m}Pa, ^{234}Pa, ^{234}U)										
Bee	2.7×10^{-3}	0.015	0.897	0.001	0.087	n.a.[a]	n.a.[a]	4.5×10^{-6}	4.4×10^{-6}	5.1×10^{-6}
Wild grass (spike)	2.8×10^{-3}	0.015	0.855	0.001	0.129	n.a.[a]	n.a.[a]	6.1×10^{-6}	n.a.[a]	6.6×10^{-6}
Earthworm	2.8×10^{-3}	0.015	0.853	0.001	0.132	n.a.[a]	1.1×10^{-5}	6.2×10^{-6}	n.a.[a]	6.5×10^{-6}
Frog	2.9×10^{-3}	0.014	0.839	0.001	0.146	5.7×10^{-5}	1.1×10^{-5}	6.3×10^{-6}	n.a.[a]	6.4×10^{-6}
Rat	2.9×10^{-3}	0.014	0.832	0.001	0.153	n.a.[a]	1.0×10^{-5}	6.2×10^{-6}	n.a.[a]	6.2×10^{-6}
Duck	2.9×10^{-3}	0.014	0.830	0.001	0.155	2.6×10^{-5}	n.a.[a]	6.1×10^{-6}	5.6×10^{-6}	6.6×10^{-6}
Deer	2.9×10^{-3}	0.014	0.825	0.001	0.161	n.a.[a]	n.a.[a]	3.8×10^{-6}	n.a.[a]	3.7×10^{-6}
Pine tree (trunk)	2.9×10^{-3}	0.014	0.825	0.001	0.161	n.a.[a]	n.a.[a]	4.7×10^{-6}	n.a.[a]	4.9×10^{-6}

Brown seaweed	2.9×10^{-3}	0.014	0.836	0.001	0.149	4.8×10^{-5}	n.a.[a]	n.a.[a]	n.a.[a]	n.a.[a]
Crab	2.9×10^{-3}	0.014	0.830	0.001	0.155	2.9×10^{-5}	n.a.[a]	n.a.[a]	n.a.[a]	n.a.[a]
Trout	2.9×10^{-3}	0.014	0.830	0.001	0.155	2.8×10^{-5}	n.a.[a]	n.a.[a]	n.a.[a]	n.a.[a]
Flatfish	2.9×10^{-3}	0.014	0.835	0.001	0.149	4.6×10^{-5}	n.a.[a]	n.a.[a]	n.a.[a]	n.a.[a]
^{239}U (progeny included: ^{239}Np, ^{239}Pu, ^{235m}U)										
Bee	3.5×10^{-4}	0.000	0.000	0.030	0.970	n.a.[a]	n.a.[a]	4.5×10^{-5}	4.4×10^{-5}	5.5×10^{-5}
Wild grass (spike)	3.7×10^{-4}	0.000	0.000	0.029	0.971	n.a.[a]	n.a.[a]	4.5×10^{-5}	n.a.[a]	5.3×10^{-5}
Earthworm	3.7×10^{-4}	0.000	0.000	0.029	0.971	n.a.[a]	8.3×10^{-5}	4.5×10^{-5}	n.a.[a]	5.2×10^{-5}
Frog	3.9×10^{-4}	0.000	0.000	0.027	0.973	1.4×10^{-4}	8.2×10^{-5}	4.5×10^{-5}	n.a.[a]	5.1×10^{-5}
Rat	4.0×10^{-4}	0.000	0.000	0.026	0.973	n.a.[a]	7.8×10^{-5}	4.4×10^{-5}	n.a.[a]	5.0×10^{-5}
Duck	4.1×10^{-4}	0.000	0.000	0.026	0.974	1.1×10^{-4}	n.a.[a]	4.3×10^{-5}	3.9×10^{-5}	5.3×10^{-5}
Deer	4.8×10^{-4}	0.000	0.000	0.022	0.978	n.a.[a]	n.a.[a]	2.4×10^{-5}	n.a.[a]	2.6×10^{-5}
Pine tree (trunk)	4.9×10^{-4}	0.000	0.000	0.022	0.978	n.a.[a]	n.a.[a]	3.1×10^{-5}	n.a.[a]	3.7×10^{-5}
Brown seaweed	4.0×10^{-4}	0.000	0.000	0.027	0.973	1.3×10^{-4}	n.a.[a]	n.a.[a]	n.a.[a]	n.a.[a]
^{240}U (progeny included: ^{240m}Np, ^{240}Np, ^{240}Pu, ^{236}U)										
Crab	4.1×10^{-4}	0.000	0.000	0.026	0.974	1.2×10^{-4}	n.a.[a]	n.a.[a]	n.a.[a]	n.a.[a]
Bee	3.6×10^{-4}	0.000	0.000	0.016	0.984	n.a.[a]	n.a.[a]	8.6×10^{-5}	8.4×10^{-5}	8.1×10^{-5}
Wild grass (spike)	4.0×10^{-4}	0.000	0.001	0.014	0.985	n.a.[a]	n.a.[a]	8.7×10^{-5}	n.a.[a]	8.1×10^{-5}
Earthworm	4.0×10^{-4}	0.000	0.001	0.014	0.985	n.a.[a]	1.7×10^{-4}	8.8×10^{-5}	n.a.[a]	8.0×10^{-5}
Frog	4.4×10^{-4}	0.000	0.001	0.013	0.987	2.1×10^{-4}	1.7×10^{-4}	8.8×10^{-5}	n.a.[a]	8.0×10^{-5}
Rat	4.7×10^{-4}	0.000	0.001	0.012	0.987	n.a.[a]	1.6×10^{-4}	8.7×10^{-5}	n.a.[a]	7.9×10^{-5}
Duck	4.9×10^{-4}	0.000	0.001	0.012	0.988	1.7×10^{-4}	n.a.[a]	8.5×10^{-5}	7.8×10^{-5}	8.3×10^{-5}

TABLE 7.7. DOSE CONVERSION COEFFICIENTS FOR NON-HUMAN BIOTA EXPOSED TO RADIOACTIVE ISOTOPES OF URANIUM (cont.)

Organism	Internal exposure					External exposure				
	Dose coefficients for internal exposure (μGy·h^{-1}·Bq^{-1}·kg)	Fractions representing contributions of various radiation types to internal dose				Aquatic environment (μGy·h^{-1}·Bq^{-1}·L)	Terrestrial environment			
		f_0	f_1	f_2	f_3		In soil (μGy·h^{-1}·Bq^{-1}·kg)	On ground (μGy·h^{-1}·Bq^{-1}·kg)	Above ground (μGy·h^{-1}·Bq^{-1}·kg)	Submersion in air (μGy·h^{-1}·Bq^{-1}·m^3)
Deer	5.8×10^{-4}	0.000	0.001	0.010	0.990	n.a.[a]	n.a.[a]	5.5×10^{-5}	n.a.[a]	4.9×10^{-5}
Crab	4.8×10^{-4}	0.000	0.001	0.012	0.988	1.8×10^{-4}	n.a.[a]	n.a.[a]	n.a.[a]	n.a.[a]
Trout	4.8×10^{-4}	0.000	0.001	0.012	0.988	1.7×10^{-4}	n.a.[a]	n.a.[a]	n.a.[a]	n.a.[a]
Flatfish	4.6×10^{-4}	0.000	0.001	0.012	0.987	1.9×10^{-4}	n.a.[a]	n.a.[a]	n.a.[a]	n.a.[a]
^{242}U (progeny included: ^{242}Np, ^{242}Pu)										
Bee	5.0×10^{-4}	0.000	0.000	0.001	0.999	n.a.	n.a.	7.1×10^{-5}	7.0×10^{-5}	6.6×10^{-5}
Wild grass (spike)	5.8×10^{-4}	0.000	0.000	0.001	0.999	n.a.[a]	n.a.[a]	7.5×10^{-5}	n.a.[a]	6.8×10^{-5}
Earthworm	6.0×10^{-4}	0.000	0.000	0.001	0.999	n.a.[a]	1.6×10^{-4}	7.7×10^{-5}	n.a.[a]	6.9×10^{-5}
Frog	6.8×10^{-4}	0.000	0.000	0.001	0.999	2.4×10^{-4}	1.5×10^{-4}	7.9×10^{-5}	n.a.[a]	7.1×10^{-5}
Rat	7.2×10^{-4}	0.000	0.000	0.001	0.999	n.a.[a]	1.5×10^{-4}	7.9×10^{-5}	n.a.[a]	7.1×10^{-5}
Duck	7.4×10^{-4}	0.000	0.000	0.001	0.999	1.8×10^{-4}	n.a.[a]	7.8×10^{-5}	7.2×10^{-5}	7.5×10^{-5}
Deer	8.4×10^{-4}	0.000	0.000	0.001	0.999	n.a.[a]	n.a.[a]	5.3×10^{-5}	n.a.[a]	4.7×10^{-5}
Pine tree (trunk)	8.3×10^{-4}	0.000	0.000	0.001	0.999	n.a.[a]	n.a.[a]	6.4×10^{-5}	n.a.[a]	6.0×10^{-5}
Brown seaweed	7.0×10^{-4}	0.000	0.000	0.001	0.999	2.2×10^{-4}	n.a.[a]	n.a.[a]	n.a.[a]	n.a.[a]
Crab	7.4×10^{-4}	0.000	0.000	0.001	0.999	1.8×10^{-4}	n.a.[a]	n.a.[a]	n.a.[a]	n.a.[a]
Trout	7.4×10^{-4}	0.000	0.000	0.001	0.999	1.8×10^{-4}	n.a.[a]	n.a.[a]	n.a.[a]	n.a.[a]
Flatfish	7.1×10^{-4}	0.000	0.000	0.001	0.999	2.2×10^{-4}	n.a.[a]	n.a.[a]	n.a.[a]	n.a.[a]

[a] n.a.: not applicable.

TABLE 7.8. STANDARD CONCENTRATION RATIOS FOR URANIUM IN REFERENCE ANIMALS AND PLANTS ADOPTED BY THE INTERNATIONAL COMMISSION ON RADIOLOGICAL PROTECTION [7.36]

Terrestrial organism	Concentration ratio (Bq/kg FW to Bq/kg DW soil)	Aquatic organism (environment)	Concentration ratio (Bq/kg FW to Bq/kg water)
Bee	1.7×10^{-2} [a]	Trout (freshwater)	8.5
Earthworm	8.8×10^{-3}	Frog (freshwater)	9.1 [b]
Wild grass	4.3×10^{-2}	Duck (freshwater)	13 [c]
Pine tree	9.9×10^{-4}		
Rat	6.5×10^{-4}	Flatfish (marine)	4.0 [a]
Deer	3.7×10^{-3} [a]	Crab (marine)	6.2 [d]
Duck	4.9×10^{-4} [a]	Brown seaweed (marine)	29
Frog	6.7×10^{-1} [b]		

Note: FW: fresh weight; DW: dry weight.

[a] Value for the generic wildlife group of the the reference animal or plant.

[b] Value derived from a related reference animal or a related generic wildlife group.

[c] Value obtained using an allometric relationship or other modelling approach.

[d] Value for the generic wildlife group within which the reference animal or plant fits for the estuarine environment.

TABLE 7.9. CONCENTRATION RATIOS FOR URANIUM IN WILDLIFE IN VARIOUS TYPES OF ECOSYSTEM [7.32]

Wildlife group: Terrestrial	Concentration ratio (Bq/kg, fresh weight whole organism:Bq/kg, dry weight soil)						
	Arithmetic mean	Standard deviation	Geometric mean	Geometric standard deviation	Minimum	Maximum	*N*[a]
Annelids	8.8×10^{-3}	n.a.[b]	n.a.[b]	n.a.[b]	n.a.[b]	n.a.[b]	1
Arthropods	1.8×10^{-2}	5.0×10^{-3}	1.7×10^{-2}	1.3	1.0×10^{-2}	2.0×10^{-2}	4
Birds: herbivorous	5.0×10^{-4}	1.1×10^{-4}	4.9×10^{-4}	1.3	4.1×10^{-4}	6.8×10^{-4}	20
Grasses and herbs	1.4×10^{-1}	4.4×10^{-1}	4.5×10^{-2}	4.6	7.7×10^{-5}	5.5	439
Grasses	1.3×10^{-1}	4.0×10^{-1}	3.7×10^{-2}	4.8	7.7×10^{-5}	5.5	280
Herbs	2.1×10^{-1}	5.5×10^{-1}	7.6×10^{-2}	4.2	2.2×10^{-3}	2.8	64
Lichens and bryophytes	2.5	4.4	1.3	3.2	2.0×10^{-2}	2.9×10^{1}	237
Mammals	5.8×10^{-3}	6.8×10^{-3}	3.7×10^{-3}	2.5	1.5×10^{-5}	2.1×10^{-2}	22
Mammals: marsupials	6.2×10^{-3}	7.9×10^{-3}	3.9×10^{-3}	2.6	8.0×10^{-4}	2.1×10^{-2}	12

TABLE 7.9. CONCENTRATION RATIOS FOR URANIUM IN WILDLIFE IN VARIOUS TYPES OF ECOSYSTEM [7.32] (cont.)

Reptiles	1.5	3.1	6.7×10^{-1}	3.6	1.3×10^{-4}	2.5	21
Shrubs	2.3×10^{-1}	6.4×10^{-1}	8.1×10^{-2}	4.3	1.4×10^{-5}	5.9	970
Trees	6.8×10^{-3}	1.4×10^{-2}	2.9×10^{-3}	3.7	1.4×10^{-5}	3.2×10^{-2}	521

Wildlife group: Freshwater	Concentration ratio (Bq/kg, fresh weight whole organism:Bq/L water)						
	Arithmetic mean	Standard deviation	Geometric mean	Geometric standard deviation	Minimum	Maximum	*N*[a]
Crustaceans	2.0×10^{2}	3.1×10^{2}	1.1×10^{2}	3.1	—[c]	—[c]	5
Fish	3.1×10^{1}	1.0×10^{2}	9.1	4.8	5.0×10^{-2}	7.6×10^{2}	1294
Fish: benthivorous	7.5×10^{1}	2.1×10^{2}	2.6×10^{1}	4.3	6.0×10^{-1}	7.6×10^{2}	99
Fish: piscivorous	2.2×10^{1}	4.0×10^{1}	1.1×10^{1}	3.4	5.1×10^{-1}	1.7×10^{2}	84
Molluscs: bivalves	5.6×10^{2}	1.3×10^{2}	5.4×10^{2}	1.3	—[c]	—[c]	3
Phytoplankton	7.1×10^{1}	4.7×10^{1}	5.9×10^{1}	1.8	4.0×10^{1}	1.8×10^{2}	40

TABLE 7.9. CONCENTRATION RATIOS FOR URANIUM IN WILDLIFE IN VARIOUS TYPES OF ECOSYSTEM [7.32] (cont.)

Reptiles	1.2×10^2	9.6×10^1	9.0×10^1	2.1	4.5×10^1	1.9×10^2	8
Vascular plants	3.7×10^2	9.9×10^2	1.3×10^2	4.2	2.9×10^1	2.7×10^3	386

Wildlife group: Marine	Concentration ratio (Bq/kg, fresh weight whole organism:Bq/L water)						
	Arithmetic mean	Standard deviation	Geometric mean	Geometric standard deviation	Minimum	Maximum	*N*[a]
Fish	8.8	6.1	7.3	1.9	2.0	1.8×10^1	9
Macroalgae	8.3×10^1	9.9×10^1	5.4×10^1	2.6	2.1×10^1	5.1×10^2	47
Molluscs	3.2×10^1	3.0×10^1	2.4×10^1	2.2×10^0	4.0	9.7×10^1	22
Molluscs: bivalves	3.5×10^1	3.5×10^1	2.5×10^1	2.3	4.0	9.7×10^1	13
Phytoplankton	2.2×10^2	2.3×10^2	1.5×10^2	2.4	1.0×10^1	6.0×10^2	10
Sea anemones and true corals	9.9×10^2	4.4×10^2	9.1×10^2	1.5	4.2×10^2	1.8×10^3	38
Vascular plants	2.4×10^2	n.a.[b]	n.a.[b]	n.a.[b]	1.7×10^2	3.0×10^2	2

TABLE 7.9. CONCENTRATION RATIOS FOR URANIUM IN WILDLIFE IN VARIOUS TYPES OF ECOSYSTEM [7.32] (cont.)

Zooplankton	3.7	4.8	2.3	2.7	1.7×10^{-1}	5.5	3

Wildlife group: Brackish	Concentration ratio (Bq/kg, fresh weight whole organism:Bq/L water)						
	Arithmetic mean	Standard deviation	Geometric mean	Geometric standard deviation	Minimum	Maximum	N[a]
Crustaceans	3.5	2.3	3.0	1.8	8.3×10^{-1}	7.6	9
Fish	4.6×10^{-1}	1.7×10^{-1}	4.4×10^{-1}	1.4	4.0×10^{-1}	5.6×10^{-1}	9
Macroalgae	3.9×10^{1}	7.8×10^{1}	1.7×10^{1}	3.6	8.6×10^{-1}	3.3×10^{2}	49
Molluscs	2.6×10^{1}	2.5×10^{1}	1.9×10^{1}	2.2	3.7	9.1×10^{1}	27
Vascular plants	1.4×10^{2}	4.6×10^{1}	1.3×10^{2}	1.4	—[c]	—[c]	3

[a] N: Number of data entries used for analysis.

[b] n.a.: not applicable for $N < 3$.

[c] —: data not available.

Irrespective of their basis of derivation, the principal outputs from an assessment of radiological impacts on non-human biota are absorbed dose rates to RAPs or to other types of reference or specific organism. The significance of such dose rates can be evaluated by comparing them with quantitative criteria such as the derived consideration reference levels recommended by the ICRP [7.37] and defined as:

> "A band of dose rate within which there is likely to be some chance of deleterious effects of ionising radiation occurring to individuals of that type of reference animal or plant (derived from a knowledge of defined expected biological effects for that type of organism) that, when considered together with other relevant information, can be used as a point of reference to optimise the level of effort expended on environmental protection, dependent upon the overall management objectives and the relevant exposure situation."

The values themselves are very similar to those that have been derived in other reviews and analyses of radiation effects data from a wider range of biota and grouped in various ways [7.38]. The derived consideration reference level values adopted by the ICRP are illustrated in Fig. 7.2.

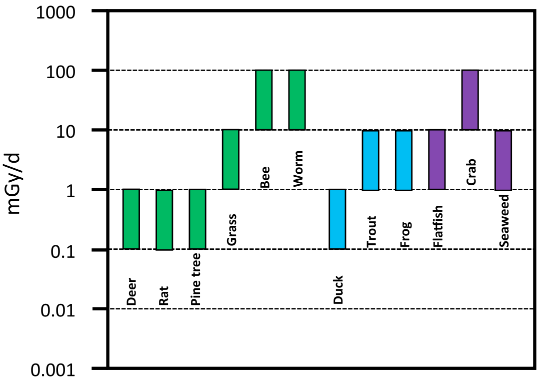

FIG. 7.2. Derived consideration reference levels grouped by terrestrial (green), freshwater (light blue) or marine (dark blue) environment. Figure reproduced from Ref. [7.37] with permission from the International Commission on Radiological Protection.

7.6. MODELS AND DATA FOR ESTIMATING CHEMICAL TOXICITY TO BIOTA

In assessing whether specific types of biota or aspects of the environment, such as habitat characteristics or biodiversity, would be adversely affected by the presence of enhanced concentrations of uranium, reference is appropriately made to EQSs. For example, the Canadian Water Quality Guidelines for the Protection of Aquatic Life [7.39] specify total recoverable freshwater guideline concentrations of 15 μg/L for long term exposure and 33 μg/L for short term exposure. These values were based on distributions of LC_{50} values (lethal concentration for 50% mortality) for ten or more species, taking the 5th percentile of the fitted distribution of these values. Similarly, soil quality guidelines for soil contact (SQG_{SC} values) are available for Canada [7.40]. These are based on toxicological data for vascular plants and soil invertebrates. A total of 7 studies were selected as acceptable for use, including 15 plant species and 4 invertebrate species. From these studies, a total of 82 acceptable no observed adverse effect concentrations, lowest observed adverse effects concentrations and effective concentration (EC_{20}) values were used. EC_{20} is the effective concentration at which 20% of the exposed population is affected. The 25th percentile of the data distribution was calculated to be 500 mg/kg and the 50th percentile was 2000 mg/kg. These then became the environmental soil quality guidelines (SQG_E values) for residential/parkland land use and for commercial and industrial land uses, respectively.

Similarly, the Netherlands has recently promulgated EQS values for uranium in waters [7.41]. These values are based on a detailed review and interpretation of the primary literature and considered chemical toxicity only. The annual average concentration was established to protect against the effects of long term exposure (includes direct ecotoxicity, secondary poisoning of predatory animals, and human exposure through fish and shellfish consumption). The maximum acceptable concentration was derived to protect against short term concentration peak effects and is based only on direct ecotoxicity. Values of 0.5 and 8.9 μg/L were adopted for the annual average concentration and the maximum acceptable concentration, respectively.

In Australia, a site specific trigger value for uranium was derived using the framework described in the Australian Water Quality Guidelines and based on ecotoxicological tests using five local test species from four trophic levels [7.42, 7.43]. With the Magela Creek region being considered of high ecological and conservation value, a trigger value protecting 99% of species was calculated, as recommended by the Australian and New Zealand Environment and Conservation Council, and by the Agriculture and Resource Management Council of Australia and New Zealand [7.44]. The resulting trigger value was 6 μg/L [7.45]. This value has been adopted by the regulator as the compliance ‘limit’ for downstream of the Ranger uranium mine, Australia [7.27, 7.45]. The lower and upper 95% confidence levels associated with this ‘limit’ are 0.3 and 103 µg/L, respectively. To capture this uncertainty in the management framework and enable uranium data to be interpreted in an environmental risk context, the 20th (0.9 µg/L) and 5th (0.3 µg/L) confidence levels of the ecotoxicity trigger value of 6 µg/L were adopted as the ‘action’ and ‘focus’ trigger values, respectively [7.27, 7.46].

Through the implementation of the Water Framework Directive 2000/60/EC [7.47], environmental quality standards in European Union Member States need to be derived to support assessments of the chemical status of the water bodies. In France, for example, a provisional EQS value of 0.3 µg/L was included for uranium chronic exposure, based on ecotoxicity data. Accounting for the bioavailability of metals, conditional EQSs were derived for different environmental conditions, leading to reference values of 1–300 µg/L [7.48]. No international recommendations have been developed for uranium as a chemically toxic element.

Reference [7.49] presents a method for relating chemical concentrations (µg [U]/L) and dose rates (µGy/h) for a specific set of reference organisms. This allows a determination as to whether chemical or radiological toxicity is more limiting for depleted, natural, low enriched and high enriched uranium in various ecological contexts.

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Chapter 8

MANAGEMENT OF SITES CONTAMINATED IN THE EXPLOITATION OF URANIUM

O. VOITSEKHOVYCH
Ukrainian Hydrometeorological Institute, Ukraine

8.1. TYPES OF INDUSTRY

8.1.1. Introduction

All minerals and raw materials contain radionuclides of natural origin. The most important in the context of radiation protection are the radionuclides in the ^{238}U (see Fig. 3.1) and ^{232}Th decay series. Uranium is just one of several elements that contribute to naturally occurring radioactive material wastes and by-products that result from processing of raw materials. The industry processing uranium-containing ores produces residues and wastes that contain uranium and other radionuclides in its decay series, and these require consideration in terms of their environmental and radiological impacts. Other industries and process materials also involve relatively high concentrations of naturally occurring radioactive material (e.g. the phosphate industry), as described in Section 9.4.

8.1.2. Uranium mining and milling

Uranium is present in the environment in many natural minerals and materials, including waters, soils and sediments. Owing to its radiological and chemical toxicity, elevated activity concentrations above natural background levels may have a negative effect on the environment and human health. The main sources of pollutants containing uranium and radionuclides in its decay series are uranium production (mining and milling) and chemical industries dealing with uranium-containing minerals (Fig. 8.1). A general overview of the uranium production cycle is provided in Refs [8.1–8.4] and is further discussed in this section.

Mining can be categorized as surface mining (open pit), underground mining and in situ leaching mining. Uranium ore is mainly mined in open pit or underground mines. The uranium content of the ore is often only 0.1–0.2%. However, in some cases, uranium concentrations in the ore may be as much as

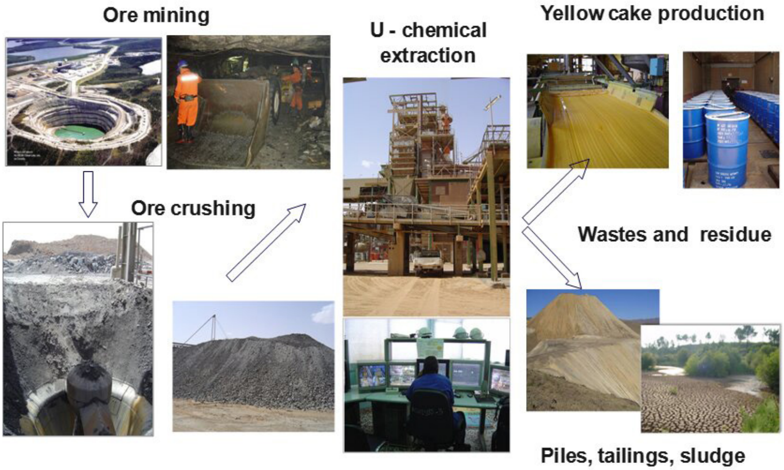


FIG. 8.1. Basic steps of the uranium production cycle (photographs taken by F.P. Carvalho, University of Lisbon and O. Voitsekhovych, Ukrainian Hydrometeorological Institute).

several per cent. The minimum concentrations that are economically suitable for industrial extraction are 0.03–0.05% [8.3]. In this case, a physical process is used to concentrate the ore and increase the grade before chemical treatment. Commonly, the 'ore' is barren rock or mineral residues that remain undissolved in the leaching process. In either case, grade control is usually achieved by measuring radioactivity as a surrogate for uranium concentration.

About 95% of the radioactivity in the uranium ore raw material is from the ^{238}U decay series and about 5% from the ^{235}U decay series, totalling about 150 kBq/kg in ore containing 0.1% U_3O_8. The activity ratio ^{235}U:^{238}U is greater than the mass ratio because of the difference in half-lives of these two radioisotopes. The ^{238}U series has 14 radioactive isotopes in secular equilibrium; thus, each represents about 11 kBq/kg.

Conventional uranium mill procedures include such steps as crushing and grinding of the ore before leaching in tanks with sulphuric or another type of acid, or with alkali to dissolve the uranium oxides, as well as extraction, precipitation and purification of the uranium concentrate (Fig. 8.2).

When the ore is processed, the ^{238}U and the very much smaller masses of ^{234}U and ^{235}U are removed. These solid residues are separated from the uranium rich solution, usually by precipitation.

The tailings residues (the residue of the ore processing after most of the ^{238}U is removed) still contain a significant amount of the original intrinsic radioactivity, including residual ^{235}U and ^{238}U, and also long lived radionuclides

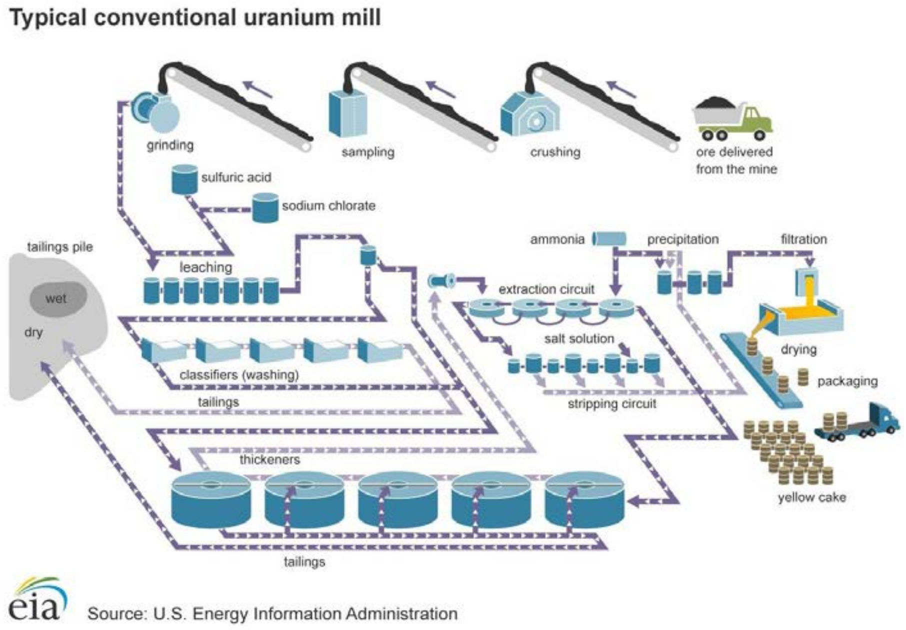


FIG. 8.2. Principal scheme of uranium milling (yellow cake production), including sequential procedures such as ore grinding, crushing, leaching (acid or alkaline solvents), extraction, and creation of residue tailings (source: US Energy Information Administration, July 2019).

of the ^{238}U decay series, such as ^{230}Th and ^{226}Ra, followed by ^{222}Rn (with radiologically significant progeny such as ^{210}Po and ^{210}Pb).

The remaining solution is filtered and the uranium is recovered in some form of ion exchange or solvent extraction system. The pregnant solution from in situ or heap leaching is treated similarly. The final chemical precipitate is filtered and dried. The final product from the mill is uranium oxide concentrate (U_3O_8), which is commonly referred to as 'yellow cake' and is typically packed into sealed 200 L steel drums ready for shipment.

8.1.3. Potential hazards from uranium mining and mill tailings

There are several types of potentially hazardous object associated with uranium mining and milling sites that may affect the surrounding environment and individuals living there. These objects, which are further discussed in this section, include open uranium pits and underground mines, waste rock piles, industrial milling sites and tailings facilities, which are usually located near the mines to limit transport, and underground leaching fields.

8.1.3.1. Conventional mining

As a result of uranium ore mining in open pits or during underground mining, a large amount of waste rock can be produced, which usually contains high concentrations of radionuclides from the uranium–thorium decay series. Low grade materials also result from mining operations. Tailings materials, which contain relatively low uranium activity concentrations and are enriched in other radionuclides of the uranium–thorium decay series, are usually located close to the mill site. In some cases, tailings materials are returned to the uranium open pits or underground mines if groundwater can be protected from contamination and the risk of radionuclide dispersion is low. Some examples are shown in Fig. 8.3 for a conventional uranium mill site in Canada, an open pit in Namibia and a former uranium pit in Kazakhstan, with subsidence of the land surface above the underground mining site and waste rock piles.

Typical mean ^{238}U activity concentrations in waste rock and contaminated soils located in the vicinity of the mine site may vary over the wide range of 0.5–5.0 Bq/g [8.4, 8.6, 8.7]. At the bottom of an open pit, meteoric water and groundwater may often have elevated activity concentrations of about 1–10 Bq/L [8.4, 8.6, 8.7].

Underground mining procedures are mainly associated with high inhalation exposures to ^{222}Rn plus its short lived progeny, and need continuous dewatering of the mine shafts. The mine waters may contain a significant amount of uranium in dissolved forms, as well as other radionuclides in the uranium–thorium decay series. In many conventional uranium mining sites, treatment of the mine water is required before it can be released into the environment.

8.1.3.2. In situ leaching

In situ leaching technology is applicable to uranium deposits located in permeable rock aquifers and enclosed in non-permeable rock (Fig. 8.4). Drill holes are used to pump the leaching liquid (e.g. ammonium carbonate, sulphuric acid) into an underground uranium-bearing aquifer and to pump out the uranium-bearing liquid [8.8, 8.9].

Significant direct environmental impacts due to contamination of groundwater may occur after the closure of in situ leaching and underground mines because significant portions of acids and dissolved uranium and other minerals remain in the aquifer, and an increasing groundwater level is typically associated with the cessation of pumping [8.9, 8.10].

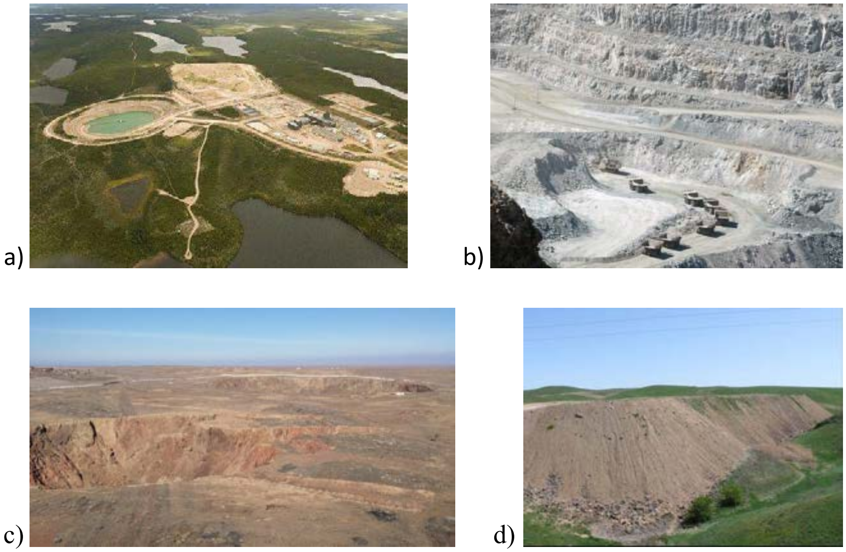
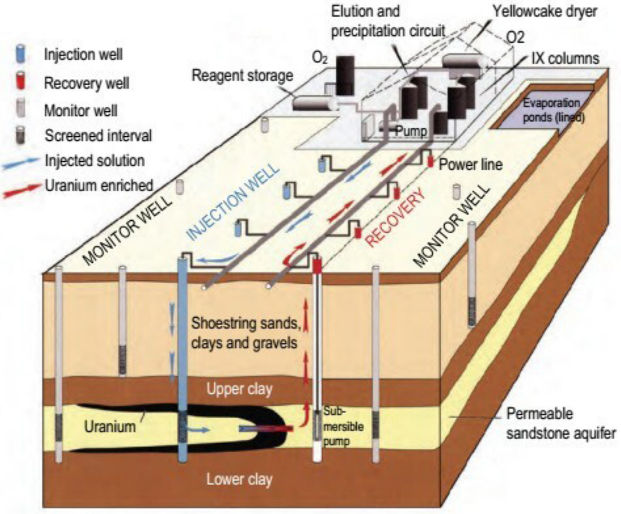


FIG. 8.3. Examples of conventional uranium mining sites where different types of hazard are present: (a) tailings management facility at Orano McClean Lake, northern Saskatchewan, Canada (reproduced from the Canadian Nuclear Safety Commission); (b) open pit uranium mine at Rossing, Namibia (1982) (photograph reproduced from Ref. [8.5]); (c) subsidence of the land surface above the former underground Vostochniy Mine site (2015) (photograph taken by O. Voitsekhovych, Ukrainian Hydrometeorological Institute); (d) remaining waste rock piles with eroded slopes at the former mine site at Kurday, Kazakhstan (2015) (photograph taken by O. Voitsekhovych, Ukrainian Hydrometeorological Institute).

8.1.3.3. Heap leaching

If the uranium content in the ore is too low to be processed through milling, heap leaching is used to remove uranium from low grade ores. The basic principle of this option is to introduce a leaching liquid (such as sulphuric acid) throughout the pile, from the top downwards, and to collect and pump the uranium-bearing liquid to a processing plant. Releases of dust, radon gas and leaching liquid are associated with the leaching process; a long term issue is the possible association of the ore, which contains mineral pyrite (FeS_2), with the uranium deposits. The pyrite can oxidize to produce sulphuric acid that can result in additional leaching of the residual uranium. A general scheme for heap leaching technology that is applied for removing uranium from the crushed ore minerals is shown in Fig. 8.5.



Note: Not to scale – diagrammatic only.

Source: Geoscience Australia.

FIG. 8.4. Scheme of in situ leaching recovery at Beverley uranium deposit, Australia (source: Commonwealth of Australia (Geoscience Australia); licensed under CC BY 4.0 [8.8]).

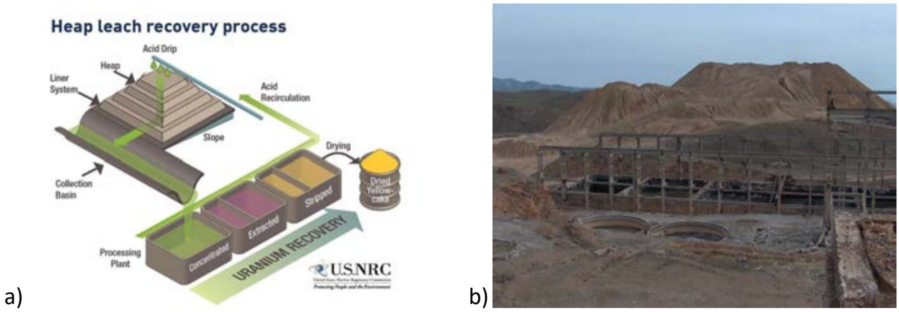


FIG. 8.5. (a) General scheme for the uranium heap leaching procedure (US Nuclear Regulatory Commission File Photo); (b) a former heap leaching facility in Taboshar, Tajikistan, with a front view of milled barren ore prepared for extraction (2006) (photograph taken by O. Voitsekhovych, Ukrainian Hydrometeorological Institute).

8.1.3.4. Uranium milling

Radiological hazards at uranium milling sites may be associated with areas of temporary storage of uranium ore raw materials and with dispersion of crushed materials. During uranium milling procedures — including transport of the radiochemical solutions, final production and the management of uranium production residues — the environment can often be impacted by spillages, dust dispersion or sludge materials produced at the milling site.

The residues (tailings) of the milling process, which contain radionuclides of the uranium–thorium decay series and many other constituents, are usually dumped in special ponds or piles. In the past, tailings materials were often dumped directly into the closest ravines or other types of depression in the landscape, creating tailings ponds adjacent to the main uranium production facility. A large amount of sludge, containing up to 85% of the initial radioactivity of the ore (the contribution of the decay products in the uranium decay series and 5–10% of the uranium initially present in the ore) is produced (see also Fig. 8.1). Several other constituents are extracted from the ore together with uranium (e.g. arsenic, iron, lead, molybdenum, selenium, vanadium) and need to be separated out from the leaching solution.

Several IAEA publications have described the best international practice for environmentally responsible site management at conventional uranium production enterprises (see Refs [8.11, 8.12]). Modern technology and application of the best practices for radiation protection for conventional uranium production under strict regulatory control can minimize contamination of the environment and significantly decrease the radiological impact of the practice. However, the most serious environmental considerations related to uranium production do not result from the uranium production process but from uranium legacy sites (ULSs) (see Section 8.2).

8.2. OVERVIEW OF LEGACY ISSUES AND REMEDIATION STRATEGIES

8.2.1. General considerations

A legacy site may be a facility or area operated in the past to standards which are not consistent with the present approach or that has been affected by major accidents and incidents, that has not been fully remediated and is radioactively contaminated at a level of concern to regulatory bodies. Currently, there is no international accepted definition of a ‘legacy site’. The issues of safety and environmental impact assessment as well as procedures for remedial action planning and its implementation at uranium production legacy sites should be considered in the context of requirements for existing exposure situations. General regulatory requirements and basic practices for remediation of legacy sites associated with past uranium production are considered in Refs [8.13–8.17].

Some useful information on remediation criteria and experience in remediation planning is given in Refs [8.13–8.15, 8.18, 8.19].

Although an operational facility may have had a disposal path when originally commissioned, political, economic or other forces out of the operator's control may have closed that disposal path, for instance for waste from decontamination. Many facilities in the former Soviet Union became legacy sites, as regulatory control was lost during the transfer process, and maintaining the necessary level of safety and management proved to be challenging during the transition to the successor authorities.

Legacy sites were often operated in the past under less rigorous regulatory regimes, with a greater focus on industrial output and less on safety. Furthermore, there was a lack of knowledge about the adverse impacts of the operation (e.g. the chemical toxicity of the uranium was not well appreciated). In addition, the operations involved naturally occurring radionuclides that were not considered dangerous in many countries, simply because they occurred naturally. This may have led to releases of material into the environment and improper management of wastes that would not occur today. ULSs are often characterized by:

- Abandoned tailings and tailings facilities;
- Unsafe conditions (e.g. unstable engineering elements of former mines and uranium extraction facilities, geotechnically unstable waste heaps, underground openings);
- Conventional safety threats (e.g. open shafts, ventilation pits, open trenches, unstable spoil heaps/tailings);
- Radiation safety issues (e.g. uncovered radioactive residues, radon, radioactive water);
- Active contamination pathways (e.g. outflows of mine drainage, surface runoff, dust, erosion) that increase the radiological hazards;
- Operation outside of normal radiation protection measures and the framework of IAEA Safety Standards Series No. GSR Part 3, Radiation Protection and Safety of Radiation Sources: International Basic Safety Standards [8.13];
- Improper decommissioning (or abandonment);
- Posing radiological hazards to people and the environment.

Examples of different types of legacy site of uranium production, which have been characterized in recent years in the framework of IAEA technical cooperation projects, are presented in Chapter 9, as well as being discussed here.

Typical uranium legacy study cases with various types of radiological hazard and impact are presented in Fig. 8.6. These include:

- Former uranium mining open pits, which are often flooded, that contain water with a high content of uranium;
- Waste rock piles and tailings facility with insufficient soil cover and unstable dam slopes subject to erosion;
- Milled barren uranium ore prepared for heap leaching or its residues after extraction;
- Acid drainage from uranium tailings creating a significant impact on the surrounding environment;
- Former uranium mining underground shafts that are flooded by groundwater, releasing uranium-bearing waters to the surface;
- Former uranium extraction facilities and contaminated industrial sites exposing people and the surrounding environment.

8.2.2. Planning remediation

A typical remediation policy should consider the national legal framework, institutional structure and applicable international conventions while providing for the allocation of responsibilities and resources, in addition to defining safety and security objectives. It should also consider public information requirements and the need for public participation in the decision making process [8.11–8.14]. The national strategy should reflect and elaborate on the goals and regulatory safety requirements as part of the policy statement. For its formulation, detailed information is needed on the current situation in the country (organizational, technical and legislative). Furthermore, large sets of site specific data on the current situation at the legacy site should be analysed (hazards characterization and monitoring data, prioritized exposure pathways and safety assessment) as a basis for justification of the optimal set of remediation measures to be proposed.

The technical solutions proposed for the remediation of sites should be politically, technically and economically feasible. When selecting a set of technological procedures, the appropriate remediation objectives should be established, and acceptable end state conditions achieved after remediation is completed. The steps in formulating and implementing the strategy include the selection of technical procedures, the allocation of responsibility for implementing the identified procedures, the establishment of supervisory mechanisms and the development of implementation plans.

Effective remediation policy should be based on the basic principles of remediation that include: (i) providing a justification for undertaking remediation; (ii) optimization of protection in relation to the remedial actions; (iii) providing

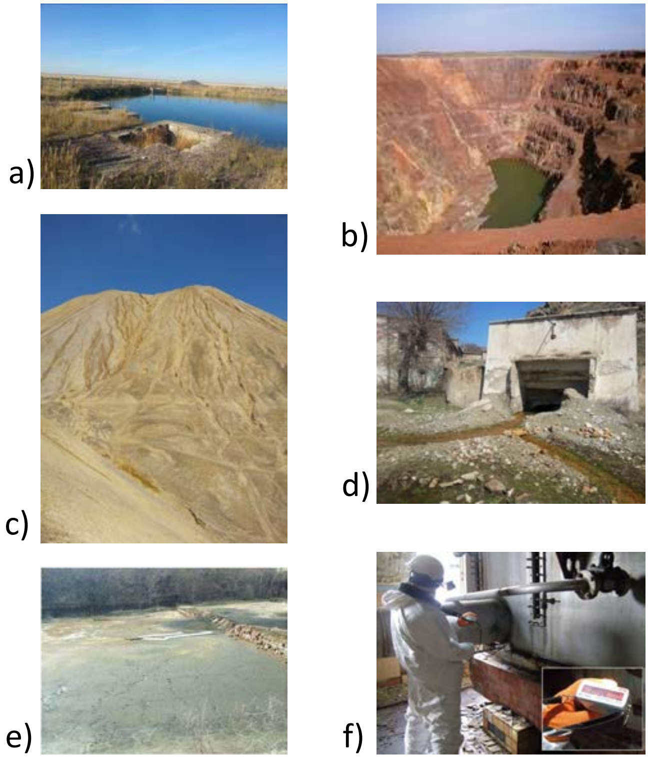


FIG. 8.6. Examples of uranium production legacy sites with various types of radiological hazard and impact. (a) Subsided mine surface complex flooded by shallow groundwaters (Kazakhstan, 2013); (b) former uranium pit (Vostochmiu mine, Kazakhstan, 2013); (c) milled barren uranium ore materials at Taboshar, Tajikistan, before it was remediated in 2019; (d) contaminated creek waters from a flooded uranium mine shaft (Taboshar, Tajikistan, 2014); (e) radioactive ^{230}*Th sludges remaining after thorium separation procedures during uranium extraction (Prydniprovskyy Chemical Plant, Kamyanske, Ukraine, 2016); (f) former uranium extraction facility at the Prydniprovskyy Chemical Plant, near a precipitation column filled with uranium extraction residues (Kamyanske, Ukraine, 2014). The photographs were taken by O. Voitsekhovych, Ukrainian Hydrometeorological Institute.*

appropriate protection of future generations and the environment; (iv) efficiency in the use of resources; and (v) providing open and transparent interactions with stakeholders [8.12].

An established remediation policy is essential for establishing the core values on which remediation is to be based. Environmental remediation policies will set the nationally agreed position and will give visible evidence of the concerns and intent of the country. The formulation of a national policy will encourage the establishment of a legal framework for ensuring coherent and consistent remediation approaches [8.12–8.14].

The decision about the necessity of remediation measures, including cleanup of contaminated sites, decontamination of uranium production facilities and removal of uranium ore-containing materials should be made on the basis of a site investigation, assessment and justified remediation action plan to be developed by authorized site operators and approved by authorized regulatory authorities.

In accordance with best international practices (see Refs [8.11, 8.12]), the decision as to the need for, and type of, remediation at a contaminated site must be based on the results of a safety assessment (estimated exposure pathways, doses and associated radiation risks in comparison to the reference doses and risks established by the regulatory body). The regulatory body should also establish remediation objectives, such as derived activity concentration levels for uranium and other radionuclides in the uranium decay series, to be compliant with environmental safety criteria for remediated sites.

As a rule, the hazardous impacts of uranium mining and uranium extraction residues arise not only from the presence of uranium (in different geochemical forms, determining its toxicity), but also from other radionuclides in the uranium decay series in the contaminated materials at the sites of mining and processing of uranium ores. Therefore, methods for identification, inventory determination, safety assessment, waste management and remediation must be applicable to all radionuclides in the uranium decay series that are associated with the contaminated sites and facilities.

However, depending on the specific technology used for uranium extraction from the uranium-containing raw materials and on the different types of residue, high activity concentrations of either ^{226}Ra or uranium isotopes can be dominant. Geochemical considerations, including oxidation conditions, may determine whether uranium migration in the drainage water and groundwater from the uranium tailings and other wastes will be significant. Furthermore, in the premises of former uranium extraction facilities, the working zone may be significantly contaminated with dispersed yellow cake materials or other materials containing high concentrations of uranium in oxidized forms. Therefore, various alternative decontamination techniques may be applicable, and the adoption of specific techniques for remediation of the contaminated sites will need to be justified considering site specific conditions.

8.2.3. Regulatory framework and remediation objectives

8.2.3.1. Basic regulatory requirements

Analyses of programmes that have been successfully implemented for the remediation of ULSs in recent decades in various countries show that these were based on sufficient planning and were implemented under State policy (see Refs [8.11, 8.12]). Good practice assumes that responsibilities for different stages of the remediation process are clearly defined and shared between State authorized bodies (ministries, departments and regional State administrations), relevant regulatory authorities and designated operators of the ULS. The legally defined scope of responsibility of the various regulatory bodies and government authorities charged with ULS management, planning and implementation of the remediation activities at the ULSs had to be defined. In addition, sources of, and mechanisms for, financing of the remediation had to be found, and legal procedures and mechanisms for securing that finance had to be put in place.

The regulatory issues and requirements that may significantly affect the efficiency and sustainability of remediation measures, and that must be considered in any remediation plans for sites contaminated with uranium raw materials (and uranium-containing residues) are listed below [8.13–8.16]:

- Identification of ULSs and hazardous areas to be remediated;
- Remediation objectives;
- Consistency of remediation planning (assessment, justification and optimization);
- Site characterization, monitoring programmes and surveillance (at the different stages, namely remediation preparedness, implementation and post-remediation period);
- Radiation protection of remediation workers and of the public;
- Radioactive discharges;
- Institutional control and conditions for release of the remediated site (or a part of it) from regulatory control;
- Public involvement in decision making (communications and awareness).

Most of these aspects are of rather common applicability and are also relevant to other situations of radioactively contaminated site management and radiation protection. Specific consideration of these issues is beyond the scope of this book. Therefore, only some important clarifications regarding the identification of uranium production legacy sites and other naturally occurring radioactive material sites contaminated with uranium-containing materials and remediation objectives are briefly addressed in Section 8.2.3.2 based on Refs [8.13–8.16].

8.2.3.2. Identification of areas that require remediation

Not all sites (or objects) related to former mining and processing of uranium ores may be identified as radiological legacy sites and involve significant health risks or environmental impact such that remediation is needed. Therefore, the identification of relevant ULSs for which remediation needs to be considered should be based on adequate criteria [8.13–8.16].

The primary method used to identify a ULS relies on available historical information on former uranium mining and processing activities, taking into consideration site characterization data, including radiological surveys and dose assessment studies. A ULS may be present if at a site, over an extended period, uranium ores and related materials containing elevated levels of natural radionuclides have been mined, stored, processed or disposed of, and if the operational conditions or disturbances from normal operations may have been such that relevant radiation exposures may occur now or in the future (i.e. there is an existing exposure situation). Indications of the presence of a ULS might include:

- Increased external dose rates due to naturally occurring radionuclides in the uranium decay series or increased radon concentrations in ambient air or inside buildings;
- Increased concentrations of naturally occurring radionuclides in the uranium decay series or some non-radioactive contaminants related to the former uranium production in groundwater or surface water, or in vegetation, caused by previous uranium mining or processing activities.

Any territory or facility related to past uranium production should not be considered a ULS if the activity concentration in the residue materials, soils, dust (aerosols) or environmental samples for radionuclides of the ^{238}U and ^{232}Th decay series is below 1 Bq/g, independent of the type, extent and land use. This criterion complies with the requirements for existing exposure situations (see paragraph 5.1 of GSR Part 3 [8.13]).

In some cases, specifically if residue materials may affect drinking water resources, the regulatory body may establish stricter criteria. In such cases, the reference dose levels may be established by the regulatory body based on safety assessment procedures and may be lower than those recommended in paragraph 5.1 of GSR Part 3 [8.13].

The reference level above which a ULS is present should be defined by the regulatory body based on criteria for the radiation exposure of individuals of the local population. Paragraph 5.8 of GSR Part 3 [8.13] states:

> "Reference levels shall typically be expressed as an annual effective dose to the representative person in the range of 1–20 mSv or other corresponding quantity, the actual value depending on the feasibility of controlling the situation and on experience in managing similar situations in the past."

The evaluation of whether a ULS exists and remedial action is required should be based on the safety and environmental assessment, with comparison of the results against the established reference levels. The currently permissible options for land use of the area (and its parts) and its surroundings must be considered. If a legally binding plan for permissible land use does not exist, land uses surrounding the site and anticipated developments should be used as the basis for this assessment. Commonly, if the ULS lies within the catchment area of a usable groundwater aquifer, the assessment should assume, independent of the current situation, use of the groundwater.

8.2.3.3. Remediation objectives

If remedial action is deemed appropriate, it must be planned and implemented such that protection and safety are optimized. Remediation at a ULS must be commensurate with the radiation risks associated with the existing and future exposure situations. To be justified, the remedial action needs to yield sufficient benefits to outweigh any radiation risks or other detriments associated with that remedial action.

Performance of a remedial action will not necessarily result in the elimination of all radioactive materials or all traces of radioactive or toxic materials related to the ULS. The optimization process may result in extensive remediation, but not necessarily restoration of previous conditions. The optimization must weigh the advantages and disadvantages of available decontamination, protection and restriction measures.

The criteria based on the reference levels recommended in GSR Part 3 [8.13] can serve as a basis for establishing a set of site specific remediation objectives to be approved by regulatory authorities establishing safe conditions for the site, allowing its conditional release from regulatory control or need for long term institutional control after site remediation. Site specific remediation objectives and safety criteria derived from established dose rate criteria can be formulated as specific activity concentrations of radionuclides in the uranium decay series in the environment. Some examples establishing such criteria for ULSs are given in

Refs [8.17, 8.18]. Some typical remediation objectives, which were applicable in many European Union Member States, are listed below:

- The soil or any materials with an activity concentration of <1.0 Bq/g for any radionuclide of the uranium–thorium decay series can be used for agricultural or industrial purposes without any restrictions. In some countries, criteria for free release from regulatory control in agricultural use is established for ^{226}Ra in soils and materials, with activity concentrations <0.2 Bq/g. Other countries have norms for radiation protection, establishing such criteria over a wide range (1–5 Bq/g), depending on how and for which purposes and conditions the materials with elevated activity concentration will be used.
- The gamma dose rate at the remediated areas should not exceed 0.2–0.3 µSv/h above local natural background (typically, total dose rates ≤0.5 µSv/h can be acceptable as a remediation objective to be achieved after ULS decontamination).
- The ^{222}Rn exhalation rate from the remediated surfaces (cleaned up soils at industrial uranium production sites or at the surface of covered uranium residue tailings) should be <0.7–1.0 $\text{Bq} \cdot \text{m}^2 \cdot \text{s}^{-1}$.
- The ^{222}Rn ambient activity concentration in air should be <250 Bq/m^3 in social and residential areas, and <1000 Bq/m^3 in industrial premises that are under regulatory control.

The criteria described above should be considered as indicative examples. In fact, for each specific safety case, such objectives should be established by regulatory bodies on the basis of safety assessment and optimization methodologies [8.13, 8.19, 8.20]. Reference [8.18] reports:

> “Buildings with a “non removable” surface contamination < 0.5 Bq/cm^2 (natural uranium) can be used further commercially or industrially. In… construction debris from demolition work with < 0.2 Bq/g Ra 226 can be released without restriction (scrap collection or recycle). At specific activities between 0.2 Bq/g and 1.0 Bq/g dumping onto contaminated areas follows for which unrestricted release is in any case not planned (heaps and tailings ponds). At specific activities > 1.0 Bq/g dumping is to be studied from a radiation protection viewpoint.
>
> “Scrap with a surface contamination < 0.5 Bq/cm^2 (after cleaning) can be released for smelting.”

Assessment of radioactivity (and specifically uranium) in water at sites contaminated with uranium-containing materials is performed based on an exposure analysis, whereby site specific, water use scenarios are considered (e.g. drinking water consumption, livestock watering, plant irrigation). It is, however, often sufficient to compare measured concentrations to screening values or guidance level values for drinking water. If water is suitable for drinking, it can generally be used without any restrictions.

Both WHO [8.21] and the European Commission [8.22] consider an effective dose rate of 0.1 mSv/a from drinking water consumption as corresponding to a very low level of risk that is not expected to give rise to any detectable adverse health effects. Accordingly, WHO defines a value of 0.1 mSv/a as the individual dose criterion for the drinking water scenario (730 L/a consumption by an adult person). In Directive 98/83/EC of the Council of the European Union [8.23], this level is referred to as 'total indicative dose'. For the determination of whether water intake may cause effective doses >0.1 mSv/a, a graded approach has been proposed by WHO and the European Commission.

In terms of screening levels for gross alpha activity (GAA) and gross beta activity (GBA), WHO sets 0.5 Bq/L and 1.0 Bq/L for GAA and GBA, respectively. The European Commission stipulates a more stringent level of 0.1 Bq/L for GAA, but also adopts 1.0 Bq/L for GBA.

If the screening levels are exceeded, then the complete radionuclide composition of the water should be determined. For assessment of the measured individual radionuclide concentrations, concentration guidance levels have been defined by WHO for all dose relevant radionuclides in drinking water. In an analogous approach, the European Commission defines reference concentration levels, *C*(der). Guidance level values and *C*(der) values for the natural radionuclides of relevance are presented in Table 8.1.

If the following additive formula is satisfied, then the individual dose criterion (respectively the total indicative dose of 0.1 mSv/a) is met and no further action is required. In the case of a mix of specific radionuclides of the uranium–thorium decay series in drinking water, the concentrations should satisfy the following formula, according to WHO and the European Commission [8.22]:

$$\sum_{i=1}^{n} \frac{C_i\left(\text{obs}\right)}{C_i\left(\text{der}\right)} \leq 1 \tag{8.1}$$

where C_i(obs) is the observed concentration of radionuclide i, C_i(der) is the derived concentration of radionuclide i and n is the number of radionuclides detected.

An intake of 2 L/d for 1 year will result in an effective dose to an adult person of 0.1 mSv [8.22]. If the above expressions are not satisfied, then a detailed analysis of the use of the water is necessary. This requires a complete exposure analysis on the basis of realistic exposure scenarios.

TABLE 8.1. WORLD HEALTH ORGANIZATION AND EUROPEAN COMMISSION LEVELS TO ASSESS RADIONUCLIDE CONCENTRATIONS IN DRINKING WATER [8.21, 8.22]

Radionuclide	Guidance level (Bq/L)	Reference concentration level, *C*(der) (Bq/L)
^{238}U [a]	10	3
^{234}U [a]	1	2.8
^{226}Ra	1	0.5
^{228}Ra	0.1	0.2
^{210}Pb	0.1	0.2
^{210}Po	0.1	0.1

[a] Uranium chemical toxicity is not assessed.

It should also be noted that uranium concentrations in drinking water are limited primarily by the chemotoxic hazards caused by intake of this element (see also Chapter 7). WHO [8.21] reports that up to a level of 30 μg/L no adverse effects on human health have been found, when water of this concentration is permanently used. However, in many countries, the limit of uranium concentration in drinking water has been set to <30 μg/L [8.21].

In many cases, near ULSs, the surface waters and groundwaters can be contaminated by chemical mining of related elements that require synergistic effects to be considered by reference to radiological and toxicological criteria in water treatment planning. Remediation measures to be taken are suitable if they are based on scientifically justified, technically feasible and economically reasonable methods that have been tested and proven in practical application, or if they are expected to be suitable in practice.

A specific consideration for designing waste management strategies and disposal of uranium-containing contaminants is the need for a very long period of legacy site management. When assessing the long term effectiveness of remediation options, future potential exposures should be estimated for periods during which non-negligible radiation exposures may occur or that capture the expected maximum of the radiation exposure and/or for which reasonably reliable assessments are possible. The regulatory body can generally assume that estimates of the radiation exposure for a period of 200 years are sufficient. If the maximum radiation exposures may occur only later, the regulatory body can require the consideration of extended periods in the assessments, but generally not more than 1000 years.

As a decision aid, a cost–benefit analysis, multi-attribute utility analysis or other appropriate quantitative evaluation method may be used. If relevant to the assessment, and if suitable approaches and parameter values are available, non-radiological impacts should be included in the quantitative evaluation. Some additional information regarding remediation objectives, site characterization, assessment remediation planning and implementation of remediation technologies can be found in Refs [8.2, 8.24, 8.25].

8.3. TYPES OF WASTE CONTAINING URANIUM

The following waste types may be generated during uranium ore extraction or treatment, or because of remediation of ULSs:

- Uranium mineralized rock materials and overburden (waste from weathered rock, possibly together with other barren rocks, which can present a direct exposure hazard to people and to the environment through the enhanced content of radionuclides in the uranium decay series, heavy metals, arsenic, dust and radon);
- Ores with subeconomic levels of mineralization or high levels of contaminants (can be considered for milling at times of high demand or towards the end of their life; otherwise, they are included in the remediation process owing to their potential for causing adverse environmental impacts);
- Mill tailings (the result of the milling process, which consists of the successive crushing and grinding of ore grade rocks to produce a relatively uniform sized, sand-like material, which is then subject to leaching by alkaline or acidic reagents to dissolve and extract the uranium);
- Waste from heap leaching operations (sludge after purification of uranium-containing milled materials and radiochemical solutions, which may also contain high concentrations of ^{230}Th and ^{226}Ra and have the potential to cause environmental and health detriments);
- Liquid wastes and residues from in situ leaching operations (solids and sludge from the neutralization of solutions; spent ion exchange resins, salt residues and used filters from water treatment; scales from pipework, pumps, valves and filters; and residues from evaporation ponds).
- Spill materials containing ‘yellow cake’ and contaminated scale materials;

- Water treatment waste (precipitates, sludges, evaporates or ion exchange resins resulting from the treatment of contaminated water during remediation operations and decommissioning, that cannot be recharged and recovered, and which may contain high activity concentrations);
- Residues from decontaminating and dismantling infrastructure, and the physical remains of infrastructure that cannot be recycled or reused (building materials, structural steel, road surface materials, parts of the mine and mill equipment, machinery, cleaning materials and agents used in decontamination of the facilities, contaminated water from high pressure hosing and washing down of the plant and equipment, and ion exchange resins used in decontamination activities);
- Contaminated soils (soils on site and off site may have become contaminated with radionuclides and heavy metals originating in the ores, or with materials such as lubricants and fuel oils, and they must undergo a proper remediation procedure or be safely disposed of);
- Waste not covered by the above categories (laboratory waste, glassware, clothing, scales from pipelines, pumps, valves and filters).

These material types require safe in situ management or relocation to alternative disposal sites during the remediation of the legacy site. In addition, the following material types may arise from the remedial action:

- Excavated top soils that have been contaminated by solid or liquid materials;
- Excavated waste rock, ore residues or tailings that may be mixed with other materials (e.g. top soils and material used as cover or liner);
- Metal components arising from equipment that cannot be reused;
- Construction materials (e.g. steel, concrete, bricks) arising from the refurbishment or demolition of buildings and other structures (e.g. settling ponds).

8.4. REMEDIATION TECHNOLOGIES

8.4.1. Containment technologies

Containment technologies are based on techniques to isolate contaminants at the site from meteoric water ingress, to limit radon emission, to discourage intrusion, and to prevent erosion of materials and contaminant transfer to surrounding areas and groundwaters (Fig. 8.7). They are potentially of importance when removal of contamination from the site is not feasible or found not to be cost effective. General principles on the justification and application

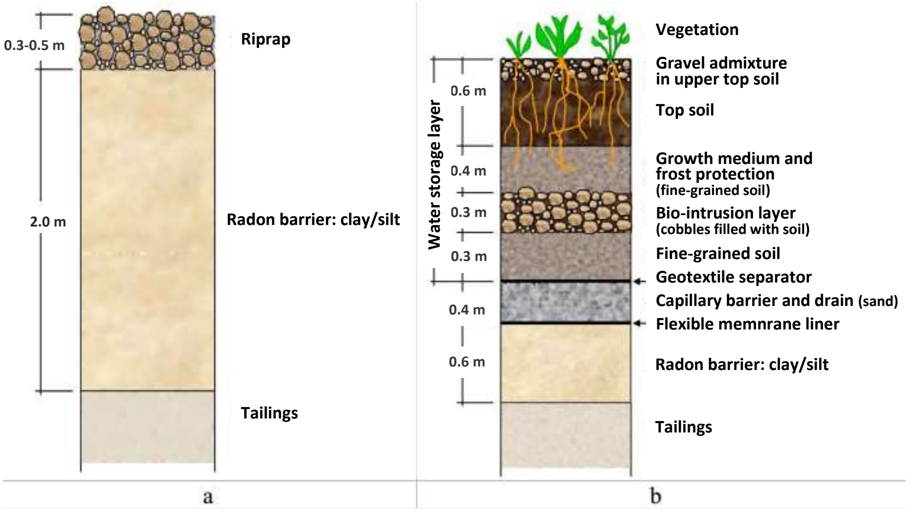


FIG. 8.7. Examples of the cover design for uranium tailings: (a) the simplest option for the Uranium Mill Tailings Remedial Action (UMTRA) Project Shiprock tailings disposal structure, USA; (b) the UMTRA Monticello containment structure, USA (after Ref. [8.25]).

of geochemical and engineering barriers for long term stable containment of uranium-containing materials are addressed in Refs [8.25–8.27].

8.4.2. Surface caps

Surface caps (covers) are artificial barriers that cover a site to restrict water infiltration, limit emission of radon from the tailings and limit natural erosive processes. They may be a standard construction practice performed in conjunction with other remediation technologies. The constructed surface cap can consist of an upper vegetated layer, a layer to prevent erosion and animal burrowing, a drainage layer and a low permeability layer. Soil and clay caps are constructed by spreading soil/clay over the contaminated area to achieve a low permeability cover. This low permeability layer is normally composed of fine-grained natural soils, but to achieve a low permeability, the soil/clay layer may incorporate bentonite, lime, cement or other materials. Asphalt caps (single-layered caps composed of bituminous asphalt) may be used when a lower susceptibility to environmental factors is to be achieved. Some other types of cap are also single layered and consist of aggregate and cement material mixtures. Synthetic membranes (based on polyvinyl chloride, polyethylene and hypalon) and sealants/stabilizers are also used to restrict erosion and surface water infiltration.

In most cases, several layers of soil materials with different functions may sufficiently reduce gamma dose rates at the surface of tailings, prevent emissions and protect the surrounding environment. The determination of what type of surface cap is most efficient depends on many factors, including radiation and ecological safety requirements, climatic conditions, cost factors and other site specific issues. The most typical cover models for uranium tailings are shown in Fig. 8.7.

In many existing tailings piles with sufficient cover, the riprap layers are covered with grassed top soils. A multilayer cover system of soils and waste rock is generally used to resist erosion, promote runoff, limit infiltration, minimize radon emissions, reduce long term maintenance and minimize animal and human intrusions. Overall, the aim is to reduce risks to human health and the environment.

8.4.3. Cut-off walls

Cut-off walls are used to prevent the potential migration of contaminants and to prevent possible groundwater contamination in the proximity of a contaminated site. They are designed according to the soil characteristics and the depth of contamination. The most common form of cut-off walls is slurry walls formed by excavation of a vertical trench. Other types are made of advanced polymer based materials, cement based grout curtains and polymeric geomembranes, semicrystalline plastics and thermoplastics, with very low permeability to gases, vapours and liquids. Innovative technologies can also include vertical cryogenic walls (interstitial water, frozen within the soil) and biological barriers (aggregation of microbes into large masses by injection of nutrients into wells).

8.4.4. Bottom barriers

Bottom barriers are horizontal subsurface barriers that prevent vertical migration by providing a ‘floor’ of impermeable material beneath the contaminated materials. As discussed above for cut-off walls, the bottom barriers can be made of different materials and may have a variety of designs. To limit downward leaching, grout injection may be used to create the required impermeable barrier below the contaminated materials. Different types of liner are also in wide use as bottom barriers. A liner is a layer of material placed beneath a landfill to prevent waste migrating out of the waste unit into other areas, particularly groundwater. A more advanced lining system may include several liners, a leachate collection and removal system between the liners, and, in the case of a landfill, a leachate collection and removal system above the liners. All modern tailings facilities have bottom barriers, preventing groundwater contamination. This contrasts with most tailings facilities constructed at the beginning of the uranium production era (the last 40–60 years of the twentieth century) in many countries that have created huge problems in preventing and controlling dispersion of highly contaminated leachates into the surrounding environment [8.28].

8.4.5. Stabilization/immobilization technologies

Reducing the mobility of contaminated material in the environment by retaining it within the host medium is a basic mechanism underlying all stabilization technologies. Stabilization technologies can generally be grouped into two major categories, in situ encapsulation and compaction [8.25, 8.28], as follows:

(a) Encapsulation technologies trap or immobilize contaminants by fully encasing the waste in a monolithic structure through the injection of grout or polymers. The end product of this process is a monolithic block of contaminated material encapsulated in grout or polymer. Mixing soil with slurry to form a cement-like matrix that immobilizes contaminated soil, increases soil strength and decreases soil permeability is one of the options applied for encapsulation.

(b) Compaction is used to consolidate soils, sludges and bulk wastes. Compaction provides a denser and more impenetrable waste form that offers less surface area exposure to natural processes that would otherwise tend to cause contaminant migration. The impact of the weight of the compactor causes shock waves within the underlying media, thereby consolidating the materials. Compaction may not be suitable for some types of waste (e.g. drums containing hazardous chemicals).

8.4.6. Treatment of waters

Various decontamination techniques require process waters that must be treated for contamination after use. The method of treatment depends on the type of contaminant, the volume of water and the flow rate of the water. Large volumes may be evaporated in ponds or tanks, and the residual evaporates collected as a sludge or crystalline mass for management and disposal as for other radioactive waste. Substantial volumes may also be treated by use of ion exchange resins, reverse osmosis plants, filtration and precipitation methods [8.20, 8.25].

The use of precipitation methods to remove radionuclides from solution is well known, the most common being bulk precipitation using lime. Such precipitation will remove most heavy metals from solution. There are also other contaminant specific methods in use, for example the use of barium chloride to remove radium from uranium mill process waters. In all such precipitation processes, the precipitate needs to be managed as a radioactive waste with an appropriate disposal strategy. Similarly, ion exchange and reverse osmosis processes will also produce sludges or precipitates that will require management and disposal.

Water treatment may be a significant challenge in the remediation of flooded underground mines, which often discharge water with a high content of uranium, radium and heavy metals into the environment. In some regions and countries (e.g. European Union Member States, United States of America), the regulatory requirements to protect groundwaters from contamination are very strict. That is the reason that high cost, site specific technologies are often considered for water treatment to achieve high water quality for possible use in irrigation and even for drinking water supply.

8.4.7. Protection of water resources during site decommissioning and remediation

At all stages of the decommissioning or remediation process, stringent precautions should be taken to ensure that no significant contamination of water resources occurs. This applies to both surface and groundwater resources. Water supplies for decontamination work should be recirculated within leak free systems to the greatest extent practicable. Water can also be used for dust suppression in contaminated areas. The discharge of these waters to the environment should be strictly controlled. Ponds and reservoirs used for containing contaminated waters and sludges should be dewatered and lined, if possible, to prevent seepage to groundwater. Earth movement and other remediation surface activities should be confined to areas where all water runoff is contained and prevented from discharging into the environment except under conditions of strict control and supervision. At remotely located sites that have been closed, or where operations are suspended, and there is no active supervision, the use of permeable reactive barriers may be considered as an additional means of water resource protection. Best practices as examples are considered in Refs [8.9, 8.12, 8.28].

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Chapter 9

CASE STUDIES

O. VOITSEKHOVYCH, T. LAVROVA
Ukrainian Hydrometeorological Institute, Ukraine

F.P. CARVALHO
University of Lisbon, Portugal

9.1. URANIUM LEGACY SITES IN CENTRAL ASIA AND UKRAINE

The relevance of the examples of uranium legacy sites and their impacts on the environment that are considered here arises from the consideration that the impacts of the radiological hazards often identified in areas of former uranium production cannot usually be managed in a simple way. The environmental impacts and economic damage from inadequate management can be very significant. In many cases, remediation of such sites and control of the associated radiological hazards requires considerable financial and intellectual resources [9.1, 9.2].

Usually, during processing of the raw material, up to 95% of the uranium is extracted from the ore concentrates. Therefore, the residues from the processed ore contain mainly ^{226}Ra and its progeny, ^{222}Rn, ^{210}Pb and ^{210}Po, as well as ^{230}Th and some other radionuclides. However, even small amounts of uranium contained in the residues may have, in specific geochemical conditions, a significant impact through contamination of groundwater and surface water [9.3, 9.4].

In some cases, high activity concentrations of uranium can be identified in micrometre sized fractions of dust particles, and aerosols of such dusts can be present at the processing facilities, creating a high potential risk of human exposure via inhalation pathways. The high solubility of submicrometre sized, uranium-containing particles in the lungs and the high radiotoxicity of such particles mean that special attention should be paid to aerosol monitoring and dust suppression in radiation protection at the workplace and in remediation planning [9.5].

Important radioactive substances that are usually present at uranium legacy sites are materials containing ^{226}Ra and other gamma emitting radionuclides in the uranium decay series. These radionuclides give rise to elevated external gamma dose rates in those locations in which uranium production residues have accumulated. Identification and assessment of the main sources of radiological hazards at uranium legacy sites is a complex task and includes a need to consider the main exposure pathways associated with all of the radionuclides in the uranium decay series. Some typical examples of radiological hazards at uranium legacy sites are illustrated and discussed in Section 9.1.1 based on assessments carried out over the past 20 years in Central Asian countries and Ukraine [9.2, 9.6–9.14].

9.1.1. Uranium legacy sites

9.1.1.1. Underground mines and mine waters

Typical examples of the environmental impacts that arise at uranium mine sites are the flooding of mine shafts or of open pits by groundwater due to closure of the mines in the absence of adequate decommissioning procedures. As a result, highly contaminated mine waters may be released at the surface, often causing significant harm to the environment as well as being a potential cause of human exposure.

Figure 9.1 provides examples of underground water leakage from uranium mines. These include water movement via mine tunnels after the cessation of uranium exploration at the settlement of Jangiabad, and examples of releases from the former mine site Charkesar-2, both in Uzbekistan [9.6], as well as an example of mine water released from adit No. 2 of the former uranium mine Taboshar, Tajikistan. The uranium activity concentrations in such mine waters vary in the range of 10–20 Bq/L [9.2]. The local population may use such waters for irrigation, livestock watering or even for drinking, creating a significant health risk. Thus, mine water treatments at such sites are necessary to bring the water into a safe state.

9.1.1.2. Uranium open pits in Tajikistan and Kazakhstan

Uranium ore mining was carried out in open pits at many sites. Such open pits are usually well connected hydraulically with groundwater horizons as well as with underground workings, shafts and transport galleries of the mines. If measures for water level regulation are terminated, the groundwater levels increase, and the contaminated water fills up the open pits, creating water bodies that look like mountain lakes, but with relatively high activity concentrations of uranium. Figure 9.2 shows examples of such former uranium mine pits in Taboshar, Tajikistan and near the former mine site of Kurday, Kazakhstan [9.2, 9.6].

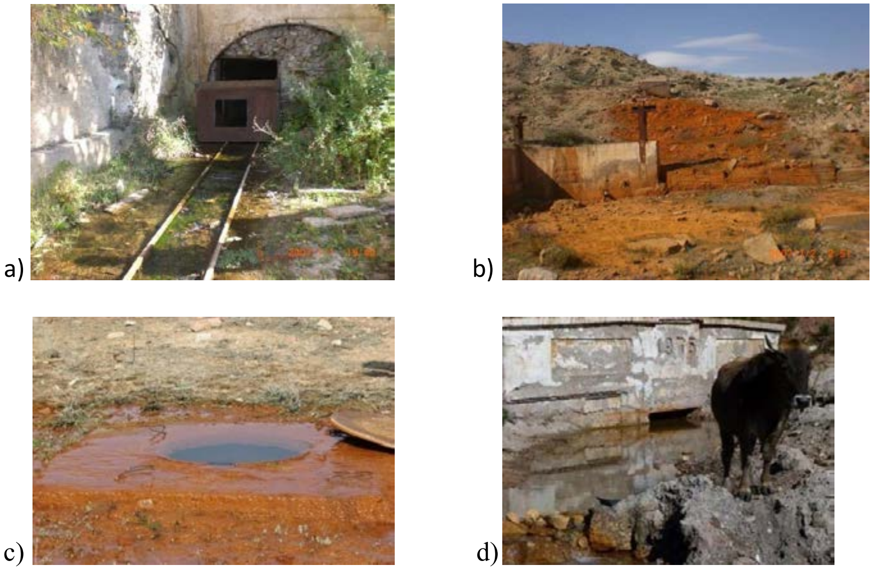

FIG. 9.1. Mine water outlets from former uranium mines: (a) water release at mine transport shaft in Jangiabad, Uzbekistan (2010); (b) surface acid mine waters with a high content of uranium and iron leach in Jangiabad, Uzbekistan (2010); (c) mine water release from the ventilation shafts in the village of Charkesar, Uzbekistan (2007); (d) adit of former mine No. 21 in Minkush, Kyrgyzstan (2013), flooded by groundwater (photographs taken by O. Voitsekhovych, Ukrainian Hydrometeorological Institute).

The activity concentrations of ^{238}U and ^{234}U in the waters of this former uranium pit in Taboshar, observed in different seasons in 2006–2008, were in the range of 20–30 Bq/L [9.2]. More detailed uranium data from Taboshar are provided in Table 9.1 [9.8].

Uranium concentrations of 1200–1400 μg/L were found in an artesian spring downgradient of the open flooded pit at Kurday, Kazakhstan [9.6]. Additional data and processes related to environmental contamination from the former uranium mine and mill sites in Ukraine and Kyrgyzstan can be found in Refs [9.9, 9.12].

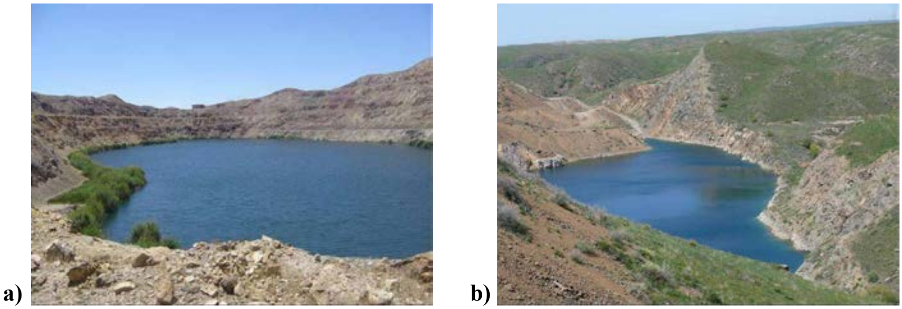

FIG. 9.2. General view of mountain lakes formed as a result of the flooding of (a) the former uranium open pit near Taboshar, Tajikistan and (b) the former mine site of Kurday, Kazakhstan (photographs taken by O. Voitsekhovych, Ukrainian Hydrometeorological Institute).

9.1.1.3. Waste rock piles

Water flows may not only wash out the leached uranium and its decay products from dispersed raw materials and from mine workings, but can also erode waste rock piles comprised of barren ore and, in some cases, protective cover materials or stored ore material, resulting in dispersion of uranium-containing materials. In some cases, such materials may contain significant concentrations of uranium and other radionuclides of the uranium–thorium decay series.

TABLE 9.1. ACTIVITY CONCENTRATIONS OF URANIUM IN WATERS COLLECTED FROM THE TABOSHAR MINE SITE, TAJIKISTAN [9.8]

Location	Activity concentrations				
	U (µg/L)	^{238}U (Bq/L)	^{234}U (Bq/L)	^{234}U:^{238}U	^{226}Ra (Bq/L)
Mine	1660	20.5	20.9	1.02	1.13
Outlet from adit No. 2	1430	17.6	17.9	1.02	0.87
Syr-Darya river	35.3	0.44	0.73	1.66	0.006

Note: Uranium in water is commonly controlled on the basis of its chemical toxicity, according to the World Health Organization guideline value of 30 µg/L.

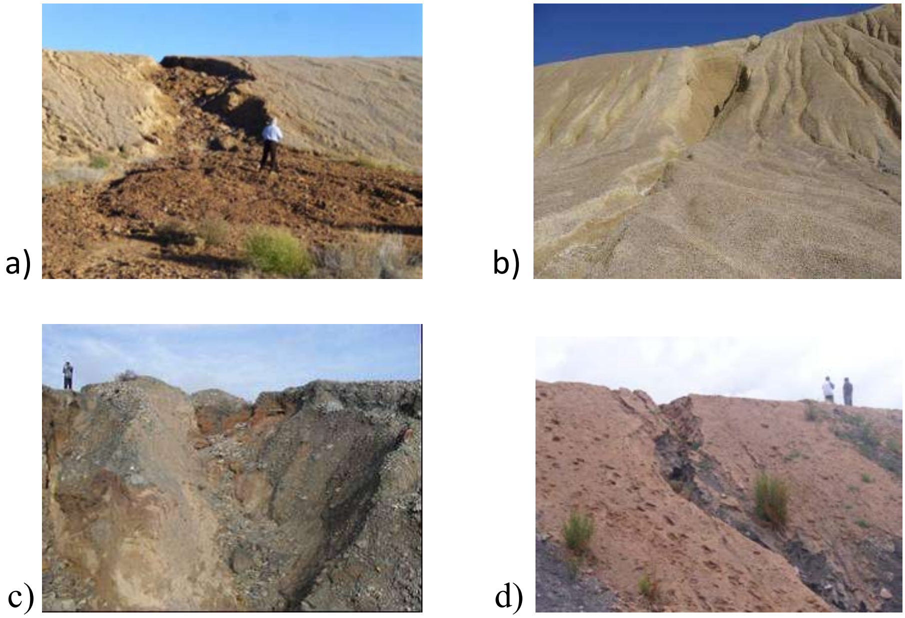

FIG. 9.3. Examples of erosion of waste rock piles located at uranium mine sites: (a) Vostochniy mine, Kazakhstan (2015); (b) barren ore waste pile at Taboshar, Tajikistan (2008); (c) Adrasman, Tajikistan (2010); (d) erosion of the protective cover and burned ash used in the past for uranium extraction at the Kadji-Say mine site, Kyrgyzstan (2006) (photographs taken by O. Voitsekhovych, Ukrainian Hydrometeorological Institute).

Typical examples of the erosion impacts caused by the water flows on the surface of waste rock piles, which are usually located close to the originating uranium mine site, are shown in Fig. 9.3.

The concentrations of uranium and other radionuclides of the uranium series in the eroded materials that are dispersed in the environment because of the impact of water flows usually correspond to their concentrations in the source material and can be quite high compared with the naturally occurring concentrations in the surrounding area. Typical remediation options are the construction of protective coatings that prevent erosion. The type of riprap, soil cover and vegetation need to be determined after consideration of the local climatic conditions.

9.1.2. Mill sites

9.1.2.1. Uranium production industrial areas

Former uranium milling facilities are usually complexes of buildings that include grinding and milling facilities, pipelines, and storage facilities for raw materials and final uranium ore concentrates and other products. During uranium production, such areas become contaminated because of seepages and dispersion of the raw uranium ore, contaminated dusts and other contaminated materials. Therefore, after cessation of uranium production and in the absence of appropriate decommissioning procedures and site decontamination actions, such areas become a significant hazard and a source of radionuclide dispersion by runoff and wind resuspension. Figure 9.4 shows examples from the Zaozerniy mine, Kazakhstan (Fig. 9.4(a)) and from Kadji-Say, near Issyk-Kul Lake, Kyrgyzstan, which is under remediation (Fig. 9.4(b)) [9.2]. Also shown are uranium ore raw materials of different size fractions (Fig. 9.4(c)) that were identified in the reloading and temporary storage area at the Base C facility and a typical view of the former uranium extraction facilities at the Prydniprovskyy Chemical Plant uranium legacy site, Ukraine, which requires cleanup and dismantling (Fig. 9.4(d)) [9.9, 9.14].

In many places, former industrial sites for uranium production have areas that were used for storage of raw materials or that have been significantly contaminated in the past during uranium ore processing. Such areas are usually characterized by elevated gamma dose rates of 10–100 μSv/h and higher, such as in the example from Ukraine given in Refs [9.9, 9.11, 9.14]. At some locations of the former Prydniprovskyy Chemical Plant uranium legacy site, the soils are mixed with fragments of dispersed particles of raw ores or uranium extraction residues of different size fractions, from several millimetres to several centimetres, which are difficult to separate from the soil matrix (Fig. 9.4(c)). The activity concentration of uranium and other radionuclides in the uranium–thorium decay series at this site varies in a wide range from <1 to 3000 Bq/g. The soils that are dominated by raw materials are usually characterized by uranium activity concentrations in equilibrium with other radionuclides in the uranium–thorium decay series [9.14].

In dry windy seasons, wind driven suspension may play a significant role in causing inhalation exposure of people. Thus, dust suppression actions and monitoring of workplaces are needed for such areas. Typical activity concentrations of ^{238}U and ^{226}Ra in aerosols collected in areas near different former uranium production facilities at the Prydniprovskyy Chemical Plant, Ukraine and other uranium legacy sites, such as at Chkalovsk, Tajikistan and at Kadji-Say, Kyrgyzstan, are given in Table 9.2 [9.2, 9.9].

In areas remote from uranium processing sites, activity concentrations of ^{238}U and ^{226}Ra in aerosols are determined by their background content in the surrounding soil and, as a rule, such activities are in the range of 0.01–0.03 mBq/m^3. However, data given in Table 9.2 show that uranium and radium activity concentrations in aerosols at former uranium mill sites or in areas adjacent to uranium processing facilities may exceed background levels by one or even two orders of magnitude. The activity concentrations of uranium and its progeny in aerosols can vary considerably depending on local conditions (e.g. wind speed, the state of the underlying surface, the moisture contents of soil cover and air, specific features of the contaminated sites). Reduction of inhalation exposure can be achieved by applying simple dust suppression measures or by using individual respiratory protection. Despite relatively high concentrations of radioactive substances in aerosols at sites of uranium production, inhalation

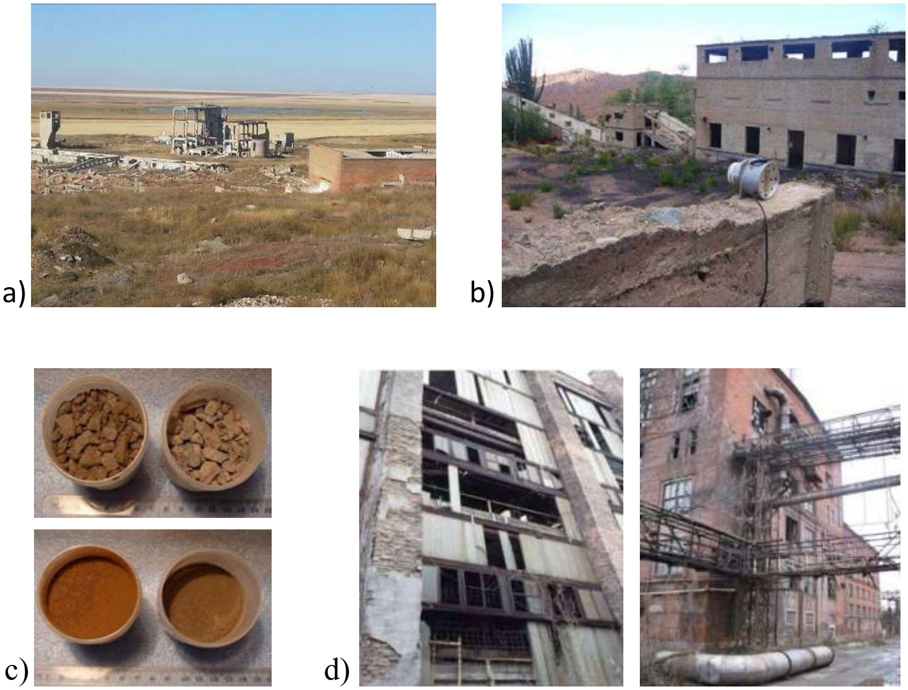

FIG. 9.4. Examples of the remaining infrastructure at former uranium mill sites: (a) former uranium mill site, Zaozerniy, Kazakhstan (2015); (b) former mill site, Kadji-Say, Kyrgyzstan (2007); (c) uranium raw materials sieved from the contaminated temporary raw material storage area at the former mill site, Prydniprovskyy Chemical Plant, Ukraine (2016); (d) current state of the former uranium extraction facilities at the Prydniprovskyy Chemical Plant (photographs taken by O. Voitsekhovych, Ukrainian Hydrometeorological Institute).

TABLE 9.2. TYPICAL ^{238}U AND ^{226}Ra ACTIVITY CONCENTRATIONS (mBq/m^3) IN AEROSOLS AT VARIOUS SITES

Radionuclide	Activity concentrations (mBq/m^3)				
	Prydniprovskyy Chemical Plant site near the uranium extraction facility, Ukraine [9.9]	Inhabited area near the Prydniprovskyy Chemical Plant, Ukraine [9.9]	Chkalovsk[a], Tajikistan (former uranium mill site) [9.2]	Chkalovsk[a], Tajikistan (town) [9.2]	Kadji-Say, Kyrgyzstan (after rain) [9.2]
^{238}U	0.05–0.30	0.01–0.02	0.13–0.18	0.01–0.05	0.02–0.04
^{226}Ra	0.10–0.15	0.01–0.03	0.04–0.06	0.03–0.05	0.02–0.03

[a] In 2016, the name of Chkalovsk town was changed to Buston.

exposures of workers and the public living in adjacent areas are relatively low compared with the potential doses that may occur owing to direct gamma radiation from contaminated facilities or materials at the mill sites containing highly contaminated residues from uranium extractions.

9.1.2.2. Tailings facilities

After uranium extraction, the residue materials contain only 5–15% of the uranium in the raw ore materials. Therefore, the radioactivity of the residues is mainly characterized by the progeny of uranium. The tailings facilities are usually located in areas adjacent to the uranium processing plants. The relative amounts and physicochemical speciation of uranium and other radionuclides in the tailings materials depend on the extraction methods used, as well as on the type and geochemistry of the uranium-containing ore materials and the geochemical conditions present in the environment. Typical examples of activity concentration of uranium and its progeny in tailings materials produced at hydrometallurgical plants in Tajikistan and Ukraine are shown in Table 9.3 [9.2, 9.11].

The main processes resulting in transport of uranium beyond the primary tailings site involve geochemical mechanisms resulting in its transformation from low oxidation states to more highly oxidized mobile forms, allowing its movement with water infiltrating into the underlying groundwater or its removal with drainage water through the dams associated with the tailings bodies.

Inundation of tailings material by water, especially in cases where the tailings facilities are located near rivers or close to river valleys, is common, and

TABLE 9.3. ACTIVITY CONCENTRATIONS OF NATURAL RADIONUCLIDES IN SAMPLES FROM URANIUM TAILINGS COLLECTED AT VARIOUS LOCATIONS

Sampling location	Activity concentrations (Bq/g)					
	^{238}U	^{226}Ra	^{230}Th	^{210}Pb	^{210}Po	Reference
Tailings, Degmai, Tajikistan	0.82–0.98	0.72–0.76	11.2–15.6	10.1–14.6	12.3–13.2	[9.2]
Tailings, Taboshar, Tajikistan	0.58–1.40	3.10–6.57	2.90–5.60	3.89–5.89	3.25–5.25	[9.2]
Tailings, Zapadnoe, Ukraine	1.50–3.25	1.54–16.2	1.60–34.8	1.52–15.2	1.60–15.1	[9.11]
Tailings, Tsentralnyy Yar, Ukraine	2.20–7.80	4.0–130	2.44–240	4.70–120	4.80–125	[9.11]

Note: Measurement uncertainties are estimated as 7–12%.

described in many technical field inspection reports. Examples of pore water seepage from a tailings body with a high content of uranium are shown in Fig. 9.5 relating to tailings pile No. 5, located on the right bank of the Mailuu-Suu river, Kyrgyzstan [9.13]. According to Ref. [9.13], in 2012, the total uranium activity concentrations (^{238}U and ^{234}U) in the drainage and seepage water from this tailings pile that entered the Mailuu-Suu river were in the range of 44–140 Bq/L (1610–6150 μg/L). The concentration of uranium in the river upstream and downstream of the outlet locations of seepage waters varied from 0.05–0.1 to 0.15–0.30 Bq/L. The water discharge of the river during the period of observation varied in the range of 20–55 m^3/s. These data indicate a relatively constant input of highly contaminated water from the tailings into the river. Despite the relatively small flow rate of the contaminated tailings water inlet, the very high activity concentration of uranium in the inlet water means that its impact on the contamination of the river is significant.

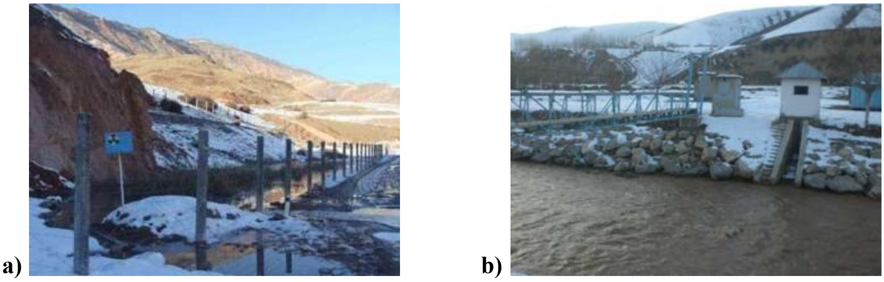

FIG. 9.5. (a) Groundwater seepage from tailings body No. 5 into the Mailuu-Suu river, Kyrgyzstan (2012); (b) monitoring sampling point on the Mailuu-Suu river, Kyrgyzstan (2011) (photographs taken by O. Voitsekhovych, Ukrainian Hydrometeorological Institute).

Seepage of interstitial water from a tailings body is quite common. Such water has a high content of uranium and usually has high mineralization and acidity. Increased acidity of interstitial water (pH 2–4) might be due to the high content of acid (sulphuric or other) in the residues of uranium production in the tailings material. Also, the natural processes of oxidation in the tailings material, for example, in association with the presence of pyrite, may generate solutions with low concentrations of sulphuric acid that stimulate leaching processes, increasing the mobility of uranium and its transport into groundwater.

Seepage waters with high mineralization and relatively high concentrations of uranium that are released into the environment and oxidized in a stream or creek can form salt deposits (e.g. based on sodium sulphate or other types of precipitate). Under certain conditions, such deposits can be crystallized, with high concentrations of uranium precipitates having a bright yellow colour with relatively high activity concentrations (up to 20 Bq/g) and high concentrations in the dissolved phase, such as those that were reported in Refs [9.2, 9.7] in the range of 50–80 mg/L (Fig. 9.6). Spring snowmelt waters or rain lead to dissolution of these soluble precipitates and transport contaminated waters into rivers. This mechanism of uranium removal from a tailings body into a river is rather common in the mountainous areas of Central Asia.

Physical processes such as unstable geodynamics, landslides, mudflows and other geotechnical failures can lead to direct transport of the tailings materials into the surrounding area. The possible consequences of such events are described in Ref. [9.10]. There are two main factors responsible for mass transport: (i) burrowing animals (Fig. 9.7(a)), which may significantly degrade the protective covers of tailings; and (ii) mudflows, which are able to transport contaminated materials large distances from the tailings (Fig. 9.7(b)).

Figure 9.7(b) shows the effect of a mudflow event in transporting contaminated materials from the tailings located at the Taboshar mill site, Tajikistan over distances of about 3.5 km. Red coloured tailings materials identified on the bottom of the local temporary stream created relatively highly contaminated areas with gamma dose rates in the range of 2–3 μSv/h (i.e. at least 20 times higher than the local gamma dose rate background).

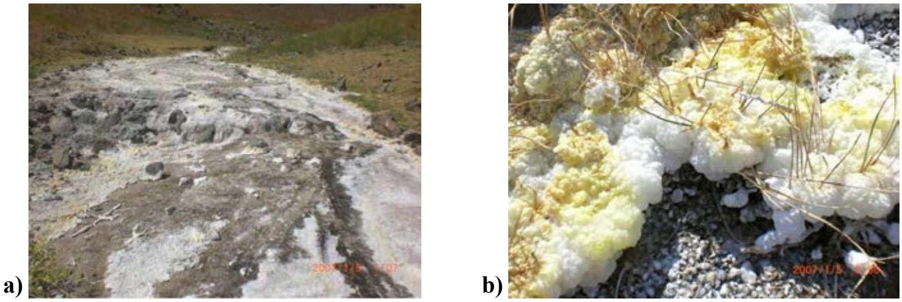

FIG. 9.6. (a) Acidic seepage water from the tailings body; (b) precipitate residues of sodium sulphate with uranium incorporated (photographs from Taboshar, Tajikistan, taken by O. Voitsekhovych, Ukrainian Hydrometeorological Institute).

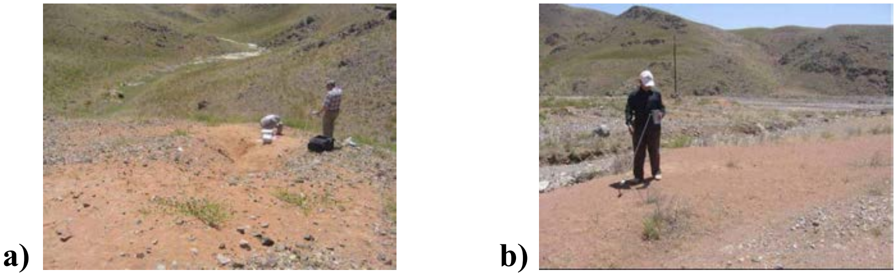

FIG. 9.7. Transport of the tailings materials to surrounding areas by burrowing animals (a) and mudflows (b). The ‘red’ materials, containing radionuclides in the uranium–thorium series from tailings in Taboshar, Tajikistan, have been transported distances of up to 3.5 km by mudflows along the Sarym-Sahly Say creek [9.2] (photographs taken in 2006 by O. Voitsekhovych, Ukrainian Hydrometeorological Institute).

9.2. URANIUM EXTRACTION FACILITIES

Acidic or sodium based processes are most frequently used for destruction of the matrices of raw materials and for sequential extraction in the industrial production of uranium. If strict regulations and adequate radiation protection are applied, radioactive contamination at the uranium production site is usually rather limited and radiation exposures are well controlled. Furthermore, in the case of planned closures of uranium production facilities and with well designed and executed decommissioning and post-closure remediation, the uranium industry does not create significant negative impacts on human health or on the environment.

However, in some cases, the decommissioning of mills and extraction facilities has been performed inadequately, and radioactively contaminated materials have been dispersed beyond the working environment and outside uranium production areas.

Examples of such negative scenarios of inadequate uranium production site safety management after the closure of an enterprise occurred around the Prydniprovskyy Chemical Plant, Ukraine, which was one of the largest facilities of the military complex of the former Soviet Union, where production of uranium for the atomic programme started in 1949 and ceased in 1992. The highest levels of radioactive contamination can be found inside the uranium processing buildings, especially in the extraction areas. Dose rates of up to 1–4 mSv/h have been measured in some locations. In many places, remaining production materials such as yellow cake and other residues containing uranium and other radionuclides in the uranium decay series have been identified (Fig. 9.8). Outdoors, radioactive contamination is present over a large part of the area of the former Prydniprovskyy Chemical Plant uranium production facility [9.9, 9.14].

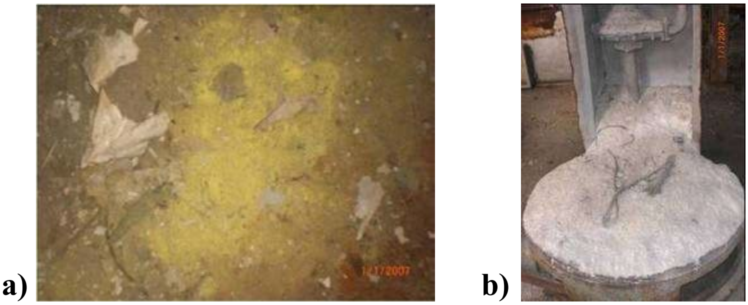

FIG. 9.8. Examples of uranium residues with a high content of uranium and other naturally occurring radionuclides in the workshops and extraction plants at the Prydniprovskyy Chemical Plant, Ukraine (photographs taken by O. Voitsekhovych, Ukrainian Hydrometeorological Institute).

Typical levels of uranium and other radionuclides in the residues from uranium production in these areas are presented in Ref. [9.14] and summarized in Table 9.4 [9.15]. The high concentrations of uranium in mineral residues identified in the area of the Prydniprovskyy Chemical Plant (on the equipment and dispersed in the former uranium production workshops) can give rise to high concentrations of radioactive substances in dust and aerosols, increasing the radiological risks from inhalation exposure.

The maximum activity concentrations in aerosols determined in monitoring programmes in the uranium extraction facilities at the Prydniprovskyy Chemical Plant legacy site were in the range of 3.1–73.4 mBq/m^3 (i.e. one to two orders of magnitude higher than those determined outside the facility area).

The most reasonable option for remediating such highly contaminated facilities is decontamination using one of the methods described in Chapter 8, supported by strict radiation protection actions and adequate radioactive waste management. If remediation safety criteria are not achieved, the former uranium production facilities need to be demolished and contaminated wastes removed from the remediated site.

TABLE 9.4. RADIONUCLIDE ACTIVITY CONCENTRATIONS IN RAW MATERIAL SPILLS FOUND IN THE URANIUM EXTRACTION AND THORIUM REMOVAL FACILITIES AT THE PRYDNIPROVSKYY CHEMICAL PLANT, UKRAINE

Type of material	Activity concentration (Bq/g)[a]			
	^{238}U	^{226}Ra	^{210}Pb	^{230}Th
Raw ore material	8.9–46.5	8.7–45.5	8.5–44.5	8.5–43.0
Raw ore material	75–100	80–110	75–100	65–90
Spill on the floor before extraction	180–360	79.0–165	17.1–40.5	27–80
Spill of yellow cake (Building 103)	1000–3200	1.5–2.5	1.5–2.5	—[b]
Spill in the thorium removal facility	0.4–3.5	7.0–34.0	9.1–34.0	45.0–3000
Sludge after thorium removal	2.21–14.1	1.70–51.5	2.60–49.5	15.2–240

[a] Data from Ref. [9.15].

[b] —: data not available.

9.2.1. Conclusions

The brief analyses given in the preceding sections demonstrate just some examples of the radiological hazards that may be relevant at legacy sites of former uranium production facilities, comprising a variety of sources, forms and possible exposure pathways. Radiological risks related to uranium legacy sites may be significant for the people living in the surrounding areas and for the environment. The presence of uranium on the environment can entail radiological and toxicological risks. Remediation actions may reduce negative effects on the environment and reduce human exposure, and they need to be case and site specific, well justified and optimized. However, a main constraint for legacy site remediation is funding.

The IAEA provides support to help Member States to ensure that appropriate regulatory systems and safety standards are in place to manage the safe and secure development of all uranium resources. The industry of conventional uranium production has an interest in supporting the success of this process, while ensuring that the remediation of legacy sites is not forgotten and no new legacy sites are created.

9.3. URANIUM IN OTHER METAL MINING AND PROCESSING INDUSTRIES

Uranium and also often thorium occur in nature with other elements in polymetallic ore deposits. This is the case, for example, for heavy mineral sand deposits (monazite), tantalite and columbite, and even for ore deposits of gold, copper, chromium and diamonds. This occurrence is often not mentioned but must be seen from two perspectives. The first is as potential additional sources of uranium for nuclear energy, the second as a neglected source of radiation exposure and environmental contamination. Occasionally, the uranium grade in the metallic ore justifies economic extraction of the uranium, but often its presence is neglected in the metal exploitation, so mineral processing may pose unperceived occupational health risks and waste disposal may require radiation safety measures.

One group of metals that has deserved increased attention is the REEs, which comprise 15 lanthanides, scandium and yttrium (thorium is sometimes included in this group as well). These elements have found application in many recent and fast growing industries such as electronics, magnets and renewable energy. These 17 elements not only are associated with each other in geological deposits, but also have a close association with actinide elements, notably uranium and thorium, which means that mining and processing them may give rise to significant radiation protection and environmental issues [9.16].

There are more than 200 minerals containing REEs, such as monazite, bastnäsite, xenotime and samarskite, but only a few are currently considered of significant economic interest. All contain variable concentrations of radionuclides in the uranium and thorium decay chains (Table 9.5) [9.17].

China is the main producer and has the largest world reserves of REEs (about 40% of the total). In China, the main REE production takes place at the Bayan-Obo mine in Inner Mongolia, which contains large amounts of lanthanum, cerium and neodymium, and has an unreported uranium and thorium content [9.18]. The general association of REEs and actinides requires the exercise of significant care in the extraction, transport and processing of REE minerals owing to the radiation protection issues that arise. It has been reported, for example, that heavy mineral sands processed for extraction of zirconium, ilmenite, rutile and garnet may give rise to enhanced radiation doses in the processing facilities, specifically in magnetic separation, mineral storage and mineral transport [9.19].

In some workplaces at mineral segregation plants processing heavy mineral sands, the external dose rate was reported to be 20–40 μSv/h. According to Ref. [9.20], the highest annual radiation exposure of a worker involved in the transport of mineral sands in Australia was estimated at about 600 μSv/h. This increased environmental effective dose rates from the local background value of 0.2 μSv/h by two to three orders of magnitude and, in many cases, it was related to the increased activity concentrations of uranium and thorium in the separated mineral products.

According to some recent studies in France, natural sands in some beach areas are significantly enriched with heavy minerals with activity

TABLE 9.5. CONCENTRATIONS OF URANIUM AND THORIUM IN THE MOST COMMON RARE EARTH ELEMENT MINERALS IN INDIA [9.17]

Mineral		Concentration (wt%)	
		Uranium (U_3O_8)	Thorium (ThO_2)
Bastnäsite	$(Ce,La)CO_3F$	Negligible	0.1–0.2
Monazite	$(Ce,La,Pr,Nd,Th,Y)PO_4$	0.2–0.4	4.5–9.5
Xenotime	$Y(PO_4)$	0.81	0.83

concentrations of ^{238}U in the range of 95–2240 Bq/kg and of ^{232}Th in range of 135–3470 Bq/kg [9.21]. In some beach sand areas of Brazil containing heavy minerals [9.22], activity concentrations of ^{238}U and ^{232}Th were estimated to be two to five times higher than was found in natural sand dunes in France. Differences in the content of uranium and thorium in the sands are determined by the type and characteristics of the minerals in the vertical profiles accumulated at the sites, because uranium and thorium concentrate in heavy minerals such as monazite and zircon [9.22]. Table 9.6 gives some typical activity concentrations for ^{238}U and ^{232}Th in some mineral sands [9.22].

Underground REE mines may pose serious radiation protection issues owing to the buildup of radon isotopes in the atmosphere of the mine and may require the development of occupational and health and safety methods, such as those developed for the Thor Lake mining project (Eco Ridge Project) in Elliot Lake, Canada [9.23]. This type of underground mining may require special personal protective equipment (respiratory protection), as well as special mine ventilation systems, ore transport conveyor belts and mine backfilling techniques [9.23]. Waste and process water from these mining and processing industries may also pose serious environmental threats and require suitable management plans.

Tantalite is another mineral displaying a strong association with uranium and thorium. Tantalite mining in open cut mines has been reported to generate radiation exposures from negligible (i.e. close to the natural radiation background) to exceedingly high dose rates, at around 20 μSv/h [9.24]. Dose rates depend on the workplace and are higher near storage of tantalite concentrates and drums containing tantalite for transport. For comparison, radiation dose

TABLE 9.6. ACTIVITY CONCENTRATIONS OF ^{238}U AND ^{232}Th IN HEAVY MINERAL SANDS [9.22]

Mineral	Activity concentration (Bq/kg)	
	^{238}U	^{232}Th
Raw sand	2.7–7.9	6.5–9.4
Ilmenite (different types)	564–1 918	1 348–1 863
Zircon (different types)	5 462–19 286	1 016–7 162
Kyanite (different types)	167–1 331	55–505
Rutile (different types)	1 249–1 737	420–738

rates from drums containing yellow cake (U_3O_8) for transport are only a few microsieverts per hour.

Another step at which radionuclide dispersal may occur is the plant site where processing of REEs and tantalite concentrates takes place. Industrial methods vary from acid dissolution of the mineral to separation of uranium and thorium by precipitation, ion exchange or other chemical procedures. Alternatively, the mineral may be processed by pyrometallurgical methods to separate uranium and thorium from the REEs. However, little information on radiation exposures is available. Assuming an average concentration of 0.4% uranium in REE ores, for a world production of about 100 000 t in 2015, 400 t of uranium could have been produced from this secondary source. Thus, the processing of these minerals may release significant amounts of radioactivity into the environment surrounding the facilities.

Among the non-conventional sources of uranium, besides phosphate minerals (considered in Section 9.4), is gold mining. Gold mining, for example in Withwatersrand, South Africa, generated milling tailings with 50–70 ppm uranium which, depending on uranium prices, have, on occasion, been exploited [9.25]. The same has been done at Cluff Lake mine, Canada [9.26].

Lignite is known to contain uranium at concentrations of up to several hundred parts per million. Radioactivity in emissions from coal fired power plants has been given attention, and the potential recovery of uranium from the coal or from coal ash has been tested. For several years, uranium was recovered in North Dakota, USA by processing lignite ashes that contained 6–8 kg U_3O_8/t. Studies were carried out on coal ash in other countries, highlighting that the thermal energy potentially available from the uranium in the coal was higher than the thermal energy obtained from coal burning [9.27, 9.28].

From the perspective of the enhanced radioactivity and radiation exposure in these non-uranium mining and processing industries, there have been significant conceptual and regulatory developments, especially with the adoption of IAEA Safety Standards Series No. GSR Part 3, Radiation Protection and Safety of Radiation Sources: International Basic Safety Standards (GSR Part 3) [9.29]. In parallel, specific publications dealing with exposure and radiation protection in various naturally occurring radioactive material industries have been published (i.e. Ref. [9.30]).

9.4. URANIUM IN PHOSPHATE MINERALS

From the late nineteenth century to the present, the use of phosphate rock as agricultural fertilizer has increased tremendously in close correlation with human population growth and food production (Fig. 9.9). World phosphate

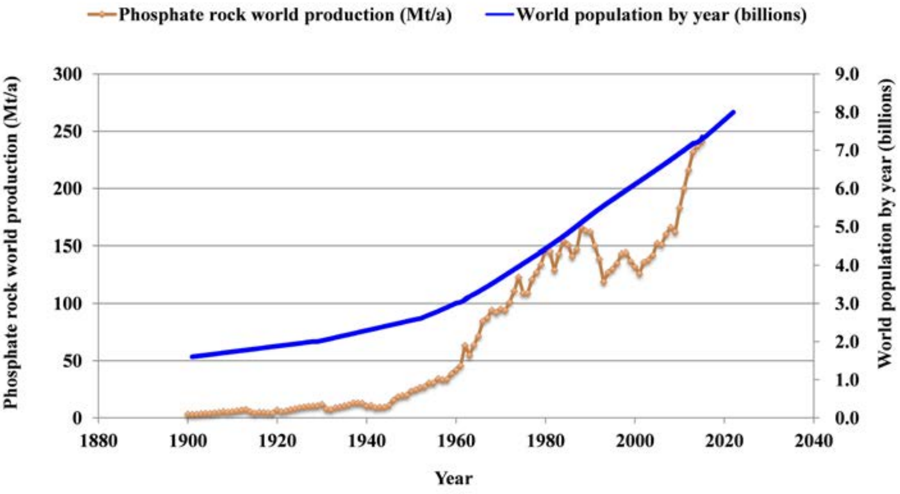

FIG. 9.9. World production of phosphate rock (data from Ref. [9.31] for the period 1900–2015) and world population growth over the past century [9.32] (data interpolation applied between 1901–1927).

rock production for 2015 was over >240 Mt/a. The use of phosphate fertilizers is likely to continue at a similar rate in the foreseeable future to support agricultural production.

The main world reserves of phosphate are located in the Western Sahara, Morocco, but important reserves also exist in North America and Asia. The state of Florida, USA and several countries around the Mediterranean basin were the main phosphate rock producers in the twentieth century, whereas the main producers of fertilizers have been China, Europe and the USA [9.33]. In the past decade, major changes have occurred in the world trade of phosphate rock following a European Union shift to the import of phosphoric acid instead of raw phosphate rock.

Phosphate rock contains varying quantities of uranium depending on the nature of the geological deposits. Uranium concentrations have been reported to range from 20 to 2000 µg/g or, in activity units, from about 0.01 to 1.6 Bq/g [U] [9.30]. Other metals, such as cadmium, arsenic and mercury, are also often present in phosphate raw materials. Reference [9.30] reports:

> "Most commercially exploited sedimentary phosphate deposits are located within a few tens of metres of the earth's surface. These deposits are usually mined by opencast techniques…In the mining of igneous deposits and some

> sedimentary deposits, the overburden and/or phosphate ore has to be broken up by drilling and blasting before it can be removed…In some operations the mined ore is exposed to high pressure water jets…This generates a slurry, which is pumped directly to the…[processing] plant".

In other cases, with hard rock operations, the ore is first crushed and then transported to the reprocessing plant by a slurry pipeline [9.30].

Reference [9.30] reports that radionuclide substitutions in the apatite structure of phosphate ore may include such cationic forms as U^{4+}, U^{6+}, Th^{4+}, Ra^{2+}, Pb^{4+}, Pb^{2+}, Po^{4+} and Bi^{3+}. It is assumed that U^{4+} substitutes for Ca^{2+} in the apatite structure because these two ions have almost identical ionic radii. The ^{238}U activity concentrations in phosphate ores are typically in the range of 0.1–0.5 to 1–5 Bq/g. These ^{238}U concentrations apply to ores of both sedimentary and igneous origin, and the uranium decay chains are generally in radioactive equilibrium.

Production of phosphoric acid from phosphate rock may be achieved through several industrial processes, but the hemihydrate process using sulphuric acid is the most common in its production for the phosphate fertilizer industry (see Ref. [9.30]). Elemental phosphorus is produced by a thermal process. During phosphate processing at fertilizer plants, fluorides need to be controlled as gases and in effluents (hydrofluoric acid and fluorosilicic acid), and about half of the fluorine partitions with the gypsum. In the process, a great deal of crud is generated, and this has generally been disposed of with phosphogypsum tailings, despite its low radionuclide content. Phosphates have a wide variety of applications in chemical industries (producing phosphoric acids and other phosphorus-containing goods) and in agriculture (for production of fertilizers and animal feeds), as well as for other uses (e.g. as phosphogypsum in the construction industry, municipal and other sectors), as illustrated in Fig. 9.10. However, their application can be constrained by the need to consider radiological impacts, because phosphate-containing products are usually associated with significant amounts of naturally occurring radionuclides.

Phosphate mineral fertilizers, mixed organic–mineral fertilizers and other derived products, such as cattle and chicken feed, are produced from phosphoric acid. In these processes, the uranium and radioactive progeny initially present in phosphate rock are distributed among phosphoric acid, fertilizers and phosphogypsum, which is the main by-product. Uranium goes partly into the fertilizer and partly into the phosphogypsum, whereas other radionuclides, such as radioisotopes of radium, lead and polonium, mainly go into the phosphogypsum by-product (Table 9.7).

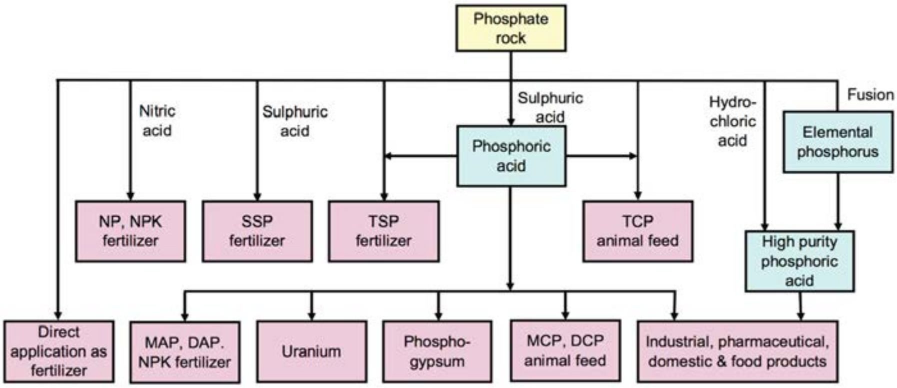

FIG. 9.10. Phosphate rock product streams (DAP: diammonium phosphate; DCP: dicalcium phosphate; MAP: monoammonium phosphate; MCP: monocalcium phosphate; NP: nitrogen–phosphorus; NPK: nitrogen–phosphorus–potassium; SSP: single superphosphate; TCP: tricalcium phosphate; TSP: triple superphosphate) (adapted from Ref. [9.30]).

TABLE 9.7. RADIONUCLIDE ACTIVITY CONCENTRATIONS IN PHOSPHATE MATERIALS FROM SENEGAL [9.34]

Material	Activity concentrations (Bq/kg)			
	^{238}U	^{226}Ra	^{228}Ra	^{40}K
Soil from the Sahara region	<70	16	9.6	13
Phosphate rock	1890	1230	14	<22
Superphosphate fertilizer (18% P_2O_5)	630	600	—[a]	<19
Phosphogypsum	320	860	3	<19

[a] —: no data available.

The equipment and pipelines used in the phosphate industry, in some cases, may be contaminated by scale having relatively high concentrations of uranium–thorium decay series radionuclides. Reference [9.30] reports:

> “Radionuclide activity concentrations in many of the process materials may exceed 1 Bq/g, with values sometimes reaching a few becquerels per gram in bulk materials (phosphate rock feedstock, phosphoric acid and

> phosphogypsum) and, occasionally, a few thousand becquerels per gram in the scale that accumulates inside pipes, vessels and filtration equipment. In terms of the Standards, therefore, wet process phosphoric acid production needs to be considered for regulation as a practice."

In phosphoric acid production plants, concentrations of uranium series radionuclides (e.g. in acid/phosphogypsum separation filters) may be enhanced and give rise to elevated occupational radiation exposures of 2–3 mSv/a. Some structures in the facilities, such as pumps and pipes used to transfer phosphoric acid, accumulate incrustations (scale) rich in ^{226}Ra and radium progeny that give rise to ambient dose rates of around 10 μSv/h in some workplaces.

An important change in the way occupational radiation protection in naturally occurring radioactive material industries is addressed was introduced by GSR Part 3 [9.29] and by European Union Council Directive 2013/59 [9.35]. The new guidelines introduced by GSR Part 3 [9.29] force attention to be paid to naturally occurring radionuclides in non-nuclear industries such as the phosphate industries.

The presence of uranium and uranium progeny in phosphorite poses both occupational radiation protection issues and environmental radiological protection issues. Many facilities producing phosphoric acid have released phosphogypsum directly into the sea for decades. Coastal locations in France, Portugal, Spain, Tunisia and the United Kingdom, among others, received those discharges and the phosphogypsum partly accumulated on the sea floor and partly dissolved in sea water. Radioactivity impact assessments in some areas led to the conclusion that enhancements of radionuclide concentrations in the coastal sea had occurred, with increases of concentrations in sediments, accumulation in molluscs and crustaceans, and transfers of radionuclides to consumers of local seafood. For example, the results of a dose assessment in the United Kingdom led to the computation of an annual effective dose of 6 mSv to members of the public through consumption of local snails and seaweeds [9.36, 9.37]. In other coastal areas, such as the location of outflow of the Scheldt river, the discharge of residues from thermal phosphor production resulted in contamination of polders in the Netherlands by ^{226}Ra, ^{210}Pb and ^{210}Po highly exceeding the natural background [9.38]. In other locations, strong radionuclide enhancement was reported in estuarine sediments (e.g. in Baie de Seine in France, the Tagus river in Portugal, Huelva in Spain).

In many countries, stockpiling of phosphogypsum on land was adopted instead of discharging it into the sea. With the closure of phosphoric acid production plants, phosphogypsum stockpiles were abandoned and a final solution has been pending. In some cases, phosphogypsum piles and former industrial infrastructures were closed, and sites remediated. In Cyprus and

Tunisia, for example, phosphogypsum stockpiles were recontoured, re-covered with soil, consolidated and integrated into the landscape with success (Taparura project, Tunisia) [9.39].

Owing to the large amount of phosphogypsum, in many countries, there have been attempts to utilize the material, rather than to accept it as a final residue. Several countries have tried to use it in applications such as road pavement, construction materials, cement additives or plaster [9.30]. The Stack Free initiative, promoted by the Florida Industrial and Phosphate Research Institute, USA, encourages the use of phosphogypsum (e.g. as a soil amendment) to consume the stockpiles that exist around the world, with the obvious advantage that phosphogypsum still contains some phosphate usable by plants and there would be no environmental liability to industry for residual waste.

However, the rise of uranium prices in 2007 dictated another path to the use of phosphogypsum. Instead of considering it a waste, phosphogypsum is now regarded as a possible non-conventional uranium source [9.40]. Chemical procedures have been developed to extract uranium from phosphate rock and these are becoming competitive. Currently, the best use of phosphogypsum and its final environmental fate remain undecided.

In the future, phosphate fertilizers and other phosphate derived products might be cleaner, and indeed there have been processes developed to remove cadmium and uranium from phosphoric acid. Cleaner production of fertilizers might be desirable, but it is also costlier and this is the reason why some of these procedures have not been implemented.

Use of phosphate materials in agriculture adds cadmium, uranium and other metals to agricultural fields and may poison soils. It was computed that regular addition of phosphate fertilizers may increase by up to 5% the concentration of uranium in top soils. The increase in uranium might not be a radiological protection issue, but the addition of ^{226}Ra and ^{226}Ra ingrown from uranium may increase radiation doses to humans through the food chain in the future, as ^{226}Ra is more water soluble and bioavailable to plants. This long term aspect has not yet been thoroughly investigated.

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ABBREVIATIONS

AMS	accelerator mass spectrometry
ATSDR	Agency for Toxic Substances and Disease Registry (USA)
BMCL	benchmark concentration lower bound
BMDL	benchmark dose lower bound
DW	dry weight
EC_{20}	effective concentration affecting 20% of the exposed population
Eh	reduction–oxidation potential
EQS	environmental quality standard
FW	fresh weight
HHB	human health benchmark
ICP-MS	inductively coupled plasma mass spectrometry
ICRP	International Commission on Radiological Protection
KPA	kinetic phosphorescence analysis
LA-ICP-MS	laser ablation inductively coupled plasma mass spectrometry
LOAEL	lowest observed adverse effect level
LSC	liquid scintillation counting
MRL	minimal risk level
NEMI	National Environment Methods Index database (USA)
NOAEL	no observed adverse effect level
PERALS	photon–electron rejecting alpha scintillation system
RAPs	representative animals and plants
SQG	soil quality guideline
TDI	tolerable daily intake
TF	transfer factor
ULS	uranium legacy site
UNSCEAR	United Nations Scientific Committee on the Effects of Atomic Radiation

CONTRIBUTORS TO DRAFTING AND REVIEW

Carvalho, F.P.	University of Lisbon, Portugal
Fesenko, S.	International Atomic Energy Agency
Harbottle, A.R.	International Atomic Energy Agency
Lavrova, T.	Ukrainian Hydrometeorological Institute, Ukraine
Lee Zhi Yi, A.	International Atomic Energy Agency
Mitchell, N.G.	Eden Nuclear and Environment Ltd, United Kingdom
Payne, T.E.	Australian Nuclear Science and Technology Organisation, Australia
Rigol, A.	University of Barcelona, Spain
Thorne, M.C.	Mike Thorne and Associates Ltd, United Kingdom
Ulanowski, A.	International Atomic Energy Agency
Vidal, M.	University of Barcelona, Spain
Voitsekhovych, O.	Ukrainian Hydrometeorological Institute, Ukraine
West, J.M.	West Consult, United Kingdom
Yankovich, T.	International Atomic Energy Agency

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