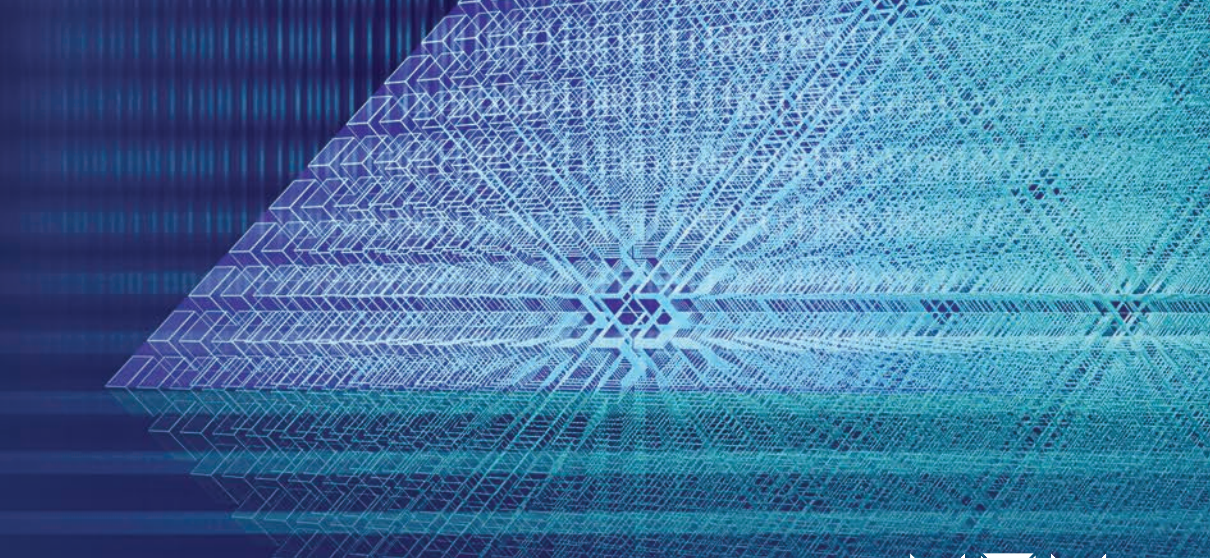

TECHNICAL REPORTS SERIES NO. 484

The Environmental Behaviour of Polonium

F. Carvalho, S. Fernandes, S. Fesenko, E. Holm, B. Howard,
P. Martin, M. Phaneuf, D. Porcelli, G. Pröhl, J. Twining

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THE ENVIRONMENTAL BEHAVIOUR OF POLONIUM

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INTERNATIONAL ATOMIC ENERGY AGENCY
VIENNA, 2017

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PO Box 100
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tel.: +43 1 2600 22417
email: sales.publications@iaea.org
http://www.iaea.org/books

© IAEA, 2017

Printed by the IAEA in Austria
November 2017
STI/DOC/010/484

IAEA Library Cataloguing in Publication Data

Names: International Atomic Energy Agency.
Title: The environmental behaviour of polonium / International Atomic Energy Agency.
Description: Vienna : International Atomic Energy Agency, 2017. | Series: Technical reports series (International Atomic Energy Agency), ISSN 0074–1914 ; no. 484 | Includes bibliographical references.
Identifiers: IAEAL 17-01115 | ISBN 978–92–0–112116–5 (paperback : alk. paper)
Subjects: LCSH: Radioecology. | Radioactive pollution. | Polonium.
Classification: UDC 539.163 | STI/DOC/010/484

FOREWORD

Polonium-210 is the main contributor to internal doses due to ingestion of radionuclides from the uranium and thorium decay series. With the expected increase in uranium mining and other industries generating naturally occurring radioactive material residues in the future, it is likely that the radiological impact of ^{210}Po will increase in importance.

The IAEA attaches great importance to the dissemination of information that can assist Member States with the implementation and improvement of activities relating to radiation safety, including the management of radioactive residues containing natural radionuclides. This publication outlines the behaviour of polonium in air, water and soil, and in the human body. The primary objective is to provide information that can be used in radiological assessments of accidental releases and routine discharges of polonium to the environment. Case studies and environmental applications of polonium isotopes are also presented.

The IAEA wishes to express its appreciation of the work of all the contributors to the drafting and review of this publication. The IAEA officers responsible for this publication were S. Fesenko and M. Phaneuf of the IAEA Environment Laboratories.

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Chapter 1

INTRODUCTION

1.1. BACKGROUND

For many years, the IAEA has supported efforts to develop publications on the environmental behaviour and use of radionuclides in response to the needs of its Member States. Since ^{210}Po is the main contributor to internal doses due to ingestion of radionuclides from the uranium and thorium decay series [1.1, 1.2], it was determined that a resource publication for polonium would be beneficial. Polonium-210 is an alpha emitting radionuclide with no radioactive progeny and produces only very-low-intensity gamma rays at very low abundance; thus, the dose largely arises from internal exposure. The main reasons for its radiological importance are its relatively high activity concentrations in certain foods and its relatively high ingestion dose coefficient.

There are seven polonium isotopes naturally present in the environment: ^{210}Po, ^{214}Po and ^{218}Po of the uranium decay series; ^{212}Po and ^{216}Po of the thorium decay series; and ^{211}Po and ^{215}Po of the actinium decay series. Polonium-210 has a half-life of approximately 138 days, which is long enough to play a significant role in many environmental processes. All of the other naturally occurring isotopes have half-lives of only 3 minutes or less. The isotopes ^{214}Po and ^{218}Po are short lived progeny of ^{222}Rn, and are important in dose assessment. However, they are best examined in relation to the environmental behaviour of radon and other short lived radon progeny. Hence, the primary focus of this publication is the environmental behaviour of ^{210}Po.

Radiation doses from ^{210}Po arise owing to natural occurrences of the radionuclide as well as to human activities. As ^{210}Po is part of the uranium decay series, it is naturally occurring and is found in varying amounts worldwide. The anthropogenic sources include uranium mining and milling activities, as well as the production of naturally occurring radioactive material (NORM), such as phosphogypsum, and oil and gas scales. With uranium mining and other industries generating NORM residues expected to grow in the future, it is likely that the radiological impact of ^{210}Po will be of increasing importance. The challenges faced have been addressed in a number of IAEA publications on radionuclide transfer in the environment, which focus on radionuclide transfers in terrestrial, freshwater and marine environments, and provide information on key transfer processes, concepts and models important in radiological assessments for all radionuclides (see Refs [1.3–1.8]).

Since 2004, the IAEA has also organized a series of projects aimed at improving environmental assessment and remediation. Through these projects, the environmental behaviour of radionuclides has been presented in other IAEA publications, which cover the following topics:

- Contaminated site characterization [1.9–1.11];
- Waste discharge, surveillance and management [1.12–1.14];
- Environmental remediation [1.15, 1.16];
- Remediation of uranium mill tailings [1.17];
- Remediation of dispersed contamination [1.18];
- Radiation protection and the management of radioactive waste in the oil and gas industry [1.19].

1.2. OBJECTIVE

This publication provides information on the environmental behaviour of polonium, which can be used in the radiological assessment of polonium occurring naturally in the environment or present due to routine discharges and accidental releases. This publication can support environmental investigations, assessments, and remediation in areas contaminated by polonium, and presents concepts, models and parameters describing the behaviour of polonium.

1.3. SCOPE

This publication explores the behaviour of polonium in atmospheric, terrestrial, freshwater and marine environments. The primary focus is the environmental behaviour of ^{210}Po, the only polonium isotope with a half-life long enough to play a significant role in environmental processes but, at 138 days, also short enough that ingrowth from its immediate progenitors ^{210}Bi and ^{210}Pb can be important when studying polonium's behaviour and for inferring kinetics and mechanisms of the processes. Information on the progenitors of ^{210}Po — in particular ^{210}Pb — is also presented to capture ^{210}Po behaviour in the environment.

In addition to presenting information on the environmental transfer of radionuclides to humans and non-human biota, this publication provides further information on the assessment of the impact of radioactive discharges (e.g. see Refs [1.12, 1.20]. Applications of ^{210}Po as a tracer of environmental processes are discussed, but industrial, military and other applications of ^{210}Po, such as occupational dose assessment, are outside the scope of this publication.

1.4. STRUCTURE

Chapter 2 introduces basic definitions and concepts for radioecology. The physical, chemical and decay properties of polonium are provided in Chapter 3, and Chapter 4 addresses sources and cycling of ^{210}Pb and ^{210}Po in the environment. Chapters 5–8 explore the behaviour of ^{210}Pb and ^{210}Po in the atmospheric, terrestrial, freshwater (including groundwater) and marine environments, respectively. Chapter 9 describes the metabolism of polonium in the human body. Radiological dose assessment models for humans and wildlife relating to the occurrence of polonium in the environment are given in Chapter 10. Appendices I–VII consider various case studies and environmental applications of polonium.

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Chapter 2

RADIOECOLOGY: BASIC DEFINITIONS AND CONCEPTS

Several generic parameters and concepts are used throughout this publication to characterize the environmental behaviour of polonium in various media. They are defined here, generally according to the recommendations of the International Commission on Radiation Units and Measurements [2.1], and follow IAEA publications on radionuclide transfers in different environments (see Refs [2.2, 2.3]). This publication has the advantage that the relatively large quantity of data for ^{210}Pb and ^{210}Po, covering a wide range of species and ecosystems, is typically described using these concepts.

2.1. BASIC PARAMETERS

There are several parameters which are commonly used to describe radionuclide partitioning between environmental compartments. These include transfer to plants or animals from soil, water or within the food chain, namely concentration ratios (CRs) or transfer coefficients (e.g. B_v, F_m and F_f), and between sediments or soil minerals and water, using distribution coefficients (K_d). These parameters are defined for equilibrium conditions, and so it is assumed that an organism or particle is in equilibrium with its surroundings in terms of its accumulation of the radionuclide of interest. The definitions of other, more specific parameters are given in Ref. [2.2]. The parameters described here are also regularly applied within existing predictive models to estimate the transfer of radionuclides in the environment and the resulting exposures for humans and non-human biota.

For terrestrial plants or wildlife, the primary indicator used to characterize radionuclide behaviour in the soil–plant system is the soil–plant CR, also often referred to as the transfer factor (TF), and it describes the transfer of radionuclides from the soil to the plant when uptake by plant roots is the only process affecting the transfer. The CR is defined as the ratio of the activity concentration of radionuclide in the plant or animal (Bq/kg, DW) and in soil (Bq/kg, DW), based on the dry weight (DW):

$$\mathrm{CR} = \frac{\text{Activity concentration of radionuclide in edible tissue } (\text{Bq/kg, DW})}{\text{Activity concentration of radionuclide in soil } (\text{Bq/kg, DW})} \quad (2.1)$$

The soil can be measured to a defined depth (e.g. 10 cm or 20 cm) and can refer to soil contamination per m^2 as an aggregated concentration ratio (CR_{agg}), particularly for materials deposited via atmospheric transfer (see Chapter 5 for a more detailed discussion). The CRs for plants are usually given for the edible parts of the plant.

A similar ratio is applied for aquatic organisms with activity concentration in water (Bq/L) or sediment (Bq/kg), and often on a fresh weight (FW) basis. The transfer of radionuclides from water to aquatic organisms includes the intake of water, sediments and feed, so the assumption is often that all those processes are aggregated.

The CR for any organism can sometimes be expressed as a bioavailable ratio (CR_{bioav}). This acknowledges that some of any environmental radionuclide (^{210}Po in this case) will be in a chemical or physical form not amenable for biological uptake. In those cases, geochemical speciation modelling (e.g. based on pH, Eh, dissolved organics or other water chemistry) or physical processing (e.g. filtration to remove particulates) will be used to estimate the bioavailable portion in the soil or water and this, in turn, can be used to estimate the CR_{bioav}.

For estimating radionuclide transfer from feeds to domestic animal food products, a transfer coefficient is widely used for both milk (F_m) and meat (F_f). This coefficient is defined as the equilibrium ratio of the radionuclide activity concentration in the milk/meat on a FW basis to the daily dietary radionuclide intake:

$$F_{m/f} = \frac{\text{Activity concentration in milk/meat (Bq/kg, FW)}}{\text{Daily radionuclide intake (Bq/d)}} \tag{2.2}$$

CRs for human food chains and biota have some differences. For human food chains, CRs are always ratios of the radionuclide activity concentrations in the edible parts of the organisms to that in the surroundings. However, for non-human species, the CR is calculated for the whole organism. This difference reflects the purpose in the application of these parameters: in the first case, the CRs are intended for assessments of radionuclide activity concentrations in food that may be ingested by humans and are used to provide a radiological dose; whereas the whole organism CRs are intended for dose assessments of the biota.

Considering abiotic compartments, K_d is one of the basic parameters used to characterize the mobility of radionuclides in the environment. The degree to which ^{210}Po is bound to solids (soils in the terrestrial environment and sediments in aquatic systems) is an important factor for determining the concentration of a radionuclide in solution, and it directly influences the fraction of radionuclide that can be incorporated by organisms (see above regarding bioavailability). Dissolved radionuclide ions can bind to solid surfaces by a number of processes that are often classified under the broad term of ‘sorption’. Polonium is particularly affected by sorption owing to its low environmental concentrations and its variable oxidation state (see Chapter 3). K_d for the partitioning of a radionuclide between the particulate phase and the dissolved phase under equilibrium conditions is similar to the aquatic CR described above and is defined as:

$$K_\mathrm{d} = \frac{\text{Activity concentration in solid phase } (\text{Bq/kg, DW})}{\text{Activity concentration in aqueous phase } (\text{Bq/L})} \tag{2.3}$$

All of the above approaches have some limitations owing to the inherent complexity and variability of natural environmental conditions. In particular, the assumption of equilibrium being established in ecosystems is debatable. Hence, the application of CRs for radiological conditions undergoing substantial temporal variations in radionuclide activity concentrations in the environmental media, for example following any accidental release, might be inappropriate.

Dynamic models are widely used for quantifying transfers of ^{210}Po in non-equilibrium situations (see Ref. [2.3]), although the data coverage to populate these models is not always sufficient for robust assessments.

2.2. CLASSIFICATIONS OF PLANTS AND ANIMALS USED FOR EXPOSURE ASSESSMENT MODELS

There are many data in the scientific literature on radionuclide transfer to plants and animals. The data are given for many different species, for whole or parts of plants and animals, as well as for different environmental conditions. However, to suit the purpose of dose assessment using predictive models such as PC-CREAM [2.4] or the ERICA Tool [2.5], the data need to represent some widely recognized, standardized conditions. Such classifications for both human and wildlife food chains can be found in IAEA publications and are discussed here (see Refs [2.2, 2.6, 2.7]).

2.2.1. Transfer to human foodstuffs

The IAEA plant classification system is based on just 14 plant groups [2.2]. All plants are categorized as cereals, maize, rice, leafy vegetables, non-leafy vegetables, leguminous vegetables, root crops, tubers, fruits, grasses (cultivated species), leguminous fodder (cultivated species), pasture (species mixture — natural or cultivated), herbs and other crops. This system has been proposed as a basis for estimating the transfer of radionuclides to plant foodstuffs in the framework of the assessment of exposures to humans through ingestion.

Plant tissues are subdivided into ten compartments: berries, buds, fruits, grains, heads, leaves, roots, seeds and pods, stems and shoots, and tubers. Not all of these ten compartments are assigned to each plant group, but only where the portion represents an edible part of a specific plant.

The transfer of radionuclides to plants greatly depends on soil properties. Existing international soil classification systems try to capture important information for plant cultivation. The soil classification system established by the Food and Agriculture Organization of the United Nations (FAO) and the United Nations Educational, Scientific and Cultural Organization (UNESCO) has 28 units (or categories) of soil and 125 subunits [2.8]. TF values are not available for subunits defined on such a detailed basis: the differences between these units in terms of radionuclide transfer are generally not substantial. Therefore, a much simpler classification system based on texture and organic matter content was suggested in Ref. [2.3], while ensuring that a reasonable amount of data are available for each category. Four soil groups — sand, loam, clay and organic soil — are defined for radiological assessments and are mostly based on grain size (clay ≤ 0.004 mm, sand ≥ 0.06–2 mm, gravel > 2 mm and loams being intermediate). The soils are grouped according to the percentage of sand and clay mineral and the organic matter content in the soil [2.3]:

> “For the mineral soils, three groups were created according to the sand and clay percentages…: ‘Sand group’: sand fraction ≥65%; clay fraction <18%; ‘Clay group’: clay fraction ≥35%; ‘Loam group’: rest of cases. A soils was 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.”

With regard to animals, there are only five categories (cattle, sheep, goats, pigs and poultry) and three groups (meat, milk and eggs) that are considered to provide transfer coefficients from daily radionuclide intake of animals and animal products consumed by humans [2.3]. The classification systems are assumed adequate to cover the variability of environmental conditions and can also be applied for the assessment of the transfer of polonium through the human consumption of food. A detailed discussion of the data and concepts used to define these categories of soil, plants and animals are given in Refs [2.2, 2.3], and many empirical data on ^{210}Po (and ^{210}Pb, where appropriate) presented in this publication use these parameters.

2.2.2. Transfer to wildlife: Reference organism concept

Since it is impossible to consider all species of plants and animals in radiological impact assessments, a classification based on a reference organisms is applied. Larsson [2.9] defines reference organism as “a series of entities that provide a basis for the estimation of radiation dose rate to a range of organisms which are typical, or representative, of a contaminated environment”. Such groups need to be selected on the basis of their representativeness for the selected environments of interest and should also allow realistic assessments, illustrating possible exposure pathways. This approach is used in the ERICA project, which develops a framework for radiation protection of the environment (see Ref. [2.5]).

A similar concept has been applied by the International Commission on Radiological Protection (ICRP), which introduced a system of discrete and clearly defined reference animals and plants for assessing radiation effects on wildlife. According to the ICRP definition [2.10]:

> “A Reference Animal or Plant is 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 reference animals and plants, covering different life stages (e.g. fish egg, adult fish). This is a basis for systematically relating radionuclide exposure to radiological dose, and then dose (or dose rate) to different types of effect, for a number of organisms that are characteristic of different types of natural environment [2.10].

In 2009, the IAEA, within the international programme Environmental Modelling for Radiation Safety (EMRAS II), initiated a working group to develop an international handbook for estimating the transfer of radionuclides to wildlife, similar to Ref. [2.2] for estimating transfer to human foodstuffs. The approach adopted for classification of wildlife was based on a reference organism concept [2.6, 2.7] and was consistent with the ICRP approach [2.11]. However, it was applied more generally, and it defines broader wildlife groups (e.g. soil invertebrate, predatory fish and terrestrial mammal; see Table 2.1). In some cases, a consideration of specific subcategories is included. This approach is also used in this publication, along with the relevant data for ^{210}Pb and ^{210}Po collected within the EMRAS II programme to harmonize environmental impact assessments.

TABLE 2.1. WILDLIFE GROUPS AND REFERENCE ORGANISMS

Broad group	Subcategory	Reference organism
	Terrestrial environments	
Amphibians	—[a]	Frog
Annelids	—[a]	Earthworm
Arachnids	—[a]	—[a]
Arthropods		
	Carnivorous	—[a]
	Detritivorous	—[a]
	Herbivorous	Bee
Birds		Duck
	Carnivorous	Duck
	Herbivorous	Duck
	Omnivorous	Duck
Ferns	—[a]	—[a]
Fungi		
	Mycorrhizal	—[a]
	Parasitic	—[a]
	Saprophytic	—[a]
Gastropods	—[a]	—[a]
Grasses and herbs		
	Grasses	Wild grass
	Herbs[b]	—[a]
Lichens/bryophytes	—[a]	—[a]
Mammals		Rat or deer
	Carnivorous	Rat
	Herbivorous[c]	Rat or deer
	Marsupial[d]	—[a]
	Omnivorous	Rat
	Rangifer spp.	—[a]
Reptiles		
	Carnivorous	—[a]
	Herbivorous	—[a]
Shrubs	—[a]	—[a]
Trees		
	Coniferous	Pine tree
	Broadleaf	—[a]

TABLE 2.1. WILDLIFE GROUPS AND REFERENCE ORGANISMS (cont.)

Broad group	Subcategory	Reference organism
Freshwater environments		
Algae	—[a]	—[a]
Amphibians	—[a]	Frog
Birds		Duck
	Carnivorous	Duck
	Herbivorous	Duck
	Omnivorous	Duck
Crustaceans	—[a]	—[a]
Fish		
	Benthic feeding[e]	—[a]
	Piscivorous[f]	Salmonid
	Forage[g]	—[a]
Insects	—[a]	—[a]
Insect larvae[h]	—[a]	—[a]
Mammals		
	Carnivorous	—[a]
	Herbivorous	—[a]
	Omnivorous	—[a]
Molluscs		
	Bivalve	—[a]
	Gastropod	—[a]
Phytoplankton	—[a]	—[a]
Reptiles	—[a]	—[a]
Vascular plants	—[a]	Wild grass
Zooplankton	—[a]	—[a]
Marine environments		
Birds		Duck
	Carnivorous	Duck
	Herbivorous	Duck
	Omnivorous	Duck
Crustaceans		
	Large	Crab
	Small	—[a]

TABLE 2.1. WILDLIFE GROUPS AND REFERENCE ORGANISMS (cont.)

Broad group	Subcategory	Reference organism
Fish		
	Benthic feeding[e]	Flatfish
	Piscivorous[f]	Salmonid
	Forage[g]	—[a]
Insects	—[a]	—[a]
Macroalgae	—[a]	Brown seaweed
Mammals		
	Carnivorous	—[a]
	Herbivorous	—[a]
	Planktivorous	—[a]
Molluscs		
	Bivalve	—[a]
	Cephalopod[i]	—[a]
	Gastropod	—[a]
Phytoplankton	—[a]	—[a]
Polychaete worms	—[a]	—[a]
Reptiles	—[a]	—[a]
Sea anemones/true corals	—[a]	—[a]
Vascular plants	—[a]	—[a]
Zooplankton	—[a]	—[a]

Source: Tables 2–4 of Ref. [2.6]. The list is based on an on-line database available at www.wildlifetransferdatabase.org

[a] —: data not available.

[b] Herb refers to any non-woody plant which does not fall into one of the other categories.

[c] Does not include *Rangifer* spp. (reindeer and caribou).

[d] All marsupials, regardless of feeding strategy.

[e] Fish feeding on benthic dwelling organisms.

[f] Fish consuming smaller fish, amphibians or birds.

[g] Fish feeding on primary producers and pelagic invertebrates and zooplankton.

[h] Insect larvae are included as the aquatic life phase is important for many species which are terrestrial as an adult.

[i] Squid, octopus and cuttlefish.

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Chapter 3

PROPERTIES OF POLONIUM

This chapter summarizes the physical and chemical properties of polonium and acts as a reference for the subsequent chapters. It includes decay data and series systematics, solution chemistry and adsorption, and methods of determining ^{210}Po activity concentrations in samples.

3.1. PHYSICAL PROPERTIES

3.1.1. Isotopes

There are 42 known isotopes of polonium in the National Nuclear Data Center Chart of Nuclides[1] and none is stable. There are seven naturally occurring radioactive isotopes of polonium, which are part of the natural decay series of long lived radionuclides (see Table 3.1 and Fig. 3.1):

— The ^{238}U series: ^{210}Po, ^{214}Po and ^{218}Po.
— The ^{235}U series: ^{211}Po and ^{215}Po.
— The ^{232}Th series: ^{212}Po and ^{216}Po.

At 138 days, however, only ^{210}Po has a half-life long enough to play a significant role in environmental processes. The decay scheme for ^{210}Po, along with those of ^{210}Bi and ^{210}Pb, which precede ^{210}Po in the ^{238}U decay series, is shown in Fig. 3.2. There are several anthropogenic isotopes with a half-life longer than a day, which are generally produced by irradiation of bismuth (see Table 3.1). The longest lived, ^{209}Po ($T_{1/2}$ = 102 a), has been used for polonium chemistry experiments and as a yield tracer of polonium during sample processing and radiochemical analyses.

3.1.2. Decay series systematics

The distribution of ^{210}Po in the environment is to be considered in the context of ^{238}U decay series systematics. The concentration of each radionuclide is controlled by the concentration of its parent, its own half-life and the amount

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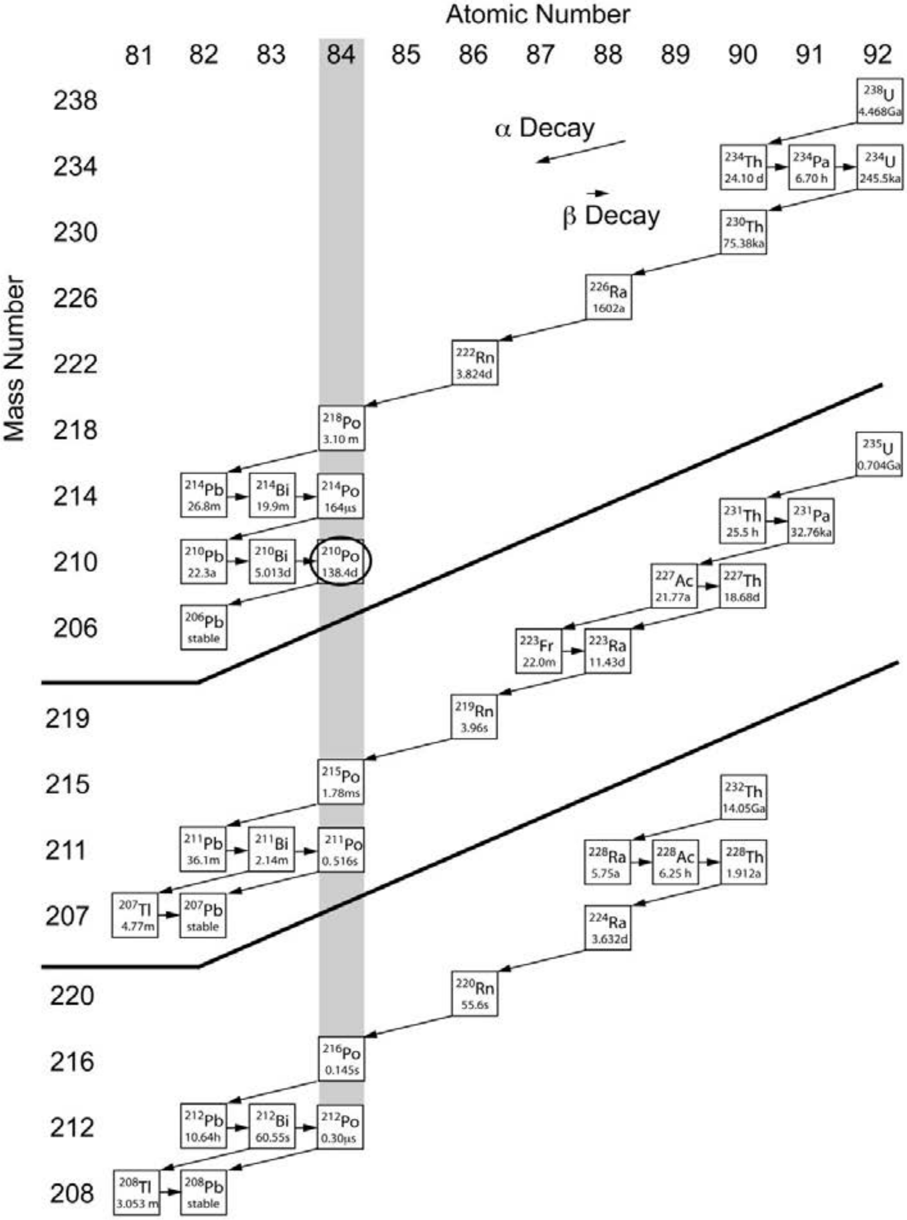
[1] See www.nndc.bnl.gov/chart

TABLE 3.1. POLONIUM ISOTOPES

	Isotope	Half-life	Decay constant (a^{-1})	Major decay modes (>1%)	Specific activity (Bq/g)	Energy of decay (MeV) (with % of total decays)	Emission energy (MeV)
Naturally occurring	^{210}Po	138.376 d	1.829 73	α	1.66×10^{14}	5.407 (>99.9%)	5.304
	^{211}Po	0.516 s	4.24×10^{7}	α	3.83×10^{21}	7.594 (98.9%) 7.024 (0.54%) 6.695 (0.52%)	7.450 6.891 6.568
	^{212}Po	0.300 μs	7.29×10^{13}	α	6.56×10^{27}	8.954 (100%)	8.785
	^{214}Po	162 μs	1.35×10^{11}	α	1.20×10^{25}	7.833 (99.99%) 7.034 (0.01%)	7.687 6.903
	^{215}Po	1.781 ms	1.228×10^{10}	α	1.09×10^{24}	7.526 (99.93%) 7.087 (0.06%)	7.386 6.955
	^{216}Po	0.148 s	1.48×10^{8}	α	1.31×10^{22}	6.906 (>99.99%)	6.778
	^{218}Po	3.07 m	1.19×10^{5}	α	1.04×10^{19}	6.115 (99.98%)	6.002
Anthropogenic ($T_{1/2} > 1$ d)	^{206}Po	8.8 d	29	α (5.45%) ε (94.55%)	2.7×10^{15}	5.327 (5.45%) 1.846 (94.55%)	5.224
	^{208}Po	2.898 a	0.239 2	α	2.19×10^{13}	5.215 (>99.99%)	5.115
	^{209}Po	115 a	6.03×10^{-3}	α	5.50×10^{11}	4.716 (0.55%) 4.977 (79.2%) 4.979 (19.8%)	4.622 4.883 4.885

Source: Data are from the Decay Data Evaluation Project (www.nucleide.org/DDEP_WG/DDEPdata.htm), where available, otherwise (for ^{206}Po and ^{208}Po) from the National Nuclear Data Center Chart of Nuclides (www.nndc.bnl.gov/chart).

Note: Transformations with intensities less than 0.01% have not been included in this table. Intensities are given as a percentage of total decay. Where more complete data are required, the original sources should be consulted.



Note: The only long lived polonium isotope, ^{210}Po, is circled.

FIG. 3.1. The naturally occurring ^{238}U, ^{235}U and ^{232}Th decay series, all of which contain polonium isotopes (Z = 84).

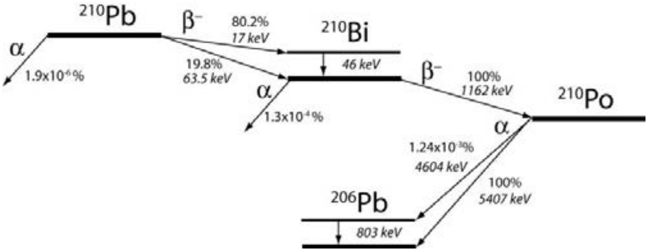


FIG. 3.2. The decay scheme for ^{210}Bi, ^{210}Pb and ^{210}Po.

of time since fractionation between the isotope and its parent occurred. For the decay series elements within samples that have been closed over long timescales, the activity of each isotope (decay rate) is the same as that of its parent (i.e. it is in secular equilibrium with its parent). For the ^{238}U decay series, which contains ^{210}Po, this can be represented by:

$$(^{238}\mathrm{U}) = (^{226}\mathrm{Ra}) = (^{222}\mathrm{Rn}) = \ldots = (^{210}\mathrm{Pb}) = (^{210}\mathrm{Po}) \qquad (3.1)$$

where the parentheses denote activity, which is equal to the number of atoms multiplied by the decay constant.

There are two important points to note:

- In secular equilibrium, the activity concentration (and, from that, the molar concentration) of a daughter radionuclide is controlled by that of the parent radionuclide, and so, ultimately, all radionuclides in the series are controlled by the longest lived parent (here ^{238}U).
- While the activities are equal, the molar abundances are inversely proportional to the decay constants, so that the molar concentrations of very short lived nuclides are very low.

The molar ratio of ^{210}Po:^{238}U is 8.5×10^{-11}, while the molar ratio of ^{210}Po:^{226}Ra is 2.4×10^{-4}. Therefore, even where uranium or radium is greatly enriched, polonium has an extremely low molar concentration — even when it exhibits high radioactivity. This has important implications for understanding its behaviour in the environment.

When any series is not in secular equilibrium due to chemical or physical fractionation, the activity of the nuclides will tend to evolve back towards secular equilibrium over a timescale determined by the half-life of the daughter isotope. The section of the ^{238}U decay series that is most relevant to understanding ^{210}Po is below ^{226}Ra. Including only those isotopes with half-lives longer than one day, this part of the series is:

$$^{226}\mathrm{Ra} \xrightarrow{1602\ \mathrm{a}} {}^{222}\mathrm{Rn} \xrightarrow{3.82\ \mathrm{d}} {}^{210}\mathrm{Pb} \xrightarrow{22.3\ \mathrm{a}} {}^{210}\mathrm{Bi} \xrightarrow{5.01\ \mathrm{d}} {}^{210}\mathrm{Po} \xrightarrow{138.4\ \mathrm{d}} {}^{206}\mathrm{Pb} \quad (3.2)$$

In secular equilibrium, the activities of all these radionuclides are the same. However, the following environmental processes can separate them:

(a) The isotope ^{222}Rn, a noble gas, can migrate. It is generally unreactive and forms no chemical bonds within environmental materials. It is sometimes released from solids by a process known as alpha recoil (see below) and, despite its relatively low aqueous solubility, it can enter any surrounding water (e.g. pore water, groundwater and lake water) or vapour phase (e.g. pore spaces, bubbles and the atmosphere), thereby separating from its ^{226}Ra parent.

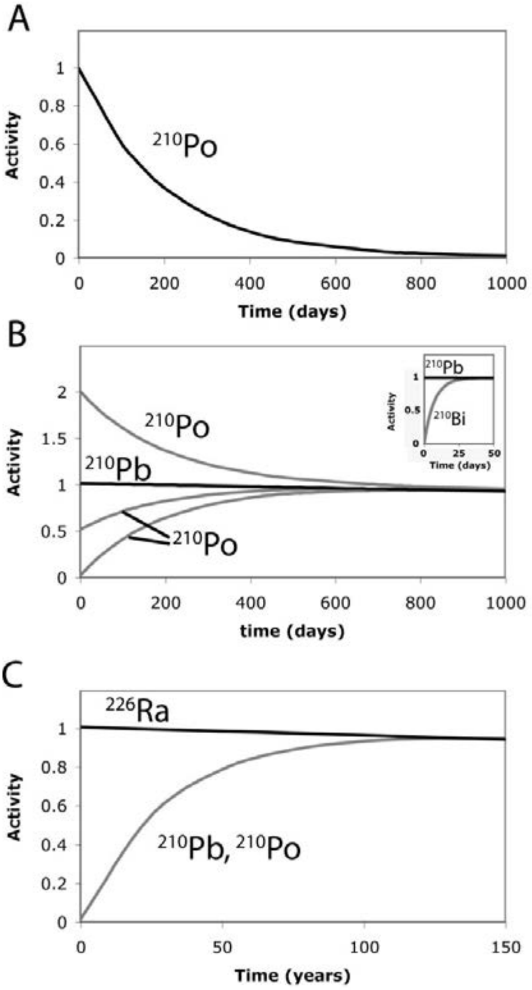
(b) The radionuclides ^{226}Ra, ^{222}Rn and ^{210}Pb, which are produced by alpha decay, and the very short lived radionuclides ^{218}Po and ^{214}Pb, which are between ^{222}Rn and ^{210}Pb, are propelled in the direction opposite to that of the alpha particle and with sufficient energy to travel approximately 20 nm in mineral structures. They can therefore escape to the surrounding water or air or come to rest in an adjacent phase [3.1, 3.2]. The direction of recoil is random for each decay, and the rate of ejection is determined only by the fraction of parent nuclides within recoil distance of mineral surfaces or channels to the surface. This is sometimes referred to as alpha recoil.

(c) Lead and polonium can volatize at high temperature in volcanic vents, wildfires (see Appendix IV) and industrial processes, and then condense or scavenge during subsequent cooling.

(d) There can be contrasting adsorption behaviour of lead, polonium and radium on particles in the atmosphere, and on surfaces in soils and aquifers, and into colloids or particulates in surface waters.

(e) Contrasting uptake, excretion and biodistribution of lead, polonium and radium can occur in biota.

The systematics of ^{210}Po under these varying circumstances are shown in Fig. 3.3. In each case, following disequilibrium, the activity of the daughter nuclide grows into secular equilibrium with the parent over a timescale relating to its half-life. To understand the evolution of ^{210}Po and the resulting radiation, the evolution of ^{210}Pb and ^{210}Bi also needs to be considered. When ^{210}Po is isolated from the other isotopes in the decay series, such as owing to the migration of ^{222}Rn and subsequent decay to ^{210}Po, it will decay unsupported. If it is then not



Note: (A) ^{210}Po is unsupported by decay of the parent ^{210}Bi (and so by decay of ^{210}Pb), for example when ^{210}Po is separated from ^{210}Pb and ^{210}Bi by precipitation or volatilization. (B) There is ^{210}Pb present, and the evolution of ^{210}Po is shown for several different starting activities. The excess or deficiency of ^{210}Po will decay according to the half-life of ^{210}Po. Such situations are found in both biological systems and abiotic environments, where ^{210}Po can be fractionated from ^{210}Pb to generate either an excess or a deficiency of ^{210}Po. The inset shows the ingrowth of ^{210}Bi, which occurs over a much shorter time period, and so the amount of initial ^{210}Bi has little effect on the overall evolution of ^{210}Po. (C) The ingrowth of ^{210}Pb is shown in the presence of ^{226}Ra, for example within barite ($BaSO_4$), which strongly incorporates radium. The different timescale for this circumstance should be noted. Over this time period, ^{226}Ra is almost constant. The activity of ^{210}Po will closely follow that of ^{210}Pb.

FIG. 3.3. The closed-system evolution of ^{210}Po under various circumstances.

subject to further additions, and losses only by decay, its activity will evolve over time according to:

$$\left({}^{210}\mathrm{Po}\right)=\left({}^{210}\mathrm{Po}\right)_0 \mathrm{e}^{-\lambda_{^{210}\mathrm{Po}}t} \tag{3.3}$$

where the activity (^{210}Po) decays from the initial value (^{210}Po)$_0$ according to the decay constant $\lambda_{^{210}\mathrm{Po}}$. The evolution is shown in Fig. 3.3, where the activity will drop to 1% of the initial value in 6.6 half-lives or about 2.5 years.

When ^{210}Pb and ^{210}Po are separated from the rest of the decay series, the activity of ^{210}Pb will evolve according to an equation analogous to Eq. (3.3), although it will decay with a half-life of 22.3 years. Its daughter, ^{210}Bi, will grow according to:

$$\left({}^{210}\mathrm{Bi}\right)=\left({}^{210}\mathrm{Pb}\right)_0 \frac{\lambda_{^{210}\mathrm{Bi}}}{\lambda_{^{210}\mathrm{Bi}}-\lambda_{^{210}\mathrm{Pb}}}\left(\mathrm{e}^{-\lambda_{^{210}\mathrm{Po}}t}-\mathrm{e}^{-\lambda_{^{210}\mathrm{Bi}}t}\right) \tag{3.4}$$

Over the time period in which the activity of ^{210}Bi approaches that of ^{210}Pb (i.e. to within 1% in 33 days), the activity of ^{210}Pb drops by only 0.3% and so can be considered constant:

$$\left({}^{210}\mathrm{Po}\right)=\left({}^{210}\mathrm{Pb}\right)_0 \left[\begin{array}{l} \dfrac{\lambda_{^{210}\mathrm{Bi}}\lambda_{^{210}\mathrm{Po}}}{\left(\lambda_{^{210}\mathrm{Bi}}-\lambda_{^{210}\mathrm{Pb}}\right)\left(\lambda_{^{210}\mathrm{Po}}-\lambda_{^{210}\mathrm{Pb}}\right)}\mathrm{e}^{-\lambda_{^{210}\mathrm{Pb}}t} \\ +\dfrac{\lambda_{^{210}\mathrm{Bi}}\lambda_{^{210}\mathrm{Po}}}{\left(\lambda_{^{210}\mathrm{Pb}}-\lambda_{^{210}\mathrm{Bi}}\right)\left(\lambda_{^{210}\mathrm{Po}}-\lambda_{^{210}\mathrm{Bi}}\right)}\mathrm{e}^{-\lambda_{^{210}\mathrm{Bi}}t} \\ +\dfrac{\lambda_{^{210}\mathrm{Bi}}\lambda_{^{210}\mathrm{Po}}}{\left(\lambda_{^{210}\mathrm{Pb}}-\lambda_{^{210}\mathrm{Po}}\right)\left(\lambda_{^{210}\mathrm{Bi}}-\lambda_{^{210}\mathrm{Po}}\right)}\mathrm{e}^{-\lambda_{^{210}\mathrm{Po}}t} \end{array}\right] +\left({}^{210}\mathrm{Po}\right)_0 \mathrm{e}^{-\lambda_{^{210}\mathrm{Po}}t} \tag{3.5}$$

Substituting values for the decay constants gives:

$$\left({}^{210}\mathrm{Po}\right)=\left({}^{210}\mathrm{Pb}\right)_0 \left[\begin{array}{l} 1.0179\mathrm{e}^{-\lambda_{^{210}\mathrm{Pb}}t} \\ +0.0376\mathrm{e}^{-\lambda_{^{210}\mathrm{Bi}}t} \\ -1.0555\mathrm{e}^{-\lambda_{^{210}\mathrm{Po}}t} \end{array}\right] +\left({}^{210}\mathrm{Po}\right)_0 \mathrm{e}^{-\lambda_{^{210}\mathrm{Po}}t} \tag{3.6}$$

Regardless of the initial activity of ^{210}Po relative to that of ^{210}Pb, the activity of ^{210}Po will evolve to within 1% of that of ^{210}Pb in 2.5 years. Over this time, the activity of ^{210}Pb drops by only 7.5% (i.e. the term drops to 0.925, and so for much of the time can be approximated to 1).

3.2. CHEMICAL PROPERTIES

While in some environments the activity concentration of ^{210}Po can be of concern, the molar concentrations in the environment are always extremely low. In the long term, the distribution of ^{210}Po is controlled by that of the parent, ^{210}Pb. The behaviour of ^{210}Po in aqueous systems is generally dominated by adsorption onto surfaces, although incorporation into colloids, biovolatilization and precipitation in sulphides can be important in some circumstances.

There have been several reviews on polonium chemistry and its behaviour in the environment [3.3, 3.4]. However, the data have been limited and some reasons for this include the following:

(a) The very high specific activity of ^{210}Po (1.66×10^{14} Bq/g; see Table 3.1) causes practical difficulties in the safe handling of even small amounts of natural polonium. The specific activities of artificially produced ^{208}Po and ^{209}Po are lower but still high.
(b) The very high specific activity also readily damages crystal lattices and materials in experiments, so it is difficult to obtain undamaged materials (e.g. precipitated minerals) for characterization.
(c) Naturally occurring ^{210}Po is very scarce. One gram of ^{238}U is generally accompanied by 75 pg of ^{210}Po. Hence, in crustal rocks with an average concentration of 2.7 ppm uranium, there is only 0.20 ng of ^{210}Po per tonne. Production of ^{208}Po and ^{209}Po requires bombardment of bismuth by neutrons in a reactor.
(d) In the environment, ^{210}Po is present at such low concentrations that it clearly does not form separate phases (see Sections 3.2.1–3.2.3 and Chapter 6 for more details). For example, groundwater typically has less than 40 mBq/L, with a maximum value of 19 Bq/L (see Section 4.5.1), which corresponds to typical groundwater concentrations of less than 0.001 fM and a maximum of only 0.45 fM.
(e) In many circumstances, ^{210}Po will decay or approach secular equilibrium with its parent, ^{210}Pb. Its distribution is therefore strictly controlled by ^{210}Pb.

3.2.1. Solution chemistry

Polonium is in Group 16 of the periodic table, which is known as the chalcogens and includes oxygen, sulphur, selenium and tellurium (see Fig. 3.4). It is generally considered to be a metalloid, with physical and chemical characteristics that are intermediate between those of metals and non-metals (along with boron, silicon, germanium, arsenic, antimony, tellurium and astatine), although it can behave more like the metals bismuth and lead (a review of the early analytical chemistry can be found in Refs [3.5–3.7]). Based on analogy with selenium and tellurium, Figgins [3.6] predicts that polonium would have stable oxidation states of −2, +2, +4 and +6, with the +4 oxidation state being the stable state in solution under oxic freshwater conditions; although Po^{2+} has also been predicted to be most stable in sea water and under reducing conditions [3.8, 3.9]. Polonium hydrolyses, forming $PoO(OH)^+$, $PoO(OH)_2$ and PoO_2 in slightly acidic to neutral pH regions and PoO_2^{-3} in alkaline solutions [3.10].

There are limited solubility data available for polonium, but it is generally assumed to be highly insoluble. Work was conducted on the precipitation of polonium from laboratory solutions in the 1950s, in particular during the development of separation procedures, and polonium was found to precipitate with various compounds of antimony, bismuth, tellurium and rare earth elements [3.5]. The solubility product for $Po(OH)_4$ is 10^{-37} [3.6]; hence, in natural waters, equilibrium activities are highly sensitive to pH [3.11]. In acidic solutions, trace polonium is precipitated by hydrogen sulphide (H_2S), along with other insoluble sulphides. Under these conditions, polonium is also highly insoluble, sinwith ce the solubility product for PoS at around 5×10^{-29} [3.6]. The concentration of dissolved polonium can therefore be strictly controlled by sulphur recycling, where the dissolution and reprecipitation of sulphur minerals occurs [3.12]. However, away from very dynamic conditions, adsorption onto particles, mineral surfaces and colloids provides the stronger control on dissolved polonium concentrations.

Polonium is present in precipitates containing radium owing to ingrowth after mineral formation. Radium is commonly precipitated in barite ($BaSO_4$). Since polonium is not readily accommodated within a crystal structure like radium, it might be released during recystallization, although laboratory experiments indicate that recrystallization does not expel a significant fraction of polonium [3.13]. In a study of the bacterial mobilization of polonium from waste gypsum, LaRock et al. [3.14] demonstrate that sulphate reducing bacteria are effective at mediating polonium release from gypsum, provided the sulphide levels resulting from their metabolism does not rise above 10 µmol/L, in which case polonium is evidently co-precipitated as a metal sulphide.

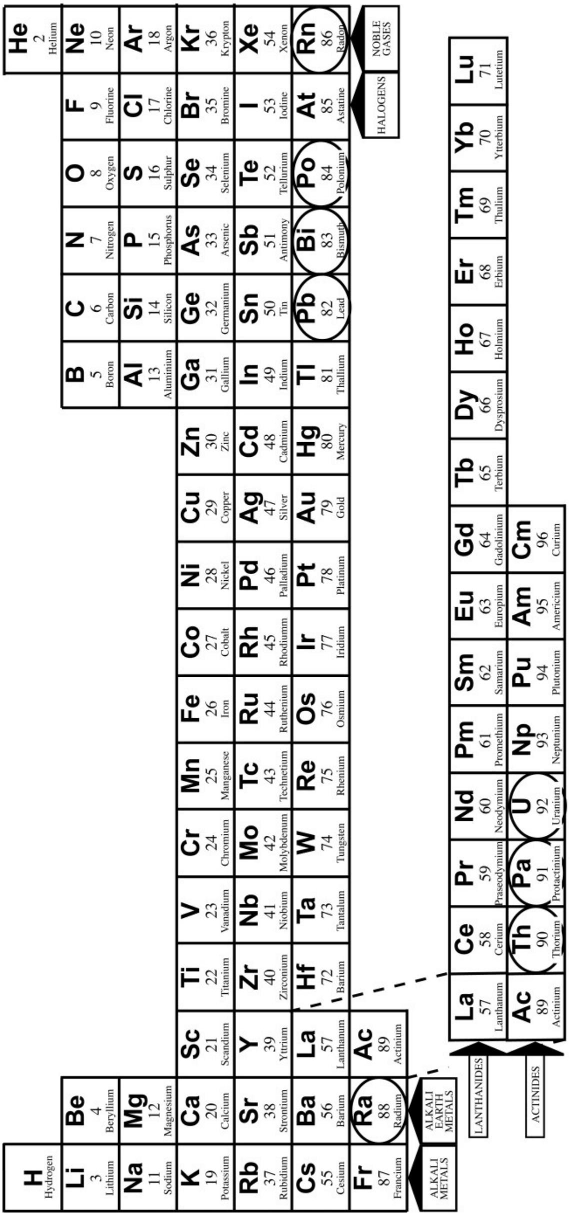


FIG. 3.4. The location in the periodic table of polonium and the elements with isotopes within the ^{238}U decay series.

Polonium forms colloids in solution [3.15], and while this may be a significant factor when there are high concentrations of polonium, or in pure solutions in laboratory experiments (see Ref. [3.10]), it is more likely that in natural environments polonium is adsorbed onto, or incorporated into, colloids formed by more abundant elements.

3.2.2. Adsorption

In most natural environments, polonium is highly surface reactive, and readily adsorbs onto mineral surfaces, particulates and colloids. However, there are limited data quantifying this. Models for the description of radionuclide sorption are still mostly based on empirical solid–liquid distribution coefficient (K_d) values of bulk materials (see Sections 2.1 and 6.4.3 for more information on K_d). This approach is the simplest sorption model available and K_d is the ratio of the concentration of radionuclide in a specified solution to the concentration of the radionuclide sorbed on a specified solid when the solution and solid are at equilibrium, with units L/kg. However, these values cannot be easily extrapolated to materials with different compositions.

There are only limited experiments to determine K_d for polonium on homogenous materials. Laboratory experiments on montmorillonite clay found measured adsorption values of around 1.5×10^3 L/kg for ^{210}Po, which were independent of both ionic strength (up to 0.2 eq/L) and pH (6 and 7) [3.10]. However, polonium that was adsorbed from solution could not be readily desorbed, with a several orders of magnitude greater proportion of polonium remaining adsorbed. It was suggested that polonium forms stable covalent surface complexes. It was also found that ^{210}Po generated by radioactivity of sorbed ^{210}Pb can be more readily desorbed, although a distribution value much higher than the adsorption experiments, of 2×10^5 L/kg, was still obtained. For other materials, batch partitioning experiments found average values of the following [3.6]:

- 8.4×10^4 L/kg for bentonite at pH10.1;
- 3.7×10^4 L/kg for a tuff (mainly plagioclase, smectite and the zeolite clinoptilolite) at pH9.4;
- 2.5×10^4 L/kg at pH10.1 for a granodiorite (largely quartz, plagioclase and K-feldspar).

These values were obtained by ultrafiltering the solution, and somewhat lower values were obtained by using 0.45 μm filters. Presumably, a significant fraction of polonium was not in true solution but rather on colloid sized particles, and this should be considered when comparing the results of different studies. Manganese (IV) oxides have often been inferred to significantly adsorb or incorporate substantial amounts of polonium in field studies (see Refs [3.16–3.18]). Pyrolusite (MnO_2) has been qualitatively shown to effectively adsorb polonium in column experiments [3.19]. However, adsorption coefficients onto manganese minerals has not been quantified in the laboratory.

Some results for partitioning experiments with soils have been published. An early study [3.20] still provides the basis for most estimates of polonium adsorption in soil, and is the basis for recent recommended values [3.21, 3.22].

Polonium partitioning has also been determined in aquatic systems, including rivers, estuaries, coastal seas and the open ocean. In these environments, ^{210}Po consistently displays K_d values of approximately 10^5, and usually slightly higher than the K_d of the parent radionuclide, ^{210}Pb [3.23, 3.24]. Values for a range of natural environments, determined from measurements of filtered waters and particulates from filters, are shown in Table 3.2.

TABLE 3.2. AQUATIC K_d IN SITU DETERMINATIONS

Environment	n	^{210}Po K_d	^{210}Pb K_d	Ref.
River	5	$(1.0 \pm 0.3) \times 10^5$	$(3 \pm 1) \times 10^4$	[3.23]
Estuary	7	$(3.3 \pm 1.5) \times 10^5$	$(2.4 \pm 1.9) \times 10^5$	[3.24]
Coastal sea, shoreline	17	$(2.8 \pm 0.9) \times 10^5$	$(1.7 \pm 1.0) \times 10^5$	[3.24]
Surface, open ocean	4	$(1–4) \times 10^4$	$(1–4) \times 10^4$	[3.23]

In rivers, ^{210}Po is mostly associated with suspended particulate matter and bottom sediments. In estuaries, ^{210}Po and lead partitioning may be controlled by the concentration of dissolved organic matter and other ligands, the nature and concentration of particulate matter, and by changes in salinity [3.23]. In coastal sea water, ^{210}Po is mostly associated with biological particles (bacteria and microplankton), while ^{210}Pb is mostly associated with the inorganic particulate fraction (clay minerals). The enhanced incorporation of ^{210}Po in particulate organic material, and also in the first levels of trophic marine chains, has been linked to metabolic processes and ^{210}Po binding to proteins [3.25–3.28]. The same occurs in open ocean waters and the degree of planktonic uptake in the euphotic zone of oceans may have a strong influence on ^{210}Po flux in oceanic water columns [3.29, 3.30].

Overall, the K_d values that are available can be used as guidelines, but there is little systematic data exploring the effects of various conditions on polonium partitioning. Furthermore, K_d values are generally values for the relative concentrations between phases and not partition coefficients for equilibrium adsorption. Further work is required to determine when polonium incorporation is reversible, and how it varies with polonium concentration.

3.2.3. Biovolatilization

Hussain et al. [3.31] find that organo-polonium compounds may comprise a substantial proportion of environmental polonium, based on experiments in which up to 50% of polonium is lost from Florida groundwater with high polonium concentrations by bubbling nitrogen. Furthermore, this effect is substantially reduced when oxidizing agents are added, suggesting that volatile organic species have been destroyed. By analogy with the behaviour of tellurium, Hussain et al. [3.31] suggest that dimethyl polonium was present. A similar experiment with groundwater from Nevada did not find significant polonium volatilization, indicating that the extent of organic polonium complexation may vary [3.18]. In addition to the implications for volatile loss of polonium to the atmosphere, these results suggest that there may be organic ligands that can control polonium adsorption behaviour in soil and groundwater.

Momoshima et al. [3.32] demonstrate that polonium is lost by volatilization in culture experiments in which microorganism growth has been enhanced in seawater and coastal sediments, and suggest that methylation of polonium is responsible. In a further, they show that when methylcobalamin, which is involved in organic methylation, is added, polonium is also lost. Similar losses are found from similar culture experiments using fresh waters and increase with enhanced bacterial growth [3.33]. Volatilization has also been demonstrated in different pure bacterial cultures [3.34].

3.3. ^{210}Po DETERMINATION

Only a short summary of ^{210}Po determination in environmental samples is presented here, since there are several studies on the topic, including a comprehensive review [3.35] and a procedure for analysis of water samples [3.36–3.38]. Owing to the volatility of polonium, high temperature sample preparation and digestion techniques, such as dry ashing and fusion with molten salts, cannot be used. To avoid polonium losses, drying should be by freeze drying or oven drying at a relatively low temperature (≤80°C).

Wet ashing is commonly used for sample dissolution, sometimes in combination with microwave digestion. Soils, sediments and other solid samples, such as filtered materials, are usually prepared using HCl, HF, HNO_3 and $HClO_4$ in varying proportions, with ashing in open vessels, pressure vessels or microwave digestion systems. Biological samples are generally treated by first drying them thoroughly at temperatures of up to 80°C or freeze drying, followed by treatment with HNO_3 (sometimes also with HCl and H_2O_2) to destroy organic matter. Polonium is pre-concentrated from water samples by a wide variety of techniques, with the most common involving one of three procedures: evaporation; co-precipitation, typically on $Fe(OH)_3$ or MnO_2; or chelation, commonly with ammonium pyrrolidine dithiocarbamate (APDC) species [3.35, 3.39].

As ^{210}Po is an alpha emitting radionuclide with a relatively short half-life and produces only very-low-intensity gamma rays and no radioactive progeny, the only practical way to determine its presence is to detect its alpha particle. The most commonly used method is alpha particle spectrometry, which utilizes a silicon surface barrier or passivated implanted planar silicon (PIPS) detectors, on the account of their excellent energy resolution, compact size, low background, excellent stability and low sensitivity to gamma radiation. Alpha spectrometry can also be performed using gridded ion chambers, which have a high efficiency and allow the use of large sources. Alternatively, alpha particle counting may be performed using a ZnS scintillator screen and photomultiplier tube, gas proportional counter or liquid scintillation counter. This approach has the disadvantage that a radioisotopic tracer cannot be used, although some liquid scintillation systems do enable separation of the ^{209}Po peak.

Whichever detection method is used, ^{210}Po has first to be chemically separated from both the sample matrix and from other alpha emitting radionuclides present in the sample. Polonium separation methods include those based on solvent extraction, ion exchange chromatography and extraction chromatography [3.35]. In addition, polonium spontaneously deposits from mild acid solutions onto metal surfaces, in particular silver, and this is very often utilized as a combined chemical separation and source preparation step [3.40]. The interference of iron during spontaneous deposition is suppressed by the addition of ascorbic acid or hydroxylamine hydrochloride (to reduce Fe^{3+}). Citric acid may also be added to suppress the effects of other ions present. Metals other than silver have been used for plating of polonium, including copper and nickel. However, analyses using these metals are more prone to interference as some ^{210}Bi or ^{210}Pb may also be deposited, resulting in an overestimation of ^{210}Po when there is a delay between deposition and counting [3.41, 3.42].

An advantage of the alpha spectrometry detection methods is that an isotopic yield tracer can be used to allow for losses during sample preparation, chemical separation and source preparation steps. Historically, ^{208}Po has been the preferred tracer, probably on the account of its greater availability. The advantage of ^{209}Po over ^{208}Po is that its E_α is further separated from that of ^{210}Po (see Table 3.1), allowing better peak resolution. In the case of non-spectrometric methods, such as total alpha counting using a ZnS scintillator screen or liquid scintillation counting, the recovery for ^{210}Po during the separation and source preparation process needs to be estimated.

Contamination of any alpha spectrometry detector by the tracer (^{208}Po or ^{209}Po) or by ^{210}Po is a potentially serious problem. This contamination can be minimized by a period of exposure of the source to air before counting, possibly due to the formation of a surface oxide film on the silver [3.5, 3.43]. A delay period of at least two days has been suggested [3.39].

A polonium spectrum with ^{209}Po used as the yield tracer is shown in Fig. 3.5. The spectrum is a simple one, consisting essentially of two singlet peaks. The spectrum obtained using ^{208}Po as a tracer is similar, but with a smaller separation between the two peaks. As described in Ref. [3.35]:

> "Once the count rates in the tracer and ^{210}Po peaks have been obtained and tailing allowed for, the contributions due to detector background and procedure blank need to be subtracted. The ratio of the net count rates in the two peaks is then used to calculate the activity concentration of ^{210}Po in the sample on the date of chemical separation of Po, taking into account the activity concentration of the tracer solution, the masses of the sample and the tracer solution used, the decay of ^{210}Po between separation and counting, the decay of the tracer between its calibration date and counting, and the α emission probabilities in the measured areas in the α-spectrum".

Finally, the results need to be corrected for decay and for ingrowth from its progenitors (especially ^{210}Pb) between the sampling date and the date of chemical

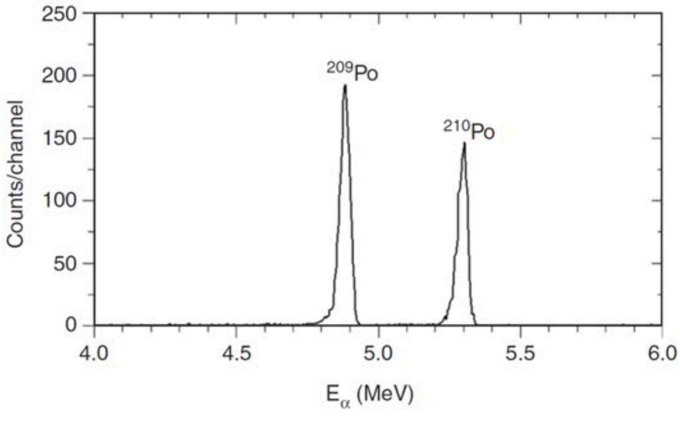


FIG. 3.5. A typical polonium alpha particle spectrum with ^{209}Po as the yield tracer.

separation of polonium from the sample for analysis. This calculation needs to be carried out separately from the decay correction to separation date because, in the former, the ^{210}Po is supported by ^{210}Pb in the sample, whereas in the latter it is unsupported (see Fig. 3.6).

In order to make the correction for ^{210}Po to the sample collection date, the ^{210}Pb activity concentration needs to be determined. The details of the ingrowth and decay calculation are dependent on the procedure followed [3.35]:

> “One common approach is that when Po is chemically separated, the solution containing the ^{210}Pb is retained and kept for an ingrowth period (usually several months). A subsequent determination of ^{210}Po from this solution is used to calculate the activity concentration of ^{210}Pb in the sample”.

An alternative approach is to use a separate determination of ^{210}Pb in the sample by another method, such as gamma spectrometry or beta counting. In general, the time delay between sample collection and ^{210}Po determination should be kept as short as practicable to avoid errors due to this decay and ingrowth correction, particularly for sample types for which the ^{210}Po:^{210}Pb activity ratio may be low (e.g. for rainwater) [3.37].

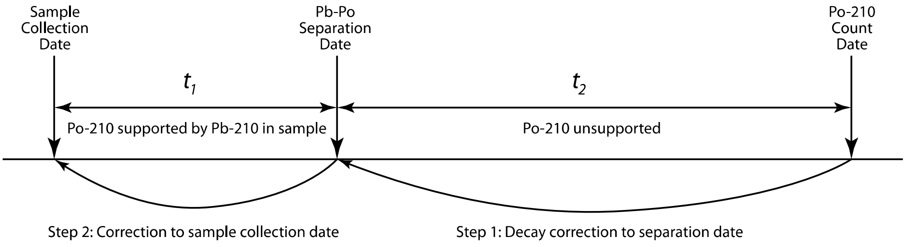


FIG. 3.6. Decay and ingrowth corrections for ^{210}Po determination, taking into account the time intervals t_1 (separation date − sampling date) and t_2 (count date − separation date).

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Chapter 4

OCCURRENCE AND CYCLING OF ^{210}Pb AND ^{210}Po IN THE ENVIRONMENT

This chapter provides a general overview of the occurrence, behaviour and cycling of ^{210}Pb and ^{210}Po in the environment and covers some major concepts and pathways (see Fig. 4.1). The data presented give a feeling for the orders of magnitude of activity concentrations which can be expected in environmental media and the relative importance of various pathways. However, the information is not comprehensive, nor does it provide details of the mechanisms involved. Environmental media and pathways are discussed in more detail in Chapters 5–10, including data on activity concentrations, transfer parameters and fluxes.

4.1. OCCURRENCE OF ^{210}Pb AND ^{210}Po IN ROCK AND SOIL

The source of virtually all ^{210}Pb and ^{210}Po in the environment is ^{238}U in the Earth's crust. In unweathered rocks, ^{210}Pb and ^{210}Po are expected to be in secular equilibrium with ^{238}U. A measure of typical natural concentrations is given by the composition of the upper continental crust (see Table 4.1), which has an average concentration of uranium of 2.7 ppm or 33 Bq/kg [4.2].

TABLE 4.1. TYPICAL URANIUM CONCENTRATIONS IN VARIOUS ROCKS

Rock type	U (ppm)	^{238}U (Bq/kg)
Basalts	0.1–1	1–12
Black shale	3–1 250	37–15 000
Gneiss	2	25
Limestone	2	25
Phosphates	50–300	620–3 700
Schist	2–5	25–62
Silicic rocks (granites, dacites)	2.2–6.1	27–75

Source: See Ref. [4.1].

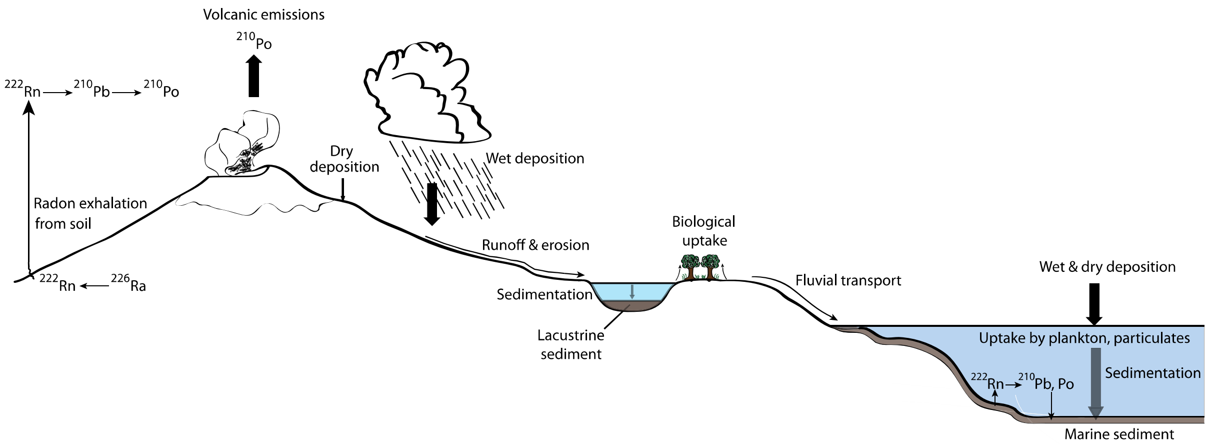

FIG. 4.1. Occurrence and cycling of ^{210}Pb and ^{210}Po in the environment.

Three major processes which transfers ^{210}Pb and ^{210}Po out of primary sources in the crust include the following:

(a) Weathering of exposed rocks at the Earth's surface;
(b) Leaching or dissolution of rocks by groundwater;
(c) Volcanic eruptions.

Surface weathering and subsequent geomorphological and geochemical processes result in the formation of soils and sediments. These processes can modify the concentration of uranium and separate the radionuclides within the uranium series decay chain. The result can be disequilibrium between the radionuclides (see Section 3.1.2). Surface soils are the interface between the atmosphere and the subsurface hydrological system, and there are radionuclide fluxes between these environmental compartments. Secular equilibrium for the ^{238}U decay chain of radionuclides in soils cannot be assumed, although the degree of disequilibrium is not large in the majority of cases.

One mechanism which causes disequilibrium in the uranium decay chain is exhalation of ^{222}Rn from surface soils to the atmosphere. This subsequently decays to ^{210}Pb, which is then deposited to the Earth's surface. Exhalation of ^{222}Rn results in a deficiency of ^{210}Pb relative to ^{226}Ra in the topsoil (primarily the top 1 m), and deposition results in an excess. Approximately 90% of ^{210}Pb deposition is by wet deposition and greatly depends on the rainfall [4.3, 4.4]. A confounding factor is ^{210}Pb in leaf litter, which can lead to excess ^{210}Pb in the surface layer [4.5, 4.6]. Since exhalation flux densities of ^{222}Rn from soils are about a hundred times greater than those from oceans, which cover more than 70% of the planet, ^{210}Pb to oceans will result in deficiency of ^{210}Pb relative to ^{226}Ra in soils.

Polonium-210 is at, or very close to, secular equilibrium with ^{210}Pb in almost all soils [4.7]. Its concentrations in surface soil vary considerably, and Table 4.2 presents data from studies over the last forty years [4.8]. In areas of normal radiation background, the concentrations most commonly lie within the range of 10–200 Bq/kg in the absence of anthropogenic influences [4.7, 4.23]. The upper ranges are mainly due to areas of uranium mining.

The concentrations of ^{210}Po in surface soils vary across a wide range, depending on the uranium concentration in the bedrock of the area, the depth of the soil sampled, climatic conditions and soil properties [4.10, 4.15, 4.22]. In addition, phosphate fertilizers commonly contain significant concentrations of ^{210}Pb and ^{210}Po [4.24, 4.25].

TABLE 4.2. CONCENTRATIONS OF ^{210}Po IN TOPSOIL

Origin	^{210}Po (Bq/kg, DW)	Ref.
Brazil	27–74	[4.9[a]]
Canada	20–22 000	[4.7, 4.10[a]]
Germany	11–210	[4.11–4.13[b]]
India	4–220	[4.14–4.16]
Malaysia	8	[4.17]
Spain	16–780	[4.18[c]]
United Kingdom	51	[4.19]
USA	10–60 70–15 000	[4.14, 4.20] [4.21[a]]
Worldwide	8–220	[4.22]

Source: See Ref. [4.8].

[a] Near a uranium mine.

[b] Near a coal plant.

[c] Near a phosphate factory.

4.2. OCCURRENCE OF ^{210}Pb AND ^{210}Po IN THE ATMOSPHERE

The primary source of ^{210}Pb in the atmosphere is ^{222}Rn exhalation from soil and its subsequent decay via short lived radon progeny. This has been estimated to give rise to a ^{210}Pb flux to the atmosphere of 3.5×10^{16} Bq/a, while fluxes from other sources are more than one order of magnitude lower [4.4]. Much of this ^{210}Pb is removed from the atmosphere by wet and dry deposition, but that fraction which does not deposit decays via ^{210}Bi to ^{210}Po, giving a flux from this source of 1.05×10^{15} Bq/a.

Volcanic eruptions are a major source of ^{210}Po in the atmosphere due to the volatility of polonium at elevated temperatures. An early study estimates the average flux to be 2.4×10^{15} Bq/a for ^{210}Po, but only 6.0×10^{13} Bq/a for ^{210}Pb [4.26]. Compared to ^{222}Rn exhalation from soil, volcanic eruptions are a highly variable source, with relatively large year to year variations.

Other sources include ^{222}Rn exhalation from the oceans (see Section 5.2.1), resuspension of soils (see Section 5.4.1), forest and savanna fires (see

Section IV.2) and the combustion of fossil fuels. Globally, these contribute less than 10% of the total fluxes of ^{210}Pb and ^{210}Po in the atmosphere [4.4], although they are sometimes important on a local or regional scale.

Both ^{210}Pb and ^{210}Po are attached to aerosol particles in the atmosphere. Radioactive decay is only a minor contributor to their removal from the atmosphere due to their relatively short residence times. The primary removal pathway is wet deposition to the Earth's surface (see Section 5.3). A secondary pathway is dry deposition to the surface, which accounts for about 10–15% of total deposition for ^{210}Pb [4.3, 4.4]. The annual, total deposition flux of ^{210}Pb ranges from several $Bq \cdot m^{-2} \cdot a^{-1}$ (as observed in the Antarctic) to several hundred $Bq \cdot m^{-2} \cdot a^{-1}$.

The distribution of ^{210}Pb and ^{210}Po in the atmosphere is complex due to the spatial and temporal variability in the sources, in wet and dry deposition fluxes, and in atmospheric dispersion processes. The highest ^{210}Pb concentrations in surface air are observed in the subtropical and temperate latitudes of the northern hemisphere owing to the relatively large land masses there. Average annual concentrations in this region mainly lie in the range of 0.2–1 mBq/m^3 [4.27]. Between the tropics, values are generally in the range of 0.1–0.5 mBq/m^3, while south of 30°S, they quickly trend down to less than 0.1 mBq/m^3, which is consistent with the radon exhalation flux density trends discussed above. These data are predominantly for continental locations, where higher surface air concentrations are on the account of greater ^{222}Rn exhalation fluxes from soils than oceans.

There is also clear evidence of a seasonality effect in ^{210}Pb and ^{210}Po concentrations in surface air [4.28–4.31]. Concentrations are normally higher during dry seasons due to higher exhalation of ^{222}Rn from the soil, greater resuspension of soil dust and lower wet deposition rates, which can vary locally.

The vertical distribution of ^{210}Pb in the troposphere is highly variable. Although mean vertical profiles of ^{222}Rn generally show a strong reduction in concentration from the surface to the upper troposphere of a factor of 100 over continental areas, for ^{210}Pb this reduction is generally less than a factor of 5, and the concentration at the upper troposphere can be higher than in the lower troposphere [4.32, 4.33]. The reason for this is scavenging of ^{210}Pb by wet and dry deposition in the lower troposphere and by precipitation from the mid-layers. The mean residence time for ^{210}Pb in the troposphere is about five days [4.28], but it is significantly longer than this in the upper troposphere, resulting in higher ^{210}Po:^{210}Pb ratios than in the lower and mid-level troposphere.

The geographical distributions of ^{210}Pb and ^{210}Po concentrations in rainwater mostly follow those in surface air. The main factors governing radionuclide concentrations in rainwater are the size of the aerosols containing ^{210}Pb and ^{210}Po, and the type and duration of rainfall [4.34]. Values for ^{210}Pb concentration in rainwater vary considerably at different locations, being most commonly in the range of 10–1000 mBq/m^3 with ^{210}Po:^{210}Pb activity ratios in the range of 0.05–0.5.

In the stratosphere, ^{210}Pb and ^{210}Po are present due to the input of ^{210}Pb and ^{222}Rn from stratosphere–troposphere exchanges, and the relatively long (approximately a year) stratospheric residence times.

4.3. OCCURRENCE OF ^{210}Pb AND ^{210}Po IN TERRESTRIAL ECOSYSTEMS

4.3.1. ^{210}Pb and ^{210}Po in plants

Terrestrial plants receive ^{210}Pb and ^{210}Po from three principal sources:

(a) Radioactive decay from ^{222}Rn already accumulated by the plant;
(b) Root uptake from the soil;
(c) Foliar uptake of ^{210}Pb and ^{210}Po deposited on the leaves.

It is generally believed that foliar uptake dominates for ^{210}Po, which is thus accumulated primarily by absorption and further translocation within the plant (see Ref. [4.35]). However, the relative importance of the root and foliar uptake pathways depends on the concentration of the radionuclides in the soil, the soil–plant concentration ratio (CR), and the rate of deposition onto plant parts above ground.

Another important consideration is that in crops such as roots, tubers, cereals, nuts and legumes whose edible portion is protected by inedible plant parts, activity concentrations should not be significantly affected by direct deposition. In contrast, observed concentrations for leafy vegetables may be up to 3–5 times higher due to deposition effects. Therefore, there can be a large variation in the radionuclide concentrations in plants, resulting from differences in radionuclide accumulation in different plant species, variable tissue distributions, climate variations and soil properties.

Sheppard et al. [4.7] report activity concentrations in plants in Canada in the range of 1–730 Bq/kg (DW) for ^{210}Pb and 0.3–460 Bq/kg (DW) for ^{210}Po. Data of the same order of magnitude were reported in 1976 for ^{210}Pb and ^{210}Po concentrations in Russian plants by Ladinskaya et al. [4.36].

Based on a wide review in 1990 of the data available for terrestrial plants [4.22, 4.36–4.39], Taskayev and Testov [4.40] report a mean value of 11 Bq/kg (DW) with a range of 0.04–111 Bq/kg (DW) for ^{210}Po activity concentrations in grassy plants sampled in areas of normal background radiation.

Activity concentrations of ^{210}Pb and ^{210}Po in areas of high natural background are found to be typically two to three orders of magnitude higher compared to normal areas. Overall, these estimates are in agreement with assessments provided in the 2004 review on ^{210}Po environmental behaviour performed by Coppin and Roussel-Debet [4.8] (see Table 4.3).

TABLE 4.3. CONCENTRATIONS OF ^{210}Po IN TERRESTRIAL PLANTS

Country	Plant	Activity conc. of ^{210}Po (Bq/kg, DW)	Ref.
Areas undisturbed by humans			
Germany	Grass and hay	1.1–29.6	[4.12]
	Grass Heather Juniper Blueberry	22–160 20–88 20–38 34	[4.11]
India	Tobacco	0.1–3.3	[4.14]
Portugal	Vegetables Fruits Trees	0.084 0.06 0.37	[4.41]
United Kingdom	Grass Lichens	6.5–29 290–370	[4.37]
World data	Grass	2.0–35	[4.42]
	Tobacco	5.6–57	[4.22]
Areas disturbed by humans			
Brazil, uranium mine at 1 km	Vegetables	1–8	[4.9]
Canada, uranium mine at Key Lake	Spruce needles Labrador tea	84 1300	[4.10]
Spain, near phosphoric acid factory	*Spartina* spp.	5.1–40.6	[4.18]
United Kingdom, Cotswolds	Grass	4–25	[4.43]

Source: See Ref. [4.8].
Note: The activity concentrations for Portugal are based on fresh weight.

4.3.2. ^{210}Pb and ^{210}Po in animals

Overall, the distributions of ^{210}Pb and ^{210}Po in animals, and their activity concentrations in different animal tissues reflect their intake with feed. The activity concentrations of ^{210}Po in animals vary across four orders of magnitude depending on the animal and organs (see Table 4.4). Lead-210 is largely retained in bones, while ^{210}Po is distributed mainly in soft tissue. Target tissues for ^{210}Po are the spleen, liver and kidneys. The ^{210}Po:^{210}Pb ratio typically exceeds unity in soft tissue; for example, ratios greater than 100 have been reported for the muscle tissue of wild boar [4.51]. However, observed ^{210}Po:^{210}Pb ratios are time dependent and can be substantially modified by continuing radioactive decay from ^{210}Pb, resulting in support for ^{210}Po in older animals.

TABLE 4.4. CONCENTRATIONS OF ^{210}Po IN ANIMAL TISSUES

Tissue	^{210}Po activity conc. (Bq/kg, FW)	Ref.
Meat		
Mutton	0.11–0.43	[4.44, 4.45]
Beef and pork	0.037–2.0	[4.46]
Liver		
Beef, lamb and mutton	0.15–120	[4.11, 4.37, 4.42, 4.47]
Chicken	0.21–1.03	[4.48]
Caribou	332	[4.10]
Kidney		
Beef, lamb and mutton	0.74–67	[4.37, 4.42, 4.47, 4.49]
Gizzards		
Chicken	0.48	[4.48]
Offal	0.19–37 $6.2 \times 10^{-2*}$	[4.22, 4.50]
Milk	$(0.33–6.7) \times 10^{-2}$	[4.12, 4.22]
Eggs	0.11–37	[4.46, 4.48, 4.50]

Source: See Ref. [4.8].

[*] Prepared for consumption.

4.4. OCCURRENCE OF ^{210}Pb AND ^{210}Po IN MARINE ECOSYSTEMS

4.4.1. ^{210}Pb and ^{210}Po in marine water

Polonium enters the marine environment by deposition from the atmosphere at the ocean surface, from the in situ radioactive decay of soluble ^{226}Ra and from the decay of ^{222}Rn gas exhaled from the sea-floor. It also enters the coastal seas with river water and sediment discharges, which includes a component from anthropogenic discharges. The resulting ^{210}Po concentrations in sea water are largely dependent on the magnitude and distribution of these sources, on ^{210}Po binding to suspended particulate matter and on particle scavenging in the water column.

Polonium-210 in coastal waters is largely associated with suspended particulate matter (around 70–80%), probably due to higher suspended loads and intensive mixing in coastal waters than in open ocean waters. Lead-210 in coastal sea water is also mainly associated with the suspended matter, although to a lesser extent than ^{210}Po. The concentrations of these radionuclides in coastal waters do not show a significant seasonal variation, and thus the atmospheric depositions do not significantly modify those concentrations.

In open waters, the ocean is stratified and vertical mixing is reduced. Within the water column, ^{210}Po is produced from radon decay. Given that radon concentrations in the water column increase from a minimum in the surface layer to a maximum in deep-sea water, and the radon distribution is mainly dependent on diffusion in the water column and marine currents, surface water concentrations of ^{210}Po in the open ocean are generally low.

Deposition from the atmosphere results in an excess of ^{210}Pb over ^{226}Ra in the surface layer of the ocean. Absorption of ^{210}Pb and ^{210}Po onto particulates and uptake by phytoplankton and zooplankton generates a downward flux of particulate associated activity. Although these particles are, in part, recycled in intermediate ocean layers, in their settling pathway through the deeper ocean layers, they adsorb and remove soluble ^{210}Pb and ^{210}Po from the water column, thus creating an imbalance between ^{226}Ra and its progeny. Some of the biogenic particulate materials generated in the upper layer of the ocean may reach the deep-sea (abyssal) floor and add ^{210}Pb and ^{210}Po to the top sediment layer. The ^{210}Pb flux arriving at the abyssal sea-floor is around double the ^{210}Pb atmospheric flux entering the oceans at the surface. The mean residence times for ^{210}Po in ocean water layers have been calculated to be around 6–12 months in the upper layer and somewhat longer, around 2 years, in the deep-sea layers [4.52, 4.53].

4.4.2. ^{210}Pb and ^{210}Po in marine organisms

The isotopes ^{210}Pb and ^{210}Po have been well studied in marine wildlife owing to their relatively high concentrations in comparison with those in terrestrial organisms. Currently known ^{210}Pb and ^{210}Po activity concentrations in marine biota cover several orders of magnitude. With a few exceptions in the case of abyssal fauna, ^{210}Po is in excess over ^{210}Pb. Reported ^{210}Po:^{210}Pb ratios are mostly in the range of 1–100 [4.54]. Carvalho [4.54] finds that organisms occupying upper trophic levels (carnivores and top predators) generally display lower ^{210}Po concentrations than planktivorous organisms (primary herbivores). Furthermore, related species occupying equivalent ecological niches in different ecosystems (e.g. filter feeding bivalves in the intertidal zone and in the abyssal sea-floor) display similar ^{210}Po activity concentrations and ^{210}Po:^{210}Pb ratios.

For higher trophic level organisms, such as shrimp, molluscs and fish, the primary source of ^{210}Po uptake is from ingested food, with very little uptake from water [4.55–4.58]. Inside these organisms, the distribution of ^{210}Po is not homogeneous, and the tissues and organs with higher concentrations are those associated with the digestive system.

Concentrations of ^{210}Pb do not increase from phytoplankton to copepods as much as ^{210}Po, and the ^{210}Pb transfer to planktivorous fish, such as sardines, is less efficient than ^{210}Po transfer. Consequently, the ^{210}Po:^{210}Pb ratio in marine food webs appears to increase with the trophic level, at about 10 in phytoplankton and zooplankton, 3–10 in herbivorous fish, 50–100 in piscivores (carnivorous fish) and 200 in the muscle of marine mammals (top predators) [4.59].

4.5. OCCURRENCE OF ^{210}Pb AND ^{210}Po IN FRESHWATER ECOSYSTEMS

4.5.1. ^{210}Pb and ^{210}Po in groundwater

Rainwater infiltrating the ground interacts with soils, sediments and rocks, and so it can accumulate polonium from a number of sources. This includes polonium produced from naturally occurring sources within soils in the unsaturated zone, polonium that has been naturally deposited onto the surface from atmospheric sources, and anthropogenic sources. These sources add to the polonium carried in the rainwater as wet deposition. Polonium is also available from progenitor nuclides in the soils and is derived from similar sources. The infiltrating waters then recharge the underlying groundwater systems.

Weathering of uranium bearing mineral phases will not normally be a major source term for ^{210}Pb and ^{210}Po in groundwater, as it typically occurs at rates that are very long compared to the decay rate of the nuclides. Rather, they are primarily available to the groundwater phase due to recoil following alpha decay of progenitor atoms in the uranium decay series (see Sections 3.1.2 and 7.2). This can take place from within the mineral grains, from absorption sites on grain surfaces, and from within the groundwater phase.

Lead and polonium are both strongly particle reactive under most conditions pertaining to groundwater. Consequently, the largest fraction of activity released from mineral grains is adsorbed onto the aquifer rocks. The distribution of lead and polonium in groundwater is therefore strictly controlled by the adsorption characteristics of the surrounding minerals. Groundwater ^{210}Po concentrations are typically in the range of 1 –30 mBq/L, although values of up to 19 Bq/L have been recorded for brines.

The strong association of ^{210}Po with mineral surfaces and particles, and its short half-life, means that significant transport of activity to a significant distance from the site of production is unusual and measured concentrations generally reflect the local supply rates and bulk adsorption coefficients. The study of ^{210}Po in groundwater is therefore primarily of interest from a human health perspective when the water is to be used for drinking or irrigation, and as a means of studying geochemical processes.

Groundwater discharge into surface waters, such as lakes, rivers and wetlands, as well as into coastal waters. These discharges can carry polonium, although the concentrations may be modified at the sediment–water interface. The relationship between polonium in groundwater and surface waters has not been studied in detail.

4.5.2. ^{210}Pb and ^{210}Po in surface water and sediments

The freshwater environment encompasses lotic (moving) water, such as rivers and streams, and standing water bodies (lentic), such as lakes, ponds, wetlands and bogs. The concentrations of ^{210}Po within moving water (e.g. a river) would be expected to reflect the environmental conditions in the catchment area as well as both natural and anthropogenic sources. The concentrations throughout the year will vary depending on variable precipitation carrying particulates from the surrounding areas to the water body.

The levels of ^{210}Po in surface water in freshwater systems are influenced by in situ decay, fluvial inputs and atmospheric deposition. Within a standing water body (e.g. a lake), there is uptake of ^{210}Pb and ^{210}Po by particulates (particularly biomass) in the water column. As these particulates settle to the bottom sediments, polonium is scavenged from the water column. Polonium has also been shown to become volatile in both fresh and marine waters by the action of microorganisms [4.60].

The levels of ^{210}Pb and ^{210}Po in sediment are influenced by the cycling of iron and manganese as the redox conditions change in the sediment. Overall, a portion of the lead and polonium in sediment may be reintroduced into the water column by diffusive processes; the remainder becomes buried by continual deposition of fresh settling matter. In addition to cycling in sediment, depending on the characteristics of the lake, the bottom layer within the water column may become anoxic during portions of the year, which will influence the levels of polonium in the water column. During the anoxic period, levels of ^{210}Pb and ^{210}Po can be significantly increased in the hypolimnion; due to differences in the cycling, the ^{210}Po:^{210}Pb ratio can be greater than one.

The activity concentrations of ^{210}Po in oxic waters are mostly in the range of 1–5 mBq/L and are higher, up to 17 mBq/L, in seasonally anoxic ponds [4.61]. On a local scale, there can be a large range of typical values.

4.5.3. ^{210}Pb and ^{210}Po in freshwater organisms

The levels of ^{210}Po have been measured in freshwater biota, but there are not as many data available as for the marine environment. When compared to marine biota, a similar pattern in freshwater biota is observed: high concentrations in plankton and lower concentrations in higher trophic biota. The distribution of ^{210}Po within an organism shows higher concentrations in the kidneys, liver and organs of the digestive system. A higher accumulation of ^{210}Po is found in the soft tissue of molluscs than in shells, whereas ^{210}Pb has been detected in higher levels in the shells. In the soft tissue of an organism, ^{210}Po is typically found at levels greater than ^{210}Pb [4.51].

Whole organism CRs for lead (FW) range from approximately 5 (amphibians) [4.62] to as high as 6000 (bivalve molluscs) [4.63]; CRs for polonium are generally higher, from 2000 (fish and vascular plants) up to 1.3×10^5 (bivalves) [4.64, 4.65]. Lead and polonium CRs for wildlife groups in freshwater ecosystems can be found in Table 7.3, Section 7.3.5.

4.6. OCCURRENCE OF ^{210}Pb AND ^{210}Po IN FOOD (INCLUDING SEAFOOD) AND DRINKING WATER

The consumption of food and drinking water is the most important exposure pathway for ^{210}Pb and ^{210}Po for humans. Data on activity concentrations of ^{210}Pb and ^{210}Po in drinking water for different countries are given in Table 4.5. Mean activity concentrations of ^{210}Po in drinking water are in the range of 0.04–7600 mBq/L, and for most regions the upper mean value is less than 4 mBq/L. Furthermore, ^{210}Po concentrations can vary by more than four orders of

TABLE 4.5. ACTIVITY CONCENTRATION OF ^{210}Pb AND ^{210}Po IN DRINKING WATER

Country	Origin	^{210}Pb (mBq/L)		^{210}Po (mBq/L)	
		Mean	Range	Mean	Range
Brazil	Mineral water	66	20–102	—[a]	—[a]
Czech Republic	Public supply	—[a]	<40–350	9	<2–71
Finland	Potable water	—[a]	0.2–21 000	—[a]	0.2–7 600
Germany	Potable water	1.5	0.2–170	0.5	0.1–40
India (Goa)	Potable water	—[a]	2.2–11.5	—[a]	1.7–7.0
Italy	Tap water Mineral water	—[a]	—[a]	—[a]	0.13–5.9 <0.04–24
Poland	Potable water	1.6	—[a]	0.5	—[a]
Portugal	Public supply	18.5	2–390	—[a]	—[a]
Romania	Water supply	—[a]	7.0–44	—[a]	7.0–44
Russian Federation	Water supply	26.9	2.3–68	2.72	1.2–7.1
Spain	Public supply	18.7	0.23–59	3.63	1.8–19.3
United Kingdom	Public supply	—[a]	40–200	—[a]	—[a]
USA	Potable water	—[a]	0.1–1.5	—[a]	—[a]

Source: See Ref. [4.66].

[a] —: data not available.

magnitude because of the diversity of water sources. High activity concentrations of ^{210}Po are found in areas of high radiation background in Finland (up to 7600 mBq/L), while the lowest values are typical for public water supply systems in countries with efficient water quality control systems, such as Italy, where the concentrations of ^{210}Po in tap water are in the range of 0.1–5.9 mBq/L. Activity concentrations of ^{210}Pb in drinking water are generally two to fivefold higher than those measured for ^{210}Pb. The mean value for most countries is in the range of 1.5–26.9 mBq/L.

Concentrations of ^{210}Pb and ^{210}Po in drinking water vary over a wide range because of variations in the concentrations of uranium and its long lived progeny, such as ^{226}Ra and ^{230}Th, in adjacent rocks, soil, atmosphere and groundwater. High levels of ^{210}Pb and ^{210}Po have been found to be associated with regions of uranium or thorium ore bodies. Mining and processing of uranium and other minerals, and geothermal springs also lead to elevated concentrations of ^{210}Pb and hence ^{210}Po in the environment.

Concentrations of ^{210}Pb and ^{210}Po in foods also vary over a wide range, reflecting differences in uranium and its progeny in soil, the atmosphere and water, differing farming practices and local features in food processing (see Tables 4.6 and 4.7).

TABLE 4.6. CONCENTRATIONS OF ^{210}Pb AND ^{210}Po IN FOODS

Product	Region	Country	^{210}Pb (mBq/kg)	^{210}Po (mBq/kg)
Milk products	North America	USA	11	—[a]
	Asia	China	16	13
		India	—[a]	15
	Europe	Germany	5–280	2–80
		Poland	18	16
		Romania	10–15	13–140
		United Kingdom	35–88	20–220
	Reference value		15	15
Meat products	North America	USA	18	—[a]
	Asia	China	140	120
		India	—[a]	440
	Europe	Germany	100–1 000	37–4 000
		Poland	98–105	99–102
		Romania	15–19	38–110
		United Kingdom	40–3 700	62–67 000
	Reference value		80	60

TABLE 4.6. CONCENTRATIONS OF ^{210}Pb AND ^{210}Po IN FOODS (cont.)

Product	Region	Country	^{210}Pb (mBq/kg)	^{210}Po (mBq/kg)
Grain products	North America	USA	33–81	—[a]
	Asia	China	34	42
		India	—[a]	15–120
	Europe	Germany	40–4 000	37–1 900
		Poland	110–160	90–140
		Romania	49–59	20–360
		United Kingdom	56–120	27–260
	Reference value		50	60
Leafy vegetables	North America	USA	41	—[a]
	Asia	China	360	430
		India	—[a]	320
	Europe	Germany	4–4 100	4–7 400
		Poland	43–51	40–67
		United Kingdom	16–3 300	37–3 300
	Reference value		80	100
Root vegetables and fruits	North America	USA	8–150	—[a]
	Asia	China	27	29
		India	—[a]	16–140
	Europe	Germany	20–4 900	22–5 200
		Poland	24–93	28–210
		Romania	19–44	12–140
		United Kingdom	18–76	—[a]
	Reference value		30	40
Fish products	North America	USA	14–1 800	150–55 000
	Europe	Germany	20–4 400	50–5 200
		Poland	81–93	3 100–3 800
		Portugal	—[a]	80–120 000
		United Kingdom	180–4 800	60–53 000
	Reference value		200	2 000

Source: See Ref. [4.67].

[a] —: data not available.

TABLE 4.7. CONCENTRATIONS OF ^{210}Pb AND ^{210}Po IN MUSCLES OF COMMON SEAFOOD

Species	Body weight (g)	No. of samples	^{210}Po (Bq/kg, FW)	^{210}Pb (Bq/kg, FW)
Sardine	51	4	66 ± 2	1.0 ± 0.02
Anchovy	5.5	6	9.4 ± 0.3	0.42 ± 0.01
Mackerel	120	4	19 ± 1	0.63 ± 0.4
Horse mackerel	200	2	5.2 ± 0.2	0.1 ± 0.02
Bigeye tuna	31×10^3	1	3.1 ± 0.09	0.46 ± 0.02
Hake	256	2	6.7 ± 0.3	0.15 ± 0.01
Red sea bream	275	1	2.4 ± 0.09	0.84 ± 0.02
Common sole	150	1	1.4 ± 0.04	0.14 ± 0.01
Skate	894	1	0.73 ± 0.03	0.12 ± 0.000 4
Squid	2 103	1	1.6 ± 0.04	0.41 ± 0.01
Common shrimp	—[a]	12	49 ± 1.5	1.1 ± 0.03
Clam	—[a]	6	152 ± 19	2.9 ± 0.1
Cockle	—[a]	5	9.4 ± 3	1.32 ± 0.06
Mussel	—[a]	12	132 ± 5	2.6 ± 0.1

Source: See Ref. [4.68].
Note: All samples were taken from the north-eastern Atlantic Ocean, off Portugal.
[a] —: data not available.

The lowest ranges in ^{210}Pb and ^{210}Po concentrations are in milk products. Two to sixfold higher concentrations were observed in meat and plant products, where root vegetables had slightly lower mean ^{210}Pb and ^{210}Po concentrations compared with other plant products. The highest concentrations were found to be in fish products, and leafy vegetables demonstrated ^{210}Pb concentrations higher than in grain products. High ^{210}Po concentrations are consistently found in seafood products, and with ^{210}Po:^{210}Pb ratios much higher than unity, usually 2–100 [4.67–4.69].

The great importance of ^{210}Po to dietary intake is found in many coastal countries, such as Japan, the Marshall Islands, Portugal and South Africa [4.67]. A global review of ^{210}Po in marine food suggests that representative concentrations are 2.4 Bq/kg in fish, 6.0 Bq/kg in crustaceans and 15.0 Bq/kg in molluscs [4.70].

Consumption of seafood varies widely, both between countries and within a single country. In addition, ^{210}Pb and ^{210}Po concentrations in seafood species vary within three orders of magnitude (see Table 4.7). If representative consumption rates are 13 kg/a of fish and 1 kg/a each of molluscs and crustaceans, the intake of ^{210}Po with these foods would be 52 Bq/a [4.67].

If there are processing or distribution delays for fish products between catch and consumption, the activity intake will be reduced owing to the radioactive decay of ^{210}Po. Statistics quoted by Aarkrog et al. [4.70] indicate that 30% of seafood is eaten fresh, 30% frozen, 20% smoked and 20% canned. Application of a correction factor of 0.6 [4.67] suggests an intake of 31 Bq/a in seafood and a weighted concentration of ^{210}Po in fish products of 2.1 Bq/kg.

4.7. ANTHROPOGENIC SOURCES OF ^{210}Po IN THE ENVIRONMENT

4.7.1. Industrial uses of human-made ^{210}Po

There are a number of industrial uses of ^{210}Po, in particular as an alpha emitter to eliminate static in manufacturing environments and to remove dust under clean conditions. Owing to its short half-life, the ^{210}Po source needs to be replaced approximately every twelve months. It has also been investigated as a heat source for thermoelectric power devices for space applications, since the energy released by decay is so great (140 W/g).

In nature, ^{210}Po occurs in extremely low concentrations; even uranium ores contain less than 0.1 mg/t. However, ^{210}Po is generated in nuclear reactors when stable ^{209}Bi is bombarded with neutrons. Around 100 g of ^{210}Po is produced annually, largely at a single facility in the Russian Federation. There have not been any widely reported releases of ^{210}Po from such facilities.

Earlier designs of nuclear weapons included a neutron source of beryllium and polonium, and so weapons programmes required additional production of ^{210}Po. In the United Kingdom, this took place at Windscale Works, Sellafield. A nuclear reactor fire there in 1957 led to the environmental release of radionuclides. In addition to a study published in 1996 on the magnitude of the release of ^{210}Po (see Ref. [4.71]), a 2007 study of ^{137}Cs, ^{131}I and ^{210}Po estimated an approximate release of 40 TBq of ^{210}Po, with a range of 14–110 TBq [4.72].

4.7.2. Environmental release of naturally occurring radioactive material

Naturally occurring ^{210}Po is continuously released from geological materials processed for industrial use. In a 1976 study, Moore et al. [4.73] estimate that anthropogenic sources could constitute up to 7% of the total ^{210}Po fluxes in the atmosphere, while elevated concentrations in soils and waters are found near related industrial activities.

4.7.2.1. Phosphate fertilizer and phosphogypsum waste

Phosphate rich rocks are extensively mined, largely to produce phosphate fertilizer. These rocks, especially those of sedimentary origin, can have high concentrations of uranium and daughter ^{210}Po. Both the products and waste of these operations can therefore release radionuclides to the environment [4.74, 4.75]. Phosphate rocks contain up to 3700 Bq/kg of uranium [4.75], with high uranium content generally corresponding to high phosphorus content. Physical processing of ore can increase radionuclide concentrations by up to 300% [4.76]. During chemical processing, phosphate rock is reacted with sulphuric acid and converted into phosphoric acid to make fertilizer, while phosphogypsum is generated as a by-product. While uranium and ^{210}Pb primarily partition into the phosphoric acid, much of the ^{226}Ra and up to 99% of the ^{210}Po enter the phosphogypsum [4.74, 4.77].

Most phosphogypsum is stored in piles (referred to as stacks), although some is discharged into surface waters. A study in the Netherlands examined the impact of phosphogypsum discharged by the fertilizer industry into the Rhine (~1.6 TBq/a) and transported by suspension to the North Sea [4.78]. The ^{210}Pb and ^{210}Po settle with particulates, with some of the ^{210}Po becoming involved in biological activity in the upper marine water column (see Chapter 8). The sediments thus remain a potential source of ^{210}Po, although much of the ^{210}Pb and ^{226}Ra — and hence probably the daughter ^{210}Po — is trapped within insoluble fractions [4.79].

Radionuclides can also escape from processing plants or phosphogypsum storage stacks. For example, a study around a fertilizer plant in the Syrian Arab Republic found elevated levels of ^{210}Po in surrounding soil, water and vegetation, attributable to deposition of daughter isotopes of ^{222}Rn escaping from the facility [4.80]. A fertilizer production facility along the Tagus estuary, Portugal, has been found to contribute radionuclides to near-shore sediments through discharge of process waters and leaching by rainwater of phosphogypsum piles [4.81]. Overall, the radiological impact of naturally occurring radioactive material industries on the European population is dominated by discharges from fertilizer plants (see Ref. [4.82]).

The spread of phosphate fertilizer, containing ^{210}Pb, which decays to ^{210}Po, can contribute to the naturally occurring radionuclides in soils. Phosphogypsum, containing ^{210}Po, has also been used as a soil amendment for depleted soils in some countries (see Ref. [4.83]).

4.7.2.2. Fossil fuel production and combustion

Fossil fuels are often rich in uranium series nuclides, since the precursor organic material was accumulated under reducing conditions favourable for incorporation of uranium. Petroleum in the United States of America can contain 0.1–40 Bq/kg of ^{226}Ra [4.82], presumably with similar ^{210}Pb and ^{210}Po activities. Production waste contains high levels of radium, with on average approximately 18 000 Bq/kg of radium in scale and 2800 Bq/kg of radium in sludge. These wastes can be significant sources of ^{210}Pb and ^{210}Po when discharged, landfilled or disposed of by land spreading [4.84]. Waters discharged from production wells can have high concentrations of ^{226}Ra, although low levels of ^{210}Po. For example, waters from Norwegian wells have up to 16 Bq/L of ^{226}Ra but only 0.2–6 mBq/L of ^{210}Po [4.85].

The behaviour of ^{210}Po in coal combustion power plants is considered by Mora et al. [4.86]. Coal typically has concentrations of 16–52 Bq/kg of ^{210}Po [4.75], which is volatilized during combustion. More than half of this is condensed onto fly ash, although some also condenses within the installations, and a small fraction remains in bottom ash. It is expected that ^{210}Pb behaves similarly. Most of the process residues are deposited onto land, although some fly ash is used in building materials [4.75].

4.7.2.3. Metal production

One clear potential source of uranium series radionuclides is tailings from uranium and radium mining sites, where leaching and runoff can disperse contaminants. As discussed in Chapter 6, lead and polonium are relatively immobile, except under highly acidic conditions. Radium is also relatively immobile [4.87], and while uranium is more readily transported, in the absence of other intermediate radionuclides, there will not be substantial ^{210}Po ingrowth over short timescales (see Chapter 3). Therefore, the spread of ^{210}Po, and of ^{210}Pb followed by ingrowth of ^{210}Po, is often through discharge of acidic waters and waste pile leachates, and the transport of airborne particles and suspended sediments. The escape of ^{222}Rn to the atmosphere and subsequent deposition of daughter nuclides is also an important route for spreading ^{210}Po (see Chapter 5). Some of the geochemistry controlling radionuclide behaviour is summarized in Ref. [4.88]. There have been various documented releases from mine tailings, in particular acute failures of containment structures: major events are listed in Ref. [4.89] and an example of the release of ^{210}Po to the environment is presented in chapter 7 of Ref. [4.90].

Ore processing for other metals can also release ^{210}Po and other radionuclides. For example, Baxter et al. [4.91] report that raw materials for a large tin smelter in the United Kingdom generally contain up to 2×10^3 Bq/kg of ^{210}Po, and there was considerable loss of ^{210}Po in stack emissions, although high concentrations were not found in surrounding soils. The technological enhancement of radionuclides in primary products or waste materials can occur during the processing of other metals as well [4.92]. Separation of zirconium also releases radionuclides from zircon, which contains high concentrations of uranium [4.93].

4.8. SUMMARY

The isotopes ^{210}Pb and ^{210}Po are widely distributed in the Earth's crust and soil. As polonium is only ever present at ultratrace levels and lead is usually present at trace levels, their behaviour is heavily influenced by major ion chemistry and processes such as adsorption, co-precipitation, and attachment to colloids in aqueous systems and to aerosols in the atmosphere.

The activity concentrations of ^{210}Pb and ^{210}Po in the topsoil vary over a wide range, with a range in most soils of 10–200 Bq/kg. The ^{210}Po:^{210}Pb ratio in soil is quite close to unity, implying that they are in secular equilibrium in soils.

Major sources to the atmosphere are decay of ^{222}Rn following its exhalation from soils, and volcanic emissions (particularly for ^{210}Po), although there are several other minor sources. Removal is primarily by deposition to the Earth's surface with precipitation.

For plants, ^{210}Po:^{210}Pb activity ratios are in the range of 0.2–0.7 in above ground biomass and 0.96–1.3 in roots, indicating different mobilities of ^{210}Pb and ^{210}Po in the soil–plant system as well as within plants. A greater variability in concentrations is observed for animals, both terrestrial and aquatic. Lead-210 is largely retained in bones, while ^{210}Po is distributed mainly in soft tissue. Target tissues for ^{210}Po are the spleen, liver and kidneys. Relatively high concentrations of ^{210}Po have been measured in marine animals, and ingestion of seafood is an important pathway for radiological dose to humans.

Both lead and polonium are strongly particle reactive under most conditions pertaining to groundwater, and most of the available activity is adsorbed onto aquifer rocks. Their distribution is therefore strictly controlled by the adsorption characteristics of the surrounding minerals. Groundwater ^{210}Po concentrations are typically in the range of 1–30 mBq/L, although values up to 19 Bq/L have been recorded for brines.

The realistic behaviour of ^{210}Po in the environment can be described if based on both predictions of environmental behaviour of ^{210}Pb as a source and transfer of ^{210}Po itself, and so the environmental behaviour of these radionuclides needs to be considered together.

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Chapter 5

^{210}Pb AND ^{210}Po IN ATMOSPHERIC SYSTEMS

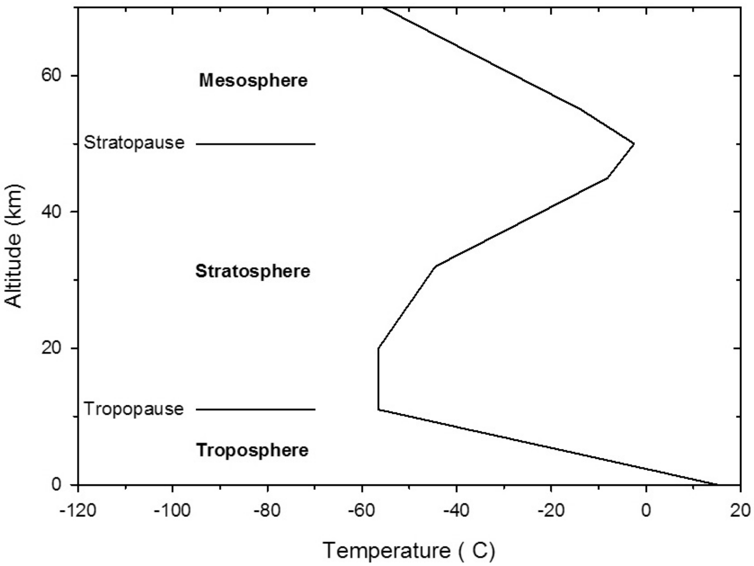
5.1. INTRODUCTION TO THE ATMOSPHERIC ENVIRONMENT

The depth of the gaseous envelope surrounding Earth is not firmly fixed but is estimated to extend, at a maximum, approximately 1000 km from the surface of the crust. Despite comprising less than 15% of the total planetary radius, the nature of the atmospheric system as a whole has an overwhelming influence on almost all the biological and geochemical conditions on Earth. Not only does it affect human exposure to external influences and how its processes occur, it also influences many of the Earth's physical characteristics.

The temperature and pressure of the atmosphere vary considerably with height above the surface. Pressure decreases exponentially with height, and altitude is often given in units of pressure (hPa) rather than distance. The temperature variation is more complex and is a crucial determinant of atmospheric mixing processes. The rate of decrease in temperature with height is termed the temperature lapse rate (°C/km), whose sign changes several times with height, giving rise to identifiable layers in the atmosphere (see Fig. 5.1). In the lowest of these, the troposphere, the lapse rate is normally positive (i.e. temperature decreases with height), resulting in rapid vertical mixing. In the stratosphere, the lapse rate is zero or negative due to absorption of solar ultraviolet radiation by ozone, and vertical mixing is slow. Above the stratosphere lie the mesosphere, thermosphere and exosphere. Together, the troposphere and stratosphere comprise the lower atmosphere, which is the region of interest here.

The actual temperature variations with height are crucial determinants of atmospheric mixing processes and are more complex than those shown in Fig. 5.1. They depend not only on the time of the year but also on variations in the structure of the atmosphere: for example, the tropopause (the boundary between the troposphere and stratosphere) is around 8 km over the poles to but 16 km over the tropics.

The troposphere can be further divided into the planetary boundary layer (PBL) and the free atmosphere. The behaviour of the PBL is strongly influenced by the Earth's surface and the wind within it is affected by surface drag. Furthermore, turbulence is common and vertical mixing is generally quite strong. However, a negative temperature lapse rate can exist within the PBL, leading to a layer of stable air above the surface called an inversion layer. This occurs when the Earth's surface is cooler than the air above, such as on nights with clear skies.



Source: See Ref. [5.1].

FIG. 5.1. Vertical structure of the lower atmosphere.

The height of the PBL is about 1 km, although this depends on surface features and atmospheric conditions.

In the atmosphere, ^{210}Bi, ^{210}Pb and ^{210}Po are attached to aerosol particles, with a diameter distribution typically covering several orders of magnitude, from around 10^{-3} µm to 10^{2} µm. The behaviour of these radionuclides reflect those of the particles to which they are attached. Airborne particles may be broadly classified into coarse particles (>1 µm) and fine particles (<1 µm). Fine particles may be further categorized into Aitken nuclei (primary aerosols: 10^{-3}–10^{-1} µm) and particles in the accumulation range (10^{-1}–10^{0} µm). These size range classifications are approximate and vary depending on the situation [5.2, 5.3].

Coarse particles are primarily generated by mechanical means (e.g. dust resuspension by wind), whereas fine particles originate from processes such as condensation of gases. Aitken nuclei form particles in the accumulation range through condensation and coagulation. Sedimentation (gravitational settling) is a significant removal mechanism for coarse particles, but this is not the case for fine particles.

The size distribution of particles to which radionuclides are attached depends on several factors, including the source of the radionuclide and the length of time it has been suspended. For example, airborne ^{210}Pb originating from resuspension of dust will be predominantly on coarse particles, whereas that produced by the decay of free ^{222}Rn present in the atmosphere is predominantly on fine particles. In the latter case, on initial production, a large proportion of the ^{210}Pb is attached to Aitken particles, but with time the proportion in the accumulation mode increases.

5.2. SOURCES OF ^{210}Pb AND ^{210}Po IN THE ATMOSPHERE

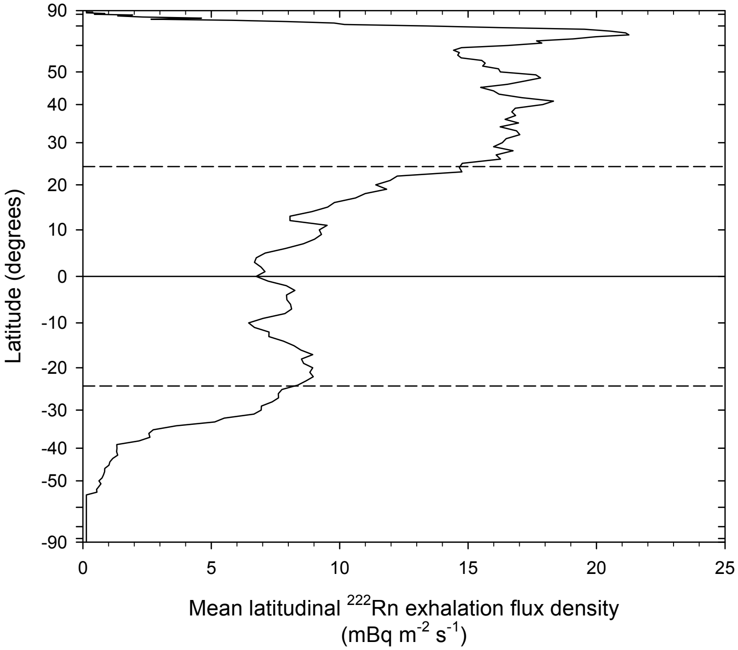
The primary source of ^{210}Pb in the atmosphere is ^{222}Rn exhalation from the Earth’s surface (mainly from soil) and its subsequent decay via short lived radon progeny to ^{210}Pb. There are several other sources, including volcanic activity, resuspension of soils, forest and savanna fires, and combustion of fossil fuel. However, these other sources have been estimated to contribute less than 5% to the total ^{210}Pb budget.

Exhalation of ^{222}Rn from the Earth’s surface with subsequent decay through ^{210}Pb to its progeny is one of the primary sources of ^{210}Po in the atmosphere. However, due to the relative volatility of polonium, some other sources are important. In particular, volcanic activity has been estimated to be as important a contributor to the total budget as the exhalation of ^{222}Rn. Each of these sources has different characteristics in terms of spatial and temporal variability, and each has a different influence on the ^{210}Po:^{210}Pb ratio and on the aerosol size distribution to which ^{210}Pb and ^{210}Po are attached [5.4]. Overall global fluxes for ^{210}Pb and ^{210}Po have been estimated to be approximately 3.64×10^{16} Bq/a and 3.82×10^{15} Bq/a, respectively [5.5].

5.2.1. Radon exhalation

Radon exhalation flux densities from soil are dependent on a number of factors, in particular the ^{226}Ra activity concentration in the soil, soil moisture, bulk density and porosity [5.6]. Although flux densities vary over several orders of magnitude, sites where they are highly elevated are small on a regional or global scale, and for the majority of soils, they are in the range of 10–50 $\text{mBq}\cdot\text{m}^{-2}\cdot\text{s}^{-1}$. Radon exhalation flux densities from the ocean surfaces have been estimated to be about two orders of magnitude lower than those from ice-free land surfaces [5.5, 5.7]. As a result of these factors, flux densities are highly variable over the Earth’s surface, and the major influences are the proportion of ice-free land followed by soil properties, including the radium activity concentration [5.8].

Figure 5.2 shows the latitudinal dependence of radon flux densities, based on the flux model of Schery and Wasiolek [5.7]. In this plot, the latitude axis is scaled such that the area enclosed by the curve between any two latitudes is



Source: Figure 1 of Ref. [5.9].

FIG. 5.2. Estimated mean annual latitudinal radon exhalation flux density.

proportional to the radon flux (Bq/s) for that area [5.10]. The region between the Tropics of Cancer and Capricorn contributes around 38% to the total flux. Mean latitudinal exhalation ^{222}Rn flux densities drop dramatically south of 30°S and north of 70°N owing to the presence of large oceans and ice-covered land areas.

The southern hemisphere contributes only about 26% of the Earth's total radon flux. The radioactive half-life of radon (3.82 days) and the mean lifetime of ^{210}Pb in the PBL, about 5–7 days [5.11], are considerably smaller than the timescale required for mixing of air between the northern and southern hemispheres. As a consequence, the lower radon flux results in lower average ^{210}Pb and ^{210}Po concentrations in the air, and lower deposition fluxes to the Earth's surface in the southern hemisphere [5.12].

The radon exhalation flux density for ice-free soil is affected by a number of factors. Of the temporally variable factors, soil moisture has the greatest influence [5.13]. The relationship is complex, but rainfall generally results in a reduction in exhalation. Most studies of seasonal variability show reductions in the wetter months: winter in temperate zones [5.14] and wet monsoon in the tropics [5.15]. However, the variability is relatively small over an interannual time frame.

Following exhalation, radon decays in the atmosphere to a series of short lived progeny (^{218}Po, ^{214}Pb, ^{214}Bi and ^{214}Po), which attach to fine (submicron) aerosol particles. The activity concentrations of these short lived progeny have been observed to be high, resulting in increased gamma dose rates close to the ground during, and immediately following, rainstorms [5.16, 5.17].

5.2.2. Volcanic activity

Volcanic activity releases thorium and uranium series radionuclides to the atmosphere, including ^{210}Pb, ^{210}Po and ^{222}Rn, with ^{210}Po in excess over ^{210}Pb in volcanic plumes due to its volatility [5.18, 5.19]. The excess is sufficiently high for ^{210}Po to be used as a tracer for long range transport of volcanic plumes [5.20]. Measurements close to Mount Sakurajima, Japan, show that ^{210}Po is predominantly attached to fine particles smaller than 2 μm [5.21].

The fluxes of ^{210}Pb and ^{210}Po to the atmosphere from this source have been estimated to be 6.0×10^{13} Bq/a and 2.4×10^{15} Bq/a, respectively [5.22]. In the case of ^{210}Po, this is of a similar order of magnitude to that from ingrowth following ^{222}Rn exhalation from soil. However, this flux is difficult to quantify accurately. Moreover, it greatly varies depending on the timing and location of volcanic activity. One approach is to use of sulphate measurements as an indicator for the influence of volcanic activity, in conjunction with ^{210}Pb and ^{210}Po measurements [5.23]. Volcanic eruptions can also lead to an injection of ^{210}Pb and ^{210}Po into the stratosphere; in a 1979 study, Lambert et al. [5.24] estimate this flux to be about 2% of that into the troposphere.

5.2.3. Resuspended soil dust

The suspension of dust can in some cases be a significant contributor to concentrations of ^{210}Pb and its progeny in the atmosphere. This is especially true in the PBL over continents. The sources often contribute only on a local or regional scale, owing to the significant role sedimentation plays in the removal of resuspended particles to the surface. Nevertheless dispersion can be significant, even over relatively large distances [5.20].

Globally, the flux has been estimated at 3.3×10^{14} Bq/a for ^{210}Pb and ^{210}Po [5.5, 5.25], which means it contributes around 1% and 10%, respectively, of the total budgets from all sources. However, because it primarily affects the PBL above significant land masses, it can be a very significant contributor to the observed concentrations at ground level.

The isotopes ^{210}Bi, ^{210}Pb and ^{210}Po are normally in secular equilibrium in topsoils, and ^{210}Pb is also normally in excess over its progeny in the troposphere. Consequently, the resuspension of dust has the effect of increasing the ^{210}Bi:^{210}Pb and ^{210}Bi:^{210}Po ratios. This can contribute to difficulties with using ^{210}Pb–^{210}Bi and ^{210}Pb–^{210}Po couples to calculate atmospheric ^{210}Pb residence times (see Section 5.4). One approach to counter this is to use a determination of a member of the uranium decay chain above ^{210}Pb to apply a correction [5.26, 5.27].

5.2.4. Biomass burning

Polonium-210 has been shown to be enriched in fire plumes, due to the volatility of elemental polonium and polonium compounds at temperatures in the low hundreds of degrees Celsius [5.28]. Estimates of total worldwide biomass burning are around (2–5) × 10^{13} kg CO_2 released per year [5.29], and the carbon content of dried plant materials is around 45–50% [5.30]. As noted in Chapter 4, concentrations of ^{210}Po in vegetable matter vary considerably, but an indicative average value is 10 Bq/kg. This would give a total annual flux of ^{210}Po from this source of around 2 × 10^{14} Bq/a if all of the available ^{210}Po were released. This release would be at ground level, and the ^{210}Po would be available for removal by wet and dry deposition processes.

On a global and time averaged scale, biomass burning should not be a significant source of ^{210}Po (nor of ^{210}Pb). Although in the case of large burning events, it can have a local or regional effect [5.20, 5.31]. The convective draught from fires can also transport ^{222}Rn to the free troposphere, with subsequent decay to ^{210}Pb [5.32]. Biomass burning is discussed in more detail in Appendix IV.

5.2.5. Wind blown sea spray and emission of volatile polonium compounds from water surfaces

The isotopes ^{210}Pb and ^{210}Po are enriched in the ocean surface microlayer, with the ^{210}Po:^{210}Pb activity ratio at around 2 [5.33]. Wind blown sea spray has been shown to be a minor contributor to the atmospheric budget, estimated at 3.7 × 10^{11} Bq/a and 7.4 × 10^{11} Bq/a for ^{210}Pb and ^{210}Po, respectively [5.5, 5.25]. In addition, there is evidence that volatile compounds, such as dimethyl polonide, can be formed and released from ocean and fresh water surfaces [5.34–5.36].

5.2.6. Anthropogenic sources

There are many anthropogenic sources of ^{210}Pb and ^{210}Po. The most significant are fossil fuel (particularly coal) burning and dispersion of phosphate fertilizers and gypsum by-products. Global fluxes are estimated to be

8.2 × 10^{12} Bq/a from fossil fuels for both ^{210}Pb and ^{210}Po, and 2.0 × 10^{13} Bq/a and 6 × 10^{11} Bq/a from phosphate for ^{210}Pb and ^{210}Po, respectively [5.5]. In addition, much of the world's biomass burning is anthropogenic (see Section 5.2.4).

Fossil fuel burning and most other anthropogenic sources impact most heavily on concentrations in urban areas. A study in Seoul, Republic of Korea [5.37], concludes that the dominant fraction of ^{210}Po in precipitation samples is linked to anthropogenic sources, primarily from the burning of coal, with minor contributions from biomass burning.

5.3. PROCESSES FOR THE REMOVAL OF ^{210}Pb AND ^{210}Po FROM THE ATMOSPHERE

Radioactive decay is only a minor contributor to the removal of ^{210}Pb and ^{210}Po on account of their short residence times in the troposphere. The primary removal pathway is wet deposition to the Earth's surface, with a secondary pathway being dry deposition. Using the estimates of global source terms for ^{210}Pb and ^{210}Po given in Section 5.2, the average global deposition flux density over the Earth's surface should be approximately 71 $\mathrm{Bq{\cdot}m^{-2}{\cdot}a^{-1}}$ and 7.5 $\mathrm{Bq{\cdot}m^{-2}{\cdot}a^{-1}}$ for ^{210}Pb and ^{210}Po, respectively. However, annual total deposition flux densities for ^{210}Pb range from several $\mathrm{Bq{\cdot}m^{-2}{\cdot}a^{-1}}$ (as observed in Antarctica) to several hundred $\mathrm{Bq{\cdot}m^{-2}{\cdot}a^{-1}}$.

5.3.1. Wet deposition

Wet deposition refers to removal of atmospheric constituents to the ground surface by precipitation (e.g. rain, snow and hail). Globally, wet deposition is the primary process for removal of ^{210}Pb and ^{210}Po from the troposphere, and occurs mainly by precipitation from either stratiform or convective clouds. The removal of ^{210}Pb and ^{210}Po essentially reflects the removal of the aerosol particles to which they are attached. The term rainout refers to the removal of aerosol which has been scavenged within the cloud, while washout refers to scavenging which occurs below the cloud during the precipitation event.

The concentrations of ^{210}Pb and ^{210}Po in precipitation are a complex result of several factors and include the following:

- Their concentrations in the air column;
- The size distribution of the aerosol to which they are attached;
- The effective height of the precipitating column;
- The relative contributions from rainout and washout;

- — The growth in raindrop size within the cloud due to ascent in convective updrafts;
- — Evaporation of the raindrop during its descent;
- — The type and duration of rainfall.

Reported values for ^{210}Pb concentration in rainwater vary considerably at different locations, being most commonly in the range of 10–1000 mBq/L[3] with ^{210}Po:^{210}Pb activity ratios of 0.005–1 or even greater. Some measured concentrations in rainwater are provided in Table 5.1, showing the variability in concentrations between locations and over extended periods at one location.

In many cases, ^{210}Pb and ^{210}Po activity concentrations are greater at the start of the precipitation event than later in the event, probably primarily due to the reduction in washout as the air below the cloud becomes depleted [5.27, 5.43]. A related effect is that activity concentrations in the precipitation are generally inversely proportional to the amount of precipitation (e.g. see Refs [5.39, 5.40, 5.44]).

TABLE 5.1. ^{210}Pb AND ^{210}Po CONCENTRATIONS AND ^{210}Po:^{210}Pb ACTIVITY RATIOS IN RAINWATER

Location	^{210}Pb (mBq/L)		^{210}Po (mBq/L)		^{210}Po:^{210}Pb		Ref.
	Range	Avg.	Range	Avg.	Range	Avg.	
Izmir, Turkey	9–198	51	2–35	8	0.03–1.09	—[a]	[5.38]
Geneva, Switzerland	60–3300	—[a]	—[a]	—[a]	—[a]	—[a]	[5.39]
Jabiru, Australia	46–1700	147	0.9–790	20	0.02–0.46	0.14	[5.27]
Galveston, USA	34–3600	132	—[a]	—[a]	—[a]	—[a]	[5.40]
Detroit, USA	105–1233	472	2.2–126	23	0.005–0.283	0.049	[5.41]
Monaco	—[a]	—[a]	—[a]	—[a]	—[a]	0.25	[5.42]

[a] —: data not available.

5.3.2. Dry deposition

Globally, dry deposition accounts for about 10–15% of total deposition for ^{210}Pb [5.5, 5.45]. However, this proportion varies spatially and temporally. The contribution from dry deposition is greater for arid regions and can exceed the wet fallout in these areas [5.46].

Dry deposition of ^{210}Pb and ^{210}Po consists of two components: activity present on resuspended dust; and activity from other sources and primarily attached to fine particles. Deposition of activity on resuspended dust will be more strongly influenced by sedimentation, although under some conditions such dust can travel thousands of kilometres. Cases of such long distance transport have been well documented for dusts originating from subtropical arid regions such as North Africa and the Middle East, Central Asia and Australia [5.20].

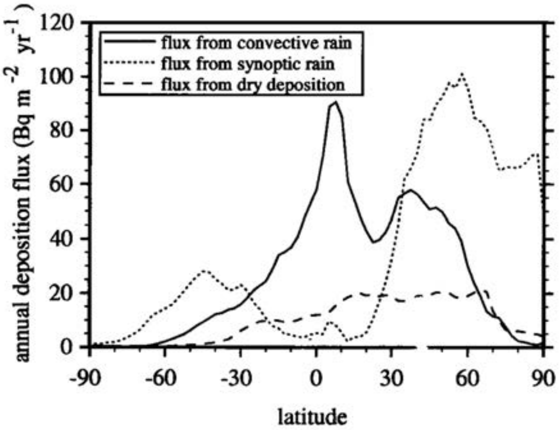
On its resuspension, the ^{210}Po:^{210}Pb activity ratio of dust can be expected to be approximately one. Since coarse particles may dominate the dry deposition, this can result in a significantly higher ^{210}Po:^{210}Pb ratio than in wet deposition [5.41].

5.3.3. Deposition fluxes

Figure 5.3 shows the results of an aerosol transport model simulation for deposition flux density of ^{210}Pb on a latitudinal basis [5.47]. There is a peak in deposition from convective rain in the tropics. The greater deposition flux in the northern hemisphere compared with the southern hemisphere is due to the higher ^{222}Rn source term (see Fig. 5.1 and Section 5.4.1). The very low deposition flux in Antarctica is due to a combination of the low southern hemisphere source term, the low precipitation rate, the lack of significant ice-free land masses close to Antarctica, and the band of strong westerly winds around 40–50°S, which act as a barrier to low level movement of air from the land masses to the north.

As discussed, wet deposition dominates the total deposition at most locations, and Fig. 5.3 shows that this is the case for all latitudinal bands. The annual wet deposition flux ($\mathrm{Bq \cdot m^{-2} \cdot a^{-1}}$) of a radionuclide can be calculated from the product of the total precipitation over the year and the (precipitation weighted) average concentration in the rainwater. Hence, both the amount of precipitation and the concentration are important in evaluating these fluxes.

An example of the importance of multiple factors in determining deposition fluxes is the waters on the east of Japan, where ^{210}Pb deposition fluxes of up to 800 $\mathrm{Bq \cdot m^{-2} \cdot a^{-1}}$ have been recorded. These high fluxes are due to a combination of air masses with high ^{210}Pb concentrations from the surface layer of the Asian continent being moved by strong winter monsoons, mixing with ascending convection clouds over the body of water between Japan and the Asian mainland,



Source: Figure 3 of Ref. [5.47].

FIG. 5.3. Zonally averaged annual deposition flux of ^{210}Pb *(*$Bq{\cdot}m^{-2}{\cdot}a^{-1}$*) due to convective rain, synoptic rain and dry deposition (reproduced with permission courtesy of American Geophysical Union).*

and subsequent precipitation (rainfall and snowfall) over the coast and mountain ridge of Japan with high ^{210}Pb concentrations in the precipitation [5.44, 5.48]. High ^{210}Po deposition fluxes and elevated ^{210}Po:^{210}Pb ratios have also been recorded in this region [5.49].

The annual amount of precipitation is usually a very strong determinant of depositional fluxes. This is true both between locations, and for interseasonal and interannual variability at the same location [5.39, 5.50]. However, Beks et al. [5.51] report that the ^{210}Pb depositional fluxes at two sites in the Netherlands were mainly correlated with the number of times that there was heavy rain or thunderstorms, rather than annual rainfall. The deposition fluxes were approximately 75 $Bq{\cdot}m^{-2}{\cdot}a^{-1}$, which is considerably lower than the studies in Japan on account of the predominantly westerly winds.

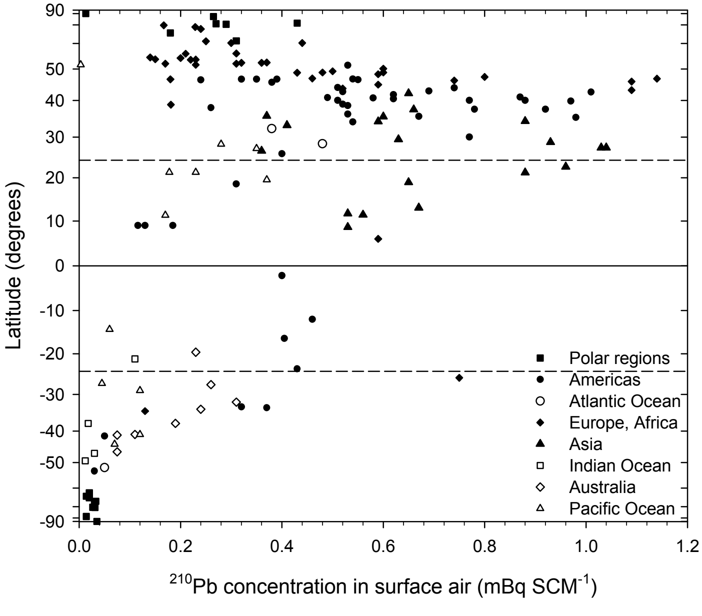
5.4. DISTRIBUTION OF ^{210}Pb AND ^{210}Po IN THE ATMOSPHERE

5.4.1. Concentrations at the land surface

In general, the most important influences on concentrations observed at the surface are the presence of significant land masses within the prevailing wind direction (as a source of ^{222}Rn), the amount of resuspension of dust, the degree of wet deposition occurring in the region and the degree of vertical mixing of the air mass (see Section 4.2). The presence of other sources, such as volcanic activity or forest fires, can also be important, especially for ^{210}Po.

Figure 5.4 shows average annual surface air concentrations of ^{210}Pb for a number of locations against latitude [5.12]. As a first approximation, the latitudinal variations in concentrations reflect the variations in the ^{222}Rn source term, which in turn, reflect the distribution of ice-free land masses. However, for any specific location, factors such as predominant wind direction, degree of vertical mixing and removal by precipitation also play important roles.

The highest ^{210}Pb concentrations in surface air are observed in the subtropical and temperate latitudes of the northern hemisphere owing to the relatively large land masses there. Values for average annual concentrations in this region mainly lie in the range of 0.2–1 mBq/m^3 [5.12]. Between the tropics, values are generally in the range of 0.1–0.5 mBq/m^3, while south of around 30°S, they quickly trend down to less than 0.1 mBq/m^3, owing to the lack of large ice-free land masses and consistent with the radon exhalation flux density trends already discussed.



Source: Figure 2.13 of Ref. [5.10].

Note: Each point represents a separate measurement location.

FIG. 5.4. Mean annual ^{210}Pb concentration in surface air as a function of latitude (reproduced with permission courtesy of Elsevier).

The concentrations in the high latitude regions of the northern hemisphere are about an order of magnitude higher than over Antarctica, primarily due to the proximity of significant land masses to the south, and low level transport from mid-latitudes to the Arctic in winter and spring, coincident with a pollution event known as Arctic haze [5.12, 5.52].

The concentrations are generally lower for the island or coastal sites (open symbols in Fig. 5.4) than for continental sites. Since Fig. 5.4 shows data for land based monitoring stations, the latitudinal average concentrations can be expected to be lower than indicated, with the island sites being more representative of the concentration over ocean areas.

There is clear evidence of a seasonality effect in ^{210}Pb and ^{210}Po concentrations in surface air [5.20, 5.53–5.55]. Concentrations are normally higher during dry seasons due to higher exhalation of ^{222}Rn from the soil, greater resuspension of soil dust and lower wet deposition rates, which can vary locally. However, the effect of air mass origin can also play a significant role in seasonality effects, depending on the local conditions [5.56].

5.4.2. Vertical profiles

Mean vertical profiles of ^{222}Rn generally show a strong reduction in concentration from the surface to the upper troposphere of a factor of 100 over continental areas [5.57–5.59]. This reflects the ground level source, and the fact that radon is only removed by radioactive decay with a half-life of 3.8 days. Nevertheless, vertical profiles are highly variable, reflecting geographical variability in the source term, as well as vertical and horizontal mixing processes in the PBL and the free atmosphere. Average profiles have been modelled using a one dimensional turbulent diffusion equation [5.60]. However, mixing processes are much more dynamic than is implied by such models, including rapid transport from the lower to the upper troposphere in convective clouds [5.61]. Discontinuities have also been observed at the PBL–free atmosphere interface, and between cloud layers and the above cloud layer [5.59].

The vertical distributions of ^{210}Pb and ^{210}Po in the troposphere are more complex than that of ^{222}Rn due to progressive ingrowth and removal by entrainment in precipitation. For ^{210}Pb, the reduction between ground level and upper tropospheric concentrations are generally less than a factor of five, and the concentration in the upper troposphere can be higher than in the cloud layer [5.57, 5.62]. The reason for this is scavenging of ^{210}Pb by wet and dry deposition in the lower troposphere, and by convective and stratiform precipitation from the mid-layers [5.53]. There is also a stratospheric contribution resulting from stratosphere–troposphere exchange, which predominantly affects upper tropospheric concentrations.

In oceanic and polar regions, the vertical distributions of ^{210}Pb and ^{210}Po can differ from those discussed above owing to the much reduced radon exhalation flux densities at the surface. Their concentrations can increase with altitude owing to a combination of the transport of radon to the free troposphere due to convective events, with subsequent formation of ^{210}Pb [5.32, 5.63] and contributions from stratospheric air [5.52]. Elevated levels of ^{210}Pb in the free troposphere over the South Pacific are attributed to radon convection due to fires, most likely in Africa [5.32]. In a study of the western hemisphere Arctic, ^{210}Pb concentrations were elevated at 3–6 km compared to those below 1 km at latitudes greater than 65°N [5.64].

Average ^{210}Pb and ^{210}Po concentrations in the stratosphere are about 0.3 mBq/SCM (standard cubic metre) at 0°C and 1000 hPa, with relatively little variation with altitude and latitude [5.22, 5.65]. As discussed in Section 5.2, some transfer of ^{210}Pb and ^{210}Po to the stratosphere can occur owing to volcanic eruptions [5.24]. Apart from this, direct transfers of ^{210}Pb and ^{210}Po to the stratosphere are negligible due to their removal by ice crystals formed in tropospheric air entering the lower stratosphere [5.66]. Rather, ^{222}Rn present in these air masses decays to ^{210}Pb, with substantial subsequent ingrowth of ^{210}Po due to the relatively long stratospheric residence times (~1 year). This transfer occurs predominantly under conditions of strong convective activity, bringing air from the lower troposphere with higher concentrations of radon and resulting in sufficient radon being transferred to support the observed stratospheric ^{210}Pb activity concentrations [5.64]. Strong convective activity, as well as tropopause folds, linked to the subtropical and polar jet streams, also result in the transfer of stratospheric air to the troposphere [5.67].

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Chapter 6

^{210}Pb AND ^{210}Po IN TERRESTRIAL SYSTEMS

6.1. DESCRIPTION OF THE TERRESTRIAL ENVIRONMENT

For the purposes of this chapter, the terrestrial environment is arbitrarily defined as encompassing the following range of related components: the atmosphere (particularly the troposphere); the lithosphere (particularly soils); and the surface and groundwater with which they are associated. Within these compartments, the terrestrial environment is also populated by a diverse range of plants and animals. The exchanges within and between these compartments are the environmental processes on which terrestrial ecosystems depend. However, the interactions among the various abiotic and biotic environments vary from location to location. The International Union of Radioecology (IUR) suggests that interaction matrices could present information on relevant processes in the terrestrial environments in a synthesized and structured way [6.1].

Each environment has its own unique chemical, physical and biological characteristics. Generic interaction matrices developed by the IUR [6.1] provide a convenient checklist for the initial evaluation of relevant environmental processes. The IUR indicates that the major components of the terrestrial environment closely relate to those discussed in the BIOMASS project [6.2]. In the IUR matrices, however, plants and animals are treated as distinct components rather than aggregated biota. Similarly, soils are subdivided into several distinct components. The main components in any terrestrial system are thus the following [6.1]:

(a) The atmosphere, particularly the troposphere, which comprises the region above the soil surface, including the regions both above and below the plant canopy.
(b) Surface aquatic bodies (i.e. rivers, streams and lakes) and groundwater, including aquifers.
(c) Vegetation, including macroscopic vegetation.
(d) Animals (macrofauna) in the terrestrial environment, inhabiting both the soil surface and areas within the soil (i.e. burrowing animals).
(e) Soil microbiota, comprising microflora and protozoa but excluding larger organisms such as earthworms and burrowing mammals.

(f) The soil, comprising minerals and organic material, aqueous solutions and the content of air in a soil sample. The characteristics of these various parts are influenced by the biotic and abiotic conditions in operation.

(g) The interface with the geosphere. This is an artificial frontier. In terrestrial environments, it will normally be represented by weathered, saturated or unsaturated materials.

6.2. MAJOR TRANSFER PROCESSES AND PATHWAYS

The decay of ^{238}U is the ultimate source of ^{210}Pb and ^{210}Po (see also Chapter 4). As a primordial radioisotope, ^{238}U is present in every component of the environment to varying degrees. The decay of ^{238}U leads to the formation of a series of different elements, including ^{210}Pb and ^{210}Po, each with its own characteristic environmental behaviour. As environmental processes are always dynamic, there may be disequilibrium between members of the decay series, depending on how open the system is to external influences (e.g. see Ref. [6.3]). The major environmental processes which cause disequilibria and the transfer of ^{210}Pb and ^{210}Po, or their relatively long lived progenitors (including ^{226}Ra and ^{230}Th), from primary sources in the crust to other compartments include the following:

- Weathering of exposed rocks;
- Leaching or dissolution of rocks by groundwater;
- Volcanic eruptions;
- Fires and burning fossil fuels;
- Exhalation of ^{222}Rn.

Given that ^{210}Pb and ^{210}Po are intrinsically linked by their radiogenic association, a combined consideration of both of these radionuclides is necessary to explain the observed patterns of ^{210}Po transfer in the environment (see Fig. 6.1).

The two main categories of biotic transfer are flora and fauna. Terrestrial plants can be exposed to ^{210}Pb and ^{210}Po via their surface (bark and cuticle of foliage), interaction with the atmosphere or via their roots, which can take up the radionuclides from the soil solution. Atmosphere–plant interactions are explored in Section 6.3. The mobility of radioisotopes in soil and their uptake by plant roots is discussed in Sections 6.4 and 6.5, and ^{210}Po transfer in animals in Section 6.6.

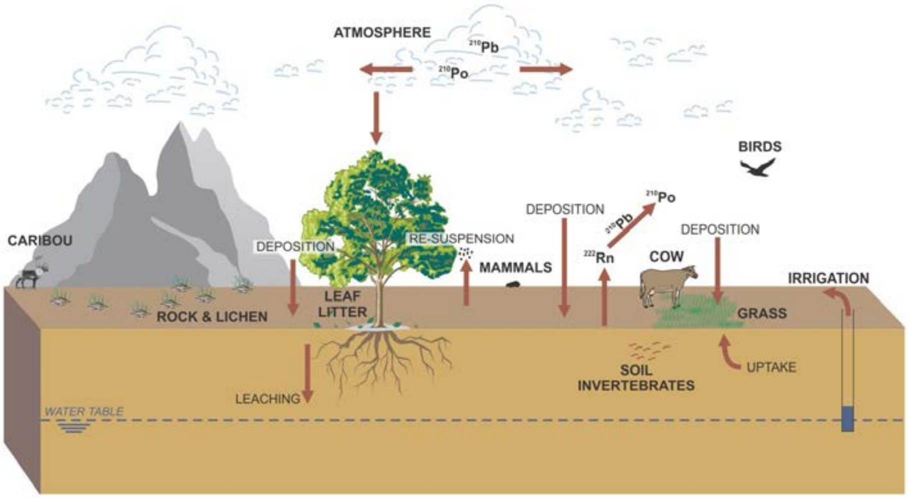

FIG. 6.1. Pathways controlling the movement and disposition of ^{210}Po in terrestrial environments.

6.3. ATMOSPHERE–PLANT INTERACTIONS

Polonium and lead activity concentrations on the surfaces of plants can be affected by many processes, including:

(a) Interception of gases, aerosols or dusts directly from the atmosphere, or by processes such as rain splash from the soil;
(b) Weathering, which results in the removal of radionuclides from plant surfaces by various physical processes and comprises the loss of radionuclides from the plant surface due to various processes, including wash-off due to rain or irrigation and surface abrasion;
(c) Biological processes (e.g. tissue ageing and leaf fall);
(d) Translocation of radionuclides from the plant surface to other, internal tissues of the plant.

References [6.4, 6.5] provide detailed discussions about the concepts and processes involved in air to plant interactions and how these processes can be quantified. This section provides a broad overview of these ideas and identifies information specific to polonium.

Plants can come into contact with ^{210}Pb and ^{210}Po by deposition of aerosols from ^{222}Rn decay or from resuspended dust. The contribution of atmospheric deposition to the total amount of radionuclides in and on plants is significant [6.6, 6.7] and can account for up to 95% [6.8]. The two basic mechanisms governing the rate of radionuclide deposition from the atmosphere to plants are wet and dry deposition processes. Wet deposition is the transport of radionuclides from the atmosphere with precipitation (rain, snow or hail); and dry deposition is a direct flux of radionuclides from the atmosphere in the form of gases or particulates. Pietrzak-Flis and Skowrońska-Smolak [6.8] find that the incorporation of the radionuclides occurs mainly as a result of wet deposition.

In addition to deposition, losses of radioactivity through weathering influence the overall amount of radioactivity retained on the plant surface. Pietrzak-Flis and Skowrońska-Smolak [6.8] measure these processes and determine experimentally that the deposition of ^{210}Po onto a variety of grasses and crops is in the range of 2–23 Bq/m^2 (6–62 Bq/m^2 in the case of ^{210}Pb), with retention proportional to their surface area and the presence of surface features (in this example possibly due to pockets on barley chaff, but may include factors such as glabrous or hirsute leaves).

With regard to translocation of the deposited radioactivity, there is unfortunately limited information available specifically on polonium, thus restricting the capacity to quantitatively model the relevant foliar uptake processes. If polonium specific data are absent, appropriate values might be identified from suitable analogues [6.9]. Although similar chemical properties of different elements do not necessarily mean similar behaviour inside the plant, this approach is still often used in practice, in the absence of other data. Analogues used for polonium include sulphur, selenium and tellurium (see Section 3.2), all of which are in Group 16 in the periodic table (chalcogens).

6.3.1. Interception

The extent to which plants can retain radionuclides depends on many physical, chemical and biological factors. The simplest parameter used for quantification is the interception fraction f, which is defined as the ratio of the activity initially retained by the standing vegetation (immediately after deposition) to the total activity deposited [6.10]. Since the interception fraction depends on the stage of development of the plant, the interception fraction is normalized to the standing biomass B (dry mass) to yield the mass interception fraction f_B (m^2/kg) [6.10]. As the leaf is the main interface between the atmosphere and vegetation, it is also possible to normalize the interception fraction to the leaf area index f_{LAI} [6.10].

There are no specific data on interception fractions for ^{210}Po. Using ^{33}S as a chemically similar analogue, an f_B of 0.35 was derived across five species based on wet deposition [6.11]. This value was slightly higher than that derived for ^{131}I (0.27) and was around three times lower compared with the other radionuclides used in the experiment (^{109}Cd, ^{144}Ce, ^{51}Cr and ^{85}Sr).

Particle size is another key parameter in determining the extent of dry deposition along with the vegetation type (often characterized by surface roughness). Interception is more effective for small particles and reactive gases [6.10]. No specific information on interception of polonium by dry deposition has been identified in the literature. However, data on interception for different particle sizes, across a range of radionuclides and vegetation types, can be found in Ref. [6.10].

6.3.2. Physical and biological weathering

Weathering is the loss of contamination from plants. The magnitude of the weathering loss of a radionuclide depends on many factors, including its solubility, strength of adsorption to the plant surface, degree of penetration into the inner flesh and leachability from the interior [6.12]. Biological factors (e.g. regular or seasonal exfoliation) also play a part in the weathering process. Thus, a complex interaction of those factors leads to the observed difference in weathering loss among radionuclides, plant species and their growth stages [6.12]. Despite this complexity, in models of environmental radionuclide behaviour, weathering is normally described by a single exponential function characterized by a first order rate constant λ_{w} or a weathering half-life T_{w} [6.12].

$$\lambda = \frac{\ln 2}{T_{\mathrm{W}}} \tag{6.1}$$

Data from numerous studies have shown limited differences in weathering between cationic species (such as caesium, cobalt, manganese, ruthenium and strontium) for most plant species, and that T_{w} is dependent on plant characteristics as well as on plant growth stage at the time of deposition [6.12]. In general, therefore, for different elements and plant groups, an assumption can be made that the weathering half-life for polonium will be in the range of 7.9–49 days (see table 1 of Ref. [6.12]).

6.3.3. Foliar translocation

Translocation is the process by which elements, water and nutrients are moved within vascular plants in the phloem and xylem, and can refer to contaminants either taken up by plants roots or absorbed from the surface of plants and transferred to other parts that have not been contaminated directly. The rate of this process reflects the mobility of an element within the plant and can vary greatly. Translocation from roots is discussed in Section 6.5. To quantify this process for material deposited directly onto the foliage, the translocation factor f_{tr} normally used represents the proportion of foliage activity per 1 m^2 of crop at the time of deposition (Bq/m^2) associated with the activity in the edible part per 1 m^2 of crop at harvest time (Bq/m^2) expressed as a percentage [6.13].

There is a lack of information on polonium translocation values; hence, an analogue could be considered here. However, there is no appropriate analogue for translocation based on foliar deposition (f_{tr}). The only analogue data available are the proportional translocation of tellurium in root vegetables, reported as 0.8 [6.14, 6.15]. However, Ham et al. [6.7] and Pietrzak-Flis and Skowrońska-Smolak [6.8] report that the movement of polonium from roots is negligible. This highlights the uncertainty associated with the analogue approach, as the experimental data for polonium have not indicated a translocation factor as high as that reported for tellurium.

6.4. MOBILITY OF POLONIUM IN SOILS

Surface soils are the interface between the atmosphere, the parent rocks and the subsurface hydrological system. There are radionuclide fluxes between these environmental compartments which govern ^{210}Pb and ^{210}Po activity concentrations and how they change in the surface soil. Generally, ^{210}Po is at, or very close to, secular equilibrium with ^{210}Pb in almost all soils [6.16]. However, the distribution of ^{210}Po in soil is rather inhomogeneous (see Chapter 4) [6.17]. The concentrations of radionuclides in surface soils vary depending on a number of processes and are discussed in Sections 6.4.1–6.4.4.

6.4.1. Resuspension

In general, three types of process are used to describe the lateral spread of contaminants which have been deposited onto surface soil [6.18, 6.19]: surface creep, saltation and resuspension. Resuspension occurs when wind exerts a force exceeding the degree of adherence of particles to the surface material. The forces in action are the weight of the particle and the adherence, as well as the aerodynamic loads relating to the flow of wind. There are two basic approaches used to model resuspension. The resuspension factor is the ratio of volumetric air concentration to soil contamination. The resuspension rate approach is based on the ratio of particle flow density to soil contamination, which depends on the resuspended material (particle size, shape and adherence), the surface type (roughness and humidity), the time lapsed after deposition, and mechanical actions (soil processing), if any.

No specific information is available for polonium. However, because the variability of measured resuspension factors and rates are very high and the accuracies of these models' predictions are rather low, there is no substantial evidence that the elemental characteristics of radionuclides influence the processes governing resuspension. This means that the data and models validated for other radionuclides can also be used for polonium. Overall, the resuspension factors measured directly after the acute depositions are around 10^{-5}/m in residential areas, on a site undergoing cleanup operations and on an arid site, and from 10^{-8}/m to 10^{-6}/m in humid areas. The resuspension factor decreases by up to 10^{-9}/m over 3–4 years after the deposition. Resuspended dust can be a very localized phenomenon or can reach global scales, such as with the resuspension of dusts from nuclear test sites in China to Japan and even further [6.20].

6.4.2. Radon exhalation

One of the major mechanisms which determines changes in ^{210}Pb concentrations (and hence ^{210}Po) in topsoil is exhalation of a fraction of the noble gas ^{222}Rn from surface soils to the atmosphere, with subsequent decay to ^{210}Pb in the atmosphere and removal to the Earth's surface by wet and dry deposition (see Section 5.3). Depending on the local soil and meteorological conditions, these processes can result in either an excess or deficiency of ^{210}Pb relative to ^{226}Ra in the topsoil (primarily in the top 1 m of soil).

Since exhalation flux densities of ^{222}Rn from soils are about a hundred times greater than that from oceans, deposition of soil-origin ^{210}Pb to the oceans can be expected to cause a deficiency of ^{210}Pb and ^{210}Po relative to ^{226}Ra in soils. Globally, approximately 90% of ^{210}Pb deposition is by wet deposition, and local deposition greatly depends on rainfall [6.21, 6.22]. A confounding factor is ^{210}Pb in leaf litter, which can lead to excess ^{210}Pb and ^{210}Po in the surface layer [6.23, 6.24]. Owing to the permanent flux from the atmosphere and other processes, activity concentrations in the topsoil (0–10 cm) tend to be higher compared with the deeper soil, where ^{210}Po is in approximate equilibrium with the ^{210}Pb and ^{222}Rn values, within a few tens of Bq/kg [6.17].

6.4.3. Partitioning of the soil Po:Pb ratio into solution

A portion of the radioactivity present in the soil will be adsorbed to soil particles, while some will dissolve into soil water and hence become available for root uptake by plants. Significant progress has been made to describe radionuclide sorption in heterogeneous solids. Nevertheless, models describing sorption are still mostly based on empirical, solid–liquid distribution coefficient (K_d) values, which are estimated from sorption studies using reasonably well characterized homogeneous surfaces (see Section 2.1).

A summary of available K_d values for lead and polonium for soils, grouped on the basis of texture and organic matter content criteria, as well as data on the effect of pH (excluding organic soils), are given in Table 6.1. Soils are grouped according to the sand and clay mineral percentages, and the organic matter content of the soil. Some variation is due to pH (8%) and clay content (24%) but not cation exchange capacity [6.25]. These numbers are estimates, since adsorption is controlled by a larger range of conditions, including the concentration of competing ions, the mineralogical and organic composition of the soils, and the grain size of the constituents (and so the available surface area for adsorption per gram).

TABLE 6.1. K_d VALUES FOR SOILS GROUPED ACCORDING TO THE TEXTURE/ORGANIC MATTER CRITERION (L/kg)

Element	Soil group	*N*	GM	GSD	AM	SD	Min.	Max.
Pb	All soils	23	2.0×10^3	1.0×10^1	1.5×10^4	3.3×10^4	2.5×10^1	1.3×10^5
	Sand	9	2.2×10^2	4	4.0×10^2	4.3×10^2	2.5×10^1	1.3×10^3
	Loam	5	1.0×10^4	3	1.5×10^4	1.6×10^4	3.6×10^3	4.3×10^4
	Clay	2	n.a.	n.a.	6.6×10^4	n.a.	5.4×10^3	1.3×10^5
	Organic	5	2.5×10^3	3	3.7×10^3	3.8×10^3	8.8×10^2	1.0×10^4
	Unspecified	2	n.a.	n.a.	5.9×10^4	n.a.	1.6×10^4	1.0×10^5
Po	All soils	44	2.1×10^2	5	5.6×10^2	1.1×10^3	1.2×10^1	7.0×10^3
	Sand	14	1.0×10^2	6	7.4×10^2	1.9×10^3	1.7×10^1	7.0×10^3
	Loam	27	2.3×10^2	4	4.6×10^2	4.6×10^2	1.2×10^1	1.8×10^3
	Clay	2	n.a.	n.a.	1.7×10^3	n.a.	7.2×10^2	2.7×10^3
	Organic	1	n.a.	n.a.	6.6×10^3	n.a.	n.a.	n.a.

Source: See Ref. [6.5].

Note: The sand group is ≥65% sand and <18% clay; the clay group is ≥35% clay; the organic group is ≥20% organic matter; and the rest is assigned to the loam group and unspecified. AM — arithmetic mean; GM — geometric mean; GSD — geometric standard deviation; n.a. — not applicable; SD — standard deviation.

Table 6.1 shows that K_d values for polonium tend to be lower than those for lead. This difference can lead to some disequilibrium between these radionuclides in the fraction assumed to be available for root transfer.

6.5. ^{210}Pb AND ^{210}Po TRANSFER FROM SOIL TO PLANTS

6.5.1. Processes governing ^{210}Pb and ^{210}Po transfer to plants

Polonium can enter plants via several routes. Most large scale contamination is generally through air deposition or from contaminated soil. Contamination can also occur via contaminated surface water (inundation and irrigation) or contaminated groundwater (upwelling and irrigation) [6.8, 6.26]. The relative importance of these pathways depends on the concentration of the radionuclides in the soil, the soil–plant concentration ratio (CR) and the rate of deposition onto plant parts above ground. An important implication is that radionuclide activity concentrations in crops such as in roots, tubers, cereals and legumes, where the edible portion is protected by inedible plant parts, should not be significantly affected by direct deposition; whereas the converse is true for leafy vegetables. Plant type and the ecosystem influence the values thus derived. Transfer factors (TFs) range across orders of magnitude, depending on the plant and soil type variations assessed [6.5].

6.5.2. ^{210}Pb and ^{210}Po transfer to mosses, plants and lichens

The factors that determine variability in radionuclide transfer to plants generally including the following [6.27]:

(a) The form in which the activity enters or is present in the soil (e.g. as particles, as aerosol or in solution);
(b) The physicochemical properties of the radionuclide;
(c) The time after entry into the soil;
(d) The type of soil and its physicochemical characteristics;
(e) The type of crop;
(f) Crop management practices (the application of fertilizers, irrigation, ploughing and liming);
(g) Climatic conditions;
(h) The experimental conditions under which the TFs were obtained.

The significance of these factors varies depending on the radionuclide of interest and the contamination scenario. Table 6.2 presents data from the 1970s on the influence of different steppe ecosystems on ^{210}Po transfer to plants.

From Table 6.2, the activity concentrations in the above ground biomass are in the range of 3.3–16.3 Bq/kg (DW) for ^{210}Po and 9.3–49.6 Bq/kg (DW) for ^{210}Pb. For both ^{210}Pb and ^{210}Po, there are higher concentrations in the underground biomass (roots). Maximum activity concentrations of ^{210}Pb and ^{210}Po occur in perennial sawn grasses, probably because of the application of mineral fertilizers containing a mixture of naturally occurring radionuclides [6.29]. The ^{210}Po:^{210}Pb ratios in plants are lower than those for topsoils, indicating higher transfer rates for ^{210}Pb compared with ^{210}Po [6.29].

Taskayev and Testov [6.29] present data on ^{210}Po transfer to 24 species of plants and mosses representative of northern taiga and collected from areas with naturally elevated radiation backgrounds (see Table 6.3).

One type of vegetation of particular interest in the assessment of ^{210}Po is lichen. Lichens are slow growing symbiotic associations (fungi plus algae or cyanobacteria) that grow on trees, rocks and soils. They have no true roots and, despite accessing some mineral nutrition from rocks or organic nutrients from bark, are highly dependent on the atmosphere for nutrition. They have a long lifespan; thus, activity concentrations of ^{210}Po are supported by ^{210}Pb ingrowth in addition to direct deposition. Lichens are important for human populations in northern climates because reindeer or caribou (which are key components of the human diet) ingest large quantities in winter. The importance of lichens has not been considered to the same extent for food chains involving herbivorous species in other climatic zones.

In a 1978 study, Ermolayeva-Makovskaya and Litver [6.30] report a high mean value of 260 Bq/kg (DW) for both ^{210}Pb and ^{210}Po in lichens of the polar and subpolar zones of the Russian Federation, from the Cola Peninsula to Chukot. Slightly lower values (70–212 Bq/kg, DW) of ^{210}Po activity concentrations in lichens in Nordic terrestrial ecosystems are presented in Refs [6.24, 6.31]. Data on lichens in northern Canada show mean activity concentrations of 275 Bq/kg (DW) for *Cladina mitis* and 622 Bq/kg (DW) for *Cetraria nivalis* [6.32]. The data indicate that the highest activity concentrations of ^{210}Pb and ^{210}Po in above ground biomass occur in mosses and lichens, followed by club-mosses and ferns, and then grasses.

6.5.3. ^{210}Pb and ^{210}Po transfer to berries, mushrooms and understory species

Several comprehensive studies in Finland and France have greatly extended the information on ^{210}Pb and ^{210}Po transfers to forest products, mainly mushrooms

TABLE 6.2. ACTIVITY CONCENTRATIONS OF ^{210}Po AND ^{210}Pb IN MEADOW PLANTS OF DIFFERENT STEPPE ECOSYSTEMS

Ecosystem	^{210}Po (Bq/kg, DW)			^{210}Pb (Bq/kg, DW)			^{210}Po:^{210}Pb ratio	
	Above ground biomass	Roots	Ratio	Above ground biomass	Roots	Ratio	Above ground biomass	Roots
Natural steppe	7.0 ± 4.4	42 ± 14	0.17 ± 0.1	17.5 ± 8.1	44 ± 15	0.41 ± 0.1	0.39 ± 0.1	0.96 ± 0.1
Saline meadow	3.7 ± 0.7	25 ± 24	0.16 ± 0.1	9.3 ± 2.6	31 ± 18	0.42 ± 0.3	0.4 ± 0.05	1.25 ± 0.2
Bog meadow	3.3 ± 2.0	23 ± 7	0.14 ± 0.1	11.8 ± 2.0	18 ± 4	0.62 ± 0.3	0.28 ± 0.05	1.25 ± 0.2
Sawn grasses	16.3 ± 6.7	—[a]	—[a]	49.6 ± 23.7	—[a]	—[a]	0.34 ± 0.08	—[a]
Trees/bushes	13.7 ± 4.4	—[a]	—[a]	32.6 ± 13.3	—[a]	—[a]	—[a]	—[a]
Mean	8.1 ± 5.9	30 ± 19	0.19 ± 0.1	21.5 ± 17	32 ± 17	0.47 ± 0.3	0.38 ± 0.12	0.97 ± 0.3

Source: See Ref. [6.28].

[a] —: data not available.

TABLE 6.3. CONCENTRATIONS OF ^{210}Po IN PLANTS OF NORTHERN TAIGA

Group	Species	^{210}Po (Bq/kg, DW)
Mosses	*Conocephalum conicum*	1480
	Polytrichum commune	353
	Mnium cinclidioides	293
	Pleurozium schreberi	292
	Sphagnum	274
Club-mosses	*Lycopodium annotinum* L.	100
	Gymnocarpium dryopteris	80
Ferns	*Dryopteris filix-mas* L.	70
Grasses	*Solidago virgaurea* L.	60
	Trientalis L.	55
	Caltha palustris	47
	Polygonum L.	46
	Geum rivale L.	39
	Filipendula ulmaria	37
	Veratrum lobelianum	34
	Anemone	34
	Chamerion angustifolium	33
	Equisetum arvense	27
	Juncus L.	20
	Sanguisorba officinalis	13

Source: See Ref. [6.29].

Note: Range is 13–1480 Bq/kg (DW); overall mean is 145 Bq/kg (DW); standard deviation is 300 Bq/kg (DW); geometric mean is 61 Bq/kg (DW); median is 42 Bq/kg (DW); geometric standard deviation is 1.1.

and berries, at different forest sites (Scots pine, Norway spruce, mixed forest and Downy birch).

Vaaramaa et al. [6.23] present data on ^{210}Pb and ^{210}Po in wild berries (blueberry and lingonberry) and mushrooms in boreal forest ecosystems in Finland. The lingonberry is an evergreen dwarf shrub and the blueberry is a deciduous plant (see Table 6.4).

TABLE 6.4. ACTIVITY CONCENTRATIONS OF ^{210}Po AND ^{210}Pb IN WILD BERRIES IN FINLAND

Wild berry	Site	^{210}Pb (Bq/kg, DW)	^{210}Po (Bq/kg, DW)
Blueberry	N. Finland	0.7 ± 0.1	3.2 ± 0.6
	S. Finland	1.7 ± 0.3	2.5 ± 0.4
Lingonberry	N. Finland	1.4 ± 0.2	2.2 ± 0.3
	S. Finland	3.2 ± 0.3	7.5 ± 1.0

Source: See Ref. [6.23].

The activity concentrations of ^{210}Pb and ^{210}Po in wild berries do not show clear dependence on the type of forest and are quite similar to those reported by Solatie et al. [6.33] for Finnish Lapland and McDonald et al. [6.34] for samples taken in England and Wales.

The highest ^{210}Pb (^{210}Po) concentrations (dry weight) in blueberry shrubs were found to be the following [6.23]:

- Lignified brown stem of the blueberry shrub: 35 (80) Bq/kg;
- Roots: 33 (48) Bq/kg;
- Green stem 30 (29) Bq/kg;
- Leaves 16 (27) Bq/kg.

For lingonberry shrubs, the highest ^{210}Po concentrations (dry weight) were found in the roots and rhizome (100 Bq/kg), followed by the current year stem (90 Bq/kg), the lignified brown stem (30 Bq/kg), and the current year and old leaves (22–26 Bq/kg) [6.23]. In general, the activity concentrations of ^{210}Pb and ^{210}Po in leaves and small tree branches were higher than those in above ground biomass of grassy plants because of higher interception of the radionuclides from the atmosphere, due to a higher leaf area index and a longer period of vegetation during the year.

Although the ^{210}Po:^{210}Pb activity ratios in the organic horizon of the studied forests was close to 0.9, in most cases the concentrations of ^{210}Po in plant tissues exceeded those of ^{210}Pb (^{210}Po:^{210}Pb varied from 1.5 to 2.5), except in the lignified stem of lingonberry where the concentration of ^{210}Pb was 1.5 times higher than ^{210}Po [6.23]. This might indicate that ^{210}Po is characterized by both a higher transfer rate than ^{210}Pb from forest soil to plant compartments and by a higher mobility within the plant. However, more precise research is needed to confirm or reject these statements.

According to the literature, fungi have been shown to accumulate stable lead to much higher concentrations than berries [6.35]. However, because of their short lifespan, direct atmospheric deposition on the surfaces of the fruiting bodies of mushrooms cannot be the major uptake route for elements [6.36]. Thus, environmental properties of the site where fungi grow can be more important for radionuclide transfer [6.23, 6.35, 6.36] provide information on ^{210}Pb and ^{210}Po transfer to many species of mushrooms.

Vaaramaa et al. [6.23] find that the activity concentrations for ^{210}Pb and ^{210}Po in mushrooms in Finland are generally higher than wild berries, especially in the case of ^{210}Po. The dry weight range for ^{210}Pb is 0.97–16.2 Bq/kg, and is much wider for ^{210}Po, 13.9–1200 Bq/kg. They find the highest mean concentration of ^{210}Po in the fruiting bodies of the following families [6.23]:

- Boletaceae: 380 Bq/kg (DW);
- Russulaceae: 32.2 Bq/kg (DW);
- Cortinariaceae: 10 Bq/kg (DW).

They find the minimum mean values for ^{210}Po for mushrooms in the following families [6.23]:

- Hygrophorus: 2.5/3.1 Bq/kg (DW) in the cap/stipe;
- Albatrellus: 3.8 Bq/kg (DW) in the whole fruit body;
- Ratio of the activity concentration between the caps and stipes: 1.6–7.1.

As for ^{210}Pb, considering the same species in different forest types, Vaaramaa et al. [6.23] find the lowest activity concentrations in mushrooms of the Boletaceae family (3.0 Bq/kg, DW) and the highest in Russulaceae (11.1 Bq/kg, DW) and Cortinariaceae (11.1 Bq/kg, DW) [6.23]. Overall, these data are similar to the ^{210}Pb concentrations (1.76–36.5 Bq/kg, DW) measured in various mushrooms collected from the forests of France [6.37].

In the northern Finland site, Vaaramaa et al. [6.23] report:

> “No differences between forest types [Norway spruce, Scots pine, Downy birch and mixed forest] were found in the concentrations of ^{210}Pb in the whole fruiting bodies (cap and stipe combined), except in the activity concentration of ^{210}Pb in *Cortinarius armillatus* L. collected from mixed forest….

....

> “The activity concentrations of ^{210}Pb were higher in the caps of the mushrooms than in the stipes for *Lactarius rufus* L. and *Russula paludosa* L. The average cap-to-stipe ratio of ^{210}Pb in the Boletaceae family was 1.0, which is in agreement with the mean ratio for ^{210}Pb (1.1) in *Xerocomus badius* L. (Boletaceae family) analyzed by Malinowska et al. [Ref. [6.38]].”

These values were statistically significantly higher than those for ^{210}Pb, reflecting higher mobility of ^{210}Po among tissues of mushrooms. Vaaramaa et al. [6.23] find that:

> “The ^{210}Po/^{210}Pb ratio in the mushrooms varied widely from 0.87 to 322 in whole fruiting bodies, mainly being higher than one. The lowest value was recorded in *R. paludosa* L. and the highest in *L. vulpinum* L. In *S. luteus* L. the ratio of ^{210}Po/^{210}Pb has been reported to be 22 and 47 in Finnish Lapland (Solatie et al. [Ref. [6.33]]). In the present study, the ratio was 17 for the same species in Scots pine forest of the northern Finland site.... The results may indicate that ^{210}Po is more effectively taken up from soil to the fruiting bodies than ^{210}Pb.”

6.5.4. ^{210}Pb and ^{210}Po transfer to agricultural plants

Several comprehensive reviews of radionuclide transfer from soil to plants, including recommended TF values for both natural and artificial radionuclides can be found in Refs [6.4, 6.5] and in Table 6.5 for lead and polonium. The TF values are for 14 plant groups according to the classifications presented in Chapter 4 (see Ref. [6.5] for assignments of individual plants to these groups, and the plant compartments considered). The TF values depend on the plants and soil types assessed. Among the lowest values for both elements is maize grain (5.2×10^{-4} for lead and 1.8×10^{-5} for polonium); while the highest are 1.0 for polonium in pasture vegetation and 25 for lead in leafy vegetables.

However, when analysing the data in Table 6.5, it should be noted that the radioactivity in the plant (or plant compartments) used for assessing the TF (or CR) values is not only acquired through root transfer. The above ground biomass might also be contaminated because of resuspension or direct deposition of radionuclides from the atmosphere [6.8] (see also Section 6.3). These atmospheric fluxes are of secondary importance for many radionuclides (such as ^{137}Cs and ^{90}Sr), but that might not be the case for ^{210}Pb and ^{210}Po, for which the foliar pathway dominates in some contamination scenarios. In this context, plants with edible compartments located above ground do not experience the same contamination conditions as root crops and tubers, which can be contaminated

TABLE 6.5. SOIL TO PLANT TRANSFER FACTORS FOR LEAD AND POLONIUM

Element	Plant group	Plant compartment	Soil group	*N*	Mean/value	GSD/SD[a]	Min.	Max.
			Temperate environments					
Pb	Cereals	Grain	All	9	1.1×10^{-2}	3.6	1.9×10^{-3}	4.8×10^{-2}
		Stems and shoots	All	4	2.3×10^{-2}	3.5	5.1×10^{-3}	9.6×10^{-2}
	Maize	Grain	All	9	1.2×10^{-3}	2.3	5.2×10^{-4}	3.8×10^{-3}
		Stems and shoots	All	3	2.8×10^{-3}	6.6	6.0×10^{-4}	2.3×10^{-2}
	Leafy vegetables	Leaves	All	31	8.0×10^{-2}	1.3×10^{1}	3.2×10^{-3}	2.5×10^{1}
			Sand	4	7.3×10^{-2}	1.5	4.9×10^{-2}	1.1×10^{-1}
			Loam	3	8.2×10^{-1}	1.0	7.9×10^{-1}	8.6×10^{-1}
			Clay	7	2.8×10^{-2}	4.1	4.1×10^{-3}	1.2×10^{-1}
	Non-leafy vegetables	Fruits, heads, berries, buds	All	5	1.5×10^{-2}	2.6×10^{1}	1.5×10^{-3}	3.9
		Stems and shoots	All	2	8.8×10^{-3}	—[b]	5.8×10^{-3}	1.2×10^{-2}

TABLE 6.5. SOIL TO PLANT TRANSFER FACTORS FOR LEAD AND POLONIUM (cont.)

Element	Plant group	Plant compartment	Soil group	*N*	Mean/value	GSD/SD[a]	Min.	Max.
Pb	Leguminous vegetables	Seeds and pods	All	17	5.3×10^{-3}	1.2×10^{1}	4.6×10^{-4}	4.9
			Sand	3	2.7×10^{-3}	3.2	6.5×10^{-4}	8.9×10^{-3}
			Loam	5	1.4×10^{-3}	4.4	6.5×10^{-4}	8.9×10^{-3}
			Clay	4	8.0×10^{-4}	1.0	4.6×10^{-4}	1.0×10^{-2}
		Stems and shoots	All	1	8.0×10^{-4}	—[b]	—[b]	—[b]
	Root crops	Roots	All	27	1.5×10^{-2}	1.6×10^{1}	2.4×10^{-4}	3.3
			Sand	5	6.4×10^{-2}	1.6	4.2×10^{-2}	1.2×10^{-1}
			Loam	5	2.3×10^{-3}	4.7	2.4×10^{-4}	1.7×10^{-2}
		Stems and shoots	All	12	6.3×10^{-2}	150	3.0×10^{-3}	1.6×10^{1}
	Tubers	Tubers	All	30	1.5×10^{-3}	7.4	1.5×10^{-4}	2.6
			Sand	5	6.4×10^{-3}	3.5	1.6×10^{-3}	3.9×10^{-2}
			Loam	17	5.2×10^{-4}	2.4	1.5×10^{-4}	2.3×10^{-3}
	Grasses	Stems and shoots	All	17	3.1×10^{-1}	1.8	1.1×10^{-1}	1.0

TABLE 6.5. SOIL TO PLANT TRANSFER FACTORS FOR LEAD AND POLONIUM (cont.)

Element	Plant group	Plant compartment	Soil group	N	Mean/value	GSD/SD[a]	Min.	Max.
Pb	Pasture	Stems and shoots	All	34	9.2×10^{-2}	4.8	2.2×10^{-3}	1.0
	Leguminous fodder	Stems and shoots	All	1	1.6×10^{-2}	n.a.[c]	n.a.[c]	n.a.[c]
Po	Cereals	Grain	All	2	2.4×10^{-4}	—[b]	2.2×10^{-4}	2.6×10^{-4}
	Maize	Grain	All	2	2.4×10^{-4}	—[b]	1.8×10^{-5}	4.7×10^{-4}
	Leafy vegetables	Leaves	All	12	7.4×10^{-3}	6.9	2.5×10^{-4}	5.0×10^{-2}
	Non-leafy vegetables	Stems and shoots	All	2	1.9×10^{-4}	—[b]	1.6×10^{-5}	3.7×10^{-4}
	Leguminous vegetables	Seeds and pods	All	4	2.7×10^{-4}	3.9	6.0×10^{-5}	1.0×10^{-3}
	Root crops	Roots	All	10	5.8×10^{-3}	4.3	2.4×10^{-4}	4.9×10^{-2}
		Stems and shoots	All	2	7.7×10^{-2}	—[b]	5.8×10^{-2}	9.7×10^{-2}
	Tubers	Tubers	All	9	2.7×10^{-3}	5.8	1.4×10^{-4}	3.4×10^{-2}
	Pasture	Stems and shoots	All	10	1.2×10^{-1}	4.2	2.2×10^{-2}	1.0
	Leguminous fodder	Stems and shoots	All	2	1.1×10^{-2}	—[b]	2.6×10^{-5}	2.2×10^{-4}

TABLE 6.5. SOIL TO PLANT TRANSFER FACTORS FOR LEAD AND POLONIUM (cont.)

Element	Plant group	Plant compartment	Soil group	*N*	Mean/value	GSD/SD[a]	Min.	Max.
			Tropical environments					
Pb[d]	Cereals	Grain	Sand	1	2.5×10^{-3}	n.a.[c]	n.a.[c]	n.a.[c]
	Grasses	Leaves	Sand	9	2.1×10^{-1}	2.2	5.9×10^{-2}	1.00
	Herbs	Leaves	Sand	2	3.7×10^{-1}	—[b]	2.0×10^{-2}	7.1×10^{-1}
	Leguminous vegetables	Grain	All	9	3.3×10^{-3}	2.4	6.5×10^{-4}	8.9×10^{-3}
			Sand	3	3.4×10^{-3}	1.3	2.8×10^{-3}	4.4×10^{-3}
			Loam	6	3.2×10^{-3}	3.0	6.5×10^{-4}	8.9×10^{-3}
	Other crops		Sand	18	2.3×10^{-1}	2.7	1.4×10^{-2}	1.0
	Non-leafy vegetables		All	2	7.0×10^{-3}	—[b]	7.0×10^{-3}	7.0×10^{-3}
	Pasture	Stems and shoots	Unspecified	1	3.0×10^{-1}	n.a.[c]	n.a.[c]	n.a.[c]
	Root crops	Roots	Loam	3	2.4×10^{-3}	1.5	1.8×10^{-3}	4.0×10^{-3}

TABLE 6.5. SOIL TO PLANT TRANSFER FACTORS FOR LEAD AND POLONIUM (cont.)

Element	Plant group	Plant compartment	Soil group	*N*	Mean/value	GSD/SD[a]	Min.	Max.
Pb[d]	Tubers	Tubers	All	16	5.7×10^{-4}	2.4	1.5×10^{-4}	2.3×10^{-3}
			Sand	1	1.6×10^{-3}	n.a.[b]	n.a.[b]	n.a.[b]
			Loam	15	5.3×10^{-4}	2.4	1.5×10^{-4}	2.3×10^{-3}
	Maize	Grain	All	6	8.5×10^{-4}	2.1	5.2×10^{-4}	3.8×10^{-3}
			Sand	1	5.2×10^{-4}	n.a.[b]	n.a.[b]	n.a.[b]
			Loam	5	9.3×10^{-4}	2.2	5.9×10^{-4}	3.8×10^{-3}

Source: See Ref. [6.5].

[a] Geometric standard deviation/standard deviation.

[b] —: data not available.

[c] n.a.: not applicable.

[d] Data for polonium not available.

through root uptake. Therefore, observed concentrations for leafy vegetables can be up to 3–5 times higher due to deposition effects. It is also known that slowly maturing plants have much lower transfer coefficients for ^{210}Po than rapidly maturing plants under greenhouse conditions [6.39].

On the other hand, the concept of soil to plant TFs is adopted as a reasonable empirical measure of plant contamination under steady state conditions, when radionuclide fluxes corresponding to different pathways are already well balanced. A general point is that the data in Table 6.5 also reflect existing differences in mobility between radionuclides in soil and tend to be in compliance with the data on K_d values given in Table 6.1. In particular, TF values for clay soils (where available) are normally about three times lower than those for light sandy soils, and this ratio is close to the ratio for the K_d values for the same radionuclides and soil types.

6.6. ^{210}Pb AND ^{210}Po TRANSFER TO ANIMALS

In this section, transfer to animals of both lead and polonium are considered because ^{210}Pb transferred to animal tissues decays to ^{210}Po. Therefore, its transfer characteristics and rates to various animals are also relevant for polonium.

6.6.1. Processes governing distribution of ^{210}Pb and ^{210}Po in animals

Lead-210 is largely retained in bone, while ^{210}Po is distributed mainly in soft tissue (see Table 6.6). Target tissues for ^{210}Po are the spleen, liver and kidneys. However, the observed ^{210}Po:^{210}Pb ratios in tissues are time dependent and can be substantially modified by continuing radioactive decay of ^{210}Pb resulting in support for ^{210}Po, as well as by the difference in biological half-lives of these radionuclides in the different tissues. The rates of these processes greatly depend on the animals and tissues of interest, and can vary across a wide range.

Following ^{210}Po administration to rats under laboratory conditions, the highest concentrations are observed in the spleen, kidney, liver and other soft tissue [6.40]. In wild animals (e.g. roe deer), the highest concentrations are observed in kidney, liver and bone [6.30].

Some of the most detailed evidence of animal dependent distribution of ^{210}Po to ^{210}Pb is given by Maslov and Testov [6.41] in the 1970s, who report data on ^{210}Pb and ^{210}Po activity in small mouse-like rodents (bank vole, six months old) and reindeer (six years old) (see Table 6.7).

Differences in the accumulation of ^{210}Po, and the ratio with ^{210}Pb, in small rodents and reindeer can be explained by the origin of the contamination. In the bank vole, the ^{210}Po accumulated in some target organs (e.g. spleen, kidney and

TABLE 6.6. CONCENTRATIONS OF ^{210}Pb AND ^{210}Po:^{210}Pb RATIOS IN BONE AND MUSCLE OF ANIMALS IN FINLAND

Animal	Bone		Muscle	
	^{210}Pb (Bq/kg, DW)	^{210}Po:^{210}Pb	^{210}Pb (Bq/kg, DW)	^{210}Po:^{210}Pb
Deer	1338	0.56	0.17	24
Moose	18.5	0.54	0.06	12.5
Wolf	9.6	0.69	0.02	100
Wolverine	22.9	0.67	0.08	90

TABLE 6.7. ACTIVITY OF ^{210}Po IN ANIMALS WITH DIFFERENT LIFESPANS

Tissue	Bank vole		Reindeer	
	^{210}Po (mBq)	^{210}Po ratio to bone	^{210}Po (mBq)	^{210}Po ratio to bone
Bone	1–7.8	1	765.9	1
Muscle	27	1.5	23.7	0.02
Liver	23	1.3	261.2	0.3
Spleen	177.6	10	45.9	0.05

Source: See Ref. [6.41].

other soft tissue) originates from the diet and not, to any great extent, from ^{210}Pb decay in the body. Therefore, the ^{210}Po:^{210}Pb ratios in different tissues of these small animals (with relatively short lifespans) typically exceed unity. In contrast, the ratios in reindeer are normally less than one (0.02–0.3), since bone contains a substantial amount of older ^{210}Pb accumulated in the animals as a consequence of their longer lifespan. Data for other species are given in Table 6.8.

The highest accumulation of ^{210}Po was observed in small rodents and earthworms, followed by reptiles and amphibians. For ^{210}Pb, the highest activity concentrations (around 50 Bq/kg, DW) were found in earthworms. The mobility of ^{210}Po varies in different species: ^{210}Po:^{210}Pb ratios were highest for rodents,

TABLE 6.8. WHOLE BODY (EXCLUDING GASTROINTESTINAL TRACT) ACTIVITY CONCENTRATIONS AND ESTIMATED ^{210}Po:^{210}Pb RATIOS IN NORDIC WILD SPECIES

Species	*N*	^{210}Po (Bq/kg, DW)		^{210}Pb (Bq/kg, DW)		^{210}Po:^{210}Pb[a]
		Mean	SD	Mean	SD	
Common shrew	9	37.2	19	0.3	0.4	258
Bank vole	8	65.1	17.3	1.5	1.1	77
Willow grouse	5	3.3	1.2	2.1	1.6	2.2
Red earthworm	2	48.6	28.7	50	8	1.0
Grey worm	5	57.2	42.3	47	23	1.5
Viper	1	20.9 21.84	—[b] —[b]	13.05 12.09	—[b] —[b]	—[b]
Frog	1	5.65 5.88	—[b] —[b]	3.39 2.98	—[b] —[b]	—[b]

Source: Adapted from Ref. [6.24].

Note: SD — standard deviation. The intestinal tracts of the worms were not removed.

[a] The ^{210}Po:^{210}Pb ratios are averages based on individual sample values rather than species means.

[b] —: data not available.

while for earthworms they were close to unity. Ratios greater than 100 have also been reported in the muscle of wild boar [6.42].

6.6.2. Quantification of ^{210}Pb and ^{210}Po transfers to agricultural animals

6.6.2.1. Absorption

The International Commission on Radiological Protection (ICRP) recommends a relatively high fractional gastrointestinal absorption value of 0.5 for polonium [6.43]. Considering ruminants in particular, data specifically for polonium gastrointestinal absorption have not been identified; however, a lower mean fractional gastrointestinal absorption value of 0.04 for lead is reported by

Howard et al. [6.44]. The value is within the range of 0.01–0.1 recommended for polonium in humans by the ICRP [6.43].

6.6.2.2. Transfer to animal food products

The major pathway for polonium transfer to animals is through ingestion of feed and water. Parameters used to estimate that transfer are the transfer coefficient — F_m and F_f for milk and flesh, respectively — and the CR (for a definition, see Chapter 2). Given its easy absorption in the gut, the transfer to meat and milk is relatively high for some animal products. As lead accumulates in bone, its transfer to milk and meat (although higher in some offal) might be expected to be lower than polonium. However, data to make the comparison are presently inadequate to test this assumption for most products (see Table 6.9).

The transfer coefficient for polonium to cow milk is based on four data sources and is lower than that for goat milk [6.45–6.48]. The difference does not mean that higher activity concentrations are expected in goat milk, as transfer coefficients include the amount of feed ingested [6.4, 6.5, 6.49]. Since CRs for milk for all ruminants are similar, it is likely that the polonium CR for cow milk would be a reasonable value to use for polonium in goat and sheep milk [6.49]. The polonium CR for beef is based on only one data source [6.50] and there is some uncertainty concerning its derivation (lack of information on weight basis), so it has not been included in Table 6.9.

Using data from the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR), the reference concentration (FW) for ^{210}Po in milk and meat is 15 mBq/kg and 60 mBq/kg, respectively [6.51]. When this is compared with a mean value (33.5 Bq/kg, DW) of natural steppe and sawn grass (see Table 6.2) and all grass species (see Table 6.3), the respective CRs are 4.5×10^{-4} and 1.8×19^{-3}. These CRs are based on specific types of ecosystem for vegetation and unassociated milk and are therefore uncertain. However, the value for cow milk is similar to Ref. [6.5]. A direct comparison of transfer of lead and polonium is only possible for cow milk. In this case, both F_m and CRs are similar for the two elements. The CR for beef would be expected to be lower than for reindeer meat, so the value given based on UNSCEAR data [6.51] might be more appropriate than the value in Ref. [6.5].

The transfer coefficient for polonium is relatively high for poultry meat and egg contents. However, this might be the result of the values being derived from unrelated feed and food samples [6.52].

The equivalent transfer values for lead are given in Table 6.10. In addition, similar values for cow milk of F_m and CR of 3.6×10^{-4} and 7.3×10^{-3}, respectively, are reported in Ref. [6.53], which also reports higher values of F_f and CR for beef (n = 3) for lead of 0.01 and 0.11, respectively. The values

TABLE 6.9. TRANSFER COEFFICIENTS AND CONCENTRATION RATIOS FOR POLONIUM TRANSFER TO ANIMALS AND FOODSTUFFS

Animal product	Transfer coefficient (d/L or d/kg)					Concentration ratio	
	N	Mean	GSD	Min.	Max.	*N*	CR
Cow milk	4	2.1×10^{-4}	1.8	8.9×10^{-5}	$3. \times 10^{-4}$	1	2.4×10^{-3}
Goat milk	2	2.2×1^{-3}	n.a.[a]	1.8×10^{-3}	2.7×10^{-3}	—[b]	—[b]
Poultry	1	2.4	n.a.[a]	n.a.[a]	n.a.[a]	—[b]	—[b]
Reindeer meat[c]	—[b]	5×10^{-2}	—[b]	—[b]	—[b]	—[b]	5×10^{-2}
Egg	1	3.1	n.a.[a]	n.a.[a]	n.a.[a]	—[b]	—[b]

Sources: See Refs [6.5, 6.31].
Note: CR — concentration ratio; GSD — geometric standard deviation.
[a] n.a.: not applicable.
[b] —: data not available.
[c] Assuming lichen intake [6.31].

incorporate review information from animal nutrition studies as well as other literature on metal transfer.

6.6.3. Transfer to terrestrial animals

The transfer of polonium to products such as game species is not quantified in Refs [6.4, 6.5], and therefore, data specifically on the human food chain are not available. However, the transfer of radionuclides to wildlife has recently been published in Refs [6.54, 6.55], in which the transfer to terrestrial animals is reported as CRs between the fresh weight whole body (excluding shell, gut and contents, feathers and pelt) and dry weight soil for a few species (see Table 6.11).

The $CR_{wo\text{-}soil}$ for polonium transfer to terrestrial animals is high compared to many other radionuclides. These values are not directly comparable to those given above for beef, as the CRs are for the whole organism (including bone), and the denominator is soil, not ingested feed. The especially high transfer of polonium to reindeer is well known and has already been discussed [6.56]. The $CR_{wo\text{-}soil}$ for different mammal categories varies widely, with a much lower value for herbivores than those carnivores or omnivores. The high $CR_{wo\text{-}soil}$ for reptiles is influenced by relatively high values from Australia [6.57] and may be due to acidic spray from mine tailings [6.58].

TABLE 6.10. TRANSFER COEFFICIENTS FOR LEAD TRANSFER TO ANIMALS AND FOODSTUFFS

Animal product	Transfer coefficient (d/L and d/kg)					Concentration ratio	
	N	Mean	GSD	Min.	Max.	*N*	CR
Cow milk	4	1.9×10^{-4}	1.0	7.3×10^{-6}	1.2×10^{-3}	1	2.4×10^{-3}
Goat milk	1	6.0×10^{-3}	n.a.[a]	n.a.[a]	n.a.[a]	1	9.0×10^{-3}
Sheep milk	1	3.5×10^{-2}	n.a.[a]	n.a.[a]	n.a.[a]	1	3.0×10^{-2}
Beef	5	7.0×10^{-4}	2.5	2.0×10^{-4}	1.6×10^{-3}	11	7.7×10^{-2}
Mutton	2	7.1×10^{-3}	—[b]	4.0×10^{-3}	1.0×10^{-2}	3	1.2×10^{-2}
Pork	—[b]	—[b]	—[b]	—[b]	—[b]	2	6.6×10^{-1}

Source: See Ref. [6.5].
Note: CR — concentration ratio; GSD — geometric standard deviation.
[a] n.a.: not applicable.
[b] —: data not available.

TABLE 6.11. TRANSFER OF POLONIUM TO THE WHOLE BODY OF TERRESTIAL ANIMALS

Wildlife group	$CR_{wo\text{-}soil}$						
	N	AM	ASD	GM	GSD	Min.	Max.
Annelid	7	1.0×10^{-1}	3.9×10^{-2}	9.6×10^{-2}	1.4	—[a]	—[a]
Bird							
Herbivorous	5	1.0×10^{-2}	2.9×10^{-3}	9.6×10^{-3}	1.3	—[a]	—[a]
Mammal	67	8.6×10^{-2}	2.1×10^{-1}	3.3×10^{-2}	4.0	2.4×10^{-4}	1.1
Carnivorous	11	1.2×10^{-1}	8.7×10^{-2}	9.7×10^{-2}	1.9	1.9×10^{-2}	1.4×10^{-1}
Herbivorous	38	2.9×10^{-3}	1.9×10^{-3}	2.4×10^{-3}	1.8	2.4×10^{-4}	9.5×10^{-3}
Omnivorous	10	2.1×10^{-1}	1.2×10^{-1}	1.8×10^{-1}	1.7	7.5×10^{-4}	2.6×10^{-1}
Rangifer spp.	199	2.5	3.7	1.4	3.0	5.9×10^{-1}	21
Reptile	15	9.5	23	3.6	4.0	1.9×10^{-2}	11

Source: See Refs [6.54, 6.55].
Note: $CR_{wo\text{-}soil}$ is the activity concentration in the whole organism (in Bq/kg, FW) divided by the activity concentration in the soil (in Bq/kg, DW). *Rangifer* spp. include reindeer and caribou. AM — arithmetic mean; ASD — arithmetic standard deviation; CR — concentration ratio; GM — geometric mean; GSD — geometric standard deviation.
[a] —: data not available.

The $CR_{wo\text{-}soil}$ for lead transfer to terrestrial animals (see Table 6.12) are generally lower than those for polonium. As for polonium, there is relatively high transfer to *Rangifer* species. The mammal values are similar for all of the subcategories. The mammal values in Tables 6.11 and 6.12 are not directly comparable to those given for beef in Tables 6.9 and 6.10 as the transfer coefficient includes dietary intake and the CR compares muscle (not whole body) with ingested feed and not soil.

TABLE 6.12. TRANSFER OF LEAD TO THE WHOLE BODY OF DIFFERENT WILDLIFE GROUPS

Wildlife group	$CR_{wo\text{-}soil}$						
	N	AM	ASD	GM	GSD	Min.	Max.
Amphibian	24	1.2×10^{-1}	5.2×10^{-1}	2.7×10^{-2}	5.6	8.8×10^{-4}	2.8×10^{-1}
Annelid	647	5.2×10^{-1}	7.5×10^{-1}	2.9×10^{-1}	2.9	2.3×10^{-3}	2.8
Arachnid	2	5.3×10^{-2}	—[a]	—[a]	—[a]	4.3×10^{-2}	6.2×10^{-2}
Arthropod	561	4.0×10^{-1}	4.7×10^{-1}	2.6×10^{-1}	2.5	4.6×10^{-3}	1.0
Bird							
Carnivorous	424	6.2×10^{-2}	1.7×10^{-1}	2.1×10^{-2}	4.4	—[a]	—[a]
Mammal	515	3.8×10^{-2}	3.6×10^{-2}	2.8×10^{-2}	2.2	2.7×10^{-4}	2.0×10^{-1}
Carnivorous	368	4.7×10^{-2}	2.8×10^{-2}	4.0×10^{-2}	1.7	8.8×10^{-3}	7.7×10^{-2}
Herbivorous	92	2.0×10^{-2}	2.7×10^{-2}	1.2×10^{-2}	2.8	1.9×10^{-3}	2.0×10^{-1}
Omnivorous	51	1.2×10^{-2}	6.3×10^{-2}	2.2×10^{-3}	6.3	2.7×10^{-4}	3.9×10^{-2}
Rangifer spp.	270	3.6	3.3	2.7	2.2	4.0×10^{-1}	18
Gastropod	47	7.3×10^{-3}	1.3×10^{-2}	3.6×10^{-3}	3.3	6.1×10^{-4}	3.8×10^{-2}
Reptile	45	3.7×10^{-1}	1.0	1.3×10^{-1}	4.3	1.4×10^{-3}	1.2
Carnivorous	32	3.8×10^{-2}	1.6×10^{-1}	8.7×10^{-3}	5.6	1.4×10^{-3}	7.0×10^{-2}

Source: See Refs [6.54, 6.55].

Note: $CR_{wo\text{-}soil}$ is the activity concentration in the whole organism (in Bq/kg, FW) divided by the activity concentration in the soil (in Bq/kg, DW). *Rangifer* spp. include reindeer and caribou. AM — arithmetic mean; ASD — arithmetic standard deviation; CR — concentration ratio; GM — geometric mean; GSD — geometric standard deviation.

[a] —: data not available.

Generally, the behaviour of ^{210}Po in terrestrial ecosystems depends on many processes which have been studied little. The available data are largely based on field observations that attempt to evaluate the role of individual factors governing the behaviour of ^{210}Po. However, it is not always possible to identify these processes in uncontrolled conditions. Furthermore, the quantity of ^{210}Po in environmental compartments depends to some extent on the ambient concentrations of ^{210}Pb and the behaviour of ^{210}Pb and ^{210}Po. Therefore, both aspects should be considered when trying to explain any observed patterns of ^{210}Po accumulation in specific environmental receptors. The generic data presented in this chapter can be used for assessing the impacts of polonium on the environment. However, these values are often based on only a few measured values or single observations, which are sometimes contradictory. Therefore, the data presented here should be considered with some caution, bearing in mind the relatively high uncertainty associated with many of the values.

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Chapter 7

^{210}Pb AND ^{210}Po IN FRESHWATER AND GROUNDWATER SYSTEMS

7.1. DESCRIPTION OF THE HYDROLOGICAL CYCLE

The continuous movement of water on, above and below the surface of the Earth is called the hydrological cycle. Throughout this cycle, water changes from one phase to another: from solid to liquid to vapour (see Fig. 7.1).

Precipitation runs off the land surface into streams and rivers to fill standing bodies of water, such as natural lakes and artificial impoundments, on its way to the sea and comes into contact with river and lake-bed sediments as it moves. Some evaporates, but a large proportion of precipitation also infiltrates the ground, where it interacts with soils and rocks. The infiltrating waters then recharge the underlying groundwater systems (see Section 7.2). Groundwater discharges into surface waters, such as lakes, rivers and wetlands, as well as into coastal waters. All these waters and discharges can carry polonium.

The surface water environment includes streams, rivers, lakes, bogs, fens and wetlands, along with the sediment in each of these water bodies (see Section 7.3). Polonium in surface water can be directly transferred to other surface water bodies, or can deposit and migrate in bottom sediments. It can also

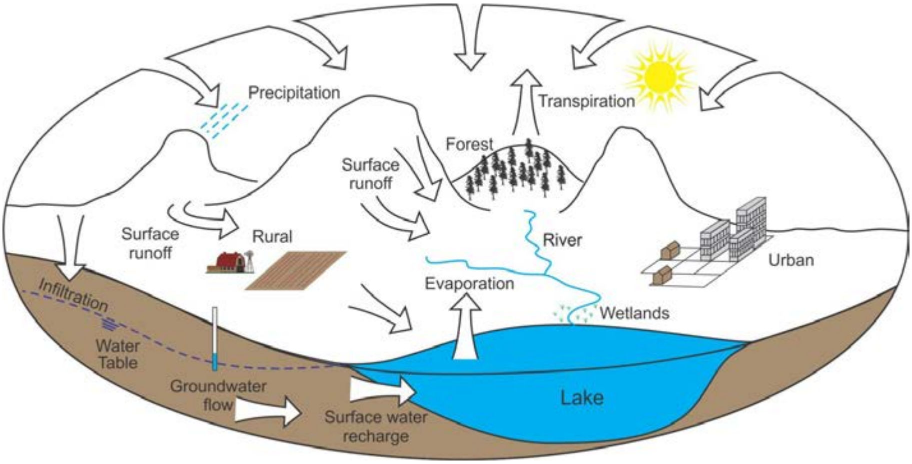


FIG. 7.1. Illustration of the hydrological cycle.

be taken up by freshwater organisms, thus entering the food chain. The transfer in surface water occurs through a combination of processes, including [7.1]:

(a) Diffusion and dispersion caused by gradients of concentration and the turbulent motion of water;
(b) Transport caused by water currents;
(c) Exchanges of contaminants between the dissolved and solid phases (suspended solids);
(d) Deposition and remobilization following interaction with suspended matter and bottom sediments.

Hydraulic processes responsible for radionuclide transfer by water are mainly independent of radionuclide properties. Exchanges of radionuclides between the dissolved and solid phases, as well as interaction with abiotic suspended matter and bottom sediments, are strongly radionuclide specific. Quantification of radionuclide redistribution in freshwater ecosystems requires the evaluation of the physicochemical properties of radionuclides in water and sediments. A description of how these processes can be represented in a quantitative manner is provided in Ref. [7.2].

Polonium in water can then transfer to biota, such as plants, phytoplankton, zooplankton, other invertebrates (including molluscs and crustaceans), fish and water based amphibians, as well as to reptiles, mammals and birds that obtain dietary components from the aquatic environment.

7.2. GROUNDWATER

7.2.1. Groundwater environment

Groundwater is an important component of the hydrological cycle and source of drinking water for humans. A simplified typical aquifer system is shown in Fig. 7.2. While water recharging the groundwater contains some ^{210}Po from rainwater, any surface deposition or releases and interactions in the vadose zone, the ^{210}Po concentrations further in the aquifer are controlled by local processes, as steady state conditions are established over very short distances and tend to break down over longer distances. In recharge zones, as precipitation enters the subsurface, there are a number of sources of polonium in the water:

- Polonium contained in the rainwater or deposited on soils from the atmosphere (i.e. wet or dry deposition; see Chapter 5);
- Polonium already in soils (see Chapter 6);

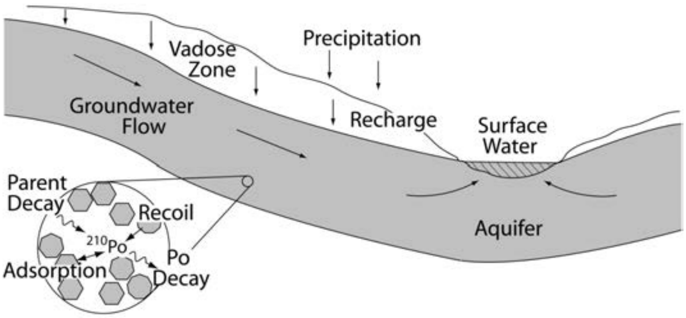


FIG. 7.2. A simplified view of groundwater flow and the dominant processes controlling the distribution of ^{210}Po within aquifers.

- Anthropogenic releases (see Chapter 4);
- Polonium produced by progenitor nuclides in the ^{238}U decay series (see Fig. 3.1, in Chapter 3) that have similar origins.

However, owing to its short half-life, ^{210}Po from sources confined to the near surface cannot migrate far before decaying away. As the water begins to interact with the surrounding minerals, both within the vadose zone and the underlying aquifers, there are a number of factors that control the distribution of polonium in the groundwater, including: the decay of the parent ^{210}Bi (which is the very short lived daughter of ^{210}Pb) that is dissolved or adsorbed; the decay of ^{210}Po, supply by recoil from mineral grains; and adsorption. The short half-life means that the concentration is determined by the conditions in the immediate area. Groundwater discharges into surface waters (see Section 7.3) and coastal waters and are thus important inputs to these environments. These inputs are likely to be moderated by interactions within sediments at the sediment–water interface, although there is insufficient data to document the general characteristics of the relationships between discharging groundwater and surface waters.

The behaviour of polonium in groundwater has been studied much less than on other radionuclides — presumably because it is relatively immobile and short lived. However, it has sometimes been found to be a significant contributor to the overall gross alpha activity of groundwater, which has in turn led to a number of regional studies.

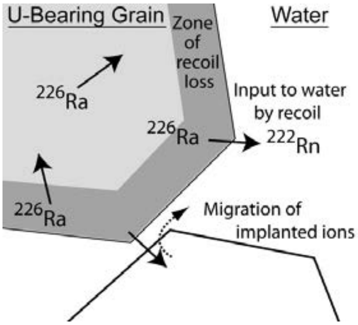
7.2.2. Sources of ^{210}Pb and ^{210}Po in groundwater

An important input of ^{210}Po is the decay of parent isotopes dissolved within the groundwater or adsorbed onto aquifer surfaces. Therefore, the distribution of progenitor atoms is important, with the distribution of uranium controlling the distribution and release of ^{230}Th, which, in turn, controls that of ^{226}Ra, which affects the release of ^{222}Rn by recoil, and so the concentrations of the short lived nuclides further along the chain (see Section 7.2.5).

A dominant input to groundwater for many constituents is weathering of aquifer rocks and thus chemical releases from mineral structures [7.3]. This is important for uranium, thorium and radium (all progenitors of ^{210}Po), and is responsible for much of the redistribution of the long lived nuclides. For short lived nuclides, however, this is generally not important compared to other sources, and so can be neglected [7.4].

The most important mechanism for the release of short lived nuclides from within mineral structures is recoil (see Figs 7.2 and 7.3). Whenever an atom is subject to energetic alpha decay, the daughter atom is recoiled in a random direction (though directly opposite to that of the alpha particle) over a distance of approximately 20 nm in most mineral lattices [7.3]. When this recoil path crosses a grain boundary (mineral surface), the daughter atom can be released to a solution in the groundwater [7.5, 7.6]. Atoms implanted into adjacent phases may escape as well, along the alpha track defect in the mineral lattice generated during the implantation event [7.3].

The stopping power of water is substantially lower than mineral lattices, and so atoms can cross pores and become implanted in adjacent grains, from which the ion may migrate back to the solution [7.3]. This distance is somewhat dependent on the lattice characteristics as well as on the decay energy (see the review in Ref. [7.4]), although the recoil characteristics of the alpha decay



Source: Figure 2 of Ref. [7.3].

FIG. 7.3. A major source of short lived nuclides in groundwater is recoil (reproduced with permission courtesy of Elsevier).

products along the decay series are similar. Quantifying this input is discussed further in Section 7.2.5.

Owing to the short half-life of ^{210}Po, its levels in groundwater are generally in a steady state, such that the removal rates are equal to the input rates. While much of the polonium is adsorbed, this may be readily exchangeable with dissolved species (see Section 7.2.5). The main removal process of adsorbed and dissolved ^{210}Pb is simply decay. Irreversible incorporation of ^{210}Po into precipitating phases is generally too slow to be a significant removal flux.

7.2.3. Groundwater concentrations

Porcelli [7.3] reports that reconnaissance studies of groundwater across the United States of America [7.7] and across California [7.8] find groundwater ^{210}Po concentrations to be typically less than 40 mBq/L. Values as high as 16 Bq/L have been recorded in a well in Finland [7.9], and brines in Australia can have values of up to 19 Bq/L [7.10]. A number of factors control the variations in concentrations of ^{210}Po in groundwater, including variations in input and removal rates. These factors are considered below, followed by an evaluation of polonium adsorption coefficients.

7.2.4. Particles and colloids in groundwater

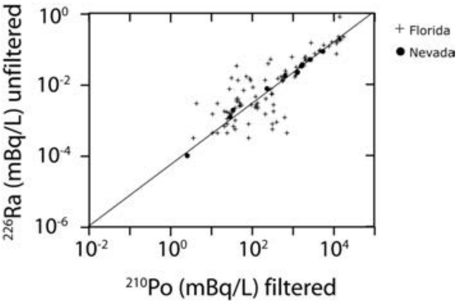
Polonium has been found to be associated with colloids and particles in groundwater, as well as being dissolved. This is expected because polonium is very particle reactive, and particles are often composed of clays and compounds of manganese and iron that have strong adsorption coefficients for polonium (see Chapter 3). Not separating these from truly dissolved constituents can lead to misleading conclusions regarding chemical behaviour. These species are usually operationally defined by filtration methods, with particles being material excluded from 0.2 μm or 0.45 μm filters, and colloids constituting material passing this filter but not molecular filters or ‘ultrafilters’ of 5–20 kilodaltons. Some large molecules, such as humic acids, are in the colloidal pool, while iron and manganese oxyhydroxides and clays may be present at sizes that can be either colloidal or particulate.

There have been several polonium speciation studies of groundwater from Finland. In low ionic strength Ca–HCO_3 waters, 35–68% of ^{210}Po was bound to particles, while more than 90% was bound to particles in higher salinity Na–Cl waters that have high concentrations of iron, manganese and humic acids [7.11]. The colloid fraction (between 5 kilodaltons and <0.45 μm) contained approximately less than 10% [7.12]. Vesterbacka et al. [7.13] report that, on average, 86% of ^{210}Pb in groundwater is found in the large particle fraction (>0.45 μm). However, for waters that are rich in organics, iron and manganese, and which have high Fe:Mn ratios, the majority of ^{210}Po is found in either the intermediate particle (100 kilodaltons and 0.45 μm) or small particle (<100 kilodaltons) fractions.

Polonium, with such low natural elemental concentrations (due to its relatively short half-life), clearly does not form its own particles but rather is adsorbed onto, or incorporated into, other phases. While early laboratory experiments suggested that polonium in simple solutions can form colloids [7.14], it is present in extremely low concentrations compared to other colloid forming elements and is highly reactive with surfaces; so that in natural environments, it is likely to be associated with colloids of mixed composition [7.12].

Studies of groundwater from aquifers in Florida [7.15] and Nevada [7.16], United States of America, provide comparisons between filtered and unfiltered samples (see Fig. 7.4). Data cluster around the line for equal concentrations, especially for the Nevada samples, although significant fractions of ^{210}Po appear to be present on particles (data above the line). However, there are significant deviations in the Florida samples. Filtering of particles from Nevada groundwater removed 5–47% of ^{210}Po [7.16]. One of the highest concentrations recorded (95 Bq/L) is for an unfiltered Florida groundwater; the filtered sample contained 17% ^{210}Po [7.17]. Such a high concentration suggests that particles with high ^{226}Ra were present. The most perplexing are some samples that appear to have higher concentrations when filtered. This can only be a sampling or analytical artefact.

The strong association of polonium with particles has a number of implications for interpreting polonium behaviour. Particles can be mobile,



Source: See Refs [7.15, 7.16].

FIG. 7.4. Data for groundwater (filtered and unfiltered) from aquifers in Florida and Nevada, United States of America.

although not necessarily at the same rate as dissolved species. There might be sampling biases in quantifying the particle load, since the particle load in samples collected from wells might not be representative of aquifer groundwater; removal of particles can occur in the filter pack around the well; or increased mobilization of particles around the well can occur from pumping, resulting in enhanced flow rates. Furthermore, if particle bound polonium is not separated, the amount of dissolved species will be overestimated. In some cases, a significant fraction of polonium can also be colloid bound. Overall, such effects should be further investigated, especially where unusual occurrences of polonium occur. Routine practice should include reporting data for both unfiltered and filtered samples, and filtration should be done during collection, or as soon as possible after, since association with particles can change during storage.

7.2.5. In situ adsorption coefficients

7.2.5.1. Quantifying polonium adsorption

As with most trace elements, the extent of ^{210}Po adsorption can be determined from the analysis of aquifer rocks or laboratory experiments. However, it is difficult to obtain a bulk average aquifer value for a process that is expected to vary substantially between samples due to the heterogeneous distribution of secondary phases. Furthermore, as discussed in Chapter 4, this has been done for polonium on only a small number of soils, sediments and aquifer materials. Porcelli [7.3] finds that:

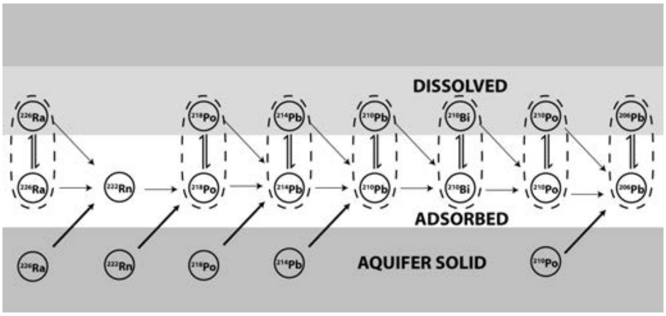
> “Studies involving decay-series systematics of nuclides in groundwater have quantified parameters of water–rock interaction from direct measurements of waters alone, with source terms and adsorption coefficients inferred from comparison between the different radionuclides. Sampling of large volumes or from a number of wells can obtain parameter values for larger volumes of aquifer. In particular, the rate of recoil supply to groundwater, which supplies all of the daughter nuclides at similar rates, and the evolution and steady state concentrations of the short-lived nuclides can be determined. Then bulk adsorption partition coefficients can be obtained.”

7.2.5.2. Calculating average aquifer adsorption coefficients

Porcelli [7.3] reports that mathematical treatments of simple aquifer models have been extensively developed [7.18–7.23] and summarized in Refs [7.4, 7.24]. These provide operational rates of transport and of water–rock interaction processes, regardless of the actual mechanisms involved, and so complement physical chemistry studies that can identify the controlling chemical processes. These studies have so far largely considered steady state groundwater profiles; that is, they assume that the conditions at each location within the aquifer do not change, and so groundwater radionuclide concentrations are constant at each location but may change along flow lines. Furthermore [7.3]:

> “In general, the decay series systematics for groundwater behavior can become quite complex, as there are a number of parameters that affect each isotope, and the resulting distribution then affects the production of the next isotope in the chain. However, the element with the simplest and most predictable behavior is ^{222}Rn, an unreactive noble gas, and the distribution of isotopes further along the ^{238}U decay series, including ^{210}Po, can be estimated from consideration of the distribution of ^{222}Rn.
>
> “The section of the ^{238}U decay series that is most directly relevant to understanding ^{210}Po is below ^{226}Ra, as shown in Fig. [7.5]. For each nuclide, the sum of dissolved and adsorbed atoms, that is the atoms that have been released from aquifer solids and are chemically exchanging, can be considered together as the mobile pool. Further, in most cases, radioisotope concentrations will have reached a steady state concentration, where the supply of the isotope from within the minerals and from decay of mobile atoms of the parent will together equal the decay rate (that is, the activity) of the mobile pool of the isotope. For the short-lived nuclides below ^{222}Rn, it is generally the case (see Porcelli and Swarzenski, 2003 [Ref. [7.24]]) that for very short-lived nuclides, recoil is much more important as an input than weathering, which can then be ignored. Also, the decay rates of short-lived adsorbed and dissolved nuclides are generally much greater than precipitation rates in groundwaters, and so this also can generally be ignored (Porelli, 2008 [Ref. [7.4]]). Of course, there are circumstances when these factors do need to be considered, e.g. in rapidly weathering horizons, or preferential rapid weathering of minerals rich in U (and so also rich in all daughter nuclides) minerals, or where abrupt changes in aquifer chemistry cause rapid precipitation. However, in the absence of evidence for these, weathering and precipitation can be ignored.”

The models have been applied to the available data to obtain recoil supply rates for ^{210}Po, as well as K_d values for both ^{210}Pb and ^{210}Pb [7.3]. Porcelli [7.3] states that the distribution of ^{210}Po can be evaluated by starting with ^{222}Rn. The radionuclide ^{222}Rn does not precipitate or adsorb, so that groundwater (^{222}Rn)



Source: Figure 3 of Ref. [7.3].

FIG. 7.5. The ^{238}U decay series from ^{226}Ra to ^{206}Pb, showing the recoil fluxes from within the aquifer solid and so supplying the mobile pool of dissolved and adsorbed radionuclides in the aquifer (reproduced with permission courtesy of Elsevier).

activity is equivalent to ^{222}Rn recoil plus the production from mobile ^{226}Ra [7.4], which is otherwise expressed as [7.3]:

$$\left(^{222}\,\mathrm{Rn}\right)_{\mathrm{W}} = b\varepsilon_{222}\left(^{226}\,\mathrm{Ra}\right)_{\mathrm{R}} + \left(^{226}\,\mathrm{Ra}\right)_{\mathrm{W}}\left(\mathrm{K}_{266}\right) \tag{7.1}$$

where

b	is the ratio of rock mass to water mass per unit volume;
ε_{222}	is the fraction of ^{226}Ra decays that recoil daughter atoms to the groundwater;
$(^{226}\mathrm{Ra})_R$	is the concentration of ^{226}Ra in the aquifer rock;
$(^{226}\mathrm{Ra})_W$	is the concentration of dissolved ^{226}Ra;

and (K_{226}) is the ratio of atoms of ^{226}Ra adsorbed to atoms in solution. Porcelli [7.3] continues:

> “The ^{222}Rn in solution will produce ^{218}Po, which will largely adsorb. In addition, there will be recoil of ^{218}Po into solution from decay of ^{222}Rn within the minerals. The decay rate of mobile ^{218}Po (adsorbed and dissolved) will equal the input rates. Mobile ^{218}Po will then produce mobile ^{214}Po, along with further recoil of ^{214}Pb, so that mobile ^{214}Pb activity will be equal to that of dissolved ^{222}Rn plus the addition of two recoil fluxes. The same will apply to ^{210}Pb (produced by decay of ^{214}Pb to the very short-lived ^{214}Bi, then the very short-lived ^{214}Po), which will have an activity of adsorbed and dissolved atoms of ^{222}Rn plus 3 recoil fluxes. Since ^{210}Bi and ^{210}Po are

> produced by low energy beta decay, there are no recoil contributions to these nuclides (see Fig. [7.5]). Therefore, the activity of mobile ^{210}Pb, ^{210}Bi and ^{210}Po will be equal."

Combining these effects together gives [7.3]:

$$\left({}^{210}\mathrm{Pb}\right)_\mathrm{W} = \frac{3b\varepsilon_{210}\left({}^{226}\mathrm{Ra}\right)_\mathrm{R} + \left({}^{222}\mathrm{Rn}\right)_\mathrm{W}}{\left(\mathrm{K}_{210_\mathrm{Pb}}\right)} \tag{7.2}$$

The activity of mobile ^{210}Pb is equal to that of mobile ^{210}Po, so that [7.3]:

$$\frac{\left({}^{210}\mathrm{Pb}\right)_\mathrm{W}}{\left({}^{210}\mathrm{Po}\right)_\mathrm{W}} = \frac{\mathrm{K}_{210_\mathrm{Po}}}{\mathrm{K}_{210_\mathrm{Pb}}} \tag{7.3}$$

If the production of ^{222}Rn is largely from recoil, so that the term for production from adsorbed ^{226}Ra can be neglected, then [7.3]:

$$\left({}^{222}\mathrm{Rn}\right)_\mathrm{W} = b\varepsilon_{222}\left({}^{226}\mathrm{Ra}\right)_\mathrm{R} \tag{7.4}$$

and

$$\mathrm{K}_{210_\mathrm{Pb}} = \frac{4}{\left({}^{210}\mathrm{Pb}\right)_\mathrm{W} / \left({}^{222}\mathrm{Rn}\right)_\mathrm{W}} \tag{7.5}$$

Porcelli [7.3] concludes by stating that there may be some circumstances in which there is sufficient ^{226}Ra adsorbed to dominate ^{222}Rn production, which will reduce $\mathrm{K}_{210\mathrm{Pb}}$ by a factor of four. However, the range in $\mathrm{K}_{210\mathrm{Pb}}$ is substantially greater and so this will generally not be a major consideration. Moreover [7.3]:

> "As discussed above, the values for $\mathrm{K}_{210\mathrm{Po}}$ and $\mathrm{K}_{210\mathrm{Pb}}$ are equal to the number of atoms adsorbed divided by the number of atoms in solution within any volume of aquifer (i.e. in units of mol/mol). In order to translate this into the more familiar values of atoms per kg of rock divided by atoms per liter of groundwater (i.e. L/kg), the porosity of the aquifer must be known. Since this is often not reported or not well constrained, the values calculated below are for moles adsorbed on aquifer solids relative to moles in solution, or mol/mol."

Further information can be found in Ref. [7.3], including site specific adsorption coefficients for groundwater in Australia, Brazil, Scandanavia and the United States of America.

7.3. SURFACE WATER

The freshwater environment encompasses lotic (moving) water, such as rivers and streams, and lentic water bodies (standing), such as lakes and ponds. The concentrations of ^{210}Po within moving water depend on the environmental conditions in the catchment area and reflecte both natural and anthropogenic sources. The concentrations throughout the year is expected to vary, since precipitation carries particulates from the surrounding areas into the water body.

Polonium in water can transfer to biota such as plants, phytoplankton, zooplankton, invertebrates, fish, water based amphibians, crustaceans, mammals and birds with dietary components from the aquatic environment (see Fig. 7.6).

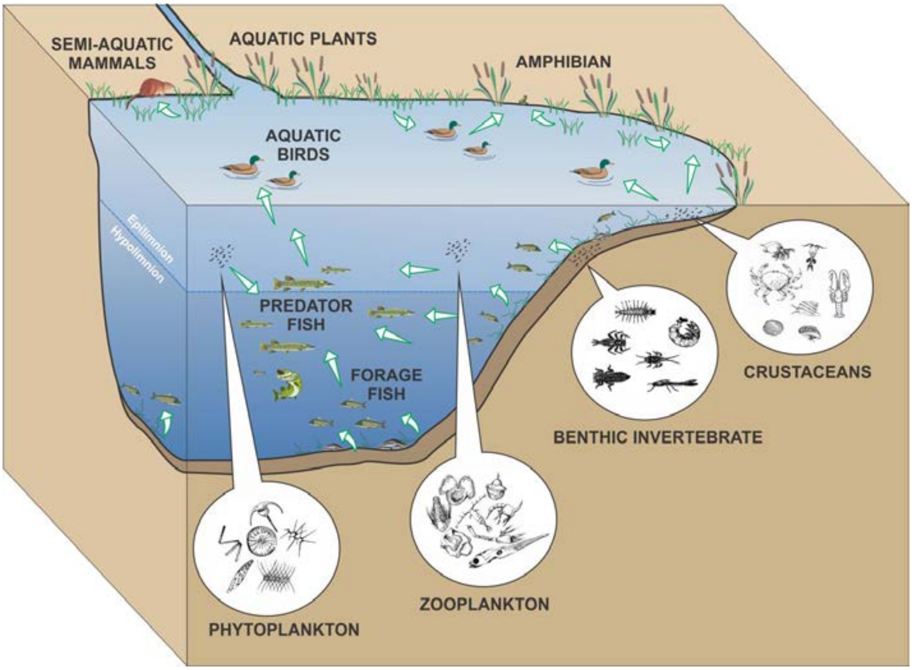


FIG. 7.6. Conceptual model of an aquatic ecosystem.

The interactions among the various abiotic and biotic environments depends on the location. The International Union of Radioecology (IUR) suggests that interaction matrices provide a convenient way to consider information on relevant processes in the aquatic environment in a synthesized and structured way [7.25]. Each environment has its own unique chemical, physical and biological characteristics. Nonetheless, the generic interaction matrices for the aquatic environment provided by the IUR [7.25] and the associated generic discussions provide a convenient checklist for initial discussions of the relevant processes.

The aquatic environment is characterized by the flow regime, water and sediment quality, and local species, including primary producers, and primary and secondary consumers. The IUR identifies the following main components for the aquatic environment:

(a) The atmosphere, which is the region above the surface;
(b) Water, including surface water bodies (e.g. rivers, streams and lakes);
(c) Abiotic suspended matter, which is inorganic particles (>0.45 μm) suspended in water;
(d) Deposited matter (sediment), which is particulate matter (inorganic and organic) deposited at the bottom of rivers, streams and lakes, including interstitial water;
(e) Primary producers, which are autotrophic organisms, such as phytoplankton, macrophytes and aquatic plants;
(f) Primary consumers, which are animals that feed on primary producers (e.g. zooplankton and macrobenthos);
(g) Secondary consumers, which are animals that feed on primary consumers (e.g. omnivorous fish);
(h) Decomposers, which comprise microflora and protozoa.

7.3.1. Sources in surface fresh water

The sources of lead and polonium in surface waters include:

— Atmospheric deposition of resuspended dusts or aerosols;
— Fluvial inputs;
— In situ decay from progenitors (particularly ^{226}Ra in sediment; depending on mixing rates, the interceding ^{222}Rn is often likely to decay before diffusing to the water surface);
— Wash-off from the terrestrial environment;
— Anthropogenic inputs (e.g. phosphate ore processing operation and uranium mining).

Radionuclides dispersed in the environment can be deposited into surface water or onto surfaces within the watershed, which, in the latter case, can represent a long term source of radionuclides for freshwater ecosystems within the affected region.

7.3.2. Levels of ^{210}Po in the freshwater environment

The activity of ^{210}Po in normally oxic waters is in the range of 1–5 mBq/L and is higher, up to 17 mBq/L, in seasonally anoxic ponds [7.26]. Table 7.1 provides a summary of ^{210}Po measurements in the freshwater environment of rivers and lakes. The amount of data for freshwater biota is less than for the marine environment (see Table 7.2).

A similar pattern in freshwater biota has been observed when compared to marine biota (see Chapter 8), in that higher concentrations have been reported in the soft tissue of molluscs than in shells, whereas ^{210}Pb has been detected in higher levels in the shells. A mollusc shell is largely composed of inorganic substances, such as calcium carbonate, while the exoskeleton of prawn consists of organic substances, such as chitin. As ^{210}Po has an affinity for organic moieties, this accounts for the relatively higher concentration of ^{210}Po in the exoskeleton of prawn [7.28].

7.3.3. Behaviour of ^{210}Po in standing water bodies

The behaviour of ^{210}Po in a water body is influenced by a combination of physical and chemical processes. Within a standing water body (e.g. lake), there is uptake of ^{210}Pb and ^{210}Po by particulates (particularly biomass) in the water column. As these particulates settle to the bottom sediment, polonium is scavenged from the water column. Within a few months, equilibrium between ^{210}Pb and ^{210}Po can be established in the sediment. However, changing redox conditions in the sediment and water column also influence the polonium levels. The chemical form (speciation) of sulphur in sediment is redox dependent and can, in turn, affect ^{210}Pb and ^{210}Po behaviour (lead forms sulphides, e.g. PbS). The cycling of polonium in sediment can be affected by microbial activity in the bottom sediment, as the release of polonium can be enhanced in sulphur enriched bacteria [7.42]. Polonium has also been shown to become volatile in both fresh and marine waters by the action of microorganisms [7.43, 7.44] (see Section 3.2).

In addition to cycling in sediment, depending on the characteristics of the lake, there can be zones within the water column that become anoxic during periods of the year, which influences the behaviour of polonium in the water column. Polonium has several oxidation states (−2, +2, +4 and +6), of which the insoluble tetravalent state Po(IV) is the most stable in oxic aqueous

TABLE 7.1. BASELINE CONCENTRATIONS OF ^{210}Po IN SURFACE WATER AND SEDIMENTS

Location	^{210}Po in water (mBq/L)	^{210}Po in sediments (Bq/kg)[a]	Ref.
Kaveri River, India[b]	0.77–1.27	14.4–26.5	[7.28]
Mutharasanallur Pond, India[b]	1.4	60	[7.29]
Rostov, Russian Federation[b]	1.8×10^{-3}	—[c]	[7.30]
Vistula River, Poland	1.94–3.21[d]	—[c]	[7.31]
Tributaries of the Vistula River, Poland	2.15–6.03[d]	—[c]	[7.31]
Oder River, Poland	1.46–2.39[d]	—[c]	[7.32]
Tributaries of the Oder River, Poland	1.02–3.64[d]	—[c]	[7.32]
Saskatchewan, Canada (control site)	—[c]	240[e]	[7.33]
Saskatchewan, Canada (unimpacted lake near the McArthur River site)	2	50[e]	[7.34]
Crystal Lake, Wisconsin, USA	1.6	—[c]	[7.35]
Bickford Reservoir, Massachusetts, USA	1.3	—[c]	[7.36]
Four Norwegian lakes	1.6–2	—[c]	[7.37]
South Alligator River, Australia (control site)	1.94 filtered 2.51 particulate	—[c]	[7.38]

[a] Fresh weight unless otherwise stated.
[b] Adapted from Ref. [7.27].
[c] —: data not available.
[d] May be influenced by anthropogenic sources.
[e] Bq/kg (DW).

solutions [7.45]. However, it can hydrolyse and form hydroxides. Polonium is influenced by the cycling of iron and manganese [7.26, 7.36, 7.46]. As oxygen levels decrease, iron and manganese are reduced and enter into solution, bringing with them transition metals, including ^{210}Pb and ^{210}Po, which are adsorbed onto

TABLE 7.2. CONCENTRATIONS OF ^{210}Po IN FRESHWATER BIOTA SPECIES

Sample	Part	^{210}Po (Bq/kg)[a]	Concentration ratios (L/kg)[a]	Ref.
Plankton[b]		19–29	$(2.1–2.5) \times 10^{4}$	[7.28]
Water hyacinth	Shoot	2.3–6.5	$(2.7–4.6) \times 10^{3}$	[7.28]
(*Eichhornia crassipes*)	Root	6.7–31	$(0.73–2.2) \times 10^{4}$	[7.29]
Bivalve mollusc	Shell	1.2–4.5	$(0.86–5.3) \times 10^{3}$	[7.28]
(*Lamellidens marginalis*)	Soft tissue	53–106	$(3.8–8.4) \times 10^{4}$	[7.29]
Bivalve mollusc	Shell	0.1[c]	—[d]	[7.39]
(*Anodonta cygnea*)	Soft tissue	19.3[c]	6.2×10^{3}[e]	
Bivalve mollusc	Soft tissue	5.7	2.1×10^{3}	[7.37]
(*Anodonta* sp.)	Shell	2.5	9.4×10^{2}	
	Whole	4.7	1.7×10^{3}	
Bivalve mollusc	Soft tissue	302–416[c]	4.7×10^{3}	[7.38]
(*Velesunio angasi*)				
Crustacean	Exoskeleton	8.6–16.6	$(0.81–1.2) \times 10^{4}$	[7.28]
(crab, prawn)	Muscle	12–20	$(0.93–1.6) \times 10^{4}$	[7.29]
Snail	Shell	0.2–3.9	$(0.14–5.3) \times 10^{3}$	[7.28]
(*Pila virens*)	Soft tissue	33–46	$(2.6–4.2) \times 10^{4}$	[7.29]
Fish	Bone	1.3–17	$(0.13–1.2) \times 10^{4}$	[7.28]
(*Mystus vittatus*,	Muscle	1.9–27	$(0.22–1.9) \times 10^{4}$	[7.29]
Orechromis mossambicus,				
Puntius chola)				
Fish	—[d]	0.36–0.43	—[d]	[7.30]
(perch, bream)[b]				
Fish[b]	Bone	0.37–0.43[c]	—[d]	[7.40]
Fish	Whole	1.0–6.5	$(0.63–9.3) \times 10^{3}$	[7.37]
	Edible	0.08–1.9	—[d]	
Fish	Bone	8–14	—[d]	[7.41]
(*Cyprinus carpio*,	Muscle	2–7	—[d]	
Sander lucioperca)	Liver	11–183		

[a] Fresh weight unless otherwise stated.

[b] Adapted from Ref. [7.27].

[c] Bq/kg (DW).

[d] —: data not available.

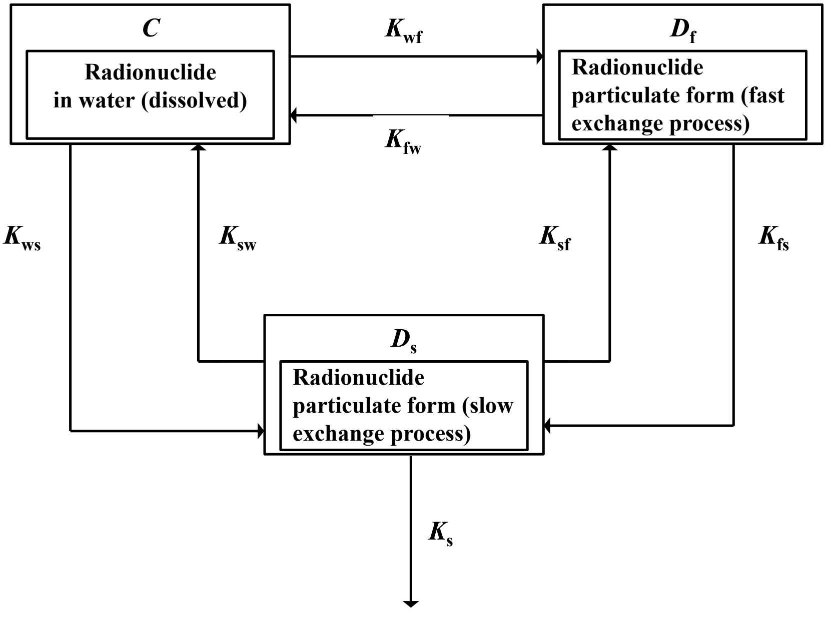
[e] L/kg (DW).

oxides. The insoluble Po(IV) is also reduced to Po(II) near the same redox potential that Mn(IV) is reduced to Mn(II) [7.36. 7.46]. The dissolved ions diffuse upwards and, as they become oxidized again, they form precipitates. Thus, changing redox conditions can have a significant effect on the levels of polonium in a water body. As a result, much higher levels of ^{210}Po have been measured in the anoxic zone of a water body (hypolimnion) compared to the epilimnion, and an enrichment of ^{210}Po compared to ^{210}Pb is observed [7.26, 7.36, 7.46].

Under certain conditions, polonium can diffuse out of the sediments and, the sediments can act as a sink for polonium.

7.3.4. Quantification of water and sediment quality

The interaction of radionuclides with bottom sediments and suspended particles is controlled by many environmental processes, such as sedimentation and resuspension (see Fig. 7.7).



Source: Figure 2 of Ref. [7.47].

Note: The fast processes are marked in the subscript by ‘f’, while the slow processes are marked by ‘s’.

FIG. 7.7. Schematic structure of contaminant fluxes and environmental compartments involved in the processes of radionuclide migration to and from bottom sediment.

Monte et al. [7.47] describe three active compartments:

- Dissolved radionuclide in water (C);
- Particulate radionuclide: rapid exchange component (D_f);
- Particulate radionuclide: slow exchange component (D_s).

Accordingly [7.47]:

> "The fluxes ($Bq \cdot s^{-1}$) from a compartment can be calculated as the product of the total amount of radionuclide in the compartment (Bq) multiplied by the 'rates' (s^{-1}) of migration (K_{wf}, K_{fw}, K_{sf}, K_{fs}, K_{ws}, K_{sw}, K_s).
>
> "The seven radionuclide fluxes can be schematized as follows:
>
> (a) Radionuclide fluxes from dissolved form to particulate form and *vice versa* — rapid exchange processes ($K_{wf} \cdot C$ and $K_{fw} \cdot D_f$);
> (b) Radionuclide fluxes from D_f to D_s and vice-versa ($K_{fs} \cdot D_f$ and $K_{sf} \cdot D_s$);
> (c) Radionuclide fluxes from water to D_s and vice-versa ($K_{ws} \cdot C$ and $K_{sw} \cdot D_s$);
> (d) Radionuclide irreversible burial in inactive sediments ($K_s \cdot D_s$)."

Under oxic conditions, an assessment of the interaction of dissolved radionuclides with solid particles in suspension or deposited, is often based on the K_d concept, assuming a presence of an equilibration between the dissolved (C_w, Bq/m^3) and the adsorbed phases (C_s, Bq/kg) of a radionuclide [7.47]:

$$K_d = \frac{C_s}{C_w} \tag{7.6}$$

There are very limited data available to determine a K_d for polonium. Ciffroy et al. [7.48] did not find a sufficient database of information. One study of polonium in a pond derived an overall K_d of 4.3×10^4 L/kg (FW) [7.29], whereas K_d values in a river were estimated at 1.5×10^5 L/kg (FW) for suspended matter to dissolved and 3×10^4 L/kg (FW) for bottom sediment to dissolved [7.49]. Application of these values should be treated with caution, as they are based on limited information and K_d values can vary widely.

7.3.5. Freshwater aquatic biota

The accumulation of ^{210}Po by aquatic biota is a dynamic process, with many pathways of exposure being involved. However, it is often simplified by assuming that abiotic and biotic media have reached steady state conditions and a concentration ratio (CR) can thus be established that relates the biota concentration to the concentration in the abiotic reference medium (water or sediment). These CRs can also be derived for those non-aquatic animals that depend on the aquatic environment, including mammals and birds.

There are two basic sets of data on radionuclide accumulation in aquatic organisms. The first approach is based on assessments of CRs for edible tissues of freshwater biota. Within the second approach, intended mainly for assessments of dose to biota, activity concentrations of radionuclides are calculated (or measured) for the whole organism. Yankovich et al. [7.50] describe methods for converting tissue concentrations to whole body values for a variety of species using CRs. The empirically derived CRs for lead and polonium are summarized in Table 7.3.

Until fairly recently, the Handbook of Parameter Values for the Prediction of Radionuclide Transfer in Temperate Environments [7.65] provided the basis for environmental assessments. Since that publication was released, new datasets have become available, and an update of that publication was considered appropriate. Two large scale reviews, in the framework of the international IAEA projects, Environmental Modelling for Radiation Safety (EMRAS I and EMRAS II), compiled the available data covering both transfers to human foodstuffs and the available quantitative data on the transfer of radionuclides to wildlife. The update was accomplished with the publication of the Handbook of Parameter Values for the Prediction of Radionuclide Transfer in Terrestrial and Freshwater Environments [7.66]. Much of the earlier data on biological uptake for many radionuclides was often on specific tissues and not the whole organism. The recent reviews were much wider in context, based on many sources and, hence, many more data for polonium were compiled. The CRs for ^{210}Pb and ^{210}Po are presented in Table 7.3 for broad wild biota groups and comprise a subset from Handbook of Parameter Values for the Prediction of Radionuclide Transfer to Wildlife [7.67] — an overview of which is provided in Ref. [7.51].

Although Ref. [7.66] represents a key reference for radioecologists, modellers and authorities by providing data for environmental impact assessments, it includes just a few freshwater CRs for lead and polonium. Hence, it has limited applicability for the impact assessment of naturally occurring radioactive material on freshwater species and their contamination of freshwater food.

From the available tissue data (see Table 7.2), the distribution of ^{210}Po within an organism shows higher concentrations in the kidney, liver and other organs in the digestive system. A higher accumulation of ^{210}Po is found in the soft tissue of molluscs than in shells, whereas ^{210}Pb has been detected at higher levels in shells. In the soft tissue of an organism, ^{210}Po is

TABLE 7.3. ^{210}Pb AND ^{210}Po CONCENTRATION RATIO ($CR_{WO\text{-}WATER}$) VALUES FOR WILDLIFE GROUPS IN FRESHWATER ECOSYSTEMS

Wildlife group	$CR_{wo\text{-}water}$							Ref.
	N	AM	ASD	GM	GSD	Min.	Max.	
^{210}Pb								
Amphibian	2	5.3	—[a]	—[a]	—[a]	1.7	8.9	[7.52]
Crustacean	5	3.9×10^1	4.7×10^1	2.5×10^1	2.6	—[a]	—[a]	[7.53]
Fish	379	2.5×10^2	7.0×10^2	8.7×10^1	4.3	2.0	7.5×10^3	[7.52–7.58]
Benthic feeding	148	1.8×10^2	6.3×10^2	4.8×10^1	5.0	3.2	7.5×10^3	[7.52–7.55, 7.58]
Forage	30	2.6×10^1	6.2×10^1	9.9	4.0	2.0	3.5×10^2	[7.52, 7.53, 7.55–7.57]
Piscivorous	201	3.5×10^2	7.8×10^2	1.4×10^2	3.8	8.3	5.7×10^3	[7.52–7.55]
Mollusc (bivalve)	32	6.0×10^3	—[a]	2.3×10^3	4.0	1.1×10^2	2.9×10^4	[7.38, 7.54, 7.59, 7.60]
Reptile	12	4.4×10^2	6.2×10^2	2.5×10^2	2.9	1.3×10^1	1.9×10^3	[7.61]
Vascular plant	21	6.2×10^1	7.0×10^1	4.1×10^1	2.5	1.3×10^1	1.9×10^2	[7.52, 7.60]

TABLE 7.3. ^{210}Pb AND ^{210}Po CONCENTRATION RATIO ($CR_{WO\text{-}WATER}$) VALUES FOR WILDLIFE GROUPS IN FRESHWATER ECOSYSTEMS (cont.)

Wildlife group	$CR_{wo\text{-}water}$							Ref.
	N	AM	ASD	GM	GSD	Min.	Max.	
^{210}Po								
Crustacean	12	8.3×10^3	7.0×10^3	6.3×10^3	2.1	1.2×10^3	1.6×10^4	[7.28, 7.29, 7.53]
Fish	203	2.0×10^3	6.6×10^3	5.9×10^2	4.8	4.9×10^1	3.7×10^4	[7.28, 7.29, 7.49, 7.53, 7.54, 7.62]
Benthic feeding	90	1.6×10^3	4.4×10^3	5.7×10^2	4.2	6.3×10^1	1.9×10^4	[7.28, 7.29, 7.49, 7.53, 7.54]
Forage	18	7.6×10^3	1.2×10^4	4.2×10^3	3.0	1.3×10^2	2.6×10^4	[7.28, 7.29, 7.53, 7.62]
Piscivorous	95	1.3×10^3	6.7×10^3	2.6×10^2	6.1	4.9×10^1	3.7×10^4	[7.53, 7.54, 7.63]
Mollusc	147	1.2×10^5	5.2×10^4	1.1×10^5	1.5	1.7×10^3	1.7×10^5	[7.28, 7.29, 7.38, 7.54, 7.62, 7.64]
Bivalve	141	1.3×10^5	4.9×10^4	1.2×10^5	1.5	1.7×10^3	1.7×10^5	
Reptile	7	3.6×10^3	2.3×10^3	3.1×10^3	1.8	1.5×10^3	7.3×10^3	[7.61]
Vascular plant	31	2.0×10^3	1.5×10^3	1.6×10^3	2.0	5.5×10^2	4.6×10^3	[7.28, 7.29, 7.62]

Source: See Ref. [7.51].

Note: $CR_{wo\text{-}water}$ is the activity concentration in the whole organism (in Bq/kg, FW) divided by the activity concentration in the water (in Bq/kg, DW). AM — arithmetic mean; ASD — arithmetic standard deviation; CR — concentration ratio; GM — geometric mean; GSD — geometric standard deviation.

[a] —: data not available.

typically found at higher levels than ^{210}Pb. Since ^{210}Po concentrations in soft tissue are normally higher than those in bones and comprise most of the edible tissues of freshwater species, the data in Table 7.3 can also be used to assess ^{210}Po concentrations in human foodstuffs. For polonium, the ratio of whole body to tissue is expected to be approximately 1.1 for freshwater fish and 2 for mammals [7.50]. However, there is a paucity of data for most species. The use of CRs in Table 7.3 represents a cautious approach to estimating ^{210}Po in edible tissues when sufficient data are unavailable. Additional reasons supporting the use of ^{210}Po CRs for assessment of human foodstuff contamination include the following [7.67]:

(a) For aquatic ecosystems, the whole organism $CR_{\text{wo-media}}$ values for bivalve molluscs, large crustaceans and marine gastropods do not include the shell. This is consistent with commonly used dosimetric approaches.
(b) For vertebrate wildlife groups, whole organism $CR_{\text{wo-media}}$ values typically do not include the gastrointestinal tract contents, although there may be some exceptions, such as when animals have been monitored live and in the case of small fish.

Table 7.3 also provides CRs for ^{210}Pb, since it serves as a source of ^{210}Po in wildlife. Such information is also often valuable for modelling purposes. The data in Table 7.3 clearly show that ^{210}Po CRs tend to be one to two orders of magnitude greater than those for ^{210}Pb, thus demonstrating higher mobility in freshwater ecosystems. The highest ^{210}Po CRs are in bivalve molluscs, followed by crustaceans and some foraging species, indicating the sensitivity of some freshwater organisms to contamination by polonium.

In summary, ^{210}Po in groundwater occurs over a wide range of concentrations. Once the differences due to release from aquifer minerals of ^{222}Rn by recoil are considered, there are very large differences due to adsorption of ^{210}Po onto surfaces. Bulk adsorption coefficients are difficult to predict because of wide variations in grain size and mineralogy over even very small spatial scales. Therefore, while earlier studies can provide some guidance on what might be expected, each aquifer needs to be characterized individually. In order to do so, accompanying data on ^{222}Rn are valuable for characterizing adsorption. Limited data also suggest that the concentrations of ^{210}Po on particles and colloids can be important under some circumstances. Future studies on the chemistry of polonium in dynamic groundwater regions, such as oxic and anoxic transitions and areas of salt water intrusion, are necessary. The behaviour of polonium within the vadose zone, which is most vulnerable to anthropogenic inputs, is also poorly understood.

The levels of ^{210}Po in surface water in freshwater systems are influenced by in situ decay, surface runoff, fluvial inputs and atmospheric deposition. Within a standing water body, ^{210}Po is removed through the settling of particulates, and its behaviour in the sediment is influenced by the cycling of iron and manganese as the redox conditions change in the sediment. This is also an important process if there are seasonal anoxic zones within the water body. More research is still needed on the partitioning of ^{210}Po onto particulates in the surface water column to fully understand the cycling of ^{210}Po and how it relates to manganese, iron and sulphur. A more comprehensive database of information needs to be developed to better understand the dynamics of ^{210}Po in the freshwater food web.

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Chapter 8

^{210}Pb AND ^{210}Po IN MARINE SYSTEMS

8.1. INTRODUCTION TO THE MARINE ENVIRONMENT

Two thirds of the planet is covered with sea water. Near the continents (neritic province), waters located between the shoreline and the shelf break, or continental margin, are called seas and are fringes of the immense blue waters beyond continental margins that make up the oceans (oceanic province). Oceans spread over all latitudes and longitudes, and their depths range from the shoreline to about 11 000 m in a few hadal trenches. The average depth of the North Atlantic, for example, is about 4500 m.

Despite the common chemical composition of sea water, oceans are by no means a uniform system. The combination of water temperature, salinity, sunlight and dissolved nutrients, such as chloride, sulphate, nitrate and other salts, creates different biotopes that are home to many species in marine ecosystems.

Biota species and biomass are unevenly distributed in the oceans. On the sea-floor, the biomass of benthic organisms decreases from the coastal waters to abyssal and hadal regions. In the pelagic domain, biomass over the continental shelf is much higher than in the open ocean. In specific areas of the continental margin, the upwelling of nutrient loaded deep water enhances phytoplankton primary productivity and contributes to the abundance of fishing resources.

8.2. POLONIUM SOURCES IN THE MARINE ENVIRONMENT

In sea water, polonium is present as a naturally occurring radionuclide and it is a companion of many other essential and non-essential dissolved elements. Polonium enters the marine environment from atmospheric deposition of ^{210}Pb and ^{210}Po at the ocean surface, from the in situ radioactive decay of ^{226}Ra dissolved in sea water, from the decay of ^{222}Rn gas exhaled from the sea-floor, and from river and anthropogenic discharges. Dissolved polonium ions rapidly adsorb onto suspended particles and are accumulated by marine organisms. The downward movement of those particles and planktonic faecal pellets leads to the depletion of polonium in surface waters, particularly in areas of enhanced primary and secondary productivity.

The atmospheric depositions of ^{210}Po originates from the decay of atmospheric radon, exhaled predominantly from landmasses and from resuspended dust. Most of the ^{210}Po measurements in surface air and in atmospheric depositions were made simultaneously for ^{210}Pb and ^{210}Po, and conclude that ^{210}Po concentrations in aerosols are minor and usually about 0.1 times ^{210}Pb concentrations (see Chapter 5). Average concentrations of ^{210}Pb and ^{210}Po in the air, as well as atmospheric deposition fluxes, vary with latitude and rainfall and are largely controlled by radon, exhalation and atmospheric concentrations, which are dependent on land mass distributions, and are modulated by global atmospheric circulation (see Chapter 5). Although direct measurements of ^{210}Pb and ^{210}Po deposited at the ocean surface have not been made, ^{210}Pb transport by global atmospheric circulation has been modelled, and ^{210}Pb deposition at the ocean surface has been estimated for specific areas to a reasonable degree of accuracy [8.1–8.3].

Ingrowth of ^{210}Po from ^{210}Pb–^{210}Bi radioactive decay in sea water takes place everywhere in the ocean and depends on ^{222}Rn concentration in sea water, which in turn is the sum of ^{222}Rn from in situ decay of dissolved ^{226}Ra and ^{222}Rn diffusing from the sea-floor following ^{226}Ra decay in sediments. The resulting ^{210}Po concentrations in sea water also greatly depend on ^{210}Po binding to suspended particulate matter and particle scavenging in the water column.

Polonium-210 also enters coastal areas with river discharges which feed the seas with chemical elements, including radionuclides of the natural radioactive series, in dissolved and solid phases. Owing to the low water solubility of polonium and its high partitioning into the solid phase (K_d), most polonium discharged by rivers is associated with suspended particulate matter and bottom sediment discharges [8.4].

The most relevant, direct ^{210}Pb and ^{210}Po anthropogenic discharges into the sea relate to non-nuclear industries, such as phosphate ore processing, oil and gas exploitation, and heavy mineral sands mining [8.5]. The better investigated case studies relate to phosphate ore processing for phosphoric acid and phosphate fertilizer manufacturing. Research conducted in several countries has shown that naturally occurring concentrations of ^{210}Po in estuaries and coastal waters receiving such phosphatic discharges were locally enhanced by factors in the range of 10–1000 [8.2, 8.6–8.10]. Often neglected but locally important, past radioactive waste dumping in the sea contained non-negligible amounts of ^{210}Po and ^{226}Ra [8.11, 8.12].

8.3. POLONIUM IN THE OCEANS

Polonium concentrations in dissolved and particulate phases have been reported for coastal waters and for the open ocean (see Refs [8.13–8.15]). Generally, polonium in coastal waters is largely associated with suspended particulate matter, with a minor fraction in the dissolved phase (<35%), probably due to the higher suspended particle loads and more intensive mixing than in open ocean waters, where polonium is mostly in the dissolved phase (see Table 8.1). In coastal sea water, ^{210}Pb is also mainly associated with suspended matter, although to a lesser extent than ^{210}Po. Neither concentration shows significant seasonal variation; thus, atmospheric depositions do not seem substantially to modify their concentrations in sea water throughout the year.

In coastal waters, concentrations of dissolved ^{210}Po are around 0.5 Bq/m^3, and usually the Po:Pb ratio is near one in the dissolved phase. In suspended particulate matter, however, ^{210}Po concentrations are generally higher than ^{210}Pb. Partitioning coefficients (K_d) for ^{210}Pb and ^{210}Po between suspended particulate matter and water (soluble fraction) have been reported for a few environments, and indicate high K_d values for ^{210}Po in the range of (2–7) × 10^5, which are on average five times higher than the K_d values for ^{210}Pb. The strong partitioning of these two radionuclides into the particulate phase, higher for

TABLE 8.1. PARTITIONING COEFFICIENT (K_d) OF ^{210}Pb AND ^{210}Po IN THE MARINE ENVIRONMENT BASED ON IN SITU DETERMINATIONS

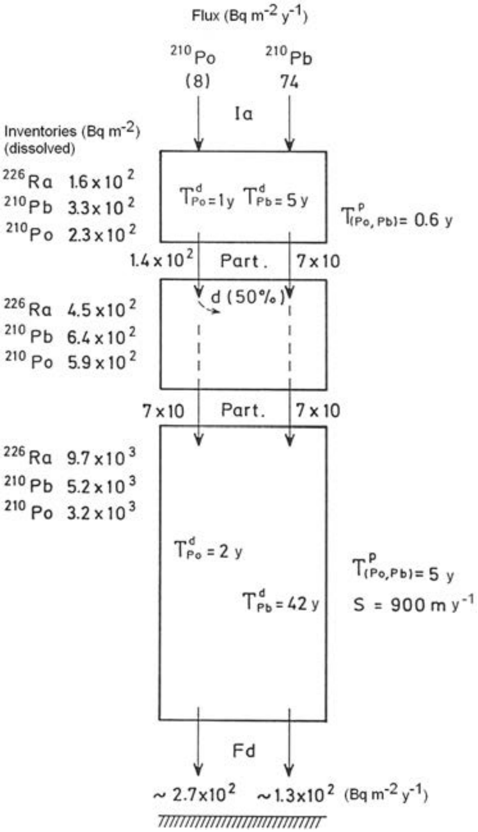
Region	Dissolved (Bq/m^3)	Particulate (Bq/m^3)	% dissolved	K_d	Ref.
^{210}Pb					
NW Mediterranean Sea, off Spain	1.27 ± 0.31	0.049 ± 0.011	96	(1.0 ± 0.57) × 10^5	[8.16]
NW Mediterranean Sea, off Monaco	1.62 ± 0.07	0.27 ± 0.02	86	(6.4 ± 0.5) × 10^4	[8.17]
NE Atlantic Ocean, Portuguese surf zone	0.54 ± 0.28	1.11 ± 0.73	33	(1.7 ± 1.0) × 10^5	[8.15]
^{210}Po					
NW Mediterranean Sea, off Spain	1.09 ± 0.37	0.27 ± 0.083	79	(7.8 ± 5.6) × 10^5	[8.16]
NW Mediterranean Sea, off Monaco	0.62 ± 0.03	0.25 ± 0.03	71	(1.6 ± 0.2) × 10^5	[8.17]
NE Atlantic Ocean, Portuguese surf zone	0.57 ± 0.13	1.87 ± 0.73	23	(2.8 ± 0.9) × 10^5	[8.15]
NE Atlantic Ocean, French surf zone	0.47 ± 0.21	0.94 ± 0.37	33	(1.2 ± 0.62) × 10^5	[8.13]

^{210}Po than ^{210}Pb, contributes to shaping the distribution of ^{210}Po in the marine environment.

In the open ocean, the water column is stratified and vertical mixing is reduced and mostly occurs in the epipelagic layer. Water stratification allows for the formation of gradients of dissolved salts and relatively stable vertical profiles of their concentrations. In the oceanic water column, ^{210}Po is produced from radon decay in the atmosphere, the decay of dissolved radon and radon exhaled from the ocean floor. Radon concentrations in the water column are lowest in the surface layer and greatest in the deep-sea water layer. Radon distribution mainly depends on diffusion in the water column and marine currents.

The isotopes ^{210}Pb and ^{210}Po are also introduced to the ocean from atmospheric depositions, and can be detected in the upper layer of the ocean, where a measurable excess of ^{210}Pb and ^{210}Po over dissolved ^{226}Ra concentrations are reported [8.16, 8.18]. Adsorption of ^{210}Pb and ^{210}Po onto particulates, followed by uptake by phytoplankton and zooplankton, allows for the removal of these radionuclides from the epipelagic layer with the downward flux of biogenic particles. Some of the particles falling from the upper oceanic layer are consumed in intermediate ocean layers and the chemical elements are recycled, contributing to supporting the abundant marine life in the oceanic mesopelagic layer. Particles, in their settling path through the deeper ocean layers, may further adsorb and remove soluble ^{210}Pb and ^{210}Po from the water column, creating an imbalance between ^{226}Ra (not adsorbed onto particles) and its particle reactive daughters, and thus prevent the formation of secular radioactive equilibrium. As a consequence of this radionuclide scavenging in the deep-sea layers, ^{210}Pb and ^{210}Po are deficient in comparison to ^{226}Ra activity concentrations in all oceans [8.19–8.22]. Although very fine particles (Stokes particles, such as clay particles) may settle with very low velocities of about 900 m/a, some of the biogenic particulate materials generated in the upper layer of the ocean, especially larger faecal pellets and carcasses of zooplankton organisms, sink rapidly and may reach the abyssal sea-floor in days or weeks, adding ^{210}Pb and ^{210}Po to the top sediment layer (see Fig. 8.1) [8.16, 8.23–8.25].

Figure 8.2 displays the vertical profiles of ^{210}Po along with those of ^{210}Pb and ^{226}Ra in the north-eastern Atlantic Ocean water column. Similar profiles have also been reported for other oceans [8.17, 8.26, 8.27]. While the concentration of dissolved ^{210}Pb can reach around 2.5 Bq/m^3 in the upper layer of the ocean, dissolved ^{210}Pb in deeper layers barely reaches 1 Bq/m^3. Dissolved ^{210}Po in the upper layer is around 1 Bq/m^3; it increases in the mesopelagic layer and decreases again, and remains at concentrations much lower than those of ^{210}Pb all the way down to the abyssal sea-floor. In particulate matter, the ^{210}Po:^{210}Pb ratio is generally consistently greater than 1.

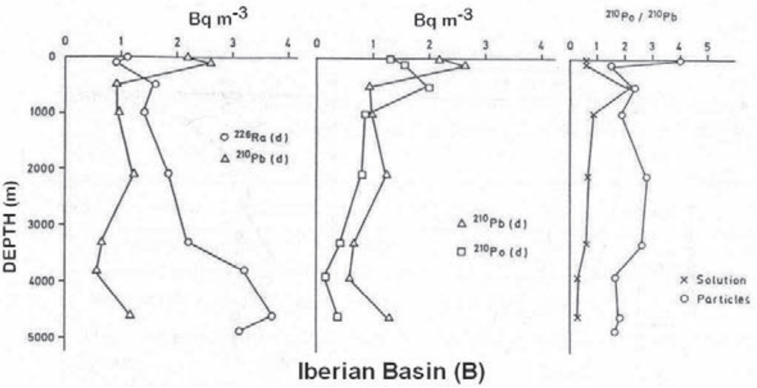


Source: Figure 2 of Ref. [8.23].

Note: d — dissolved phase; Fd — deposition flux on the sea-floor; Ia — atmospheric input of ^{210}Pb (Bq·m^{-2}·a^{-1}); T — mean residence time (years); p — particulate phase; Part. — radionuclide flux associated to the particle flux (Bq·m^{-2}·a^{-1}).

FIG. 8.1. Box model of the water column in the north-eastern Atlantic Ocean, with inventories and mean residence times of ^{210}Pb and ^{210}Po, and deposition fluxes at the sea-floor.

The mean residence time of ^{210}Po in ocean water layers was calculated for several oceans and values are about 0.5–1 years in the upper layer and somewhat longer, around 2 years, in the soluble phase of deep-sea layers [8.16–8.18, 8.23, 8.25]. Accumulation of ^{210}Po deposited onto the oceanic bottom sediments has generally not been measured, as all available reports focus on ^{210}Pb measurements [8.15]. The ^{210}Po flux arriving at the abyssal sea-floor in the north-eastern Atlantic Ocean was estimated as twice that of the ^{210}Pb flux and, in turn, the settling ^{210}Pb flux at the abyssal sea-floor was computed as twice that of the ^{210}Pb atmospheric flux entering at the surface of the ocean. Furthermore,



Source: Based on Ref. [8.23].

FIG. 8.2. Profiles of ^{226}Ra, ^{210}Pb and ^{210}Po in the water column of the north-eastern Atlantic Ocean.

while the ^{210}Po:^{210}Pb ratio in atmospheric particle depositions at the surface of the ocean is around 0.1, in the sediment depositions onto the abyssal sea-floor of the north-eastern Atlantic Ocean, the ^{210}Po:^{210}Pb ratio is around 2 (see Fig. 8.1).

8.4. ACCUMULATION AND TURNOVER OF ^{210}Pb AND ^{210}Po IN MARINE ORGANISMS

Carvalho [8.28] reports that:

> “Polonium (^{210}Po) and radioactive lead (^{210}Pb) in marine organisms have attracted the attention of scientists because of their relatively high concentrations in comparison with those in terrestrial organisms and also because ^{210}Po concentrations in marine biota often are much enhanced in comparison to those of ^{210}Pb, its radioactive grandparent [8.29, 8.30]. The first determinations of ^{210}Po in marine biota had shown high concentrations of largely unsupported ^{210}Po [i.e. above the ^{210}Pb concentration] in plankton, 110 Bq·kg^{-1} (dry weight), and soon other reports added information on unsupported ^{210}Po in marine fish, crustacean and whales [Refs [8.29, 8.31–8.33]]. Early measurements did record also high ^{210}Po concentrations in some marine species, such as the lanternfish, and in certain internal organs such as the pyloric *caecca* of tuna fish [Refs [8.34, 8.35]]. Pioneer data were reviewed by Cherry and Shannon [Ref. [8.29]] who highlighted the importance of ^{210}Po as a major

> internal source of the radiation dose received in the tissues of marine organisms.
>
> “Further work revealed the distribution of ^{210}Po in crustacean tissues, including the euphausiid *Meganicthyphanes norvegica*, and the role of zooplankton in the removal of ^{210}Po from the ocean surface layer [Refs [8.36–8.38]].”

Early work on polonium distribution in marine crustaceans highlighted elevated concentrations in the hepatopancreas and suggested that food could be the main source of polonium in invertebrates and the release of faecal pellets its main excretion route [8.36, 8.39–8.43].

The understanding of accumulation mechanisms and the biokinetics of many radionuclides in marine biota has been the goal of extensive experimental work carried out over decades in many radioecology laboratories. However, in contrast to, for example, fission products and transuranium elements, not much experimental work has been performed on the biokinetics of polonium in marine species and aquatic species in general (see the early works in Refs [8.44, 8.45]). This is understandable because, as a pure alpha emitter, polonium is not easily amenable to measurement by direct spectrometric techniques and even its measurement by total alpha counting is not possible without some type of relatively expensive and time consuming method. This difficulty was likely the main reason for the long persistent gap in knowledge on polonium uptake pathways, metabolism and turnover rates in marine biota, leaving polonium biokinetics open to guess work or assumptions by inference from indirect evidence.

The development of alpha spectrometry equipment and the production of artificial polonium isotopes allowed the use of double tracer techniques, involving three polonium isotopes, to investigate the role of the food ingestion pathway (food labelled with ^{208}Po) and sea water (labelled with naturally occurring and added ^{210}Po), and, using ^{209}Po as an isotopic tracer, for determination of radiochemical yield and internal analytical quality control of every sample [8.46].

Using such a double tracer technique, it is possible to quantify food and water as polonium sources, as well as polonium elimination by excretion and radioactive decay. This allows a mathematical description of the kinetics of polonium uptake and elimination in marine organisms. Carvalho and Fowler [8.46] model the polonium biokinetics using the following mass balance equation:

$$\frac{\mathrm{d}Q_t}{\mathrm{d}t} = I_\mathrm{w} W C_\mathrm{w} + AFC_\mathrm{f} - (k+\lambda)Q_t \tag{8.1}$$

where

Q_t is the activity of polonium in the organism at time t (Bq);

I_w is the uptake rate constant fom water (mL/g organism d^{-1}) by all processes, namely drinking and surface adsorption;

W is the weight of the organism (g);

C_w is the polonium concentration in water (Bq/mL);

A is the digestive assimilation efficiency or absorption efficiency of polonium from food;

F is the amount of food ingested by an organism per day (g/d);

C_f is the concentration of polonium in food (mBq/g);

K is the elimination rate constant of polonium (d^{-1}) from the organism;

and λ is the radioactive disintegration rate constant of ^{210}Po ($\lambda_{210\mathrm{Po}} = 0.005\,01\ \mathrm{d}^{-1}$); $\lambda_{208\mathrm{Po}} = 0.000\,656\ \mathrm{d}^{-1}$). Each term in this equation can be considered separately for assessing and describing the kinetics of polonium accumulation in the organism from water and food only.

Expressed in terms of activity concentration, the polonium concentration in an organism in steady state equilibrium of exchange with the environment (C_ss) is described by [8.46]:

$$C_\mathrm{ss} = \left(\frac{A \cdot C_\mathrm{f}}{(k+\lambda)}\right)\frac{F}{W} \tag{8.2}$$

In a set of laboratory experiments using shrimp, prawn and fish, Carvalho and Fowler [8.47] show that polonium absorbed in the internal tissues of biota is almost exclusively taken up from the ingested food and absorbed through the digestive pathway.

In shrimp, it was experimentally verified that, in spite of the presence of dissolved ^{210}Po in sea water, there was only minor absorption of ^{210}Po ions from water. Furthermore, this minor absorption did not take place through gill epithelium, as it does for many other ionic elements, but rather the tiny amounts of ^{210}Po absorbed from sea water were due to the drinking water reflex of marine organisms for osmotic balance, and so this polonium was absorbed through the gut wall in the same manner as polonium from food [8.46, 8.47].

Furthermore, it was demonstrated that crustaceans, such as shrimp and prawn, in sea water containing dissolved ^{210}Pb and ^{210}Po, easily accumulated ^{210}Po from ingested food into their internal organs, while ^{210}Pb absorption from food was minor. In addition, dissolved ^{210}Pb in sea water was adsorbed onto exposed surfaces, such as the exoskeleton and gills, without significant absorption into internal organs. This differentiated uptake, facilitating gut absorption of food-borne ^{210}Po in comparison to food-borne ^{210}Pb, explains the increased Po:Pb ratio (>1) systematically found in internal tissues of marine biota.

Moreover, Carvalho and Fowler [8.46] show that the ingestion pathway accounted for over 97% of ^{210}Po intake in shrimp, and for over 99% in fish internal tissues. The absorption of polonium from ^{208}Po labelled food closely matches the gut absorption efficiency of protein from diet in prawn and fish. Once absorbed, this ^{208}Po is rapidly distributed in internal tissues according to the distribution pattern of naturally occurring ^{210}Po. Further experiments show that polonium absorbed though the digestive pathway and accumulated in the liver of teleost fish binds to proteins, such as ferritin and metallothioneins, that strongly bind ^{210}Po and are able to retain significant amounts of this radioelement in internal tissues [8.48].

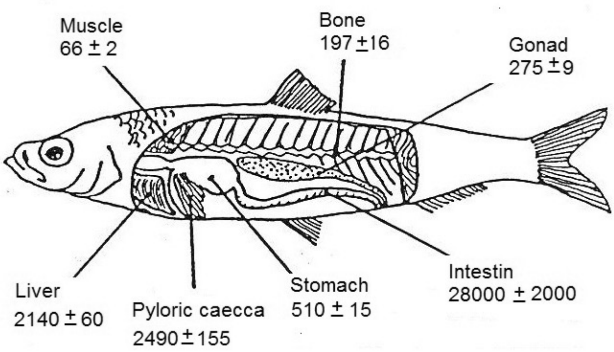
Experiments carried out with phytoplankton cells verify their rapid uptake of ^{210}Po from liquid culture media. Using these polonium labelled cells to feed mussels, it can be shown that ^{210}Po is rapidly absorbed and accumulates in the soft tissue of mussels. It can be concluded that filter feeding molluscs, such as mussels, in spite of filtering large volumes of sea water containing dissolved ^{210}Po, accumulate ^{210}Po in internal tissues mainly from ingested food [8.49].

Experiments using the same polonium double tracer technique also show that ^{210}Po dissolved in the liquid media of microalgal cultures can be rapidly adsorbed onto and absorbed into phytoplankton cells. In addition, through ingestion of these cells, ^{210}Po was transferred to herbivorous (planktivorous) zooplankton [8.50–8.52].

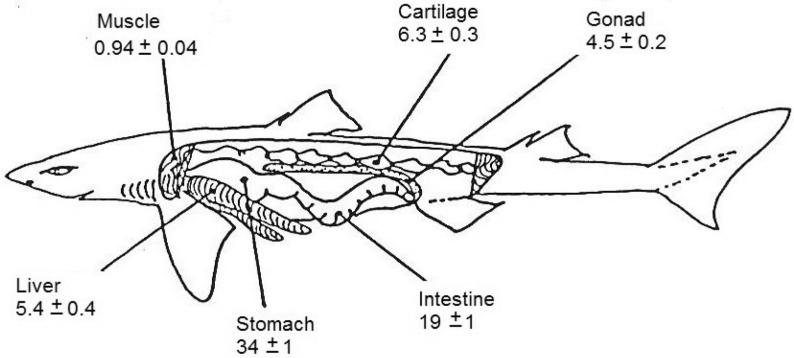
From kinetic studies on ^{210}Pb and ^{210}Po in marine shrimp, Carvalho and Fowler [8.47] conclude that ^{210}Po has a biological half-life of 10–11 days in whole body shrimp ($T_{1/2}$ = 7 days in the hepatopancreas; $T_{1/2}$ = 28 days in muscle). At 4–11 days, ^{210}Pb has a shorter half-life in whole body shrimp and a much shorter half-life in internal organs ($T_{1/2}$ = 4 days in the hepatopancreas; $T_{1/2}$ = 2 days in muscle), which may further contribute to higher ^{210}Po:^{210}Pb ratios in marine biota.

The distribution of polonium in internal organs and tissues has been reported for many marine organisms, including molluscs, crustaceans, fish and mammals. Higher ^{210}Po concentrations are systematically found in the liver and digestive tract, and lower ones in muscle tissue. For example, ^{210}Po concentrations in fish tissues vary enormously from species to species (see Fig. 8.3), but there is a general pattern of distribution: lower concentrations are consistently measured in muscle tissue and higher concentrations are found in the gut and liver, mostly in relation to the digestive organs (see Section 8.5.2).

This distribution pattern is common to teleost (bony fish) and elasmobranchs (cartilaginous fish), and cartilaginous fish generally display lower ^{210}Po concentrations than teleost fish — most probably on account of



Sardina pilchardus



Scyliorhinus canicula

Source: See Ref. [8.53].

FIG. 8.3. Polonium-210 in fish organs of a teleost fish (above) and a cartilagineous fish (below).

their physiology and the biochemical composition of their tissues [8.54, 8.55]. In several species of teleost fish and marine mammals, ^{210}Po variations in the muscle tissue were correlated with red muscle content [8.56]. As red muscle has a higher myoglobin content, this association with ^{210}Po was considered a likely biochemical association.

High ^{210}Po concentrations were reported in 1982 for the hepatopancreas of some mid-water pelagic shrimp, giving rise to high absorbed radiation doses [8.57]. Surprisingly high ^{210}Po concentrations were also measured in the gonads of several fish species [8.54]. This was assumed to relate to the accumulation of proteins in the egg, but high ^{210}Po in eggs may expose the genetic material to relatively high radiation doses. For example, in the case of the common sardine, Carvalho [8.54] estimate that effective absorbed dose from ^{210}Po could reach 170 mSv/a in the gonads and even higher in some other tissues. It has been suggested that this could merit further investigation, as this dose might have biological effects at a genetic level and might play a role in the genetic variability of organisms.

The search for genotoxic effects (micronuclei and DNA strand breaks) induced by ^{210}Po was undertaken in mussels from the Brazilian coast, with naturally occurring ^{210}Po concentrations averaging 155.6 ± 7.9 Bq/kg (FW) in their soft tissue. At this level of ^{210}Po concentrations, the internal dose rate was 0.02 mGy/d (7.3 mGy/a) and no genotoxic effects were observed. However, this dose rate was lower than the suggested dose threshold for inducing effects — 10 mGy/d [8.58]. High ^{210}Po concentrations are also reported in marine mammals at levels that are unusual by human standards (see Appendix III).

8.5. POLONIUM IN MARINE BIOTA IN SEVERAL OCEAN REGIONS

The ^{210}Pb and ^{210}Po activity concentrations reported in marine biota are spread over several orders of magnitude. The radionuclide levels do not relate to seawater depths, ecosystems or geographical locations [8.28]. This section reviews ^{210}Po data pertinent to several ocean regions and many taxonomic groups, with comments on ecology and trophic relationships in order to aid data interpretation.

8.5.1. Intertidal zone

Carvalho [8.28] reports that:

> "The intertidal rocky shore and also soft bottom areas (e.g., in estuaries) of the Atlantic Ocean and of the Mediterranean Sea are zones of dense biological habitats with high biomass and typical populations such as those of seaweeds, mussel and oyster beds, and characteristic fish species (e.g., Blenniidae). Similar dense populations may be found on the rocky shores of most temperate regions [Refs [8.59–8.61]]."

In the intertidal zone, ^{210}Po accumulated in macrophytic algae comes from ^{210}Po dissolved in sea water. The first likely uptake mechanism is surface adsorption by simple contact. Adsorption of dissolved radionuclides onto the surface of algal blades by diffusion mechanisms across the blade surface boundary layer also apply to transuranic elements, such as plutonium and americium, with similar ‘piston velocities’, and it is not considered an active biological uptake mechanism (for further information, see the early work in Refs [8.62–8.64]).

Carvalho [8.28] reports that the ^{210}Po:^{210}Pb ratios in macrophytic algae are consistently higher than unity, often ranging from three to ten. This seems to be due to preferential adsorption of ^{210}Po onto the algal surface. Analyses of ^{210}Pb and ^{210}Po distribution in seaweed in the 1980s confirmed that most ^{210}Po is bound or adsorbed onto external surfaces [8.65]. Later work also confirmed passive adsorption of metals to microalgal cell walls and fast kinetics of ^{210}Po and ^{241}Pu sorption in marine phytoplankton and zooplankton cells [8.66–8.68].

Detailed studies report ^{210}Po concentrations in seaweed averaging 4.8 Bq/kg (FW) (range: 1.6–9.1 Bq/kg) in the coastal area of Portugal in the north-eastern Atlantic Ocean [8.28, 8.54]. Carvalho [8.28] reports that differences among algae classes are small, and *Fucus* sp. (Phaeophyceae), often used as a bioindicator species, is the seaweed with the highest ^{210}Po concentration (9.1 Bq/kg, FW). Concentrations of ^{210}Po in *Fucus vesiculosus* at Pirou, the French Channel, are 13.7 Bq/kg (FW) [8.13]. Concentrations in the range of 11–26 Bq/kg (FW) are reported for several seaweed species on the Tamil Nadu coast, India, in a tropical environment [8.69]. Since ^{210}Po accumulation in seaweed is operated by sorption mechanisms, those slight variations in ^{210}Po concentrations in seaweed might be due to the sorption characteristics of algal coatings. Furthermore, adsorption of radionuclides might also be affected by water temperature, water mixing and suspended particulate matter load, especially in the surf zone.

Polonium-210 accumulation in animal species is very different to that in seaweed. Different mollusc species living in the same intertidal rocky shore can display very different ^{210}Po concentrations. For example, Carvalho [8.28] finds the following concentrations in molluscs at the north-eastern Atlantic coast:

— Limpet (*Patella aspera*): A herbivore mollusc feeding on seaweed grasped from the rock surface — 11.6 Bq/kg (FW);
— Mediterranean mussel (*Mytilus galloprovincialis*): A filter feeding mussel whose diet is organic suspended particles — 132 Bq/kg (FW);
— Winkle (*Littorina littorea*): A carnivore gastropod — 283 Bq/kg (FW).

Although these molluscs all live in the same zone, ^{210}Po is concentrated in their soft tissue at different levels, which reflects that these species occupy distinct biotopes and trophic levels although they live together in the same zone [8.28]. Comparing and interpreting results for organisms from rocky shores is generally straightforward. However, this might not be the case for results from soft bottom organisms.

Intertidal, sediment dwelling bivalve molluscs which live in soft bottoms at the temperate north-eastern Atlantic coast, such as the clams *Tapes decussatus* and *Ensis siliqua*, exhibit dry weight concentrations of 760 Bq/kg and 225 Bq/kg, respectively. Another clam (*Anadara granosa*) from a similar biotope in the tropical environment of Mumbai Harbour, India, exhibited 313 Bq/kg (DW) and another burrowing bivalve (*Tonna dolium*) on the Tamil Nadu coast, India, exhibited 132 Bq/kg (FW) [8.69, 8.70]. Although living in similar environments, these different species might have different feeding strategies and diets, which would account for the differences. It should be recognized that results of radionuclide analysis in sediment burrowing molluscs may be affected by incidental parameters, such as the presence of sediment in the gut that is sometimes difficult to remove. While some researchers remove the gut sediment prior to analysis by dissection, washing or depuration, others do not. Thus without a common methodology in sample preparation, comparing results and interpretation can be problematic.

This difficulty is not present in every case, especially when studying organisms that do not ingest bulk sediment to process organic matter as food. Although all crab species inhabit sandy and muddy substrates, their feeding strategies involve food selection instead of ingestion of bulk sediment [8.71]. Extensive analysis of ^{210}Po in ten species of brachyuran crabs from the intertidal zone of the Gulf of Mannar, on the Indian coast of the Bay of Bengal, show that ^{210}Po concentration (FW) in crabs varies according to their food habits: it is low in herbivores, 45.9 Bq/kg; and significantly higher in carnivores, up to 221.2 Bq/kg.

Heyraud et al. [8.72] examine whether mussel samples from different latitudes might reflect the differences in atmospheric depositions of ^{210}Pb and ^{210}Po (see Refs [8.73–8.80] and Table 8.2.). As depositions in the southern hemisphere are up to ten times lower than those in the northern hemisphere (see Section 5.3), they hypothesize that ^{210}Pb and ^{210}Po in mussels might reflect that difference. Their results show that mussels from European and South African coasts display similar average radionuclide concentrations and no noticeable latitude effect can be identified. Furthermore, mussels from the coasts of Denmark and the United Kingdom with low seawater temperature and strong seasonal temperature fluctuation, when analysed for ^{210}Pb and ^{210}Po, display an average and range of ^{210}Po concentrations comparable to mussels from tropical zones on the coast of Brazil and from the coast of the Mediterranean Sea.

Charmasson et al. [8.73] compare ^{210}Pb and ^{210}Po accumulated in filter feeding mussels from intertidal areas of the Mediterranean coast and mussels from deep-sea hydrothermal vents of the Menez Gwen field, in the Mid-Atlantic

TABLE 8.2. ACTIVITY CONCENTRATIONS (AND RANGE) OF ^{210}Po AND ^{210}Pb IN MUSSEL (*Mytilus* spp.) SOFT TISSUES

Region	DW:FW	Concentration (Bq/kg, DW)		Po:Pb (range)	Ref.
		^{210}Po	^{210}Pb		
Adriatic Sea, Italy	0.16	112 (50–142)	—[a]	—[a]	[8.75]
Aegean Sea, Turkey	0.19	305 (52–1344)	28.0 (6–167)	12.1 (3.1–25.0)	[8.76]
English Channel					
Pirou, French coast	0.17	296	12.2	25.4	[8.13]
Irish Sea, Ireland	0.12	222 (80–468)	11.9 (4–25)	15.9	[8.14]
Mediterranean Sea					
Croatia	—[a]	54–460[b]	—[a]	—[a]	[8.77]
Toulon, France	0.17	203.1	41.3	8.6	[8.73]
NE Atlantic					
Portugal open coast	0.13	759 (460–1470)	45 (23–96)	17	[8.80]
(oligotrophic)	0.13	210	1	21	[8.80]
Portugal Tejo Estuary	—[a]	289–589	—[a]	—[a]	[8.74]
(mesotrophic)					
Ré Island, France					
North Sea					
United Kingdom	—[a]	104–3124	2.8–284	—[a]	[8.79]
South Atlantic					
Brazil	—[a]	1995	—[a]	—[a]	[8.78]
Brazil	—[a]	156[b] (54–460)	—[a]	—[a]	[8.57]
Cape of Good Hope, South Africa	—[a]	409 (170–1290)	5.7 (1.2–16.2)	86 (22–218)	[8.72]

[a] —: data not available.

[b] Fresh weight.

Ridge, the latter exposed to sulphide rich vent waters, and report similar concentrations despite all the environmental differences. Results for coastal clams and deep-sea clams from soft bottoms of the north-eastern Atlantic Ocean, both sediment burrowing, suspension feeding bivalves, also reveal similar ^{210}Pb and ^{210}Po concentrations [8.28]. These results suggest that, in spite of different environmental conditions in distinct ecosystems, similar organisms with similar feeding strategies and at the same trophic level in the food chain accumulate ^{210}Po up to comparable concentrations.

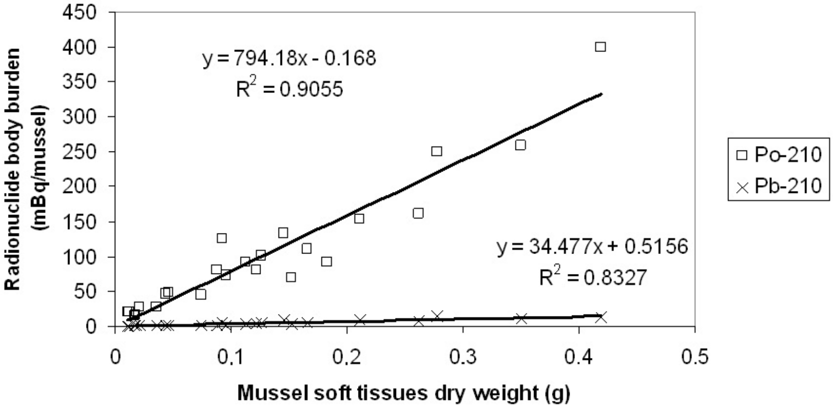
Coastal mussels, often of the family Mytillidae, are used worldwide as sentinel organisms to monitor contamination in coastal areas following the concept of the pilot International Mussel Watch programme of the United Nations Educational, Scientific and Cultural Organization to report ^{210}Po in mussels in many regions. In the Mediterranean Sea, for example, ^{210}Po has been determined in mussels from the coast to assess the radiological risk from radioactive atmospheric depositions (mainly ^{137}Cs) from the Chernobyl accident compared to ^{210}Po. It was concluded that, in spite of widespread deposition of ^{137}Cs in the Mediterranean Sea, the radiation dose to mussels and to mussel consumers would be a minor fraction of that from naturally occurring ^{210}Po. A similar finding was deteremined in 1995 by the then IAEA Marine Environment Laboratory, Morocco [8.81]. However, ^{210}Po concentrations for mussels also vary widely [8.82].

Mussels have also been used as sentinel organisms to monitor the impact of local discharges of radionuclides which include ^{210}Po, for example from phosphate industries. In some coastal areas subject to these effluents, ^{210}Po enhancement can be observed in mussels and other marine molluscs, while in other cases, surprisingly no ^{210}Po enhancement could be detected [8.8, 8.79]. The ^{210}Po concentrations in mussels in different regions around the world vary across a wide range of values, and results from specific areas with anthropogenic discharges often cannot be considered significantly different on a statistical basis (see Table 8.2). An examination of published regional ^{210}Po datasets reveals wide ranges of concentration in mussels from the same coastal region, collected at the same time of the year and by the same research team. For example, the concentrations of naturally occurring ^{210}Po in mussels from 27 locations around Ireland were in the range of 80–468 Bq/kg (DW) [8.14], which indicates that wide variations truly do exist among samples and are not artefacts. An interesting finding is that ^{210}Po concentrations in mussels from the most pristine areas are sometimes even higher than those in mussels from areas subject to anthropogenic discharges of ^{210}Po, thus rendering coastal monitoring results difficult to interpret (see Refs [8.13, 8.14, 8.83]).

This interest in the use of mussels as biomonitors led to a detailed investigation of ^{210}Pb and ^{210}Po bioaccumulation in mussels, the effects of environmental parameters and the physiological condition index of mussels on ^{210}Po concentration [8.84], and the assessment of the allometric effect on ^{210}Po concentration [8.85]. One important finding is that the fluctuation of ^{210}Po in mussel soft tissue throughout the year seems at first glance to be a consequence of the seasonal ^{210}Pb–Po atmospheric deposition flux. Instead, Carvalho et al. [8.85] verify that it is an effect of the seasonal fluctuation of the physiological condition of mussels. They report that during winter, when mussels have less fat, the ^{210}Po concentration per kilogram in the soft tissue is higher than in summer. In summer, when the accumulation of lipids and ripe gonads increase the physiological condition index of mussels, ^{210}Po activity concentration expressed on a mass basis exhibit lower values. However, when ^{210}Po concentration is expressed per mussel (^{210}Po body burden), ^{210}Po values are nearly stable over the year, and the main seasonal fluctuation is of lipid (fat) reserves, not of ^{210}Po [8.15]. Carvalho et al. [8.85] describe the allometric effect on ^{210}Po concentration as very pronounced in mussels and, for example, an increase in mussel shell length from 2.5 cm to 5.0 cm can correspond to a decrease of 50% in ^{210}Po concentration in mussel soft tissue (see Figs 8.4 and 8.5).

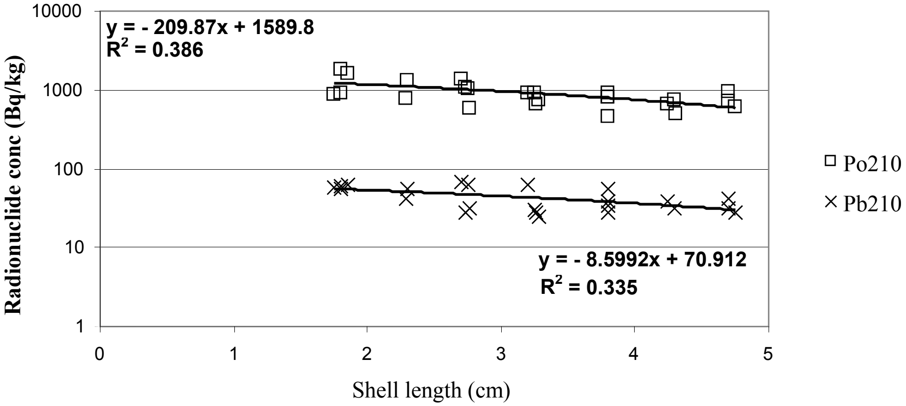
These findings on mussels, based on the determinations of ^{210}Pb and ^{210}Po, lead Carvalho et al. [8.85] to recommendations that can be applied to all contaminants and include the following:

(a) To select carefully mussels of the same size class to avoid allometric effects on ^{210}Po concentrations;
(b) To determine the physiological condition index of mussels to correct for the fat content;
(c) To take and pool a minimum number of individuals in each sample to minimize inter-individual variation;
(d) To allow for statistically valid comparison of results from different places.



Note: Based on the data presented in Ref. [8.85].

FIG. 8.4. Variation of ^{210}Pb and ^{210}Po body burden in mussel soft tissue as a function of shell length.



Note: Based on the data presented in Ref. [8.85].

FIG. 8.5. Allometry of ^{210}Pb and ^{210}Po concentrations in mussels as a function of shell length.

An improved sampling methodology can achieve comparable results in coastal monitoring using molluscs.

8.5.2. Coastal seas

Carvalho [8.28] reports that:

> “Amongst the large diversity of fish species in the waters above continental shelf, the Clupeidae (sardine, herring, and sprats), the Scombridae (mackerel) and Thunidae (tuna), have a special importance due to their abundance compared with other fish and to their major place in the human diet. While these are all pelagic fishes, other also very common fish species are demersal [i.e. live near the sea-floor and feed on benthic organisms], such as the sea hake (*Merlucius merlucius*), the pouting (*Trisopterus luscus*) and the small benthic sharks, such as the spotted catshark (*Scyliorhinus canicula*).”

With regard to ^{210}Po transfer from the marine environment to human populations, the consumption of seafood from coastal seas is the main pathway and therefore has received much attention in the assessment of radiation exposure (see Refs [8.70, 8.86–8.90]).

Fish species from many coastal regions around the world have been analysed, either on a whole body basis or in segregated organs and tissues, and the ^{210}Po concentrations are spread over a very wide range of values which has challenged interpretation (see Table 8.3). Attempts have been made to find an

TABLE 8.3. ACTIVITY CONCENTRATIONS OF ^{210}Po AND ^{210}Pb IN FISH TISSUE FROM COASTAL AREAS OF THE NORTH ATLANTIC

Species and region	Tissue	DW:FW	^{210}Po (Bq/kg)	^{210}Pb (Bq/kg)	Po:Pb
Sardine (*Sardina pilchardus*) Madeira Island, Portugal	Muscle	0.23	66	1.0	66
	Liver	0.28	2 140	6.2	345
	Gonad	0.27	275	1.8	156
	Bone	0.39	197	26	7.5
	Stomach	0.27	510	2.5	199
	Caecca	0.23	2 490	138	18
	Intestine	0.24	28 000	100	281
Chub mackerel (*Scomber japonicas*) Madeira Island, Portugal	Muscle	0.26	19	0.63	30
	Liver	0.56	1 035	7.3	141
	Gonad	0.25	183	22	8
	Bone	0.39	42	11	3.7
Blue jack mackerel (*Trachurus picturatus*) Madeira Island, Portugal	Muscle	0.28	8.9	0.7	13
	Liver	0.28	615	134	4.6
	Gonad	0.17	56	1.1	51
	Bone	0.38	43	15	2.8
White sea bream (*Diplodus sargus*) Lisbon, Portugal	Muscle	0.27	0.52	0.20	2.6
	Liver	0.31	28	14.5	1.9
	Gut	0.28	39	1.13	34
	Caecca	0.25	69	5.0	14
	Bone	0.53	25	31	0.8
Cod (*Gadus morhua*) Newfoundland, Canada	Muscle	0.19	0.628	0.040	16
	Liver	—[a]	6.59	0.08	81
	Gonad	—[a]	1.87	0.014	14
	Bone	—[a]	0.41	0.61	0.7
European hake (*Merlucius merlucius*) Sesimbra, Portugal	Muscle	0.19	6.4	0.15	43
	Liver	0.64	10.8	1.51	7
	Gonad	0.17	52.0	1.26	41
	Bone	0.30	7.8	0.77	10

Source: See Refs [8.28, 8.83].

[a] —: data not available.

explanation for ^{210}Po variation based on allometry (body size and metabolic rates of species), diet composition, water depth, latitude and other environmental parameters.

For example, Dahlgaard [8.83] attempts to relate ^{210}Po concentration levels in fish with variation of environmental parameters in the Baltic Sea. Average ^{210}Po concentrations (FW) in frozen fish fillets are 0.35 Bq/kg (cod), 0.65 Bq/kg (herring) and 0.96 Bq/kg (plaice). The differences in ^{210}Po between species are statistically significant. However, an attempt to find a correlation between ^{210}Po concentrations in fish and the salinity gradient, which in the Baltic–North Sea region is in the range of 8–35%, shows no statistically significant effect either on fish or on mussels.

A 1988 study of herring, cod and flounder in the Baltic Sea (Gulf of Gdansk, Poland) found that the highest concentrations were measured in the digestive organs, in particular the intestine [8.91]. Furthermore, the contribution of the digestive organs to the total ^{210}Po accumulated in the fish body was positively correlated with the degree of fullness of the stomach, while the ^{210}Po concentration in fish decreased if food was lacking in the stomach.

The ^{210}Po concentration (^{210}Po whole body burden or ^{210}Po concentration in muscle) can allow for ranking species into different ^{210}Po concentration levels. Heyraud and Cherry [8.40] tentatively interpret this as being dependent on the diet of each species. For example, ^{210}Po concentrations are much lower in herbivorous fish (e.g. white sea bream, *Diplodus sargus*) than in planktivorous fish (e.g. sardine, *Sardina pilchardus*) [8.28, 8.54]. Research was also published in 1989 on the effect of diet composition on ^{210}Po concentrations in fish, in an attempt to apply ^{210}Po as an indicator of diet, at least in clupeoid fish [8.92].

While some parameters play a role in constraining ^{210}Po concentrations, none is sufficient to provide a key explanation for such a wide range of concentrations determined in fish and other biota living in the same environment. A different approach, rooted in previous findings, has been proposed for interpreting ^{210}Po concentrations in marine biota on the basis of food chains in the ocean and the trophic position of organisms (see Refs [8.28, 8.46, 8.48, 8.54]). This interpretation is based on evidence provided by a comparison of ^{210}Po levels in species that are taxonomically related, often with very similar feeding strategies and especially occupying the same trophic level in the food chain [8.28].

One example is that the ^{210}Po concentration level in the muscle of planktivorous sardines (*Sardina pilchardus*) from the north-eastern Atlantic Ocean (see Table 8.3) is comparable to that in similar fish of the tropical sea environment off the Tamil Nadu coast, India, where ^{210}Po concentrations (FW) in planktivorous fish muscle are 190 Bq/kg for *Sardinella longiceps* and 116Bq/kg for *Eleuteronema tetradactylum* [8.28, 8.69]. Similarly high ^{210}Po concentrations are reported for the anchovy (*Engraulis encrasicolous*) in the Adriatic Sea, and for the anchovy and the sprat (*Sprattus sprattus*) in the Black Sea — all of which are planktivorous fish species that consistently exhibit ^{210}Po concentrations higher than all other fish from the same areas [8.75, 8.93].

Another example is the similar ^{210}Po concentration levels in mesopelagic fish species, such as the lantern fish and hatchet fish, from the Pacific Ocean and the North Atlantic Ocean, but with a similar diet based on small pelagic zooplankton crustaceans and occupying the same trophic level (see Refs [8.28, 8.54, 8.94]).

A last example, reported by Carvalho [8.28], is the ^{210}Po concentration in large predators of coastal seas, such as dolphins (*Delphinus delphis*), with average ^{210}Po concentrations of 49 Bq/kg in the dorsal muscle and 107 Bq/kg in the liver, which are generally higher than in fish [8.95, 8.96]. The diet of these marine mammals (Delphinidae) is exclusively based on seafood and they are at the top of food chains in coastal seas. Dolphins concentrate ^{210}Po exclusively from the diet (i.e. ^{210}Po transferred along the food chain) — there is practically no absorption from water. Interestingly, other Delphinidae from the tropical waters of Brazil in the southern hemisphere and from temperate waters off the Portuguese coast exhibit very similar ^{210}Po concentrations, reinforcing the absence of an influence from geographical and climatic regions and the key role of trophic level and diet on ^{210}Po accumulation (see Appendix III).

Some trophic relationships and ^{210}Po concentration levels are easy to interpret and correlate, such as ^{210}Po transfer from phytoplankton to herbivorous zooplankton, from plankton to planktivorous fish, or from fish to marine mammals, as described above; while others might be more challenging. For example, Hassona et al. [8.97] report ^{210}Po concentrations (FW) for coral reef fish species from the Red Sea, a tropical environment with complex food webs in the sublittoral zone, according to their general dietary habits:

- Carnivorous species: 1.8 Bq/kg (range: 0.25–6.42 Bq/kg);
- Herbivores: 2.2 Bq/kg (range: 1.52–3.80 Bq/kg);
- Omnivorous fish: 2.5 Bq/kg (range: 0.72–5.04 Bq/kg).

Although Hassona et al. [8.97] interpret ^{210}Po concentrations as a reflection of the dietary habit of the species, differences in ^{210}Po concentrations among groups are minor.

Eventually, and particularly in reef ecosystems, one aspect that might need refinement and further detailed work is the understanding and classification of the dietary habits of fish and a proper relationship with ^{210}Po accumulation. There are at least four types of alimentary canal in marine herbivorous fish which can be modelled using analogies with chemical reactors [8.98]. Herbivores can consume food of different qualities, with different digestion rates and different transit times; and they can process food in non-acidic to highly acidic stomachs, which can modify ^{210}Po uptake. Furthermore, carnivorous fish in coral reef ecosystems involuntarily ingest large amounts of coral fragments while capturing their prey. Studies have shown that sea-birds that find food in the sea exhibit ^{210}Po concentrations that reflect the food habits and ^{210}Po concentrations in their prey [8.99, 8.100].

8.5.3. Epipelagic zone

In the oceanic province, ^{210}Pb and ^{210}Po concentrations in the upper layer of the ocean are controlled mainly by atmospheric deposition and, to a much lesser extent, by the decay of ^{226}Ra dissolved in sea water [8.28]. A 1988 study found the average ^{210}Po concentration in this oceanic upper layer to be around 1 mBq/L [8.22]. The upper layer of the ocean, or epipelagic zone, contains a very significant biomass of phytoplankton and zooplankton [8.59, 8.61]. Phytoplankton exhibits ^{210}Po concentration factors of around 10^4–10^6 and Po:Pb ratios of around 3, which are lower than those observed in some zooplankton groups (see Refs [8.101–8.103]).

Investigations made of ^{210}Po accumulation in zooplankton report a high concentration in euphausiids (krill) and that ^{210}Po concentration increases from phytoplankton to macrozooplankton to mesozoopklankton [8.39, 8.54, 8.102]. The concentrations of ^{210}Pb and ^{210}Po in zooplankton faecal pellets and their role in the rapid removal of these radionuclides from the epipelagic zone have triggered research on the vertical transport of elements carried by relatively large biogenic particles in the water column (see Refs [8.24, 8.26, 8.27, 8.36–8.38]). However, studies on plankton always face the difficulty that plankton samples obtained with towed nets are based on the size of organisms, being a mixture of species rather than plankton organisms sorted by phylogenetic group, ecological niche or trophic level. Organism size is not an indication of trophic level. Furthermore, even small planktonic organisms have diverse strategies of food selection and may occupy different trophic levels regardless of their similar size [8.104].

There are data obtained by analysing plankton sorted by species or genus rather than by analysing bulk plankton samples. Results on ^{210}Po based on species separation has increased the understanding of ^{210}Po accumulation from sea water to phytoplankton to mesozooplankton to krill (*Eupasia superba*) in the Antarctic Ocean [8.105]. Furthermore, a comparison of ^{210}Po datasets from Antarctica, the Mediterranean and the north-eastern Atlantic Ocean reveals that the range of ^{210}Po concentrations are similar and unrelated to seawater temperature or geographical region (see Refs [8.54, 8.101, 8.102, 8.105]).

Reporting on the north-eastern Atlantic Ocean, Carvalho [8.28] finds that:

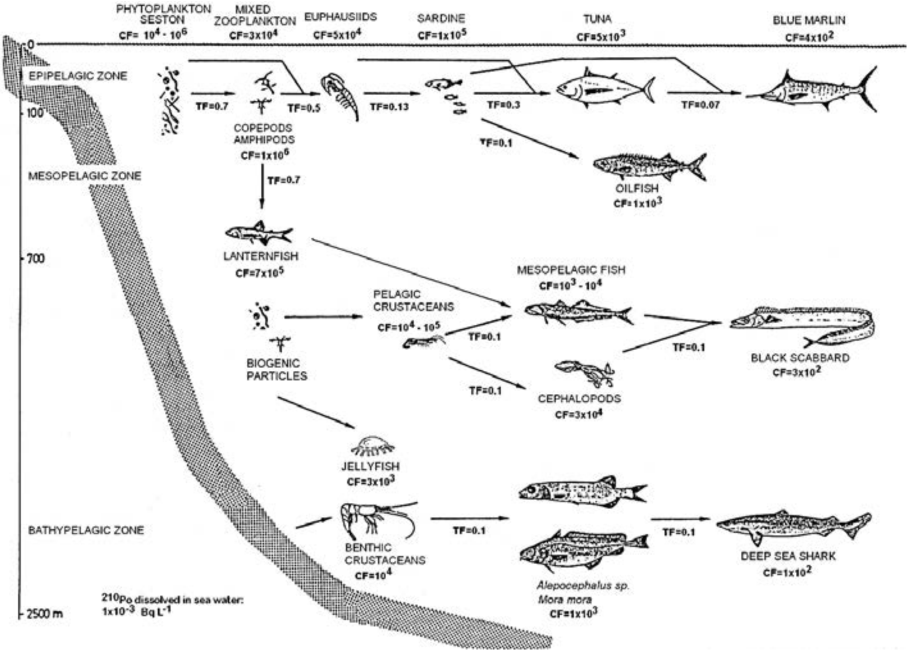
> “The groups of small zooplanktonic crustaceans such as ostracods, amphipods, copepods, euphausiids and mysids, displayed the highest ^{210}Po activity concentrations in marine organisms ranging from 84 $Bq\cdot kg^{-1}$ in euphausiids [generally carnivores] to 2900 $Bq\cdot kg^{-1}$ in amphipods [mostly planktivorous herbivores]. Other important zooplanktonic groups, such as the chaetognaths (*Sagitta* spp.), pteropods (*Creseis* spp., *Diacria trispinosa*) and pelagic polychaetes contained ^{210}Po concentrations in that range and Po:Pb ratios also higher than jellyfish, generally between 5 and 10 and occasionally higher.
>
> “Planktonic fish larvae (Mycthopidae, Alepochephalidae) and cephalopod larvae (Loligonodae, Ommastrephidae) displayed ^{210}Po concentrations similar to other zooplankton taxa as reported above, but with Po:Pb around 6 and not different from their adult fish stages while this ratio went up to 157 in crustaceans (mysids).”

Carvalho [8.28] reports that large predator teleosts of the north-eastern Atlantic Ocean epipelagic zone, such as tuna (*Thynnus obesus* and other tuna species), feed upon clupeoid fish, squid and zooplankton, while the blue marlin (*Makaira nigricans*) feeds upon clupeoid fish, and the oil-fish (*Ruvettus pretiosus*) eats several pelagic fish species (see Fig. 8.6). Polonium-210 concentration in tuna muscle is 3.0 ± 0.1 Bq/kg, with a Po:Pb ratio of 6.6, in blue marlin muscle 0.4 ± 0.2 Bq/kg and in oil-fish 0.7 ± 0.02 Bq/kg, thus also much lower than in tuna.

Carvalho [8.28] finds that:

> “Other oceanic large predators on the top of food chains, such as the sperm whale (*Physeter catodon*) feeding upon deep water cephalopods, contained ^{210}Po and ^{210}Pb concentrations in muscle tissue roughly comparable to tuna.”

One interesting question is raised by ^{210}Po concentrations in the same or closely related species, occupying the same trophic level in different oceans. For example, ^{210}Po concentrations in tuna muscle from the Atlantic Ocean, the Pacific Ocean and the Mediterranean Sea are within one order of magnitude. However, taking into account the uncertainty of determinations, they are significantly different and are in the range of 3–53.3 Bq/kg (FW) (see Table 8.4). As tuna are at the top of marine food chains and do not absorb ^{210}Po from water, different ^{210}Po concentrations in tuna must be a consequence of different ^{210}Po



Source: See Fig. 2 of Ref. [8.28].

FIG. 8.6. Polonium-210 in pelagic food chains in the ocean (reproduced with permission courtesy of Elsevier).

TABLE 8.4. ACTIVITY CONCENTRATIONS (AND RANGES) OF ^{210}Po IN TISSUES OF TUNA FISH FROM SEVERAL OCEANS

Region	Species	Tissue	^{210}Po concentration (Bq/kg, FW)	Ref.
NE Atlantic Ocean				
Azores Islands	Several	Muscle	5 (3–8)	[8.53]
		Liver	288 (278–297)	
		Bone	20 (5–51)	
Pacific Ocean,				
S. California	Several	Muscle	11–48	[8.34]
		Liver	144–777	
		Gonad	85–278	
Japan	*Thunnus thynnus*	Muscle	24.4 ± 1.6	[8.85]
Marshall Islands	Bonito	Muscle	36.9 (21.5–53.3)	[8.86]

concentrations in the first trophic levels of the food chain (i.e. in phytoplankton and zooplankton in their environment). The ^{210}Po analyses of plankton from the open ocean, especially from oligotrophic waters in the Pacific Ocean, actually show that ^{210}Po concentrations in zooplankton are very much affected by plankton density (see Ref. [8.101] for a model to describe the relationship). Low biomass prompts higher concentration levels in phytoplankton, which is transferred to zooplankton, and so forth. Polonium-210 concentrations in zooplankton in oligotrophic regions of the Pacific Ocean are indeed much higher than ^{210}Po concentrations in zooplankton of the North Atlantic Ocean. This is propagated in the food chain and reflected by ^{210}Po concentrations in muscle tissue of tuna fish of both oceans [8.5, 8.87].

8.5.4. Mesopelagic and bathypelagic zones

Zooplankton from the deep waters of the north-eastern Atlantic Ocean display very different concentrations according to the type of organism. Carvalho [8.28] reports that deep-sea medusa (*Atolla* sp. and *Crossota* sp.), salpes (*Salpa* spp.) and *Pyrosoma* sp. — all gelatinous organisms — contain the lowest ^{210}Po concentrations measured in marine organisms at 0.7–35 Bq/kg (with Po:Pb ratios generally < 5). Their ^{210}Po concentrations are similar to those determined in epipelagic jellyfish (i.e. Hydromedusae *Sarsia* spp. and *Obelia* spp.).

Research on mesopelagic decapod shrimp shows the existence of specialized diets, with the consumption of ^{210}Po rich organic particles in the oceanic water column, leading in some species to very high concentrations of ^{210}Po. High ^{210}Po concentrations are reported, especially in the hepatopancreas of some shrimp species [8.106] and in the liver and gut of some mesopelagic fish feeding on particles and zooplankton [8.54, 8.94]. This accumulation leads to unusually high internal radiation doses from the alpha emission of ^{210}Po, reaching values of 3–5 Sv/a, reported in the 1980s, in the hepatopancreas of shrimp and liver and other internal organs of small mesopelagic fish (*Argyropelecus* and *Myctophum*) [8.54, 8.57].

Carvalho [8.28] analyses more than 50 fish species living in the mid-water column and sampled near Madeira Island, at the Great Meteor East Seamountin of the Madeira Abyssal Basin, and in the Porcupine Abyssal Basin. Generally, most mesopelagic and bathypelagic fish species are small (0.5–60 g adult weight), but there are large differences in their ^{210}Po concentrations, suggesting feeding regimes based on different prey and different trophic levels (see Fig. 8.6).

Large predators in mesopelagic and bathypelagic domains include teleosts, such as *Aphanopus carbo* and *Mora mora*, and black sharks, such as *Scynodesmus* sp., all with low ^{210}Po concentrations, around 1 Bq/kg in muscle tissue, and comparable to concentrations in other fish in other ocean layers occupying similar trophic levels [8.28].

8.5.5. Abyssal zone

The amount of data available on abyssal species is much less than for other oceanic regions. Nevertheless, the available data show that ^{210}Pb and ^{210}Po concentrations determined in ascidians, polychaetes, Ophiuridae, cephalopods, crustaceans and fish are similar to concentrations reported for organisms at other oceanic depths (see Table 8.5).

Carvalho [8.28] finds that:

> "Also ^{210}Po concentrations measured in the giant abyssal mysid *Gnatophausia ingens* and Eryonidae crabs, 18 ± 2 and 22 ± 2 Bq kg^{-1} respectively, were close to the ^{210}Po concentration measured in the coastal shrimp *Leander serratus*, 25 ± 0.8 Bq kg^{-1}".

In Carvalho [8.28], a small bivalve mollusc from the abyssal plain (*Sillicula fragilis*) displays ^{210}Po concentrations in soft tissue comparable to the north-eastern Atlantic clam (*Tapes decussatus*) and the North Sea clam (*Mya arenaria*) (see also Ref. [8.84]). Unusually high ^{210}Po concentrations, 1.6×10^3 Bq/kg (DW) were reported in the 1980s for infaunal xenophytophores, large protozoans that live in sediment and are closely related to foraminifera, from a hadal trench in the Pacific Ocean, which might be exposed to radiation doses comparable to the dose in the hepatopancreas of mid-water shrimp [8.57, 8.107].

Carvalho [8.28] concludes that:

> "One singularity of abyssal fauna shall be highlighted. In 3 out of the 15 abyssal species analyzed the Po:Pb ratios were around the unity. In the abyssal fish *Nematonurus* (*Coryphaenoides*) *armatus*, a rattail fish characteristic of the abyssal zone, the ^{210}Po:^{210}Pb ratio in muscle was 0.5, which was uncommon amongst all fish analyzed [8.84]. These low Po:Pb ratio values are probably linked to the long fasting periods of abyssal fauna [Ref. [8.108]]."

TABLE 8.5. ACTIVITY CONCENTRATIONS (AND RANGES) OF ^{210}Po AND ^{210}Pb (FW) IN BENTHIC FAUNA OF THE PORCUPINE ABYSSAL PLAIN, THE NORTH-EASTERN ATLANTIC OCEAN AND HYDROTHERMAL VENTS IN THE MID-ATLANTIC RIDGE

Species and tissue	*N*	DW:FW	^{210}Po (Bq/kg ± SD)	^{210}Pb (Bq/kg ± SD)	Po:Pb
Ascidian					
Chitonanthus abyssorum	4	0.15	38 ± 2	38 ± 2	1
Polychaetes	3				
Gut		0.22	27 ± 2	11 ± 0.5	2.5
Remainder		0.18	52 ± 2	13 ± 0.5	4
Asteridae					
Astropecten sp., wt = 0.6 g	3	0.38	50 ± 3	7.6 ± 0.4	6.4
Astropecten sp., wt = 13 g	1	0.23	171 ± 13	25 ± 2	6.7
Bivalve mollusc (soft tissue)					
Silicula fragilis	1	0.16	124 ± 8	123 ± 4	1
Bathymodiolus azoricus[a]	—[b]	0.19	123.9 ± 21.4	—[b]	—[b]
Isopods	2				
Whole body		0.23	27 ± 1	1.91 ± 0.09	14
Gut		0.41	94 ± 3	11.4 ± 0.3	8.2
Exoskeleton		0.68	194 ± 5	8.4 ± 0.2	23
Amphipods					
Eurythenes grillus	3	0.20	32 ± 10	28 ± 11	1.1
Eurythenes grillus[a]	26	0.28	104.1 (62.4–285.9)	26.5 (11.8–67.4)	3.9
Cephalopods					
Vampiroteuthis infernalis	1	0.07	39 ± 3	6.9 ± 0.3	5.6
Fish					
Nematonurus (C.) armatus					
Muscle	9	0.17	0.31 ± 0.28	0.60 ± 0.55	0.5
Liver	9	—[b]	3.58 ± 2.29	2.50 ± 1.64	1.4
Skin	4	—[b]	1.94 ± 0.73	0.68 ± 0.33	2.8
Bone	4	—[b]	3.25 ± 1.96	0.85 ± 0.24	3.8

Source: See Ref. [8.83] unless otherwise indicated.

Note: SD — standard deviation.

[a] Source is Ref. [8.73].

[b] —: data not available.

8.6. POLONIUM AND LEAD CONCENTRATION RATIOS IN MARINE ORGANISMS

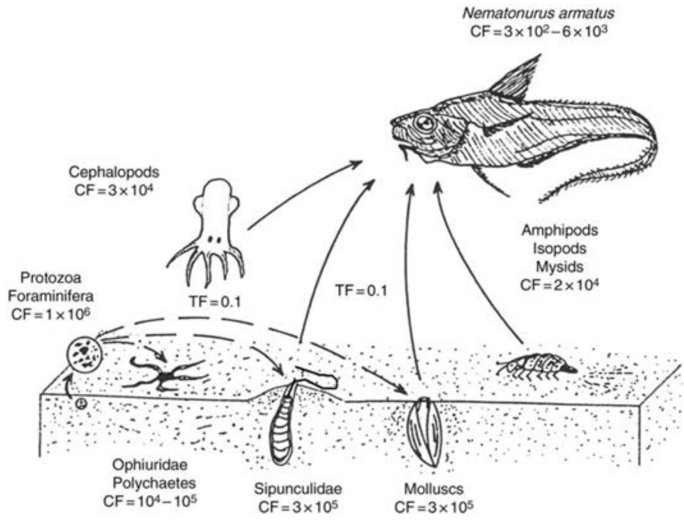
Concentration ratios (CR) are defined as the ratio of activity concentration in the organism (Bq/kg, FW) to the activity concentration in filtered sea water (Bq/L). Determinations of CRs are frequently performed when comparing bioaccumulation of radionuclides in different locations and seas and are used in modelling. As this ratio uses the concentration of radionuclide dissolved in sea water, it allows for an easy identification of organisms that concentrate more of the radionuclide, which might be of interest for further study and use in environmental radiological monitoring programmes as biomonitors. Organisms that concentrate a particular radionuclide might also be the main transfer route of that radionuclide to humans.

However, computation of CRs does not imply that sea water is the source of radionuclide to the organism. Indeed, it is food and not sea water that is the source of internally accumulated ^{210}Po. Nevertheless, the information on CRs for biota groups is useful to model the transfer of radionuclides in food chains and to predict potential radiation exposures in specified ecosystems and food webs. This is the main purpose of the data compilation and recommended CRs in IAEA publications such as the Handbook of Parameter Values for the Prediction of Radionuclide Transfer to Wildlife [8.109]. Furthermore, CRs of ^{210}Po concentrations in sea water are not different from those of similar species in coastal areas (see Fig. 8.7).

8.7. TRANSFER OF ^{210}Pb AND ^{210}Po IN MARINE FOOD CHAINS

In the past, the interpretation that ^{210}Po concentration in tissues of marine biota could be a reflection of ^{210}Po content in their diet was gaining credence, but the relative importance of water and food as ^{210}Po sources remained unknown. The use of radioactive tracers clarified that diet is the almost exclusive source of ^{210}Po accumulated in the internal organs of marine organisms [8.46, 8.47]. Since then, several studies have adopted this point of view and interpreted ^{210}Po data in marine biota in the light of food chains and trophic levels (see Refs [8.28, 8.52, 8.110, 8.111]).

A consistent view of ^{210}Po transfer in marine food chains in the pelagic environment is depicted in Fig. 8.7. Whole body ^{210}Po CRs are depicted, relating ^{210}Po concentrations in biota to ^{210}Po concentrations in sea water. Furthermore, ^{210}Po transfer factors (TFs) relating ^{210}Po concentrations in predator tissues to ^{210}Po concentrations in the diet of each species are indicated.



Source: Figure 1 of Ref. [8.84].
Note: Concentration ratios are equivalent to concentration factors (CFs).

FIG. 8.7. Transfer factors (TFs) and concentration ratios for ^{210}Po for the demersal food chain at the abyssal north-eastern Atlantic Ocean (reproduced with permission courtesy of Oxford Journals).

Although these are simplified trophic chains and crude representations of real food webs in the oceans, they are based on the known diet of species and predator–prey relationships [8.28]. The advantage is that it clearly shows that in the same oceanic ecosystem there are organisms often in the same zoological group, such as fish, that display very different concentration factors and thus highlight that water cannot be the main source of ^{210}Po to organisms. Furthermore, organisms with similar ^{210}Po CRs can be found in different regions and ecosystems without any relationship between them.

Carvalho [8.28] reports that ^{210}Po TFs from prey to predator are often around 0.1, with interesting deviations such as 0.7 in the trophic link from phytoplankton to copepods, 0.3 in the trophic link from small pelagic fish and euphausiids to tuna, and 0.7 in the transfer of ^{210}Po from zooplankton to the lantern fish (Myctophidae). Carvalho [8.28] proposes that these ^{210}Po TFs are similar to energy transfer in marine food chains, known as eco-trophic coefficients, and that this strongly suggests an association between ^{210}Po transfer and energy transfer in marine food chains. Furthermore [8.28]:

> “Based on ^{210}Po concentration data in marine biota combined with ^{210}Po behaviour in biological systems, such as ^{210}Po binding to amino acids and proteins [Ref. [8.48]], and experimental research on ^{210}Po and ^{210}Pb assimilation in marine species [Refs [8.46, 8.47, 8.49]], we interpret ^{210}Po transfer in marine food chains as a tracer of protein transfer from prey to predator tissues. In every ecosystem of the ocean, the accumulation of ^{210}Po in the marine biota thus reflects the trophic position of the species in the food web rather than the geographic location, water depth, or other environmental parameter.
>
> “The behaviour of ^{210}Pb in the food chains seems slightly different. ^{210}Pb concentrations do not increase from phytoplankton to copepods as much as ^{210}Po, and the ^{210}Pb transfer to planktivorous fish, such as sardines, is less efficient than ^{210}Po transfer.”

At higher trophic levels, such as in marine mammals, ^{210}Po levels are high, while ^{210}Pb levels are as low as in lower trophic levels. The Po:Pb ratio in marine food webs seem to increase with trophic level, from around 10 in phytoplankton and zooplankton, 3–10 in herbivorous fish and 50–100 in piscivores (carnivorous fish), rising to 200 in the muscle tissue of marine mammals [8.96].

In summary, the concentrations of dissolved ^{210}Pb and ^{210}Po in the oceans vary within a relatively narrow range. Differences between regions and ocean layers do exist, and there are geochemical and geographical reasons for this, such as ^{210}Pb and ^{210}Po concentrations in surface water of the oceans and land mass distribution. However, these differences are relatively small and are not sufficient to explain the wide range of ^{210}Po concentrations in biota, especially when species coexist in the same water body or region. Carvalho [8.28] finds that:

> “Po-210 and ^{210}Pb concentrations were determined in marine species from all the ecological zones in the ocean, from the shoreline to abyssal depths. In the tissues of all species the ^{210}Po concentration values spread across 5 orders of magnitude, from 0.5 Bq kg^{-1} in jellyfish to (2.8 ±0.2) × 10^4 Bq kg^{-1} in the gut wall of the common sardine [*Sardina pilchardus*] and even to (3.33 ± 0.17) × 10^4 Bq kg^{-1} in the gut walls of the pelagic shrimp *Plesionika edwardsi*.”

Furthermore, ^{210}Po concentrations seem to be similar in the tissues of a coastal bivalve mollusc and in a deep-sea mollusc, and in the muscle tissue of tuna fish in the Pacific Ocean and the Atlantic Ocean, provided that those species occupy a similar position in the food chain. Conversely, ^{210}Po concentrations in marine biota might be very different in species living in the same water mass, and therefore exposed to the same ^{210}Po concentrations in water, as reviewed in this appendix for many species in marine ecosystems.

The concentration of dissolved ^{210}Po in suspended particulate matter, including living microorganisms, such as bacteria and phytoplanton cells, probably starts with surface adsorption on ultra fine particles and accumulation of polonium in bacteria and phytoplankton as an element similar to sulphur in the building blocks of proteins — the amino acids. According to Carvalho [8.28], the enhancement of ^{210}Po concentrations is very pronounced in biota feeding on bacteria and phytoplankton at the base of marine food chains, such as observed in small zooplankton organisms (e.g. copepods and mysids), but also in large planktivorous organisms, such as mussels, sardines and blue whales, and it is transferred to carnivores, such as marlins, dolphins and sperm whales. Carvalho [8.28] concludes that:

> "Actually, the ^{210}Po activity concentration levels recorded in marine organisms and ^{210}Po:^{210}Pb ratios may depend upon the number of trophic levels in the food chain. The less trophic links exist in the food chain, the higher will be the ^{210}Po concentration in the top predator tissues."

Furthermore, in oligotrophic regions of the oceans, more ^{210}Po is concentrated per unit of mass of phytoplankton and zooplankton, and this ^{210}Po is transferred to upper trophic levels in the food chain, while in coastal seas with high plankton biomass, the reverse is observed (i.e. plankton display lower ^{210}Po concentrations) [8.101]. In marine organisms [8.28]:

> "^{210}Po:^{210}Pb ratios mostly ranged from near 1 up to 100, and occasionally higher, showing the enhancement of ^{210}Po concentrations comparatively to ^{210}Pb in internal organs, especially in those related to digestive functions and the gonad.
>
> ....
>
> "Regarding the ^{210}Po transfer in marine food chains, ^{210}Po concentrations in trophic levels and in characteristic species highlighted the similarities of ^{210}Po transfer factors with energy transfer rates in marine food chains."

This has been experimentally verified in a few species using a double tracer technique [8.46, 8.47]. Carvalho [8.28] suggests that:

> "^{210}Po follows protein transfer in food chains and may thus give a surrogate measure of the fraction of prey tissues that is incorporated in predator's tissues in marine food webs."

With regard to ^{210}Po transfer from the marine environment to human populations, the consumption of seafood from coastal seas is the main pathway and deserves much more attention in radiation exposure assessment. Humans, as consumers of seafood, are also elements of sea-borne food chains and, according to their dietary habits, will have different intakes of ^{210}Po and thus different internal radiation doses. This was pointed out in early studies on seafood consumers and documented in dietary studies for several countries [8.70, 8.86, 8.88]. This exposure of humans to ^{210}Po is not constant and, at present and due to overfishing, humans are increasingly fishing down the marine food chains, which implies that current trends of wild seafood consumption are likely to increase ^{210}Po ingestion and the collective radiation dose in seafood prone populations [8.28].

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Chapter 9

POLONIUM IN HUMANS

9.1. INTRODUCTION

Polonium-210 is one of the most radiotoxic natural radioactive isotopes known to humans due to its relatively long half-life (138 days) compared to many radon progeny in the uranium series, its high specific activity (166 TBq/g) and its emission of high linear energy transfer alpha radiation (5.304 MeV) [9.1]. On account of its high specific activity, it is also one of the rarest elements in nature. In addition to occurring naturally, it can also be produced by neutron activation of ^{209}Bi. The fractional intestinal uptake was given as 10–90% by Hill [9.2] in 1965 and as 50% by the International Commission on Radiological Protection (ICRP) in 1993 [9.3]. The daily intake is in the range of 37–370 mBq for a European diet [9.2], but higher daily intake occurs in seafood rich countries, such as Japan (480–690 mBq) [9.4]. The biological half-life is in the range of 26–100 days (see Table 9.1). There is a prompt excretion of about 3% within a period of 9 days [9.6].

With a half-life of 22.3 years, ^{210}Pb is the parent nuclide of ^{210}Po and builds up from the decay of ^{222}Rn in the uranium series. The inert noble gas ^{222}Rn is generated in all soils and rocks during the decay of ^{226}Ra and is subject to exhalation from the ground to the atmosphere. The daughter radionuclides of ^{222}Ra are then deposited by rain, wind and gravitational settling onto terrestrial vegetation. Hence, humans are exposed to radioactive polonium as a consequence of natural processes, and its estimated contribution to the annual background effective dose is 120 μSv/a [9.1].

Polonium accumulates readily in biota, mainly owing to its affinity for protein [9.2], and hence easily passes through the food chain to humans. For example, ^{210}Po has been found to bind to unidentified proteins with high molecular weight (70 kilodaltons), which could be haemocyanin or metal binding enzymes [9.15]. Protein rich foods, such as crustaceans and other shellfish, contain the highest activity concentrations of ^{210}Po. Elevated body burdens of ^{210}Po are also observed in humans that consume large amounts of reindeer meat, for example subarctic native populations [9.16, 9.17].

Most polonium enters the body through the consumption of food and then reaches the gastrointestinal tract, from which much of it is excreted with faeces. Polonium and lead absorbed into plasma are distributed throughout the soft tissue of the body and accumulate in the liver and kidneys [9.6]. The accumulation

TABLE 9.1. A SURVEY OF AVAILABLE DATA FOR FRACTIONAL UPTAKE AND BIOLOGICAL HALF-LIFE OF ^{210}Po IN HUMANS

Matrix	*N*	Gastrointestinal uptake fraction	Biological half-life (days)	Ref.
Crab meat	7	0.60–0.94	—[a]	[9.5]
Caribou meat	14	0.31–0.70	100	[9.6]
Normal food	15–63	0.35	83	[9.7]
	7	0.1–0.3	—[a]	[9.8]
	—[a]	0.05–0.45[b]	50	[9.9]
	—[a]	0.1	30–60	[9.10]
	—[a]	—[a]	73 (Inuit), 114 (Caucasians)	[9.11]
Po in water	5	0.38–0.78	31	[9.12]
Puncture wound (accident)	1	—[a]	26	[9.13]
Shellfish	7	0.44–0.52	28–51	[9.14]
	—[a]	0.50	50	[9.3]

[a] —: data not available.

[b] Depending on the organ.

of ^{210}Pb in the skeleton provides another source of ^{210}Po over time. Since ^{210}Po delivers the main absorbed dose of the Pb–Bi–Po decay chain, due to its alpha emission (E_{α} = 5.3 MeV), the behaviour of this isotope is of great interest.

Human biomonitoring is conducted mainly by urine and faecal sampling. Blood sampling, however, has the advantage of being an integrated indicator of polonium deposits across the metabolically active parts of the body; hence, changes in the polonium body burden should be reflected in the blood concentration. There is an absence, however, of suitable analogues (i.e. elements with similar biochemical behaviour to polonium) to help to understand the metabolic pathways in the human body which influence ^{210}Po biokinetics. Polonium is a chalcogen element and, hence, it has chemical similarities to the elements of the oxygen group in the periodic table (Group VIA). According to Waska et al. [9.15], ^{210}Po has similar pathway characteristics in biological matter as another chalcogen, selenium.

Most of the data for polonium activity concentrations in human tissues and other matrices are from the 1960s and 1970s. The analytical procedures at that time did not undergo any inter-calibration or quality control [9.18]. Yield determinants were often not used for the analytical procedure and a recovery of 100% was assumed, meaning that the true activity concentrations of ^{210}Po in the samples were often underestimated. Furthermore, the results might be affected by in vitro buildup of ^{210}Po from ^{210}Pb. Samples need to be analysed shortly after collection, otherwise ^{210}Pb would also have to be determined to correlate the result to the day of collection.

9.2. INGESTION AND INHALATION

The average daily intake of naturally occurring ^{210}Po for people following a typical European diet is estimated to be 37–370 mBq [9.2, 9.19]. Populations that consume marine food, such as crustaceans and shellfish, can have a much higher intake, and this can be exacerbated for people living in areas of low oceanic primary productivity, such as on the islands of the central Pacific Ocean [9.20] (see also Chapter 8). Certain critical groups, such as radium dial painters, exhibit exceptionally high levels [9.2, 9.21]. It is also clear that populations that consume caribou and reindeer have higher concentrations in their body [9.11, 9.16]. A health risk evaluation for ingestion of ^{210}Po has been performed by Scott [9.10] and an internal dose assessment using a biokinetic model is reported by Li et al. [9.9].

The main sources of ^{210}Po (and its progenitor ^{210}Pb) in humans are food, water and inhalation, with additional in vivo contributions from ^{222}Rn dissolved in the body as well as from ^{210}Pb and ^{226}Ra in bones. A 1966 study shows indications of higher levels of ^{210}Po in the organs of smokers [9.22]. The intake via inhalation is much smaller than that from food [9.7], even at uranium milling facilities [9.23].

9.3. DISTRIBUTION IN TISSUES

The highest activity concentrations of ^{210}Po in humans are found in the liver, kidneys and bone. The content in the skeleton and muscle tissue comprises about 70% of the total body burden. For bone, a significant proportion is due to buildup from ^{210}Pb [9.24]. Activity concentrations generally increase with age by accumulation of ^{226}Ra and ^{210}Pb. For other animals, the concentrations are also highest in the liver and kidneys, but the fraction of body content is different due to a large portion being contained in the fur [9.25]. Table 9.2 shows the results of a survey of ^{210}Po activity concentrations and the ^{210}Po:^{210}Pb activity ratio in different human organs. Almost all of the data are from the 1960s and 1970s,

TABLE 9.2. POLONIUM IN HUMAN ORGANS

Matrix	Activity concentration (mBq/kg, FW)	Provenance	Ref.
Lung	326	Former USSR	[9.7]; bb = 1.7%; ^{210}Po:^{210}Pb = 1.4
	318	USA, smokers	[9.2]
	118	USA, non-smokers	[9.2]; ^{210}Po:^{210}Pb = 1.7
	200	United Kingdom	[9.16]
	189	USA	[9.17]; ^{210}Po:^{210}Pb = 0.75
Lung parenchyma	290	USA, smokers	[9.22]
	93	USA, non-smokers	[9.22]
Lung bronchi	470	USA, smokers	[9.22]
	370	USA, non-smokers	[9.22]
Lung nodes	4 000	USA, smokers	[9.22]
	1 400	USA, non-smokers	[9.22]
Liver	970	Former USSR	[9.7]; bb = 8.7%; ^{210}Po:^{210}Pb = 2.2
	400	USA	[9.24]; bb = 1.7%
	625	United Kingdom	[9.16]
	7 000	Radium dial painters	[9.2]; ^{210}Po:^{210}Pb = 1.0
	740	USA, smokers	[9.2]
	550	USA, non-smokers	[9.2]; ^{210}Po:^{210}Pb = 2.1
Liver	529	USA	[9.17]; ^{210}Po:^{210}Pb = 2.1
	503	Children, AK, USA	[9.17]; ^{210}Po:^{210}Pb = 2.1
	530	USA, smokers	[9.22]
	220	USA, non-smokers	[9.22]
	750	Former USSR	[9.26]
Bone	1 070	United Kingdom	[9.16]
	55 000	Radium dial painters	[9.2]; ^{210}Po:^{210}Pb = 1.0
	8 700[a]	USA	[9.24]; bb = 63%
	1 070	Former USSR	[9.7]; bb = 50.2%
	1 700	—[b]	[9.26]

TABLE 9.2. POLONIUM IN HUMAN ORGANS (cont.)

Matrix	Activity concentration (mBq/kg, FW)	Provenance	Ref.
Bone (femur)	1 006	Children, AK, USA	[9.17]; ^{210}Po:^{210}Pb = 0.82
Blood	7 900	Radium dial painters	[9.21]; ^{210}Po:^{210}Pb = 0.7
	292[c]	Lapland, Finland	[9.27]
	38[c]	Helsinki, Finland	[9.27]
	370[c]	Sami, male, Sweden	[9.28]; ^{210}Po:^{210}Pb = 3.3
	180[c]	Sami, female, Sweden	[9.28]; ^{210}Po:^{210}Pb = 2.6
Muscle	111	Former USSR	[9.7]; bb = 17.6%; ^{210}Po:^{210}Pb = 0.8
	222	USA	[9.24]; bb = 17%
	22	USA, smokers	[9.22]
	37	USA, non-smokers	[9.22]
Hair	2 000–11 000[d]	Sweden	[9.29]; ^{210}Po:^{210}Pb = 2.2
	900 000	Radium dial painters	[9.2]; ^{210}Po:^{210}Pb = 1.0
	3 300	Former USSR	[9.7]; ^{210}Po:^{210}Pb = 2.2
	9 000–22 000[d]	India, non-smokers	[9.30]
	23 000[d]	India, smokers	[9.30]
	800–14 000[d]	USA, non-smokers	[9.31]
	1 200–12 000[d]	USA, smokers	[9.31]
	5 300	Former USSR	[9.26]
Kidney	640	United Kingdom	[9.16]
	970	Former USSR	[9.7]; bb = 1.2%; ^{210}Po:^{210}Pb = 2.8
	9 300	Radium dial painters	[9.2]; ^{210}Po:^{210}Pb = 9
	758	USA, smokers	[9.2]
	555	USA, non-smokers	[9.2]; ^{210}Po:^{210}Pb = 3.2
	384	USA	[9.17]; ^{210}Po:^{210}Pb = 2.6
	277	Children, AK, USA	[9.17]; ^{210}Po:^{210}Pb = 2.6
	330	USA, smokers	[9.22]
	210	USA, non-smokers	[9.22]

TABLE 9.2. POLONIUM IN HUMAN ORGANS (cont.)

Matrix	Activity concentration (mBq/kg, FW)	Provenance	Ref.
Placenta	122	United Kingdom	[9.16]
	740–4 000	Reindeer and caribou consumers, Canada	[9.16]; ^{210}Po:^{210}Pb = 6.3
	400	N. Canada	[9.11]
Gonads	144	USA, smokers	[9.2]
	104	USA, non-smokers	[9.2]
	255	USA	[9.17]; ^{210}Po:^{210}Pb = 1.1
Testis	144	United Kingdom	[9.16]
Spleen	117	USA	[9.17]; ^{210}Po:^{210}Pb = 1.8
	22	Children, AK, USA	[9.17]; ^{210}Po:^{210}Pb = 0.6
	220	Former USSR	[9.7]; bb = 0.8%; ^{210}Po:^{210}Pb = 0.2
Pancreas	107	USA	[9.17]; ^{210}Po:^{210}Pb = 1.8
Thyroid	189	USA	[9.17]
Heart	19	USA	[9.17]
	70	USA, smokers	[9.22]
	37	USA, non-smokers	[9.22]
Eye, choroid and iris	1 000–10 000	United Kingdom	[9.32]
Bone marrow	24	United Kingdom	[9.33]

Note: bb — body burden.

[a] mBq/kg, ash.

[b] —: data not available.

[c] mBq/L.

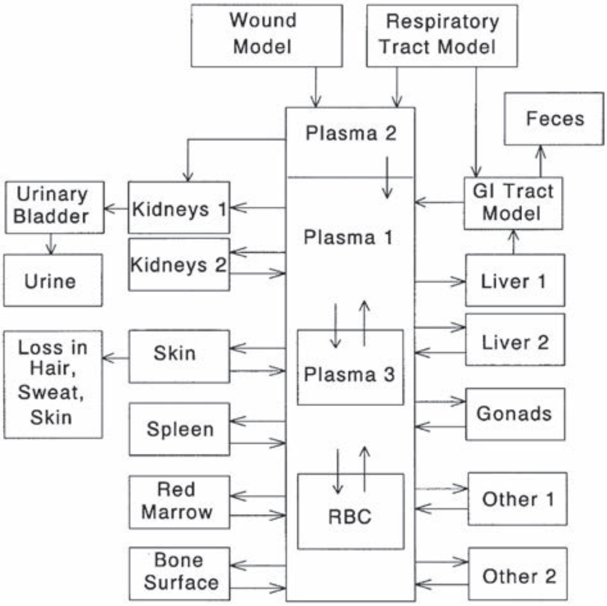
[d] mBq/kg (DW).

when access to autopsy samples was permitted. However, estimation of body content can be achieved by analysing urine and faeces. Hair also seems to be a good indicator of polonium contamination [9.12].

Polonium has an affinity for proteins. For example, gelatin is reported to contain 1500 mBq/kg of ^{210}Po and insulin 23 000–70 000 mBq/kg [9.2]. In the past, people with diabetes may have been exposed to elevated levels of ^{210}Po in natural insulin. Insulin is now made artificially; it is no longer extracted from the pancreas and does not contain any ^{210}Po. However, another critical group might be people undergoing dialysis. If polonium became associated with protein molecules in the body too large to pass through the membrane during dialysis, the radionuclide might build up in the body. This has not yet been investigated.

9.4. RETENTION AND BIOLOGICAL HALF-LIVES

The kinetics of polonium are rather complex (see Fig. 9.1) and are influenced by its wide variety of potential oxidation states (−2, +2, +4 and +6; see Section 3.2). Polonium can be released from one organ and recirculated.



Source: Figure 5 of Ref. [9.34].

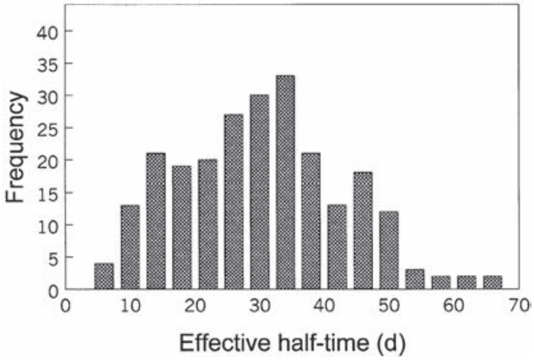
FIG. 9.1. Compartments of the systemic model, paths of movement of polonium between compartments, and connections between the systemic model and user-supplied models of the respiratory tract, gastrointestinal tract and wounds (reproduced with permission courtesy of Elsevier).

The in vivo concentration of ^{210}Pb causes buildup of ^{210}Po, especially in bone where the biological half-life of ^{210}Pb is long. In addition, ^{226}Ra plays a role as a precursor of ^{210}Po. Several studies report values between 10 and 60 days for the effective biological half-life of ^{210}Po in humans (see Fig. 9.2), and fractional uptake following ingestion ranges from a few percent to 90%. The dose factor is thereby quite uncertain. All those factors probably depend, to a large degree, on the composition of the foodstuff with which polonium is ingested.

The highest activity concentrations in humans are found in the liver and, to a lesser extent, in the kidneys. It has also been found that hair is a good indicator of polonium contamination and that hair is an important excretion route in addition to faecal and urinary routes [9.11]. Due to its complex biokinetics and its array of exposure and excretion pathways, the biological and effective half-lives of polonium are highly variable (see Fig. 9.2).

Leggett and Eckerman [9.34] find that:

> "Although there is a relatively large database on the biological behavior of polonium in man and laboratory animals, some important aspects of the biokinetics of polonium in man have not been characterized with much certainty. A central problem is that a substantial portion of the data on urinary excretion of ^{210}Po by human subjects may not be reliable. Although relatively detailed data are available from a controlled study on human subjects, these data are limited not only by potential errors in the urinary excretion data but also by the fact that the state of health of the subjects could have affected the biokinetics of polonium."



Source: Figure 2 of Ref. [9.34].

FIG. 9.2. Distribution of effective half-lives derived from urinary excretion records for a large number of workers using polonium (reproduced with permission courtesy of Elsevier).

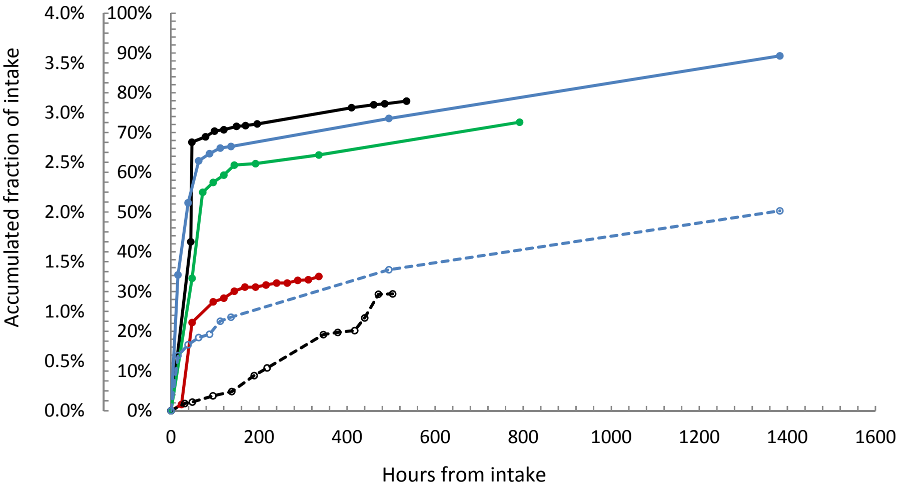
Hence, the results might be poorly representative of the general population. Conclusions about data on laboratory animals are complicated by the variation in biokinetics of polonium due to differences between “species, route of exposure, and the chemical form of polonium taken into the body” [9.34].

The human gastrointestinal uptake fraction f_1 has been established by a number of different studies, with a wide range of results (for fractional uptake and biological half-life for ^{210}Po, see Table 9.1). Thomas et al. [9.6] show that a total ingestion of 20 Bq with caribou meat resulted in a maximum of 3.2 Bq/d in faecal excretion over 4–6 days after intake and a maximum of 0.32 Bq/d of urinary excretion from 5 to 10 days after ingestion. From these results, a gastrointestinal uptake fraction of 0.56 ± 0.04 was established. In the 1950s, Silberstein et al. [9.8] published gastrointestinal uptake fractions as low as 0.1–0.3. The ICRP [9.3] has increased its reported gastrointestinal uptake fraction from 10% to 50%, which, according to other studies, still seems too small. Hunt and Allington [9.5] report gastrointestinal uptake fractions in the range of 0.6–0.94, with a mean of 0.76, after analysing a study in 1991 in which seven volunteers ingested crab meat with a total ^{210}Po activity intake of up to 44.2 Bq. A range of 10–25% for daily faecal excretion and a maximum daily urine excretion of 0.2–0.3% is reported. Additional data from the 1970s are provided by Landinskaya et al. [9.7], who analysed tissue from humans and estimated their intake from normal food, and in Ref. [9.11], which reports on the retention of ^{210}Po in Inuit and Caucasians.

In a 2012 study on radioactive polonium biokinetics by Henricsson [9.12], volunteers ingested ^{209}Po in a $PoCl_4$ solution. The excretion via faeces and urine of four of the subjects is shown in Fig. 9.3. It is well known that there are some critical groups, such as those who eat reindeer or caribou and those who consume large quantities of marine food (particularly molluscs and crustaceans).

In conclusion, there have been limited data generated on the kinetics and organ distribution of polonium in humans since the 1970s. Only about eight studies have been performed and, of these, only three experimental studies were conducted after the 1970s. Considering that ^{210}Po is the largest contributor of radiological dose to humans (from alpha particle emission), this is surprising.

There have been some critical population groups identified, such as those who consume large amounts of seafood or consume caribou and reindeer. However, the large variations in fractional uptake and biological residence time results in a large uncertainty in the estimated dose factor for oral intake of ^{210}Po. The fractional intake probably depends on the food composition with which polonium is consumed, which will vary between regions and lifestyles [9.16]. It would be of interest to conduct a survey of the main food for ingestion of ^{210}Po in different countries, perhaps by analysing hair, which could be a good indicator of internal contamination [9.11, 9.30].



Source: Data from Ref. [9.12].

FIG. 9.3. Accumulated excretion of ^{209}Po via faeces.

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Chapter 10

RADIOLOGICAL DOSE ASSESSMENT FOR ^{210}Po TO HUMANS AND OTHER BIOTA

10.1. MODELS AND DATA FOR ESTIMATING INTERNAL EXPOSURES TO HUMANS

Dosimetric models to estimate internal exposures in humans, subsequent to incorporation of radionuclides via ingestion and inhalation, are developed by the International Commission on Radiological Protection (ICRP). The most recent models for ^{210}Po are described in ICRP Publications 67 [10.1] and 72 [10.2] for ingestion, and in ICRP Publications 71 [10.3] and 72 [10.2] for inhalation. The end points of those models are dose coefficients for ingestion and inhalation, which are calculated for six age groups (3 month old children; 1, 5, 10 and 15 year old children; and adults). The dose coefficients are based on the equivalent dose rates received until the age of 70 years following an acute intake at the age of intake: for a one year old child, the dose contributions are integrated over a period of 69 years; for adults, the dose is integrated over a period of 50 years.

The key processes in the biokinetic models are absorption of the radionuclides into the blood and the distribution and retention in the organism. Hence, the time integrated concentrations in the organs and tissues and the number of disintegrations during the integration period are calculated from these quantities. For a specific organ, the integrated absorbed dose is calculated from the following:

(a) The number of decays during the integration period;
(b) The energy per decay;
(c) The energy deposited in a specific organ from radionuclides within the organ;
(d) The energy deposited in a specific organ from radionuclides decaying in other organs and tissues.

The model parameters applied in the biokinetic models are derived from all available and appropriate experiments and observations on the uptake and distribution of radionuclides, many of which determined from animal studies.

Following ingestion, the transfer of ^{210}Po in the ICRP model from the gut to the blood (resorption) is age dependent. The highest resorption is applied for infants (3 months) with 100%, and 50% for all other age groups [10.1].

Distribution and retention are described by complex model simulation of the fluxes of ^{210}Po between different organs and tissues. The most important process for long term retention is the exchange of ^{210}Po between the kidneys, liver and skeleton.

To calculate dose per unit intake values, it is assumed that for ^{210}Po entering the systemic circulation, fractions of 0.05, 0.1, 0.1, 0.3 and 0.45 are deposited in the spleen, kidneys, red bone marrow, liver and the rest of the body, respectively [10.1], where it is retained with a half-life of 50 days. For ^{210}Po that has entered the excretory system, a urinary:faecal excretion ratio of 1:2 is assumed.

The transfer to the blood from the lungs depends on the age and the chemical form of ^{210}Po. In general, the ICRP takes into account three absorption classes: slow (S), medium (M) and fast (F) retention. In Tables 10.1–10.4, the dose coefficients for ^{210}Po are summarized for ingestion and inhalation. These are given as effective dose per unit intake (Sv/Bq), and all of the organs considered in Refs [10.1, 10.3] are listed. For ingestion of ^{210}Po, the organs with the highest dose coefficients are the kidneys, liver, bone surface, red bone marrow and spleen. For inhalation, the dose coefficients for the lung and the extrathoracic airways are also important. There are large differences between the dose coefficients for the different absorption classes, in particular when comparing organ doses (see Tables 10.2–10.4). To estimate the effective dose from ^{210}Po, the highest dose coefficients are calculated for the absorption class S, which are about a factor of seven higher than for inhalation class F. This is because the less soluble materials remain in the lungs longer and thus provide more exposure to those tissues. The ranking of the organ doses also varies with the inhalation class.

10.2. DOSE CONVERSION FACTORS FOR WILDLIFE

10.2.1. General assumptions

Plants and animals can be exposed to ionizing radiation from radionuclides in the environment by both external and internal exposure. The United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) [10.4] reports that:

> “101. Radionuclides distributed in the environment lead to external exposure of an organism living in or close to a medium that contains radionuclides. The external exposure of biota is the result of complex and non-linear interactions of various factors:

TABLE 10.1. DOSE COEFFICIENTS FOR INGESTION OF ^{210}Po

Organ/tissue	Dose coefficient (Sv/Bq)					
Age at intake (years)	<1	1–2	2–7	7–12	12–17	>17
f_1 value	1	0.5	0.5	0.5	0.5	0.5
Adrenals	5.7×10^{-6}	2.0×10^{-6}	9.8×10^{-7}	5.8×10^{-7}	3.4×10^{-7}	2.8×10^{-7}
Bladder	5.7×10^{-6}	2.0×10^{-6}	9.8×10^{-7}	5.8×10^{-7}	3.4×10^{-7}	2.8×10^{-7}
Bone surface	8.0×10^{-5}	2.3×10^{-5}	8.9×10^{-6}	4.8×10^{-6}	2.8×10^{-6}	1.6×10^{-6}
Brain	5.7×10^{-6}	2.0×10^{-6}	9.8×10^{-7}	5.8×10^{-7}	3.4×10^{-7}	2.8×10^{-7}
Breast	5.7×10^{-6}	2.0×10^{-6}	9.8×10^{-7}	5.8×10^{-7}	3.4×10^{-7}	2.8×10^{-7}
Gastrointestinal tract						
Oesophagus	5.7×10^{-6}	2.0×10^{-6}	9.8×10^{-7}	5.8×10^{-7}	3.4×10^{-7}	2.8×10^{-7}
Stomach	5.7×10^{-6}	2.0×10^{-6}	9.8×10^{-7}	5.8×10^{-7}	3.4×10^{-7}	2.8×10^{-7}
Small intestine	5.7×10^{-6}	2.0×10^{-6}	9.8×10^{-7}	5.9×10^{-7}	3.4×10^{-7}	2.8×10^{-7}
Upper large intestine	5.8×10^{-6}	2.1×10^{-6}	1.0×10^{-6}	6.1×10^{-7}	3.6×10^{-7}	2.9×10^{-7}
Lower large intestine	6.0×10^{-6}	2.3×10^{-6}	1.1×10^{-6}	6.7×10^{-7}	3.9×10^{-7}	3.2×10^{-7}
Colon	5.9×10^{-6}	2.1×10^{-6}	1.1×10^{-6}	6.4×10^{-7}	3.7×10^{-7}	3.0×10^{-7}
Kidney	1.8×10^{-4}	6.2×10^{-5}	3.4×10^{-5}	2.3×10^{-5}	1.6×10^{-5}	1.3×10^{-5}
Liver	1.1×10^{-4}	4.0×10^{-5}	2.0×10^{-5}	1.3×10^{-5}	8.5×10^{-6}	6.6×10^{-6}
Muscle	5.7×10^{-6}	2.0×10^{-6}	9.8×10^{-7}	5.8×10^{-7}	3.4×10^{-7}	2.8×10^{-7}
Ovaries	5.7×10^{-6}	2.0×10^{-6}	9.8×10^{-7}	5.8×10^{-7}	3.4×10^{-7}	2.8×10^{-7}
Pancreas	5.7×10^{-6}	2.0×10^{-6}	9.8×10^{-7}	5.8×10^{-7}	3.4×10^{-7}	2.8×10^{-7}
Red bone marrow	8.1×10^{-5}	2.6×10^{-5}	1.2×10^{-5}	6.4×10^{-6}	3.8×10^{-6}	2.6×10^{-6}
Respiratory tract						
Extrathoracic airways	5.7×10^{-6}	2.0×10^{-6}	9.8×10^{-7}	5.8×10^{-7}	3.4×10^{-7}	2.8×10^{-7}
Lungs	5.7×10^{-6}	2.0×10^{-6}	9.8×10^{-7}	5.8×10^{-7}	3.4×10^{-7}	2.8×10^{-7}
Skin	5.7×10^{-6}	2.0×10^{-6}	9.8×10^{-7}	5.8×10^{-7}	3.4×10^{-7}	2.8×10^{-7}
Spleen	2.2×10^{-4}	7.6×10^{-5}	4.1×10^{-5}	2.5×10^{-5}	1.6×10^{-5}	1.1×10^{-5}
Testes	5.7×10^{-6}	2.0×10^{-6}	9.8×10^{-7}	5.8×10^{-7}	3.4×10^{-7}	2.8×10^{-7}
Thymus	5.7×10^{-6}	2.0×10^{-6}	9.8×10^{-7}	5.8×10^{-7}	3.4×10^{-7}	2.8×10^{-7}
Thyroid	5.7×10^{-6}	2.0×10^{-6}	9.8×10^{-7}	5.8×10^{-7}	3.4×10^{-7}	2.8×10^{-7}
Uterus	5.7×10^{-6}	2.0×10^{-6}	9.8×10^{-7}	5.8×10^{-7}	3.4×10^{-7}	2.8×10^{-7}
Remainder	9.3×10^{-5}	3.9×10^{-5}	2.1×10^{-5}	1.3×10^{-5}	8.2×10^{-6}	6.6×10^{-6}
Effective dose	2.6×10^{-5}	8.8×10^{-6}	4.4×10^{-6}	2.6×10^{-6}	1.6×10^{-6}	1.2×10^{-6}

Source: See Ref. [10.1].

TABLE 10.2. DOSE COEFFICIENTS FOR INHALATION OF ^{210}Po, INHALATION CLASS F

Organ/tissue	Dose coefficient (Sv/Bq)					
Age at intake (years)	<1	1–2	2–7	7–12	12–17	>17
f_1 value	0.2	0.1	0.1	0.1	0.1	0.1
Adrenals	1.6×10^{-6}	1.1×10^{-6}	4.9×10^{-7}	2.9×10^{-7}	1.7×10^{-7}	1.4×10^{-7}
Bladder	1.6×10^{-6}	1.1×10^{-6}	4.9×10^{-7}	2.9×10^{-7}	1.7×10^{-7}	1.4×10^{-7}
Bone surface	2.3×10^{-5}	1.3×10^{-5}	4.5×10^{-6}	2.4×10^{-6}	1.4×10^{-6}	8.0×10^{-7}
Brain	1.6×10^{-6}	1.1×10^{-6}	4.9×10^{-7}	2.9×10^{-7}	1.7×10^{-7}	1.4×10^{-7}
Breast	1.6×10^{-6}	1.1×10^{-6}	4.9×10^{-7}	2.9×10^{-7}	1.7×10^{-7}	1.4×10^{-7}
Gastrointestinal tract						
Oesophagus	1.6×10^{-6}	1.1×10^{-6}	4.9×10^{-7}	2.9×10^{-7}	1.7×10^{-7}	1.4×10^{-7}
Stomach	1.6×10^{-6}	1.1×10^{-6}	4.9×10^{-7}	2.9×10^{-7}	1.7×10^{-7}	1.4×10^{-7}
Small intestine	1.6×10^{-6}	1.1×10^{-6}	4.9×10^{-7}	2.9×10^{-7}	1.7×10^{-7}	1.4×10^{-7}
Upper large intestine	1.7×10^{-6}	1.1×10^{-6}	5.0×10^{-7}	3.0×10^{-7}	1.7×10^{-7}	1.4×10^{-7}
Lower large intestine	1.8×10^{-6}	1.2×10^{-6}	5.3×10^{-7}	3.2×10^{-7}	1.8×10^{-7}	1.5×10^{-7}
Colon	1.7×10^{-6}	1.1×10^{-6}	5.2×10^{-7}	3.1×10^{-7}	1.7×10^{-7}	1.6×10^{-7}
Kidney	5.1×10^{-5}	3.4×10^{-5}	1.7×10^{-5}	1.1×10^{-5}	7.7×10^{-6}	6.4×10^{-6}
Liver	3.1×10^{-5}	2.2×10^{-5}	1.0×10^{-5}	6.6×10^{-6}	4.1×10^{-6}	3.3×10^{-6}
Muscle	1.6×10^{-6}	1.1×10^{-6}	4.9×10^{-7}	2.9×10^{-7}	1.7×10^{-7}	1.4×10^{-7}
Ovaries	1.6×10^{-6}	1.1×10^{-6}	4.9×10^{-7}	2.9×10^{-7}	1.7×10^{-7}	1.4×10^{-7}
Pancreas	1.6×10^{-6}	1.1×10^{-6}	4.9×10^{-7}	2.9×10^{-7}	1.7×10^{-7}	1.4×10^{-7}
Red bone marrow	2.3×10^{-5}	1.4×10^{-5}	6.1×10^{-6}	3.2×10^{-6}	1.8×10^{-6}	1.3×10^{-6}
Respiratory Tract						
Extrathoracic airways	1.6×10^{-6}	1.1×10^{-6}	5.0×10^{-7}	3.0×10^{-7}	1.7×10^{-7}	1.4×10^{-7}
Lungs	1.7×10^{-6}	1.1×10^{-6}	5.2×10^{-7}	3.1×10^{-7}	1.9×10^{-7}	1.6×10^{-7}
Skin	1.6×10^{-6}	1.1×10^{-6}	4.9×10^{-7}	2.9×10^{-7}	1.7×10^{-7}	1.4×10^{-7}
Spleen	6.3×10^{-5}	4.2×10^{-5}	2.0×10^{-5}	1.3×10^{-5}	7.8×10^{-6}	5.5×10^{-6}
Testes	1.6×10^{-6}	1.1×10^{-6}	4.9×10^{-7}	2.9×10^{-7}	1.7×10^{-7}	1.4×10^{-7}
Thymus	1.6×10^{-6}	1.1×10^{-6}	4.9×10^{-7}	2.9×10^{-7}	1.7×10^{-7}	1.4×10^{-7}
Thyroid	1.6×10^{-6}	1.1×10^{-6}	4.9×10^{-7}	2.9×10^{-7}	1.7×10^{-7}	1.4×10^{-7}
Uterus	1.6×10^{-6}	1.1×10^{-6}	4.9×10^{-7}	2.9×10^{-7}	1.7×10^{-7}	1.4×10^{-7}
Remainder	3.3×10^{-5}	2.2×10^{-5}	1.0×10^{-5}	6.5×10^{-6}	4.0×10^{-6}	3.3×10^{-6}
Effective dose	7.4×10^{-6}	4.8×10^{-6}	2.2×10^{-6}	1.3×10^{-6}	7.7×10^{-7}	6.1×10^{-7}

Source: See Ref. [10.3].

TABLE 10.3. DOSE COEFFICIENTS FOR INHALATION OF ^{210}Po, INHALATION CLASS M

Organ/tissue	Dose coefficient (Sv/Bq)					
Age at intake (years)	<1	1–2	2–7	7–12	12–17	>17
f_1 value	0.2	0.1	0.1	0.1	0.1	0.1
Adrenals	6.4×10^{-7}	3.5×10^{-7}	1.6×10^{-7}	9.7×10^{-8}	5.7×10^{-8}	4.9×10^{-8}
Bladder	6.4×10^{-7}	3.5×10^{-7}	1.6×10^{-7}	9.7×10^{-8}	5.7×10^{-8}	4.9×10^{-8}
Bone surface	8.9×10^{-6}	4.1×10^{-6}	1.5×10^{-6}	7.9×10^{-7}	4.6×10^{-7}	2.8×10^{-7}
Brain	6.4×10^{-7}	3.5×10^{-7}	1.6×10^{-7}	9.7×10^{-8}	5.7×10^{-8}	4.9×10^{-8}
Breast	6.4×10^{-7}	3.5×10^{-7}	1.6×10^{-7}	9.7×10^{-8}	5.7×10^{-8}	4.9×10^{-8}
Gastrointestinal tract						
Oesophagus	6.4×10^{-7}	3.5×10^{-7}	1.6×10^{-7}	9.7×10^{-8}	5.7×10^{-8}	4.9×10^{-8}
Stomach	6.5×10^{-7}	3.5×10^{-7}	1.6×10^{-7}	9.7×10^{-8}	5.7×10^{-8}	4.9×10^{-8}
Small intestine	6.5×10^{-7}	3.5×10^{-7}	1.7×10^{-7}	9.8×10^{-8}	5.8×10^{-8}	5.0×10^{-8}
Upper large intestine	7.1×10^{-7}	3.9×10^{-7}	1.8×10^{-7}	1.1×10^{-7}	6.3×10^{-8}	5.4×10^{-8}
Lower large intestine	8.2×10^{-7}	4.7×10^{-7}	2.2×10^{-7}	1.3×10^{-7}	7.3×10^{-8}	6.2×10^{-8}
Colon	7.6×10^{-7}	4.3×10^{-7}	2.0×10^{-7}	1.2×10^{-7}	6.7×10^{-8}	5.7×10^{-8}
Kidney	2.0×10^{-5}	1.1×10^{-5}	5.7×10^{-6}	3.8×10^{-6}	2.7×10^{-6}	2.2×10^{-6}
Liver	1.2×10^{-5}	7.1×10^{-6}	3.4×10^{-6}	2.2×10^{-6}	1.4×10^{-6}	1.2×10^{-6}
Muscle	6.4×10^{-7}	3.5×10^{-7}	1.6×10^{-7}	9.7×10^{-8}	5.7×10^{-8}	4.9×10^{-8}
Ovaries	6.4×10^{-7}	3.5×10^{-7}	1.6×10^{-7}	9.7×10^{-8}	5.7×10^{-8}	4.9×10^{-8}
Pancreas	6.4×10^{-7}	3.5×10^{-7}	1.6×10^{-7}	9.7×10^{-8}	5.7×10^{-8}	4.9×10^{-8}
Red bone marrow	9.1×10^{-6}	4.6×10^{-6}	2.1×10^{-6}	1.1×10^{-6}	6.3×10^{-7}	4.6×10^{-7}
Respiratory tract						
Extrathoracic airways	3.2×10^{-5}	2.4×10^{-5}	9.9×10^{-6}	6.6×10^{-6}	3.6×10^{-6}	3.5×10^{-6}
Lungs	1.1×10^{-4}	8.1×10^{-5}	5.1×10^{-5}	3.5×10^{-5}	3.1×10^{-5}	2.6×10^{-5}
Skin	6.4×10^{-7}	3.5×10^{-7}	1.6×10^{-7}	9.7×10^{-8}	5.7×10^{-8}	4.9×10^{-8}
Spleen	2.5×10^{-5}	1.4×10^{-5}	6.8×10^{-6}	4.2×10^{-6}	2.7×10^{-6}	1.9×10^{-6}
Testes	6.4×10^{-7}	3.5×10^{-7}	1.6×10^{-7}	9.7×10^{-8}	5.7×10^{-8}	4.9×10^{-8}
Thymus	6.4×10^{-7}	3.5×10^{-7}	1.6×10^{-7}	9.7×10^{-8}	5.7×10^{-8}	4.9×10^{-8}
Thyroid	6.4×10^{-7}	3.5×10^{-7}	1.6×10^{-7}	9.7×10^{-8}	5.7×10^{-8}	4.9×10^{-8}
Uterus	6.4×10^{-7}	3.5×10^{-7}	1.6×10^{-7}	9.7×10^{-8}	5.7×10^{-8}	4.9×10^{-8}
Remainder	1.0×10^{-6}	5.6×10^{-7}	2.7×10^{-7}	1.6×10^{-7}	1.0×10^{-7}	8.4×10^{-8}
Effective dose	1.5×10^{-5}	1.1×10^{-5}	6.7×10^{-6}	4.6×10^{-6}	4.0×10^{-6}	3.3×10^{-6}

Source: See Ref. [10.3].

TABLE 10.4. DOSE COEFFICIENTS FOR INHALATION OF ^{210}Po, INHALATION CLASS S

Organ/tissue	Dose coefficient (Sv/Bq)					
Age at intake (years)	<1	1–2	2–7	7–12	12–17	>17
f_1 value	0.02	0.01	0.01	0.01	0.01	0.01
Adrenals	4.4×10^{-8}	1.9×10^{-8}	8.5×10^{-9}	5.0×10^{-9}	2.8×10^{-9}	2.4×10^{-9}
Bladder	4.4×10^{-8}	1.9×10^{-8}	8.5×10^{-9}	5.1×10^{-9}	2.9×10^{-9}	2.4×10^{-9}
Bone surface	6.1×10^{-7}	2.2×10^{-7}	7.7×10^{-8}	4.1×10^{-8}	2.3×10^{-8}	1.4×10^{-8}
Brain	4.4×10^{-8}	1.9×10^{-8}	8.5×10^{-9}	5.0×10^{-9}	2.8×10^{-9}	2.4×10^{-9}
Breast	4.4×10^{-8}	1.9×10^{-8}	8.5×10^{-9}	5.0×10^{-9}	2.8×10^{-9}	2.4×10^{-9}
Gastrointestinal tract						
Oesophagus	4.4×10^{-8}	1.9×10^{-8}	8.5×10^{-9}	5.0×10^{-9}	2.8×10^{-9}	2.4×10^{-9}
Stomach	4.9×10^{-8}	2.1×10^{-8}	9.6×10^{-9}	5.7×10^{-9}	3.2×10^{-9}	2.7×10^{-9}
Small intestine	5.5×10^{-8}	2.6×10^{-8}	1.2×10^{-8}	7.0×10^{-9}	3.8×10^{-9}	3.2×10^{-9}
Upper large intestine	1.1×10^{-7}	6.5×10^{-8}	2.8×10^{-8}	1.7×10^{-8}	8.6×10^{-9}	7.2×10^{-9}
Lower large intestine	2.4×10^{-7}	1.5×10^{-7}	6.4×10^{-8}	3.9×10^{-8}	2.0×10^{-8}	1.6×10^{-8}
Colon	1.7×10^{-7}	1.0×10^{-7}	4.4×10^{-8}	2.6×10^{-8}	1.3×10^{-8}	1.1×10^{-8}
Kidney	1.4×10^{-6}	5.9×10^{-7}	2.9×10^{-7}	2.0×10^{-7}	1.3×10^{-7}	1.1×10^{-7}
Liver	8.4×10^{-7}	3.8×10^{-7}	1.7×10^{-7}	1.2×10^{-7}	7.0×10^{-8}	5.7×10^{-8}
Muscle	4.4×10^{-8}	1.9×10^{-8}	8.5×10^{-9}	5.0×10^{-9}	2.8×10^{-9}	2.4×10^{-9}
Ovaries	4.4×10^{-8}	1.9×10^{-8}	8.5×10^{-9}	5.0×10^{-9}	2.8×10^{-9}	2.4×10^{-9}
Pancreas	4.4×10^{-8}	1.9×10^{-8}	8.5×10^{-9}	5.0×10^{-9}	2.8×10^{-9}	2.4×10^{-9}
Red bone marrow	6.2×10^{-7}	2.4×10^{-7}	1.1×10^{-7}	5.6×10^{-8}	3.1×10^{-8}	2.3×10^{-8}
Respiratory tract						
Extrathoracic airways	5.7×10^{-5}	4.4×10^{-5}	1.9×10^{-5}	1.3×10^{-5}	7.1×10^{-6}	6.9×10^{-6}
Lungs	1.5×10^{-4}	1.1×10^{-4}	7.2×10^{-5}	4.9×10^{-5}	4.3×10^{-5}	3.5×10^{-5}
Skin	4.4×10^{-8}	1.9×10^{-8}	8.5×10^{-9}	5.0×10^{-9}	2.8×10^{-9}	2.4×10^{-9}
Spleen	1.7×10^{-6}	7.2×10^{-7}	3.5×10^{-7}	2.2×10^{-7}	1.3×10^{-7}	9.6×10^{-8}
Testes	4.4×10^{-8}	1.9×10^{-8}	8.5×10^{-9}	5.0×10^{-9}	2.8×10^{-9}	2.4×10^{-9}
Thymus	4.4×10^{-8}	1.9×10^{-8}	8.5×10^{-9}	5.0×10^{-9}	2.8×10^{-9}	2.4×10^{-9}
Thyroid	4.4×10^{-8}	1.9×10^{-8}	8.5×10^{-9}	5.0×10^{-9}	2.8×10^{-9}	2.4×10^{-9}
Uterus	4.4×10^{-8}	1.9×10^{-8}	8.5×10^{-9}	5.0×10^{-9}	2.8×10^{-9}	2.4×10^{-9}
Remainder	1.0×10^{-7}	4.8×10^{-8}	2.2×10^{-8}	1.4×10^{-8}	8.3×10^{-9}	7.5×10^{-9}
Effective dose	1.8×10^{-5}	1.4×10^{-5}	8.6×10^{-6}	5.9×10^{-6}	5.1×10^{-6}	4.3×10^{-6}

Source: See Ref. [10.3].

> — The geometrical relation between the source of the radiation and the target;
> — The activity levels of the radionuclides in the environment;
> — The materials in the environment and their shielding properties;
> — The radionuclide-specific decay properties characterized by the radiation type, the energies emitted and their emission probabilities; and
> — The habitat and size of the organism."

For estimating exposures to wildlife, models have been developed for deriving dose conversion coefficients (DCCs) for a set of plants and animals. Since it is impossible to consider all species of flora and fauna explicitly in dose assessment, DCCs are provided for a set of reference organisms that were selected within the FASSET and ERICA project to represent typical members of common ecosystems (see Refs [10.5–10.7]). The reference organisms considered here are summarized in Table 10.5. For calculating DCCs for internal and external exposures, the following simplifying assumptions were made:

(a) The radionuclides are uniformly distributed within the body and the supporting media.
(b) The shape of all organisms is approximated by spheres and ellipsoids.

10.2.2. Dose conversion factors for flora and fauna

UNSCEAR [10.4] reports that (see also Refs [10.8–10.11]):

> "103. The exposure due to radionuclides incorporated into the organism is determined by the activity concentrations in the organism, the size of the organism, and the type and the energy of the emitted radiation. A key quantity for estimating internal doses is the absorbed fraction of energy $\phi(E)$, which is defined as the fraction of energy emitted by a radiation source that is absorbed within the target tissue, organ or organism. In the simplest case, the organism is assumed to be in an infinite homogeneous medium and to have a uniform activity concentration throughout its body. The densities of the medium and the organism's body are assumed to be identical. Under these conditions, both internal (D_{int}) and external (D_{ext}) dose conversion coefficients (DCCS; the DCC is defined as either the absorbed dose or the absorbed dose rate, according to the circumstances, per unit activity concentration of the relevant radionuclide in the organism or medium) for monoenergetic radiation can be expressed as a function of the absorbed fraction…:
>
> $$D_{\mathrm{int}} = E \cdot \phi(E) \;\text{ and }\; D_{\mathrm{ext}} = E \cdot (1 - \phi(E)) \qquad (4)\text{"}$$

TABLE 10.5. REFERENCE ORGANISMS DEFINED IN FASSET/ERICA, AND REFERENCE ANIMALS AND PLANTS AS DEFINED BY THE INTERNATIONAL COMMISSION ON RADIOLOGICAL PROTECTION

ERICA reference organisms (examples)	ICRP reference organisms	Habitat	Mass (kg)
Terrestrial environment			
Detritivorous invertebrates (woodlouse)	n.a.[a]	On and in soil	1.7×10^{-4}
Insects	Bee	In air	5.9×10^{-4}
Lichen and bryophytes (bryophyte)	n.a.[a]	On soil	1.1×10^{-4}
Gastropods (snail)	n.a.[a]	On soil	1.4×10^{-3}
Grasses and herbs	Wild grass	On soil	2.6×10^{-3}
Soil invertebrates (earthworm)	Earthworm	In soil	5.2×10^{-3}
Amphibians (frog)	Frog	On soil	0.031
Bird eggs	Duck egg	On soil	0.050
Burrowing mammals (rat)	Rat	In soil	0.31
Reptiles (snake)	n.a.[a]	On soil	0.74
Wading birds (duck)	Duck	On soil	1.3
Large mammals (deer)	Deer	On soil	250
Trees	Pine tree	On soil	470
Shrubs	n.a.[a]	On soil	—[b]
Marine environment			
Phytoplankton	n.a.[a]	In water	6.5×10^{-11}
Zooplankton	n.a.[a]	In water	6.1×10^{-5}
Sea anemones/true corals	n.a.[a]	In water	1.8×10^{-3}
Macroalgae	Brown seaweed	In water	6.5×10^{-3}
Benthic molluscs	n.a.[a]	In water	1.6×10^{-2}
Polychaete worms	n.a.[a]	In water	1.7×10^{-2}
Vascular plants	n.a.[a]	In water	2.6×10^{-2}

TABLE 10.5. REFERENCE ORGANISMS DEFINED IN FASSET/ERICA, AND REFERENCE ANIMALS AND PLANTS AS DEFINED BY THE INTERNATIONAL COMMISSION ON RADIOLOGICAL PROTECTION (cont.)

ERICA reference organisms (examples)	ICRP reference organisms	Habitat	Mass (kg)
Pelagic fish	n.a.[a]	In water	0.57
Crustaceans	Crab	In water	0.75
Benthic fish	Flat fish	In water	1.3
Reptiles (marine turtle)	n.a.[a]	In water	140
Mammals	n.a.[a]	In water	180
Sea anemones/true corals (colony)	n.a.[a]	In water	200
Wading birds	Duck	In water	1.3
Freshwater environment			
Phytoplankton	n.a.[a]	In water	2.1×10^{-12}
Zooplankton	n.a.[a]	In water	2.4×10^{-6}
Crustaceans	n.a.[a]	In water	1.6×10^{-5}
Insect larvae	n.a.[a]	In water	1.8×10^{-5}
Vascular plants	n.a.[a]	In water	1.1×10^{-3}
Gastropods	n.a.[a]	In water	3.5×10^{-3}
Bivalve molluscs	n.a.[a]	In water	7.1×10^{-2}
Pelagic fish	Salmonid/trout	In water	1.3
Benthic fish	n.a.[a]	In water	1.5
Mammals	n.a.[a]	In water	3.9
Birds	Duck	In water	1.3
Amphibians	Frog	In water	3.1×10^{-2}

Source: See Ref. [10.7].

[a] n.a.: not applicable.

[b] —: data not available.

Ulanovsky et al. [10.11] find that:

> "The second equation is an approximation that — in a strict sense — only holds if the organism and the surrounding medium are of the same density and elemental composition."

UNSCEAR [10.4] continues:

> "104. Absorbed fractions for photon and electron sources uniformly distributed in soft-tissue spherical bodies immersed in an infinite water medium have been systematically calculated by Monte Carlo simulation [Ref. [10.10]]. The calculations covered a particle energy range of 10 keV to 5 MeV, a range for the mass of the body from 10^{-6} to 10^{3} kg….

…….

> "109. The approach was also applied to the calculation of the absorbed fractions for non-aquatic animals and their internal exposures. With the use of the absorbed fractions for spheres and the suggested re-scaling and interpolation techniques, a set of internal DCCs has been calculated for all reference animals and plants [Ref. [10.10]]."

In Ulanovsky and Pröhl [10.12], DCCs for external exposure are calculated separately for aquatic and terrestrial reference animals and plants. For aquatic organisms, which are immersed by water, external exposure is calculated according to equations above.

For terrestrial reference animals and plants, the estimation of external exposures is more complex, since soil, air and organic matter differ considerably in composition and density [10.11, 10.13]. Therefore, the derivation of DCCs is based on radiation transport simulated for monoenergetic photons by means of Monte Carlo techniques [10.11, 10.13]. Owing to the complexity of the processes and the variability of life forms, reference geometries, as defined by energy, contaminated media, and organism size and shape, are considered in detail by Taranenko et al. [10.13]. External exposures under conditions for which specific calculations have not been performed, can be estimated with sufficient accuracy by interpolation. The DCCs for ^{210}Po are summarized in Table 10.6 for terrestrial organisms, and in Tables 10.7 and 10.8 for aquatic organisms.

The values for internal exposure are given in units of dose rate per unit activity in the organisms (μGy/h per Bq/kg). The values are given for absorbed dose (i.e. radiation weighting factors to account for the effectiveness of the

TABLE 10.6. DOSE CONVERSION COEFFICIENTS FOR TERRESTRIAL REFERENCE ORGANISMS FOR ^{210}Po FOR INTERNAL AND EXTERNAL EXPOSURE

Reference organism (example)	Internal exposure				External exposure		
	DCC (µGy/h per Bq/kg)	f_1 (%)	f_2 (%)	f_3 (%)	In soil/ infinite (µGy/h per Bq/kg)	On soil/ planar (µGy/h per Bq/m^2)	On soil/ volume (µGy/h per Bq/kg)
Woodlouse	3.1×10^{-3}	100	0	0	4.6×10^{-9}	2.9×10^{-11}	1.7×10^{-9}
Bees	3.1×10^{-3}	100	0	0	—[a]	2.9×10^{-11}	1.7×10^{-9}
Terrestrial lichens	3.1×10^{-3}	100	0	0	—[a]	1.5×10^{-11}	9.1×10^{-10}
Terrestrial gastropods (snail)	3.1×10^{-3}	100	0	0	4.6×10^{-9}	2.9×10^{-11}	1.7×10^{-9}
Terrestrial grasses (wild grass)	3.1×10^{-3}	100	0	0	—[a]	6.5×10^{-11}	1.7×10^{-9}
Soil invertebrates (earthworm)	3.1×10^{-3}	100	0	0	4.5×10^{-9}	—[a]	—[a]
Terrestrial amphibians (frog)	3.1×10^{-3}	100	0	0	4.5×10^{-9}	2.9×10^{-11}	1.7×10^{-9}
Bird eggs (duck egg)	3.1×10^{-3}	100	0	0	—[a]	2.8×10^{-11}	1.7×10^{-9}
Terrestrial burrowing mammals (rat)	3.1×10^{-3}	100	0	0	4.3×10^{-9}	2.8×10^{-11}	1.7×10^{-9}
Terrestrial reptiles (snake)	3.1×10^{-3}	100	0	0	4.1×10^{-9}	2.7×10^{-11}	1.6×10^{-9}
Large terrestrial mammals (deer)	3.1×10^{-3}	100	0	0	—[a]	1.4×10^{-11}	8.6×10^{-10}
Trees (pine tree)	3.1×10^{-3}	100	0	0	—[a]	2.0×10^{-11}	1.4×10^{-9}
Terrestrial shrubs	—[a]	—[a]	—[a]	—[a]	—[a]	3.1×10^{-11}	1.6×10^{-9}
Terrestrial wading birds (duck)	3.1×10^{-3}	100	0	0	—[a]	2.6×10^{-11} 2.3×10^{-11}	1.6×10^{-9} 1.5×10^{-9}

Source: See Ref. [10.13].

[a] —: data not available.

TABLE 10.7. DOSE CONVERSION COEFFICIENTS FOR MARINE REFERENCE ORGANISMS FOR ^{210}Po FOR INTERNAL AND EXTERNAL EXPOSURE

Reference organism (example)	Internal exposure				External exposure
	DCC (µGy/h per Bq/kg)	f_1 (%)	f_2 (%)	f_3 (%)	DCC (µGy/h per Bq/L)
Marine phytoplankton	2.0×10^{-7}	100	0	0	3.1×10^{-3}
Zooplankton	3.1×10^{-3}	100	0	0	4.9×10^{-9}
Sea anemones/true corals	3.1×10^{-3}	100	0	0	4.8×10^{-9}
Macroalgae (brown seaweed)	3.1×10^{-3}	100	0	0	4.9×10^{-9}
Benthic molluscs	3.1×10^{-3}	100	0	0	4.7×10^{-9}
Vascular plants	3.1×10^{-3}	100	0	0	4.7×10^{-9}
Marine worms (polychaete worm)	3.1×10^{-3}	100	0	0	4.8×10^{-9}
Marine pelagic fish	3.1×10^{-3}	100	0	0	4.4×10^{-9}
Marine crustaceans (crab)	3.1×10^{-3}	100	0	0	4.3×10^{-9}
Marine benthic fish (flatfish)	3.1×10^{-3}	100	0	0	4.5×10^{-9}
Marine reptiles	3.1×10^{-3}	100	0	0	2.3×10^{-9}
Marine mammals (dolphin)	3.1×10^{-3}	100	0	0	2.2×10^{-9}

Source: See Ref. [10.13].

different radiation types in causing effects to biota are not applied, since the discussion on this topic is ongoing [10.14]).

To estimate the possible impact of different radiation qualities, the contributions of the different types of radiation to the DCCs for internal exposure are given separately in the tables. These contributions are quantified through the factors f_1, f_2 and f_3, respectively, where f_1 specifies densely ionizing radiation (alpha particles and spontaneous fission fragments), f_2 represents the fraction of low energy beta radiation (E < 10 keV), and f_3 gives the contributions of higher energy beta radiation (E > 10 keV) and gamma radiation of all energies.

TABLE 10.8. DOSE CONVERSION COEFFICIENTS FOR FRESHWATER TERRESTRIAL REFERENCE ORGANISMS FOR ^{210}Po FOR INTERNAL AND EXTERNAL EXPOSURE

Reference organism (example)	Internal exposure				External exposure
	DCC (µGy/h per Bq/kg)	f_1 (%)	f_2 (%)	f_3 (%)	DCC (µGy/h per Bq/L)
Freshwater phytoplankton	6.3×10^{-9}	100	0	0	3.1×10^{-3}
Freshwater zooplankton	3.1×10^{-3}	100	0	0	4.9×10^{-9}
Freshwater crustaceans	3.1×10^{-3}	100	0	0	4.9×10^{-9}
Freshwater insect larvae	3.1×10^{-3}	100	0	0	4.9×10^{-9}
Freshwater plants (vascular plant)	3.1×10^{-3}	100	0	0	4.9×10^{-9}
Freshwater gastropods	3.1×10^{-3}	100	0	0	4.8×10^{-9}
Freshwater molluscs (bivalve mollusc)	3.1×10^{-3}	100	0	0	4.6×10^{-9}
Freshwater pelagic fish (trout)	3.1×10^{-3}	100	0	0	4.3×10^{-9}
Freshwater benthic fish	3.1×10^{-3}	100	0	0	4.3×10^{-9}
Freshwater mammals (muskrat)	3.1×10^{-3}	100	0	0	3.9×10^{-9}
Amphibians (frog)	3.1×10^{-3}	100	0	0	4.7×10^{-9}
Wading birds (duck)	3.1×10^{-3}	100	0	0	4.2×10^{-9}

Source: See Ref. [10.13].

Consequently, any user-defined radiation weighting factors w_{r} can be applied to these fractions to calculate radiation weighted DCCs:

$$D_{\mathrm{int}}^{\mathrm{wt}} = D_{\mathrm{int}} \sum w_r \cdot f_r \tag{10.1}$$

where D_{int} represents the DCC as provided in Tables 10.6–10.8.

The discussion on the derivation of radiation weighting to be applied in dosimetry for flora and fauna is still ongoing. Radiation weighting factors for alpha radiation are reviewed in Ref. [10.14]. For screening models being applied to obtain conservative results, radiation weighting factors of 10 and 20 for deterministic and stochastic end points, respectively, were considered to be appropriate to obtain conservative results. To evaluate radiological impacts on flora and fauna, deterministic end points are relevant [10.7].

Values for external exposures for aquatic organisms are given for a total immersion in water in units of μGy/h per Bq/L. For terrestrial organisms, various different source geometries are considered, depending on the habitat of the organism considered [10.13]:

(a) For on or above soil organisms, two geometries are considered:
  - A planar source, the source being at a depth of 0.3 cm to account for surface roughness (μGy/h per Bq/m^2);
  - A 10 cm thick volume source (soil density: 1.6 g/cm^3)(μGy/h per Bq/kg).

(b) For in soil organisms, the values are given for organisms living in the middle of a volume source with a thickness of 50 cm (soil density: 1.6 g/cm^3) (μGy/h per Bq/kg).

The decay properties are reflected in the contributions of alpha, beta and gamma radiation as quantified in Tables 10.6–10.8 by f_1, f_2 and f_3, respectively. Only alpha radiation contributes to the internal exposure. Due to the short range of alpha particles, external exposure due to ^{210}Po in the environment is negligible [10.13]. Only very small organisms, with dimensions in the order of the range of alpha particles (ca. 100 μm), can be exposed externally [10.13]. The short range of alpha particles means that all energy emitted is absorbed locally. Therefore, the DCCs for internal exposure are the same for all organisms, with the exception of very small organisms, with dimensions that are in the order of the range of alpha particles [10.13].

10.2.3. Non-homogeneous distribution

The DCCs for internal absorbed dose rate are calculated assuming a homogeneous distribution of radionuclides within the body [10.15]. However, this is often not the case: for example, iodine accumulates in the thyroid, polonium in the liver and transuranic elements in the kidney. The influence of inhomogeneous distributions on the average whole body dose is investigated by Gómez-Ros et al. [10.15]. In the case of ^{210}Po, for which only alpha radiation is relevant for internal exposure, the exposure due to ^{210}Po in tissues and organs is directly proportional to the ^{210}Po activity in the organ.

When a radionuclide accumulates in a specific organ, the dose will be higher than the whole body average. The dose in the specific organ is enhanced by a factor that can approximately be estimated from the ratio of whole body mass to organ mass.

10.2.4. Application of dose conversion coefficients

The DCCs can be used to estimate external and internal exposures for organisms living in different habitats. The total dose is calculated as the sum of external and internal exposure. The internal exposure of a terrestrial or aquatic organism (D_{int}) is estimated from the activity concentration in soil or water (C_s, C_w), concentration ratios ($\text{CR}_{\text{soil-organism}}$, $\text{CR}_{\text{water-organism}}$) and the DCC for internal exposure according to:

$$\text{Terrestrial organisms: } D_{\text{int}} = C_s \cdot \text{CR}_{\text{soil-organism}} \cdot \text{DCC}_{\text{int}} \tag{10.2}$$

$$\text{Aquatic organisms: } D_{\text{int}} = C_w \cdot \text{CR}_{\text{water-organism}} \cdot \text{DCC}_{\text{int}} \tag{10.3}$$

Dose rates can also be estimated directly from measured radionuclide concentrations in biota. External exposures are directly estimated from the activity in relevant media and the appropriate DCC. Some organisms move between different habitats owing to their lifestyle or in different stages of development. This effect can be accounted for through a simple superposition of the DCCs for isolated habitats.

Ulanovsky and Pröhl [10.12] provide a simple example: an aquatic benthic organism is considered that lives on the interface of water and sediment. Assuming that sediment density and composition are close to those of water, and that the radionuclide concentrations in water C_w and sediment C_s are known, the external dose to such an organism can be calculated according to:

$$D_{\text{ext}} = (0.5C_w + 0.5C_p)\,\text{DCC}_{\text{ext}} \tag{10.4}$$

where D_{ext} is the DCC for the radionuclide in water. The factor of 0.5 is to account for the habitat at the interface of water and sediment; the organism is exposed to 50% of the radionuclides in sediments and water, respectively.

A more complex exposure has to be considered, for example, for a wading bird, such as a duck, that lives in three different habitats: on soil, above soil and on the water surface [10.12]. For assessing the total external exposure of a duck, the time spent in these three habitats has to be known.

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Appendix I

GEOCHRONOLOGICAL APPLICATIONS OF POLONIUM

I.1. INTRODUCTION

The systematics of the decay of ^{210}Pb to ^{210}Po (through the decay of ^{210}Bi) can be used to determine ages and residence times over several half-lives of ^{210}Po, in the order of approximately 500 days. This can be applied to situations in which ^{210}Po is strongly fractionated from ^{210}Pb so that substantial disequilibrium is generated.

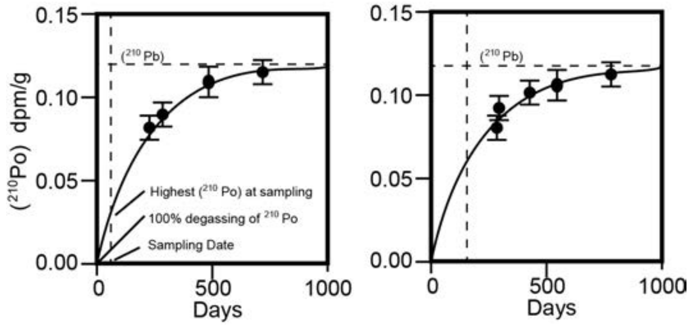
I.2. DATING VOLCANIC ROCKS

There have only been a few examples of dating using ^{210}Po. The method was first developed for, and mostly applied to, volcanic rocks, especially submarine basalts where volcanic events are not so readily observed. In a rock that forms incorporating ^{210}Pb and ^{210}Po in disequilibrium, the ^{210}Po will evolve according to the following equation [I.1]:

$$\left({}^{210}\mathrm{Po}\right)=\left({}^{210}\mathrm{Po}\right)_0 \mathrm{e}^{-\lambda_{^{234}\mathrm{Po}}t}+\left({}^{210}\mathrm{Pb}\right)\left(1-\mathrm{e}^{-\lambda_{^{234}\mathrm{Po}}t}\right) \tag{I.1}$$

For time periods substantially longer than the half-life of ^{210}Bi, the ingrowth of this intermediate nuclide can be ignored. Samples of recent volcanics are analysed for (^{210}Po) soon after sample collection, and subsequent analyses can be used to determine ^{210}Pb. An age can then be determined once an estimate of (^{210}Po)$_0$ has been made. This method was first adapted by Rubin et al. [I.1] to determine the age of mid-ocean ridge basalts from a volcanically active region. The method relies on disequilibrium within the basalt at the time of solidification and no subsequent gains or losses of ^{210}Pb or ^{210}Po. Since ^{210}Po can be completely volatilized at temperatures over 400°C, it might be expected that the samples completely lose ^{210}Po during eruption. However, the retention of some ^{210}Po cannot be discounted due to incomplete degassing as basalts are quickly cooled, especially under increased pressures at large ocean depths.

Rubin et al. [I.1] measure the (^{210}Po) in each sample over two years after collection (see Fig. I.1). The simple observation of ^{210}Po ingrowth demonstrates that the samples erupted within approximately two years. Through



Source: Figure 1 of Ref. [I.1].

FIG. I.1. Polonium-210 activities per gram in two samples of mid-ocean ridge basalts from the East Pacific Rise (reproduced with permission courtesy of Nature).

repeated analyses of each sample, best-fit ingrowth curves can be determined which defined the maximum value of (^{210}Po) (equal to (^{210}Pb)), and when (^{210}Po) = 0. This approach puts a greater constraint on ^{210}Po ingrowth than a simple determination of (^{210}Pb) once secular equilibrium is achieved. While the time of eruption is constrained between when (^{210}Po) = 0 and the time of collection, the actual eruption time could be between these if the basalt did not completely lose all ^{210}Po and $(^{210}\text{Po})_0 > 0$. The lowest measured value of (^{210}Po) is approximately 0.25 and represents the highest possible initial value. Age windows, corresponding to $(^{210}\text{Po})_0 = 0\text{–}0.25$, of up to several months are then obtained [I.1]. Rubin et al. [I.2] find that ^{210}Po–^{210}Pb dates of fresh basalts at a ridge section are consistent with eruption during recent recorded seismic events, and Johnson et al. [I.3] find the age of a seamount sample to be approximately five months. Basalt from Samoa is assumed to have completely lost ^{210}Po during subaerial eruption, and an age of around 260 days was determined [I.4].

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Appendix II

RADIOCAESIUM, ^{210}Po AND ^{210}Pb IN WOLVES

II.1. INTRODUCTION

Since the Chernobyl accident, ^{137}Cs ($T_{1/2}$ = 30.07 years) activity deposition in Sweden has varied in the range of 1000–100 000 Bq/m^2, and more than half of this activity has now decayed. The isotope ^{210}Po ($T_{1/2}$ = 138 days) originates from ^{222}Rn exhaled from the ground and its intermediate ^{210}Pb. The more even terrestrial distribution of ^{210}Po depends on the uranium content of the ground, the topography of the surrounding land mass and the amount of wet deposition. Persson [II.1] estimated the annual deposition of ^{210}Pb in central Sweden in 1970 to be around 63 Bq/m^2. The integrated activity of ^{210}Pb, supposing that the same amount is decaying as is introduced by precipitation, gives 2000 Bq/m^2. The ^{210}Po:^{210}Pb activity ratio in precipitation is around 0.1–0.2. The integrated deposition is then slightly lower for ^{210}Po than for ^{210}Pb, since it takes about two years for ^{210}Po to come into radioactive equilibrium with ^{210}Pb.

Polonium-210 and ^{137}Cs can accumulate in the food chain, and especially in reindeer, which can contain enhanced concentrations in soft tissue after feeding on lichen during the winter. A study by Gjelsvik et al. [II.2] published in 2014 measured activity levels of ^{137}Cs, ^{210}Po, and the ^{210}Po:^{210}Pb ratio in different tissues of 28 wolves in Sweden to determine the aggregated transfer factor (TF) and to calculate doses to the animals.

II.2. MATERIALS AND METHODS

Gjelsvik et al. [II.2] examined samples of muscle, blood, liver and kidney 28 wolves killed in Sweden in 2009 and 2010 (see Table II.1). The samples for analysis were prepared by drying and grinding, and ^{137}Cs was measured with high purity germanium gamma spectrometry. The ^{210}Po activity was determined with alpha spectrometry (ion implanted silicon detectors) after wet ashing and spontaneous deposition onto silver discs. The radiochemical yield determinant was ^{209}Po; and ^{210}Pb was analysed with dry incineration of a larger amount where Po evaporates. After six months, the buildup ^{210}Po from ^{210}Pb was analysed. The ingrowth rate gives the contribution of ^{210}Pb, and the ^{137}Cs and ^{210}Po concentrations were decay corrected to date of death.

TABLE II.1. DATA FOR SAMPLED WOLVES

Wolf	County	Community	Sex	Weight (kg)	Age (years)
1	Jämtland	Härjedalen	F	32.1	1
2	Jämtland	Härjedalen	M	34.9	1
3	Dalarna	Malung	M	40	0
4	Dalarna	Malung	M	41	1
5	Dalarna	Malung	F	28	0
6	Dalarna	Malung	F	29	0
7	Örebro	Degerfors	M	48	1
8	Norrbotten	Gällivare	M	47	1
9	Jämtland	Härjedalen	F	32.6	1
10	Gävleborg	Ockelbo	M	46.5	1
11	Gävleborg	Ockelbo	F	34	1
12	Dalarna	Malung-Sälen	M	47	6
13	Dalarna	Ludvika	M	28	0
14	Dalarna	Ludvika	F	36	4
15	Dalarna	Vansbro	M	38	2
16	Dalarna	Ludvika	M	—[a]	8
17	Dalarna	Ludvika	F	36	1
18	Gävleborg	Ljusdal	M	49	6
19	Västra Götaland	Mellerud	M	49.5	3
20	Värmland	Munkfors	F	40	1
21	Värmland	Sunne	M	32	0
22	Värmland	Eda	M	28	0
23	Örebro	Örebro	F	37.5	1
24	Värmland	Arvika	F	38	3
25	Örebro	Lindesberg	F	29	0
26	Värmland	Torsby	M	48	1
27	Värmland	Sunne	M	40	5
28	Västra Götaland	Dals-Ed	F	38	5

Source: Table 1 of Ref. [II.2].

[a] —:data not available.

II.3. RESULTS AND DISCUSSION

The activity concentrations of ^{137}Cs and ^{210}Po in different tissues from the wolves are Table II.2. The information comprises summaries from Refs [II.2–II.4].

After uptake, polonium mainly accumulates in the liver and kidneys, while caesium is more homogenously distributed in the body (but is lower in bone). Food is the major intake pathway for both caesium and polonium. For ^{210}Po, there is also in vivo buildup from ^{210}Pb. Reindeer have enhanced levels of both ^{137}Cs and ^{210}Po, especially after winter due to the consumption of lichen. However, during the summer, 74% of the wolf diet consists of moose. The aggregated TF for ^{210}Po shows large variations (0.000 5–0.27) depending on the organ (see Table II.2). Gjelsvik et al. [II.2] report that:

> "The bio-kinetics of polonium in the body is rather complicated. The fractional uptake by man has been reported from 0.1 to 0.8. The biological residence time has been reported to be between 20 and 80 days [Ref. [II.5]]. Thomas et al. [Ref. [II.3]] have studied the transfer of ^{210}Po and ^{210}Pb through the lichen–caribou–wolf food chain of northern Canada where caribou was the main food item for the wolves. Their results are in good agreement with our data. Thomas et al. [Ref. [II.3]] reported ^{210}Po/^{210}Pb activity ratios of 30–56 for liver and kidney. Fuller [Ref. [II.6]] estimated that the average daily intake was 2.7 kg per day when eating caribou and 5.4 kg per day when eating moose."

The fresh weight activity concentrations are: 7.5–13 Bq/kg in muscle; 145–369 Bq/kg in the liver; 142–343 Bq/kg in the kidneys; and 20–29 Bq/kg in bone. The lower values are from a region with less ^{210}Po in lichen and is thus reflected in the food chain. Gjelsvik et al. [II.2] continue that:

> "Howard et al. [Ref. [II.7]] estimated the transfer coefficient F_{f} (d kg^{-1}) of ^{210}Po from caribou to wolves as the ratio of activity concentration in the muscle of wolf (Bq kg^{-1} w.w.) divided by the daily amount intake of caribou meat (kg w.w. d^{-1}) with the activity concentration of ^{210}Po in the quantity Bq kg^{-1} w.w. which gave a value of 0.089 d kg^{-1} w.w."

Using the same coefficient for the Swedish wolves, the food would contain 8–170 Bq/kg (DW). The caribou from Canada contained 30–50 Bq/kg (DW) in muscle, 500–1300 Bq/kg (DW) in the kidneys and 550–1400 Bq/kg (DW) in the liver, depending on the region [II.3]. In the muscle of moose from Finland, sampled in March, the concentrations were 25 Bq/kg and in reindeer, 45 Bq/kg [II.4]. Persson [II.8] reported in 1972 concentrations

TABLE II.2. SUMMARY OF DATA ON ^{137}Cs, ^{210}Po AND ^{210}Pb IN WOLVES

Matrix	^{137}Cs (Bq/kg)	^{210}Po (Bq/kg)	^{210}Po:^{210}Pb	Remarks	Ref.
Blood[a]	—[b]	17	19	Canada, Snowdrift, 1993	[II.3]
	4–959	2.2–54	2.3–54	Sweden, 2010–2011	[II.2]
Bone[c]	—[b]	4.2–25	0.29–0.7	Canada, Baker Lake, 1993	[II.3]
	—[b]	20	0.6	Canada, Snowdrift, 1993	[II.3]
	—[b]	4.8	0.26	E. Finland, 1968	[II.4]
	—[b]	11–67	0.29–0.7	N. Finland, 1968	[II.4]
Liver[d]	—[b]	1 400	58	Canada, Baker Lake, 1993	[II.3]
	—[b]	551	30	Canada, Snowdrift, 1993	[II.3]
	—[b]	6.8	22	E. Finland, 1968	[II.4]
	—[b]	49–310	26–115	N. Finland, 1968	[II.4]
	36–4 050	20–523	9.4–56	Sweden, 2010–2011	[II.2]
Kidney[d]	—[b]	1 300	58	Canada, Baker Lake, 1993	[II.3]
	—[b]	539	28	Canada, Snowdrift, 1993	[II.3]
	31–3 453	24–942	1.8–7.9	Sweden, 2010–2011	[II.2]
Muscle[d]	—[b]	49	—[b]	Canada, Baker Lake, 1993	[II.3]
	—[b]	29	—[b]	Canada, Snowdrift, 1993	[II.3]
	350	1.2	57	E. Finland, 1968	[II.4]
	270–7 000	0.3–1.0	30–80	N. Finland, 1968	[II.4]
	70–8 410	1–43	2–77	Sweden, 2010–2011	[II.2]

	Aggregated transfer factor (Po·kg^{-1}·m^{-1})	Ref.
Liver	0.012–0.23	[II.2]
Kidney	0.001–0.27	
Muscle	0.000 5–0.021 0.01–0.26	

Source: Adapted from Ref. [II.2].

[a] Bq/L.

[b] —: data not available.

[c] Bq/kg, fresh weight.

[d] Bq/kg, dry weight.

in the muscle of Swedish reindeer as 16–48 Bq/kg, 28–220 Bq/kg in the kidneys and 88–148 Bq/kg in the liver. Kauranen et al. [II.4] observed in 1971 lower concentrations in wolves, lynx and wolverine in southern Finland compared to northern Finland [II.2].

The gastrointestinal uptake of ^{210}Po is higher than ^{210}Pb. Consequently, the ^{210}Po:^{210}Pb activity ratio increases with level in the food chain. In lichen, the ratio is slightly less than 1, and in reindeer about 5 in the liver and 20 in muscle [II.8]. Thomas et al. [II.3] find ^{210}Po:^{210}Pb activity ratios of 28–58 for the liver and kidneys of wolves, which is in fair agreement with the the data in Ref. [II.2].

In the south of Sweden, moose is the major food item for wolves. The activity concentration is generally higher in reindeer than in moose but in the north the concentrations do not differ much. However, the data do not support the tendency for higher concentrations in northern Sweden in contrast to the results in Ref. [II.4] for Finland.

Rather high concentrations of ^{137}Cs are found in areas with high deposition from the Chernobyl accident, such as in Gävleborg county. However, wolves move long distances to find their prey.

Gjelsvik et al. [II.2] report that the activity ratio of ^{210}Po:^{137}Cs in liver is in the range of 0.011–4.0, in muscle 0.002–0.05 and in kidneys 0.012–2.1. This differential bio-distribution shows the much larger accumulation of ^{210}Po in the liver and kidneys relative to ^{137}Cs. The large spread in the data is due to the variability in ^{137}Cs deposition in different areas and also reflects the range over which wolves travel to obtain prey.

The dose conversion factors in humans for ^{137}Cs and ^{210}Po are 1.2×10^{-8} Sv/Bq and 7×10^{-7} Sv/Bq, respectively. This is for a person with a body weight of 70 kg and using a quality factor of 20 for ^{210}Po. The dose is not applicable to animals, given the lack of specific tissue weighting factors for non-human biota, and only the absorbed dose in grays can be calculated. The absorbed dose depends on the biological residence time, organ distribution and, for ^{137}Cs, the body weight as well (a substantial fraction of the gamma energy from 662 keV photons escapes from the body depending on body mass). Gjelsvik et al. [II.2] report that:

> "The biological half-time of caesium in the human body is about 70 days but is supposed to be shorter for wolves both for caesium and polonium. Under an equilibrium situation doses can be calculated even if the biological half-time is not known but this is not possible under a dynamic situation. We suppose a stable condition and 35 days half-life. The maximal dose from ^{137}Cs to a wolf would be 3000 μGy per year…. The maximal dose from ^{210}Po to liver would be 2600 μGy per year."

For estimating the aggregated TFs for ^{137}Cs, charts of known deposition in Sweden from the Chernobyl accident should be used. Estimates of the aggregated TF can be as high as 0.1 but with large variations.

II.4. SUMMARY

There are significantly high activity concentrations of caesium and polonium in wolves, and the concentrations are higher in areas where reindeer are a significant part of the diet. Wolves are believed to prefer organs such as the liver and kidneys, which contain higher quantities of polonium. However, the proportion of different organs eaten is less important for ^{137}Cs, since it is more homogenously distributed in the body. A high Po:Cs ratio might indicate that moose is a major food source. Particularly high activity concentrations of ^{137}Cs are found in tissues from wolves from areas with high fallout from the Chernobyl accident. Further studies of wolves from the summer season could yield important information on food habits.

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Appendix III

^{210}Po IN MARINE MAMMALS

III.1. INTRODUCTION

Marine mammals encompass 125 species in 20 families that merge in 5 different mammalian groups [III.1]:

(a) Cetaceans (whales, dolphins and porpoises);
(b) Sirenians (manatees and dugongs);
(c) Pinnipeds (sea lions, walruses and seals);
(d) Marine and sea otters;
(e) Polar bears.

These marine mammalian groups are more closely related to one or another terrestrial mammalian group (e.g. horses, bears or elephants) than they are to each other. However, all marine mammals have one thing in common: they derive virtually all of their food from the marine environment [III.1–III.3]. Jefferson et al. [III.1] report that:

> “Marine mammals are not randomly distributed in the world’s oceans. It has long been known, for example, that certain species are found exclusively or primarily in waters of a particular depth, temperature range, or oceanographic regime, and not in areas lacking one or all of these characteristics. For most species, however, little is known of the particular factors that cause them to be found in one area and not in another that appears, qualitatively at least, the same.
>
> “One major factor affecting productivity, and thus indirectly influencing the distribution of marine mammals, is the pattern of major ocean currents.
>
> …….
>
> “Wherever oceanic conditions promote high nutrient content, it is likely that some species of marine mammal will be present to exploit that richness. Thus, the presence of marine mammals and other high order predators and consumers in an area is related primarily to prey, and secondarily to the water conditions supporting that productivity.”

Mammals do not substantially absorb chemical elements, including radionuclides, directly from sea water, as do fish through the gill epithelium. The mammalian diet is exclusively based on common marine species, and the large majority of radionuclide intake takes place through ingestion and inhalation.

III.2. MARINE MAMMAL FOOD CHAINS

In general, marine mammals are at the top of marine food chains, for example: the blue whale, the largest animal, feeds on zooplankton (krill); the sperm whale feeds on large size pelagic cephalopods and deep-sea sharks; the dolphins of coastal seas (*Delphinus delphis* and other Delphinidae) feed on common coastal fish species such as Clupeidae and Carangidae fish; sea otters and seals feed on crustaceans, bivalves and gastropods; and the killer whale feeds on sea otters and penguins. In their environments, marine mammals occupy different trophic levels (planktivorous, piscivores and carnivores) and often occupy the top position in their food webs, but marine mammals may, in turn, have humans, sharks and bears, as well as larger cetaceans and pinnipeds as predators [III.3, III.4].

Owing to their position in marine food webs and following international conventions on enhancing the protection and conservation of marine mammals, studies have been conducted on several species to investigate contamination by pesticide residues, polychlorinated biphenyls, heavy metals and other toxicants, as well as on reproductive success and health impairment (e.g. Refs [III.5, III.6]). Several reports have also covered the accumulation of radionuclides in marine mammals. The radiosensitivity of marine mammals is often comparable to that of terrestrial mammals. Thus knowledge on food chain transfer and radionuclide absorption into internal organs can be used as a proxy for radionuclide accumulation and radiation effects in other mammals, including humans [III.7–III.9].

III.3. BIOACCUMULATION AND BIODISTRIBUTION

Although there have been limited studies on radioactivity in marine mammals, current knowledge includes data on ^{210}Po activity concentrations for some species as well as computed whole body radiation doses from from ^{210}Po and several other natural and artificial radionuclides [III.10–III.13]. Most ^{210}Po analyses on marine mammals have been performed on samples from beach stranded animals or on samples from by-catch netted in tuna fishing gear, which explains the scattered nature of data, both in species and geography.

In 1974, Cherry and Shannon [III.14] published a review of the few results for ^{210}Po reported. Since then, more data have been published ,but they do not yet encompass most marine mammal families and, furthermore, many reports focus on determination of artificial radionuclides. This is the case, for example, for reports on: ^{137}Cs and $^{239+240}$Pu in harbour porpoises and seals with regard to local radioactive waste discharges from Sellafield, the United Kingdom; temporal trends of global radioactive fallout from nuclear weapon tests in seals from Lake Baikal, the Russian Federation; and ^{137}Cs geographical distribution in oceans and specific coastal areas (such as Antarctic seas) determined in dolphins, porpoises and seals (see Refs [III.10, III.12, III.15–III.20]).

Less attention has been given to measuring naturally occurring radionuclides in marine mammals, with the exception of ^{40}K, which has often been used to account for the natural radiation background and for comparison with internal radiation dose received from artificial radionuclides [III.10, III.13, III.18]. However, current concerns regarding radiation protection of non-human biota and the definition of reference species for environmental radiological risk assessment has tilted radioecologists' research interests to increasing knowledge about the actual radiation dose received by biota [III.7–III.9, III.21–III.23]. Furthermore, it has been recognized that, in marine organisms at least, the radiation dose from internally deposited alpha emitters, such as ^{210}Po, is much larger than that from internally deposited gamma emitters, such as ^{137}Cs and other artificial radionuclides [III.24, III.25].

The results on ^{210}Po in internal tissues of marine mammals show large inter-individual variation. In the same species, and for individuals collected in the same season and same area, ^{210}Po concentrations in muscle tissues vary over relatively wide ranges. For example, in the common dolphin of the north-eastern Atlantic Ocean, ^{210}Po concentrations (FW) are in the range of 9–87 Bq/kg, averaging 56 Bq/kg [III.13]. A wide activity concentration (FW) range of 48–102 Bq/kg is also found in the South Atlantic Franciscana dolphin, averaging 66.1 Bq/kg [III.26]. Of all marine mammals analysed so far, ^{210}Po concentrations in muscle fall in the range of 1–102 Bq/kg (FW) (see Table III.1).

Polonium-210 concentrations (FW) in other organs are higher than in muscle, averaging 123 Bq/kg in the liver and 110 Bq/kg in the kidneys of the north-eastern Atlantic common dolphin [III.13], and are even higher in the liver of the South Atlantic Franciscana dolphin, averaging 315 Bq/kg [III.26] (see Table III.1).

Analysis of the data indicates that in each marine mammal species, the highest ^{210}Po concentrations are consistently in the liver and kidneys, followed by muscle. Lower concentrations are measured in mammary glands, gonads, bone and fat tissue (see Table III.1). In each species, the liver to muscle ^{210}Po concentration ratio is always much higher than unity and among individual

TABLE III.1. ACTIVITY CONCENTRATIONS OF ^{210}Po IN MARINE MAMMALS

Region	Species	^{210}Po (Bq/kg, FW)	Ref.
NE Atlantic	Common dolphin		[III.13]
	Muscle	56 ± 32	
	Liver	123 ± 42	
	Kidney	110 ± 49	
	Gonad	11 ± 2	
	Fat tissue	11 ± 11	
	Bone	4.63 ± 0.12	
	Mammary gland	29.5 ± 0.9	
	Striped dolphin		[III.13]
	Muscle	42.0 ± 0.8	
	Mammary gland	8.2 ± 0.2	
	Pilot whale		[III.13]
	Muscle	13.7 ± 0.3	
	Fat tissue	0.70 ± 0.06	
North Atlantic	Sperm whale		[III.25]
	Muscle	5.0 ± 0.2	
South Atlantic	Franciscana dolphin		[III.26]
	Muscle	66.1 ± 18.9	
	Liver	315 ± 168	
	Guiana dolphin		[III.26]
	Muscle	24.6 ± 17.2	
	Liver	243 ± 213	
	Clymene dolphin		[III.26]
	Muscle	15.7 ± 1.1	
	Atlantic spotted dolphin		[III.26]
	Muscle	37.1 ± 36.4	
	Liver	125.7 ± 6.6	
	Bottlenose dolphin		[III.26]
	Muscle	5.8 ± 6.3	
	Liver	180.5 ± 244.0	

TABLE III.1. ACTIVITY CONCENTRATIONS OF ^{210}Po IN MARINE MAMMALS (cont.)

Region	Species	^{210}Po (Bq/kg, FW)	Ref.
South Atlantic	Common dolphin		[III.26]
	Muscle	9.34 ± 0.7	
	Liver	54.3 ± 3.2	
	Common dolphin[a]		[III.27]
	Muscle	86 ± 2	
	Liver	122 ± 0.4	
	Kidney	180 ± 71	
	Gonad	3.6 ± 0.5	
	Fat tissue	6.4 ± 0.6	
	Bone	26 ± 0.5	
	Whale[a]		[III.27]
	Muscle	1.1 ± 0.03	
	Liver	18 ± 0.3	
	Kidney	43.1 ± 0.9	
	Gonad	1.8 ± 0.1	
	Bone	2.5 ± 0.2	
Pacific	Common dolphin		[III.28]
	Muscle	4.5	
	Sperm whale		[III.29]
	Muscle	23.3 ± 0.4	
Atlantic	Fin whale		[III.29]
	Muscle	1.9 ± 0.1	
	Kidney	9.07 ± 0.30	
	Fat tissue	1.3 ± 0.1	
Arctic	Ringed seal		[III.11]
	Muscle	20.9	
	Liver	98.7	
	Kidney	82.7	
Bering Sea	Pacific walrus		[III.30]
	Muscle	28.7 ± 17	
	Liver	189 ± 157	
	Kidney	174 ± 85	

TABLE III.1. ACTIVITY CONCENTRATIONS OF ^{210}Po IN MARINE MAMMALS (cont.)

Region	Species	^{210}Po (Bq/kg, FW)	Ref.
Bering Sea	Bearded seal		[III.30]
	Muscle	27.1 ± 2.3	
	Liver	207 ± 30.4	
	Kidney	128 ± 20.5	
Swedish coast	Grey and harbour seals		[III.19]
	Muscle	2.5–20	
	Liver	6–63	
	Kidney	5–65	
	Gonad	0.5–1.8	
Greenland, Bylot Sound	Seal		[III.31]
	Muscle	8.1 ± 0.8	
	Liver	43 ± 4	
Antarctica	Leopard seal		[III.15]
	Muscle	1.6 ± 0.1	
	Liver	11.5 ± 0.3	
	Kidney	15.8 ± 0.4	
	Crabeater seal		[III.15]
	Muscle	1.3 ± 0.2	
	Liver	22.5 ± 3.3	
	Kidney	11.4 ± 2.4	
	Weddell seal		[III.15]
	Muscle	5.0 ± 2.5	
	Liver	48.8 ± 22.5	
	Kidney	33.0 ± 11.7	

Note: The arithmetic mean and standard deviation are recalculated from the original data.

[a] Converted to fresh weight using dry weight to fresh weight conversion factors determined in dolphins, as follows: muscle: 0.31; liver: 0.25; kidney: 0.22; gonad: 0.22; fat tissue: 0.64; bone: 0.47; mammary gland: 0.24 [III.13].

dolphins, this ratio varies from 2 to 39 [III.26]. High ^{210}Po concentrations in the liver are thought to reflect ^{210}Po absorption from recently ingested food (within a few hours), as in terrestrial mammals. From human and animal data, it can be inferred that one week after intake the liver retains, on average, 30% of the ^{210}Po absorbed into the blood, while 8–10% is retained in the kidneys, 9–10%

in blood and 5–6% in the skeleton [III.32]. Polonium-210 in the muscle and in systemic circulation is mostly derived from ingestion as well. Bone is the long term site for deposition of ^{210}Pb, which decays into ^{210}Po. Concentrations of ^{210}Pb and ^{210}Po in dolphin bone were measured in near radioactive equilibrium [III.13]. It is interesting to note that radionuclide distribution modelling in the ringed seal concluded that probably not more than 1% of the ^{210}Po in the ringed seal soft tissue would be from ingrown ^{210}Po derived from decay of ^{210}Pb deposit in the bone [III.11].

Polonium-210 activity concentration determined in the dorsal muscle of dolphins is always sampled in the same muscle and body zone, and is therefore comparable. It shows an increase with dolphin body length and thus with age [III.13], which suggests a steady increase of ^{210}Po body burden, probably due to the higher ingestion rate by larger individuals and increasing contribution of ^{210}Po from ^{210}Pb in the bone as the animal ages, such as observed in humans (see Chapter 9). In dolphins and whales, other authors have reported an allometric variation in ^{210}Po concentration, with lower ^{210}Po in larger animals [III.27]. However, body size is not the only parameter which influences ^{210}Po concentration in marine organisms. A relationship was also identified between high ^{210}Po concentrations in muscle and muscle tissue myoglobin content, ranging from low in white muscle to high in red muscle [III.27]. This is likely due to a strong association between ^{210}Po and metal binding proteins (such as found with ferritin in fish), contributing to enhancing ^{210}Po accumulation in red muscle tissue compared to white muscle tissue [III.33].

Among individuals of the same species, such as observed in the common dolphin, the variable ^{210}Po concentrations in muscle and other internal organs might be dependent on diet composition and ^{210}Po assimilation efficiency from recently ingested food. This receives strong support from previous studies in other marine species, such as fish and shrimp [III.34, III.35], as well as in humans [III.36].

The diet of marine mammals is not constant every day and changes over the lifespan of the species and to vary from one feeding ground to another. In spite of this, ^{210}Po ingestion is still the main source of ^{210}Po accumulated in internal organs. Therefore, the differences in average ^{210}Po concentrations between species might reflect the different diets of marine mammals. With this assumption in mind, ^{210}Po concentrations (FW) in muscle tissue are: 10–100 Bq/kg in dolphins; 2.5–20 Bq/kg in seals off the Swedish coast; 5 Bq/kg in sperm whales; and as low as 1.1 Bq/kg in South Atlantic whales (see Table III.1). All of these mammals have different food chains which could assist data interpretation. However, data interpretation is difficult because samples have origins in different oceanic regions and ecosystems, and were obtained at different times. This is important because whales, for example, starve during migration. Furthermore, in most cases the exact muscle sampled was not recorded or described, and ^{210}Po results for each species have different statistical weight, in many cases being represented by only a single sample.

III.4. REGIONAL DIFFERENCES

Consistent datasets for the same ecosystem are even more scant. For the Antarctic Ocean food chain, however, Roos et al. [III.15] report ^{210}Po concentrations in muscle tissue for single individuals of the Weddell seal, the crabeater seal and the leopard seal of 16, 4.1 and 5.0 Bq/kg (FW), respectively. In the Antarctic food web, the crabeater seal specializes in krill and other crustaceans; the leopard seal eats krill, fish and, on occasion, crabeater seals; and the Weddell seal feeds on fish, squid, krill and, infrequently, penguins [III.3]. While krill and other crustaceans generally display relatively higher ^{210}Po concentrations, penguins were reported to display relatively low ^{210}Po concentration in their flesh, 1.5–2.2 Bq/kg (FW) [III.15]. Therefore, the different ^{210}Po levels in these three mammal species seem to reasonably mirror their feeding habits.

Conversely, ^{210}Po concentrations could be expected to be similar in the same or related species from different oceanic regions when they occupy the same ecological niche and trophic level. This was the case for dolphins in the tropical South Atlantic and temperate north-eastern Atlantic Ocean [III.13, III.26].

Globally, ^{210}Po transfer in marine food chains seems to be associated with protein transfer and ^{210}Po concentration in consumer tissues mostly reflects ^{210}Po levels in the prey and thus the trophic level occupied by marine species [III.37]. Although ^{210}Po concentrations in mammals are also expected to reflect ^{210}Po food chain transfer and their trophic level, the low number of marine mammal species and specimens analysed so far is not sufficient to draw clear conclusions.

While polonium concentrations in marine mammal tissues are highly variable and depend on absorption from food, ^{40}K concentrations determined in tissues of porpoises, bottlenose dolphins and seals do not show wide inter-individual variation within the same species [III.10, III.13, III.20]. This is consistent with the homeostatic control of ^{40}K concentration in the internal organs of mammals.

The generally higher ^{210}Po concentrations in marine mammal tissue compared to terrestrial mammals, such as muscle tissue from cows, lambs and rabbits (see Chapter 6), is a direct consequence of the ^{210}Po concentration in the diet. While the diet of marine mammals is exclusively based on marine prey that consistently have ^{210}Po:^{210}Pb activity ratios well above unity, the diet of terrestrial mammals (which can be herbivorous, omnivorous or strictly carnivorous) is generally lower in ^{210}Po than in ^{210}Pb, at least for herbivorous and omnivorous species. Similarly, some human populations (e.g. in Japan and Portugal) have a diet rich in marine food ($^{210}Po:^{210}Pb > 1$ in the diet), with average ^{210}Po intake substantially higher than for populations with a continental diet ($^{210}Po:^{210}Pb < 1$) [III.38, III.39]. These divergent radionuclide intakes are likely to be reflected in ^{210}Po concentrations in human tissues and whole body burdens; however, this has not yet been evaluated (see Chapter 9).

Radiation doses due to natural (^{210}Pb, ^{210}Po and ^{40}K) and artificial (^{137}Cs) radionuclides accumulated in internal organs have been computed for some marine mammals. Due to its alpha radiation emission, ^{210}Po is the main contributor to the absorbed radiation dose and is estimated to account for more than 97% of the dose in dolphins [III.13, III.26, III.40].

In 1982, ^{210}Po activity concentrations in dolphin muscle tissue mostly fell in the range of 10–100 Bq/kg (FW), and are two to three orders of magnitude higher than the average activity concentration in tissues of the human body, 0.2 Bq/kg (FW) [III.41]. From internally accumulated ^{210}Po, the whole body radiation dose for dolphins is around 1.5 μGy/h and 1.9 μGy/h for *Delphinus delphis* and *Pontoporia blainvillei*, respectively. Such natural radiation doses received by these marine mammals (~15 mGy/a) are unusually high compared to humans who receive an average dose from internal ^{210}Po of about 6.2×10^{-4} μGy/h [III.26, III.37]. Nevertheless, such dose rates in marine mammals are still below the threshold radiation dose rate of 100 μGy/h, at which biological effects have been observed in other mammals [III.9, III.42].

Further study of ^{210}Po in marine mammals would be of great value, in particular to understand how they cope with relatively high radiation doses and to identify detoxification and repair mechanisms for genetic damage caused by radiation in internal tissues.

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Appendix IV

POLONIUM RELEASES FROM VEGETATION AND FOREST FIRES

IV.1. INTRODUCTION

Humans absorb ^{210}Po much more than other heavy radioactive elements, such as thorium and uranium [IV.1]. In the human body, the main origin of ^{210}Po is in food and water; however, it can also be accumulated from the air through inhalation and lung absorption [IV.2, IV.3] (see also Chapter 9). Early studies from the 1970s on ^{210}Pb and ^{210}Po activity concentrations in atmospheric aerosols reported the occurrence of higher than expected ^{210}Po activities and abnormally high ^{210}Po:^{210}Pb activity ratios (i.e. >1) [IV.4, IV.5]. Those reports often attributed these exceptional ratios to volcanic emissions (see Ref. [IV.5]). Enhanced ^{210}Po concentrations and ^{210}Po:^{210}Pb activity ratios were also attributed to releases from industrial sources and vegetation fires, but these were usually reported with scarce experimental evidence in support of that attribution [IV.4, IV.6]. Later work measuring ^{210}Pb and ^{210}Po in smoke from savannah fires in Africa showed that smoke particles can contain high ^{210}Pb and ^{210}Po levels and increase the usually low activity concentration ratios in surface air aerosols in the region [IV.7, IV.8].

The determination of radionuclides in effluent releases to the atmosphere (gases and particulate matter) from non-nuclear industries also reveal enhanced concentrations of ^{210}Po and occasionally high ^{210}Po:^{210}Pb ratios. These are reported, for example, for industries such as coal fired power plants, metal smelters and elemental phosphorus producing plants [IV.9]. High ^{210}Po concentrations and other naturally occurring radioactive material (NORM) in effluents from these industries are generally associated with relatively high temperatures used in their core processes.

In 1982, the United Nations Scientific Committee on the Effects of Atomic Radiation compiled what was known about radioactive emissions from NORM industries, pointing out that substantial enrichment of natural radioactive elements with low boiling point could be observed within the wastes generated in some of those industries [IV.2]. Later, reports were provided of the existence of high activity deposits and scales of certain volatile radioisotopes, particularly ^{210}Po, in filters, ash and on the surfaces of the furnaces and subsequent devices where the processes take place [IV.10–IV.13]. Mora et al. [IV.13] attempt to model the behaviour of ^{210}Po in these industries was made based on its physical properties, particularly its low melting point.

Morawska [IV.14] investigates burning wood in stoves for house heating as a source of contaminants to indoor air. The ^{210}Po concentrations are found to be enhanced in smoke from domestic wood combustion and to contribute to poor indoor air quality; although in modern houses the smoke is efficiently extracted to the atmosphere. Nonetheless, measurable concentrations of ^{210}Po are reported in association with soot and organic compounds in the extracted smoke [IV.15]. Enhanced ^{210}Po levels and elevated ^{210}Po:^{210}Pb ratios in aerosols in the atmosphere of Seoul, Republic of Korea, are associated with coal burning [IV.10].

High concentrations of ^{210}Po in cigarette smoke and enhanced radiation doses to the smokers' lung have been reported. Although tobacco leaves have moderate ^{210}Po concentrations, ^{210}Po volatilization and re-concentration in smoke particles are associated with the high temperature of tobacco combustion [IV.16–IV.19].

Altogether, evidence from investigations of NORM industries, wood combustion and cigarette smoke case studies has sufficiently underpinned the case for enhanced ^{210}Po activity concentrations occurring in particulates formed during the transformation of common raw materials that undergo high temperature processes. Despite this knowledge, assessment of ^{210}Po and other natural radionuclides released by forest and bush fires has been essentially under-investigated in atmospheric radioactivity research.

IV.2. VEGETATION AND FOREST FIRES

Wildfires burn many thousands of hectares of forest and bush around the world every year. Although global statistics of vegetation fires are far from complete, analysis of satellite data for 2000 revealed that 350 million ha were affected by vegetation fires [IV.20]. The plant biomass burned annually can vary according to the ecosystem affected, but reaches many millions of tonnes. This combustion releases considerable amounts of carbon, particles and gases (including toxic organic substances) to the atmosphere [IV.21–IV.23]. More than 200 volatile substances have already been identified in forest fire smoke, including toxic gases, such as carbon monoxide, formaldehyde, acrolein, hydrocarbon compounds and dioxins [IV.15, IV.23, IV.24]. Smoke particles also adversely affect the respiratory tract and their inhalation in areas impacted by forest fires have been connected to a higher frequency of asthma and other lung diseases [IV.23]. Furthermore, smoke rapidly travels around the globe and raises concerns about air quality in areas far from forest fires [IV.25].

Compared with what is known about the environmental stressors described in Section IV.1, the release of radionuclides by vegetation fires has been less investigated and attention has been focused almost exclusively on artificial radionuclides. One example is the work undertaken to evaluate resuspension of artificial radioactivity, such as ^{137}Cs, released by the Chernobyl accident, in 1986, and deposited onto soils and forests in Eastern Europe [IV.26–IV.31]. This resuspension was observed recently, following summer forest fires in Belarus and Ukraine. These forest fires reinitiated atmospheric transport processes to carry ^{137}Cs to central, northern and western European countries, although on a much smaller scale than in 1986 [IV.1, IV.32, IV.33].

However, besides radioactivity of artificial origin, vegetation fires are also likely to release naturally occurring radionuclides, such as ^{14}C and ^{40}K, and members of the uranium, thorium and actinium decay series [IV.1]. Plants absorb naturally occurring radionuclides through root and foliar uptake, and accumulate them throughout their tissues (see Chapter 6). So far, the radiological impact of smoke from forest or grass fires has scarcely been investigated [IV.32, IV.34]. Nevertheless, work done on ^{137}Cs resuspension by forest fires in Eastern Europe allows for comparison of measurements of naturally occurring radionuclides in surface air in Finland. The air activity concentrations of resuspended ^{137}Cs are of little radiological significance and are comparable to, or lower than, those of naturally occurring radionuclides in aerosol particles [IV.32]. Artificial transuranic radionuclides and total alpha radioactivity associated with smoke from prescribed fires have also been measured at the Savannah River Site and other woodland areas in the southern United States of America [IV.34].

With fire, natural radionuclides concentrated in plants are released to the atmosphere as smoke components. It is known that concentrations of naturally occurring radionuclides are generally low in vegetation. Atmospheric radionuclides, such as the long lived ^{222}Rn progeny, ^{210}Pb, ^{210}Bi and ^{210}Po, deposit onto the surface of leaves and bark, where the highest ^{210}Po concentrations in plants are usually measured. In the tree trunk and branch wood, ^{210}Po content is usually lower than in leaves [IV.35]. The burning of this wood still releases some ^{210}Po along with other compounds [IV.15]. More significant than combustion of tree wood are the ^{210}Po releases by grass and bush vegetation fires to the atmosphere (see Table IV.1).

Carvalho et al. [IV.1] hypothesize that the release of radionuclides from plant biomass can occur through two mechanisms:

> “One consists in the volatilization of elements [such as polonium] when the flame temperature exceeds the volatilization point of those elements. The other is the enrichment through enhancement of element concentrations in ash/smoke particles (refractory fraction) due to reduction of plant material mass with the volatilization of water, organic compounds, and elements of low volatilization point.”

TABLE IV.1. RADIONUCLIDE ACTIVITY CONCENTRATIONS IN PLANTS AND IN SMOKE FROM PLANT COMBUSTION (Bq/kg, DW)

	^{238}U	^{234}U	^{226}Ra	^{210}Pb	^{210}Po	^{232}Th
Leaves	—[a]	—[a]	—[a]	9.4	1.05	—[a]
Trunk	—[a]	—[a]	—[a]	4.3	0.14	—[a]
Smoke of:						
Eucalyptus wood	65 ± 5	88 ± 7	318 ± 65	52.2 ± 4	125 ± 10	35 ± 3
Acacia wood	11 ± 1	36 ± 4	503 ± 165	440 ± 19	611 ± 28	<29
Bush wildfire	169 ± 7	167 ± 7	2492 ± 639	369 ± 36	4422 ± 186	—[a]
Pinewood wildfire	204 ± 11	217 ± 12	3547 ± 1678	—[a]	3972 ± 157	117 ± 8

Source: Table 1 of Ref. [IV.1] (see also Refs [IV.24, IV.35]).

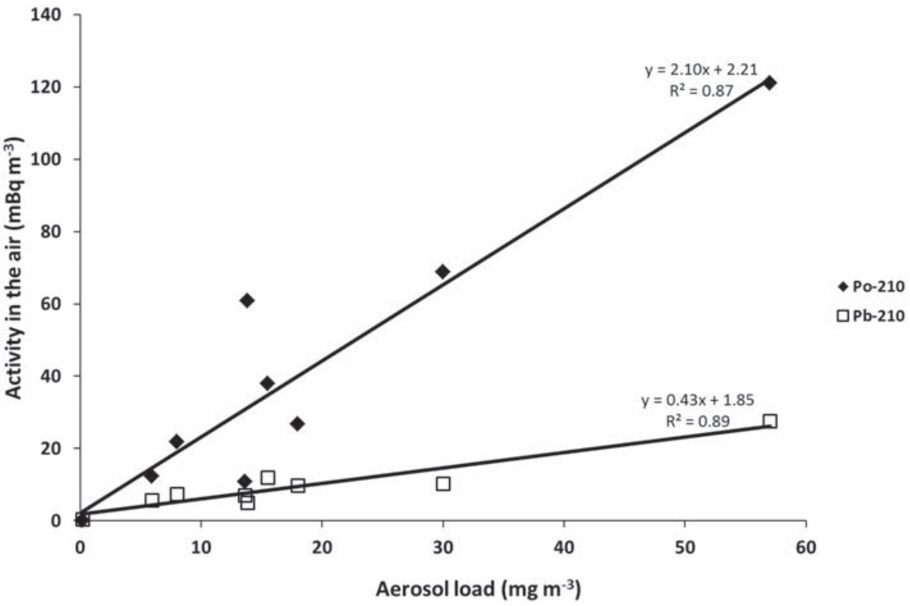
[a] —: data not available.

In contrast to the more refractory elements, such as uranium and thorium, Carvalho et al. [IV.1] report that polonium is volatile at lower temperatures and can be released to the atmosphere as gaseous compounds.

More research has recently been conducted on ^{210}Po and other natural radionuclides released from vegetation by wildfires [IV.24, IV.35]. Carvalho et al. [IV.1] report that in aerosols sampled in the vicinity of fires, "the radionuclide activities per unit of air volume was systematically higher with higher particle load in the air, suggesting that smoke particles collected on filters were the vehicle for transport of radioactivity in the atmosphere" (see Fig. IV.1). The ^{210}Po activity concentrations in smoke particles were enriched by a factor of 100–1000 over those in the vegetation on a mass basis [IV.24]. Furthermore, Carvalho et al. [IV.1] find (see also Refs [IV.35, IV.36]):

> "The proportions of radionuclides in the smoke were modified in comparison with background aerosols and with unburned plants. For example, the concentration ratios ^{210}Po/^{210}Pb in vegetation before the fire were often about 0.1, while in the smoke from vegetation fires this ratio increased up to 12, revealing an extraordinary enrichment of smoke particles in ^{210}Po."

Paatero et al. [IV.32] report that the ^{210}Po:^{210}Pb activity ratio in air increased by almost an order of magnitude when smoke particulates were present. Carvalho et al. [IV.36] determine ^{210}Po in smoke from vegetation fires was determined in several particle size classes of smoke aerosols sampled with a cascade impactor [IV.36]. This reveals an enrichment of the activity concentration of

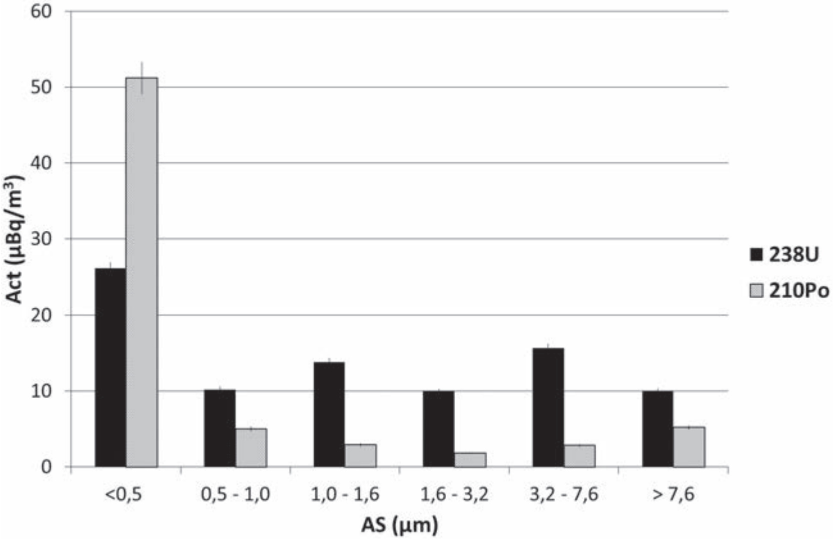


Source: Figure 1 of Ref. [IV.24].
Note: Correlations are statistically significant at $p < 0.01$.

FIG. IV.1. Aerosol load in surface air with smoke from vegetation fires, and ^{210}Pb and ^{210}Po radioactivity in the air (reproduced with permission courtesy of EIT Press).

radionuclides in aerosol particles collected near the vegetation fire compared to reference aerosol samples collected in other areas in the absence of vegetation fires [IV.1]. In the case of ^{210}Po, this enrichment is observed mostly in the smallest particles, with aerodynamic diameters less than 0.5 µm. In contrast, the distribution of refractory elements, such as ^{232}Th and ^{238}U, in smoke particle aerosols across the range of size classes show reasonable homogeneity [IV.1]. Owing to the much higher number of smaller particles (<0.5 µm) per unit volume, these are the main contributors for inhaled activity, particularly for ^{210}Po (see Fig. IV.2.)

Polonium atoms in the gas phase are positive ions and can easily be recaptured by electrostatic charges onto the aerosol particles, and smaller particles (coagulation nuclei) can gradually enter accumulation mode and later settle as coarse aerosol particles [IV.14]. Considering the ^{210}Po radionuclide distribution in aerosol fractions, inhalation of smoke in the vicinity of a vegetation fire can lead to inhalation of significant activity. Inhalation of up to 70 mBq/m^3, mostly due to the ^{210}Po fraction associated with very fine particles, has been estimated [IV.35]. Prolonged exposure to smoke from vegetation fires



Source: Figure 2 of Ref. [IV.36].

FIG. IV.2. Radionuclide concentrations in aerosol particles in the smoke from a forest fire sorted by aerodynamic size classes (AS) (reproduced with permission courtesy of Elsevier).

might lead to enhanced radiation doses to the lung, especially from ^{210}Po, that could be comparable or even higher than the radiation dose received by heavy cigarette smokers. This exposure to ionizing radiation is sufficient justification for recommending respiratory protection, in particular for firefighters and members of the public in areas heavily affected by smoke from vegetation and forest fires [IV.35, IV.36]. Increasing worldwide consumption of coal is also likely to enhance ^{210}Po levels in the atmosphere and its impact on public health should be carefully monitored [IV.10].

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Appendix V

POLONIUM IN TOBACCO AND DOSES TO HUMANS

V.1. INTRODUCTION

Tobacco (*Nicotina rustica*) is in the Solonaceae family, together with bell pepper (capsicum), tomato and potato, and there are many species in the tobacco genus, *Nicotiana*. It was originally grown as a crop in Central America and is named after the Caribbean island of Tobago. Most tobacco is smoked, but other forms exist, such as snuff and chewing tobacco. Nicotine, the active substance in tobacco, is present in lower doses in potato plants, tomato plants and, together with cocaine, in coca plants. It is named after a French ambassador to Portugal, Jean Nicot, who introduced snuff, taken through the nose, to the French court in 1560.

V.2. ^{210}Pb AND ^{210}Po IN TOBACCO

Inhalation of tobacco smoke is ranked second to food as a source of ^{210}Pb and ^{210}Po in humans (see Ref. [V.1] for a review), and there is a clear correlation between lung cancer and smoking, and evidence of a synergistic effect when smoking and elevated radon levels are combined [V.2–V.4]. There are several publications which deal with ^{210}Pb and ^{20}Po in tobacco and the transfer to humans (see Table V.1). Activity concentrations are in the range of 2.8–37 mBq/g. There are few data on activity concentrations in pipe tobacco, cigar tobacco and snuff. A cigarette contains about 1 g of tobacco.

When ^{210}Pb is also analysed, it is generally in radioactive equilibrium to ^{210}Po. In addition, while all results are of the same order of magnitude, the activity concentrations of ^{210}Pb and ^{210}Po in tobacco vary depending on the geographical region in which the tobacco is grown and harvested. If foliar deposition is the main source for the radionuclides, the ^{210}Po:^{210}Pb activity ratio should be lower than one at harvest. However, owing to the time elapsed from harvest to purchase, equilibrium has usually been attained in tobacco products.

TABLE V.1. EXAMPLES OF ^{210}Po ACTIVITY CONCENTRATIONS IN TOBACCO

Country	Activity per gram (mBq/g)	Activity per cigarette (mBq)	Ref.
Various	6.6–24	7.4–23	[V.4]
Egypt	12.6–24	9.7–21.8	[V.5]
Greece (tobacco leaves)	6–18	5–15	[V.6]
Poland	5.7–33	5.7–33.2	[V.7]
Portugal	2.8–37	2.6–27.7	[V.8]
Sweden (snuff)	6–60	—[a]	[V.9]
USA	13.3–20.7	12.6–17.8	[V.10]
Cigarettes	10.5–22		
Pipe tobacco	7.6–13.9	—[a]	
Cigar tobacco	10.5–30		
USA		12–18	[V.11]
Pipe tobacco	7–13	—[a]	
Cigar tobacco	17	—[a]	

[a] —: data not available.

V.3. TRANSFER TO HUMANS AND RADIOLOGICAL DOSES

The effective dose conversion factors for ^{210}Pb and ^{210}Po for inhalation by adults are 1.1×10^{-6} Sv/Bq and 3.3×10^{-6} Sv/Bq, respectively [V.12]. The dose conversion factors by ingestion are slightly lower, at 6.7×10^{-7} Sv/Bq and 1.2×10^{-6} Sv/Bq, respectively.

Additional studies from the 1960s include information on the radionuclide contents in the wrapping paper, filters, cigarette ash, post-smoking filters and cigarette ends, as well as in the mainstream and sidestream smoke, and that sometimes a smoking machine was used for the radionuclide budget in the different matrices [V.10, V.11] (see Table V.2).

There are large variations in the data. Apart from the variation of radionuclide concentrations in tobacco, the authors often have different estimates of the amount of smoke inhaled and the fraction of ^{210}Po retained in the lungs. Different dose conversion factors have been used where recommendations have changed over time. Sometimes, calculations are based on assumed lung

TABLE V.2. ACTIVITY CONCENTRATIONS IN DIFFERENT SMOKING MATRICES AND ANNUAL DOSES (SMOKING 20 CIGARETTES PER DAY) FROM ^{210}Po

Study	Activity per gram (mBq/g)	Dose (μSv/a)
Skwarzek et al. [V.7]		—[a]
Filter	0.1–4.3	
Paper	1.5	
Filter after smoking	0.3–9.1	
Ash only	0.43–14.7	
Ferri and Baratta [V.11]		165–460 (lungs)
Filter after smoking	1.1–2.0	
Inhaled	1.7–4.0	
End+ash	4.3–7.1	
Khater [V.5]		193 (effective dose)
Filter	0.9–9.7	
Paper	0.3–8.1	
Filter after smoking	1.5–12.6	
Ash only	2.2–35	
Carvalho and Oliveira [V.8]		420 (lungs)
Inhaled	0.97–1.52	
End+ash	0.3–4.9	
Black and Bretthauer [V.10]		—[a]
Inhaled	2.6	
End only	7.0	
Watson [V.4]	—[a]	5–220 (lung)
Papastefanou [V.6]	—[a]	47–135 (effective dose)

[a] —: data not available.

concentrations. The quality factor used to estimate dose from alpha radiation (now 20) is not always given in these studies. As for less common exposure routes, using tobacco as snuff gives a lower dose than smoking. It would be of interest to study the impact of ^{210}Po derived from using water pipes (hookahs).

A fraction of the radionuclides will transfer to the smoker and will also expose persons in the vicinity. Smokers have higher concentrations of these radionuclides in their lungs but also in other organs, such as the liver and kidneys (see Chapter 9). The dose to the lungs is much higher from smoking than from breathing outdoor ambient air, and a correlation exists between the prevalence of lung cancer in smokers with those living in houses with high radon concentrations.

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Appendix VI

ESTIMATION OF AEROSOL RESIDENCE TIMES IN THE ATMOSPHERE

VI.1. AEROSOL RESIDENCE TIME

The residence time (τ) for a radionuclide in a parcel of air (or any other environmental compartment) represents the average time an atom of that radionuclide is present in the air parcel before it is removed [VI.1]. If N is the total number of atoms of the radionuclide in the air parcel, I is the rate of introduction from sources (e.g. by ingrowth from a parent or transfer from another air parcel) and R is the rate of removal (e.g. by radioactive decay, or wet or dry deposition), then:

$$\frac{\mathrm{d}N}{\mathrm{d}t} = I - R \tag{VI.1}$$

If N is a constant, then steady state conditions hold, and $I = R$. In this case, the residence time is given by:

$$\tau = \frac{N}{R} = \frac{N}{I} \tag{VI.2}$$

In the case of the total amount of ^{222}Rn in the atmosphere, the only significant removal process is radioactive decay. Under the assumption that a steady state condition applies, then:

$$\tau = \frac{N}{\lambda N} = \frac{1}{\lambda} = 5.5 \text{ days} \tag{VI.3}$$

However, for a portion of the whole atmosphere, such as the planetary boundary layer (PBL), the average residence time for ^{222}Rn will be less than this due to mixing, which results in additional net removal from the PBL to the free troposphere. These residence times are not long in comparison with characteristic mixing times in the troposphere; as a result, ^{222}Rn is not well mixed either horizontally or vertically.

The ^{222}Rn progeny, ^{210}Pb, ^{210}Bi and ^{210}Po, are attached to aerosols in the atmosphere, and their behaviour reflects that of those aerosols. In the troposphere, aerosol residence times range from days to weeks, depending on the size of the aerosol, height above ground level and the prevailing meteorological conditions. In contrast, residence times in the stratosphere are of the order of one year [VI.1].

The degrees of disequilibrium between ^{222}Rn and ^{210}Pb, and between ^{210}Pb and its radioactive progeny, ^{210}Bi and ^{210}Po, have sometimes been used to calculate apparent atmospheric aerosol residence times [VI.2–VI.4]. For this purpose, concentrations of the pair (^{222}Rn, ^{210}Pb) in air can be used [VI.5]. For the pairs (^{210}Pb, ^{210}Bi) and (^{210}Pb, ^{210}Po), concentrations in air [VI.6–VI.8], in precipitation [VI.9–VI.11] and in dry fallout [VI.11] have been used.

In the case that ^{210}Pb, ^{210}Bi and ^{210}Po are produced solely by decay of ^{222}Rn in the atmosphere, that their production and removal are in steady state at the time and place of measurement, and that the removal term follows first order kinetics, then:

$$\frac{\mathrm{d}N_2}{\mathrm{d}t} = N_1\lambda_1 - N_2\left(\lambda_2 + \lambda_\mathrm{R}\right) = 0 \tag{VI.4}$$

where 1 is a parent radionuclide, 2 is its immediate daughter, λ_1 and λ_2 are their radioactive decay constants, and λ_R is the first order rate constant for removal of aerosols by all processes. The mean aerosol residence time is:

$$\tau_\mathrm{R} = \frac{1}{\lambda_\mathrm{R}} \tag{VI.5}$$

For the individual parent–progeny pairs, the apparent aerosol residence time is given by [VI.2, VI.5]:

$$\tau_\mathrm{R}\left(\mathrm{Rn, Pb}\right) = \frac{1}{\lambda_\mathrm{Pb}}\left(\frac{A_\mathrm{Pb}}{A_\mathrm{Rn}}\right) \tag{VI.6}$$

$$\tau_\mathrm{R}\left(\mathrm{Pb, Bi}\right) = \frac{1}{\lambda_\mathrm{Bi}}\left(\frac{A_\mathrm{Bi}}{A_\mathrm{Pb} - A_\mathrm{Bi}}\right) \tag{VI.7}$$

$$\tau_\mathrm{R}\left(\mathrm{Pb, Po}\right) = \frac{-b + \sqrt{b^2 - 4ac}}{2a} \tag{VI.8}$$

where

$$A_X = N_X \lambda_X;$$

$$a = A_{\mathrm{Pb}} - A_{\mathrm{Po}};$$

$$b = -A_{\mathrm{Po}}\left(\frac{1}{\lambda_{\mathrm{Bi}}} + \frac{1}{\lambda_{\mathrm{Po}}}\right);$$

and $c = \dfrac{-A_{\mathrm{Po}}}{\lambda_{\mathrm{Bi}}\lambda_{\mathrm{Po}}}$.

In each case, these should represent the residence time of the aerosols to which ^{210}Pb is attached. In practice, the apparent residence times calculated from these three radionuclide pairs often do not agree (e.g. see Refs [VI.2, VI.5]). Table VI.1 shows a summary of apparent residence times calculated for tropospheric aerosols from a number of studies [VI.6, VI.12]. The residence times are highly variable. In general, those obtained from the (^{210}Pb, ^{210}Po) pair are higher than for the other two pairs. However, relatively high residence times are also sometimes obtained for the (^{210}Pb, ^{210}Bi) pair.

Resuspension of dust can be an interfering factor in these calculations, since ^{210}Pb and its progeny will be at, or close to, secular equilibrium on this dust. This factor can be greater for the larger aerosol size fractions [VI.14, VI.23, VI.24]. It can also be greater for dry fallout than for precipitation [VI.11]. One approach to this problem is to use the determination of a member of the uranium decay chain above ^{210}Pb to apply a correction to the measured ^{210}Pb, ^{210}Bi and ^{210}Po concentrations to allow calculation of corrected residence times [VI.6, VI.10].

In the case of the (^{210}Pb, ^{210}Po) pair, another factor is the influence of significant sources of ^{210}Po from the Earth's surface due to the volatility of polonium, such as volcanic emissions, biomass burning and coal combustion (see Chapter 5). The influence of such sources is expected to be much lower in the case of (^{210}Pb, ^{210}Bi). In addition, the mean residence time of ^{210}Bi (τ_{Bi} = 7.2 d) is more comparable to aerosol residence times in the troposphere than that of ^{210}Po. Consequently, a residence time calculation based on (^{210}Pb, ^{210}Bi) is more reliable than one based on (^{210}Pb, ^{210}Po).

The apparent residence times obtained represent an average for the activity in the air or precipitation sample. They can therefore represent an average of residence times for several air masses which have mixed, such as a mixture of upper tropospheric and lower tropospheric air with different residence times [VI.5, VI.24]. For precipitation samples, they can represent an average of residence times for in-cloud rainout and below-cloud washout, and mixing of air masses during the storm [VI.9, VI.10].

TABLE VI.1. APPARENT AEROSOL RESIDENCE TIMES OBTAINED FOR ATMOSPHERIC OR PRECIPITATION SAMPLES

Location	τ_R(Rn, Pb) (d)	τ_R(Pb, Bi) (d)	τ_R(Pb, Po) (d)	Ref.
Portugal				
Savacem	—[a]	0.5–82 (5.7)	7–757 (33)	[VI.6]
USA				
Oak Ridge, TN	—[a]	4.8–15.3 (8.2)	—[a]	[VI.12]
Boulder, CO	2.2–3.4	1.59–13.0 (5.4)	11–77	[VI.2, VI.5, VI.13]
Argonne, IL	—[a]	6–67	33–66	[VI.14]
Madison, WI	—[a]	—	9.6	[VI.15]
Fayetteville, AR	—[a]	2.4–25.6 (8.5)	—[a]	[VI.16]
Fayetteville, AR	—[a]	3–240 (20)	2–320 (40)	[VI.9]
Poker Flat, AK	—[a]	—[a]	11.9–32	[VI.7]
Eagle, AK	—[a]	—[a]	0–38.9	[VI.7]
Switzerland				
Jungfraujoch	—[a]	—[a]	1–12 (6)	[VI.17]
Germany				
Freiburg	—[a]	—[a]	20	[VI.18]
France				
Gif-sur-Yvette	6.5 6.77	8.8–10.5 7–9	—[a]	[VI.19]
United Kingdom				
Milford Haven	6.77	—[a]	40	[VI.20]
India				
Mumbai	—[a]	8	—[a]	[VI.21]
Egypt				
El-Minia	—[a]	4.3–12.86 (9.83)	—[a]	[VI.22]

Source: Table 3 of Ref. [V.12].
Note: Averages in parentheses.
[a] —: data not available.

The use of the (^{210}Pb, ^{210}Bi) and (^{210}Pb, ^{210}Po) pairs relies on determination of these radionuclides in air filter or precipitation samples. The ^{210}Bi and ^{210}Po determinations should be carried out as soon as practicable after collection of the sample in order to minimize uncertainties resulting from decay and ingrowth corrections [VI.25]. When determining the sample ^{210}Po activity, ingrowth from both ^{210}Pb and ^{210}Bi between sample collection and analysis needs to be allowed for. In this case, a separate ^{210}Bi determination can be used to avoid unquantifiable errors in the ingrowth and decay correction [VI.8, VI.26].

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Appendix VII

USE OF ^{210}Po AS A RADIOTRACER IN BIOGENIC PARTICULATE FLUX STUDIES IN THE OCEANS

VII.1. INTRODUCTION

Polonium-210 is present in the world's oceans, and ^{210}Po sources and its distribution and dynamics in the water column are described in more detail in Chapter 8. The distribution of ^{210}Po in the water column of the open ocean is generally similar in various oceanic regions. In the upper layer of the ocean, ^{210}Po activity concentration in water is lower than that of ^{210}Pb due to the overriding importance of atmospheric deposition where ^{210}Po:^{210}Pb is roughly around 0.1. In the mesopelagic zone, under the euphotic/epipelagic layer, ^{210}Po is in excess relative to ^{210}Pb, probably due to release of ^{210}Po soluble forms from sinking faecal pellets and biogenic debris originating from the upper layer. In the bathypelagic and abyssopelagic zones, ^{210}Po concentrations are lower than ^{210}Pb, and although continuously produced in the water from ^{226}Ra–^{210}Pb radioactive decay, ^{210}Po is also continuously removed by sinking particulate matter [VII.1].

The mean residence time of ^{210}Pb and ^{210}Po radionuclides can be determined in the water column in several oceanic regions using the disequilibria between ^{226}Ra, ^{210}Pb and ^{210}Po. In general, ^{210}Po residence time in the upper layer of the ocean is short, around 0.6–0.7 a^{-1} [VII.1–VII.4]. This short residence time is due to rapid ^{210}Po adsorption onto particles such as viruses, bacteria, plankton, organic debris and inorganic particles that make the gravitational flux of settling particles. Furthermore, ^{210}Po is taken up into the cells of phytoplankton and becomes associated with soluble proteins in the cytosol [VII.5]. Zooplankton, as phytoplankton grazers, easily absorb ^{210}Po with cytosolic proteins of ingested phytoplankton, while metals, including ^{210}Pb, are segregated and remain mostly associated with the ejected cell debris.

The grazing activity of zooplankton in the epipelagic zone processes and removes particulate matter from the ocean surface layer, mostly packing it into faecal pellets that rapidly sink throughout the water column reach the abyssal sea-floor in a relatively short time [VII.6, VII.7]. The role of zooplankton in the removal of ^{210}Po and other elements from the epipelagic layer of the ocean has long been known and the associated transport of contaminants (e.g. plutonium from fallout) in the oceans has been investigated [VII.8–VII.11].

Besides phytoplankton and zooplankton, all organisms of marine food chains in the upper layer of the ocean contribute through similar processes, although not quantified yet, to concentrate ^{210}Po, starting from ionic ^{210}Po in the soluble phase and gradually incorporating it into large particles and biota tissues. With the seasons, and in line with the highs and lows of primary productivity in the epipelagic layer, ^{210}Po removal rates and flux to the deep-sea with the sinking particles vary throughout the year [VII.3, VII.12].

VII.2. USE OF ^{210}Po AS A RADIOTRACER

Studies on the biogenic particle flux to the deep ocean started in the 1960s with the supply of organic carbon from the euphotic zone to the permanently dark deep-sea layers and the sustainability of deep-sea food chains. In the 1970s, the existence of hydrothermal vents and deep-sea food chains based on the synthesis of organic matter by sulphur reducing bacteria were discovered, which sustain localized deep-sea food chains that do not depend on sunlight to synthesize organic compounds. For most of the ocean sea-floor, however, deep-sea life depends on the organic matter synthesized in the euphotic zone and exported to the deep sea [VII.13].

Investigations on the carbon cycle have highlighted the importance of the oceans as a sink for atmospheric carbon. Phytoplankton in the oceans, similar to forests on land, is a major sink for atmospheric CO_2 [VII.14, VII.15]. Dissolution of atmospheric CO_2 in the ocean is followed by incorporation of CO_2 into organic molecules through photosynthesis by phytoplankton, and this incorporation is followed by organic carbon recycling in, and export from, the euphotic zone with the sinking particle flux. This association of oceanic organic matter production and particulate matter export, conceptualized as the ‘biological carbon pump’, contributes to controlling CO_2 levels in the atmosphere and buffering the global climate system [VII.14, VII.16]. Therefore, the downward transfer and sequestration of carbon in the oceans is a central issue in global climate change.

Studies on particulate organic carbon, particulate inorganic carbon and biomineral (e.g. calcium and silicon) transfers in the ocean looked into natural tracers to help quantify transfer fluxes [VII.3, VII.7]. The behaviour of several radionuclides from the uranium radioactive series in the oceans has been assessed, especially ^{210}Pb, ^{210}Po and ^{234}Th, and their potential as natural radiotracers for particle flux [VII.17].

Phytoplankton productivity is connected to enhanced ^{210}Po absorption from sea water, and association of this naturally occurring radionuclide with the organic matter cycle in the surface ocean layer has been established [VII.15, VII.18].

Progress has been made in the direct measurement of the biogenic particle flux in the ocean, especially due to the use of drifting or moored particle interceptor devices or sediment traps [VII.6]. Determination of radionuclide concentrations in samples of particulate material indicate a high association of several particle reactive radionuclides, such as ^{210}Pb, ^{210}Po, $^{239+240}$Pu and ^{234}Th, with the total flux of particulate and detrital material [VII.18–VII.20]. However, recent work has shown a better correlation between ^{210}Po and particulate organic carbon fluxes [VII.3]. Owing to ^{210}Po binding to amino acids and incorporation into proteins in living organisms, the use of ^{210}Po as a tracer for particulate organic carbon flux in the ocean water column seems to have a scientific basis and has recently been explored. Polonium-210 seems to offer a good indirect means of computing the flux of organic particles and thus particulate organic carbon in the ocean [VII.3, VII.18–VII.20].

Although ^{210}Pb is also a particle reactive radionuclide, as is ^{234}Th, ^{210}Pb is adsorbed onto organic and inorganic surfaces but is not significantly absorbed into marine organisms to the extent that ^{210}Po is (see Chapter 8). For suspended particulate matter in coastal environments throughout the year, both ^{210}Pb and ^{210}Po total concentrations in water are positively correlated with the load of suspended particulate matter ($p < 0.01$). However, ^{210}Po concentration in particulate matter is better correlated with chlorophyll-a ($p < 0.10$), while ^{210}Pb was correlated with the inorganic particle residue ($p < 0.01$) but not with chlorophyll-a [VII.21]. All evidence suggests that ^{210}Po is bound, and mostly incorporated in, living matter, while ^{210}Pb seems to undergo adsorption onto the mineral and detrital particle fraction. Other naturally occurring radionuclides, such as ^{234}Th, have been used in combination with ^{210}Po to determine the mean residence time of particles in the surface ocean layer [VII.3]. It seems that ^{210}Po is a suitable tracer in oligotrophic conditions (non-bloom of phytoplankton), while ^{210}Pb and ^{234}Th display a better correlation and are thus better tracers of the inorganic fraction of particulate flux. Different natural radiotracers can provide different information and, actually, the measurement of ^{210}Po and ^{234}Th might be complementary to better understand the particle flux [VII.3, VII.7, VII.19, VII.20, VII.22].

Further research could be on the relationship of radionuclides and particulate organic carbon in particles of different size classes, and on the binding and partitioning (K_d) of radiotracers and organic particles. Further studies on ^{210}Po binding adsorption and absorption by microscopic life forms in the ocean are still needed, as are studies on the release rates of ^{210}Po from sinking particles (carcasses and faecal pellets).

Determination of ^{210}Po in association with other parameters is a tool to assess the organic particle flux in various regions of the ocean, and it is expected that by using these natural radiotracers a global picture of the biominerals and particulate organic carbon fluxes in the world's oceans will emerge.

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ABBREVIATIONS

APDC	ammonium pyrrolidine dithiocarbamate
CR	concentration ratio
DCC	dose conversion coefficient
DW	dry weight
FW	fresh weight
ICRP	International Commission on Radiological Protection
K_d	distribution coefficient
NORM	naturally occurring radioactive material
PBL	planetary boundary layer
TF	transfer factor

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