

# **Safety Reports Series**

## **No. 114**

# **Exposure due to Radionuclides in Food Other than During a Nuclear or Radiological Emergency Part 1: Technical Material**

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EXPOSURE DUE TO  
RADIONUCLIDES IN FOOD  
OTHER THAN DURING A  
NUCLEAR OR RADIOLOGICAL  
EMERGENCY

PART 1: TECHNICAL MATERIAL

The Agency's Statute was approved on 23 October 1956 by the Conference on the Statute of the IAEA held at United Nations Headquarters, New York; it entered into force on 29 July 1957. The Headquarters of the Agency are situated in Vienna. Its principal objective is "to accelerate and enlarge the contribution of atomic energy to peace, health and prosperity throughout the world".

SAFETY REPORTS SERIES No. 114

# EXPOSURE DUE TO RADIONUCLIDES IN FOOD OTHER THAN DURING A NUCLEAR OR RADIOLOGICAL EMERGENCY

PART 1: TECHNICAL MATERIAL

JOINTLY SPONSORED BY  
THE FOOD AND AGRICULTURE ORGANIZATION  
OF THE UNITED NATIONS,  
INTERNATIONAL ATOMIC ENERGY AGENCY  
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INTERNATIONAL ATOMIC ENERGY AGENCY  
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## FOREWORD

Radionuclides of both natural and human-made origin are present at various concentrations in food, resulting in exposure to ionizing radiation and an internal radiation dose. IAEA Safety Standards Series GSR Part 3 establishes basic requirements for the protection of people and the environment against harmful effects of ionizing radiation. Requirement 51 of GSR Part 3 states that **“The regulatory body or other relevant authority shall establish reference levels for exposure due to radionuclides in commodities.”** Paragraph 5.22 of GSR Part 3 further states that

“The regulatory body or other relevant authority shall establish specific reference levels for exposure due to radionuclides in commodities such as construction materials, food and feed, and in drinking water, each of which shall typically be expressed as, or be based on, an annual effective dose to the representative person that generally does not exceed a value of about 1 mSv.”

These requirements are in section 5 of GSR Part 3, which addresses existing exposure situations.

Currently, the associated recommendations in the IAEA Safety Standards to advise Member States on how these requirements ought to be implemented are very limited. Guidance has previously been developed and published by other international organizations with wider responsibilities for the quality of food and drinking water. Specifically, criteria for the assessment and management of radionuclides in drinking water in existing exposure situations have been published by the World Health Organization (WHO) and are referred to in the international food standards of the Joint FAO/WHO Codex Alimentarius Commission (Codex). For food, the Codex standards also include guideline levels for several radionuclides that are important for food in international trade. These guideline levels apply to radionuclides contained in foods destined for human consumption and traded internationally that have been contaminated following a nuclear or radiological emergency (i.e. both accidents and malevolent actions). However, there is a lack of information and practical guidance for assessing and therefore controlling exposures in existing exposure situations due to radionuclides in food. This Safety Report includes information pertaining to many different radionuclides, both human-made and of natural origin. However, the emphasis of the technical material contained in this publication is on a number of radionuclides of natural origin because, in general, it is these radionuclides that contribute significantly to ingestion dose in practice.

The purpose of this Safety Report is to provide Member States with technical information that can be used as a basis to assess and, if necessary, manage exposure to radionuclides in food in existing exposure situations. This includes information on the observed distributions of concentrations of natural radionuclides in various food products, the use of dietary surveys to assess ingestion doses and radionuclide concentrations in natural mineral waters and wild foods. This technical information could be useful in providing the scientific and technical foundation for a future guidance (published as IAEA-TECDOC-2011) on implementing relevant GSR Part 3 requirements, as they relate to radionuclides in food and drinking water.

This Safety Report is jointly sponsored by the Food and Agriculture Organization of the United Nations (FAO) and WHO. The IAEA wishes to acknowledge the contributions made by K. Kelleher (Ireland) in drafting and reviewing this report. The IAEA gratefully acknowledges the contribution of experts from the FAO, WHO and the project's international steering group of experts from IAEA Member States. The IAEA officers responsible for this publication were C. Blackburn of the Joint FAO/IAEA Centre of Nuclear Techniques in Food and Agriculture, T. Colgan, J. Brown and P.P. Haridasan of the Division of Radiation, Transport and Waste Safety.

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# 1. INTRODUCTION

## 1.1. BACKGROUND

### 1.1.1. Radionuclides in food

Radionuclides of both natural and human-made origin are present in various concentrations throughout the environment. The various sources of radionuclides in food are discussed in Section 2. In both terrestrial and aquatic ecosystems, these radionuclides can be transferred to plants and animals that are consumed by humans, thereby resulting in an exposure to ionizing radiation and an internal radiation dose. The activity concentration of a given radionuclide in a specific food can be highly variable, depending on many factors, including its chemical form and speciation. The ability of radionuclides to transfer into food is not the focus of this publication; suffice it to say that radionuclides can be detected in foods, though generally at low concentrations. Their mobility and transfer into food may depend on the characteristics of the ecosystem in which the radionuclide is present as well as physical, chemical and biological processes. For example, the degree to which the radionuclide is affixed to soil particles (in terrestrial ecosystems) or sediments (in aquatic ecosystems) determines how it will move through the environment and be assimilated by living organisms, such as plants and animals [1].

Food is essential for life, providing our bodies with carbohydrates, fats, fibre, minerals, protein, vitamins and water. Good nutrition is vital for good health and disease prevention. Food is central not only to our health but also our psychology and culture. As such, food and food quality are highly emotive issues. The physical phenomenon of radioactivity can also generate strong feelings, which explains why the subject of radioactivity in food is equally emotive.

Individual diets are highly variable, but, in general, the foods that we eat reflect our state of food security, and this can be defined according to four basic principles:

- (a) Availability — for example, the quantity and quality of food that can be produced or purchased;
- (b) Accessibility — the individual's ability to acquire appropriate foods (i.e. given their legal, political, economic and social arrangements);
- (c) Utilization — the ability to make proper use of food in terms of storage, adequate sanitation, preparation and the provision of sufficient energy and nutrients;

- (d) Stability with time — is food available, is it accessible and can it be utilized at all times?

Local soil and climatic conditions and access to natural resources such as forests and aquatic ecosystems have played an important role in determining the foods that are available to local populations. More recently, however, the globalization of the food production industry, the commercial development of aquaculture and a large reduction in transport costs have broadened the range of foods available in all regions of the world. Food availability has increased, and increasing numbers of people can access and utilize a broad range of different foods in addition to those produced locally.

Since 1976, WHO has maintained the Global Environment Monitoring System (GEMS) — Food Contamination Monitoring and Assessment Programme, commonly known as GEMS/Food [2]. This database provides detailed consumption data for 17 ‘cluster diets’, each of which is a combination of average national diets. Detailed dietary information has also been published by many national and regional agencies and is widely used to assess the nutritional quality of the diets and identify the presence of agrochemicals, additives and contaminants in order to evaluate potential health risks.

Information on dietary consumption is also an essential tool in assessing the radiation dose received from radionuclides in the diet; this is discussed in Section 3. Given the large variability in individual food preferences and differences in regional consumption patterns, monitoring programmes may need to include a component of the measurements of radionuclides in both individual foods and in samples of ‘total diet’. When sampling the total diet, the issues that arise primarily relate to the representativeness of samples and the identification of subgroups of the population receiving higher than average radiation doses. Sampling of individual food products can be helpful in identifying those foods or, perhaps more correctly, those food–radionuclide combinations that contribute disproportionately to individual dose. Measured concentrations of radionuclides in individual food products can also assist in supporting national and international trade.

### **1.1.2. IAEA Safety Standards**

Until relatively recently, the IAEA Safety Standards addressed criteria for controlling public exposure to radiation from radionuclides in food only

in the context of nuclear or radiological emergencies.<sup>1</sup> This changed in 2014 with the inclusion of safety requirements for existing exposure situations in IAEA Safety Standard Series No. GSR Part 3, Radiation Protection and Safety of Radiation Sources: International Basic Safety Standards [3], including for radionuclides in food.

Requirement 51 of General Safety Requirements (GSR) Part 3 [3] states that **“The regulatory body or other relevant authority shall establish reference levels for exposure due to radionuclides in commodities.”** Paragraph 5.22 of GSR Part 3 [3] further states:

“The regulatory body or other relevant authority shall establish specific reference levels for exposure due to radionuclides in commodities such as construction materials, food and feed, and in drinking water, each of which shall typically be expressed as, or be based on, an annual effective dose to the representative person that generally does not exceed a value of about 1 mSv.”

These requirements are in section 5 of GSR Part 3 [3], which addresses existing exposure situations.

Paragraph 5.1 of GSR Part 3 [3] defines the scope of the requirements addressing existing exposure situations. In the case of commodities such as food and drinking water, the requirements apply to the following:

- (a) Exposure due to radionuclides deriving from past activities that were never subject to regulatory control or that were subject to regulatory control but not in accordance with the requirements of GSR Part 3 [3];
- (b) Exposure due to radionuclides deriving from a nuclear or radiological emergency, after the emergency has been declared to be ended;
- (c) Exposure due to radionuclides of natural origin, regardless of activity concentration.

In summary, in accordance with GSR Part 3 [3], radiation exposure from the consumption of food and drinking water in non-emergency situations is required to be managed as an existing exposure situation through the establishment and use of reference levels and needs to consider both natural and human-made radionuclides.

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<sup>1</sup> IAEA safety standards related to regulatory control of discharge limits also consider the levels of radioactivity in food and water; for example, IAEA Safety Standards Series No. GSG-9, Regulatory Control of Radioactive Discharges to the Environment [4].

### 1.1.3. Exposure situations

In the IAEA Safety Standards, radiation exposure is categorized according to three broad circumstances that individuals may experience, namely planned exposure situations, emergency exposure situations<sup>2</sup> and existing exposure situations. These are defined in the IAEA Safety Glossary [5] and have the following meanings:

- (a) A planned exposure situation arises from the planned operation of a radiation source or from a planned activity that results in an exposure due to a source.
- (b) An emergency exposure situation arises as a result of an accident, a malicious act or other unexpected event and requires prompt action in order to avoid or to reduce adverse consequences.
- (c) An existing exposure situation is a situation of exposure that already exists when it is necessary to take a decision on the need for control.

Planned exposure situations introduce new sources of radiation exposure; they are a matter of choice and are normally subject to some form of control or authorization by the regulatory body. On the other hand, emergency exposure situations and existing exposure situations are both situations that are not a matter of choice — when they occur, a decision needs to be taken on what actions, if any, are justified to reduce exposure. Emergency exposure situations develop into existing exposure situations, often referred to as the ‘recovery phase’, following a nuclear or radiological emergency.

Radionuclides present in food and drinking water can arise as a result of any of the three exposure situations. For example:

- (a) A regulated activity such as an authorized discharge from a nuclear facility or a hospital is a planned exposure situation.
- (b) Emergency exposure situations may result in the accumulation of radionuclides in food and drinking water that may persist into the recovery phase.
- (c) Primordial radionuclides present in soil represent an existing exposure situation.

For any given radionuclide, it might not always be possible to identify its origin precisely. For example, a food sample could contain <sup>137</sup>Cs from several

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<sup>2</sup> Emergency exposure situations are outside the scope of this publication. Requirements for emergency exposure situations are established in IAEA Safety Standards Series No. GSR Part 7, Preparedness and Response for a Nuclear or Radiological Emergency [6].

different sources: nuclear weapons testing, authorized discharges from a nearby licensed facility and unplanned releases from a previous accident. However, the IAEA Safety Standards require that radiation doses from food and drinking water in the diet be managed as either an emergency exposure situation or an existing exposure situation.

#### **1.1.4. Existing international guidance**

As mentioned above, the safety requirements relevant to radionuclides in food and drinking water are established in section 5 of GSR Part 3 [3], which covers existing exposure situations. Currently there are no associated recommendations or guidance in the IAEA Safety Standards to advise Member States on how these requirements ought to be implemented. However, some related guidance has previously been developed and published by other international organizations with wider responsibilities for the quality of food and drinking water.

Criteria for the assessment and management of radionuclides in drinking water in existing exposure situations have been published by WHO [7]. These criteria cover both natural and human-made radionuclides. The international food standards of the Joint FAO/WHO Codex Alimentarius Commission (Codex) include guideline levels for 20 key radionuclides that are important for food in international trade that has been contaminated as a result of a nuclear or radiological emergency [8]. These Codex guideline levels are based on conservative assumptions, and the radionuclides included are those that are important for uptake into the food chain and are typically contained in releases from nuclear installations as a result of an accident or malevolent act or are used in radioactive sources in large enough quantities to be significant potential contributors to levels in foods if there were to be an incident. Radionuclides of natural origin are generally excluded from consideration in the Codex guideline values, but  $^3\text{H}$ ,  $^{14}\text{C}$  and  $^{235}\text{U}$  are included because they may also be human-made and meet the preceding criteria.

IAEA-TECDOC-1788 [9] summarized the international standards and guidance for different exposure situations that relate to radionuclides in food and drinking water. It also identified a number of gaps and inconsistencies in guides that provide recommendations and guidance on how to comply with the safety requirements. The differences between the various regulatory and guidance documents include differences in scope, radiation protection criteria and terminology. Specifically, there is a lack of information and practical guidance for controlling exposures in existing exposure situations attributable to natural radionuclides in food.

Management of radionuclides in food and drinking water can also be considered in the broader context of all goods (i.e. commodities in general) containing radionuclides — either intentionally added or present adventitiously — that are supplied to the public. A discussion document, prepared jointly by the Autoridad Regulatoria Nuclear of Argentina and the IAEA [10], suggests that it may be impractical “to use dosimetric quantities [i.e. radiation dose] as the primary basis for controlling the presence of radioactivity in consumer goods” as “These quantities are generally unmeasurable...and their estimation requires modelling, often with substantial subjective uncertainties.” Instead, the document, along with other national and international guidelines, suggests using the range of observed concentration, such as activity concentration per unit weight or per unit volume, as the basis for control.

In order to address the gaps and inconsistencies identified in Ref. [9], and in response to an IAEA General Conference resolution, a project on Radionuclides in Food and Drinking Water in Non-Emergency Situations was initiated. An international Steering Group of experts, under the chairmanship of a leading expert from Norway, together with a joint Secretariat of the FAO, IAEA and WHO, was convened to direct and manage the project.

The first meeting of the Steering Group was held in December 2017 and included representatives from FAO, the United Nations Environment Programme (UNEP) (the UNSCEAR secretariat) and WHO. The Steering Group agreed that during 2018 the work would initially be focused on quantifying the range of radiation doses typically received from natural radionuclides in food. It was agreed that the Steering Group would meet annually to advise on and guide the work to be undertaken under the project.

At its meeting in September 2019, the Steering Group agreed on the production of a publication to summarize the technical work undertaken within the project. The present report is the result of this request. The Steering Group also asked the secretariat to develop a discussion paper on how to harmonize the approach to managing natural and human-made radionuclides in both food and drinking water. This latter document has resulted in a separate IAEA TECDOC publication.<sup>3</sup>

#### **1.1.5. Justification of protective actions and optimization of protection and safety**

Exposures from radionuclides in food and drinking water are governed by the principles of justification of protective actions and optimization of protection

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<sup>3</sup> Exposure Due to Radionuclides in Food Other Than During a Nuclear or Radiological Emergency, IAEA-TECDOC-2011 (2022).



and safety. The relevant requirements with respect to the exposure of members of the public to radionuclides in food and drinking water are outlined in Requirement 48 of GSR Part 3 [3]. Therefore, any protection strategy and remedial or protective actions related to exposure to radionuclides in food and drinking water are to be applied accordingly to ensure that they are commensurate with the risks involved and that any benefits will outweigh any detriments associated with their implementation.

## 1.2. OBJECTIVE

The principal objective of this Safety Report is to provide the Member States of the FAO, IAEA and WHO with technical information that can be used to assess and manage radionuclides in food in existing exposure situations, consistent with the approach of WHO for radionuclides in drinking water. This includes information on the observed distributions of concentrations of natural radionuclides in various food products, the use of total diet and other studies to assess ingestion doses, and information on radionuclide concentrations in natural mineral waters.

This Safety Report is intended to provide the scientific and technical foundation for a future guidance on implementing Requirement 51 of GSR Part 3 [3], as it relates to radionuclides in food and drinking water. Guidance provided here, describing good practices, represents expert opinion but does not constitute recommendations made on the basis of a consensus of Member States.

## 1.3. SCOPE

This Safety Report considers exposure due to radionuclides in food other than during an emergency exposure situation. It addresses radionuclides of both natural and human-made origin in food. Natural mineral waters that are sold as foods are addressed in the standards of the Joint FAO/WHO Codex Alimentarius Commission [11] and are therefore within the scope of the Safety Report.

The main focus of this Safety Report is on natural radionuclides in food, which is the major omission in current international guidance such as the Codex Alimentarius [8], where the emphasis is on human-made radionuclides and incidents. For individual food products, data are provided that characterize the observed worldwide variability of the important natural radionuclides in terms of parameters such as the arithmetic mean, the median and an upper

bound percentile of the population.<sup>4</sup> These parameters will be of use to those who measure radionuclides of natural origin in food products and derive statistics to characterize their sample. Further, in terms of the radiation dose from the intake of radionuclides in the whole diet, different dose assessment methodologies are presented, and the advantages and disadvantages of each are discussed. Information is also provided on the approaches used for managing non-radioactive contaminants in food.

Advice received from the IAEA Radiation Safety Standards Committee, expert consultants and other interested parties indicates that any future guidance and approach to managing natural radionuclides in food could be based on activity concentrations as well as radiation dose. This Safety Report recognizes that activity concentrations in foods are fundamental and therefore lays the foundation for future work in this area by deriving parameters that characterize the activity concentrations of several natural radionuclides in certain foods. Note that these parameters are not intended to become established as limits for natural radionuclides in food. The utility of these population parameters is that others may use them as comparators for their sample statistics.

Criteria for controlling food in emergency exposure situations are outside the scope of the Safety Report.

#### 1.4. STRUCTURE

This Safety Report presents and summarizes the technical material that could be used to support national policies and strategies on the evaluation of radiation doses from radionuclides present in the diet. An additional Technical Document (published as IAEA-TECDOC-2011) will discuss the various issues to be taken into account in implementing Requirement 51 of GSR Part 3 [3] and the use of the information presented in this publication.

Following this introductory section, Section 2 of this Safety Report addresses the sources of natural and human-made radionuclides in food. Section 2 also summarizes the outcomes of previously published national and international reviews of radionuclides in food throughout the world.

The various approaches for estimating the dietary intake of radionuclides in food are outlined in Section 3. This section highlights the strengths and limitations

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<sup>4</sup> In statistics, a parameter is a numerical value that describes a characteristic of an entire population (for example, a population median is the median of a complete set of a defined group), whereas a statistic is a numerical value characteristic of a sample (for example, a sample median is the median of the set of measurement results collected as a subset of the entire population).

of these approaches. In addition, alternative approaches to estimating dietary intake of radionuclides are also discussed. These approaches could be used in the absence of food consumption data or radioactivity in food monitoring data.

Section 4 reviews and summarizes the ingestion dose from over 50 years of national dietary studies. Further, the variability of ingestion dose as a function of age and dietary study type from these studies is investigated, and the ingestion dose from previous studies in natural mineral water, aquaculture and wild foods is discussed.

A comprehensive dataset of natural radionuclides in food products was compiled as part of the development of this publication. These data were collated by radionuclide and food subcategory. Section 5 outlines the approach taken to collate these data and the methodologies employed to derive population parameters, including confidence intervals of the population means and 95th percentile values for a number of combinations of radionuclide and food subcategories. In some instances, this approach was applied to individual food products or species.

Section 6 summarizes the key technical findings and identifies a number of knowledge gaps that it would be helpful to address and could be the focus of future work.

The Safety Report identifies and discusses the essential components of national programmes to assess radiation doses from the diet for both the general population and those subgroups likely to receive elevated doses (the 'representative person').

The Appendix contains additional information on the process for the selection of natural radioactivity in food monitoring data for inclusion in the statistical analysis outlined in Section 5.

There are three annexes to this report. Annex I summarizes the radioanalytical techniques that could be used to identify and quantify natural radioactivity in foods. Annex II contains the results from the statistical analyses of natural radionuclides in foods as described in Section 5, and Annex III provides additional information on the statistical analyses at the food category levels for molluscs.

## **2. RADIONUCLIDES IN FOOD**

### **2.1. SOURCES OF RADIONUCLIDES IN FOOD**

There are many natural and human-made radionuclides present throughout the environment, all of which are potentially available for incorporation into food and drinking water. Natural radionuclides consist of both cosmogenic and primordial radionuclides. Human-made radionuclides in the environment arise from several sources, including authorized discharges, nuclear fallout from the testing of nuclear weapons and large scale accidental releases. A small number of radionuclides that are produced artificially are also found naturally.

The different sources of radionuclides in the environment are discussed below.

#### **2.1.1. Cosmogenic and primordial radionuclides**

There are more than thirty so-called cosmogenic and primordial radionuclides present in the environment, with half-lives from a few seconds up to millions of years. Cosmogenic radionuclides are produced constantly through the interaction of cosmic radiation with stable nuclides in the upper atmosphere. Cosmogenic radionuclides include  $^3\text{H}$ ,  $^7\text{Be}$ ,  $^{14}\text{C}$ ,  $^{22}\text{Na}$ ,  $^{32}\text{P}$  and  $^{35}\text{S}$ . However, at the surface of the Earth the concentrations of many of these radionuclides in food and drinking water are low and they can be difficult, if not impossible, to detect.

Primordial radionuclides are those that were produced when the Earth was formed and have sufficiently long half-lives that they are still detectable in the environment. Best known are the three radioactive series, headed by  $^{238}\text{U}$ ,  $^{235}\text{U}$  and  $^{232}\text{Th}$ . The radionuclide  $^{235}\text{U}$  and its progeny are generally relatively unimportant as a source of radiation exposure in the environment because the concentrations of  $^{235}\text{U}$  are so much lower than those of  $^{238}\text{U}$  and  $^{232}\text{Th}$ . A number of regions throughout the world have high natural background radiation levels. Food farmed in these regions can have elevated levels of natural radionuclides as a result of the uptake of uranium and thorium series radionuclides from the soil and water. A non-exhaustive list of such areas includes Ramsar (Islamic Republic of Iran) [12], Yangjiang (China) [13], Kerala (India) [14] and Mamuju (Indonesia) [15]. This publication discusses uranium isotopes in the context of ingestion dose as a result of radiotoxicity. However, consideration of the total uranium content also needs to be taken into account, as the predominant risk from uranium exposure is chemical toxicity [7].

Other well known primordial radionuclides with long half-lives include  $^{87}\text{Rb}$ ,  $^{115}\text{In}$  and  $^{147}\text{Sm}$ , but these and others are present in the environment in such low concentrations that they are insignificant in terms of radiation exposure.

Another important primordial radionuclide is  $^{40}\text{K}$ , which is found in varying concentrations throughout the environment. Potassium is readily incorporated into plants and crops, for which it is an essential element in the control of photosynthesis, the activation of growth related enzymes and many other essential biological mechanisms.  $^{40}\text{K}$  represents 0.012% (120 ppm) of natural potassium and so is itself present in all foods. Potassium (and therefore  $^{40}\text{K}$ ) is also present in most water supplies. The levels of natural potassium and therefore  $^{40}\text{K}$  in the human body are more or less uniformly distributed and are under homeostatic control. The United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) has estimated an annual effective dose due to  $^{40}\text{K}$  of 165  $\mu\text{Sv}/\text{year}$  for adults and 185  $\mu\text{Sv}/\text{year}$  for children [16].

### **2.1.2. Authorized discharges**

In managing low level liquid and gaseous wastes produced in the nuclear and other industries, regulatory bodies may authorize the release of such wastes to the environment. These discharges are subject to strict regulatory control such that the radiological impact on humans and the environment complies with national and international standards for radiation safety [4]. Discharges occur routinely from nuclear facilities, such as power reactors and reprocessing plants, as well as other licensed operations, such as those at hospitals and research laboratories. Most of the discharges consist of radionuclides of human-made origin, but some also include natural radionuclides.

Some industrial activities involving the extraction and processing of minerals and other commercial ores are also performed under regulatory control. The mining of uranium and thorium historically has always been subject to regulatory control. In some States, other activities such as the mining of gold, rare earths and other ores are also regulated to control the radiation doses received by workers and the public and to protect the environment. These industries involving naturally occurring radioactive material will often discharge natural radionuclides into the environment.

### **2.1.3. Nuclear fallout**

The above ground testing of nuclear weapons that took place between 1945 and 1980, but peaked in the late 1950s and early 1960s, released radionuclides directly into the environment. Those radionuclides that were produced in sufficiently large quantities and have sufficiently long half-lives to make

them of long term significance are  $^3\text{H}$ ,  $^{14}\text{C}$ ,  $^{90}\text{Sr}$  and  $^{137}\text{Cs}$ . The total global deposition of these four radionuclides has been estimated to be  $186 \times 10^3$  PBq (for  $^3\text{H}$ ), 213 PBq (for  $^{14}\text{C}$ ), 622 PBq (for  $^{90}\text{Sr}$ ) and 948 PBq (for  $^{137}\text{Cs}$ ) [16]. These four radionuclides may still be found in low but measurable quantities in the environment and, apart from  $^3\text{H}$ , are particularly important as a source of ingestion dose from the diet.

Large amounts of shorter lived radionuclides were also produced, including  $^{95}\text{Zn}$ ,  $^{103,106}\text{Ru}$ ,  $^{131}\text{I}$ ,  $^{140}\text{Ba}$ ,  $^{141}\text{Ce}$  and  $^{144}\text{Ce}$ . These are no longer detectable in the environment.

Nuclear fallout can be of particular concern in foods produced in the vicinity of legacy nuclear weapons test sites, such as Semipalatinsk (Kazakhstan) [17] and the Marshall Islands in the South Pacific [18].

#### **2.1.4. Unregulated activities**

There are many circumstances in which waste materials containing elevated concentrations of radionuclides are present in the environment as a result of practices that may not, at the time, have been subject to regulatory control from a radiation perspective. These include tailings from uranium and other mining activities. As with authorized activities, the radionuclides present may contaminate the environment directly or as a result of water runoff, while resuspension and root uptake may transfer radionuclides, such as those from the uranium and thorium series radionuclides, to plants and crops.

#### **2.1.5. Large scale accidental releases**

A number of nuclear and other accidents have released significant quantities of radionuclides to the environment. These include the Kyshtym accident in 1957 [19], the accident at the Chornobyl Nuclear Power Plant (NPP) in 1986 [20] and the accident at the Fukushima Daiichi NPP in 2011 [21].

The source term (amount of activity released) with respect to important long lived radionuclides for each of the three accidents is given in Table 1.

### **2.2. NATIONAL AND INTERNATIONAL REVIEWS**

#### **2.2.1. Natural radionuclides**

UNSCEAR published ‘reference values’ of activity concentrations for natural radionuclides in food in its 1993 Report [22]. These were subsequently updated in 2000 [19], as summarized in Table 2. These reference values were

TABLE 1. SOURCE TERM FOR MAJOR UNPLANNED RELEASES OF RADIONUCLIDES TO THE ENVIRONMENT

Place	Estimated release (PBq)				Reference
	Sr-90	Cs-134	Cs-137	Pu-239/240	
Kyshtym	2	— <sup>a</sup>	—	—	[19]
Chornobyl	10	~47	~85	0.031	[20]
Fukushima	$3.3 \times 10^{-3}$ -0.14	8.3-50	7-20	$0.9 \times 10^{-7}$ - $6.4 \times 10^{-6}$	[21]

<sup>a</sup> —: data not available.

TABLE 2. REFERENCE VALUES FOR CONCENTRATIONS OF RADIONUCLIDES IN THE URANIUM AND THORIUM SERIES IN FOOD PRODUCTS AND DRINKING WATER [16]

Food category	Activity concentration (Bq/kg)									
	U-238	Th-230	Ra-226	Pb-210	Po-210	Th-232	Ra-228	Th-228	U-235	
Milk products	0.001	0.0005	0.005	0.015	0.015	0.0003	0.005	0.0003	0.00005	
Meat products	0.002	0.002	0.015	0.08	0.06	0.001	0.01	0.001	0.00005	
Grain products	0.02	0.01	0.08	0.05	0.06	0.003	0.06	0.003	0.001	
Leafy vegetables	0.02	0.02	0.05	0.08	0.1	0.015	0.04	0.015	0.001	
Roots and fruits	0.003	0.0005	0.03	0.03	0.04	0.0005	0.02	0.0005	0.0001	
Fish products	0.03	0.01	0.1	0.2	2	0.01	— <sup>a</sup>	0.1	—	
Drinking water	0.001	0.0001	0.0005	0.01	0.005	0.00005	0.0005	0.00005	0.00004	

<sup>a</sup> —: data not available.

based on the most representative and widely available data at that time. The reference values have been developed for use in dose assessments and, when applied to averaged worldwide consumption data, indicate that the annual age weighted dose from  $^{40}\text{K}$  is approximately 170  $\mu\text{Sv}$ , and the dose from radionuclides of the uranium and thorium decay series is approximately 140  $\mu\text{Sv}$  (almost entirely due to  $^{228}\text{Ra}$ ,  $^{226}\text{Ra}$ ,  $^{210}\text{Pb}$  and  $^{210}\text{Po}$ ), which results in a total worldwide averaged ingestion dose from natural radionuclides of about 310  $\mu\text{Sv}/\text{year}$ . Somewhat higher doses are received by infants and children, and somewhat lower doses are received by adults. More than 90% of the dose is attributable to natural radionuclides in food and the remainder to natural radionuclides in drinking water.

The complexity of these types of calculations is underlined by considering how the reference value for  $^{210}\text{Po}$  in fish products has been derived. The food group 'fish products' consists of fish, crustaceans and molluscs, for which the estimated average activity concentrations are 2.4 Bq/kg, 6 Bq/kg and 15 Bq/kg, respectively. The representative annual consumption rates are 13 kg for fish and 1 kg each for crustaceans and molluscs. However, because of the relatively short half-life of  $^{210}\text{Po}$  (138 days), radioactive decay between the time of catch and consumption needs to be considered. Aarkrog et al. [23] provide statistics for the percentage of seafood that is eaten fresh, frozen, smoked and canned and the typical time delays prior to consumption for each process.<sup>5</sup> When all these factors are taken into account, the reference activity concentration for fish products is 2 Bq/kg (2000 mBq/kg).

These calculations are based on production and consumption patterns at the time. They are not necessarily representative of current production and consumption patterns, neither generally nor between and within individual countries. One important development in recent years is the growth of the aquaculture industry and the impact this has had on both the market for seafood and the associated radionuclide concentrations. This is discussed further in Section 4.4.

It is reasonable to question the usefulness of a reference value for activity concentration for  $^{210}\text{Po}$  in fish products when the activity concentrations and consumption rates for fish, shellfish and molluscs are so different. This has been recognized by UNSCEAR, which has modified its food categories and now provides radionuclide and consumption data separately for freshwater fish, saltwater fish, crustaceans and molluscs (see Section 3.1). Table 3 gives the activity concentration in each food category that would result in an individual

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<sup>5</sup> Other natural radionuclides and many human-made radionuclides of interest have longer half-lives, and so the time delay prior to consumption has a very small effect and can be ignored in dose calculations.



TABLE 3. CALCULATED ACTIVITY CONCENTRATIONS OF NATURAL RADIONUCLIDES IN EACH FOOD GROUP THAT WOULD GIVE 1 mSv IN A YEAR USING WORLDWIDE POPULATION WEIGHTED CONSUMPTION RATES

Food category	Activity concentration (Bq/kg) to give 1 mSv in a year								
	U-238	Th-230	Ra-226	Pb-210	Po-210	Th-232	Ra-228	Th-228	U-235
Milk products	342	73	22	22	13	67	55	214	314
Meat products and offal	505	108	33	33	19	99	81	316	464
Grain products	171	37	11	11	6	33	27	107	157
Vegetables and fruit	97	21	6	6	4	19	16	60	89
Fish (freshwater)	3 900	835	254	254	146	763	627	2 440	3 580
Fish (saltwater)	2 960	635	193	193	111	580	476	1 850	2 720
Crustaceans	20 200	4 330	1 320	1 320	758	3 950	3 250	12 630	18 550
Molluscs	13 890	2 980	906	906	521	2 720	2 230	8 680	12 760

dose of 1 mSv in a year on the basis of the worldwide weighted consumption rates quoted in Section 3.1. Thus, an annual consumption of 1.6 kg of molluscs with an average activity concentration of 521 Bq/kg of  $^{210}\text{Po}$ , with a committed effective dose per unit intake of  $1.2 \times 10^{-6}$  Sv/Bq for adults, corresponds to an annual dose of 1 mSv:

$$\text{Activity concentration (Bq / kg)} = \frac{1 \times 10^{-3} \text{ Sv}}{1.6 \text{ kg} \times 1.2 \times 10^{-6} \text{ (Sv / Bq)}} = 521 \text{ Bq / kg} \quad (1)$$

On the other hand, because the average annual consumption of cereals is so much higher (130 kg), a much lower activity concentration of about 6 Bq/kg corresponds to an annual dose of 1 mSv.

## 2.2.2. Human-made radionuclides

The activity concentrations of human-made radionuclides in food vary significantly and depend on the source of these radionuclides, the time elapsed since their release into the environment and the ability to transfer into food from the environment. Sources of human-made radionuclides may include fallout from nuclear weapons tests as well as discharges of radionuclides from isotope production industries, nuclear power generating facilities, research laboratories and hospitals. In addition, residual levels of human-made radionuclides may remain in the environment over many years following accidents, sometimes at levels much higher than those before the accident (as discussed further in Section 4.5).<sup>6</sup> Below is a summary of the levels of human-made radionuclides in foods in existing exposure situations in some countries and regions worldwide.

### 2.2.2.1. Australia

In 2019, the Australian Radiation Protection and Nuclear Safety Agency conducted a study on radiation doses from the average Australian diet [24]. As part of this study, 268 food samples, including individual foodstuffs, drinks and infant food, were analysed for  $^{137}\text{Cs}$ ,  $^{134}\text{Cs}$ ,  $^{60}\text{Co}$  and  $^{241}\text{Am}$ . Only 21 of these samples (8%) had measurable activity concentrations of  $^{137}\text{Cs}$ , ranging from 0.061–0.389 Bq/kg, and activity concentrations for all other radionuclides were not detected and therefore reported as below the limits of detection.

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<sup>6</sup> The levels of human-made radionuclides in food included in this review do not include emergency exposure situations.

2.2.2.2. Belarus, the Russian Federation and Ukraine

A 2006 IAEA report on the environmental consequences of the accident at the Chornobyl NPP [20] includes measurement data of radioactivity in foods. The report summarizes the data for measured activity concentrations of  $^{137}\text{Cs}$  in foods produced in highly contaminated and less highly contaminated areas in Belarus, the Russian Federation and Ukraine (see Table 4).

The report indicates that there has been a slow decrease in the levels of human-made radionuclides in food in the decades following the accident at the Chornobyl NPP. The levels of radiocaesium ( $^{137+134}\text{Cs}$ ) in agricultural food products in areas affected by the accident at the Chornobyl NPP are typically below national, regional and international action levels. This is as a result of agricultural countermeasures implemented following the accident and due to natural processes, such as soil migration of radionuclides and radioactivity decay.

2.2.2.3. Canada

Health Canada has been conducting total diet studies since 2000 [25]. The main purpose of these studies is to monitor chemical contaminants in the foods that are typically consumed by the Canadian population according to WHO dietary survey guidelines. The contaminants measured as part of the total diet

TABLE 4. MEAN  $^{137}\text{Cs}$  ACTIVITY CONCENTRATIONS IN AGRICULTURAL FOOD PRODUCTS ACROSS CONTAMINATED AREAS OF BELARUS, THE RUSSIAN FEDERATION AND UKRAINE (2000–2003) [20]

Contaminated district (soil deposition range)	Grain	Potato	Wheat	Milk
	Bq/kg		Bq/L	
<i>Belarus</i>				
Gomel (185 kBq/m <sup>2</sup> )	30	10	220	80
Mogilev (37–185 kBq/m <sup>2</sup> )	10	6	100	30
<i>Russian Federation</i>				
Bryansk (185 kBq/m <sup>2</sup> )	26	13	240	110
Kaluga, Tula, Orel (37–185 kBq/m <sup>2</sup> )	12	9	42	20
<i>Ukraine</i>				
Zhytomyr, Rovno (185 kBq/m <sup>2</sup> )	32	14	400	160
Zhytomyr, Rovno (37–185 kBq/m <sup>2</sup> )	14	8	200	90

studies include trace elements, pesticides, industrial chemicals and radionuclides (both natural and human-made).

The levels of  $^{40}\text{K}$ ,  $^{210}\text{Pb}$ ,  $^{134}\text{Cs}$ ,  $^{137}\text{Cs}$  and  $^{131}\text{I}$  in food and the monitoring results reported from 2015 to 2017 covered approximately 480 composite food samples that included individual ingredients and infant food. All the food samples analysed reported human-made activity concentrations that were below the limits of detection of approximately 1 Bq/kg for all radionuclides measured. Furthermore, a review of the radioactivity in food monitoring data from 2000 to 2010 identified only very few samples that had measurable activity concentrations of  $^{137}\text{Cs}$  above the limit of detection of approximately 2 Bq/kg.

#### 2.2.2.4. *The European Union*

Under Article 36 of the Euratom Treaty, all European Union (EU) Member States are obliged to report environmental radioactivity monitoring data to the European Commission on a regular basis. These data are reviewed and compiled by the European Commission Joint Research Centre into the Radioactivity Environmental Monitoring data bank (REMdb). The REMdb was created after the accident at the Chornobyl NPP in 1986 and has compiled over 30 years of radioactivity monitoring data that include human-made radionuclides in food and drinking water [26].

The latest publicly available dataset from the REMdb contains monitoring data from 2007 to 2011 and includes more than 15 000 measurements in milk samples and approximately 5500 measurements in complete meals [27] (data from 2012 onward are expected to be made available once the next monitoring report is published). These data cover all 27 European Union Member States and the United Kingdom (UK). These measurements are compared to a reporting level corresponding to a dose level of 1  $\mu\text{Sv}$ , which is deemed of no radiological significance by the European Commission [28].

A total of 17 293 measurements of  $^{137}\text{Cs}$  made in milk samples were reported, of which only 1204 (7%) were above the reporting level of 0.5 Bq/L. Of the 5683 measurement results for  $^{90}\text{Sr}$ , only 39 (<1%) were above the reporting level of 0.2 Bq/L.

#### 2.2.2.5. *Japan*

In Japan, following the nuclear accident at the Fukushima Daiichi NPP, the Ministry of Health, Labour and Welfare initiated a comprehensive monitoring programme for human-made radioactivity in food to ensure that the levels of  $^{137}\text{Cs}$  and  $^{134}\text{Cs}$  were below the corresponding Japanese limits implemented in April 2012 [29].

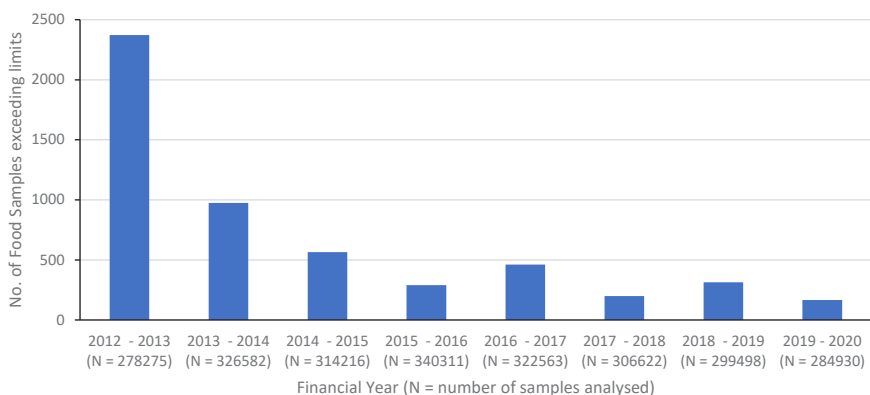


FIG. 1. Number of food samples exceeding the national reference level of Japan for radiocaesium of 100 Bq/kg in food samples [30].

The monitoring programme is conducted by the local governments of the 47 prefectures in Japan, and between 2012 and 2019 the average number of samples analysed was approximately 310 000 samples per year. The monitoring programme has demonstrated that the levels of radiocaesium in food has been decreasing steadily year on year since the accident. Between 2012 and 2013 the percentage of samples exceeding the limits stood at 0.9%, whereas between 2019 and 2020 the percentage had dropped to 0.1%. Similarly, the number of food samples with measurable radiocaesium activity concentrations is very low when compared to the total number of samples measured [30].

Figure 1 outlines the number of food samples exceeding the Japanese national reference level of 100 Bq/kg for radiocaesium in food from fiscal year (FY) 2012–2013 to FY 2019–2020.

In 2020, the food samples with  $^{137}\text{Cs}$  activity concentration above 100 Bq/kg were in wild foods.

#### 2.2.2.6. United States of America

The US Food and Drug Administration has been conducting total diet studies since 1961. This programme was initiated to evaluate the levels of radioactivity in food as a result of nuclear weapons fallout. The annual survey has since evolved to identify approximately 800 contaminants and nutrients in 280 foods and beverages that are sampled from retail outlets across the United States of America (USA) [31]. The food samples are prepared as they would be for human consumption and analysed for radionuclides and other contaminants. The human-made radionuclides analysed as part of the total diet studies are  $^{90}\text{Sr}$ ,

$^{137}\text{Cs}$ ,  $^{106}\text{Ru}$  and  $^{131}\text{I}$ , and the latest available results cover 2006 to 2014 [32]. A total of 3740 food samples were analysed for  $^{137}\text{Cs}$ , of which only 3 (<0.1%) detected  $^{137}\text{Cs}$  above the limit of detection of 5 Bq/kg. Strontium-90 analyses were conducted on 3498 food samples, and 285 (8%) of these samples had  $^{90}\text{Sr}$  activity concentrations above the limit of detection of 0.1 Bq/kg.

#### 2.2.2.7. *Other studies*

Most national studies conducted on human-made radionuclides in food contain few, if any, measurements of human-made radionuclides other than  $^{137}\text{Cs}$  and  $^{90}\text{Sr}$ . However, monitoring of food for other human-made radionuclides is sometimes conducted at or close to nuclear facilities such as nuclear power plants, nuclear reprocessing facilities and research establishments. For example, the United Kingdom routinely reports levels of  $^{14}\text{C}$ ,  $^{60}\text{Co}$ ,  $^{99}\text{Tc}$ ,  $^{239,240}\text{Pu}$  and  $^{241}\text{Am}$  in seafood close to the Sellafield nuclear fuel production and reprocessing facility [33]. Similar monitoring is conducted in France on seafood, meat and milk samples sourced from the vicinity of the Cap de la Hague reprocessing facility on the north-west coast of France [34]. The Korean Nuclear Society has monitored fish for  $^{131}\text{I}$  in water bodies close to a radioisotope production facility in Daejeon, Republic of Korea [35].

In addition, specific studies have also been conducted on the long term accumulation of human-made radionuclides in foods as a result of fallout from nuclear accidents. These studies highlight specific regions and consumers of food that continue to be affected by human-made radionuclides even after the end of a nuclear emergency. These studies are primarily focused on human-made radionuclides in wild foods (Section 4.5.2), but can also include, for example, domestic animals grazing on contaminated lands. For example, elevated  $^{137}\text{Cs}$  activity concentrations as a result of the accident at the Chernobyl NPP in 1986 were measured in the meat of sheep grazing in a number of upland areas of the United Kingdom, including Cumbria, North Wales, Scotland and Northern Ireland. These elevated  $^{137}\text{Cs}$  activity concentrations led to restrictions on the slaughter and consumption of some sheep meat in affected regions until 2012, over 25 years after the accident [36]. Table 5 summarizes the results of the surveys reviewed.

### 2.3. SUMMARY

The concentrations of uranium and thorium series radionuclides are highly variable and can range over several orders of magnitude. This variability can be as a result of geographical location, climate, agricultural practices, food processing

TABLE 5. SUMMARY OF <sup>137</sup>Cs, <sup>134</sup>Cs AND <sup>90</sup>Sr ACTIVITY CONCENTRATIONS FROM DIET SURVEYS AND MONITORING PROGRAMMES [24–26, 30, 32]

Country	Year(s)	Radionuclide	No. of samples analysed	No. of samples >limit of detection (%)	Activity concentration of samples higher than the limits of detection or reporting levels		
					Min. (Bq/kg)	Max. (Bq/kg)	Median (Bq/kg)
Australia	2019	Cs-137	268	8	0.061	0.389	0.103
Canada	2015–2017	Cs-137	480	0	— <sup>a</sup>	—	—
		Cs-134	479	0			
EU27 + UK	2007–2011	Cs-137 (milk)	17 293	7 <sup>b</sup>	0.5	29	0.85
		Sr-90 (milk)	5 683	0.6 <sup>b</sup>	0.2	44.8	1
Japan	2019–2020	Cs-137	40 486	1	0.393	560	10
		Cs-134		0.02	2.9	37	5.2
USA	2006–2014	Cs-137	3 740	0.1	10.8	93.3	—
		Sr-90	3 498	8	0.1	1.12	0.148

<sup>a</sup> —: data not available.

<sup>b</sup> The percentage of samples reported for the EU27 + UK are those above the European Union defined reporting level and not above a limit of detection.

and the types of foods consumed. For example, the levels of <sup>210</sup>Po in shellfish are typically higher than the levels of <sup>210</sup>Po in terrestrial foods such as grain and milk products. On average, the most significant contributors to ingestion dose are, in descending order, <sup>210</sup>Po, <sup>210</sup>Pb, <sup>228</sup>Ra and <sup>226</sup>Ra (<sup>40</sup>K is also important but not at all amenable to control and therefore excluded from consideration). These four radionuclides have been estimated to contribute over 95% of the ingestion dose from the uranium and thorium series radionuclides and, for this reason, are the radionuclides of primary focus when investigating natural radionuclides in food.

For human-made radionuclides, the majority of over 70 000 measurements conducted in the surveys reviewed are below the limits of detection or reporting

levels for  $^{137}\text{Cs}$ ,  $^{134}\text{Cs}$ ,  $^{90}\text{Sr}$  and other human-made radionuclides, indicating that the human-made radionuclides in food are generally at a level that does not pose a significant health risk to consumers. However, there are specific areas and regions that have been contaminated through fallout from past nuclear weapons testing and nuclear accidents. Both animals and plants from these regions continue to accumulate human-made radionuclides that can subsequently be consumed by individuals, leading to elevated ingestion doses (see also Section 4.5.2). Radiocaesium, particularly  $^{137}\text{Cs}$ , and  $^{90}\text{Sr}$  are the most commonly measured human-made radionuclides in foods, the sources of which are fallout from atmospheric nuclear weapons testing and past nuclear accidents. Additional human-made radionuclides, such as  $^{60}\text{Co}$ ,  $^{106}\text{Ru}$ ,  $^{131}\text{I}$  and  $^{239,240}\text{Pu}$ , are also measured in foods, but these radionuclides are typically only measured as a result of being discharged into the environment from nearby nuclear or research facilities. This highlights the need to consider the discharge of radionuclides from facilities in regions when establishing food monitoring programmes.

The range of activity concentrations reported for  $^{137}\text{Cs}$ ,  $^{134}\text{Cs}$  and  $^{90}\text{Sr}$  varies greatly between the different regions. This could be as a result of the measurement techniques used to measure the food samples and the past practices or events that have occurred in the different regions. For example, Japan is the only country of the regions reviewed that had food samples with measurable concentrations of  $^{134}\text{Cs}$ ; this was a result of the Fukushima Daiichi NPP accident in 2011. This accident also gives rise to the highest median and maximum values of  $^{137}\text{Cs}$  compared to other countries reviewed. The higher activity concentrations noted in the case of Japan are mainly attributed to wild foods and to the results of targeted sampling focusing on items that concentrate radionuclides.

### **3. ESTIMATION OF DIETARY INTAKES**

#### **3.1. INTRODUCTION**

This section outlines the approaches that could be used to estimate the dietary intake of radionuclides in a population. It discusses the determination of appropriate national food consumption data for use in dietary surveys and alternative sources of food consumption data from international studies. The most common practical approaches used to conduct dietary surveys are summarized, including the advantages and disadvantages of each approach. In addition, two alternative approaches are discussed that could be used if food consumption



data are not available or comprehensive dietary surveys are not possible, given resource constraints.

### 3.2. FOOD CONSUMPTION DATA

The dietary intake of radionuclides is often estimated on the basis of dietary assessment methodologies that have been developed primarily for other, non-radiological, purposes. For example, a range of methodologies have been developed to collect nutritional information and assess the relationships between diet and disease. An FAO resource guide has been published [37] that provides an overview of different dietary assessment methods to estimate food consumption data, including their application, validity, strengths and limitations. This resource guide also provides guidance for selecting methodologies, particularly in low resource situations, and identifies two general categories of methodologies:

- (a) Indirect methods that utilize secondary data (e.g. food supply, agricultural statistics, food expenditure) for assessing diets;
- (b) Direct methods that collect primary dietary data from individuals who are representative of a population or population group.

Indirect methods do not provide an indication of individual intakes but are useful for identifying trends in food availability and consumption across different geographical regions.

#### 3.2.1. Indirect methods

The FAO guidance identifies two main types of indirect studies. These are:

- (a) Surveys of national diet based on food balance sheets;
- (b) Household consumption and expenditure surveys.

##### 3.2.1.1. *National food balance sheets*

Each year, the FAO compiles country specific food balance sheet data on the basis of available information for around 100 food groups in approximately 185 countries [38]. Gross national food supply is calculated by adding the total quantity of food produced and the total quantity imported. Adjustment for exports and changes in food stock levels are also included. Net food availability is determined by subtracting the food used for animal feed, seeds and other

purposes as well as losses in the supply chain. Food availability<sup>7</sup>, expressed as kilograms per capita per year, is calculated by dividing net food availability by a country's population. Food availability can be linked to food composition data and presented as per capita energy intake (kilocalories per day), protein intake (grams per day) and fat intake (grams per day).

These data provide an overview of national intakes and provide an indication of overall trends. However, they do not provide information on individual variability or reflect seasonal changes. They do not include information on non-purchased foods (e.g. wild foods or food consumed from private gardens). As the FAO balance sheets are based on trade, the consumption data for water include only bottled water and exclude other water sources, such as public and private water supplies. As such, the consumption data for water could be highly inaccurate. Nevertheless, these data provide a valuable basis for assessing intakes on a national level and provide the basis of other analyses, as outlined in more detail below.

#### *3.2.1.2. Household consumption and expenditure*

Household food consumption has been defined as “the total amount of food available for consumption in the household, generally excluding food eaten away from the home unless taken from home” [39]. Participant households typically keep a record of expenditure, quantity of food purchased and types of food consumed during a defined time period, for example one to four weeks. These surveys provide an evaluation of food consumption at the household level. Surveys of this type are routinely conducted in countries to determine socioeconomic information, such as consumer price indices, and to investigate trends in poverty and income distribution [40]. These household surveys are unable to provide detailed information on the variability of food consumption within the same household and they do not take cooking methods or food losses into consideration. However, this form of survey is a relatively inexpensive and straightforward approach for tracking food consumption patterns in a household compared to direct methods (Section 3.2.2).

As an example, the National Food Survey, undertaken in the UK between 1940 and 2000, was one of the longest running continuous surveys of household food consumption and expenditure in the world [41]. Each year, data were collected for individual households for a period of seven days using a log book that was completed by the person responsible for domestic food arrangements

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<sup>7</sup> Food availability is defined by the FAO as the availability of sufficient quantities of food of appropriate quality, supplied through domestic production or imports (including food aid).

in each household. This information was used for various purposes, including evaluating temporal trends in food expenditure and consumption. The data collected were also used to derive representative national food intake rates, for the purpose of assessing intakes of radionuclides. This included deconstructing the components of combined foods using standardized recipes to provide generic intake rates for the main food groups, which could be used in the absence of specific intake data [42].

### **3.2.2. Direct methods**

Direct methods can be classified into two groups:

- (a) Prospective methods, involving recording the diet at the time of consumption;
- (b) Retrospective methods on the basis of recall after the food has been consumed, such as 24 hour recall and food frequency questionnaires.

These methods are used, among other things, to identify trends in food consumption, food and nutrient intakes and to evaluate associations between diet and disease [37].

Retrospective methods, such as food frequency questionnaires, have the disadvantage of being dependent on the memory of the respondents and their ability to remember all food product and portion sizes consumed over the reference period. However, these methods are considered to be a 'time effective' method and are commonly used, for example, as part of large epidemiological studies. The FAO provides a detailed description of the advantages and disadvantages of these and other methods [37].

Prospective methods include estimated food records, weighed food records and duplicate diet methods. The latter methods are relatively resource intensive, depending on the amount of detail needed in the survey.

The estimated food record approach involves respondents continuously documenting all foods and beverages consumed during a predefined period (e.g. one to seven days). The respondents may provide information on food preparation, cooking methods and mixed dishes. This method provides information on food and nutrient intakes at an individual level, although there may be a self-reporting bias, as respondents may be selective with the foods they choose to report. However, it is possible to validate estimated food records by using weighed food records and biomarkers.

The weighed food record involves respondents weighing and recording all foods and beverages consumed for a measurement period of between one and seven days. Leftovers may also be weighed or estimated in this process. Foods consumed away from home may also be recorded separately, and possibly

weighed, depending on the design of the survey. This approach places a high burden on both the respondents and researchers but provides perhaps the most precise measure of individuals' food and nutrient intake.

### **3.2.3. International food consumption data**

Food safety authorities in many countries undertake national dietary surveys, as outlined in Sections 3.1.1 and 3.1.2. This may be infrequent or not always possible due to a lack of resources. In these cases, countries can use internationally compiled consumption data, such as FAO food balance sheets (FAOSTAT), the WHO Global Environment Monitoring System — Food Contamination Monitoring and Assessment Programme, commonly known as GEMS/Food cluster diets, or a modified form of these cluster diets used by UNSCEAR.

#### *3.2.3.1. FAO food balance sheets (FAOSTAT)*

Every year, the FAO requests food balance sheets (as described in Section 3.2.1.1) from its member countries. It then compiles the data and makes them available online [38]. These data are typically raw or semiprocessed agricultural commodities and provide an estimate of the mean quantities of foods available for consumption in each country. However, waste at the household and individual level cannot be accounted for, and therefore consumption rates that are based on this approach are higher than estimates based on actual food consumption data.

#### *3.2.3.2. WHO GEMS/Food cluster diets*

WHO has established the Global Environment Monitoring System — Food Contamination Monitoring and Assessment Programme, commonly known as GEMS/Food, to provide information on the levels of chemical contaminants in food and the consequent dietary intakes. This information is collected from worldwide collaboration centres and recognized national institutions. As part of this work, WHO has developed an approach to describe the various diets around the world based on the analysis of food availability from FAO food balance sheets [38]. The WHO GEMS/Food cluster diets dashboard maintains a suite of 17 'cluster diets', which has been established on the basis of similarities in national dietary patterns rather than being grouped regionally [43]. These are shown in Fig. 2. WHO also hosts this information in an online database containing summary statistics for 37 national food surveys from 26 countries [44].

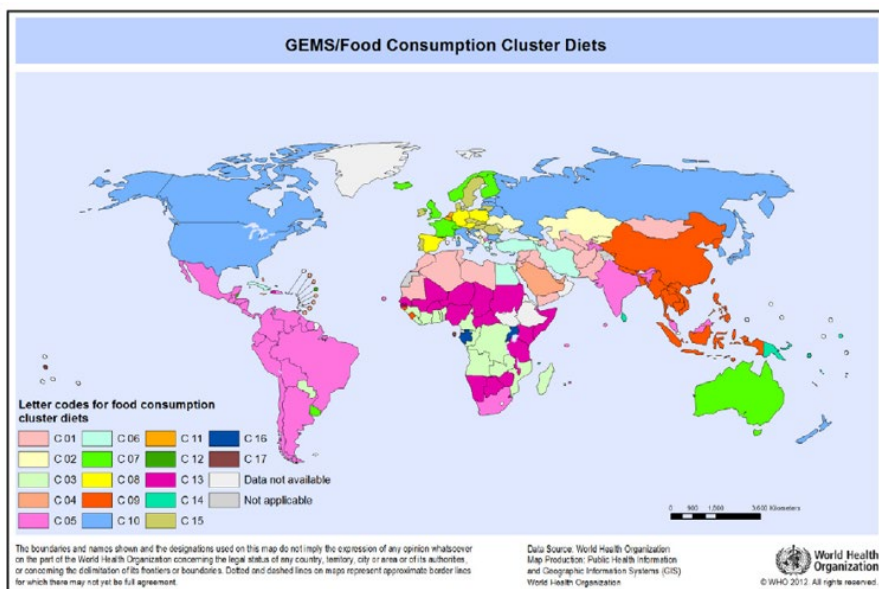


FIG. 2. WHO GEMS/Food cluster diet regions [43]. Reproduced with permission from WHO.

The WHO GEMS/Food databases thus provide a useful starting point for understanding national food consumption rates for the purpose of assessing intakes of radionuclides, particularly in the absence of other data. However, these cluster diets are not suitable to assess the dietary intakes of specific populations, for example children, or to estimate intakes of food in periods shorter than a lifetime.

### 3.2.3.3. UNSCEAR 2016 regional data

UNSCEAR has developed a method for estimating public exposures due to radioactive discharges, in part to allow an assessment of exposures arising from different forms of electricity generation. This was published in 2016 [45]. Information regarding the location and habits of the population, specific to the regions in which discharges occurred, were collated. Consumption rates were therefore derived for the regions for which population distribution information was available from the UNEP Division of Early Warning and Assessment Global Resource Information Database (GRID) [46]. The categories of terrestrial foods were simplified into four categories on the basis of radionuclide transfer factors and general consumption habits worldwide. The broad regions were categorized as follows: Polar (Arctic and Antarctic), North America, Latin America and

TABLE 6. ANNUAL AVERAGE PER CAPITA CONSUMPTION RATES FOR TERRESTRIAL FOODS BY UNEP REGION [45]

UNEP region	Annual average per capita consumption rate (kg)							
	Cereals	Vegetables and fruit	Milk and dairy products	Meat and offal	Freshwater fish	Marine fish	Crustaceans	Molluscs
Africa	130	220	31	17	2.7	6.6	0.1	<0.02
Asia and Pacific	140	240	45	30	7.8	6.9	1.4	2.4
Europe	110	280	120	65	3.4	13	0.9	1.0
Latin America and Caribbean	110	210	77	63	1.6	5.9	0.6	0.4
North America	88	260	120	100	4.5	8.2	2.8	1.4
West Asia	140	180	43	34	1.3	4.5	0.3	<0.02
World average <sup>a</sup>	130	230	65	44	5.7	7.5	1.1	1.6

<sup>a</sup> Worldwide average of the values for each region weighted by population density.

the Caribbean, Africa, Europe, West Asia, Asia and the Pacific. Representative consumption rates for terrestrial and aquatic foods were derived by reanalysing the information from the WHO GEMS/Food database, which is in turn derived from the FAO food balance sheets [47]. These are indicative of habits averaged over the entire population. Population weighted world average values were derived for terrestrial foods, freshwater fish and marine foods (Table 6). These data are generalized values that were developed for a specific assessment and therefore do not represent individual consumption habits. However, they may be a useful starting point for understanding the possible intakes of key food groups and for designing more detailed investigations.

### 3.3. DIETARY SAMPLING METHODS

Various approaches can be used to estimate the intakes of radioactivity from diet, depending on the various approaches for estimating food consumption outlined in Section 3.1. The choice of which method to use depends on the objectives and resources available. This section outlines five approaches that could be used for the monitoring of nutrients, contaminants, chemical substances and residues and, in this case, radionuclides. These are total diet studies, market basket studies, duplicate diet studies, canteen meal studies and monitoring of individual foods.

#### 3.3.1. Total diet and market basket studies

A total diet study (TDS) consists of selecting, collecting and analysing commonly consumed food products purchased at a retail level. The purchased food is processed for consumption, and representative food groups are pooled into proportions consumed and homogenized prior to analysis for radionuclides. The food samples are analysed for radionuclides and doses are calculated using consumption rate data representative of the local, regional or national population.

For a market basket study (MBS), a large selection of food products are collected from consumer points of sale in proportions approximating the consumption patterns of interest. Minimal sample preparation methods are conducted prior to analysis (e.g. Codex guidelines methods for analysis and sampling [48]). This differs from the TDS approach, as the samples are analysed as purchased, as opposed to as consumed.

TDSs first started in the USA to assess intakes of radionuclides, notably  $^{137}\text{Cs}$  and  $^{90}\text{Sr}$ , from fallout from nuclear weapons testing. The survey was initiated in the late 1950s with the analysis of milk and was extended in the early 1960s to cover the total diet using food collected from supermarkets, grocery

stores and fast food restaurants. Foods were prepared for analysis to represent how they were consumed [49].

Sometimes national authorities undertake MBSs and TDSs to estimate intakes of radionuclides via food by different groups of the population (e.g. by age and sex). In this case, foods that represent the majority (typically 95%) of the national diet are purchased according to their contribution to the diet, assigned to aggregated or individual food groups and analysed for radionuclides in order to estimate the total exposure from all food groups. An MBS or TDS involves establishing the following elements: the list of foods to be considered, the potential radionuclides to consider, data on food consumption and the necessary resources (including laboratory facilities and, in the case of a TDS, kitchen facilities).

These approaches have the potential to provide information on average intakes and on the contribution of individual food groups to the total intake of radionuclides. It is possible to adapt the methodology to take account of regional differences, but this approach is not generally used to target specific population groups. It is, however, possible to use this approach to identify situations that may warrant further study, for example by a duplicate diet study.

Further information on TDSs and MBSs can be found in guidance provided by WHO, the FAO and the European Food Safety Authority [50, 51], and elsewhere [52].

Following the accident at the Chornobyl NPP in 1986, Sweden monitored levels of  $^{137}\text{Cs}$  in food, which included the collection of market basket samples in eight major towns across the country [53]. This is also an approach that was used extensively in Japan after the Fukushima Daiichi NPP accident in 2011 [54].

TDSs are no longer generally used for measuring radionuclides in the total diet due to the very low levels of human-made radionuclides in foods from fallout from global atmospheric weapons testing, which was the primary focus of such assessments. For example, Canada stopped using TDSs for radionuclides in 2007, having had a programme since 1992. The US continues to measure  $^{137}\text{Cs}$  and  $^{90}\text{Sr}$  in diet samples in four regions, selecting market baskets from three cities in each region per year.

Some special studies have been carried out. An example is a study in Lebanon, where gamma emitting radionuclides were measured in 77 foods combined into 12 food groups [55].

Few countries have included naturally occurring radionuclides in their TDS programmes. In 1998 measurements of  $^{210}\text{Po}$ ,  $^{226}\text{Ra}$  and  $^{232}\text{Th}$  were made in the market baskets collected in the USA, and all concentrations were below the limits of detection [56]. China included  $^{210}\text{Pb}$ ,  $^{210}\text{Po}$ ,  $^{226}\text{Ra}$  and  $^{228}\text{Ra}$  in the first two years of its TDS programme to obtain a baseline of activity concentrations in foods [57]. The UK has in the past measured activity concentrations of some



naturally occurring radionuclides in the diet as part of its monitoring programme, but this was last reported in 2009 [58].

The advantages of the TDS and MBS approaches are that they provide easily understandable information on the dietary intake of radionuclides in food for the use of regulatory authorities and the public, and they are also able to identify which food groups are the principal sources of particular radionuclides. In addition, if analyses are conducted on individual foods collected, it allows the assessment of doses received by the entire population or by selected populations, as long as food consumption data are available for each of the population groups of interest. For TDSs, the methodology also takes into account the effect of food preparation on the radioactivity levels in foods.

The disadvantages of the TDS and MBS approaches are that they cannot be used to estimate ingestion of radionuclides at the individual level or for small groups that are deemed high risk, since these studies are based on average food consumption rates. In addition, this approach can involve significant resources, depending on the number of food groups being considered and whether the studies are ongoing; especially for TDSs, where food preparation is necessary prior to analysis.

### **3.3.2. Duplicate diet and canteen meal studies**

In duplicate diet studies (DDSs) and canteen meal studies (CMSs), participants are requested both to keep a record of all foods and beverages consumed and to retain duplicate portions throughout a specific time period. This approach provides a direct assessment of intakes of nutrients and contaminants in individual diets in the form in which the food was prepared. These studies can also be part of the food record and weighed food record studies mentioned previously and can be used in studies of well defined populations. Both duplicate diet and canteen meal studies can be supplemented with additional information, such as a record of food consumed and photographs of the food consumed.

These studies can be conducted by participants in households or in large canteens or restaurants to estimate radionuclides ingested for a particular population group. The two approaches are defined as follows:

- (a) The DDS is a method for estimating dietary intake through the collection and analysis of identical portions of foods and beverages consumed by an individual over a period of time. The whole diet consumed is combined and analysed for radionuclides and doses are calculated.
- (b) In a CMS, prepared (as consumed) meals are purchased in a restaurant (e.g. a university dining hall or business cafeteria, etc.) and pooled together (all items: meats, vegetables, fruits, beverages, etc.) for analysis. This may

be a one time collection or may be over a period of time. The meals are analysed for radionuclides, and doses are calculated.

Examples of the DDS approach can be seen in the Aomori Prefecture in Japan, where 80 duplicate diet samples were collected from 100 participants over a period of four years and analysed for 11 natural and human-made radionuclides to estimate the ingestion dose of office workers, fishers and agricultural workers [59]. A baseline survey of background radiation levels in Karnataka on the south-west coast of India also used the DDS approach to estimate the levels of seven natural and human-made radionuclides in vegetarian and non-vegetarian meals consumed by the local population [60].

The CMS approach is the recommended approach for the analysis of complete meals in the EU for the purposes of assessing dose to the population [61]. In 2014, Ireland used the CMS approach to estimate the ingestion dose from natural radioactivity in food through the collection of meals from a large restaurant facility on a university campus [62]. This approach has also been used in Seville, Spain [63], where 24 representative diet samples from a university canteen were collected over a six year period from 2007 to 2012 and analysed for  $^{210}\text{Po}$ . Samples of complete meals for breakfast, lunch and dinner were collected over a five day period once every three months. A composite diet sample was produced from the five days of sampling to represent the sample for each quarter of the year. Although not specifically addressed in the paper, this suggests that dietary choice and the (seasonal) variability of specific foods can be important parameters in determining individual dose.

The advantages of the duplicate diet or canteen survey approach is that analysis is undertaken on food as it would actually be consumed. Therefore, any changes in radionuclide concentration in food due to post-production practices and subsequent processing and preparation have already taken place. In addition, detailed food consumption data are not needed. Furthermore, if estimating exposure to small groups or individuals, these approaches involve relatively few resources.

The disadvantages of the approach are that the studies are typically suitable only for a small or specific group of any given population. It is also very resource intensive for participants to purchase and prepare duplicate meals and, as such, these types of studies can rarely be conducted for more than seven days. For this reason, the data may not be representative of long term consumption or exposure patterns.

### **3.3.3. Monitoring of individual food products**

It would be difficult to monitor radionuclide levels in every different individual food product that makes up a varied diet. Although it is possible that some non-staple food products could have enhanced levels of radionuclides, they are unlikely to be eaten in large amounts. The estimate of radionuclide intake will therefore be reasonable if only staple food products are considered. For this reason, this approach is ideally suited to estimate intakes in communities or population groups with diets that are dominated by a limited range of staple food products. An example is in the estimation of intakes for infants or young children. For more complex diets, additional criteria would need to be applied to ensure the representativeness of the diet, which leads to more complex sampling strategies on a much larger scale either geographically, or in terms of the number of food samples needed, or both.

The types of food products requiring analysis as part of these studies include those that are consumed in large quantities by the population or group of interest and food products that may not be consumed regularly but could potentially contain high levels of radioactivity (Section 5). Therefore, some information on the amounts and types of food products consumed is a prerequisite for conducting surveys using individual foods. In addition, if a realistic estimate of intakes of radionuclides is needed, then appropriate processing and preparation of food, such as washing and cooking, is also necessary.

This approach is more accurate than the total diet/market basket approach that sometimes uses composite samples that can result in the dilution of radionuclides to a level below limits of detection if there is a large range of radionuclide concentrations in different foods. When analysing individual food products, there are no such dilution effects. In addition, individual foods containing elevated levels of radioactivity are more easily identified. The individual food product approach also allows the easy identification of population groups of greater risk.

The main disadvantage of this approach is the requirement for analytical resources. The analysis of individual food products as opposed to composite or duplicate meals requires significant analytical capabilities and resources to process a large number of individual food samples.

### **3.3.4. Summary of dietary sampling methods**

There is no preferred or recommended approach for sampling food to estimate radioactivity in diets. The various survey approaches described have various advantages and disadvantages that need to be considered when selecting the appropriate survey for the determination of intake of radioactivity in the diet.

The choice of approach also needs to take into account the resources available to conduct such surveys, whether surveys are already being conducted that could be extended to include the measurement of radioactivity and whether data currently exist on the levels of radioactivity in foods. Some of the considerations involved in selecting the most suitable dietary survey for radioactivity in food are outlined in Table 7.

The total diet–market basket approach is routinely used in countries for the monitoring of contaminants in foods and to determine intakes in the diet of the whole population. If such surveys already exist, then it may be suitable to extend the survey for the collection and analysis of foods or composite meals

TABLE 7. SUMMARY OF FACTORS FOR CONSIDERATION WHEN CHOOSING SAMPLING SURVEYS

Method		TDS <sup>a</sup> /MBS <sup>b</sup>	DDS <sup>c</sup> /CMS <sup>d</sup>
Suitability of approach	Large scale	Suitable	Unsuitable
	Small scale	Unsuitable	Suitable
Participant burden		n.a. <sup>e</sup>	Large
Field staff burden		Medium: extensive sample collection, storage and transport	Large: two or more house visits, food storage instructions, storage containers
Individual consumption data provided		No	Yes
Processing of consumption data		Yes	No
Additional comments		Food consumption data needed	Cost of paying participants for food
Timescale		Medium to long term	Short term

<sup>a</sup> TDS: total diet study.  
<sup>b</sup> MBS: market basket study.  
<sup>c</sup> DDS: duplicate diet study.  
<sup>d</sup> CMS: canteen meal study.  
<sup>e</sup> n.a.: not applicable.

for radionuclides, as the organizers of such surveys may already have sufficient resources to conduct such a survey and will have collated sufficient data on the consumption patterns of the population as a whole. These surveys provide a more accurate estimate of dietary intake when compared to other approaches, but they are also more complex and costly. For example, the German Federal Institute for Risk Assessment is conducting a TDS for the German population, and radionuclides in food are being assessed as part of this study [64]. To establish and maintain these types of surveys requires considerable resources on an ongoing basis. As such, this approach may not be suitable for the exclusive monitoring of radioactivity in food. In addition, as these types of studies are primarily focused on the entire population, they may not be suitable for smaller population groups, such as the more highly exposed members of the population.

The duplicate diet approach is suitable for small groups of the population, such as the more highly exposed members of the population and does not involve any consumption data or require consumption surveys to be conducted in advance. Ideally, duplicate food samples are taken from individuals who are representative of the most highly exposed members of the population, namely the 'representative person'. The representative person could be determined through the use of habits surveys and, once identified, it may be possible for such individuals to conduct a DDS or to have one conducted on their behalf, for example, if the representative person is an infant. However, this may not be practical and requires significant effort and responsibility on the part of the participants.

Individual food surveys can be conducted for the population as a whole or for the more highly exposed members of the population, but consumption data are needed for any survey of this type. If these are not already available, a consumption survey would need to be established prior to any sampling or analysis being carried out. Significant analytical resources are also needed for the analysis of individual food products on an ongoing basis, and these may not always be available.

In the absence of extensive resources, pre-existing food surveys, habit surveys or information on the levels of radioactivity in staple foods, the most straightforward approach for establishing intakes of radionuclides in the diet would be either the duplicate diet or the canteen meal approach. To overcome any issues related to reliance on participants, the canteen meal approach could be used. This negates the need for active participation by consumers in that food collected is already prepared for consumption, and food consumption data are not needed. The issue related to long term food consumption habits could be addressed through routine sampling periodically, for example, by collecting food samples over a period of a year for one week every quarter.

More information on the selection of the appropriate approach for the sampling of food for ingestion dose along with their advantages and

disadvantages can be found in the WHO Guidelines for the Study of Dietary Intakes of Chemical Contaminants [65].

### **3.3.5. Other approaches**

#### *3.3.5.1. Site specific habit surveys*

In situations where there is an identified source of radionuclides, it may be helpful to undertake focused surveys to gather information on the habits of people close to, or affected by, that source [66]. Such surveys may be designed to evaluate the habits of a given community or of those most likely to have the highest intake of radionuclides or receive the highest exposures from that source due to their location or behaviour (often referred to as the critical group or representative person). Such surveys are sometimes undertaken around nuclear facilities in order to determine the extent to which people may receive radiation doses via the different exposure pathways. These surveys may also be designed to estimate the dietary intake rates of local foods and may be targeted at collecting information on the intakes of specific foods by specific groups of people. For example, a habit survey conducted in the vicinity of a naval dockyard site (that contained radioactive waste as a result of the refitting and dismantling of nuclear submarines) in Rosyth, UK, derived adult, infant and child consumption rates for foods consumed from the survey area of interest [67].

Habit surveys may also be combined with monitoring programmes to provide information about both the intakes of key local foods and the activity concentrations of radionuclides within those foods. However, these surveys are resource intensive and are likely to be warranted only in situations where there is a significant identified or suspected source of radionuclides.

#### *3.3.5.2. Use of transfer factors*

In the absence of relevant information on consumption patterns or the levels of radionuclides in food consumed by individuals or the population, an alternative approach can be utilized to estimate the levels of radioactivity in food for human consumption. The IAEA has published two technical reports on the transfer of radionuclides in terrestrial and freshwater environments (Technical Reports Series No. 472) [68] and concentration factors for biota in the marine environment (Technical Reports Series No. 422) [69]. These publications use the concepts of the soil to plant transfer factor ( $F_v$ ) and the concentration ratio ( $CR$ ), which can be used to estimate the transfer of radionuclides to plants and animals in the terrestrial environment, while the concentration factor ( $CF$ ) is used to estimate the transfer of radioactivity to biota in the marine environment.

The applicability and limitations of these approaches are also outlined in these publications.

Technical Reports Series No. 472 includes  $F_v$  values for vegetables (leafy, non-leafy and root), grains and fruits for temperate, non-temperate, subtropical, tropical and alpine ecosystems.  $CR$  values for agricultural systems are available that estimate the transfer of radionuclides to animals and animal products, including milk, meat and eggs. The transfer of radionuclides from the environment to wild foods such as mushrooms, berries and game is also addressed.

The information available on freshwater ecosystems includes transfer factors for edible aquatic plants and  $CR$  values for freshwater invertebrates and freshwater fish tissues. In addition, information is also provided on the application of food processing activities that can affect the concentrations of radionuclides in food. Technical Reports Series No. 422 provides  $CF$  values for biota in the marine environment with  $CF$  values derived in accordance with activity concentrations in filtered seawater. The  $CF$  values published include fish, crustaceans, molluscs, macroalgae (seaweed), cephalopods and mammals such as seals that can be consumed by indigenous populations.

The  $F_v$ ,  $CR$  and  $CF$  values are based on numerous assumptions, for example that the activity concentration in biota is in equilibrium with its surroundings, which is rarely the case, and, for some radionuclides, these factors are derived from those that are analogous to the element in question. Furthermore, appropriate soil, river sediment and marine sediment measurement data in the area or region of interest also need to be available to use this approach in order to estimate radioactivity concentrations in foods. As a result of the assumptions made and the potential variability in soil and sediment measurement data, these values have large uncertainties associated with them and are only to be used in the event that no direct information is available on the levels of radioactivity in food. However, measurements made in soil and sediments could also be used as a scoping tool in identifying locations or regions that have the potential to transfer elevated levels of radionuclides to foods produced in that area. This would assist in the development of a targeted food monitoring programme in areas where resources are limited.

### **3.3.6. Discussion**

To conduct an appropriate assessment of ingestion doses from radionuclides in food, information is needed on the amount and types of food consumed by the population, population group or individuals of interest. Food consumption data may be published by national authorities, such as food regulators, and these could be used for radionuclide intake assessments. These national consumption data can be derived using indirect or direct methods through the use of national

food balance sheets or food frequency questionnaires, for example. If national consumption data are not available, other sources of food consumption data are available from the FAO, WHO, UNSCEAR and UNEP. However, these are normally compiled from data from several countries, and this needs to be accounted for, to the extent possible, in radionuclide intake assessments.

The choice of food sampling techniques used for radionuclide intake assessment is dependent upon a number of factors, including the purpose and scale of the study, the target population and the resources available. Several different approaches have been outlined, along with the advantages and disadvantages of each. In the absence of information or data on activity concentrations in foods, soil to plant transfer factors, concentration ratios or concentration factors can be used to estimate the activity concentration in edible portions of foods consumed by the population.

## **4. ANALYSIS OF DIETARY EXPOSURE STUDIES AND PATHWAYS**

### **4.1. INTRODUCTION**

This section reviews and summarizes previously published studies on radiation doses from the diet and in natural mineral waters, identifying the radionuclides of interest from their contribution to annual effective dose [5], hereafter referred to as dose.<sup>8</sup> The dietary exposure pathways that have the potential to be significant contributors to ingestion dose are also discussed; these are, in general, related to fish and fish products, aquatic plants, wild foods and game animals.

### **4.2. DIETARY DOSE STUDIES**

An evaluation of previously published studies of doses from dietary intake was conducted to evaluate the dose arising from the ingestion of radioactivity in foods and identify the significant natural and human-made radionuclides contributing to dose from the diet. The variability of ingestion dose as a function of age and dietary study types was also investigated.

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<sup>8</sup> These evaluations are dependent on the literature surveyed.



#### 4.2.1. Literature survey

A literature survey was conducted to identify published studies on ingestion dose from radionuclides in the diet. This literature survey identified 217 scientific papers and reports covering the period from 1957 to 2019. These papers and reports were reviewed to determine whether they contained sufficient information and data for inclusion in the evaluation. The criteria for the inclusion of published studies in the evaluation were as follows:

- (a) The dose assessment was based on estimates of dose from the total diet from one or more radionuclides. Studies where only a single or a few foods were considered were excluded.
- (b) Studies stating that the foods considered in the dose assessment reflected the majority of the diet were considered in conjunction with the quality of the paper.
- (c) Doses from drinking water were excluded as being outside the scope of the review.
- (d) Doses from  $^{40}\text{K}$  were excluded.<sup>9</sup>

In total, 127 of the 217 publications identified by the literature survey met the above criteria and were included in the evaluation. These 127 publications covered 46 countries worldwide.

The studies included in the evaluation fell into the four main categories that were broadly defined by the methodology used to collect the food samples (TDS, DDS, MBS and CMS). A summary of the key elements of these study types is given in Section 3. Table 8 shows the type of assessment, the countries where the ingestion dose was assessed and the number of studies of each type that were undertaken. Some publications reported doses from more than one type of assessment approach, resulting in 158 surveys from the 127 publications.

The dose assessments listed in Table 8 included both natural and human-made radionuclides. Table 9 shows the countries in which studies have been undertaken on natural or human-made radionuclides, or both.

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<sup>9</sup> In cases where the dose from  $^{40}\text{K}$  was included in the total dose, the dose attributable to  $^{40}\text{K}$  was subtracted from the total dose using data available in the publication or using the UNSCEAR estimates of 165  $\mu\text{Sv}/\text{year}$  (adults) and 185  $\mu\text{Sv}/\text{year}$  (children) for the dose arising from  $^{40}\text{K}$  [16].

TABLE 8. COUNTRIES THAT HAVE CARRIED OUT THE DIFFERENT TYPES OF DIETARY ASSESSMENTS FROM THE LITERATURE SURVEY

Assessment type <sup>a</sup>	No. of countries	Countries (No. of studies)
CMS	8	Austria (1), Cuba (1), Germany (1), India (1), Ireland (1), Poland (1), Spain (2), United States of America (1)
DDS	9	Brazil (1), China (3) <sup>b</sup> , India (5), Japan (18), Poland (1), Russian Federation (1), Ukraine (2), United Kingdom (5), United States of America (1)
MBS	34	Bangladesh (1), Belarus (1), Brazil (3), China (3) <sup>c</sup> , Costa Rica (1), Croatia (1), Cuba (2), Denmark (3) <sup>d</sup> , Finland (1), France (1), Ghana (1), Iceland (1), India (10), Iran, Islam. Rep. (2), Israel (1), Italy (2), Japan (9), Republic of Korea (4), Kuwait (1), Marshall Islands (1), Morocco (1), New Zealand (1), Norway (3), Pakistan (4), Philippines (2), Poland (3), Romania (1), Russian Federation (1), Spain (5), Sudan (1), Sweden (1), Syrian Arab Republic (1), United States of America (4), Viet Nam (2)
TDS	17	Australia (1), Bangladesh (1), Brazil (2), China (3), Germany (1), India (3), Italy (3), Japan (5), Republic of Korea (2), Lebanon (1), Pakistan (1), Philippines (1), Portugal (1), Sri Lanka (1), United Kingdom (2), United States of America (4), Viet Nam (1).

<sup>a</sup> CMS: canteen meal study; DDS: duplicate diet study; MBS: market basket study; TDS: total diet study.

<sup>b</sup> Including one study conducted in Taiwan, China.

<sup>c</sup> Including two studies conducted in Taiwan, China.

<sup>d</sup> Including one study conducted in the Faroe Islands and one study conducted in Greenland.

The radionuclides included in the dose assessments were:

- Twelve natural radionuclides (excluding <sup>40</sup>K): <sup>87</sup>Rb, <sup>210</sup>Pb, <sup>210</sup>Po, <sup>224</sup>Ra, <sup>226</sup>Ra, <sup>228</sup>Ra, <sup>228</sup>Th, <sup>230</sup>Th, <sup>232</sup>Th, <sup>234</sup>U, <sup>235</sup>U and <sup>238</sup>U;
- Eleven human-made<sup>10</sup> radionuclides: <sup>3</sup>H, <sup>14</sup>C, <sup>35</sup>S, <sup>60</sup>Co, <sup>90</sup>Sr, <sup>129</sup>I, <sup>131</sup>I, <sup>134</sup>Cs, <sup>137</sup>Cs, <sup>239+240</sup>Pu and <sup>241</sup>Am.

<sup>10</sup> Both <sup>3</sup>H and <sup>14</sup>C occur naturally and as a result of human activities. For the purposes of this survey, they were considered to be human-made.

TABLE 9. COUNTRIES WHERE THERE ARE DIETARY ASSESSMENTS FOR NATURAL RADIONUCLIDES AND/OR HUMAN-MADE RADIONUCLIDES

Radionuclides	No. of countries	Countries (No. of studies)
Natural	23	Bangladesh (1), Brazil (5), China (2), Cuba (1), France (1), Germany (1), Ghana (1), India (13), Italy (3), Japan (4), Korea, Rep. (4), Marshall Islands (1), Morocco (1), Pakistan (4), Poland (3), Portugal (1), Philippines (2), Romania (1), Spain (4), Syrian Arab Republic (1), United Kingdom (2), United States of America (1), Viet Nam (1)
Human-made	23	Belarus (1), China (1) <sup>a</sup> , Costa Rica (1), Croatia (1), Denmark (3) <sup>b</sup> , Finland (1), Germany (1), Iceland (1), Iran, Islam. Rep. (1), Israel (1), Italy (2), Japan (20), Korea, Rep. (1), Lebanon (1), Norway (1), Pakistan (1), Philippines (1), Russian Federation (2), Sudan (1), Sweden (1), Ukraine (1), United Kingdom (5), United States of America (8)
Both natural and human-made	15	Australia (1), Austria (1), Brazil (1), China (5) <sup>c</sup> , Cuba (2), India (3), Iran, Islam. Rep. (1), Ireland (1), Japan (3), Kuwait (1), New Zealand (1), Norway (2), Spain (2), Sri Lanka (1), Ukraine (1)

<sup>a</sup> Study conducted in Taiwan, China.

<sup>b</sup> Including one study conducted in the Faroe Islands and one study conducted in Greenland.

<sup>c</sup> Including one study conducted in Taiwan, China.

In general, studies that have included natural radionuclides have primarily focused on the uranium and thorium series radionuclides. Specifically, <sup>210</sup>Po, <sup>210</sup>Pb, <sup>226</sup>Ra and <sup>228</sup>Ra have been measured, as these radionuclides are known to be significant contributors to dose from the diet [16]. The studies that include human-made radionuclides are principally concerned with <sup>137</sup>Cs and <sup>90</sup>Sr due to their presence in the environment as a result of nuclear weapons fallout and past nuclear accidents, such as those at the Chornobyl and Fukushima Daiichi NPPs.

#### 4.2.2. Total doses

A number of the surveys reviewed as part of the evaluation estimated the total annual dose from the diet at a national or regional level. A summary of these survey estimates of total annual dose from all radionuclides (excluding the dose from <sup>40</sup>K) is given in Table 10.

TABLE 10. TOTAL ANNUAL EFFECTIVE DOSE FROM ALL RADIONUCLIDES IN THE DIET (EXCLUDING <sup>40</sup>K)

Country	ICRP age group	Total dose (mSv/year)	Notes
Australia [24]	Infant (<1 year)	0.26	Fed with milk formula
	Infant (<1 year)	0.02	Fed with breast milk
	Child (1–2 years)	0.30	
	Child (2–7 years)	0.18	
	Child (7–12 years)	0.19	
	Child (12–17 years)	0.22	
	Adult (>17 years)	0.06	
Austria* [70]	Adult (>17 years)	0.33	99.6% from natural radioactivity
Brazil [71]	Adult (>17 years)	0.44	São Paulo region
Brazil [72]	Adult (>17 years)	0.43	Rural areas
	Adult (>17 years)	0.29	Urban areas
France [73]	Adult (>17 years)	0.32	Typical seafood consumer nationally
	Adult (>17 years)	0.19	Light seafood consumer nationally
	Adult (>17 years)	0.73	Typical seafood consumer at seaside sites
	Adult (>17 years)	≤2.6	High rate seafood consumer at seaside sites
Germany [74]	Infant (<1 year)	0.27	
	Child (1–2 years)	0.18	
	Child (2–7 years)	0.14	
	Child (7–12 years)	0.14	
	Child (12–17 years)	0.16	
	Adult (>17 years)	0.04	
Ireland [75]	Adult (>17 years)	0.10	95% from natural radioactivity
Japan [76]	Adult (>17 years)	0.43	96% from natural radioactivity

TABLE 10. TOTAL ANNUAL EFFECTIVE DOSE FROM ALL RADIONUCLIDES IN THE DIET (EXCLUDING <sup>40</sup>K) (cont.)

Country	ICRP age group	Total dose (mSv/year)	Notes
New Zealand [77]	Child (1–2 Years)	<0.12	86% from natural radioactivity
	Child (2–7 years)	<0.09	85% from natural radioactivity
	Child (12–17 years)	<0.07	Female, 82% from natural radioactivity
	Child (12–17 years)	<0.05	Male, 81% from natural radioactivity
	Adult (>17 years)	<0.09	Female, 85% from natural radioactivity
	Adult (>17 years)	<0.15	Male, 90% from natural radioactivity
Norway* [78]	Infant (<1 year)	0.34	98% from natural radioactivity
	Child (12–17 years)	0.25	98% from natural radioactivity
	Adult (>17 years)	0.19	98% from natural radioactivity
Norway [79]	Infant (<1 year)	0.39	99% from natural radioactivity
	Adult (>17 years)	0.29	95% from natural radioactivity
United Kingdom [80]	Adult (>17 years)	0.19	92% from natural radioactivity
Viet Nam* [81]	Adult (>17 years)	0.20	Red River Delta region

\* The <sup>40</sup>K dose was subtracted from the estimated reported dose in these studies.

For some of these surveys, where information was not explicitly given regarding the contribution to the dose from natural radionuclides, the assumption has been made that the total dose is solely from natural radionuclides, with the contribution from human-made radionuclides being negligible. The surveys in Austria, Ireland, Japan, New Zealand, Norway and the United Kingdom have derived the dose arising from both natural and human-made radionuclides separately.

The summary of the results outlined in Table 10 indicates that the overall annual effective dose from natural and human-made radioactivity in the diet is typically well below 1 mSv/year. The only exception is in the study in France for a cohort of adults living in seaside towns who consume large quantities of

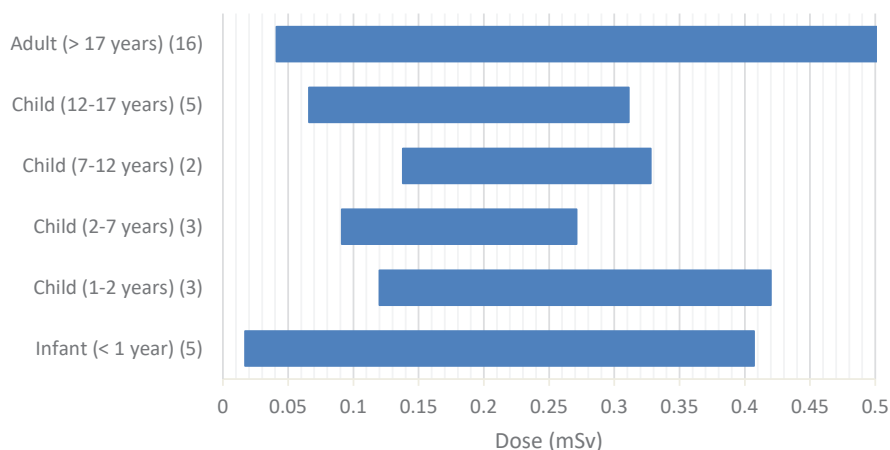


FIG. 3. Range of dose from ingestion for ICRP age groups with the number of dose estimates for each age group in parentheses.

seafood [73]. However, the authors state that for the majority of the French population the assessed dose ranges between approximately 0.2 mSv/year and 0.3 mSv/year. For those studies that assessed the dose arising from both natural and human-made radionuclides in foods, the majority of the dose is as a result of natural radionuclides in the diet.

When the various estimates of the annual ingestion dose have been compiled as a function of age, there is no significant difference between ingestion doses for infants, children and adults. This suggests that the higher dose coefficients for children and infants are offset by lower consumption rates. Figure 3 shows the range of ingestion doses for the various International Commission on Radiological Protection (ICRP) age groups included in the review.<sup>11</sup>

UNSCEAR has estimated a worldwide age weighted ingestion dose of 0.144 mSv/year [16]. In comparison, the geometric mean of the doses in the total diet included in this review is 0.19 mSv/year.

#### 4.2.3. Doses as a function of radionuclide

To understand the contribution to dose from individual radionuclides, the data compiled were further evaluated to determine the relative contribution of different radionuclides to the total dose. An initial evaluation for each radionuclide

<sup>11</sup> The dose results from the French seaside sites have not been included, as these represent a very specific cohort of the French adult population.

was conducted on all dose assessments compiled (i.e. including all dietary study types and age groups). The geometric mean and range of doses for each radionuclide in order of the contribution to dose is shown in Table 11 and Fig. 4.

It can be concluded that approximately 90% of the dose from ingestion is due to uranium and thorium series radionuclides, and 10% of the dose is due to other radionuclides. The four main contributors to the dose are  $^{210}\text{Po}$ ,  $^{210}\text{Pb}$ ,  $^{228}\text{Ra}$  and  $^{226}\text{Ra}$ , in order of priority. These together contribute over 95% of the dose from all uranium and thorium series radionuclides. This is also observed in the UNSCEAR estimates of effective dose arising from uranium and thorium series radionuclides [16].

The range of values shown in Table 11 differs from those shown in Section 4.2.2 and Table 10, as additional individual radionuclide data are included. Section 4.2.2 focuses on publications that include dose from the total diet, whereas these data include surveys that only considered individual or a small number of radionuclides in individual foods. Very large variability in the doses arising from each of the radionuclides is specified. In a small number of studies, the dose from ingestion is above 1 mSv/year for the uranium and thorium series radionuclides  $^{210}\text{Po}$  (maximum = 3.6 mSv/year),  $^{228}\text{Ra}$  (maximum = 2.7 mSv/year) and  $^{226}\text{Ra}$  (maximum = 2.4 mSv/year). The maximum dose associated with  $^{210}\text{Po}$  is from a US study on  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  in the diet in the Marshall Islands. This high dose from  $^{210}\text{Po}$  is as a result of high  $^{210}\text{Po}$  activity concentrations in seafood in conjunction with the very high seafood consumption rates of the residents of the Marshall Islands [82]. The  $^{228}\text{Ra}$  dose of 2.67 mSv is from a Ghanaian study with  $^{228}\text{Ra}$  activity concentrations in food that were two orders of magnitude higher than those found in other studies, which was due to the high consumption rate of cassava and plantains in the diet [83]. The study that has the highest reported dose from  $^{226}\text{Ra}$  is from Kuwait [84]. However, the doses estimated in this study are expressed as 90th, 95th and 99th percentile values for  $^{226}\text{Ra}$  and would therefore be considered an upper bound rather than a geometric mean ingestion dose value.

The dose from ingestion of other radionuclides is two orders of magnitude lower than that from uranium and thorium series radionuclides. Apart from  $^{14}\text{C}$ , which has contributions from both natural (cosmogenic) and human-made sources, the most significant contributors to ingestion dose from human-made radionuclides are radiocaesium ( $^{134}\text{Cs}$  and  $^{137}\text{Cs}$ ) and  $^{90}\text{Sr}$ . The studies that have estimated the largest dose from these human-made radionuclides are typically those surveys that have been conducted shortly after the large scale nuclear accidents at the Chernobyl and Fukushima Daiichi NPPs. One study in the Ovruc region in Ukraine [85] estimated an ingestion dose of 4.95 mSv/year from  $^{137}\text{Cs}$  on the basis of a duplicate diet survey from samples taken in the region in 1990.

TABLE 11. GEOMETRIC MEAN AND RANGE OF DOSES FOR EACH RADIONUCLIDE<sup>a</sup>

Radionuclide (No. of values)	GM (mSv/year)	Range (mSv/year)
U and Th series radionuclides		
Po-210 (92)	0.14	0.001–3.640
Pb-210 (60)	0.05	0.001–0.409
Ra-228 (29)	0.028	0.002–2.670
Ra-226 (61)	0.017	0.001–2.402 <sup>b</sup>
Th-228 (11)	0.006	$0.3 \times 10^{-3}$ –0.3942
Th-232 (52)	$1 \times 10^{-3}$	$4.1 \times 10^{-5}$ –0.3
Ra-224 (1)	$1 \times 10^{-3}$	— <sup>c</sup>
Th-230 (8)	$0.7 \times 10^{-3}$	$0.1 \times 10^{-3}$ –0.023
U-238 (54)	$0.6 \times 10^{-3}$	$0.1 \times 10^{-3}$ – $1 \times 10^{-3}$
U-234 (22)	$0.4 \times 10^{-4}$	$0.1 \times 10^{-3}$ – $1 \times 10^{-3}$
U-235 (8)	$0.2 \times 10^{-4}$	$1.7 \times 10^{-6}$ – $7 \times 10^{-3}$
Other radionuclides		
C-14 (24)	$8.5 \times 10^{-3}$	$4.6 \times 10^{-3}$ – $14 \times 10^{-3}$
Cs-134/137 (276) <sup>d</sup>	$3.5 \times 10^{-3}$	$0.1 \times 10^{-3}$ –4.95
Sr-90 (111)	$2.6 \times 10^{-3}$	$0.1 \times 10^{-3}$ –0.35
Rb-87 (4)	$2.0 \times 10^{-3}$	$1 \times 10^{-3}$ – $6 \times 10^{-3}$
I-129 (4)	$1.9 \times 10^{-3}$	$1.9 \times 10^{-3}$ – $2.1 \times 10^{-3}$
Am-241 (14)	$9.6 \times 10^{-4}$	$0.1 \times 10^{-3}$ – $21 \times 10^{-3}$



TABLE 11. GEOMETRIC MEAN AND RANGE OF DOSES FOR EACH RADIONUCLIDE<sup>a</sup> (cont.)

Radionuclide (No. of values)	GM (mSv/year)	Range (mSv/year)
S-35 (4)	$7.4 \times 10^{-4}$	$0.3 \times 10^{-3}$ – $1.4 \times 10^{-3}$
Co-60 (5)	$6.8 \times 10^{-4}$	$0.5 \times 10^{-3}$ – $1 \times 10^{-3}$
Pu-239/240 (41)	$2.4 \times 10^{-4}$	$0.1 \times 10^{-3}$ – $4.7 \times 10^{-3}$
H-3 (5)	$1.0 \times 10^{-4}$	$1.0 \times 10^{-4}$

- <sup>a</sup> Limit of detection values have been assumed to be absolute values. Dose estimates for adults, children and infants for all study types have been included.
- <sup>b</sup> The highest reported <sup>226</sup>Ra value is a 99th percentile value from Kuwait.
- <sup>c</sup> —: data not available.
- <sup>d</sup> See text box on radiocaesium values.

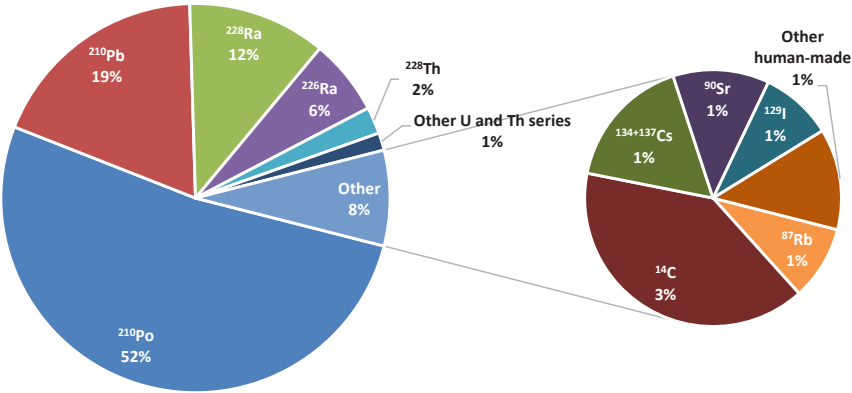


FIG. 4. Contributions to annual ingested dose from radionuclides.

### Dose from radiocaesium ( $^{134}\text{Cs}$ and $^{137}\text{Cs}$ )

Not all scientific publications reviewed as part of this assessment provided information on the ingestion dose from radiocaesium. In those that did, the dose was reported in various ways. Some reported the dose from  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  separately, whereas some reported them as a single ingestion dose, namely as radiocaesium or  $^{134}\text{Cs} + ^{137}\text{Cs}$ . The table below summarizes the number of publications reporting the ingestion dose in the three different ways.

Radionuclide	<i>n</i>
$^{137}\text{Cs}$	182
$^{134}\text{Cs}$	58
$^{134+137}\text{Cs}$	43

The presence of  $^{134}\text{Cs}$  in food in the surveys reviewed is a direct result of environmental releases following the accidents at the Chernobyl and Fukushima Daiichi NPPs. The papers reporting  $^{134}\text{Cs}$  ingestion doses were reviewed to ensure that the reported doses were not those received during the emergency exposure phase of these nuclear emergencies. The only  $^{137}\text{Cs}$  or  $^{134}\text{Cs}$  ingestion doses considered as part of this assessment were those that were reported more than one year from the time of the accident. There were 27 publications that did not meet this criterion.

The ingestion dose from  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  was calculated from the geometric mean of all  $^{134}\text{Cs}$ ,  $^{137}\text{Cs}$  and  $^{134+137}\text{Cs}$  ingestion doses reported that fulfilled the criteria outlined above.

The radiocaesium geometric mean calculated was  $3.5 \times 10^{-3}$  mSv/year ( $n = 256$ ).

Two of the publications reviewed also estimated the dose arising from the short lived radionuclide  $^{131}\text{I}$ . The first study<sup>12</sup>, published in Germany shortly after the accident at the Chernobyl NPP, reported measurable quantities of  $^{131}\text{I}$  in total diet samples in 1986. These measurements and subsequent dose estimates are not included in this review, as they were assessed in the first few months after a large scale nuclear accident and are not relevant to non-emergency exposure situations. The second study, from New Zealand [77], estimated an upper bound of  $^{131}\text{I}$

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<sup>12</sup> DIEHL, J. F., FRINDIK, O., MILLER, H. Radioactivity in total diet before and after the Chernobyl reactor accident. The situation in the Federal Republic of Germany. Zeitschrift für Lebensmittel-Untersuchung und Forschung. **189** (1989) 36-38. Not included in the present review.

dose based on minimum detectable activities of  $^{131}\text{I}$  measurements in food. Since no measurable  $^{131}\text{I}$  activity concentrations were reported in food samples from New Zealand, they were also omitted from the survey. None of the publications considered in this review included information on  $^{131}\text{I}$  activity concentrations in environmental samples or food as a result of discharges from facilities other than nuclear facilities after an accident, for example discharges from hospitals or research institutes, and therefore no conclusions can be drawn regarding the dose arising from  $^{131}\text{I}$  discharges from these facilities.

#### 4.2.4. Comparison with UNSCEAR estimates

Further investigation of the dose to adults from uranium and thorium series radionuclides was conducted, and the results were compared to the age weighted dose estimates from the ingestion of uranium and thorium series radionuclides previously published by UNSCEAR [16].

The generic dose assessment carried out by UNSCEAR identifies  $^{228}\text{Ra}$ ,  $^{226}\text{Ra}$ ,  $^{210}\text{Pb}$  and  $^{210}\text{Po}$  as the key radionuclides contributing to ingestion dose [16]. These four radionuclides have been estimated to contribute to over 95% of the age weighted annual effective dose of approximately  $140\ \mu\text{Sv}$  (Fig. 5) [16]. However, UNSCEAR notes that the concentrations of natural radionuclides in food are highly variable and can range over several orders of magnitude. Consequently, individual doses can also vary widely between countries and population groups due to differences in climate, agricultural practices and diet.

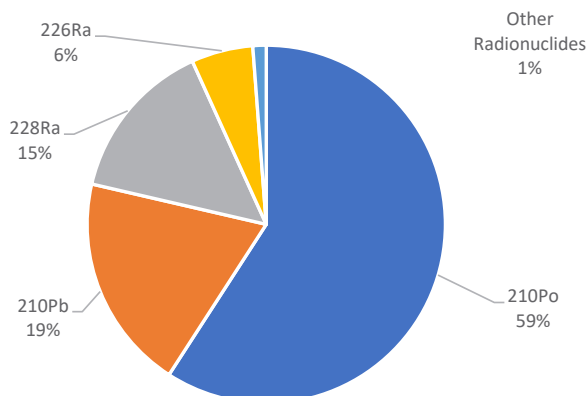


FIG. 5. The UNSCEAR estimated annual age weighted effective dose from the ingestion of uranium and thorium series radionuclides [16].

A comparison of annual ingestion dose from uranium and thorium series radionuclides in food based on this review and UNSCEAR age weighted annual effective dose is shown in Table 12 and Fig. 6.

These data indicate that the overall estimated ingestion dose for adults, children and infants from the uranium and thorium series radionuclides is approximately 50% higher than the age weighted annual dose published by UNSCEAR. However, the key radionuclides contributing the majority of the dose are the same. Polonium-210 contributes more than half of the total dose from uranium and thorium series radionuclides in both evaluations, and the percentage contributions from other radionuclides are broadly in agreement. In all cases the doses estimated for the uranium and thorium series radionuclides in this review are higher than those estimated by UNSCEAR. This could be because some of the publications included in our review focused on regions where the levels of natural radionuclides were known to be elevated, while others included information on consumers with higher than average consumption, such as high rate seafood consumers. In contrast, UNSCEAR focused on overall annual intake rates on the basis of worldwide reference values of uranium and thorium series radionuclides in foods and age weighted, averaged dose to the general population worldwide.

TABLE 12. COMPARISON OF ANNUAL DOSE TO ADULTS, INFANTS AND CHILDREN FROM URANIUM AND THORIUM SERIES RADIONUCLIDES BASED ON THIS REVIEW AND UNSCEAR AGE WEIGHTED ANNUAL DOSE [16]

Radionuclide	Total ingestion dose (mSv)	
	This review (% contribution)	UNSCEAR 2000 (% contribution)
Po-210	0.14 (56%)	0.085 (59%)
Pb-210	0.05 (20%)	0.028 (19%)
Ra-228	0.031 (13%)	0.021 (15%)
Ra-226	0.017 (7%)	0.008 (6%)
Other radionuclides	0.01 (4%)	0.002 (1%)

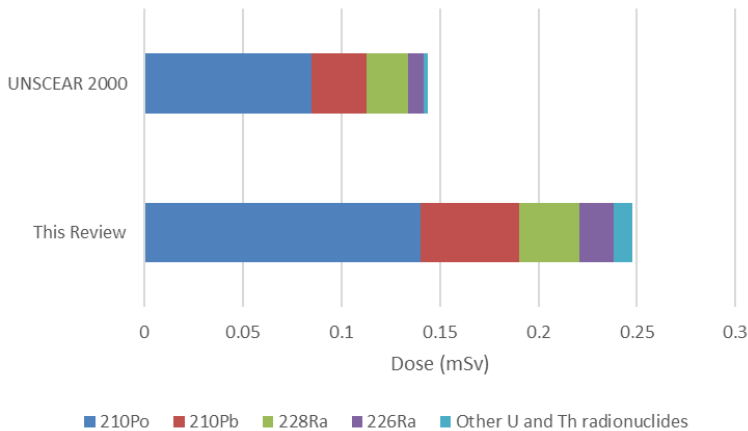


FIG. 6. Comparison of annual dose from uranium and thorium series radionuclides based on this review and the UNSCEAR 2000 Report's annual effective dose to adults [16].

#### 4.2.5. Doses as a function of age

The data presented in Table 12 were segregated further into the doses for the different age groups and reviewed to determine whether any significant differences in dose as a result of age could be observed. ICRP Publication 101a [86] recommends the use of three rather than six age categories for estimating annual dose to the representative individual in prospective assessments. However, since the majority of the studies included in the review preceded ICRP Publication 101a, the data are presented using the previously recommended six age categories of Infant, Child (1–2), Child (2–7), Child (7–12), Child (12–17) and Adult (>17 years).

The data for the different age groups are presented in Fig. 7 for the four natural radionuclides that have been found to typically contribute most to the dose from dietary intakes if they are present in food, namely <sup>210</sup>Po, <sup>210</sup>Pb, <sup>226</sup>Ra and <sup>228</sup>Ra. Figure 7 shows that although only tentative observations can be drawn due to the very limited data for infants and children, the doses for children are broadly similar to those for adults, indicating that higher dose coefficients for children and infants are balanced to a large extent by lower consumption rates, as is also discussed with respect to the total doses shown in Fig. 4.

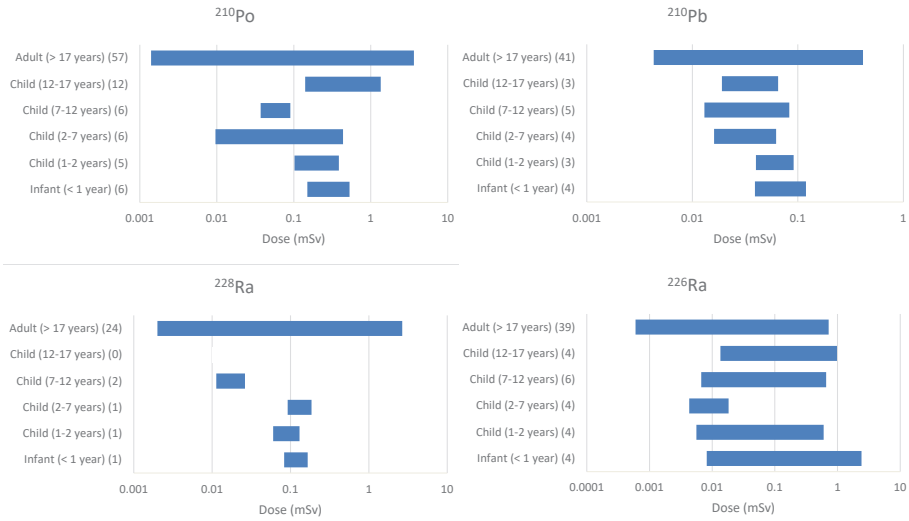


FIG. 7. Annual dose from dietary intakes for  $^{210}\text{Po}$ ,  $^{210}\text{Pb}$ ,  $^{226}\text{Ra}$  and  $^{228}\text{Ra}$  as a function of age. The number of dose estimates for each age group is in parentheses.

#### 4.2.6. Doses as a function of dietary survey type

For six of the studies reviewed, doses have been estimated using different dietary study types. A comparison of the doses estimated for each of these studies is summarized in Table 13. This small number of studies shows that the dose estimates are very similar regardless of the study type used.

#### 4.2.7. Discussion

This literature survey identified 127 publications that reported estimated doses arising from radioactivity in the diet from 46 countries worldwide. The majority of studies reviewed were conducted using the MBS approach. The publications reviewed contained information on the dose for 12 natural radionuclides and 11 human-made radionuclides. The surveys of doses from human-made radionuclides focused on radiocaesium and  $^{90}\text{Sr}$  due to their presence in food as a result of nuclear weapons fallout and past nuclear accidents.

Diet surveys that focused on total dose from ingested radionuclides show that the overall annual dose is typically estimated as being below 1 mSv/year. The mean estimated annual dose from a review of these surveys is 0.19 mSv/year (GM,  $n = 36$ ) for all age groups, and the dose is dominated by natural radionuclides in the diet.

TABLE 13. DOSES ESTIMATED USING DIFFERENT DIETARY SURVEY TYPES WITHIN A SINGLE STUDY

Country	Radionuclide	Dose (mSv/year)			
		Study type <sup>a</sup>			
		MBS	DDS	TDS	CMS
India, Gudalore [87]	Po-210	0.700	0.682		
India, Southern Tamil Nadu [88]	Po-210	0.076	0.052		
Japan [89]	Sr-90	0.0009		0.0007	
	Cs-137	0.0004		0.0003	
Japan [90]	Cs-134/137	0.0035	0.0020		
USA, Washington DC [91]	Sr-90	0.0046		0.0052	
Poland [92]	Ra-226	0.0045	0.005		0.0053
	Pb-210	0.0208	0.035		0.0313
	Po-210	0.054	0.0539		0.0526

<sup>a</sup> MBS: market basket study; DDS: duplicate diet study; TDS: total diet study; CMS: canteen meal study.

A review of the dose from each individual radionuclide indicates that the four main contributors to ingestion dose (not including <sup>40</sup>K) are the natural radionuclides <sup>210</sup>Po, <sup>210</sup>Pb, <sup>228</sup>Ra and <sup>226</sup>Ra. The contribution of human-made radionuclides to dose is approximately 10% of the overall dose in the publications reviewed. However, very large variability in the dose from individual radionuclides is observed. This could be a result of surveys being conducted in high natural background radiation areas and surveys focusing on consumers with higher than average ingestion of radionuclides, such as high seafood consumers. This could also be a factor contributing to the difference in the overall dose to adults from the naturally occurring radionuclides when compared to published data from UNSCEAR [16]. The results of this review indicate that the overall dose (not including <sup>40</sup>K) to adults is approximately 0.19 mSv/year, whereas UNSCEAR estimated a value closer to 0.12 mSv/year. The main difference is a higher dose from <sup>210</sup>Po in this study. Comparing the dose from the total diet as a

function of age indicates that there is not any significant variability between ages. However, this is based on a very limited dataset. The number of studies where data for the different dietary study types can be compared is very small, so it is important to consider the most appropriate survey type for a given situation to determine the dose from the ingestion of radionuclides, as discussed in Section 3.

#### 4.3. NATURAL RADIONUCLIDES IN NATURAL MINERAL WATER

Natural mineral waters originate in underground water-bearing strata and are characterized by their mineral content, which is a result of chemical processes under natural conditions. Mineral waters are collected from natural or drilled sources under conditions that ensure microbiological purity and avoid external pollution of the supply [11].

The chemical composition of natural mineral waters is determined by a number of factors, including residence time of the water, redox conditions, types of adsorption, kinetics of mineral phases and, most importantly, the underlying geology of the water source [93]. Bedrock contains naturally occurring radionuclides from the uranium and thorium decay series, such as  $^{210}\text{Po}$ ,  $^{210}\text{Pb}$ ,  $^{228}\text{Ra}$  and  $^{226}\text{Ra}$ , that can be readily transferred to the mineral waters. These mineral waters are subjected to minimal treatment prior to bottling, and treatment is carried out only on the condition that the mineral content is not modified in its essential constituents [11]. This could result in enhanced levels of naturally occurring radionuclides in mineral water intended for human consumption.

At present, there are no international criteria for radioactivity in natural mineral waters. WHO has published international guidelines for drinking water quality that include criteria to determine the safety of drinking water with respect to its radioactivity content [7]. However, natural mineral waters are sold as foods, and existing food standards, the Codex Alimentarius, do not provide criteria for radionuclides in natural mineral waters [11]. The Codex Standard for Natural Mineral Waters [11] does not provide any criteria for radionuclides, whereas the standard for bottled or packaged drinking waters (other than natural mineral waters) [94] states that they need to comply with the radiological criteria set out in the WHO guidelines for drinking water quality.

In 2019, the average worldwide per capita consumption of bottled waters was 55 L/year, with consumption in some countries being as high as 280 L/year (Mexico), 230 L/year (Thailand) and 200 L/year (Italy) per capita [95]. Bottled natural mineral waters can represent a large proportion of the overall bottled waters consumed, with 82% of all bottled waters consumed in Europe being natural mineral waters [96].



As natural mineral waters can contain enhanced levels of natural radioactivity and their consumption worldwide can be significant, numerous studies have been conducted investigating the levels of natural radionuclides in them, and estimates have been made concerning the ingestion dose arising from their consumption. The existing publications were reviewed to determine:

- (a) The radionuclides with the highest activity concentrations;
- (b) The dose arising from natural radioactivity in natural mineral waters;
- (c) Which radionuclides are the largest contributors to dose;
- (d) How the radionuclide concentrations and doses compare to those for other foods.

#### **4.3.1. Literature survey**

A review of scientific literature from 1970 to 2020 was conducted to identify publications that contain measurements of naturally occurring radionuclides in mineral waters. The publications were reviewed to determine whether they included any measurements of radionuclides in the uranium or thorium decay series in mineral waters for human consumption. Specifically, publications were reviewed that considered mineral waters being used as water supplies and mineral waters commercially bottled for consumption (non-carbonated and carbonated). In some cases, these publications included measurements in mineral waters that were not destined for human consumption, for example mineral waters used in health spas for medical, bathing and recreational purposes [97], and these measurements were excluded from the review. In addition, publications included bottled water products that did not meet the criteria to be defined as a natural mineral water, and these were also excluded. In most cases, countries assessed natural mineral waters originating from within their own country, but some measured imported bottled natural mineral waters (e.g. in Japan), and these were included in the review [98].

The review identified 143 papers, of which 72 papers covering 35 countries worldwide included measurements in mineral waters that fulfilled the criteria outlined above. A dataset of 2540 measurements containing 10 radionuclides of the uranium and thorium series was compiled from the literature reviewed.

#### **4.3.2. Results**

The range of values for each of the 10 radionuclides was determined, along with the geometric mean of the values. A summary of the data and the results of the analysis is given in Table 14. These activity concentrations represent the mineral waters ‘as sampled’ and not ‘as consumed’. In most cases, the samples

TABLE 14. SUMMARY OF RESULTS FROM THE REVIEW OF  
NATURAL MINERAL WATERS

Radionuclide	No. of measurements	Activity concentration (mBq/L)		
		Min.	Max.	Geometric mean
Pb-210	131	0.18	34 000	75
Po-210	123	0.15	110	2.5
Ra-226	1 000	$1.0 \times 10^{-3}$	19 600	26
Ra-228	293	0.10	5 609	29
Th-228	25	$4.8 \times 10^{-3}$	152	0.7
Th-230	24	$6.0 \times 10^{-4}$	2.40	0.01
Th-232	160	$7.0 \times 10^{-4}$	3 390	1.5
U-234	189	0.16	464	7.1
U-235	72	0.02	17	0.7
U-238	523	0.01	2 240	4.9

were bottled mineral waters for consumption, but some were sampled at source and may not reflect the final activity concentrations as consumed when taking into consideration the ingrowth and decay of the uranium and thorium series radionuclides over time.

Overall, the geometric mean of activity concentrations indicates that  $^{210}\text{Pb}$  activities in bottled waters (75 mBq/L) are higher than those from other uranium and thorium series radionuclides. The activity concentrations of  $^{226}\text{Ra}$  and  $^{228}\text{Ra}$  are broadly similar, with geometric mean values of 26 mBq/L and 29 mBq/L, respectively. All other radionuclides have geometric means of less than 10 mBq/L.

The highest activity concentrations measured in individual samples were for  $^{210}\text{Pb}$  (34 Bq/L),  $^{226}\text{Ra}$  (20 Bq/L) and  $^{228}\text{Ra}$  (5.6 Bq/L). The highest  $^{210}\text{Pb}$  activity concentration was measured in a bottled mineral water sample from the Tyrol region of Austria, which is known to have high  $^{222}\text{Rn}$  activity concentrations [99]. The  $^{210}\text{Pb}$  activity can be attributed to the ingrowth from

$^{222}\text{Rn}$  in the mineral water after bottling, which could indicate that the  $^{222}\text{Rn}$  in the bottled water is supported by  $^{226}\text{Ra}$ . The highest  $^{226}\text{Ra}$  activity concentration was detected in a mineral water spring in Saratoga, New York, USA [100], which has been known to contain high  $^{226}\text{Ra}$  activity concentrations for over a century. The highest  $^{228}\text{Ra}$  activity concentration measured was from a mineral water sample collected in the Caxambú region in Brazil [101], which is known to have high levels of naturally occurring radiation [102]. Other high values, such as those in  $^{238}\text{U}$  and  $^{232}\text{Th}$ , were also noted. A  $^{238}\text{U}$  activity concentration of 2.24 Bq/L (180  $\mu\text{g/L}$ ) was measured in a mineral water well in Bulgaria [103]. A  $^{232}\text{Th}$  activity concentration of 3.39 Bq/L was detected in a bottled mineral water from Malaysia [104]. Although these values are relatively large, they do not have a significant impact on ingested dose (Section 4.3.3).

There is also large variability of all uranium and thorium series radionuclides in the data, with activity concentrations varying between three and eight orders of magnitude for the mineral waters investigated. This large variation can be attributed to the factors outlined previously, namely the underlying geology of the water source and other considerations, such as the residence time of the water, redox conditions, types of adsorption and desorption in the water system and kinetics of the mineral phases [93].

#### 4.3.3. Dose from natural mineral waters

The ingestion dose arising from each of the radionuclides in the uranium and thorium series in the dataset was estimated using the following equation:

$$A_i \times M(A) \times e_{\text{ing}} = \text{ingestion dose (mSv/year)} \quad (2)$$

where

$A_i$  is the activity concentration of the radionuclide of interest (i) in the water (Bq/L);

$M(A)$  is the volume of mineral water consumed per year (L/year);

and  $e_{\text{ing}}$  is the committed effective dose per unit intake of the radionuclide of interest (mSv/Bq).

The activity concentrations of the radionuclides used to estimate ingestion doses were the calculated geometric mean values of the 10 radionuclides (Table 14). The volume of mineral water consumed was conservatively assumed to be 55 L per year, which was based on the worldwide average consumption of bottled waters in 2019 [95], and the committed effective dose per unit intake

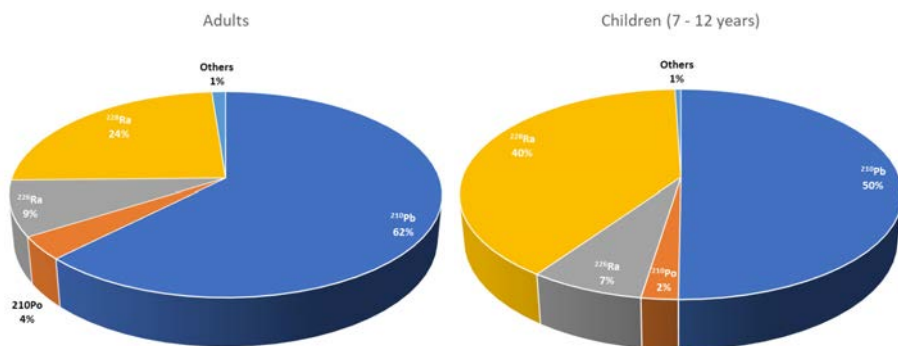


FIG. 8. Contribution of natural radionuclides to ingestion dose from natural mineral waters based on the calculated geometric mean values from the data compiled.

of the radionuclides was that of adults and children (age 7–12)<sup>13</sup> from ICRP Publication 60 [105]. The estimated ingestion dose is  $4.5 \times 10^{-3}$  mSv/year for adults and  $1.6 \times 10^{-2}$  mSv/year for children. The percentage contribution to estimated ingestion dose from each of the radionuclides is summarized in Fig. 8.

For adults, the calculated ingestion dose is dominated by  $^{210}\text{Pb}$ , which accounts for over 60% of the ingestion dose ( $2.8 \times 10^{-3}$  mSv/year). The next most significant contributor is  $^{228}\text{Ra}$  (24%), followed by  $^{226}\text{Ra}$  (9%); although their activities are broadly similar, the dose from  $^{228}\text{Ra}$  is higher as a result of its higher committed effective dose per unit intake. Polonium-210 accounts for 4% of the dose, with the other six uranium and thorium series radionuclides ( $^{228}\text{Th}$ ,  $^{230}\text{Th}$ ,  $^{232}\text{Th}$ ,  $^{234}\text{U}$  and  $^{238}\text{U}$ ) accounting for the remaining 1% of dose.

The overall estimated ingestion dose to children (aged 7–12) is higher for each of the radionuclides when compared to adults. The ingestion dose is also dominated by  $^{210}\text{Pb}$ , which accounts for 50% ( $7.8 \times 10^{-3}$  mSv/year). The ingestion dose from  $^{228}\text{Ra}$  is much higher, at  $6.2 \times 10^{-3}$  mSv/year, compared to that of adults ( $1.1 \times 10^{-3}$  mSv/year), and the contribution to dose is also much larger at 40%. This is due to the higher  $^{228}\text{Ra}$  ingestion dose coefficient for children compared to adults. However, it is assumed here that the consumption rates for adults and children are identical. No consideration is given to a lower consumption rate for children, which may counterbalance the higher ingestion dose coefficient.

<sup>13</sup> The dose to children (age 7–12) was estimated, as this was deemed to be the most sensitive age group for children with respect to dose. For children younger than this age group, it was assumed that the consumption rates of natural mineral waters would be significantly lower than those for older children.

The ingestion dose to adults and children for the highest activity concentrations for each of the radionuclides was also estimated, assuming the average annual worldwide consumption of bottled water (Table 15). These results, and those in Fig. 8, indicate that, in the majority of cases, the estimated doses for each of the radionuclides are below 1 mSv/year for adults and children.

For adults, the estimated ingestion dose using the highest <sup>210</sup>Pb activity concentration from Austria could be above ~1 mSv/year if the consumption rate of this bottled water was greater than 55 L per year. Similarly, the ingestion dose from <sup>228</sup>Ra in Caxambú, Brazil and <sup>226</sup>Ra in Saratoga, New York, USA, could result in an ingestion dose of ~1 mSv/year if consumed in large quantities. For a child (age 7–12), given a <sup>210</sup>Pb activity concentration of 34 000 mBq/L, the estimated ingestion dose is 3.5 mSv/year. Similarly, for <sup>228</sup>Ra, the ingestion dose to an adult is 0.2 mSv/year, whereas for a child it increases to 0.9 mSv/year.

TABLE 15. ESTIMATED INGESTION DOSE (mSv/YEAR) BASED ON MAXIMUM ACTIVITY CONCENTRATIONS IN THE DATA COMPILED<sup>a</sup>

Estimated dose from maximum activity (mSv/Bq per year)	Maximum activity (mBq/L)	Adult ingestion dose (mSv/year)	Children (age 7–12) ingestion dose (mSv/year)
Pb-210	34 000	1.28	3.52
Po-210	110	$7.19 \times 10^{-3}$	$1.56 \times 10^{-2}$
Ra-226	19 600	0.31	0.88
Ra-228	5 609	0.21	1.19
Th-228	152	$5.96 \times 10^{-4}$	$1.24 \times 10^{-3}$
Th-230	2.4	$2.75 \times 10^{-5}$	$3.14 \times 10^{-5}$
Th-232	3 390	$4.25 \times 10^{-2}$	$5.36 \times 10^{-2}$
U-234	464	$1.24 \times 10^{-3}$	$1.87 \times 10^{-3}$
U-235	170	$4.35 \times 10^{-5}$	$6.58 \times 10^{-5}$
U-238	2 240	$5.49 \times 10^{-3}$	$8.30 \times 10^{-3}$

<sup>a</sup> Assuming a worldwide average annual consumption of 55 L/year.

#### 4.3.4. Discussion

For natural mineral waters, the four most important radionuclides contributing to dose are  $^{210}\text{Pb}$ ,  $^{228}\text{Ra}$ ,  $^{226}\text{Ra}$  and  $^{210}\text{Po}$ . However, this differs from other foods in the diet, as the activity concentration and dose contribution from  $^{210}\text{Po}$  are lower than those from the  $^{210}\text{Pb}$  and radium isotopes. Typically,  $^{210}\text{Po}$  dominates the ingestion dose in food. However, the behaviour of  $^{210}\text{Po}$  in water is different from that in organic matrices;  $^{210}\text{Po}$  is highly insoluble in water and readily adsorbed onto aquifer rocks, which can account for its low activity concentration in natural mineral waters [106]. Therefore, any future guidelines developed for the management of radioactivity in food need to consider any potential differences in the radiochemical composition of natural mineral waters and other foods.

The ingestion dose from natural mineral waters is, in most cases, not a significant contributor to overall ingestion dose from natural radionuclides. The estimated ingestion dose, assuming a consumption rate of 55 L/year and using the geometric mean values of activity concentrations from the data compiled as part of this study, is 5  $\mu\text{Sv}/\text{year}$ , which is 2% of the UNSCEAR estimated ingestion dose of approximately 300  $\mu\text{Sv}/\text{year}$  from food and drinking water [107]. This dose of 5  $\mu\text{Sv}/\text{year}$  is also approximately half of the UNSCEAR estimated value of 10  $\mu\text{Sv}/\text{year}$  for the ingestion of natural radionuclides in drinking water. If consumption rates of natural mineral water are above the worldwide average of 55 L/year (as is the case in some countries) or the mineral water supply is known to have elevated levels of  $^{210}\text{Pb}$ ,  $^{228}\text{Ra}$  and, to a lesser extent  $^{226}\text{Ra}$ , then the ingestion dose may become a significant contributor to total dose.

This literature review also indicates that the radionuclide measured most often is  $^{226}\text{Ra}$ . However, this review indicates that  $^{210}\text{Pb}$  and  $^{228}\text{Ra}$ , which are primarily beta emitters, are important radionuclides contributing to the dose from natural mineral waters. Radiochemical separation techniques are typically needed when measuring these radionuclides in water samples (see Annex I), when the activity concentrations are typically in the mBq/L range, but this is not always straightforward. However, it may be possible to estimate upper limits for both  $^{210}\text{Pb}$  and  $^{228}\text{Ra}$  activity concentrations using gamma spectrometry once appropriate systems and techniques are utilized. These would include the use of low energy, low background gamma spectrometry systems that have been calibrated appropriately, with adequate corrections being conducted to correct for self-absorption of the low energy  $^{210}\text{Pb}$  gamma emission of 46 keV. Furthermore, the samples need to be sealed and stored for a sufficient time to allow for the  $^{228}\text{Ra}$  progenies,  $^{228}\text{Ac}$  and  $^{212}\text{Pb}$ , to reach secular equilibrium so that the  $^{228}\text{Ra}$  activity concentration can be determined.

For example, a  $^{210}\text{Pb}$  activity concentration of 25 Bq/L in mineral water would lead to a dose of  $\sim 1$  mSv/year if a consumption rate of 55 L/year is assumed. This level of  $^{210}\text{Pb}$  in mineral water samples can be measured by gamma spectrometry once adequate self-absorption corrections have been conducted [108]. Similarly, a  $^{228}\text{Ra}$  activity concentration of 25 Bq/L in mineral water would lead to a dose of  $\sim 1$  mSv/year if a consumption rate of 55 L/year is assumed, and these measurements can also be carried out using gamma spectrometry.

#### 4.4. AQUACULTURE

Aquaculture is the production, under controlled conditions, of fish, shellfish, algae and other aquatic plants. The aquaculture industry operates in both the freshwater and marine environments and has two main components: fishery products (which includes fish, crustaceans, molluscs and other aquatic animals) and aquatic plants (of which the principal component is seaweed). The production of fishery products by aquaculture is often referred to as fish farming.

The scale of aquaculture operations is highly variable, from large industrial and commercial entities to much smaller scale facilities providing an important food supply to isolated rural villages. Economically, the aquaculture industry is dominated by the production of fishery products, which is currently worth over US \$250 billion annually. By comparison, the annual production of aquatic plants has an economic value of approximately US \$13.3 billion [109].

Global aquaculture production for both fishery products and aquatic plants is dominated by China, whose output exceeds that of the rest of the world combined. Other Asian countries are also large producers of both fishery products and aquatic plants. Some form of aquaculture is practised in almost every country in the world.

##### 4.4.1. Fishery products

The aquaculture industry for fishery products has grown steadily since the 1980s and currently represents just under 50% of the worldwide production from all sources. Over the next 10 years it is anticipated that the volume sourced from global capture fisheries will remain relatively stable at between 86 million tonnes and 93 million tonnes, while the contribution from aquaculture will increase to approximately 120 million tonnes [109].

According to the FAO [109], 466 different species of finfish, 109 molluscs and 64 crustaceans are currently reared, or have been reared in the past, through aquaculture. However, many of these are produced in small amounts,

and the world market is dominated by a small number of key species, as shown in Table 16.

As mentioned above, worldwide aquaculture production is dominated by China and other Asian countries. This is shown in Fig. 9. China dominates the market for freshwater fish (54%), crustaceans (30%) and molluscs (83%). For marine fish, China and Norway each represents approximately 20% of global production.

Table 17 provides the worldwide production of various species and clearly shows that freshwater aquaculture is dominated by fish, whereas molluscs dominate production in marine and coastal waters [109]. The production of certain species is practised to a minimal extent, or not at all, in certain regions of the world.

TABLE 16. MAIN FISHERY PRODUCTS PRODUCED BY AQUACULTURE [109]

Category	No. of key species	Contribution of key species	Important species
Fish	20	84%	Carp, pangasius (catfish), salmon and tilapia
Crustaceans	6	92%	Crab, crawfish, prawn and shrimp
Molluscs	9	90%	Clam, mussel, oyster and scallop

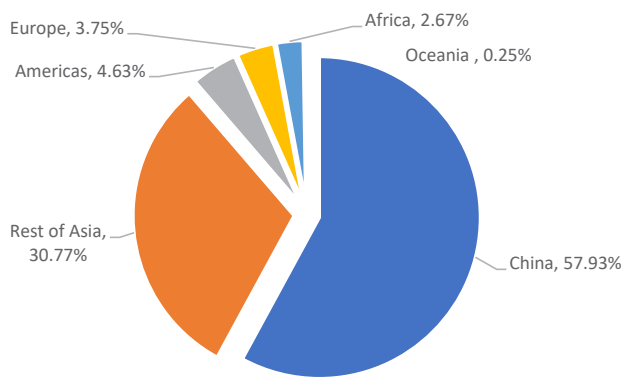


FIG. 9. Global contribution to aquaculture fish production by region.



TABLE 17. GLOBAL PRODUCTION OF FISHERY PRODUCTS IN INLAND AND MARINE WATERS (10<sup>3</sup> TONNES, LIVE WEIGHT) [109]

	Africa	Americas	Asia	Europe	Oceania
FRESHWATER					
Fish	1 893	1 139	43 406	508	5
Crustaceans	— <sup>a</sup>	73	3 579	—	—
Molluscs	—	—	207	—	—
Other	—	1	528	—	—
MARINE (including coastal)					
Fish	291	1 059	3 995	1 892	92
Crustaceans	6	888	4 834	—	6
Molluscs	6	640	15 876	680	102
Other	—	—	387	—	5
TOTAL	2 196	3 800	72 812	3 080	205

<sup>a</sup> Where no value is reported, either the annual production is less than 10<sup>3</sup> tonnes or production data are unavailable.

In terms of the accumulation of radionuclides by fishery products, one important consideration is whether or not the species is artificially fed or is allowed to source nutrients directly from the environment in which it is growing. The decision to feed or not to feed is primarily an economic one: fishmeal is often the major cost for any aquaculture enterprise, but this is outweighed by greatly increased yield of product per unit volume of water.

One would anticipate that the concentration of radionuclides in fishery products that are not fed artificially would reflect the concentrations in the environment and be similar to the concentrations observed in the same species produced in the wild; on the other hand, one would expect the concentration in fed fishery products produced by aquaculture to reflect the concentrations

in the feed rather than in the environment. This is supported by the work of Yamamoto et al. [110].

The diet of fish needs to contain carbohydrates, fats, protein, vitamins and minerals in the right proportions, depending on the individual species, the stage of the growth cycle and the environmental conditions. In the case of farmed carnivorous fish, fishmeal and fish oils represent approximately 50% of the feed content, with the remainder consisting of cereals (corn, soya, wheat), proteins such as amino acids and fatty acids, vitamins and other essential products. For herbivorous and omnivorous fish, the diet normally does not contain fishmeal or fish oils, although recently these have been included in feeds as minor components [111]. Approximately 45% of worldwide aquaculture activities are intensively managed, meaning that there is complete control of water quality, and all the nutritional requirements of the raised animals are met by providing feed. Molluscs are normally not artificially fed, while for both fish and crustaceans, feeding depends on the species, the environment in which they are reared and the management system that is in place [109].

Fishmeal is produced in many countries, and the fish used for its production will depend on the species available locally at any one time — several different species may be used by a given producer and these will also vary over time, depending on availability. Fishmeal is normally made from small marine fish that are too small for human consumption, as well as the trimmings of larger fish from commercial fisheries. As a general rule, 5 kg of fish produce 1 kg of fish meal, and it is assumed that 1 kg of fishmeal produces 1 kg of edible fish [112].

As discussed in Section 5, the radionuclide of most importance as a source of radiation dose from the consumption of fish and shellfish is  $^{210}\text{Po}$ , with the highest concentrations often observed in molluscs, followed by crustaceans and small fish (such as anchovies and whitebait). Larger fish generally have lower  $^{210}\text{Po}$  concentrations. Additionally, wild marine fish tend to have higher concentrations of  $^{210}\text{Po}$  than wild fish sourced from freshwater environments. It is also important to note that there is very wide variability in the radionuclide concentrations observed between and within individual species, with no clear geographical dependence.

There are very limited published scientific data on the activity concentrations of natural radionuclides in fishery products produced by aquaculture. A study in Norway [113] compared the concentrations of  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  in 100 samples of farmed salmon with those in other wild marine species. The concentrations of  $^{210}\text{Po}$  were found to be 10 to 100 times lower in the farmed salmon, whereas the  $^{210}\text{Pb}$  concentrations are broadly similar (Table 18). Radium-228 and  $^{226}\text{Ra}$  were also measured in farmed salmon but were below the detection limit in all 100 samples analysed.

TABLE 18. NATURAL RADIONUCLIDES IN MARINE FISH IN NORWAY [113]

Species	Radionuclide activity concentrations (Bq/kg, fresh weight)	
	Po-210	Pb-210
Salmon — farmed	0.003–0.023	0.03–0.07
Cod — wild	0.09–2.8	0.02–0.07
Haddock — wild	1.1–1.8	—
Saithe — wild	0.7–1.0	—
Redfish — wild	0.16	—
Herring — wild	0.6–8.5	0.06
Mackerel — wild	1.3–5.4	0.06

This is consistent with the conclusions of an earlier study conducted in the UK [114], which measured the concentration of a number of natural and human-made radionuclides in farmed rainbow trout and Atlantic salmon. While no comparable data were available for wild trout and salmon, the author referred to the concentrations of these radionuclides in other marine fish, noting that:

“The  $^{210}\text{Pb}$  values are comparable with those obtained for farmed fish in the present study, whereas the  $^{210}\text{Po}$  values are generally about an order of magnitude higher. However, both  $^{210}\text{Pb}$  and  $^{210}\text{Po}$  in farmed fish were at concentrations consistent with natural levels.”

Smith [114] also calculated transfer factors from fishmeal to the animals and provided measured ratios between the activity concentration of five radionuclides in the flesh of farmed fish and the fishmeal with which they were fed. These ratios are given in Table 19, clearly indicating that the concentration of all measured radionuclides in the two species of farmed fish studied is lower than the corresponding activity concentration in the feed. This is particularly the case for  $^{210}\text{Po}$  in farmed Atlantic salmon, for which, based on the median value of the ratio, the activity concentration is 150 times lower in the fish than in the feed.

TABLE 19. RATIO<sup>a</sup> OF ACTIVITY CONCENTRATIONS IN FARMED FISH AND FISHMEAL [114]

Radionuclide	Atlantic salmon		Rainbow trout	
	Range	Median	Range	Median
C-14	0.13–0.91	0.34	<0.52–0.56	0.47
Tc-99	— <sup>a</sup>	0.19	—	0.19
Cs-137	0.49–>1.3	0.82	0.43–>0.93	0.56
Pb-210	<0.0093–0.17	0.12	<0.007–0.12	0.10
Po-210	0.0031–0.024	0.0068	0.0061–0.033	0.013

<sup>a</sup> Activity concentration in the flesh of fish divided by that in the fishmeal.

The transfer of radionuclides to farmed fish was of particular interest in Japan after the Fukushima Daiichi NPP accident in 2011, where <sup>137</sup>Cs was the radionuclide of most radiological significance. In an experiment with controlled <sup>137</sup>Cs concentrations in water and feed, Ref. [110] observed that:

“radiocaesium contamination in salmonids is mainly via the food chain and that direct intake from water via the skin, gut, and (or) gills has no major direct impact on muscle tissue concentrations.”

Japan currently applies a national limit of 100 Bq/kg to all nationally produced food; a corresponding limit of 40 Bq/kg has been established for <sup>137</sup>Cs in fish feeds [115].

4.4.2. Aquatic plants

Aquatic plants<sup>14</sup> can be subdivided into two main components: macroalgae and microalgae. Macroalgae (more commonly referred to as seaweeds) are

<sup>14</sup> Plants such as rice and watercress are also grown in water but are not algae. As such, they fall outside the definition of aquatic plants used by the FAO.

plant-like organisms that generally live attached to rock or other hard substrata in coastal areas. They are photosynthetic and can manufacture their own food. They have an ‘anchor’ to attach themselves to surfaces such as rocks, with a stem to hold the blade or thallus. Seaweeds can grow in rivers and freshwater lakes as well as in the marine environment. They are a source of biologically active compounds, including proteins and polysaccharides with many uses in nutrition and biomedicine.

Microalgae are single cell microscopic organisms capable of converting solar energy to chemical energy through photosynthesis. They exist individually or in chains or groups in both freshwater and marine environments. Microalgae, along with bacteria, form the base of the food web and provide energy for all the trophic levels above them. Unlike higher plants, such as seaweeds, microalgae do not have roots, stems or leaves. The main uses of microalgae are as a component of biofuels and animal feeds. Because they are rich in micronutrients, microalgae are also used as dietary supplements for humans.

Aquatic plants grow in both freshwater and marine environments; they are collected in the wild and are also farmed. The farming of aquatic plants is dominated by seaweeds, with a much smaller production of microalgae. The industry has increased more than twofold since 1995 and represents a market that is currently worth approximately US \$13 billion [109]. This is still only ~5% of the value of the market for farmed fishery products. The bulk of the farmed seaweed market is for human consumption.

Seaweeds are classified according to their pigmentation, as follows:

- (a) Brown — with fucoxanthin as the dominant pigment;
- (b) Green — containing chlorophyll;
- (c) Red — with phycoerythrin pigment.

However, red seaweeds do not always appear red in colour. Sometimes, these are purple, yellowish brown, light to dark green or pink. Brown seaweeds are usually large, and some species can reach lengths of several metres. Red and green seaweeds are usually smaller, generally ranging from a few centimetres to approximately a metre in length.

Over the past 50 years, the seaweed and microalgae industries have undergone a dramatic global expansion. In the past, harvesting of wild seaweeds was the only source, but over time the demand for seaweed has gradually outstripped the supply from natural sources, and methods for farming seaweed have been developed. Today, seaweed for human consumption comes mainly from farming rather than natural sources [116].

China is the leading world producer of farmed seaweeds (mainly *Porphyra*, *Saccharina* and *Undaria*), contributing 47.8% of the total worldwide production,

followed by Indonesia (38.7%), the Philippines (5.7%) and the Republic of Korea (4.5%).

Historically, major global seaweed production came from the brown seaweeds such as *Undaria* and *Saccharina*. From 2010 onwards, there was a marked increase in red seaweed production, mainly due to the increased production of *Kappaphycus* in Indonesia brought about by the rapid expansion of cultivation areas. The major producing countries of eucheumatoids (*Kappaphycus* and *Eucheuma*) in the South-East Asian region are Indonesia, Malaysia and the Philippines. Disease outbreaks [117–119], the poor quality of cultivars and the adverse effects of climate change [120] have affected output in recent years.

Seaweeds have long been known as useful indicators of radionuclides in the marine environment [121] and are used routinely in national environmental monitoring programmes to monitor the impact of authorized and unplanned discharges from nuclear and other facilities [122, 123]. Following the Fukushima Daiichi NPP accident in 2011, different seaweeds collected locally showed concentrations of  $^{137}\text{Cs}$  that were some 8 to 50 times higher than those in seawater [124]. Seaweeds are also known to accumulate heavy metals, including those with radionuclide analogues, such as lead and cobalt [125].

Radionuclide concentrations in seaweeds are recognized as being from 10 to 30 000 times higher than the concentrations in seawater. These concentration ratios — (nuclide concentration in algae) : (nuclide concentration in seawater) — were derived from the scientific literature and are documented in international reviews [69, 126]. The data are updated continuously and are directly accessible through the IAEA's Marine Information System (MARIS) database [127]. The most abundant data are for brown seaweeds and for the artificial radionuclides  $^{137}\text{Cs}$ ,  $^{239,240}\text{Pu}$ ,  $^{99}\text{Tc}$ ,  $^{60}\text{Co}$ ,  $^{90}\text{Sr}$  and  $^{110\text{m}}\text{Ag}$ , as well as the natural radionuclides  $^{40}\text{K}$ ,  $^{226}\text{Ra}$ ,  $^7\text{Be}$  and  $^{228}\text{Ra}$ . This is because *Fucus vesiculosus* in particular has been utilized as a bioindicator for understanding the levels of radionuclides in the marine environment. In recent years, radionuclide data for some edible species of red seaweed have been added to the database.

The exposure pathways by which exposure of humans from seaweeds and microalgae is possible are as follows:

- (a) Direct consumption;
- (b) Consumption of dietary and other food supplements;
- (c) Consumption of food grown on soils where seaweeds are used as a fertilizer;
- (d) Use as animal feed (both terrestrial and marine) for animals subsequently consumed by humans;
- (e) Use in such as cosmetics, body moisturizers, shampoo, etc.;
- (f) Thalassotherapy (seaweed baths).

Pathways (e) and (f) both represent external exposure to the skin, and the associated doses would be expected to be extremely low. For pathways (c) and (d), unless the radionuclide present in the food is specific to the algae and not the soil, it may not be possible to differentiate between the percentage of a given radionuclide coming from the environment (soil, water or marine sediment) and from the algae. The limited information available suggests that these pathways are also minor. The principal exposure pathways are (a) and (b), with the associated radiation doses being dependent on the extent to which seaweed is consumed directly and the extent to which it is a component of other food products that are also consumed.

Marine seaweeds are consumed particularly in the Asian East Pacific coastal countries [109]. The reported per capita annual consumption rates in 2013 (the latest year for which data are available) were 22 kg/year, 9.2 kg/year and 0.9 kg/year, respectively, in the Republic of Korea, China and Japan. For both China and the Republic of Korea, the consumption of marine seaweeds has increased significantly over the past 50 years, while in Japan the consumption rate has remained steady [128, 129]. The main species consumed are *Saccharina* spp. (Konbu), *Undaria* spp. (Wakame) and *Porphyra* spp. (Nori); the latter two species are produced by aquaculture. Chinese, Japanese and Korean communities living outside their homeland might be expected to have similar consumption patterns.

In Japan, Ota et al. [130] undertook a detailed study of the natural radionuclides  $^{238}\text{U}$ ,  $^{232}\text{Th}$ ,  $^{226}\text{Ra}$ ,  $^{210}\text{Pb}$  and  $^{210}\text{Po}$ , as well as the artificial radionuclides  $^{137}\text{Cs}$ ,  $^{90}\text{Sr}$  and  $^{239,240}\text{Pu}$ , in the diet, including seaweed, in the period from 1989 to 2005. Using a consumption rate of 5.33 kg/year fresh weight for seaweeds, the associated annual individual radiation dose was estimated as 0.014 mSv. The  $^{238}\text{U}$ ,  $^{226}\text{Ra}$ ,  $^{90}\text{Sr}$  and  $^{239,240}\text{Pu}$  concentrations in *Saccharina Japonica* and *Undaria pinnatifida* were among the highest observed in all foods. The total annual effective dose for the Japanese population from all radionuclides in the total diet was calculated as 0.8 mSv and so the contribution to the dose from the consumption of seaweed was ~2% (Table 20).

Population groups present in countries not normally associated with the consumption of seaweeds may traditionally consume seaweeds at particularly high rates. For example, the critical group of laver bread consumers in Wales received individual doses of around 1 mSv annually between 1956 and 1971 [131]. The doses were primarily attributable to human-made radionuclides, of which the most important was  $^{106}\text{Ru}$ , discharged from the nearby Sellafield reprocessing plant; the contribution from natural radionuclides was not evaluated. In the Kuril Islands, annual consumption rates of up to 50 kg of *Laminaria* and *Fucus* seaweeds have been reported [132]. *Porphyra* (known locally as Karengo) is also an important constituent of the traditional Maori diet in New Zealand [133].

TABLE 20. THE ANNUAL EFFECTIVE DOSE IN JAPAN FROM  
SEAWEED INGESTION [130]

Nuclide	Activity concentration (Bq/kg, fresh weight)	Consumption (kg)	Effective dose (mSv/year)
U-238	2.7	5.33	$6.3 \times 10^{-4}$
Th-232	0.076	5.33	$9.3 \times 10^{-5}$
Ra-226	2.8	5.33	$4.2 \times 10^{-3}$
Pb-210	1.3	5.33	$4.8 \times 10^{-3}$
Po-210	3.3	5.33	$4.2 \times 10^{-3}$
Cs-137	0.50	5.33	$3.5 \times 10^{-5}$
Sr-90	1.1	5.33	$1.6 \times 10^{-4}$
Pu-239/240	0.0076	5.33	$1.0 \times 10^{-5}$
TOTAL			0.014

**4.4.3. Discussion**

It is clear that aquaculture is an increasingly important industry and, in the future, a larger percentage of our fishery products will be produced in this way. From the perspective of radioactivity and radiation doses to consumers, there is limited evidence to suggest that the concentrations of  $^{210}\text{Po}$ , in particular, may be at least 10 times lower in farmed fish that are artificially fed compared with those captured in the wild. This is because the transfer of  $^{210}\text{Po}$  from fishmeal to the flesh of the animal is low.

Molluscs produced by aquaculture are normally not fed artificially, and therefore one would expect that the activity concentrations in the flesh, and the associated radiation doses to consumers, would be similar to those in the wild. This is important, as molluscs tend to contain the highest concentrations of radionuclides, in particular  $^{210}\text{Po}$ , of all fishery products.

This has implications for national monitoring programmes. It may not always be possible to be certain whether a particular sample is produced by aquaculture or has been captured in the wild, particularly if it is purchased in a supermarket. Even if it is possible to determine that a particular sample is produced by aquaculture,



whether or not it has been artificially fed is unlikely to be known without direct follow-up with the producer. In many instances, this will not be possible. National authorities are therefore likely to observe a large variation in the concentration of radionuclides in environmental samples, and this may need to be taken into account in dose assessments. For TDSs, this is less important, as one is assessing the total dose from the diet rather than from its individual components.

In the case of seaweeds, the limited information available suggests that this is a minor contributor to radiation doses from the diet. The same is true of microalgae. However, certain subgroups of the population may be high rate consumers of seaweed, and, depending on the various radionuclides present, the associated radiation doses may be relatively high.

The information currently available is very limited, and that makes it difficult to state with any certainty the impact of aquaculture on the radiation doses received by consumers. There is a need for additional data on the concentrations of various natural radionuclides, in particular  $^{210}\text{Po}$ , in farmed fishery products with comparative data for the same species caught in the wild.

#### 4.5. WILD FOODS

Certain food products originating from seminatural ecosystems have a high uptake of specific radionuclides compared with agricultural products. Depending on the environmental conditions, these radionuclides may be natural or human-made. It is important to consider the consumption of such wild foods where these foods may be the dominant source of exposure for certain subgroups of the population.

The concentrations of natural radionuclides in both wild and farmed fish and shellfish have already been covered in Sections 2.2.1 and 4.4. The present section is limited to the following:

- (a) Natural radionuclides in terrestrial wild foods and marine mammals;
- (b) Human-made radionuclides in terrestrial and freshwater wild foods, since the available data show that such products may contain elevated concentrations of these radionuclides.

The activity concentration of a given radionuclide in a given wild food product is dependent on several factors, including:

- The radionuclide concentrations present in the environment;
- The physical and chemical properties of the environmental media (e.g. soil, water) and the chemical form of the radionuclide;

- The specific uptake capability of a particular organism, including intake from their diets, in the case of animal products.

#### **4.5.1. Natural radionuclides in terrestrial wild foods**

Most wild foods do not display substantially elevated levels of natural radionuclides compared with cultivated foods, but highly elevated levels have been observed in some wild species. Activity concentrations are not expected to vary significantly from year to year, except where human activities change the natural state of the environment.

An overview of activity concentrations observed in areas with assumed normal levels of natural radioactivity and areas with known or suspected high levels is presented in Table 21. These data were compiled mainly from a dataset of natural radioactivity in food products from the scientific literature and cover the years 1998 to 2017 (see the Appendix).

Considering the high dose coefficient for  $^{210}\text{Po}$  [105], it follows from Table 21 that  $^{210}\text{Po}$  is generally the main contributor to ingestion doses from wild foods in areas assumed to contain natural radiation within the normal range, followed by  $^{210}\text{Pb}$ . Geometric mean values indicate that other naturally occurring radionuclides are of less importance overall, although in these cases there are fewer data available. Activity concentrations of  $^{210}\text{Po}$  that are considerably higher than the UNSCEAR reference levels (see Section 4.1.4) are found, particularly in reindeer and caribou meat, the meat of marine mammals and several species of mushrooms.

In areas with enhanced concentrations of natural radionuclides, present naturally or as a result of industrial activities, other wild foods and radionuclides may also be significant contributors to the dose and can be important to consider, depending on the situation.

The topic of  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  transfer to different organisms is discussed in more detail in Ref. [106].

##### *4.5.1.1. Edible plants and mushrooms*

The concentrations of  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  in wild berries and other wild edible plants, such as nettles and crab apples, are somewhat enhanced compared to UNSCEAR reference values for fruits; however, concentrations are still low in areas with assumed normal levels of radioactivity. In areas with enhanced levels of natural radioactivity, elevated  $^{226}\text{Ra}$  concentrations have been observed in edible plants (Table 21).

Significant differences are apparent between the different species of mushrooms. Mushroom fruiting bodies have a short lifespan, and the high

TABLE 21. ACTIVITY CONCENTRATIONS OF NATURAL RADIONUCLIDES OBSERVED IN WILD FOODS, Bq/kg, FRESH WEIGHT

		Areas with assumed normal concentrations				Areas with known or suspected high concentrations			
		AM <sup>a</sup>	GM <sup>b</sup>	Min.	Max.	n	Min.	Max.	n
<b>Edible plants and mushrooms</b>									
Berries	Po-210	0.24	0.18	0.021	1.1	53	0.016	0.3	2
	Pb-210	0.29	0.23	0.027	1.2	53	0.2		1
	Ra-226	0.22	0.055	0.013	2.3	15	0.53	151	9
	Ra-228						0.61		1
	U, Th			0.000009	0.084	31	0.0014	0.05	10
Other edible plants <sup>c</sup>	Po-210	0.27	0.14	0.03	0.91	9	0.12	1.8	6
	Pb-210	0.38	0.23	0.04	1.0	9	0.079	4.5	6
	Ra-226	0.17	0.087	0.009	0.64	7	0.18	140	1
	Ra-228						0.41	7.3	5
	U, Th			<0.00019	0.070	28	<0.01	0.44	21
Mushrooms	Po-210	9	1.7	0.039	219	137	0.01	460	3
	Pb-210	0.48	0.32	0.04	3.7	162	0.62	7.4	2
	Ra-226	0.14	0.08	0.031	5.8	10	0.39	8.9	2
	Ra-228	0.22				1	0.26	0.43	2
	U, Th	0.40		<0.0023	9.2	25	0.016	0.71	2

TABLE 21. ACTIVITY CONCENTRATIONS OF NATURAL RADIONUCLIDES OBSERVED IN WILD FOODS, Bq/kg, FRESH WEIGHT (cont.)

		Areas with assumed normal concentrations				Areas with known or suspected high concentrations		
		AM <sup>a</sup>	GM <sup>b</sup>	Min.	Max.	n	Min.	Max.
<b>Meat products</b>								
Reindeer/caribou (meat) [134]	Po-210	9.7	9.2	5.5	16	7	14	1
	Pb-210	3.4	2.2	0.12	6.9	18	1.1	1
	Ra-226						0.21	1
Other game (meat) <sup>d</sup> [135]	Po-210	0.36	0.18	0.01	6.7	85		
	Pb-210	0.25	0.20	0.059	0.46	10		
	U, Th			$5.7 \times 10^{-5}$	0.017	45		
Marine mammal (meat) <sup>e</sup> [136]	Po-210	21	13	1.1	86	30		

<sup>a</sup> Arithmetic mean  
<sup>b</sup> Geometric mean.  
<sup>c</sup> Wild edible vegetables, herbs and fruits (other than berries).  
<sup>d</sup> Wild edible birds, boar/pig and deer (other than reindeer).  
<sup>e</sup> Different species of seal, whale and dolphin. It is unknown whether all species included are commonly consumed.

$^{210}\text{Po}$  concentrations observed in some species are not likely to be due to aerial deposition but rather due to a species specific affinity for uptake. For example, some, but not all, species of the *Boletaceae* family have been found to contain particularly high levels of  $^{210}\text{Po}$ . While observed  $^{210}\text{Po}$  concentrations range from 0.07–0.58 Bq/kg in the red capped scaber mushroom, values ranging from 7.3–220 Bq/kg have been found in foxy bolete, both members of the *Boletaceae* family (Table 22) in areas with normal levels of naturally occurring radioactivity. The species specific differences in uptake from soil emphasize the importance of considering the species actually consumed when assessing exposure. None of the species display very high levels of  $^{210}\text{Pb}$ , suggesting a preferential uptake of  $^{210}\text{Po}$ .

#### 4.5.1.2. *Meat products*

The elevated  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  concentrations observed in reindeer and caribou meat, ranging from 5.5–16 Bq/kg and 0.12–6.9 Bq/kg, respectively, are associated with their diet consisting largely of lichen, especially during the winter. Lichens are slow growing perennials with a large surface area that efficiently retain deposited radionuclides. Hence, they accumulate more  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  than most grazing plants, as summarized by Skuterud et al. [137]. Somewhat enhanced  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  concentrations compared with UNSCEAR reference levels are also observed in other game, although to a lesser extent than reindeer and caribou.

Marine mammalian meat, such as seal, whale and dolphin, is consumed in large quantities by some population groups. These products contain high levels of  $^{210}\text{Po}$  compared to agricultural meat products and most marine fish species. As summarized in Ref. [106], the mean activity concentrations in different species and regions ranged from 1.3–27 Bq/kg in seals, 1.1–23 Bq/kg in whales and 4.5–86 Bq/kg in dolphins. It has been hypothesized that the high  $^{210}\text{Po}$  concentrations in these marine mammals are associated with the high content of red muscle tissue, exhibiting a higher myoglobin content [138].

Internal organs, such as liver, kidney and spleen, have been shown to contain higher  $^{210}\text{Po}$  concentrations than muscles in a variety of species [106]. For example, Macdonald et al. [139] reported geometric mean concentrations of  $^{210}\text{Po}$  of 89–1064 Bq/kg in liver and 56–478 Bq/kg in kidney of caribou in northern Canada. Kauranen et al. [135] reported  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  liver concentrations of 8.9–48 Bq/kg and 0.89–3.1 Bq/kg, respectively, in Eurasian elk in Finland — approximately one order of magnitude higher than in muscle. A similar trend has been seen in marine mammals [138]. For population groups that consume large quantities of offal, radiological assessments need to take the added dose from these products into consideration.

TABLE 22.  $^{210}\text{Po}$  AND  $^{210}\text{Pb}$  ACTIVITY CONCENTRATIONS IN SELECTED SPECIES OF MUSHROOMS FROM AREAS WITH ASSUMED NORMAL LEVELS OF NATURAL RADIOACTIVITY, Bq/kg, FRESH WEIGHT

Family	Species	AM <sup>a</sup>	GM <sup>b</sup>	Min.	Max.	n
<i>Boletaceae</i>	Foxy bolete ( <i>Leccinum vulpinum</i> )	Po-210	48	27	7.3	220
		Pb-210	0.25	0.23	0.1	0.41
	Orange birch bolete ( <i>Leccinum versipelle</i> )	Po-210	25	22	8.2	48
						12
	Birch bolete ( <i>Leccinum scabrum</i> )	Po-210	15	9.7	1.9	52
		Pb-210	0.29	0.25	0.1	0.8
	Red capped scaber mushroom ( <i>Leccinum aurantiacum</i> )	Po-210	0.28	0.23	0.07	0.58
		Pb-210	0.11	0.096	0.04	0.26
<i>Agaricaceae</i>	Parasol mushroom ( <i>Macrolepiota procera</i> )	Po-210	1.0	0.95	0.34	1.7
		Pb-210	0.77	0.74	0.46	1.3
<i>Russulaceae</i>	Tall brittlegill ( <i>Russula paludosa</i> )	Po-210	0.79	0.65	0.2	2.4
		Pb-210	0.49	0.42	0.2	1.2

<sup>a</sup> Arithmetic mean

<sup>b</sup> Geometric mean.

#### 4.5.2. Human-made radionuclides in wild foods

Elevated concentrations of human-made radionuclides are most likely to be present in terrestrial and aquatic environments as a result of unplanned releases, such as previous large scale nuclear accidents. Apart from these acute releases, long term chronic releases can also occur as a result of industrial processes, which may or may not be regulated. In the context of this report, which focuses on chronic exposure due to radionuclides in the diet, we are primarily interested in the recovery phase after such large scale accidents. These are generally referred to as ‘existing exposure situations’. There is no established time frame for when the emergency phase ends and the existing exposure situation commences — this depends on the specific conditions of the accident. However, for the purposes of this publication, the existing exposure situation commences one year after the accident. By this time, releases to the environment have ended and the situation is moving towards greater stability. The existing exposure can continue for many years thereafter.

The radionuclides  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  (jointly referred to as ‘radiocaesium’) are often the more important human-made radionuclides of interest in existing exposure situations. This is due to the relatively long physical half-lives of  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  and the effective transfer of radiocaesium in the food chain. In general, highly organic soils with low levels of caesium fixing clays and low potassium levels will tend to have higher transfer to plants. Under these conditions, radiocaesium may remain available for uptake by plants and mushrooms for decades, and the effective ecological half-life of  $^{137}\text{Cs}$  may eventually approach its physical half-life in some species and ecosystems. In a mobile form (not particle bound), radioactive strontium (primarily  $^{90}\text{Sr}$ ) is readily available for root uptake by plants, and the uptake increases with low calcium levels in soil and low pH.

In existing exposure situations, intake of  $^{131}\text{I}$  is no longer relevant due to its short physical half-life of only eight days. The various radionuclides of plutonium and americium are absorbed by organisms only to a small extent and are therefore considered to be of little importance to ingestion doses from food consumption.

Because transfer factors and ecological half-lives for wild foods are greatly dependent on the time that has passed since fallout and environmental conditions, they are specific to the time and location of a given study and not necessarily applicable to other situations. However, lessons can be learned from past exposure situations that have resulted in the accumulation of human-made radionuclides in wild foods. Three selected cases that have been widely studied are:

- (a) *Fukushima Prefecture after the Fukushima accident in 2011.* Approximately 70% of the surface area of the Prefecture consists of forested areas and received on the order of  $10\text{--}100\text{ kBq/m}^2$   $^{137}\text{Cs}$  [140]. Japanese forest soils

tend to have plentiful amounts of clay minerals, which results in a lower transfer of radiocaesium to plants and animals compared with European forests [141–143].

- (b) *Vulnerable uplands and forests in Norway after the accident at the Chornobyl NPP in 1986.* Mean  $^{137}\text{Cs}$  deposition levels per municipality in Norway varied from virtually none up to  $100 \text{ kBq/m}^2$  [144]. Organic soils poor in mineral nutrients led to high and persistent radiocaesium transfer in many forest and upland ecosystems. Higher radioactive fallout from the accident at the Chornobyl NPP occurred in the regions nearby and is summarized in, for example, Ref. [107]. However, Norway is used here to exemplify radiosensitive forest and upland ecosystems.
- (c) *The Marshall Islands after nuclear weapons testing carried out between 1945 and 1958.* The islands are characterized by a tropical climate and coral soil, consisting primarily of calcium carbonate with no clay minerals, and low in organic matter, potassium and other nutrients. Plant availability is therefore different from most of the areas that have been studied [18, 82].

#### 4.5.2.1. Edible plants and mushrooms

High transfer has been observed in wild forest berries and in edible vegetative plant parts of some species. Food samples collected in 2013–2014 in Kawauchi Village located in the Fukushima Prefecture in Japan showed that while 0.1% and 1.2% of cultivated vegetables and fruits, respectively, exceeded the regulatory limit of  $100 \text{ Bq/kg}$  for radiocaesium, 32.8% of edible wild plants and mushrooms exceeded the same limit [145]. For example, Tsuchiya et al. [146] found high concentrations in select species of Japanese wild edible plants (*sansai*) in Kawachi Village in Fukushima Prefecture, with the highest radiocaesium concentrations being in *Koshiabura* (leaf shoots of *Chengiopanax sciadophylloides*), containing on average  $2800 \text{ Bq/kg}$  fresh weight. Recent  $^{137}\text{Cs}$  measurements in Norwegian wild berries only occasionally exceed  $1000 \text{ Bq/kg}$  [147].

The results reported by Robison et al. [148] from samples collected in 1975 on Bikini Island showed a very high uptake of  $^{137}\text{Cs}$  and  $^{90}\text{Sr}$  in different wild growing edible plants. For example,  $^{90}\text{Sr}$  and  $^{137}\text{Cs}$  concentrations of  $640$  and  $3350 \text{ Bq/kg}$ , respectively, were observed in bread fruit, and  $31$  and  $1900 \text{ Bq/kg}$ , respectively, in coconut milk. While the uptake in these foods is not much higher than that observed in garden vegetables and domesticated animals roaming freely on the island, the case serves as an important example of how local soil properties and diets need to be taken into account when assessing the radiological impact of wild foods.



It has been well documented that wild mushrooms have a great capacity for accumulating some mineral nutrients and metals as well as radiocaesium [1]. Whereas there is a high correlation between  $^{137}\text{Cs}$  and potassium uptake in plants, this is not observed in mushrooms. Potassium values are within a narrow range in mushrooms, regardless of species; however,  $^{137}\text{Cs}$  concentrations vary widely, suggesting that the mechanism for uptake is different from that of potassium in mushrooms. In Kawauchi Village in 2015, the median  $^{137}\text{Cs}$  values for each sampled species ranged from  $<7$  to 1300 Bq/kg [145]. Norwegian monitoring data reveal values reaching 10 000–30 000 Bq/kg during the first few years after the accident at the Chornobyl NPP and a very slow decline since the mid-1990s. The mean levels still exceed 1000 Bq/kg for selected species and districts [147].

Because of the very high radiocaesium transfer to some species, the appearance of mushrooms in the late summer and early autumn may cause a large temporary increase in radiocaesium levels in herbivores that forage for mushrooms, especially in years with a high mushroom abundance.

While mushrooms in general have a higher uptake of radiocaesium than plants, the reverse appears to be the case for radiostrontium [149, 150].

#### 4.5.2.2. *Meat products*

Concentrations of radiocaesium are relatively high in muscle because radiocaesium behaves similarly to potassium in living organisms. High concentrations have been observed in a variety of game animals compared with those in domesticated animals grazing on cultivated fields. For example, elevated uptake has been observed in different types of deer [1].

Reindeer are particularly susceptible to accumulating radiocaesium. As is also the case for natural radionuclides, the very high concentrations of radiocaesium observed during the first years after a radioactive fallout are due to a diet high in lichens, particularly in winter. Radiocaesium levels in individual semidomesticated reindeer reached 150 000 Bq/kg in the most affected areas in Norway following the accident at the Chornobyl NPP [151]. More than 30 years after the accident, individual reindeer are still found to exceed 3000 Bq/kg in Norway.

Unusually high transfer of radiocaesium has also been observed in wild boar (*Sus scrofa*). Wild boar are omnivores, and their diet changes during the seasons — from eating mainly plants in the spring and summer, to burrowing for roots, tubers, larvae and earthworms in the autumn and winter, leading to increased levels of contamination. Samples of wild boar in Fukushima Prefecture displayed mean radiocaesium levels ranging from 610 to 2700 Bq/kg in the period 2012–2015, with individual samples containing up to 33 000 Bq/kg [152]. European studies have also found high and persistent transfer to wild boar after

the accident at the Chornobyl NPP, as seen, for example, in the German Bavarian forests [153, 154].

Domesticated animals grazing exclusively in seminatural ecosystems prior to slaughter have a diet identical to that of game animals and ingest radiocaesium in similar quantities. This is the case with sheep in several countries, for example. Concentrations up to 40 000 Bq/kg were measured in Norwegian sheep after the accident at the Chornobyl NPP [151], and countermeasures are still necessary to reduce  $^{137}\text{Cs}$  levels below the regulatory limit of 600 Bq/kg in the most afflicted districts. The highest overall radiocaesium levels in sheep were reached in 1988 due to very high abundance of wild mushrooms that year. High concentrations of  $^{137}\text{Cs}$  were also observed in mountain sheep grazing upland pastures in the UK and in Ireland in the years following the accident at the Chornobyl NPP [36, 155].

Usually, very little  $^{90}\text{Sr}$  is found in edible animal tissues because  $^{90}\text{Sr}$  is accumulated in calcium rich animal organs, particularly bones and teeth.

#### 4.5.2.3. *Freshwater fish*

A high transfer of radiocaesium to freshwater fish has been well documented in some areas [107]. In Kawauchi Village (Fukushima Prefecture, Japan) in 2013–2014, freshwater fish were some of the most contaminated food items, with 39% of samples exceeding the regulatory limit of 100 Bq/kg [145]. In some river systems in the Hamadori region (along the east coast of the Fukushima Prefecture, where the reactor is located), some species continue to show high concentrations. For example, activity concentrations of  $^{137}\text{Cs}$  of up to 25.6 kBq/kg were measured in white spotted char in 2016 in forest streams [156, 157].

Laboratory experiments with dace established a concentration ratio ( $CR$ ) from water to fish of  $\sim 10$ . By contrast, the  $CR$  values observed in the field were 1240–12 900, clearly demonstrating that the  $^{137}\text{Cs}$  in the water is not the source of the  $^{137}\text{Cs}$  in the flesh. Analysis of the gut content of fish caught in the wild showed that the diet includes both terrestrial and aquatic insects [157]. Fish with a diet of algae containing similar concentrations of  $^{137}\text{Cs}$  showed much lower concentrations of  $^{137}\text{Cs}$  in the flesh, demonstrating the importance of bioavailability in determining the accumulation of  $^{137}\text{Cs}$  in freshwater fish.

Shallow nutrient poor lakes with little turnover are particularly vulnerable to high transfer of radiocaesium to freshwater fish. For example, the transfer of  $^{137}\text{Cs}$  to fish is observed in a nutrient poor subalpine Norwegian lake that has been studied closely since the accident at the Chornobyl NPP and is situated in an area with a mean  $^{137}\text{Cs}$  deposition of 130 kBq/m<sup>2</sup>. In this instance,  $^{137}\text{Cs}$  concentrations in brown trout (*Salmo trutta*) peaked in 1987, with a range of 1070–8400 Bq/kg [158]. For comparison, predatory fish in the large Kiev

Reservoir, located in an area receiving 555 kBq/m<sup>2</sup> of <sup>137</sup>Cs, contained 1000–7000 Bq/kg in the same year [107].

Radionuclides in lakes and rivers may be removed relatively quickly by water transport or by adsorption and sedimentation, often resulting in a quick drop in radionuclide levels in freshwater fish in the first period after the fallout. However, runoff of radionuclides from the catchment area, as well as remobilization from sediments, can represent a long term source of continuous contamination of the freshwater ecosystem and can lead to a slow long term decline in the following decades. In the above mentioned Norwegian reference lake, <sup>137</sup>Cs levels in brown trout declined with an ecological half-life of three to four years during the first years; however, since 2002, the rate of decline has been very slow, approaching the physical half-life of <sup>137</sup>Cs [158].

The transfer of artificial radionuclides in terrestrial and freshwater ecosystems is discussed in more detail in Refs [1, 72].

#### **4.5.3. Discussion**

Certain types of wild foods in different types of ecosystems can accumulate both natural and human-made radionuclides to high concentrations. These include mushrooms, berries and the meat of wild animals. Freshwater fish have also been shown to accumulate high concentrations of human-made radionuclides under certain conditions. In the majority of situations, <sup>210</sup>Po appears to be the most important natural radionuclide, while <sup>137</sup>Cs is the human-made radionuclide likely to contribute most to radiation doses through the consumption of food.

There are no international standards for natural radionuclides in food. Following unplanned releases of <sup>137</sup>Cs to the environment, concentrations in wild foods may exceed national or regional maximum permissible levels or Codex Alimentarius guideline levels for years or even decades. Individual doses will depend on the local radiation situation and lifestyle, in particular for subgroups of the population such as hunters, fishers and those who collect wild foods from the forest. National authorities may need to develop specific advice, and in some cases apply controls, to manage the associated radiation doses.

## 5. STATISTICAL ANALYSIS OF MEASUREMENT DATA FOR NATURAL RADIONUCLIDES IN FOOD

### 5.1. INTRODUCTION

#### 5.1.1. Background

UNSCEAR has published reference values for natural radionuclides in individual food groups, including drinking water, in its 1993 Report [22], and these were updated in 2000 [19]. These compiled data have generated considerable interest in the topic of natural radionuclides in food, and measurement results are widely reported in the scientific literature around the world.

UNSCEAR presented summary data for nine natural radionuclides, namely  $^{238}\text{U}$ ,  $^{235}\text{U}$ ,  $^{232}\text{Th}$ ,  $^{230}\text{Th}$ ,  $^{228}\text{Th}$ ,  $^{228}\text{Ra}$ ,  $^{226}\text{Ra}$ ,  $^{210}\text{Pb}$  and  $^{210}\text{Po}$ . The prevalent radionuclide  $^{40}\text{K}$  is also naturally occurring but was not included in the UNSCEAR reference values (typical worldwide activity concentrations) because levels of  $^{40}\text{K}$  in the human body are more or less uniformly distributed, since levels of natural potassium in the body are under homeostatic control. The six food groups for which reference value data are reported by UNSCEAR are: meat products, milk products, fishery products, grain products, leafy vegetables, and root vegetables and fruits; drinking water is also included (see Section 2).

The UNSCEAR compiled data demonstrate that separate radionuclide–food groups have different ranges of activity concentrations. The purpose of this work is to produce estimates for the global distribution of radioactivity levels in food groups for each of these radionuclides in order to support guidance to Member States on managing them. Clearly, measurements are not available for the entire population of each food product across the world for each radionuclide. The distribution of activity concentration in the whole population has therefore been estimated for each food product using the measurements of activity concentrations that are available, which represent a sample of the whole population.<sup>15</sup> The next section will describe how these sample sets were chosen.

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<sup>15</sup> In statistics, a ‘population’ is the complete set of a defined group, and the ‘sample’ is a representative subset of the population. In this case, the sample is the set of measurement results we have collected, while the population is the complete set of activity concentrations for the radionuclide–food group of interest.

### 5.1.2. Data description

The data on natural radionuclides in food compiled by UNSCEAR included measurement data up to 1998. Since then, a significant amount of additional data has become available. This work analysed this additional data to improve understanding of the levels of radioactivity in food products. Data were collected for nine radionuclides in food products ( $^{238}\text{U}$ ,  $^{235}\text{U}$ ,  $^{232}\text{Th}$ ,  $^{230}\text{Th}$ ,  $^{228}\text{Th}$ ,  $^{228}\text{Ra}$ ,  $^{226}\text{Ra}$ ,  $^{210}\text{Pb}$ ,  $^{210}\text{Po}$ ) measured in the time period 1998–2017.

There were two separate sources of data available for this analysis. Information relating to measurements of natural radionuclides (uranium and thorium decay series) in foods was compiled from available published data as well as those provided to the IAEA from its Member States. At the beginning of the project, it was not clear whether these sample sets were representative of the food populations under investigation. Therefore, one goal of the analysis was to establish whether the data contained any biases.

## 5.2. DATA COLLECTION

Two separate approaches to data collection were used:

- (a) A review of the literature in peer reviewed scientific journals;
- (b) A request to IAEA Member States to provide data collected as part of their monitoring programmes.

It was decided that data from each of the two collection routes would be compiled and analysed separately. The reason for this approach was that the scientific literature in peer reviewed journals might be expected to focus on situations where elevated concentrations have been detected, or are expected, whereas data from IAEA Member States that include data from environmental monitoring programmes are more likely to avoid this type of bias because they routinely sample food in the production and public supply chains. There was therefore an expectation, to be checked at a later time, that the distribution of activity concentrations of natural radionuclides in food published in the scientific literature might be higher than that observed in data collected as part of ongoing monitoring programmes. If this was found to be the case, as was anticipated, merging both datasets would be inappropriate from the viewpoint of statistical evaluation.

An early decision was taken to report all measurement data in terms of fresh weight, as this is how most foods are consumed. It was also decided that

drinking water would be excluded, although a separate assessment was carried out of natural radionuclides in natural mineral waters (see Section 4.3).

### **5.2.1. Published data from scientific literature**

Prior to undertaking the literature review, criteria were established to ensure the consistent selection of relevant publications and the quality of the data to be included. The criteria for accepting or rejecting publications, the associated quality assurance criteria, and the procedures for managing and compiling data that were discussed and agreed with the FAO, UNSCEAR and WHO are described in the Appendix.

Publications were identified by the IAEA Library. These covered scientific publications published in the period 1998–2017. The total number of papers that met the search criteria was 320. Subsequently, a further 3 papers were identified, bringing the total number of papers for screening to 323.

The measurement data for each radionuclide–food product combination were compiled in a spreadsheet, where all relevant information was recorded. Measurement data reported as dry weight were converted to fresh weight using agreed procedures. This spreadsheet contained approximately 8000 individual measurements for radionuclides in food. These data comprise observations of natural radioactivity in food from many different regions of the world.

### **5.2.2. Monitoring programme data from Member States**

A request to provide measurement data on naturally occurring radionuclides in food products collected as part of environmental monitoring programmes or projects was sent to all IAEA Member States in March 2018. Twenty-eight Member States<sup>16</sup> provided data, with a further six replying that they had no relevant data. Data that were already included in the data from the literature review were removed, as they would constitute duplicate data. Data not relevant to the project were also excluded (for example, some of the datasets included measurements of radionuclides in drinking water). Where data were unclear, this was followed up and clarified with the provider.

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<sup>16</sup> Data were provided by Australia, Bahrain, Belgium, Bosnia and Herzegovina, Brazil, Croatia, the Czech Republic, Egypt, France, Germany, Ghana, Hungary, Indonesia, Ireland, the Islamic Republic of Iran, Japan, the Republic of Korea, the Netherlands, New Zealand, Norway, Romania, the Russian Federation, Slovenia, Switzerland, the Syrian Arab Republic, the United Arab Emirates, the United Kingdom and the United States of America.

### 5.3. STATISTICAL ANALYSIS

Many datasets of measurements across different sciences show a skewed distribution, especially when underlying effects are multiplicative, mean values are low, variances are observed to be large and values cannot be negative. Log-normal distributions (Fig. 10) often fit such skewed distributions closely [159, 160]. Therefore, the statistical analyses that were conducted started with the reasonable premise that the activity concentrations of natural radionuclides in foods products are log-normally distributed. The analyses were not dogmatic, if the data could not be characterized by log-normal distributions, then a more appropriate distribution would have been sought. However, the premise of log-normality was found to hold and be useful. Not only was it able to facilitate an efficient review of the available data, but it also provided a deeper insight into the variability of natural radionuclides such as  $^{210}\text{Po}$ ,  $^{210}\text{Pb}$ ,  $^{228}\text{Ra}$  and  $^{226}\text{Ra}$  in foods worldwide.

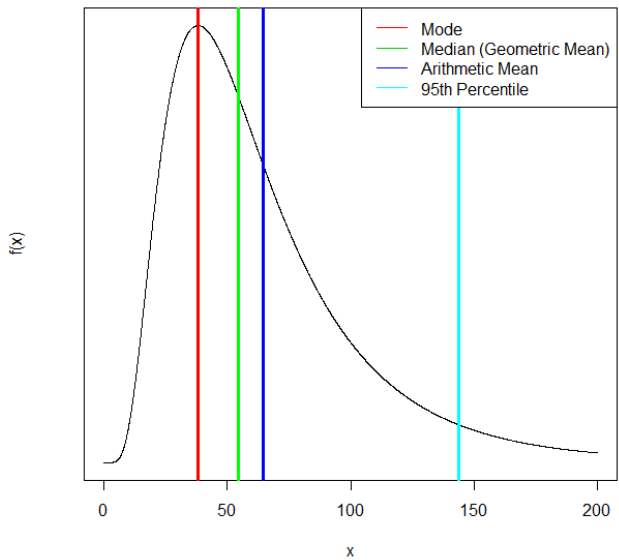


FIG. 10. Example of a log-normal distribution indicating the mode, median (geometric mean), arithmetic mean and 95th percentile values.

The goal of this analysis is to use the sample data to estimate various parameters of the population, namely the median<sup>17</sup>, arithmetic mean and 95th percentile of the population distribution for radionuclides in food. These estimates could provide a basis for guidance relating to natural radionuclides in food.

The statistical analysis of the food measurement data was conducted using R Statistics [161] and consisted of a series of stages, as outlined in Fig. 11.

The initial stages of the analyses focused only on the data collated from the scientific literature — the ‘published dataset’.

The compiled datasets contained information on 38 variables associated with the data and these were used to assist in the formal statistical analyses. However, a formal analysis of the data using all 38 variables was not practical.



*FIG. 11. Summary of statistical analysis process.*

<sup>17</sup> For log-normal distributions the median and geometric mean are identical.



Instead, eight key variables were selected as being of most relevance for the formal statistical analysis of foods to meet the objectives of this work:

- (a) Food category;
- (b) Food subcategory;
- (c) Food product;
- (d) Radionuclide;
- (e) Country;
- (f) Region based on UNEP classification [46];
- (g) WHO GEMS/Food cluster diet region [43];
- (h) Activity concentration (fresh weight).

The purpose of the analysis was to predict the activity concentration variable. It was found that the variables of most use for predicting were radionuclide, food subcategory and, in some cases, food product. Other variables could be used for statistical analysis in the future if needed.

### **5.3.1. Statistical analysis of published and Member State datasets**

The datasets categorized food products at three different levels: food category (e.g. fish products), food subcategory (e.g. crustacean) and food product (e.g. crab). The first step was to determine the appropriate level at which to conduct the statistical analyses.

As mentioned previously, the population is assumed to follow a log-normal distribution. The first stage of the analysis was to establish whether the sample data were representative of a log-normal distribution.

Analysis at the food category level showed that the sample was not representative of a log-normal distribution (quantile–quantile (Q–Q) plots (Section 5.3.2) indicated that the sample was not from a single homogeneous population). This indicated that a subcategorization of the data was necessary, and the options available were food subcategory or region (based on UNEP classification). Analysis by region was conducted, and it was found that this variable did not yield useful outputs. Therefore, it was necessary to conduct statistical analyses at the food subcategory level. In some cases, when the analysis at the food subcategory level did not yield useful outputs, further analysis was carried out at the food product level.

Once it had been determined that the analyses were to be conducted at the food subcategory level, a more detailed review of the datasets was conducted to identify potential errors and outliers in the measurement data sample. The published dataset contained data divided into 21 food subcategories, covering 9 radionuclides, yielding a total of 189 potential subsets for analysis (Table 23).

TABLE 23. FOOD CATEGORIES AND SUBCATEGORIES IN THE PUBLISHED AND MERGED DATASETS

Fish products	Fruit	Grain	Meat products	Milk products	Vegetables	Miscellaneous
Bivalves	Fruit	Grain	Eggs	Liquid milk	Leafy vegetables	Beverages
Crustaceans			Meat	Milk products	Root vegetables	Herbs
Freshwater fish			Offal			Honey and sugar
Non-bivalve molluscs						Mushrooms
Saltwater fish						Nuts
						Seaweed
						Uncategorized

However, not all of these 189 subsets contained sufficient measurement data for rigorous statistical analysis. Similarly, the Member States dataset also contained 189 equivalent subsets, of which some had insufficient data for analysis.

The detailed review of the published and Member States datasets identified a number of potential errors, which were checked and corrected, where necessary. For example, potential errors in the units for activity concentrations were identified for a number of measurement data, as well as incorrect conversions from dry to fresh weight. Where possible, potential errors associated with the data were checked with either the author(s) of the published papers or national contact points for the organizations that submitted Member State data and corrections made.

Having resolved issues with the measurement data for both the published dataset and Member States dataset, it was found that the sample data at the radionuclide–food subcategory level was representative of a log-normal distribution for each dataset.

While each dataset was found to be representative of a log-normal distribution, it was not clear whether the published data were biased and displayed

higher activities than the Member States data (Section 5.1). To establish whether a bias existed, the published and Member States data were merged. The distribution of the merged dataset was examined using Q–Q plots and formal goodness of fit hypothesis tests to determine whether the merged data were representative of a single homogeneous population.

This process identified a further set of potential errors in both datasets. As previously, these were either clarified and corrected through liaison with the authors and data providers or deleted entirely from the spreadsheet. Having resolved issues with the merged dataset, it was found that the sample data at the radionuclide–food subcategory level were representative of a log-normal distribution.<sup>18</sup> This was a significant result, which enabled the analysis to be conducted on the merged dataset, leading to estimation of the population parameters.

One of the issues identified was the incorrect unit conversion of measurement data. For example, from mBq/kg to Bq/kg and from µg/kg to Bq/kg (see 5.3.1.1).

#### 5.3.1.1. *Conversion of total U and Th measurement results to activity concentrations*

A number of papers reviewed and included in the datasets for food include total uranium and total thorium measurements in µg/kg and mg/kg, which have been typically determined via inductively coupled plasma mass spectrometry (ICP-MS) or other mass spectrometry methods. These values have been converted to Bq/kg by assuming that the total uranium measured is <sup>238</sup>U and the total thorium measured is <sup>232</sup>Th.

The conversion from total U to <sup>238</sup>U activity is calculated using the specific activity of <sup>238</sup>U, which is  $12.4 \times 10^{-3}$  Bq/µg.

The conversion from total Th to <sup>232</sup>Th activity is calculated using the specific activity of <sup>232</sup>Th, which is  $4.1 \times 10^{-3}$  Bq/µg.

For example, if the total U content is 1 µg/g of the sample, then the overall activity is:

$$1 \frac{\mu\text{g}}{\text{g}} \times 12.4 \times 10^{-3} \frac{\text{Bq}}{\mu\text{g}} = 12.4 \times 10^{-3} \frac{\text{Bq}}{\text{g}} = 12.4 \frac{\text{Bq}}{\text{kg}} \quad (3)$$

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<sup>18</sup> The only exception was for <sup>228</sup>Ra in saltwater fish (see Table 28).

Similarly, if the total Th content is 1 µg/g of the sample, then the overall activity is:

$$1 \frac{\mu\text{g}}{\text{g}} \times 4.1 \times 10^{-3} \frac{\text{Bq}}{\mu\text{g}} = 4.1 \times 10^{-3} \frac{\text{Bq}}{\text{g}} = 4.1 \frac{\text{Bq}}{\text{kg}} \quad (4)$$

### 5.3.2. Statistical analysis of merged datasets

Having established that the samples are representative of the log-normal populations, point estimates of the median and 95th percentile values and the confidence interval estimate of the arithmetic mean of the population distribution were determined. An iterative process of analysis and reanalysis was undertaken for each radionuclide in order to produce a series of datasets for categories of food products, with the foods in each dataset being representative of a log-normal distribution for the activity concentration of that particular radionuclide.

For the radionuclide–food subcategory subsets that contained a sufficient sample size<sup>19</sup>, the log transformed measurement data were inspected to identify any outliers. Three different plots of the log transformed data were considered: box plot, histogram and normal Q–Q plot. These three plots were used not only to identify outliers but also to verify the log-normal distribution of the data.

As an additional check on the merged dataset, the sample data were ranked by activity concentrations. If the values from one dataset tended to be consistently higher than the other, it was a sign that merging the two datasets might not be appropriate.

Once the Q–Q plot was produced, potential outliers from the log-normal distribution were examined in more detail. In a lot of cases, there were obvious reasons for these outliers, such as the samples being located in high natural background radiation areas or in areas located near facilities, such as mines, discharging elevated levels of natural radioactivity into the environment. In these cases, the outliers were excluded from the analysis, as the measured activity concentrations are elevated for a particular reason and do not describe the typical behaviour of these radionuclides in food.

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<sup>19</sup> If the sample subset contained a very small sample, then the median result of the measurement data was reported (Annex II). Determination of an appropriate sample size was based on consideration of the power of the goodness of fit tests that were used to check log-normality and also on the accuracy of the estimates produced, in particular for the confidence intervals for the mean and the 95th percentile values.

Outliers at the lower end of the sample distribution could sometimes be attributed to the measurement technique. For example, analyses conducted by ICP-MS typically reported much lower activity concentrations in foods compared to other techniques that could only report minimum detectable activities or limits. The removal of outliers was not straightforward and relied on a thorough review of the literature, clarification of measurement data reported by Member States and expert judgement.

Once these outliers were removed, new Q–Q plots of the remaining data were produced, and the homogeneity of the data was checked. Q–Q plots are an especially useful tool for identifying whether a sample of data comes from a single homogeneous population or from multiple populations. Large deviations from the straight line in a Q–Q plot and data following S-shaped curves are especially strong indicators that the data do not come from a single homogeneous population but instead from multiple populations. In this case, it was an indicator that the food subcategory needed to be split further into individual foods for statistical analysis. An example of analysis at the different food category levels is outlined in Annex III.

Once the log-normal distribution of the dataset was confirmed visually through the Q–Q plot and any outliers were addressed appropriately, the log-normality of the data was validated through formal goodness of fit hypothesis tests (Shapiro–Wilk, Anderson–Darling and Kolmogorov–Smirnov).

Having determined through Q–Q plots and goodness of fit tests that the sample was from a single homogeneous log-normal population, it could be used to estimate the necessary population parameters. Specifically, estimates were produced for the population arithmetic mean, the population median and the upper 95th percentile of the population (Tables 24–29).

If the initial distribution was not log-normally distributed, further investigation of the data was performed. This involved checking for transcription errors, determining whether the measurements were conducted in areas of high natural radiation, checking for unit conversion errors and reviewing measurement techniques (Section 5.3.3.1).

As a further check, the empirical percentage of observations in the sample that was above the estimated 95th percentile was determined. If the empirical percentage was very different from the predicted 5%, then this indicated that, despite meeting the other tests used, the model was not able to predict the sample data appropriately and therefore further investigation was necessary.

These different checks cannot completely determine whether the model fit is perfect. However, the analysis procedure employed, which combined graphical tools with formal hypothesis testing and final validation with empirical data, was the most robust approach possible.

The confidence interval estimate of the population arithmetic mean was calculated as follows. The activities  $X$  were log transformed to obtain  $Y = \log(X)$ , and these were used in the following formula [162]:

$$\bar{Y} + \frac{S_Y^2}{2} \mp Z_{\text{crit}} \sqrt{\frac{S_Y^2}{n} + \frac{S_Y^4}{2(n-1)}} \quad (5)$$

where

$n$  is the sample size;

$\bar{Y}$  and  $S_Y$  are the sample mean and the sample standard deviation of the log transformed activities, respectively;

and  $Z_{\text{crit}}$  is the critical value from the standard normal distribution.

### 5.3.3. Results of statistical analyses

The median confidence interval for the population arithmetic mean and upper 95th percentile values are reported in Bq/kg, fresh weight, and summarized in Figs 12–15 and Tables 24–29. Statistical analyses were conducted for food subcategories and some food products for  $^{210}\text{Po}$ ,  $^{210}\text{Pb}$ ,  $^{226}\text{Ra}$  and  $^{228}\text{Ra}$ . The median values of the radionuclide–food subcategory subsets that were deemed to have an insufficient sample size for analysis are presented in Annex II.

#### 5.3.3.1. Statistical analysis of $^{238}\text{U}$ and $^{232}\text{Th}$

The measurement techniques used for the determination of  $^{238}\text{U}$  and  $^{232}\text{Th}$  have been divided into two broad categories: ‘indirect’ and ‘direct’. Indirect measurement techniques are those that do not measure the  $^{238}\text{U}$  and  $^{232}\text{Th}$  isotopes directly; instead, their progeny are measured, and the assumption is made that these progeny are in secular equilibrium with the  $^{238}\text{U}$  and  $^{232}\text{Th}$  parent isotopes. One of the most common indirect methods is gamma spectrometry, where the assumption is made that the short lived radon decay products such as  $^{214}\text{Pb}$  and  $^{228}\text{Ac}$  are in equilibrium with  $^{238}\text{U}$  and  $^{232}\text{Th}$ , respectively (after allowing for suitable ingrowth of these daughter products over a given time). This technique is routinely used for the measurement of  $^{238}\text{U}$  and  $^{232}\text{Th}$  in soil samples and building materials, where this assumption is typically valid. However, this is typically not the case for food products, where partitioning of natural radionuclides is modified through various biological factors and subsequently through processing of food products for human consumption [19]. Direct methods measure the  $^{238}\text{U}$  and  $^{232}\text{Th}$  radionuclides directly; the most common direct approaches are

alpha spectrometry and ICP-MS, which involve radiochemical separation of the radionuclides from the food samples prior to analysis.

When conducting the statistical analysis for  $^{238}\text{U}$  and  $^{232}\text{Th}$  in food products, both direct and indirect measurement techniques were included, resulting in significant difficulties in the statistical analysis.

As a result of the significant difficulties in analysing the  $^{238}\text{U}$  and  $^{232}\text{Th}$  data with both direct and indirect methods included, separate statistical analyses were conducted on activity concentration data determined by direct and indirect measurement techniques for the published dataset. Member States data could not be used, as there was no information available on the measurement techniques used. For example, the published dataset for  $^{238}\text{U}$  in grain contains 79 measurements from 17 publications. Twenty-two of these measurements were gamma spectrometry measurements with results reported that were below detection limits and excluded from the analysis. The indirect measurement techniques used to determine the  $^{238}\text{U}$  activity concentrations were gamma

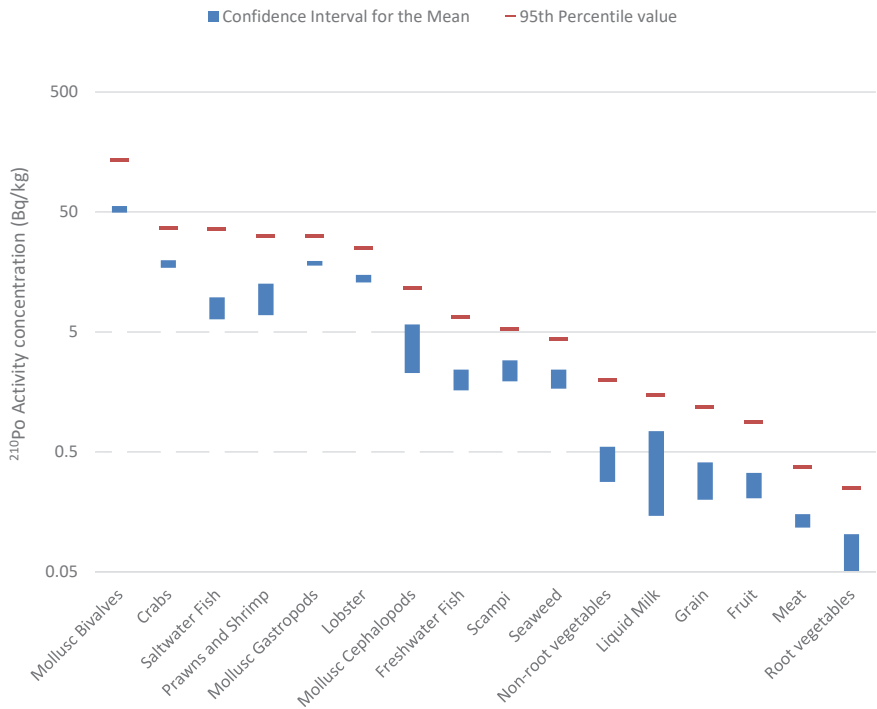


FIG. 12. Polonium-210 activity in food confidence interval for the arithmetic mean and 95th percentile values.

TABLE 24. SUMMARY OF <sup>210</sup>Po ACTIVITY CONCENTRATIONS IN AQUATIC FOOD

Food	Number of data points ( <i>n</i> ) above detection limits	Median (Bq/kg)	Lower confidence interval for the arithmetic mean (Bq/kg)	Upper confidence interval for the arithmetic mean (Bq/kg)	95th percentile (Bq/kg)	Note
Mollusc bivalves	663	40.37	49.47	55.97	134.0	
Crabs	191	16.40	17.10	19.80	37.0	
Saltwater fish	386	2.40	6.38	9.70	36.0	
Prawns and shrimp	59	6.40	6.89	12.60	32.0	
Mollusc gastropods	292	17.48	17.89	19.49	32.0	UK data only
Lobster	143	12.74	12.95	14.91	25.0	
Mollusc cephalopods	25	2.35	2.27	5.76	11.6	
Freshwater fish	222	0.99	1.63	2.42	6.7	
Norwegian lobster/scampi	38	2.00	1.94	2.90	5.3	
Seaweed	43	1.70	1.68	2.42	4.3	



TABLE 25. SUMMARY OF <sup>210</sup>Po ACTIVITY CONCENTRATIONS IN TERRESTRIAL FOOD

Food	Number of data points (n) above detection limits	Median (Bq/kg)	Lower confidence interval for the arithmetic mean (Bq/kg)	Upper confidence interval for the arithmetic mean (Bq/kg)	95th percentile (Bq/kg)	Note
Non-root vegetables	216	0.12	0.28	0.55	2.00	
Liquid milk	57	0.07	0.15	0.75	1.50	
Grain	119	0.14	0.20	0.41	1.50	The 95th percentile value was rounded from 1.2 Bq/kg to 1.5 Bq/kg to ensure that the percentage of results in the sample set above the 95th percentile value was 5%
Fruit	161	0.12	0.20	0.33	0.87	
Meat	203	0.10	0.12	0.15	0.37	
Root vegetables	46	0.04	0.05	0.10	0.25	Due to the small sample size, a conservative 95th percentile value of 0.25 Bq/kg was selected

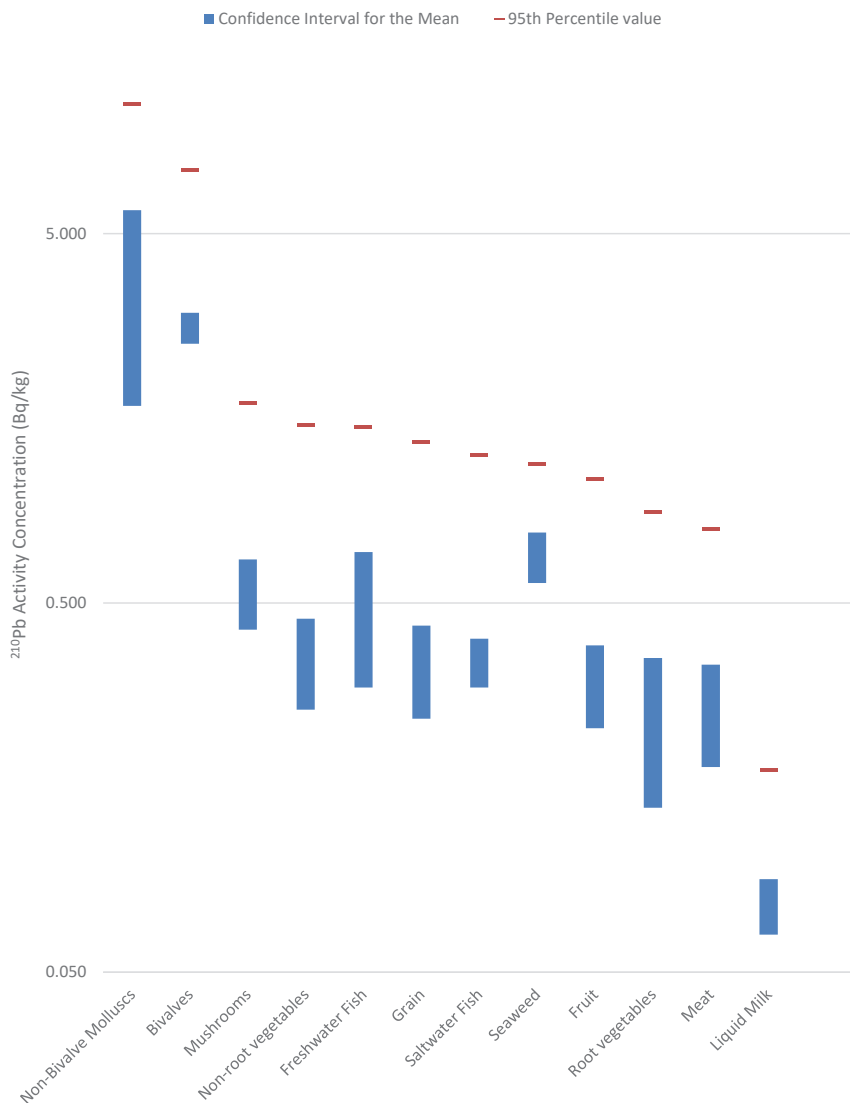


FIG. 13. Lead-210 activity in food confidence interval for the arithmetic mean and 95th percentile values.

spectrometry and solid state nuclear track detectors. The direct methods used were alpha spectrometry, ICP-MS, internal neutron activation analysis and fluorimetry.

The published dataset for  $^{232}\text{Th}$  in grain contains 126 measurements from 19 publications. Twenty-two of these measurements were below detection limits and were excluded from the analysis. The indirect and direct measurement

TABLE 26. SUMMARY OF <sup>210</sup>Pb ACTIVITY CONCENTRATIONS IN AQUATIC FOOD

Food	Number of data points ( <i>n</i> ) above detection limits	Median (Bq/kg)	Lower confidence interval for the arithmetic mean (Bq/kg)	Upper confidence interval for the arithmetic mean (Bq/kg)	95th percentile (Bq/kg)	Note
Mollusc non-bivalves	18	2.00	1.71	5.78	11.24	Small dataset
Mollusc bivalves	351	1.99	2.52	3.05	7.40	
Freshwater fish	59	0.23	0.30	0.69	1.50	
Saltwater fish	330	0.20	0.30	0.40	1.40	The 95th percentile value was rounded from 1.8 Bq/kg to 1.5 Bq/kg to ensure that the percentage of results in the sample set above the 95th percentile value was 5%
Seaweed	27	0.62	0.57	0.78	1.19	
Crustaceans	129	0.14	0.16	0.22	0.53	

TABLE 27. SUMMARY OF <sup>210</sup>Pb ACTIVITY CONCENTRATIONS IN TERRESTRIAL FOOD

Food	Number of data points ( <i>n</i> ) above detection limits	Median (Bq/kg)	Lower confidence interval for the arithmetic mean (Bq/kg)	Upper confidence interval for the arithmetic mean (Bq/kg)	95th percentile (Bq/kg)	Note
Mushrooms	132	0.32	0.42	0.66	1.73	
Non-root vegetables	235	0.13	0.26	0.45	1.50	
Grain	153	0.15	0.24	0.43	1.50	
Fruit	160	0.14	0.23	0.38	1.10	
Root vegetables	90	0.07	0.14	0.36	0.88	
Meat	60	0.15	0.18	0.34	0.79	
Liquid milk	71	0.06	0.06	0.09	0.18	

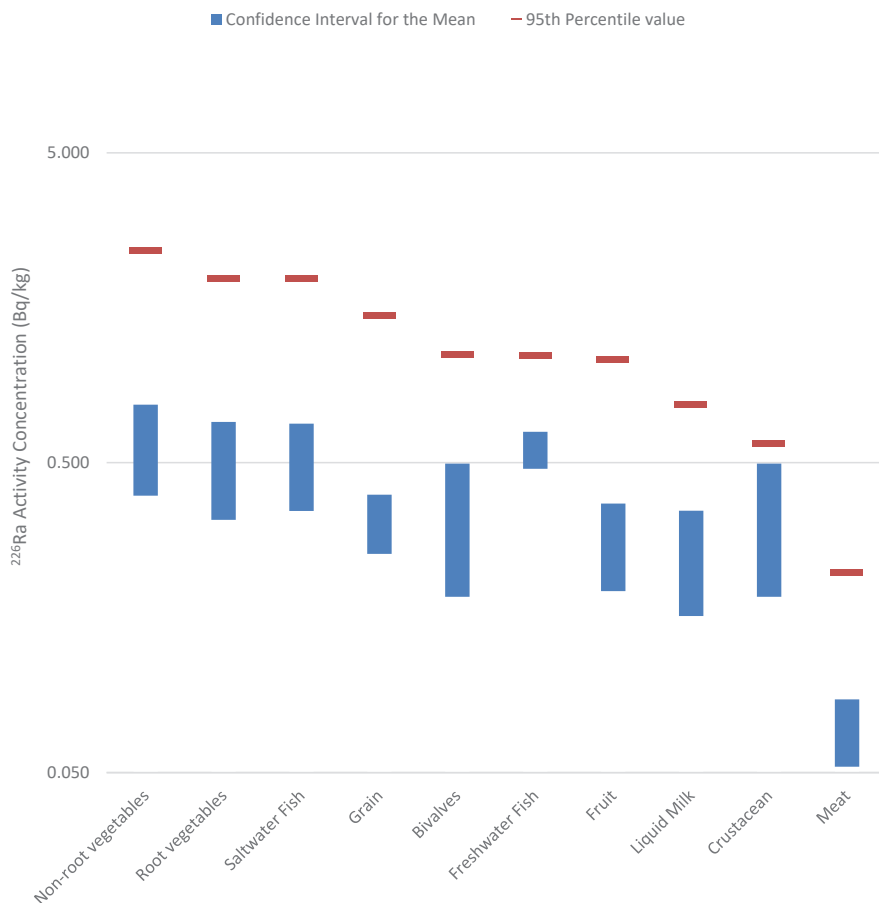


FIG. 14. Radon-226 activity in food confidence interval for the arithmetic mean and 95th percentile values.

techniques used to determine the  $^{232}\text{Th}$  activity concentrations were identical to those for  $^{238}\text{U}$ , apart from fluorimetry, which was not used as a direct technique.

The highest measured  $^{238}\text{U}$  and  $^{232}\text{Th}$  activity concentrations reported were those that used the indirect gamma spectrometry measurement technique. The indirect methods reported activity concentrations that were higher than those reported using direct methods (Table 30).

The median activity concentrations for the direct methods are three orders of magnitude smaller than those determined using the indirect methods. Results from the statistical analysis also indicate significant differences in 95th percentile values.

TABLE 28. SUMMARY OF <sup>226</sup>Ra ACTIVITY CONCENTRATIONS IN FOOD

Food	Number of data points ( <i>n</i> ) above detection limits	Median (Bq/kg)	Lower confidence interval for the arithmetic mean (Bq/kg)	Upper confidence interval for the arithmetic mean (Bq/kg)	95th percentile (Bq/kg)	Note
Leafy vegetables	324	0.11	0.39	0.77	2.40	
Root vegetables	239	0.11	0.33	0.68	2.10	
Saltwater fish	95	0.27	0.35	0.67	2.00	
Grain	292	0.12	0.25	0.39	1.50 <sup>a</sup>	
Mollusc bivalves	37	0.16	0.18	0.50	1.10	
Freshwater fish	59	0.48	0.48	0.63	1.10	
Fruit	193	0.08	0.19	0.37	1.10	
Liquid milk	147	0.05	0.16	0.35	0.77	
Crustacean	20	0.06	0.18	0.50	0.58	Small dataset
Meat	90	0.04	0.05	0.09	0.22	

<sup>a</sup> The 95th percentile value was rounded from 1.1 Bq/kg to 1.5 Bq/kg to ensure that the percentage of results in the sample set above the 95th percentile value was 5%.

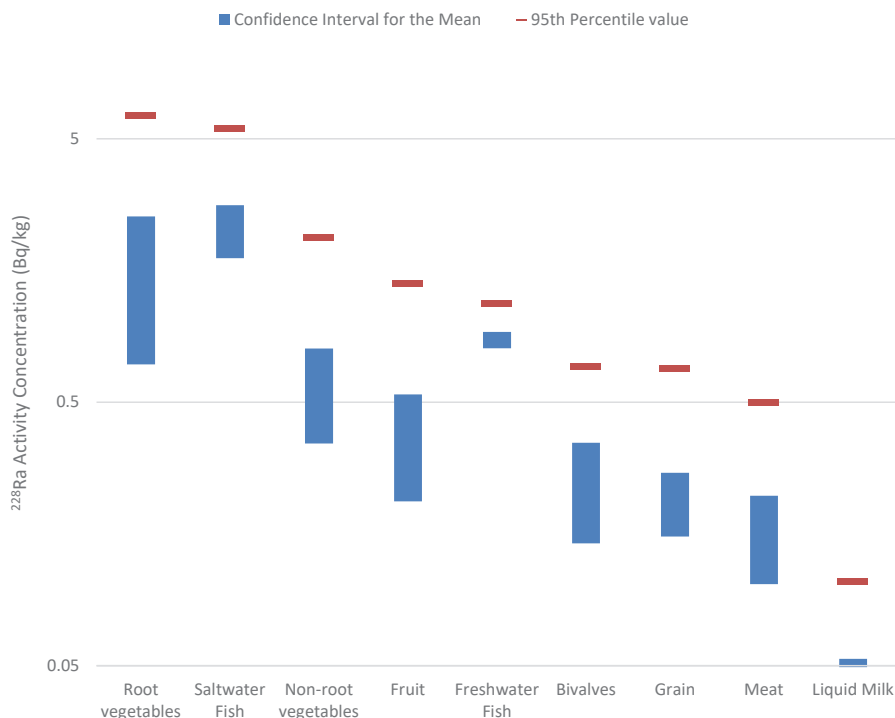


FIG. 15. Radium-228 activity in food confidence interval for the arithmetic mean and 95th percentile values.

Reviewing the number of measurements for  $^{238}\text{U}$  and  $^{232}\text{Th}$  in grain samples, it is also clear that indirect measurement methods, mainly gamma spectrometry, are the most common measurement technique for both  $^{238}\text{U}$  and  $^{232}\text{Th}$ . This approach is likely to be the most common, as it is the most straightforward to use and does not involve any radiochemical separation prior to analysis. However, the assumption that both  $^{238}\text{U}$  and  $^{232}\text{Th}$  are in equilibrium with  $^{226}\text{Ra}$  and  $^{228}\text{Ra}$ , respectively, cannot be used for food products. Although this approach cannot be used for the measurement of  $^{238}\text{U}$  and  $^{232}\text{Th}$  in food samples, other gamma spectrometry methods are available for the measurement of  $^{238}\text{U}$  using the short lived progeny  $^{234}\text{Th}$  and  $^{234\text{m}}\text{Pa}$  [163].

TABLE 29. SUMMARY OF <sup>228</sup>Ra ACTIVITY CONCENTRATIONS IN FOOD

Food	Number of data points ( <i>n</i> ) above detection limits	Median (Bq/kg)	Lower confidence interval for the arithmetic mean (Bq/kg)	Upper confidence interval for the arithmetic mean (Bq/kg)	95th percentile (Bq/kg)	Note
Root vegetables	82	0.31	0.70	2.54	6.1	
Saltwater fish <sup>a</sup>	43	1.75	1.76	2.80	5.5	
Non-root vegetables	188	0.12	0.35	0.80	2.1	
Fruit	94	0.11	0.21	0.54	1.4	
Freshwater fish	32	0.84	0.80	0.93	1.2	Small dataset
Mollusc bivalves	19	0.17	0.15	0.35	0.68	Small dataset
Grain	90	0.12	0.15	0.27	0.67	
Meat	46	0.09	0.10	0.22	0.50	
Liquid milk	101	0.04	0.04	0.05	0.10	

<sup>a</sup> Published data only.



TABLE 30. SUMMARY OF <sup>238</sup>U AND <sup>232</sup>Th IN GRAIN STATISTICAL ANALYSIS USING DIRECT AND INDIRECT METHODS

Nuclide	Measurement technique	No. of measurements (n)	Median activity (Bq/kg)	Mean (Bq/kg)		95th percentile (Bq/kg)
				Lower confidence interval <sup>a</sup>	Upper confidence interval <sup>a</sup>	
U-238	Direct	24	$5.5 \times 10^{-3}$	$17.2 \times 10^{-3}$	0.27	0.23
	Indirect	55	3.2	3.15	12.32	27
Th-232	Direct	17	$2.9 \times 10^{-3}$	$5.5 \times 10^{-3}$	0.12	0.11
	Indirect	109	1.1	2.04	4.35	12

<sup>a</sup> These are the 95% confidence intervals of the population arithmetic mean.

## 6. SUMMARY

### 6.1. INTRODUCTION

Previous sections have reviewed and summarized the available measurement data on natural and human-made radionuclides in food, including natural mineral water. Studies of doses from the total diet undertaken by TDS, MBS, DDS and CMS have also been reviewed. This has allowed the relative contribution of different radionuclides to individual doses to be quantified. The accumulation of radionuclides in wild foods and the potential impact of aquaculture on the radiation doses received by consumers of fishery products have also been evaluated.

A review has been undertaken of measurement data for the years 1998–2017 of the activity concentrations of nine natural radionuclides from the uranium and thorium series —  $^{238}\text{U}$ ,  $^{235}\text{U}$ ,  $^{234}\text{Th}$ ,  $^{232}\text{Th}$ ,  $^{228}\text{Th}$ ,  $^{228}\text{Ra}$ ,  $^{226}\text{Ra}$ ,  $^{210}\text{Pb}$  and  $^{210}\text{Po}$  — in foods destined for human consumption. These data were sourced from over 300 publications in the scientific literature and, in addition, monitoring data were provided by national authorities in 28 Member States. A review of the scientific literature also identified radionuclides other than those in the uranium and thorium series that can be contributors to ingestion dose. These include human-made radionuclides such as  $^{90}\text{Sr}$ ,  $^{134}\text{Cs}$ ,  $^{137}\text{Cs}$  and  $^{90}\text{Sr}$ , which exist in the environment as a result of nuclear weapons fallout, discharges from nuclear facilities and past accidents. The levels of  $^{14}\text{C}$ , a cosmogenic radionuclide, in food can also be elevated as a result of these human practices.

The main results and conclusions of the work undertaken are summarized below. Some areas for further work are also identified.

### 6.2. POTASSIUM-40

Potassium-40 is an important radionuclide found in varying concentrations in all foods. Absorption of potassium, and therefore also of  $^{40}\text{K}$ , by the body is controlled by metabolic processes. For this reason,  $^{40}\text{K}$  delivers a fixed annual radiation dose of  $\sim 170\text{ }\mu\text{Sv}$  to all individuals. Small variability in this dose is observed on the basis of gender and age. The dose from  $^{40}\text{K}$  cannot be eliminated or controlled, and so it is excluded from consideration under the IAEA Safety Standards. For this reason, the concentrations of  $^{40}\text{K}$  in various foods are not documented in this report, and the dose from its ingestion is not considered.

### 6.3. URANIUM AND THORIUM SERIES RADIONUCLIDES IN FOOD

Four radionuclides —  $^{226}\text{Ra}$ ,  $^{210}\text{Pb}$  and  $^{210}\text{Po}$  from the uranium decay series and  $^{228}\text{Ra}$  from the thorium decay series — together represent approximately 90% of the total dose from all radionuclides in the diet. These are worldwide average values, and there will always be variability at the national, regional, local and individual levels. Seasonal variability is also observed, reflecting availability or personal choice in the consumption of foods at different times of the year. Together, these four radionuclides contribute an annual individual dose that is typically 0.24 mSv. From the limited data available, there appears to be no significant difference in the doses received by infants, children and adults.

Polonium-210 is the radionuclide that contributes most to individual dose, representing approximately 52% of the total dose, that is, 0.14 mSv of the 0.24 mSv referred to above. However,  $^{210}\text{Po}$  is known to concentrate preferentially in fishery products, and high consumers of molluscs, in particular, can receive individual doses of up to a few mSv from this radionuclide alone.

The aquaculture industry, also known as fish farming, currently contributes approximately 50% of the total worldwide fishery products consumed. This is expected to increase in future years. Based on very limited data, it would appear that the  $^{210}\text{Po}$  concentrations are considerably lower, by a factor of 10–100, in farmed fish that are artificially fed compared with those that are not fed or are captured in the wild. Molluscs, which contain the highest concentrations of  $^{210}\text{Po}$  of all foods, are normally not artificially fed, and therefore the concentrations will not be lower in those that are produced by aquaculture.

Most wild foods do not display high concentrations of natural radionuclides. However, elevated concentrations of a few tens of Bq/kg of  $^{210}\text{Po}$  have been observed in reindeer and caribou meat, the meat of marine mammals and several species of wild mushrooms.

The reported concentrations of  $^{238}\text{U}$  and  $^{232}\text{Th}$  in food appear to be highly dependent on the analytical technique used. Alpha spectrometry and mass spectrometry methods measure the radionuclides of interest directly. Other analytical techniques, for example gamma spectrometry, involve inferring the activity concentrations of these radionuclides from the activity concentrations of related radionuclides that have been measured. This latter technique can give results that are higher by an order of magnitude or more than the direct measurements if incorrect assumptions are made about the equilibrium of  $^{238}\text{U}$  and  $^{232}\text{Th}$  with the short lived radon decay products of  $^{214}\text{Pb}$  and  $^{228}\text{Ac}$ . For this reason, the use of analytical techniques that measure the presence of  $^{238}\text{U}$  and  $^{232}\text{Th}$  directly or appropriate gamma spectrometry techniques, such as those described in Ref. [163], are preferred.

Natural mineral waters are classified as a food and are covered by the standards of the Joint FAO/WHO Codex Alimentarius Commission. In general, the average radiation doses from consumption are low, typically  $\sim 10 \mu\text{Sv}$  or less in a year. Unlike other foods, the dominant radionuclide in terms of dose is  $^{210}\text{Pb}$  rather than  $^{210}\text{Po}$ . Certain individual natural mineral waters show highly elevated concentrations of natural radionuclides and, at typical consumption rates, could deliver annual radiation doses of up to a few mSv.

#### 6.4. OTHER RADIONUCLIDES IN FOOD

There are several sources of human-made radionuclides throughout the environment, and they are therefore present in the terrestrial and aquatic ecosystems from which food is sourced. The emergency situations that exist in the immediate aftermath of a nuclear accident are outside the scope of this review. However, the longer recovery phase (non-emergency situation), when the concentrations of human-made radionuclides in food may still be elevated compared to those that were present prior to the accident, is included.

Apart from the four natural radionuclides mentioned above, the other important radionuclides that contribute to dose in non-emergency situations are  $^{14}\text{C}$ ,  $^{90}\text{Sr}$  and  $^{137+134}\text{Cs}$ . Carbon-14 occurs naturally in the environment but is also produced by nuclear fuel cycle activities. Caesium-134, which has been released to the environment as a consequence of previous nuclear accidents, can be important in the short term but, because of its relatively short half-life of 2.1 years, it is of less significance in the longer term. Iodine-131 is an important source of radiation dose to the thyroid gland in the immediate aftermath of a nuclear accident but has normally decayed to insignificant concentrations by the time the emergency has ended, and the long term recovery phase has begun. Other human-made radionuclides, such as plutonium radionuclides and  $^{241}\text{Am}$ , are poorly assimilated into plants and animals and are therefore relatively unimportant as a source of radiation dose from the diet.

In general,  $^{14}\text{C}$ ,  $^{90}\text{Sr}$  and  $^{137+134}\text{Cs}$  contribute  $\sim 5\%$  to the ingestion dose. Of this, over half comes from  $^{14}\text{C}$ , with the remainder split equally between  $^{137+134}\text{Cs}$  and  $^{90}\text{Sr}$ . As is the case for the four radionuclides in the uranium and thorium decay series, these are worldwide average values that may not represent the situation nationally, particularly for those Member States that have been affected by past nuclear accidents. All other radionuclides together represent less than 3% of the average ingestion dose from the diet.

Typical concentrations of  $^{137+134}\text{Cs}$  and  $^{90}\text{Sr}$  in most foods are a few Bq/kg or less. As a result of past nuclear accidents, in parts of Belarus, Japan, the Russian

Federation and Ukraine, the concentrations in some foods are higher by up to an order of magnitude.

Wild foods, such as the meat of game animals, forest mushrooms and berries, and Japanese mountain vegetables (sansai), are known to concentrate  $^{137+134}\text{Cs}$ , and elevated concentrations can persist for many decades. Domesticated animals grazing organic soils can also accumulate  $^{137+134}\text{Cs}$  in their flesh. Concentrations of some tens of thousands of Bq/kg have been observed in wild foods in Europe in the aftermath of the accident at the Chornobyl NPP from 1986 up to the current time. Somewhat lower, but still elevated, concentrations have been observed in Japan following the Fukushima Daiichi NPP accident in 2011.

Elevated concentrations of  $^{137+134}\text{Cs}$  have also been observed in fish inhabiting nutrient poor freshwater lakes. As for wild foods, concentrations of several tens of Bq/kg have been reported. Strontium-90 also accumulates in freshwater fish. Because it concentrates in the bones, the concentrations in the flesh tend to be lower than those of  $^{137+134}\text{Cs}$ .

The concentrations and associated doses from human-made radionuclides in natural mineral waters were not evaluated. Most natural mineral waters come from underground sources, and it can therefore be assumed that the concentrations of human-made radionuclides are negligible.

## 6.5. ACTIVITY CONCENTRATIONS IN INDIVIDUAL FOODS

The measurement data on the activity concentrations of individual radionuclides in foods have been collated into 8 food categories comprising a total of 20 different food subcategories. The food subcategory datasets have been tested for log-normality, and appropriate statistical analyses have been applied to derive the 95th percentile values of the population. In some cases, the log-normality of the data comprising the food subcategory could not be verified, and subsequent statistical analyses were conducted at the individual food or species level. In many cases, this involved the removal of outliers originating from food products coming from high natural background radiation areas or areas influenced by discharges of naturally occurring radioactive material from mining or other industrial processing facilities. Comparison with the 95th percentile value allows national authorities to determine whether their own measurements fall within or outside the worldwide distribution for that specific radionuclide and food.

The 95th percentile value has been derived only for those datasets where the total number of observations is relatively large and the data behave homogeneously; that is, they approximate well to a log-normal distribution. This is the case for the four most important radionuclides of  $^{228}\text{Ra}$ ,  $^{226}\text{Ra}$ ,  $^{210}\text{Pb}$  and

$^{210}\text{Po}$  in most food subcategories and, where appropriate, individual foods. For datasets for which it has been decided that detailed statistical analysis is not appropriate, only the median value of the dataset is reported.

The available measurement data for human-made radionuclides such as  $^{137}\text{Cs}$  and  $^{90}\text{Sr}$  are unduly influenced by monitoring programmes undertaken in the aftermath of the accidents at the Chernobyl NPP in 1986 and the Fukushima Daiichi NPP in 2011. An added issue is that many analyses are undertaken for the purpose of ensuring compliance with regulatory limits. Consequently, results are often reported as being below the limit of detection. These two factors mean that it has not been possible to determine 95th percentile values for the activity concentration of these radionuclides in food subcategories.

Limited monitoring data exist for  $^{14}\text{C}$  in foods and are primarily gathered through environmental monitoring programmes at or close to nuclear facilities that discharge  $^{14}\text{C}$  to the environment. Approximately 80% of  $^{14}\text{C}$  dose typically arises from  $^{14}\text{C}$  produced in the atmosphere as a result of cosmic ray interactions, and the remaining 20% originates from human-made sources, primarily discharges from nuclear facilities.

## 6.6. DIETARY STUDIES

A number of different dietary survey approaches have been used in national programmes. These are TDSs, MBSs, DDSs and CMSs. Each of these approaches has advantages and disadvantages that need to be considered in order to determine the appropriate approach for any given population or region. A key factor is the resources available to conduct such a survey.

The advantages of TDSs and MBSs are that they provide information on the average intake of radionuclides in the diet and the dose contribution from individual foods or food groups over the entire population. However, they are generally not used for specific population groups or individuals because of the resources required.

The DDS and CMS approaches are typically used for individuals and targeted population groups and require fewer resources than TDSs and MBSs because of their smaller scale. These surveys involve the food being analysed in a 'table ready' or 'as consumed' state; consumption rate data are not needed. As these studies are normally conducted over a relatively short time period and target specific groups, they are not suitable for determining long term consumption or exposure patterns for the whole population.

## 6.7. KNOWLEDGE GAPS

While much information has been published in recent years on the radiation doses received by people who consume forest foods, the focus has been on those areas of the world directly affected by past nuclear accidents. Much less information is available on the radiation doses received by indigenous populations in other parts of the world, many of whom also source their food from the natural and seminatural environments.

The health benefits of seaweed are widely recognized. While the current worldwide consumption rate is low, this is increasing, and an extensive aquaculture industry is in operation. Seaweed and seaweed extracts are also used in multiple and diverse industries. While there is no indication that radionuclides have accumulated to high concentrations in seaweed or in its extracts, it would be wise to keep this under review as the industry expands and diversifies.

Given the increasing importance of the aquaculture industry and the fact that seafood is an important source of radiation dose from the diet, the lack of data on the transfer of both natural and human-made radionuclides from fish feed to fishery products needs to be addressed. One issue is to better document the activity concentration of  $^{210}\text{Po}$  in farmed fish that are artificially fed compared with those that are not fed or are caught in the wild. Ideally, the activity concentration in the same species coming from the three different production pathways would be compared. Another issue is the development of guidelines or standards for the radionuclide content of fish feed to ensure that national standards for the same radionuclides in food can be met.

Carbon-14 is a relatively significant contributor to ingestion dose compared to human-made radionuclides such as  $^{90}\text{Sr}$  and  $^{137}\text{Cs}$ . Therefore, further information and measurement data on the levels of  $^{14}\text{C}$  in foods ingested would be useful, particularly where discharges of  $^{14}\text{C}$  into the environment are known to exist and can contribute to ingestion dose through uptake in the food chain.





## Appendix

### LITERATURE REVIEW AND DATA COMPILATION

Specific activity concentration data for naturally occurring radionuclides (i.e. those of the uranium and thorium decay series) in food were collected using two approaches:

- (a) A literature review of scholarly publications such as manuscripts in scientific journals that report measurements of natural radioactivity in food;
- (b) Requests to all IAEA Member States for data on natural radioactivity in food that might be collected as part of their monitoring programmes.

These data were collated as two separate datasets: one for the literature review and one relating to data provided by Member States. These datasets were reviewed separately and, where appropriate, data for individual radionuclides from each database were merged to perform statistical analysis on naturally occurring radionuclides in foods. This appendix outlines the steps taken to collate the data and describes the information contained within the datasets that were used in statistical analysis.

#### I.1. LITERATURE REVIEW

The IAEA Library supports research activities by providing a specialized collection of nuclear information on the peaceful uses of nuclear energy and its applications. Library services include the provision of up to date and reliable print and electronic information resources plus expert research and personalized information services to library users. A literature search was conducted using these expert services to identify as many appropriate publications as possible between 1998 and 2017. The search terms chosen were intended to find publications reporting radioactivity in food measurements for the following radionuclides:  $^{238}\text{U}$ ,  $^{235}\text{U}$ ,  $^{228}\text{Th}$ ,  $^{230}\text{Th}$ ,  $^{232}\text{Th}$ ,  $^{226}\text{Ra}$ ,  $^{228}\text{Ra}$ ,  $^{210}\text{Pb}$  and  $^{210}\text{Po}$  (i.e. those included in the 2000 UNSCEAR report [16]).

All aspects of the literature review were discussed and agreed in advance with the FAO, UNSCEAR and WHO.

The data search included the use of electronic information resources to search for information in the following publications:

- Peer reviewed journals;
- Reports issued by national/regional organizations or government departments/ministries;
- Reports issued by intergovernmental organizations (e.g. the EC);
- University dissertations (e.g. doctoral theses).

Manuscripts identified by the search were reviewed to ensure that the data were original measurements and not a review of previously published data.<sup>20</sup>

Additional checks were carried out to determine the quality and usefulness of the data. The quality assurance checks were as follows:

- (a) Is it clear that the data are reported on a wet or dry weight basis?
- (b) Does the publication contain information about quality control procedures that are in operation in the measurement laboratory?
- (c) Was the food measured destined for human consumption (i.e. not a laboratory or field experiment)?
- (d) Can the data be extracted? (For example, if the data are only in graph format they cannot be used; data available in text or tabulated form can be used.)
- (e) Have the data been captured only once? (For example, multiple papers could be published from the same dataset; these data would be included only once.)

Publications that did not meet these criteria were excluded from the review. The total number of publications included in the review was 326.

Quality checks were conducted at each stage of the selection process to ensure the appropriate application of the acceptance and rejection criteria. The quality checks were conducted internally and by an independent external auditor, who is an expert in this scientific field. These audits were fully documented. Any queries that arose related to the inclusion or exclusion of publications or data as a result of the internal or external audits were resolved and documented by a third party. The accepted data were compiled into a 'published food data' spreadsheet, with each measurement assigned a unique key, along with a primary key associated with the relevant publication.

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<sup>20</sup> Some additional relevant publications were identified by reviewing the listed references in review papers.

## I.2. DATA PROVIDED BY IAEA MEMBER STATES

An official letter was sent to all IAEA Member States requesting monitoring data on natural radioactivity in food to supplement the data already collated as part of the literature review. Data for natural radioactivity in food monitoring between 1998 and 2017 were requested to ensure that the data covered the same time period as those collected as part of the literature review.

Each Member State was provided with a formatted spreadsheet that included a set of guidelines to assist in the compilation of natural radioactivity in food data. Each Member State was also asked to nominate a national contact person to assist with any follow-up inquiries.

Twenty-eight Member States provided data, with a further six replying that they had no relevant data.<sup>21</sup> Following review by the IAEA, further correspondence with the nominated contact person was necessary to clarify the information provided, for example where data appeared to be incomplete or questionable.

These data were then compared to data compiled as part of the literature review in order to avoid overlap or duplication of radioactivity in food measurements. Once these checks were completed, all the individual Member State data were compiled into a 'national food data' spreadsheet, with each measurement assigned a unique key.

In cases where duplication of data with the published food data spreadsheet was identified and confirmed, the information was excluded from the national food data spreadsheet and retained in the published food data spreadsheet.

## I.3. DATASETS OF ACTIVITY CONCENTRATIONS IN FOOD

The published food dataset and national food dataset were developed as two separate spreadsheets. Statistical analyses were used to check whether it was appropriate to combine radionuclide data from each dataset into one. There could be bias in each dataset; for example, a routine monitoring programme might be more likely to report low or 'not detected' activity concentrations,

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<sup>21</sup> Data were provided by Australia, Bahrain, Belgium, Bosnia and Herzegovina, Brazil, Croatia, the Czech Republic, Egypt, France, Germany, Ghana, Hungary, Indonesia, Ireland, the Islamic Republic of Iran, Japan, the Republic of Korea, the Netherlands, New Zealand, Norway, Romania, the Russian Federation, Slovenia, Switzerland, the Syrian Arab Republic, the United Arab Emirates, the United Kingdom and the United States of America.

whereas information in peer reviewed journal articles might tend to report unusually high activity concentrations because these would be of interest to the research community. If this were so, it would skew the data and would give a bias to the published food dataset, indicating that merging the two datasets might not be possible or would be complicated by the need to employ weighting methods to adjust the data accordingly. Fortunately, this was not found to be an issue in practice.

Where there were sufficient datapoints for analysis, it was generally found that data from each of the two datasets could be combined. Statistical analysis showed that the published food dataset and the national food dataset seemed to be equally representative samples of the radionuclide concentrations found in food in general. Nevertheless, an indication of the dataset origin was included in the statistical analysis of the combined datapoints when determining statistics for a number of food category–radionuclide combinations.

The approach of combining data from each dataset permitted the appropriate statistical analysis of a number of merged datasets for key radionuclide–food combinations. For publications or Member States who reported activity in dry weight rather than in wet (or fresh) weight, a conversion factor was applied to convert the values to fresh whenever possible. The fresh weight activity calculation was derived using the following formula:

$$\text{Fresh weight activity concentration} = \text{dry activity concentration} \times \text{fraction of dry weight}$$

To determine the appropriate conversion factor, the publication or information from the Member State was reviewed, and if no value was provided in the paper, then other sources were consulted to find a representative factor.

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## **Annex I**

### **ANALYTICAL TECHNIQUES TO IDENTIFY AND QUANTIFY RADIONUCLIDES IN FOOD**

#### **I-1. INTRODUCTION**

Various analytical techniques can be used to identify and quantify radionuclides in food and drinking water. The analytical technique used in any analysis is dependent upon a number of factors, including the following:

- The radionuclide or radionuclides of interest — consideration needs to be given to the type of radioactive decay, whether it is alpha, beta or gamma, and the half-life of the radionuclide(s) of interest.
- The levels of radioactivity present — the abundance of the radionuclides present will have an impact on the type of procedure used. The procedure will also need to have appropriate sensitivity and limits of detection to detect the levels of radioactivity present. Interfering radionuclides may have to be removed before measurement.
- The resources available to conduct the analysis — including the equipment, consumables, radioactive tracers and carriers for radiochemical separations, radiometric measurement equipment, time needed for analysis and human resources available.
- The robustness of the technique — all of the analytical techniques used need to be validated to ensure that the technique can stand up to scrutiny and the results and associated uncertainties are accurate.
- Quality assurance and control — the laboratory undertaking these techniques needs to have an adequate quality assurance system and techniques to ensure ongoing proficiency of the measurements through quality control checks and participation in interlaboratory comparisons and proficiency tests.

This annex summarizes the various analytical techniques that can be used in determining the activity concentration of radionuclides in food and drinking water samples on the basis of current international best practice. The methods described can measure the radionuclides of interest to activity concentrations that are typically found in food and drinking water samples. Additional information on each of the procedures outlined is available in the references.

## I-2. SCREENING METHODS FOR RADIONUCLIDES IN WATER

Screening methods for radioactivity in drinking water are based on the gross alpha and gross beta activity measurements in samples [I-1 to I-3]. Gross alpha and gross beta measurements are robust and simple and save time and resources for initial screening of radioactivity in drinking water. However, as this is only a screening method, there are limitations to this approach. Principally, screening cannot accurately quantify the specific radionuclides in drinking water samples and the gross alpha and beta activities determined are relative to the standards used when calibrating the screening techniques. WHO has set screening levels of 500 mBq/L for gross alpha activity and 1000 Bq/L for gross beta activity. If either of these screening levels is exceeded, then analysis of individual radionuclides is recommended. In these cases, where accurate quantification of radionuclides in drinking water samples is necessary, the specific analytical techniques outlined in Section I-4 of this annex could be used.

Numerous screening methods have been developed for gross alpha and gross beta activity in water samples [I-4 to I-6]. The two most common approaches are:

- Gross alpha and gross beta screening using liquid scintillation counting (LSC);
- Gross alpha and gross beta screening by a combination of evaporation and analysis using gas proportional counting (GPC).

### I-2.1. Gross alpha and gross beta screening using LSC

The measurement principles for LSC screening are as follows:

- (a) Acidification of the sample;
- (b) Thermal preconcentration;
- (c) Mixing the concentrated sample with an appropriate scintillant;
- (d) Analysis of the sample on an LSC with alpha–beta discrimination capabilities.

In most cases, drinking water samples do not contain excessive amounts of suspended particulate matter, as they would be unpalatable for human consumption [I-7]. Therefore, filtration of drinking water samples is typically not necessary prior to screening. Drinking water samples can be taken from the distribution point, such as a tap, and need to be acidified using nitric acid to prevent adsorption of the radionuclides present onto the sampling container.

The sample can be evaporated slowly on a hotplate at approximately 80°C to concentrate the sample tenfold, thus improving the sensitivity of the measurement. The range of total dissolved solids in water samples is typically less than 600 mg/L and thermal preconcentration is not an issue. However, if a precipitate appears during this concentration step, the concentration factor will need to be reduced to prevent the formation of salts.

An aliquot of the concentrated sample can then be mixed with a commercially available scintillant in a glass or polyethylene scintillation vial. The scintillant chosen needs to be suitable for alpha–beta separation. The sample to scintillant ratio is normally an 8:12 volume; for example, in a 20 ml scintillation vial there is 8 ml of sample and 12 ml of scintillant.

The samples can be counted on a low level LSC that has the necessary alpha–beta discrimination capabilities and has been calibrated using appropriate certified alpha and beta emitting radioactive standards. These certified standards can also be used to optimize the alpha and beta discrimination setting on the LSC. The choice of the alpha and beta emitting standards used for calibration is important, as the screening method is relative to the standards used. If the screening technique is primarily being used for screening natural radioactivity, then the alpha and beta standards chosen also need to be naturally occurring radionuclides. For example,  $^{238}\text{U}$  or  $^{230}\text{Th}$  as an alpha source and  $^{40}\text{K}$  as a beta source. For artificial radionuclides,  $^{241}\text{Am}$  or  $^{239}\text{Pu}$  can be used as an alpha source and  $^{90}\text{Sr}/^{90}\text{Y}$  as a beta source.

To avoid any issues arising as a result of chemical quench in the samples, the method can be optimized to ensure that all samples and calibration standards are at a constant quenching level. For example, the International Organization for Standardization (ISO) has taken this approach by ensuring that samples are standardized to a constant pH of 2.7 upon sampling, and so the pH is estimated to reduce to pH 1.7 after thermal preconcentration [I–5]. Minimal adjustments can be made to the sample pH at this stage to ensure that the pH is at 1.7, which is the same pH as the certified reference solutions used for calibration.

## **I–2.2. Gross alpha and gross beta screening using evaporation and GPC**

To screen drinking water samples using GPC, a sample is slowly evaporated onto a stainless steel planchette using a hotplate or infrared lamp. The sample residue is then measured using an alpha–beta GPC.

The volume of sample needed for this analysis is dependent upon the amount of residue after sample evaporation. The volume needs to be calculated to ensure that the mass of sample residue evaporated onto the planchette is between 50 mg and 100 mg. The sample is acidified, using nitric acid to prevent the adsorption of radionuclides onto the walls of the sampling container, but

this needs to be kept to a minimum, as the addition of acid can increase the salt content of the sample residue after sample evaporation. Normally, the sample is acidified using 10 ml of concentrated nitric acid per litre of sample.

Evaporation of the sample can be carried out by placing the planchette on a hotplate or under an infrared lamp. The temperature during evaporation is kept at or below 80°C to prevent spitting of the sample and the loss of volatile radionuclides such as  $^{210}\text{Po}$  that could be present. The sample is gradually transferred to the planchette using a pipette to ensure that the residue is distributed uniformly over the entire surface and is as thin as possible. This is to ensure that the self-absorption of the alpha and beta particles is minimized.

The counting efficiencies are dependent on the amount of self-absorption of the alpha and beta particles. The problem of self-absorption is much greater in the case of alpha activity because of the greater degree of interaction between alpha particles and matter. Therefore, it is necessary to determine the alpha and beta counting efficiencies as a function of residue mass.

The alpha and beta counting efficiencies can be determined for drinking water samples containing a range of residues and a self-absorption efficiency curve can be derived, plotting the variability of alpha and beta counting efficiencies as a function of residue mass. Similar to the LSC method outlined in Section I-2.1, the standards chosen for calibration are dependent upon the type of radioactivity likely to be present in the drinking water samples of interest, that is, whether the radionuclides of interest are primarily naturally occurring or human-made. The types of alpha standards used can include  $^{241}\text{Am}$  and  $^{239}\text{Pu}$  for human-made radioactivity and uranium for naturally occurring radionuclides. Beta standards can include  $^{40}\text{K}$  for naturally occurring radioactivity and  $^{90}\text{Sr}/^{90}\text{Y}$  or  $^{137}\text{Cs}$  for human-made radioactivity.

To determine a self-absorption efficiency curve, a set of calibration sources with the same activity but increasing residue mass are prepared. The residue mass can be varied through the inclusion of stable salts such as sodium carbonate ( $\text{Na}_2\text{CO}_3$ ) and is typically varied between 50–100 mg. For example, to determine the self-absorption efficiency curve for an alpha standard, 10 purified water samples could be prepared using increasing amounts of  $\text{Na}_2\text{CO}_3$  solution and spiked with the same amount of  $^{241}\text{Am}$  standard solution. The samples are prepared in the same manner as outlined above, and a function relating the calibration efficiency to the residue mass can be derived.

Analysis of planchettes is always to be conducted as soon as is practicable after the evaporation of the drinking water sample because:

- Humidity can lead to the absorption of moisture in the atmosphere where the planchettes are being stored. This leads to an increase in mass on the planchette that can impact on the counting efficiency. Absorption affects

can be minimized through the storage of planchettes in a desiccator prior to counting.

— There can be ingrowth of radon progeny over time.

Following the evaporation of the sample, the planchette is counted using a GPC that has been calibrated to ensure that the alpha and beta crosstalk on the instrument has been minimized to reduce misclassification.

Both of the LSC and GPC measurement techniques described have similar limits of detection. Given a sample count time of approximately 12 h, detection limits between 5–10 mBq/L are attainable. However, the LSC method does have numerous advantages over the GPC approach. Sample preparation for GPC is more time consuming and less straightforward than for the LSC approach. The evaporation of the water samples onto a planchette can lead to further losses of volatile radionuclides such as  $^{210}\text{Po}$ , and GPC techniques cannot measure low level beta emitters ( $<0.1$  MeV) such as  $^{210}\text{Pb}$  and  $^{228}\text{Ra}$ . Therefore, the LSC approach is increasingly becoming the preferred technique for gross alpha–beta screening. In addition, the LSC approach can handle water samples with a greater amount of total dissolved solids.

These screening techniques can be used only for the screening of gross alpha and beta activities in water samples and are not suitable for use for the analysis of food samples. Analysis of food samples using the GPC approach would not be practical given the interferences that would arise due to the presence of organic matter in samples. There are also difficulties in attaining a thin source on the planchette, leading to issues with self-absorption in the sample residue. The measurement of solid samples using LSC is not suitable, as this can lead to significant issues with colour and chemical quenching in LSC samples. Radioanalytical techniques that could be used to purify food samples prior to such screening tend to have a high degree of specificity for particular radionuclides that could lead to the loss of a significant proportion of the radioactivity present before analysis.

### I-3. METHODS FOR SPECIFIC RADIONUCLIDES

Some of the analytical techniques outlined in the subsequent sections can be used to measure one or more specific radionuclides, depending on their properties. The analysis of food and drinking water samples by gamma spectrometry can measure the activity concentrations of a number of gamma emitting radionuclides concurrently. The analysis of alpha and beta emitting radionuclides is, in general, more complex, involving a number of separation and purification steps prior to analysis, and these will be dealt with individually.

### **I-3.1. General requirements for analytical techniques**

#### *I-3.1.1. Sample collection*

Food samples collected for radioactivity analysis need to be representative of the food of interest. This may involve the collection of a number of samples to take into consideration variability as a result of geographical or temporal trends. In addition, sample size is also an important factor to consider, as this has an influence on the limits of detection for radioanalytical techniques. The sample size needed is evaluated prior to collection to ensure that the radioanalytical techniques employed can reach an appropriate detection limit for the radionuclides of interest. The time interval between sampling and analysis also needs to take into consideration the half-lives of the radionuclides of interest to ensure that they remain above detection limits during this time period.

#### *I-3.1.2. Sample pretreatment*

Sample pretreatment for food and drinking water samples is carried out to obtain a homogeneous sample, preconcentrate radionuclides of interest, remove unwanted residues and organic matter, introduce stable carriers or radioactive yield tracers, if necessary, and prepare the sample for the radiochemical procedures that may follow.

Preliminary sample pretreatment for food samples comprises drying and homogenization that will concentrate the radionuclides present and ensure that any aliquots taken for subsequent analysis are representative of the bulk sample. Ashing can also be carried out but, in these instances, care is needed to ensure that there are no losses of radionuclides of interest because of their volatility at increased temperatures.

Samples can be dried to a constant dry weight in an oven at 80–105°C or using a freeze drier. Once the sample has been dried to a constant weight the dry sample can be ground using a pestle and mortar or laboratory homogenization equipment (such as a food blender, food processor, homogenizer or rotor mill) and passed through a laboratory test sieve (to prevent clumping of the sample). The drying and homogenization of food samples is advisable prior to any analysis or radiochemical technique, as this process provides a uniform sample matrix and acts as concentration step that increases the measurement sensitivity.

Drinking water samples are acidified at the time of sampling to prevent loss of radionuclides due to plate-out on the walls of the sampling container and to prevent the buildup of biological material. This can be achieved by keeping the sample at a pH of 6–8, which typically equates to 10 ml of concentrated HCl per 1 L of sample [I-8]. Water samples can be evaporated to preconcentrate the

radionuclides of interest, and evaporation can be conducted using a hotplate or infrared (evaporation) lamps, but care is needed to avoid loss of the sample through spattering.

Samples can be further concentrated after drying through dry ashing in a laboratory furnace. This step also removes any organic matter in the sample that could interfere in radiochemical separations [I-9]. Dry ashing normally takes place over a period of 24–72 h, where the sample is ashed in stages of increasing temperature from room temperature to 450°C. The ashing time is dependent upon the quantity and type of food. For example, 1 L of milk can be ashed over a period of approximately 72 h by placing the sample in the furnace at room temperature and increasing the temperature steadily by 0.5°C/min until it reaches 150°C. The temperature stays at 150°C for 2 h before being increased to 450°C at 1°C/min until it reaches a final temperature of 450°C, where it remains for 18 h before cooling.

Food samples require digestion prior to radiochemical separation techniques. This is to ensure that samples are completely free of organic matter and insoluble residues that can interfere with radiochemical separation. Following ashing, samples are dissolved by wet digestion on a hotplate using nitric (HNO<sub>3</sub>) and hydrochloric (HCl) acids with a hydrogen peroxide (H<sub>2</sub>O<sub>2</sub>) catalyst. In most cases, this digestion procedure is sufficient for complete dissolution of food samples. Other digestion techniques, such as the use of hydrofluoric acid and microwave digestion [I-10], have also been used in the digestion of samples, but these would be necessary only if the sample contained silicates or refractory particles, which is not the case in food samples. Fusion techniques have also been used for other environmental matrices, but these are not suitable for food samples because of the small sample size capable of being treated using this approach [I-11].

The chemical yield of any radiochemical separation technique is determined by the addition of a stable carrier or radioactive tracer to the original sample before analysis. A stable isotopic carrier similar to the radioisotope being determined can be added to the sample before radiochemical separation, and the amount of carrier retained at the end of the analysis can be determined gravimetrically or by mass spectrometry. This approach can be seen in analyses of strontium that use stable strontium nitrate or yttrium nitrate carriers and in analyses of radium that can use the chemically analogous but stable barium carrier. Radioactive tracers are used in a similar manner. This is most common in the determination of the yield in alpha spectrometry measurements. In these cases, the yield from the analysis can be determined through the addition of another alpha emitting isotope of the same element. For example, <sup>209</sup>Po is used as the yield tracer for <sup>210</sup>Po analysis (Section I-1.1.4).

### *I-3.1.3. Radiochemical separation*

Radiochemical separation is typically not necessary for gamma emitting radionuclides. Radionuclides that are pure alpha or beta emitters in most cases need radiochemical separation before measurement. Alpha emitting radionuclides have to be separated radiochemically from those with similar alpha energies to avoid overlapping peaks in measurement spectra. Similarly, beta particles produce much broader or continuous spectra and can only be measured after separation from other beta emitting radionuclides. Radionuclides behave similarly to their stable isotopes, and so radiochemical separations are based on the same traditional methods of chemical separation, namely precipitation, ion exchange chromatography and solvent extraction. More recently, developments in the field of radiochemical separation have led to the combining of elements of ion exchange and solvent extraction, producing commercially available extraction chromatography resins that have proven very effective in the separation of lanthanides and actinides.

### *I-3.1.4. Source preparation for alpha spectrometry*

There are two methods of source preparation for alpha spectrometric measurement: electrodeposition and microprecipitation. Electrodeposition provides better energy resolution, but the process takes longer. Microprecipitation onto filter paper is a rapid method but requires the use of hydrogen fluoride, which is a limiting factor in some laboratories. There can also be deposition issues, as the layer may be too thick on the filter paper. There have been studies in which the efficiency of both techniques has been evaluated [I-12].

For electrodeposition, an electrodeposition cell is needed to carry out the deposition. Very few commercial options are available. It is common in laboratories to design custom made cells with liquid scintillation vials or to order specially made cells composed of either polytetrafluoroethylene (PTFE) or polyvinylidene fluoride (PVDF; TECAFLON). An example of a cell design is described in the literature [I-13, I-14]. Along with the cell, an anode, a stainless steel disc for deposition and a cathode are needed. The anode is a thin platinum wire, and the cathode is a stainless steel-INOX base. A power supply is used to provide a current to allow the deposition to take place. The equipment comes at an initial high cost, but the equipment is reusable.

For microprecipitation, isotopes can be precipitated using cerium fluoride or neodymium fluoride. The sample solution is mixed with chemicals and is vacuum filtered through a cellulose membrane fibre with a pore diameter of 0.1  $\mu\text{m}$ . The filter paper is dried by an infrared lamp and can be mounted on an aluminium or stainless steel disc ready for alpha spectrometry analysis.



### **I-3.2. Direct measurement of gamma emitting radionuclides**

In the past, gamma spectrometry systems were based on the use of scintillation detectors, principally sodium iodide and, to a lesser extent, caesium iodide and bismuth germanate detectors. However, these have largely been replaced with semiconductor detection systems. These initially consisted of lithium drifted germanium and lithium drifted silicon detectors. The more technologically advanced detectors are now based on the use of high purity germanium (HPGe). The HPGe systems have a much higher energy resolution and a much lower background count rate when compared to sodium iodide systems and are the preferred method for the measurement of low level environmental radioactivity in a laboratory.

For HPGe gamma spectrometry systems to function correctly, they have to be kept at a low temperature (approximately  $-190^{\circ}\text{C}$ ), and this requires constant cooling. The most cost effective and convenient approach to cooling the HPGe detectors is with liquid nitrogen. More recently, electronic cooling systems have been developed for use in situations where a constant supply of liquid nitrogen is not available. Sodium iodide detectors are cheaper and have a higher detector efficiency than HPGe detectors. Furthermore, they can operate effectively at room temperature (as long as the temperature remains stable). However, their poor energy resolution and high background count rate can make the measurement of low level environmental samples more difficult.

The analysis of food and drinking water samples for gamma emitting radionuclides does not involve extensive preparation or radiochemical separation. However, food and drinking water samples can be preconcentrated via drying, ashing or evaporation, if necessary. The gamma emitting radionuclides of interest need to be distributed uniformly throughout the sample matrix and be representative of the bulk sample. This can be achieved through homogenization of the sample for analysis.

Samples to be analysed as part of dietary intake studies are prepared in the normal manner for consumption, and only the edible parts of the food are analysed. Food samples are dried and homogenized for analysis in the manner outlined above (Section I-3.1) and placed in an appropriate container, such as a Marinelli beaker, for analysis. Drinking water samples can be poured directly into the analysis container after sample pretreatment (i.e. acidification).

#### *I-3.2.1. Radiocaesium*

Radiocaesium ( $^{134}\text{Cs}$  and  $^{137}\text{Cs}$ ) can be measured readily using a properly calibrated gamma spectrometry system. Calibration of these systems can be conducted experimentally using standard sources, numerically using computer

codes and semiempirically using a combination of standard sources and computer codes [I–15].

Determination of the activity concentration of the radionuclides in a sample is dependent upon the detector efficiency, energy resolution, sample count time, sample size, self-attenuation and coincidence summing effects. Each of these factors can have an impact on the sensitivity and minimum detectable activity of a measurement. Both sample size and detector efficiencies can be maximized using large volume Marinelli beakers (0.5–2 kg) that are tailored to the dimensions of the gamma detector. These Marinelli beakers enable the gamma rays from the sample to interact with the top and sides of the detector’s active area, thus increasing overall efficiency.

Self-attenuation occurs when gamma photons are absorbed by the sample matrix and cannot reach the detector and is dependent upon the density and composition of the sample matrix. Coincidence summing occurs as a result of radionuclides emitting at least two gamma photons concurrently. The effects of coincidence summing increase with gamma detector efficiency and decrease with source–detector distance. To minimize coincidence summing effects, samples can be analysed further away from the detector. The most common approaches to correct for attenuation and coincidence summing effects are dedicated software packages, Monte Carlo codes and empirical equations used in conjunction with transition measurements of varying sample matrix densities.

For radiocaesium, the activity concentrations of <sup>137</sup>Cs and <sup>134</sup>Cs can be determined through measurements of the principal gamma emission lines from both isotopes that range from 563 keV to 802 keV, as outlined in Table I–1.

TABLE I–1. RADIOCAESIUM GAMMA EMISSION ENERGIES (keV) AND INTENSITIES (%) [I–16]

<sup>134</sup> Cs (T <sub>1/2</sub> = 2.064 years)		<sup>137</sup> Cs (T <sub>1/2</sub> = 30.05 years)	
Energy (keV)	Intensity (%)	Energy (keV)	Intensity (%)
563.246	8.342		
569.330	15.368		
604.720	97.63	661.657	84.99
795.86	85.47		
801.95	8.694		

Caesium-137 has only one significant gamma transition at 662 keV, whereas  $^{134}\text{Cs}$  has many, with the most intensive being at 605 keV and 796 keV. Given the relatively complex decay scheme of  $^{134}\text{Cs}$ , quantification of this radionuclide is susceptible to coincidence summing effects, and corrections for these effects are necessary and can be conducted using the methods outlined above.

#### *I-3.2.2. Other radionuclides*

Gamma spectrometry systems are typically calibrated using commercially available calibration sources containing gamma emitting radionuclides that cover an energy range from approximately 40 keV to 2000 keV. Therefore, they have the potential to measure radionuclides other than radiocaesium, including  $^{210}\text{Pb}$ ,  $^{241}\text{Am}$ ,  $^{226}\text{Ra}$  progeny and  $^{228}\text{Ra}$  progeny.

Lead-210 produces a 46.5 keV gamma photon through internal conversion when decaying to  $^{210}\text{Bi}$ . This is a relatively low gamma energy with a low abundance of 4.6% and is difficult to detect in food and water samples due to:

- Self-attenuation in the sample;
- The relatively low counting efficiency of ordinary HPGe detectors at energies below 100 keV;
- The interference of background counts at low energies as a result of Compton scattering;
- The relatively low activity concentrations of  $^{210}\text{Pb}$  found in typical food and water samples, ranging from 10–100 mBq/kg.

Therefore, measurement of  $^{210}\text{Pb}$  using gamma spectrometry for food and water samples is not appropriate.

Americium-241 produces a 59 keV gamma photon with an emission probability of 36% and can be used to measure  $^{241}\text{Am}$  directly from some high activity samples, but normally this cannot be used to determine activity concentrations of  $^{241}\text{Am}$  in food or drinking water samples where activity concentrations are expected to be significantly lower. The primary decay mode for  $^{241}\text{Am}$  is alpha decay. Therefore,  $^{241}\text{Am}$  is typically determined via alpha spectrometry after the appropriate radiochemical separation techniques (Section I-4.1.3).

The naturally occurring radionuclides  $^{226}\text{Ra}$  and  $^{228}\text{Ra}$  can also be measured via gamma spectrometry using their short lived progeny if they are assumed to be in secular equilibrium.  $^{226}\text{Ra}$  can be measured using the mean value of the  $^{214}\text{Pb}$  (352 keV) and  $^{214}\text{Bi}$  (609 keV and 1765 keV) gamma peaks after an ingrowth period of 28 days if the sample has been hermetically sealed during this time to

prevent the escape of radon gas. Radium-226 can also be calculated in the same manner using the mean values of the  $^{228}\text{Ac}$  (911 keV) and  $^{212}\text{Pb}$  (239 keV) peaks as the ingrowth period for Radium-228 progeny is adequately covered by the  $^{226}\text{Ra}$  ingrowth.

However, as with  $^{241}\text{Am}$  and  $^{210}\text{Pb}$ , this measurement technique can be utilized only for direct measurement of samples with relatively high  $^{226}\text{Ra}$  and  $^{228}\text{Ra}$  activity concentrations, such as soil or other industrial waste products (naturally occurring radioactive material) where activity concentrations for  $^{226}\text{Ra}$  and  $^{228}\text{Ra}$  are in the tens to hundreds of Bq/kg [I-17]. The activity concentrations of  $^{226}\text{Ra}$  and  $^{228}\text{Ra}$  contained in food and drinking water are typically three orders of magnitude lower than those found in samples of soil or naturally occurring radioactive material [I-17] and will need preconcentration and radiochemical separation prior to analysis in order to determine their activity concentrations (Section I-4.1.8).

The assessment of  $^{40}\text{K}$  in food and drinking water samples is not a requirement for dose assessments because the dose contribution from this radionuclide remains constant in healthy humans regardless of the amount ingested, as the levels of potassium, and hence  $^{40}\text{K}$ , in the body are controlled by metabolic processes in the human body. However,  $^{40}\text{K}$  can serve as a quality control check on food samples being measured by gamma spectrometry. Potassium-40 can be measured using the 1460.55 keV peak. The levels of  $^{40}\text{K}$  in different foodstuffs can vary, but the range of activities within these food groups is relatively small. Typical  $^{40}\text{K}$  activity concentrations can be seen in Table I-2.

## I-4. RADIOCHEMICAL TECHNIQUES

### I-4.1.1. *Strontium-90*

Strontium-90 decays by beta emission to  $^{90}\text{Y}$ , which has a half-life of 64 h and a very high maximum beta energy of 2.27 MeV, which increases its radiotoxicity. The properties of these radionuclides are outlined in Table I-3.<sup>1</sup>

Strontium-90 and  $^{90}\text{Y}$  exist in secular equilibrium in environmental samples and, since both are beta emitters, radiochemical separation is necessary before measurement. Strontium-90 and  $^{90}\text{Y}$  are measured by either LSC or GPC. The

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<sup>1</sup> Other radioactive isotopes of Sr and Y, such as  $^{89}\text{Sr}$  and  $^{91}\text{Y}$ , are not expected to be present in any environmental samples in non-emergency situations, and therefore potential interferences from these isotopes are not addressed.

TABLE I-2. TYPICAL CONCENTRATIONS OF <sup>40</sup>K IN FOOD [I-18]

Food	Typical concentration (Bq/kg)
Lettuce	20-40
Milk	40-60
Tomatoes	60-80
Bananas	80-100
Chicken	80-100
Potatoes	140-180
Cod	140-170
Beef	150-200
Wheatgerm	300-350

TABLE I-3. PROPERTIES OF <sup>90</sup>Sr AND <sup>90</sup>Y ISOTOPES [I-16]

Nuclide	T <sub>1/2</sub>	Principle decay mode	Maximum beta energy (MeV)
Sr-90	29.9 years	$\beta$	0.54
Y-90	2.67 days	$\beta$	2.27

relatively high beta energies of <sup>90</sup>Sr and <sup>90</sup>Y ensure that measurement using either of these approaches is relatively straightforward after radiochemical separation.

The yield tracers used for <sup>90</sup>Sr analysis can be stable Sr or Y carriers, such as strontium nitrate and yttrium nitrate, which are added at the beginning of the analysis, and chemical yield can be determined by the amount of Sr or Y in the final sample solution after separation. The amount of stable tracer can be determined gravimetrically or using mass spectrometry. Another yield tracer that can be used is <sup>85</sup>Sr (T<sub>1/2</sub> = 65 days, E<sub>gamma</sub> = 514 keV (96%)), a gamma emitting Sr isotope that can be measured in the sample solution after radiochemical separation using gamma spectrometry.

Radiochemical separation of  $^{90}\text{Sr}$  or  $^{90}\text{Y}$  for environmental samples can be conducted through a combination of selective precipitations, extraction chromatography and solvent extraction techniques [I-19]. One major challenge in radiochemical separation of  $^{90}\text{Sr}$  from samples is the adequate separation of strontium from calcium. Calcium is an abundant element in most samples, chemically the most equivalent to strontium, and can interfere with the separation of strontium. It is also important to remove alkaline earth metals such as potassium during radiochemical extraction, as the presence of  $^{40}\text{K}$  in samples can interfere with measurement of  $^{90}\text{Sr}$  using LSC or GPC.

Sample pretreatment for analysis of food samples for  $^{90}\text{Sr}$  is as outlined in Section I-3.1.1. After ashing, the sample is dried to a residue on a hotplate awaiting radiochemical extraction.

The  $^{90}\text{Sr}$  activity concentrations in water samples are typically very low, and therefore large sample volumes of at least 10 L are needed to reach suitable detection limits. Preconcentration of the water samples can be achieved by evaporation or by a carbonate precipitation [I-20, I-21].

The traditional approach for radiochemical extraction of Sr from samples is selective precipitation, where a series of precipitations to remove interfering radionuclides, earth metals and alkali metals are conducted. The first step in this process is the removal of calcium from the sample by a nitrate precipitation. Standard procedures, conducted routinely in the past but now replaced by newer techniques, applied concentrated fuming  $\text{HNO}_3$ . Using this approach, 100%  $\text{HNO}_3$  is added to the sample precipitate until the concentration reaches 70%. When the nitrites reach 70%, the strontium precipitates as strontium nitrate, and the calcium is retained in the solution [I-22]. One-step nitrate precipitation may not be sufficient to remove all of the calcium from the precipitate, and it may have to be repeated two or more times. This method requires a large quantity of chemicals, and there are health and safety implications when using fuming nitric acid.

Alternative approaches have been used that utilize  $\text{NaOH}$  [I-23] or 65%  $\text{HNO}_3$  [I-19]. The precipitate is dissolved in water and a carbonate precipitation is conducted by adding ammonium carbonate to reduce the volume of the sample. The next step is to separate strontium from barium and radium isotopes in the sample by a chromate precipitation. The precipitate is dissolved in a weak acid, and sodium chromate is added and adjusted to pH 4. Barium and radium are removed from solution by the precipitate and another carbonate precipitation is carried out on the solution. This precipitate is again dissolved in a weak acid, iron chloride is added and the solution is adjusted to pH 8 or 9 to precipitate ferric hydroxide. The ferric hydroxide precipitation removes yttrium, thorium, radium progeny and lanthanides from the solution. The final carbonate precipitation is carried out, and the precipitate is dried and weighed to determine the chemical

yield. This precipitate can be counted by GPC or dissolved in a weak acid and mixed with scintillant in the glass vial for counting by LSC.

The extraction chromatography Sr resin can be used to analyse  $^{90}\text{Sr}$  in food and drinking water samples. The separation of strontium using the Sr resin is conducted in an 8 M  $\text{HNO}_3$  solution, where strontium is very tightly bound to the resin, whereas other alkali metals such as calcium and radium are not bound or are very weakly bound and can be removed readily through rinsing with 8 M  $\text{HNO}_3$  [I-24]. Prior to separation of Sr using the Sr resin it will be necessary to preconcentrate strontium and other alkaline earth metals using an oxalic or phosphate precipitation [I-25]. A common approach to preconcentration in food and drinking water samples is calcium phosphate precipitation [I-20, I-26]. Water samples can be preconcentrated to 1 L prior to precipitation. Approximately 20 g of food sample is dried, homogenized and ashed, and a small quantity of concentrated nitric acid is added to digest the organic compounds; 1.25 M calcium nitrate and 3.2 M ammonium hydrogen phosphate are added to the sample, and it is adjusted to pH 9 or 10 using 12 M NaOH to form a calcium phosphate precipitate. The precipitate is then dissolved in concentrated  $\text{HNO}_3$ :1 M  $\text{Al}(\text{NO}_3)_3$  and is ready for loading onto the Sr resin column. The Sr resin column is preconditioned using 8 M  $\text{HNO}_3$ , and the sample is loaded onto the column, retaining strontium and eluting other alkali metals and yttrium. The column is further washed with 8 M  $\text{HNO}_3$  to ensure the complete removal of barium and calcium. If plutonium and neptunium are in the sample, they can be removed from the column through washing with 3 M  $\text{HNO}_3$ ; 0.05 M oxalic acid. Strontium is eluted from the column using 0.05 M  $\text{HNO}_3$ . A portion of the solution can be dried on a planchette and measured by GPC or it can be added to a glass vial with scintillant for analysis by LSC.

Another approach for the analysis of  $^{90}\text{Sr}$  in food or drinking water samples is solvent extraction using the organic solvent di-(2-ethylhexyl)phosphoric acid (HDEHP) to extract the  $^{90}\text{Y}$  from the sample. The yttrium extracted from the sample can be analysed by Cherenkov counting using an LSC. Food and drinking water samples are pretreated as outlined above with the addition of a stable yttrium nitrate carrier solution. After ashing, the samples are digested using concentrated  $\text{HNO}_3$  and dried on a hotplate. The resulting residue is dissolved in 1 M HCl and the solution is adjusted to a pH between 1.1 and 1.2. This solution is placed in a separation funnel with 10% HDEHP in toluene and shaken for 1–2 min. The yttrium is extracted from the solution into the HDEHP solution and separated from interfering elements such as uranium, thorium, radium and their decay products, as well as isotopes of caesium, potassium and strontium. After a weak acid wash using 0.08 M HCl, the yttrium is back extracted into 3 M  $\text{HNO}_3$  by shaking for a further 1–2 min. The 3 M  $\text{HNO}_3$  solution is adjusted to pH 10 through the addition of ammonia solution, and the yttrium will form an yttrium

hydroxide precipitate. The precipitate is dissolved using 1 ml concentrated  $\text{HNO}_3$  and immediately transferred to a liquid scintillation vial and topped up with water for Cherenkov counting using a low level LSC. After measurement on the LSC, the yield of the analysis is determined gravimetrically by acidimetric titration of the vial.

There are several approaches to determining  $^{90}\text{Sr}$  activity using LSC or GPC. If strontium is extracted using selective extraction or extraction chromatography, the  $^{90}\text{Sr}$  present can be determined before interferences from the rapid ingrowth of  $^{90}\text{Y}$ . However, this would be necessary only if a rapid assessment was needed. The best approach would be to wait for the  $^{90}\text{Y}$  in the sample to reach secular equilibrium with the  $^{90}\text{Sr}$  (after approximately three weeks) and measure the total beta spectrum of both, as this method improves the counting efficiency. If  $^{90}\text{Y}$  has been extracted using solvent extraction, the samples need to be counted as soon as possible after radiochemical separation, since the  $^{90}\text{Y}$  is no longer supported by  $^{90}\text{Sr}$  and has a relatively short half-life. The advantage of this approach is the much higher counting efficiency for Cherenkov counting by LSC when compared to standard LSC techniques using scintillant.

All of the above approaches to  $^{90}\text{Sr}$  measurements in food and drinking water samples have limits of detection on the order of mBq/kg. The solvent extraction technique using HDEHP measures  $^{90}\text{Y}$  by Cherenkov counting, whereas the selective precipitation and extraction chromatography methods need to wait for the ingrowth of  $^{90}\text{Y}$  before counting. Therefore, if a much more rapid analytical method is needed, solvent extraction would be the best approach. However, if  $^{90}\text{Sr}$  is being analysed as part of sequential extractions, then extraction chromatography is the ideal approach (see Section I-5).

#### *I-4.1.2. Plutonium*

Plutonium isotopes exist in the environment as a result of nuclear weapons testing, nuclear accidents and discharges from nuclear reprocessing facilities and power plants. The most common Pu isotopes in the environment are outlined in Table I-4, together with their decay properties.

The most straightforward approach for measuring alpha emitting Pu isotopes is via alpha spectrometry, as this technique can reach limits of detection as low as 1 mBq per sample. In addition, the radiochemical extraction and purification steps are less onerous compared to mass spectrometry techniques such as ICP-MS and thermal ionization mass spectrometry, and alpha spectrometry is a relatively low cost procedure, considering the time required to undertake the analysis and the cost of measurement equipment. However, alpha spectrometry is not capable of distinguishing between the alpha peaks of  $^{239}\text{Pu}$  and  $^{240}\text{Pu}$  because



TABLE I-4. PROPERTIES OF PLUTONIUM ISOTOPES [I-16]

Nuclide	T <sub>1/2</sub> (years)	Principle decay mode	Alpha energy and intensity (MeV (%))
Pu-238	87.7	$\alpha$	5.499 (71%) 5.456 (29%)
Pu-239	$2.4 \times 10^4$	$\alpha$	5.157 (71%) 5.144 (15%)
Pu-240	$6.6 \times 10^3$	$\alpha$	5.168 (73%) 5.124 (27%)
Pu-241	14.3	$\beta$	n.a. <sup>a</sup>

<sup>a</sup> n.a.: not applicable.

their alpha energies overlap (Table I-4), and therefore they have to be determined together as <sup>239+240</sup>Pu.

Plutonium-241 is a beta emitter and is measured by LSC. This LSC measurement can be conducted by stripping an alpha source from a disc prepared for alpha spectrometry after analysis [I-27], or an aliquot of the Pu extracted from a sample can be taken for LSC analysis prior to electrodeposition [I-28]. This will not be addressed in detail here, as <sup>241</sup>Pu would not be a significant exposure pathway from ingestion compared to the alpha emitting Pu isotopes. However, the <sup>241</sup>Pu progeny, <sup>241</sup>Am, is of more concern from an exposure perspective, and analysis of this radionuclide is discussed in Section I-4.1.3.

To measure plutonium by alpha spectrometry, the Pu isotopes are first extracted from the sample matrix to remove unwanted metals and other alpha emitting radionuclides to prevent spectral interferences. If radiochemical separation is not fully effective, isotopes of americium, uranium and thorium and their progeny can contaminate the alpha source, and this is of particular concern with <sup>241</sup>Am, <sup>210</sup>Po and <sup>224</sup>Th, as they either partially or completely overlap with Pu spectra peaks. The presence of unwanted metals in the sample solution can prevent the electrodeposition of a thin source, which can result in degradation of the alpha spectrum with a decrease in peak resolution, causing overlapping of alpha peaks.

The yield tracer most commonly used in plutonium analysis using alpha spectrometry is <sup>242</sup>Pu (4.98 MeV). This tracer can sometimes be present in nuclear waste samples, but it would not be present in detectable amounts in

food or drinking water samples. Plutonium-236 could also be used for alpha spectrometry, but it has a relatively short half-life ( $T_{1/2} = 2.58$  years), and so  $^{242}\text{Pu}$ , with its longer shelf life, is the preferred option.

Plutonium extraction from samples, as with other actinides, can be conducted by solvent extraction, anion exchange chromatography and extraction chromatography. These separation techniques rely on the properties of the various oxidation states of this element.

Food samples can be prepared for Pu analysis as outlined in Section I-3.1.

The activity concentrations of plutonium in drinking water are typically very low, and large sample volumes of 100–1000 L may be needed for analysis. In these instances, preconcentration will be necessary. This can be accomplished by co-precipitation with ferric hydroxide, ferrous hydroxide or manganese dioxide. Alternatively, the water sample can be passed through filter fibres impregnated with manganese dioxide.

Solvent extraction techniques for plutonium are mainly used in nuclear fuel reprocessing and waste management processes. These techniques use organic solvents such as tributyl phosphate (TBP), which is used in the separation of Pu in the PUREX process [I-29]. However, solvent extraction is not commonly used for environmental samples and will not be addressed here.

Anion exchange chromatography has been used for the extraction of Pu from environmental samples since the 1960s and is one of the most straightforward approaches, as extraction for alpha spectrometry only requires a single anion exchange column. This has resulted in the publication of a number of standard test methods that have been developed using this approach [I-30 to I-32]. The method is based on the formation of strong anionic complexes of tetravalent Pu with high concentrations of  $\text{HNO}_3$  or sometimes  $\text{HCl}$ .

Once a food or drinking water sample has been pretreated via ashing and digestion and/or co-precipitation using ferric hydroxide, for example, potassium metabisulfite is added to reduce all species of Pu to Pu (III). The sample solution is adjusted to 8 M  $\text{HNO}_3$ , converting Pu (III) to Pu (IV), and loaded onto a preconditioned anion exchange column. The column is washed using 8 M  $\text{HNO}_3$ . Uranium, americium, transition metals and most other elements are not retained on the column and pass through when the sample solution is loaded onto the column. Any minor amounts left on the column are removed during the 8 M  $\text{HNO}_3$  wash. Thorium is washed from the column using 12 M  $\text{HCl}$ . Pu is then eluted from the column by reducing the Pu (IV) to Pu (III) using a 2 M  $\text{HCl}$ –0.1 M  $\text{NH}_2\text{OH}$  solution. The eluate can then be prepared for alpha spectrometry analysis by electrodeposition on a stainless steel disc [I-28].

Plutonium can be extracted from environmental matrices using extraction chromatography resins such as TRU, TEVA and UTEVA. These can be used

individually or in tandem to separate Pu from other interfering radionuclides or to perform sequential extractions of radionuclides in samples (see Section I-5.).

For example, TRU resin can be used to separate Pu and other actinides in  $\text{HNO}_3$  solutions [I-33]. Once a food or drinking water sample has been prepared for analysis using the steps outlined above, the actinides in the sample are preconcentrated by co-precipitation with calcium fluoride. The precipitate is dissolved in 2 M  $\text{HNO}_3$  and 200 mg of boric acid (the addition of boric acid complexes any insoluble fluorides remaining in the sample solution). The oxidation state of Pu in the sample solution is adjusted to Pu (IV) through the addition of 50 mg of sodium nitrite. The TRU resin column is preconditioned in the same manner, with the washing of the column with a solution of 2 M  $\text{HNO}_3$  and 0.01 M  $\text{NaNO}_2$ , and the sample solution is passed through the column followed by 2 M  $\text{HNO}_3$ -0.01 M  $\text{NaNO}_2$  wash. This will retain all actinides on the TRU column and remove all other interfering radionuclides and other matrix constituents. Am is then stripped from the column using 4 M HCl (and the solution can be prepared for electrodeposition and alpha spectrometry, if necessary). Pu is subsequently eluted using 4 M HCl-0.01 M  $\text{TiCl}_3$  solution. The Pu elution can be prepared for alpha spectrometry analysis using electrodeposition onto a stainless steel disc.

Once the Pu samples have been electrodeposited onto a stainless steel disc, the samples can be counted for an appropriate time in an alpha spectrometer. For food or drinking water samples that are likely to contain trace amounts of Pu isotopes, the count time would be on the order of three or four days. Assuming an absolute counting efficiency of 30-40%, chemical recovery greater than 80% and a sample size of approximately 10 g, the limits of detection would be as low as 20-50 mBq/kg.

#### *I-4.1.3. Americium-241*

Americium-241 ( $T_{1/2} = 433$  years) decays 100% by alpha emission to  $^{237}\text{Np}$ . The primary alpha particles emitted have energies of 5.49 MeV (84%) and 5.44 MeV (13%), and the decay also produces a low energy gamma photon (59 keV). However, gamma spectrometry is not suitable for the detection of low

levels of  $^{241}\text{Am}$  in food and drinking water samples (see Section I-3.2). The preferred method is alpha spectrometry, which requires radiochemical separation.<sup>2</sup>

The yield tracer used for the analysis of  $^{241}\text{Am}$  by alpha spectrometry is  $^{243}\text{Am}$ . Americium-243 ( $T_{1/2} = 7370$  years) emits alpha particles with energies of 5.28 MeV (87%) and 5.23 MeV (11%). The energies of these alpha particles are sufficiently separated from those of  $^{241}\text{Am}$  on an alpha spectrum to allow for analysis and activity calculations using alpha spectrometry.

Samples for alpha spectrometry analysis need to be free of interfering alpha emitting radionuclides, especially  $^{238}\text{Pu}$  and  $^{210}\text{Po}$ , whose alpha energies overlap with those of  $^{241}\text{Am}$  and  $^{243}\text{Am}$ . The sample pretreatment and radiochemical separation techniques used for the separation of  $^{241}\text{Am}$  from other interfering radionuclides are those outlined in Section I-3.1. Food samples are homogenized and digested in the same way, and an  $^{243}\text{Am}$  yield tracer is added before radiochemical separation. Drinking water samples would typically require preconcentration prior to radiochemical separation, and this can be conducted by co-precipitation using hydroxides, oxalates or manganese dioxide. For example, calcium oxalate precipitation can concentrate the americium and other actinides in the sample and is also effective at removing iron. This can be achieved through the addition of calcium chloride and oxalic acid to a sample solution and heating. By adjusting the pH of the solution to pH 1.5 using ammonia, the americium is co-precipitated with calcium oxalate, and the precipitate is ready for radiochemical extraction after dissolution [I-22].

As with plutonium, the radiochemical separation techniques used for americium are solvent extraction, ion exchange chromatography and extraction chromatography. The two most common approaches are ion exchange chromatography and extraction chromatography, which use co-precipitation and solvent extraction techniques before and during radiochemical separation to concentrate americium.

Americium separations can be conducted by using consecutive ion exchange columns prior to analysis by alpha spectrometry. A combination of anion and cation exchange columns in conjunction with calcium oxalate precipitation and solvent extraction can be used for americium separation [I-34]. After sample preconcentration using manganese dioxide, the sample is prepared

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<sup>2</sup> Curium is also extracted as part of the Am radiochemical processes outlined in this section. The activity concentrations of curium in environmental samples are very low compared to  $^{241}\text{Am}$ , and the alpha energies of the Cm isotopes do not overlap with those of  $^{241}\text{Am}$  or the  $^{243}\text{Am}$  yield tracer. Therefore, the presence of Cm in the environmental samples or spectra is not of concern from an analytical perspective.

in an 8 M  $\text{HNO}_3$  solution and passed through an anion exchange column that retains plutonium. The eluate is evaporated and dissolved in water before further preconcentration using a double calcium oxalate precipitation. The precipitate is converted to a chloride medium and passed through a column containing both cation and anion exchange resins that retain Po, Th and Fe. Solvent extraction is conducted on the eluate using dibutyl N-N-diethyl carbamyl phosphonate to remove calcium from the sample solution, and a final extraction is conducted in an anion exchange column to remove earth metals before the Am is eluted using a 1.5 M  $\text{HCl}$ –methanol solution. The eluate can be prepared for electrodeposition on a stainless steel disc with subsequent analysis by alpha spectrometry.

The radiochemical separation of americium can be carried out using the Eichrom TRU, TEVA and DGA extraction chromatography resins [I–35]. These extraction chromatography resins are typically used sequentially or in conjunction with anion exchange chromatography to separate americium from actinides and other interfering lanthanides. These sequential separations are, in most cases, used in combined procedures in the determination of americium and other radionuclides in environmental samples (see Section I–5). IAEA methods use calcium fluoride or calcium oxalate co-precipitation in conjunction with the TRU resin to extract  $^{241}\text{Am}$  from environmental samples [I–33]. Using these methods, the separation of Am from other actinides in environmental samples can be accomplished by loading a 3 M  $\text{HNO}_3$  sample solution onto a preconditioned TRU column and eluting the americium using a 4 M  $\text{HCl}$  solution. The Am eluted from the column may also contain lanthanides, and these need to be separated using an additional radiochemical separation step. After elution from the TRU column, the sample solution is evaporated and dissolved in 2 M  $\text{NH}_4\text{SCN}$ –0.1 M  $\text{HOOH}$  and loaded onto a preconditioned TEVA column. Lanthanides are washed from the column using 1 M  $\text{NH}_4\text{SCN}$ –0.1 M  $\text{HOOH}$ , and Am is eluted using 2 M  $\text{HCl}$  [I–36]. Alternatively, the lanthanides can be removed by means of anion exchange chromatography [I–37]. Using this approach, the sample solution eluted from the TRU column is converted to a nitrite and evaporated. The residue is dissolved in 1 M  $\text{HNO}_3$ –95%  $\text{CH}_3$  solution and loaded onto a preconditioned anion exchange column. The lanthanides are eluted from the column using a 0.1 M  $\text{HCl}$ –0.5 M  $\text{NH}_4\text{SCN}$ –80%  $\text{CH}_3\text{OH}$  solution. Americium is then eluted using 1.5 M  $\text{HCl}$ –86%  $\text{CH}_3\text{OH}$ . The eluted solution containing the americium can be prepared for alpha spectrometry by electrodeposition on a stainless steel disc.

All of the procedures outlined above have limits of detection of 1 mBq/sample or lower, assuming an 80% recovery and an absolute alpha counting efficiency of the alpha spectrometry system of 30–40% [I–38].

#### *I-4.1.4. Polonium-210*

There is a standardized approach for the analysis of polonium that can be applied to the assessment of the radionuclide in foodstuffs and water [I-39]. A comprehensive review is available on the analysis of polonium for different matrices, discussing various radiochemical techniques before plating onto a disc by spontaneous deposition [I-40]. Studies on polonium and its natural occurrence in the environment have also been reviewed with a discussion of typical activities and the dose arising from the consumption of foodstuffs and drinking water [I-41, I-42].

Water and food samples are pretreated for analysis using the techniques outlined in Section I-3.1. However, due to the known volatility of polonium at higher temperatures, it is best not to exceed a maximum of 80°C or 90°C for drying and, for the same reason, ashing is not appropriate. Instead, wet digestion is a better option, as high temperatures are not necessary.

Before sample preparation, the tracer is added. The two radioisotopes of polonium normally used as yield tracers are  $^{208}\text{Po}$  and  $^{209}\text{Po}$ . Polonium-208 is more widely available than  $^{209}\text{Po}$ , but  $^{209}\text{Po}$  is more desirable, as its alpha emission energy peak has a larger separation from the  $^{210}\text{Po}$  peak, so it is less likely to overlap with tailing from the  $^{210}\text{Po}$  peak. Furthermore,  $^{209}\text{Po}$  ( $T_{1/2} = 125.2$  years) has a much longer half-life than  $^{208}\text{Po}$  ( $T_{1/2} = 2.89$  years), giving  $^{209}\text{Po}$  standards a much longer shelf life than  $^{208}\text{Po}$  standards.

The aim of the procedures outlined is to digest the sample in chloride form prior to source preparation. Therefore, digestion is carried out using concentrated HCl.

Polonium is preconcentrated in the water samples using either evaporation or co-precipitation. Care is needed if evaporating the sample because of the volatility of polonium. This approach is not very practical when reducing a large sample volume because of the time that would be needed to reduce the volume when evaporating water samples at 80°C or 90°C. If no evaporation or preconcentration step is carried out, digestion and spontaneous deposition can be sufficient to isolate polonium. However, for food samples, filtration after digestion and before spontaneous deposition is necessary.

Co-precipitation is usually carried out during sequential extractions using extraction chromatography for multiple radionuclides.  $\text{MnO}_2$  [I-39, I-41] or  $\text{Fe}(\text{OH})_3$  are the main co-precipitates used. For the  $\text{MnO}_2$  co-precipitation, 0.2 M  $\text{KMnO}_4$  and 0.3 M  $\text{MnCl}_2$  are added to the sample, followed by stirring for an hour; the quantity of chemicals needed is dependent on the volume of sample. The pH is adjusted to 8–9 with  $\text{NH}_3$ . The precipitate is left to settle overnight and is centrifuged. The precipitate is collected and dissolved in 2 M HCl with  $\text{H}_2\text{O}_2$ . The sample is heated to decompose  $\text{H}_2\text{O}_2$  to prepare it for radiochemical

extraction. For co-precipitation with  $\text{Fe}(\text{OH})_3$ ,  $\text{Fe}^{3+}$  ( $\text{Fe}(\text{OH})_3$ ) is added to the water sample. After 30 min of stirring, the sample solution is adjusted to pH 9 or pH 10 with concentrated  $\text{NH}_4\text{OH}$ . Once the precipitate is formed, it is left overnight to settle. As with the  $\text{MnO}_2$  procedure, the precipitate is collected, centrifuged and dissolved in 2 M  $\text{HCl}$  with  $\text{H}_2\text{O}_2$  [I-43].

Following co-preparation, polonium can be chemically separated from the sample solution and other radionuclides present using ion exchange chromatography, extraction chromatography and, to a lesser extent, solvent extraction.

The use of anion exchange columns is preferred over cation exchange columns, as the  $^{210}\text{Po}$  is highly retained by the column in  $\text{HCl}$  medium and can be easily stripped, even in dilute  $\text{HNO}_3$  concentrations [I-42].

Sr Spec extraction chromatography resin can be used for sequential separation of  $^{210}\text{Po}$ ,  $^{210}\text{Bi}$  and  $^{210}\text{Pb}$ . The column is conditioned with 2 M  $\text{HCl}$ , and the sample is loaded onto the column and washed with 2 M  $\text{HCl}$ , which elutes  $^{210}\text{Bi}$ ; and 6 M  $\text{HNO}_3$  is added to the column to elute the  $^{210}\text{Po}$ , separating it from  $^{210}\text{Pb}$ . The  $^{210}\text{Po}$  is dried and redissolved in  $\text{HNO}_3$ ,  $\text{HCl}$  and  $\text{H}_2\text{O}_2$  to destroy any organic material from the column. Finally, the residue is dissolved in 0.5 M  $\text{HCl}$  and is ready for source preparation by spontaneous deposition [I-39].

Polonium isotopes can be plated onto a disc from a mildly acidic solution (0.1–0.5 M  $\text{HCl}$ ) using spontaneous deposition. Spontaneous deposition can be conducted using commercially available or laboratory made ‘spontaneous deposition’ or ‘auto-deposition’ kits. The basic set-up of these kits consists of a container for the sample solution, a stirring mechanism, a source of heat and a disc to plate the polonium. Silver is the most common disc used for spontaneous deposition. Other metals such as copper and nickel have been used for deposition, but these are not as efficient as silver, as there is a greater likelihood of interference from  $^{210}\text{Bi}$  or  $^{210}\text{Pb}$  depositing onto the disc [I-41]. Ascorbic acid is added to the sample solution prior to deposition to reduce  $\text{Fe}^{3+}$  to  $\text{Fe}^{2+}$  in the sample, as the former valence state can disturb the plating by lowering  $^{210}\text{Po}$  recoveries and cause bad resolution of alpha peaks [I-42]. The sample solution is heated to approximately 80–90 °C for 5–6 h [I-40]. Electrodeposition onto stainless steel discs is also an option, but there is little benefit to using this approach compared to spontaneous deposition.

The most common measurement technique for  $^{210}\text{Po}$  is alpha spectrometry using a passivated implanted planar silicon detector. The emission peaks of interest of polonium and two tracers are presented in Table I-5.

A typical  $\text{Po}$  spectrum consists of two singlet peaks: the  $^{210}\text{Po}$  peak and the tracer peak. The  $^{210}\text{Po}$  activity can be derived from the activity of the yield tracer peak.

TABLE I-5. PROPERTIES OF POLONIUM ISOTOPES [I-16, I-44]

Nuclide	T <sub>1/2</sub> (years)	Decay mode	Alpha energy and intensity (MeV (%))
Po-208	2.898	$\alpha$	5.115 (99.99%)
Po-209	115	$\alpha$	4.883 (79.2%) 4.885 (19.8%)
Po-210	0.379	$\alpha$	5.304 (99.99%)

Detection limits of as low as 20 mBq/g and 2 mBq/L for organic samples have been reported for drinking water [I-39, I-40].

#### I-4.1.5. Lead-210

There are a number of different approaches for the analysis of  $^{210}\text{Pb}$ . It can be measured directly by gamma spectrometry, but it can be difficult to measure because of the low energy of the emitted gamma radiation (Section I-3.2). In this section, the analysis of  $^{210}\text{Pb}$  focuses on two techniques: GPC and LSC. Using these techniques,  $^{210}\text{Pb}$  can be determined by measuring the beta emissions from  $^{210}\text{Pb}$  directly and/or indirectly from its progeny  $^{210}\text{Bi}$  or  $^{210}\text{Po}$ . After radiochemical separation has been carried out, the source preparation technique chosen will depend upon the radionuclide being measured and the measurement technique. The activity concentrations found in food can be quite low, and this needs to be considered when determining sample size and sample preparation.

For organic matrices, a stable lead carrier is added to the sample before digestion. A stable carrier containing  $\text{Pb}^{2+}$  element is added to the sample to determine the recovery of the sample.  $\text{Pb}(\text{NO})_3$  and  $\text{PbCl}$  are chemical compounds that can be used as a carrier. The chemical recovery can be determined gravimetrically or by ICP-MS. The samples can be pretreated using the methods outlined in Section I-3.1. Following pretreatment, the sample is dried and dissolved in concentrated HCl.

For a water sample, the stable lead carrier is added after filtration and acidification. The lead is preconcentrated in the water samples using co-precipitation with either  $\text{MnO}_2$  [I-39, I-45] or  $\text{Fe}(\text{OH})_3$ , as outlined in Section I-3.1.1.

Unlike  $^{210}\text{Po}$ , which can rely upon spontaneous deposition to separate interfering radionuclides,  $^{210}\text{Pb}$  has to be chemically separated from the sample matrix. Lead-210 also has to be separated from its  $^{210}\text{Bi}$  progeny during separation,



as the measurement of  $^{210}\text{Pb}$  relies on fresh ingrowth of this radionuclide after separation. Separation techniques include ion exchange chromatography and extraction chromatography [I-39, I-40, I-46, I-47]. Using these separation techniques, sequential separations of naturally occurring radionuclides can be conducted [I-39].

As mentioned above, the strontium resin can be used for sequential separation of  $^{210}\text{Po}$ ,  $^{210}\text{Bi}$  and  $^{210}\text{Pb}$ . The column is conditioned with 2 M HCl and the sample is loaded onto the column and washed with 2 M HCl, which elutes the  $^{210}\text{Bi}$ . The time and date of the separation of  $^{210}\text{Bi}$  need to be recorded. This allows the ingrowth of  $^{210}\text{Bi}$  to be determined for activity calculations. If conducting sequential analysis to determine  $^{210}\text{Po}$ , 6 M  $\text{HNO}_3$  is added to the column to elute the  $^{210}\text{Po}$ , separating it from the  $^{210}\text{Pb}$ , and 6 M HCl is then added to elute the  $^{210}\text{Pb}$ . The  $^{210}\text{Pb}$  fraction is dried and redissolved in HCl and  $\text{H}_2\text{O}_2$  to destroy any organic material from the column. Concentrated  $\text{H}_2\text{SO}_4$  is added to the solution to form a sulphate precipitate with the lead carrier. If the radiochemical yield is determined gravimetrically, precipitation of lead is necessary. The precipitation step serves as a further purification of the  $^{210}\text{Pb}$ , as well as determining the yield. Lead can be precipitated into a variety of compounds, namely  $\text{PbSO}_4$  [I-48, I-49],  $\text{PbC}_2\text{O}_4$  [I-50, I-51] and  $\text{PbCrO}_4$  [I-52]. After precipitation and centrifugation, the precipitate is washed with deionized water and centrifuged until the pH is neutral.

For GPC, the precipitate is dried on a preweighed aluminium planchette with an infrared lamp. The recovery is calculated by the weight of the stable carrier on the planchette. The sample is stored for at least 14 days prior to measurement. This storage period is to allow for the ingrowth of  $^{210}\text{Bi}$ , as its emissions are detected in order to determine the activity of  $^{210}\text{Pb}$ . The beta energy of  $^{210}\text{Bi}$  is much higher than that of  $^{210}\text{Pb}$  and is much easier to measure (see Table I-6). There is negligible contribution from  $^{210}\text{Pb}$ , as its energy is so low. Typical efficiencies for  $^{210}\text{Bi}$  are ~40%, with minimum detectable activity values of 3 mBq/sample. The  $^{210}\text{Pb}$  activity concentrations are determined by Bateman's equation, which is described by Johansson [I-53].

TABLE I-6. PROPERTIES OF  $^{210}\text{Pb}$  AND  $^{210}\text{Bi}$  [I-16]

Nuclide	$T_{1/2}$ (years)	Decay mode	Maximum beta energy and intensity (KeV (%))
Pb-210	22.23	$\beta$	63.5 (100%)
Bi-210	5 days	$\beta$	1161.2 (99.99%)

For LSC, the precipitate and solution are filtered on a preweighed membrane filter, dried at 80°C for an hour and cooled in a desiccator prior to determining the final weight of the filter paper. The membrane filter is washed with 3 M  $\text{HNO}_3$  to dissolve the precipitate. The solution is dried and redissolved in 2 M  $\text{HNO}_3$  before the sample is transferred to a polyethylene LSC vial containing 15 mL of scintillant that is optimized for alpha and beta discrimination. ICP-MS can also be used to determine the recovery for LSC measurements. An aliquot of sample is taken prior to separation and measured. During the source preparation for the LSC vial, any leftover sample solution is measured by ICP-MS, and the difference in measurements determines the recovery of lead [I-54].

In liquid scintillation measurement, it is possible to measure the sample as soon the sample is prepared, whereas for GPC the sample is stored for a minimum of 14 days to allow for the ingrowth of  $^{210}\text{Bi}$ . However, for LSC there will always be a small amount of interference from  $^{210}\text{Bi}$ , which needs to be taken into consideration during the analysis. The longer the sample is stored prior to counting, the greater the interference from ingrowth of  $^{210}\text{Bi}$ . Interferences can be corrected through the use of various energy windows on the LSC [I-52]. Similar detection limits are seen for both techniques.

Lead-210 can also be determined indirectly using its daughter product,  $^{210}\text{Po}$ . After plating the  $^{210}\text{Po}$  onto the disc, as previously described in Section I-3.1, the samples can be kept for a period of approximately six months to allow for the ingrowth of any  $^{210}\text{Po}$  arising from the decay of  $^{210}\text{Pb}$  in the intervening time period. The ingrown  $^{210}\text{Po}$  can be plated again with a new aliquot of polonium tracer as a yield tracer, and the  $^{210}\text{Pb}$  activity concentration can be determined again using Bateman equations [I-55].

#### *I-4.1.6. Uranium*

For the analysis of uranium, traditional radiometric methods are the normal approach. The ISO has developed a standardized approach for the analysis of uranium isotopes using chemical extractions and alpha spectrometry analysis after the preparation of a thin source on a disc or filter paper [I-14]. Alpha spectrometry offers the most straightforward approach to the determination of the three key isotopes of uranium, namely  $^{234}\text{U}$ ,  $^{235}\text{U}$  and  $^{238}\text{U}$ . The properties of these U isotopes and the radiochemical tracer used for alpha spectrometry are outlined in Table I-7.

Mass spectrometry methods for uranium analysis, such as ICP-MS, are also available, and very low limits of detection can be achieved. However, these techniques are expensive and more complex, as additional purification and measurement components may be needed. For example, nebulizers are necessary to detect the less abundant uranium isotopes [I-56].

TABLE I-7. PROPERTIES OF NATURALLY OCCURRING URANIUM AND  $^{232}\text{U}$  [I-16]

Nuclide	$T_{1/2}$ (years)	Decay mode	Alpha energy and intensity (MeV (%))
U-234	$2.45 \times 10^5$	$\alpha$	4.774 (71.37%) 4.722 (28.42%)
U-235	$7.04 \times 10^8$	$\alpha$	4.397 (57.19%) 4.366 (18.80%)
U-238	$4.4 \times 10^6$	$\alpha$	4.198 (77.5%) 4.151 (22.3%)
U-232	70.6	$\alpha$	5.320 (69.1%) 5.263 (30.6%)

Water and food samples are pretreated for analysis using the techniques outlined above in Section I-3.1. Uranium-232 is used as a yield tracer, as the energy peaks from  $^{232}\text{U}$  do not overlap with the other isotopes of uranium.

The first step to isolate uranium in the sample is preconcentration. This is achieved by co-precipitation using ferric hydroxide ( $\text{Fe}^{3+}$ ). After the addition of  $\text{Fe}(\text{OH})_3$  to the sample, ammonia is added to adjust the pH to 9, forming a precipitate. This precipitate is centrifuged and dissolved in 6 M  $\text{HNO}_3$ . The sample can be separated in two ways, through either anion exchange or extraction chromatography. A combination of both anion exchange chromatography and extraction chromatography can also be used for high purification of uranium or for conducting sequential separations of different radionuclides, including uranium [I-57, I-58].

For anion exchange, the column is conditioned in 8 M HCl. The sample is dried, and the residue is redissolved in 8 M HCl. The sample solution is loaded onto the column. The column is washed twice with 8 M HCl to remove any thorium and radium. The column is then washed with 3 M HCl to elute neptunium and plutonium. A further wash of the column with 7 M  $\text{HNO}_3$  is made to remove iron from the column. Finally, the uranium is eluted using 0.1 M HCl and collected for source preparation [I-14].

For extraction chromatography, TEVA and UTEVA columns are stacked prior to radiochemical extraction. Thorium causes the greatest interference with uranium, so the principal aim is the separation of these isotopes. The columns are conditioned with 3 M  $\text{HNO}_3$ . The sample is dried and the residue is redissolved

in  $\text{Al}(\text{NO}_3)_3\text{--HNO}_3$  solution. The sample solution is passed through the column. The column is rinsed twice with 3 M  $\text{HNO}_3$ , which elutes the uranium from the TEVA column and retains it on the UTEVA column. The columns are then separated. The UTEVA column is rinsed with 3 M  $\text{HNO}_3$ , followed by a 9 M  $\text{HCl}$  wash to convert the column to chloride form. The column is washed with  $\text{HCl}$ –oxalic acid solution to ensure the removal of thorium, plutonium and neptunium. Finally, the uranium is eluted from the column using 1 M  $\text{HCl}$ . The sample is collected in a beaker and is ready for alpha spectrometry [I–14, I–59].

Typical detection limits for uranium isotopes are 5 mBq/L for 500 mL of water and 5 mBq/kg for 10–100 g of food (which can be reduced to 5 g if the sample is ashed during sample pretreatment).

#### *I–4.1.7. Thorium*

The two most common naturally occurring thorium isotopes are  $^{232}\text{Th}$  and  $^{230}\text{Th}$ . Thorium-232 and  $^{228}\text{Th}$  are part of the thorium decay series, with  $^{232}\text{Th}$  comprising more than 99.5% of the mass of naturally occurring thorium. Thorium-230 is the next most abundant isotope in nature (<0.05%) and is part of the  $^{238}\text{U}$  decay series. The remaining three thorium isotopes,  $^{227}\text{Th}$ ,  $^{231}\text{Th}$  and  $^{234}\text{Th}$ , exist in only trace amounts in nature, have relatively short half-lives compared to  $^{232}\text{Th}$ ,  $^{230}\text{Th}$  and  $^{228}\text{Th}$  and do not contribute significantly to radioactivity in food or drinking water, and measurement of these isotopes is not considered here (Table I–8).

Thorium-232,  $^{230}\text{Th}$  and  $^{228}\text{Th}$  are alpha emitters and are typically measured using alpha spectrometry techniques that require the addition of the artificial thorium isotope  $^{229}\text{Th}$  ( $E_\alpha = 5.168$  MeV) prior to radiochemical separation [I–60]. Mass spectrometry techniques such as ICP-MS and OE-MS can also be used to determine the activity concentrations of these long lived isotopes [I–61]. The decay of  $^{232}\text{Th}$  to  $^{228}\text{Ra}$  produces gamma radiation, but its intensity is too low to allow the direct determination of  $^{232}\text{Th}$  by gamma spectrometry. In addition, the gamma photon with the highest intensity occurs at 63.8 keV (0.26%) and is subject to self-attenuation at such a low energy. In some cases,  $^{232}\text{Th}$  can be measured via its  $^{228}\text{Ac}$  progeny, but this approach can only be utilized if there is equilibrium with the radionuclides in the decay scheme, namely  $^{232}\text{Th}$ ,  $^{228}\text{Ra}$  and  $^{228}\text{Ac}$ . This cannot be assumed for food and drinking water samples, and therefore this approach is not suitable for these matrices.

To analyse a food or drinking water sample for thorium, one or more radiochemical separation techniques need to be used to separate the thorium isotopes from other interfering alpha emitting radionuclides, such as  $^{210}\text{Po}$ ,  $^{232}\text{U}$ ,  $^{234}\text{U}$ ,  $^{237}\text{Np}$ ,  $^{238}\text{Pu}$  and  $^{241}\text{Am}$ . The radiochemical separation techniques that can be

TABLE I-8. NATURALLY OCCURRING THORIUM ISOTOPES [I-16]

Nuclide	$T_{1/2}$ (years)	Decay mode	Alpha energy and intensity (MeV (%))
Th-227	0.051	$\alpha$	5.977 (23%) 6.038 (24%)
Th-228	1.91	$\alpha$	5.42 (73%) 5.34 (26%)
Th-230	$7.5 \times 10^4$	$\alpha$	4.69 (76%) 4.62 (23%)
Th-231	0.0029	$\beta$	n.a. <sup>a</sup>
Th-232	$1.4 \times 10^{10}$	$\alpha$	4.01 (79%) 3.95 (21%)
Th-234	0.066	$\beta$	n.a.

<sup>a</sup> n.a.: not applicable.

used are co-precipitation in conjunction with solvent extraction, anion exchange chromatography or extraction chromatography.

Solvent extraction techniques utilize various organic extraction reagents such as TBP, TEAC (Trolox equivalent antioxidant capacity) assay kits and ethylenediaminetetraacetic acid (EDTA) to extract thorium isotopes in nitric acid systems, but this approach is typically used for the recovery of thorium from rare earths and in the production and processing of thorium for use as fuels in experimental and test reactors, rather than for analyses of radioactivity in food or drinking water [I-62 to I-64].

Co-precipitation can be used to separate thorium from other alkali and alkaline earth metals. An iron phosphate, calcium phosphate or manganese oxide co-precipitation can separate thorium and other actinides from other interfering radionuclides [I-65, I-66]. Once separated from these earth metals, further separation from other actinides is needed, which is performed via ion exchange chromatography or extraction chromatography.

The separation of thorium using anion exchange columns is a two stage process. First, the sample solution is passed through an anion exchange column in a strong chloride form (i.e. 8 M HCl). This retains the alpha emitting radionuclides uranium, protactinium, polonium, plutonium and neptunium on the column.

Thorium, along with americium, curium and radium (if not already removed by co-precipitation), passes through this column. A second anion exchange column in a nitrate form (i.e. 8 M HNO<sub>3</sub>) retains the thorium on the column and allows radium, americium and curium to pass through. The thorium retained on the column can then be eluted separated using a weak nitric acid [I-67].

An alternative approach to anion exchange chromatography is the use of commercially available extraction chromatography resins, such as DGA, Diphonix, TRU, UTEVA and TEVA [I-24]. The most commonly used resin for thorium extraction is TEVA. Thorium and other actinides can be separated from other interfering radionuclides in a sample using co-precipitation with calcium phosphate. The precipitate is then dissolved in 3 M HNO<sub>3</sub> and is passed through the TEVA column, where the thorium is retained on the column with plutonium and neptunium, while uranium and americium pass through. Thorium is then eluted from the column using 9 M HCl, with the other actinides being retained [I-68].

The thorium solution eluted from the anion exchange column or extraction chromatography column is typically electrodeposited onto a disc and counted via alpha spectrometry.

The alpha energies of the four alpha emitting thorium isotopes are outlined in Table I-8. These energies range between 3.9 MeV (<sup>232</sup>Th) and 6 MeV (<sup>227</sup>Th). The alpha energies of the four naturally occurring isotopes and the <sup>229</sup>Th tracer are sufficiently different to be identified on the same alpha spectrum. The <sup>229</sup>Th tracer added to the sample prior to radiochemical separation is used to determine the yield and activity concentration of the <sup>232</sup>Th and <sup>230</sup>Th isotopes in the sample. Thorium-227 activity concentrations in nature are very low, and alpha emissions are not normally observed on the alpha spectrum. However, the alpha emitting thorium progeny <sup>224</sup>Ra, <sup>212</sup>Bi, <sup>220</sup>Rn, <sup>216</sup>Po and <sup>212</sup>Po will be observed on the spectrum as ingrowth occurs, but they do not interfere with the thorium isotope peaks.

#### *I-4.1.8. Radium*

The IAEA has published an analytical quality review of different analytical methodologies for the determination of radium isotopes in environmental samples [I-69]. An extensive review on radium and the different analytical techniques that can be employed has also been published by Jia and Jia [I-70]. Radium-226 is much more straightforward to measure than <sup>228</sup>Ra, as it is an alpha emitter with a much longer half-life, and many radiochemical extraction techniques are available. Radium-228 is much more difficult to analyse, as it is a low energy beta emitter. However, it decays to <sup>228</sup>Ac, a beta emitter with a much higher energy, and this can be used to measure <sup>228</sup>Ra indirectly. Actinium-228

has a short half-life of 6.15 h, and this also needs to be taken into consideration during analysis. The properties of <sup>226</sup>Ra, <sup>228</sup>Ra and <sup>228</sup>Ac are outlined in Table I–9.

The other naturally occurring radionuclides, <sup>223</sup>Ra and <sup>224</sup>Ra, will not be considered in this overview of analysis, as their half-lives are relatively short compared to those of other radium isotopes, and their contribution to dose is negligible.

Water and food samples are pretreated for analysis using the techniques outlined in Section I–3.1. The most commonly used radioactive tracers for radium analysis are <sup>223</sup>Ra, <sup>224</sup>Ra, <sup>225</sup>Ra and <sup>133</sup>Ba. While it is more advantageous to use tracers that are the same isotope as the radionuclides of interest, their half-lives are very short, and they may not be readily available to most laboratories. Radium and barium are chemically very similar, as they are in the same elemental group. However, care is needed during analyses because the small differences between them can have an effect on determining the recovery of Ra when using the Ba tracer, as demonstrated by Sill [I–71]. Barium-133 ( $E_{\text{gamma}} = 356 \text{ keV}$  (62%)) has a half-life of 10.5 years and can be used as a radioactive yield tracer in these radiochemical analyses, with the yield determined after chemical extraction using gamma spectrometry. A non-radioactive barium carrier, such as barium chloride, can also act as a tracer and carrier, and the yield when using stable barium can be determined gravimetrically [I–72].

Radium is preconcentrated by co-precipitation. Co-precipitation with barium sulphate (or lead sulphate) is the most common approach and can be used for the separation and purification of radium [I–69]. This co-precipitation step is conducted by adjusting the water sample pH to 0–1 with hydrochloric acid. The barium carrier is added and the water sample is boiled for 10 min; 9 M sulphuric

TABLE I–9. PROPERTIES OF <sup>226</sup>Ra, <sup>228</sup>Ra AND <sup>228</sup>Ac [I–16]

Nuclide	T <sub>1/2</sub>	Decay mode	Alpha/beta energy and intensity (MeV (%))
Ra-226	1 600 years	$\alpha$	4.78 (94%)
Ra-228	5.75 years	$\beta$	0.046 (100%)
Ac-228	6.15 h	$\beta^-$	2.123 (100%)
		$\gamma$	0.015 (37%) 0.911 (26.2%) 0.968 (15.9%) 0.338 (11.4%)

acid is added to the boiled solution to start the formation of the precipitate and is left boiling for a further 30 min. The sample is left to cool and to allow the precipitate to settle [I-71, I-72]. Depending on the measurement technique, either the sample can be prepared for gamma spectrometry by filtering and drying the precipitate or further radiochemical separation can be conducted. The quantity of barium carrier varies depending on the stage at which the barium is separated from radium. If further chemical separation steps are necessary, then 75 µg to 110 mg of  $\text{Ba}^{2+}$  can be added [I-69].

Co-precipitation with  $\text{MnO}_2$  can also be used when the preconcentration of a number of radionuclides is necessary [I-73]. The sample is purged with  $\text{N}_2$  for 2 h to eliminate any dissolved  $\text{CO}_2$ . This is completed by adding 0.2 M  $\text{KMnO}_4$  and adjusting the pH to 8–9 with  $\text{NH}_3$ ; then 0.3 M  $\text{MnCl}_2$  is added, and the solution is left to bubble for an hour and the precipitate is left to settle. The precipitate is collected and washed with 0.2%  $\text{NH}_4\text{Cl}$  and filtered. The filtrate is dissolved by 1.2 M  $\text{HCl}$  and 1%  $\text{H}_2\text{O}_2$ , left to evaporate to dryness, redissolved in 5 M  $\text{HCl}$  and evaporated again to decompose any peroxides in the sample [I-73].

Barium sulphate co-precipitation is used if radium is the only radionuclide of interest in the water sample [I-74]. However, if other radionuclides in the sample are also being measured in the sample as part of a sequential analysis, then this approach cannot be used (see Section I-5). Evaporation of the water sample can be performed prior to co-precipitation to increase the sample volume for better detection limits for the likes of gamma analysis, but co-precipitation alone is usually sufficient for the preconcentration if further radiochemical separation steps are being carried out.

Following sample preconcentration, radium analysis can be carried out by a number of radiochemical separation techniques that are based on ion exchange chromatography and extraction chromatography techniques.

Ion exchange chromatography can be used for food samples. After the organic material of the food sample has been destroyed, the sample is filtered, and the filtrate is evaporated to dryness. The residue is redissolved in 9 M  $\text{HCl}$  after sample digestion and loaded onto a strongly basic anion exchange column. The sample is passed through the column, separating it from uranium and  $^{210}\text{Po}$ . The sample solution is dried and redissolved in 8 M  $\text{HNO}_3$  and passed through another strongly basic anion exchange resin. The solution is in a nitrate medium, causing the thorium to be retained on the column while allowing the radium to pass through. The radium fraction is dried and dissolved in hydrochloric acid (pH 1.5) and loaded onto a cation exchange column. After the addition of a highly concentrated hydrochloric acid, radium and barium will elute from the column. The barium yield tracer can be measured by gamma spectrometry and the  $^{226}\text{Ra}$  by alpha spectrometry or liquid scintillation counting [I-71].



Extraction chromatography using the commercially available Mn resin, Ln resin and DGA resin columns can also be used to separate  $^{226}\text{Ra}$  and  $^{228}\text{Ra}$  from samples [I-75]. After sample pretreatment and the addition of  $^{133}\text{Ba}$ , a sample residue is redissolved to pH 2 using concentrated HCl. The solution is adjusted to pH 6–7 using 6 M NaOH and loaded onto a Mn resin column. The  $\text{MnO}_2$  resin in the column is dissolved using 4 M HCl–15%  $\text{H}_2\text{O}_2$ , and the solution is stored for 36 h to allow for the ingrowth of  $^{228}\text{Ac}$ . Following storage, the solution is passed through stacked Ln resin and DGA resin columns. The Ln resin column removes uranium and thorium, and the DGA resin retains  $^{228}\text{Ac}$ . The radium and barium pass through both columns, and the eluate is evaporated to dryness and set aside. The  $^{228}\text{Ac}$  retained on the DGA column is eluted using 0.5 M HCl, and this eluate is measured by LSC immediately after counting to determine the  $^{228}\text{Ra}$  activity concentration. The Ra–Ba residue is dissolved in 0.1 M HCl, and further elutions are carried out using the Ln resin column to remove other interferences prior to microprecipitation onto a filter and counting by alpha spectrometry.

Another commercial product, Radium Rad Disks by 3M, is a further form of solid phase extraction specifically for radium in water samples. They are membranes that act like filters and are made of up inert polymers, to which the radium isotopes selectively adhere [I-76]. These discs are expensive, and high volumes of 2 M  $\text{HNO}_3$  are needed, but they can be reused. The water sample is filtered through a mounted EMPORE Radium Rad disc membrane, and the  $^{226}\text{Ra}$  and  $^{228}\text{Ra}$  in the sample are retained. For  $^{228}\text{Ra}$  analysis, the filter is left in a Petri dish for 14–28 days to allow for ingrowth of  $^{228}\text{Ac}$ . The filter is prepared in the mount again and the  $^{228}\text{Ac}$  is then eluted using 0.5 M  $\text{HNO}_3$ . The solution can be precipitated with yttrium oxalate or by fluoride microprecipitation and then counted by GPC [I-77] or LSC [I-78]. Radium-226 measurement can be conducted by LSC or the filter can be mounted on a disc for alpha spectrometry [I-79].

The IAEA has developed a method for the rapid determination of  $^{226}\text{Ra}$  and  $^{228}\text{Ra}$  in drinking water samples. Co-precipitation using the barium sulphate method is used to preconcentrate a large sample volume to reach low detection limits. Following this, the  $\text{Ba}(\text{Ra})\text{SO}_4$  precipitate is dissolved in EDTA solution along with ammonium sulphate and glacial acetic acid is added to form another precipitate. The solution is boiled for 10 min and cooled followed by centrifugation. This is repeated twice more to ensure purification. The precipitate is washed in deionized water and then hot EDTA is added to suspend the precipitate.  $\text{BaSO}_4$  is added to the vial and a scintillant cocktail is added [I-80].

Measurement of radium isotopes can be carried out using gamma spectrometry, alpha spectrometry ( $^{226}\text{Ra}$ ) and LSC ( $^{226}\text{Ra}$  and  $^{228}\text{Ra}$ ).

Measurements of food or drinking water samples using gamma spectrometry will involve co-precipitation and radiochemical separation prior to measurement

to attain appropriate limits of detection. The activities for  $^{226}\text{Ra}$  and  $^{228}\text{Ra}$  using gamma spectrometry are determined through measurement of their short lived progeny. Following co-precipitation or radiochemical separation, samples need to be sealed and stored for approximately 28 days to ensure that  $^{226}\text{Ra}$  and its progeny have reached secular equilibrium. The progeny measured are  $^{214}\text{Pb}$  (295 keV and 352 keV) and  $^{214}\text{Bi}$  (609 keV) for  $^{226}\text{Ra}$  and  $^{228}\text{Ac}$  (338 keV and 911 keV) for  $^{228}\text{Ra}$ . Typical detection limits using gamma spectrometry  $^{226}\text{Ra}$  are 0.1–1 mBq and 100 mBq for  $^{228}\text{Ra}$  [I–81].

Liquid scintillation counting can be used in the rapid determination of  $^{226}\text{Ra}$  and  $^{228}\text{Ra}$  activities in drinking water, as the ingrowth of radium progeny is not needed. As discussed above, the Rad discs can simply be added to a vial with scintillant cocktail and counted, and the IAEA has also developed a rapid assessment method for the analysis of these isotopes using this method [I–80].

Alpha spectrometry is the preferred method for  $^{226}\text{Ra}$  analysis, as the limits of detection can be up to two orders of magnitude lower than those used in gamma spectrometry or LSC [I–70]. Source preparation for alpha spectrometry requires a thin and even source layered on a stainless steel, silver or nickel disc. A variety of techniques, including direct evaporation, co-precipitation and electrodeposition, can be used to prepare such sources. However, the preferred method is the microprecipitation of a sample after chemical extraction onto a disc [I–71]. Electrodeposition techniques can also be conducted, but they involve the addition of a Ra tracer, and these are not readily available and have a relatively short half-life.

Mass spectrometry is another measurement method for the determination of  $^{226}\text{Ra}$  and  $^{228}\text{Ra}$ . ICP-MS is the most common form of mass spectrometry to determine the concentration of  $^{226}\text{Ra}$  and the  $^{226}/^{228}\text{Ra}$  isotopic ratio. This approach requires further purification of samples after the radiochemical extraction techniques outlined above to remove Ba from the sample solution, as Ba can cause interferences in the Ra spectrum in ICP-MS systems [I–82].

#### *I–4.1.9. Carbon-14*

Carbon-14 ( $T_{1/2} = 5730$  years) is a low level beta emitter ( $E_{\text{max}} = 156$  keV), and measurement of this isotope can be conducted only after complete removal from the sample of interest. Removal of  $^{14}\text{C}$  from food and water samples is carried out using a combustion furnace [I–83].

Food samples for  $^{14}\text{C}$  analysis are pretreated by drying and homogenization as outlined in Section I–3.1. The addition of a yield tracer or stable carrier is not necessary, as the combustion furnace is assumed to have 100% efficiency in removing  $^{14}\text{C}$  from the sample. However, the effectiveness of the recovery of the

combustion system is checked periodically by the use of  $^{14}\text{C}$  standards to monitor furnace performance.

Approximately 0.5–1 g of food sample is placed into a quartz sample boat and loaded into a quartz tube in the sample furnace. The combustion furnace is set to  $900^{\circ}\text{C}$  to ensure complete conversion of the carbon to a gaseous oxidized state. The combustion gases from the sample contain  $^{14}\text{C}$  in the form of carbon dioxide ( $^{14}\text{CO}_2$ ) and carbon monoxide ( $^{14}\text{CO}$ ). These gases are carried via airflow into a second furnace containing a Pt–alumina catalyst and pure oxygen, which oxidizes the  $^{14}\text{CO}$  to  $^{14}\text{CO}_2$ . The  $^{14}\text{CO}_2$  released from the furnace can be trapped in a number of different ways, depending on the measurement technique.

Carbon-14 can be measured by LSC, GPC or atomic mass spectrometry. The most straightforward measurement technique for food samples is low level LSC. To prepare a  $^{14}\text{C}$  sample for LSC analysis, the  $^{14}\text{CO}_2$  released from the furnace is bound to an amine. Commercially available amines, such as Perkin Elmer Carbosorb and Meridian Biotechnologies CarbonTrap solvents, have been specifically designed to capture  $^{14}\text{CO}_2$ . The CarbonTrap solvent is loaded into a gas bubbler and attached to the outlet of the combustion furnace before combustion of the sample. The  $^{14}\text{CO}_2$  expelled from the furnace is passed through the CarbonTrap in the bubbler, forms a carbamate with the amine and is retained in the CarbonTrap. A fraction of the CarbonTrap is transferred to a glass scintillation vial and a scintillation cocktail, such as Meridian Biotechnologies CarbonCount, is added. The vial is counted by LSC, and the  $^{14}\text{C}$  activity concentration can be determined. The limit of detection for this measurement technique is typically 10 Bq/kg, which is sufficient for food samples.

## I-5. COMBINED RADIOANALYTICAL PROCEDURES FOR THE DETERMINATION OF RADIONUCLIDES IN ENVIRONMENTAL SAMPLES

Radiochemical separation techniques can be used sequentially to separate and measure the activity concentrations of several different radionuclides in the same sample matrix. The majority of these sequential separations use extraction chromatography resins that are capable of separating the actinides in various oxidation states. The IAEA has developed a number of methods for the sequential extraction of actinides and other radionuclides using extraction chromatography columns in conjunction with co-precipitation, solvent extraction and ion exchange chromatography techniques [I-33, I-37, I-69, I-84]. For example, Fig. I-1 outlines a sequential radiochemical procedure for the determination of  $^{90}\text{Sr}$ ,  $^{241}\text{Am}$  and Pu isotopes using LSC and alpha spectrometry [I-37].

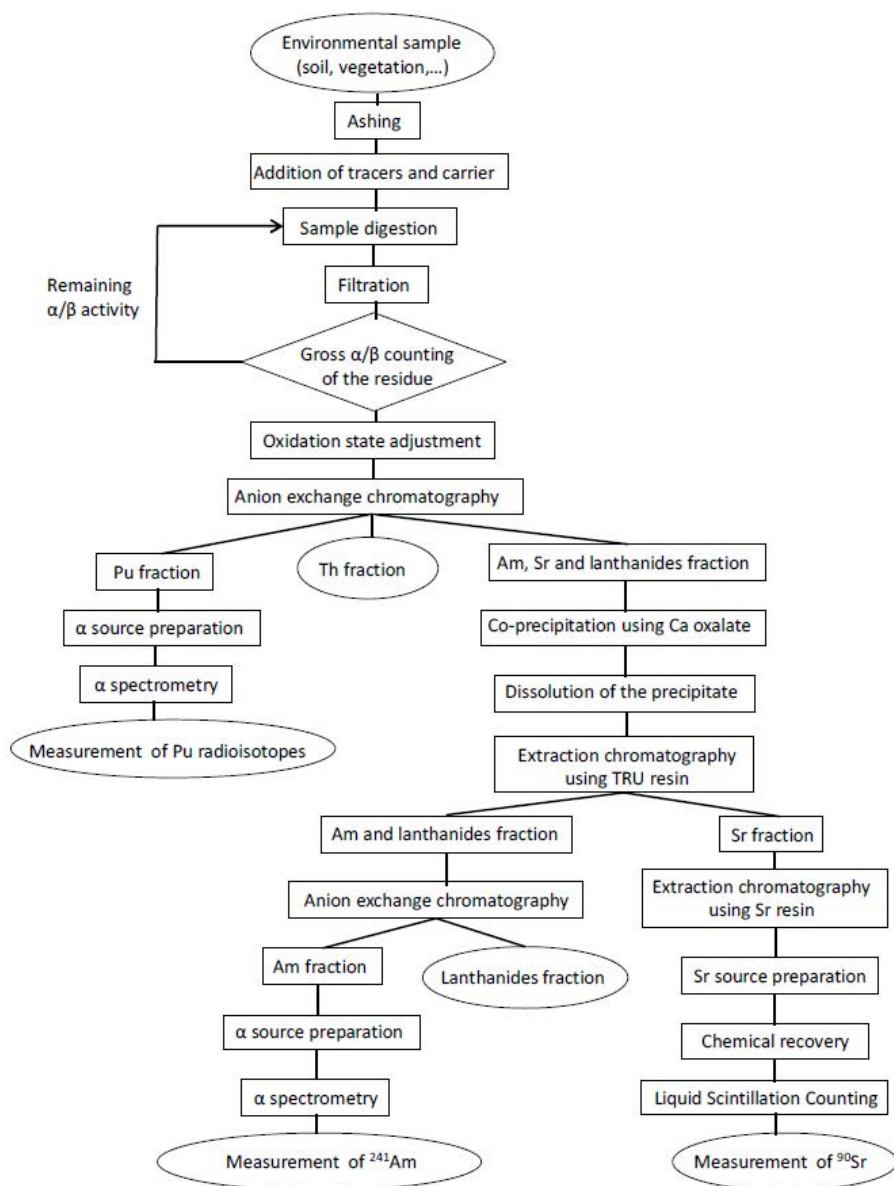


Fig. I-1. Flow diagram outlining the sequential extraction procedure for Pu, Am and Sr in environmental samples [I-37].

The methods developed by the IAEA are principally for use in environmental matrices but can be modified accordingly for the measurement of food and drinking water samples, if necessary.

## I-6. GENERAL QUALITY CONTROL CONSIDERATIONS

To ensure that all of the methods outlined in this annex are reliable, consistent and fit for purpose, consideration needs to be given to the quality of the analytical technique, the results produced and the overall approach to quality in the laboratory conducting the measurements.

The quality of the analysis and the results begins with the appropriate validation of the analytical techniques and continues with adequate quality assurance and quality control on an ongoing basis. This is achieved through the following:

- (a) The use of quality control samples during analysis. Quality control standards include blank samples, calibration checks, sample duplicates, recovery checks and carrier or yield checks. These can be tracked on control charts and reviewed on a regular basis for acceptability and investigation of trends.
- (b) Equipment checks and maintenance. A programme of routine quality control checks is needed for all the equipment used to ensure that there are no changes in counting efficiency, background count rates or, in the case of spectrometry systems, energy. An adequate programme of preventative maintenance for equipment also needs to be implemented.
- (c) Participation in proficiency testing schemes and interlaboratory comparisons. Participation in these schemes acts as an external reference for the analytical technique of interest. A number of national and international bodies routinely conduct proficiency testing and interlaboratory comparisons. For example, the IAEA's Analytical Laboratories for the Measurement of Environmental Radioactivity (ALMERA) network conducts interlaboratory comparisons on environmental matrices on an annual basis [I-85].
- (d) Traceability of radioactive standards. Radioactive standards and tracers used for method validation and ongoing routine analysis need to be certified and traceable to a national or international standard. These standards also need to be tested on a regular basis to ensure that there are no significant changes in the activity concentrations as a result of, for example, plate-out of the standard on the standard container or evaporation.
- (e) Measurement uncertainty. The uncertainty, limit of detection and decision threshold for any result are to be determined for each analysis, taking into consideration all sources of uncertainty. Guidance on the evaluation of

measurement uncertainty for radioactivity measurements is available from the ISO and IAEA [I–86, I–87].

Other factors to be considered to ensure the overall maintenance of quality in a measurement laboratory include:

- Training of staff;
- Development of standard test procedures to ensure consistency of the analytical technique regardless of the analyst;
- Maintenance of records, such as sample documentation, equipment calibration and maintenance records and staff training records.

### I–7. SUMMARY

A summary of the tracers, radiochemical procedures, measurement techniques and limits of detection is provided in Table I–10.

TABLE I–10. OVERVIEW OF METHODS USED FOR RADIOACTIVITY ANALYSIS IN FOOD AND DRINKING WATER SAMPLES

Radionuclide	Yield tracer/carrier	Radioanalytical technique	Measurement technique	Typical detection limit
Gross alpha Gross beta	Not needed	Evaporation	Gas proportional counting Liquid scintillation counting	5 mBq/L
Cs-137	Not needed	Drying and homogenization	Gamma spectrometry	0.5 Bq/kg
Sr-90	Strontium nitrate Yttrium nitrate	Extraction chromatography Solvent extraction (HDHEP)	Liquid scintillation counting Cherenkov counting	1–20 mBq/kg

TABLE I–10. OVERVIEW OF METHODS USED FOR RADIOACTIVITY ANALYSIS IN FOOD AND DRINKING WATER SAMPLES (cont.)

Radionuclide	Yield tracer/carrier	Radioanalytical technique	Measurement technique	Typical detection limit
Pu-238 Pu-239/240	Pu-242	Anion exchange chromatography Extraction chromatography (TRU resin)	Alpha spectrometry	20–50 mBq/kg
Am-241	Am-243	Ion exchange chromatography Extraction chromatography (TRU resin)	Alpha spectrometry	1–20 mBq/kg
Po-210	Po-209	Co-precipitation Spontaneous deposition	Alpha spectrometry	2–20 mBq/kg
Pb-210	Pb <sup>2+</sup>	Co-precipitation Extraction chromatography (Sr resin) Anion exchange chromatography	Gas proportional counting Liquid scintillation counting	3–30 mBq/kg
Ra-226	Ba-133 Ba <sup>2+</sup>	Co-precipitation Ion chromatography Extraction chromatography	Gamma spectrometry Liquid scintillation counting Alpha spectrometry	0.1–1 mBq/kg
Ra-228	Ba-133 Ba <sup>2+</sup>	Co-precipitation — barium sulphate Extraction chromatography	Gamma spectrometry Liquid scintillation counting	100 mBq/kg

TABLE I–10. OVERVIEW OF METHODS USED FOR RADIOACTIVITY ANALYSIS IN FOOD AND DRINKING WATER SAMPLES (cont.)

Radionuclide	Yield tracer/carrier	Radioanalytical technique	Measurement technique	Typical detection limit
U-238 U-235 U-234	U-232	Anion exchange chromatography Extraction chromatography (TEVA and UTEVA) Electrodeposition	Alpha spectrometry	10 mBq/kg
Th-230 Th-232	Th-229	Anion exchange chromatography Extraction chromatography (TEVA resin)	Alpha spectrometry	1–20 mBq/kg
C-14	Not needed	Combustion	Liquid scintillation counting	10 Bq/kg

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## Annex II

### STATISTICAL ANALYSES OF NATURAL RADIONUCLIDES IN FOOD

A large volume of data has been collated on natural radioactivity in food from the scientific literature and from Member States (Section 5.2). Statistical analyses have been conducted for  $^{210}\text{Po}$ ,  $^{210}\text{Pb}$ ,  $^{226}\text{Ra}$  and  $^{228}\text{Ra}$  in various foods to determine the variability of the naturally occurring radionuclides in foods and, in some cases, to derive the upper 95th percentile of the population. A full description of the statistical approach is provided in Section 5.3. The results from the statistical analyses of  $^{238}\text{U}$  and  $^{232}\text{Th}$  are not reported, given the inherent issues with the measurement techniques used (Section 5.3.3.1).

The results from the statistical analyses are presented in the tables in Section 5.3.3. Statistical analyses could not be conducted on all radionuclide–food subcategories due to time constraints, and some samples did not have a sufficient sample size for appropriate analyses. The determination of an appropriate sample size was based on a consideration of the power of the goodness of fit tests that were used to check log-normality and also on the accuracy of the estimates produced, in particular for the confidence intervals for the mean and the 95th percentile values.

The median values of the radionuclide–food subcategories where statistical analyses were not performed are outlined in Table II–1.

TABLE II-1. MEDIAN ACTIVITY CONCENTRATIONS OF RADIONUCLIDE-FOOD SUBCATEGORIES NOT INCLUDED IN STATISTICAL ANALYSES

Food subcategory	Median activity concentration (Bq/kg, fresh weight)								
	Pb-210	Po-210	Ra-226	Ra-228	Th-228	Th-230	Th-232	U-235	U-238
Beverages	0.115	0.042	0.211	0.415	4.650	—	0.025	—	0.004
Bivalves	*	*	*	*	0.471	0.545	0.326	0.031	0.824
Crustaceans	*	*	*	0.398	0.043	0.016	0.017	—	0.034
Eggs	0.090	0.069	0.109	0.090	0.043	0.249	0.005	—	0.081
Freshwater fish	*	*	*	*	0.655	0.310	2.810	2.505	0.012
Fruit	*	*	*	*	0.026	0.040	0.007	0.000	0.011
Grain	*	*	*	*	0.090	0.002	0.400	0.025	0.124
Herbs	0.569	0.882	0.326	0.180	2.900	0.557	0.058	0.003	0.091
Honey and sugar	0.107	0.230	0.049	0.188	0.142	0.014	0.002	—	0.008
Liquid milk	*	*	*	*	0.033	0.230	0.038	0.011	0.130
Meat	*	*	*	*	0.040	0.004	0.008	0.021	0.019



TABLE II-1. MEDIAN ACTIVITY CONCENTRATIONS OF RADIONUCLIDE-FOOD SUBCATEGORIES NOT INCLUDED IN STATISTICAL ANALYSES (cont.)

Food subcategory	Median activity concentration (Bq/kg, fresh weight)								
	Pb-210	Po-210	Ra-226	Ra-228	Th-228	Th-230	Th-232	U-235	U-238
Milk products	0.388	0.150	0.078	0.139	0.113	0.020	0.150	0.002	0.013
Mushrooms	*	0.580	0.300	0.260	—	0.073	0.048	0.007	0.037
Non-bivalve molluscs	*	*	0.230	0.210	—	—	0.060	—	0.549
Non-root vegetables	*	*	*	*	0.042	0.060	0.027	0.014	0.029
Nuts	0.126	0.101	0.231	0.530	12.600	0.011	9.408	—	0.006
Offal	0.520	0.719	—	—	—	—	0.026	—	0.023
Root vegetables	*	*	*	*	0.042	0.008	0.179	0.010	0.089
Saltwater fish	*	*	*	*	0.028	0.010	0.090	0.001	0.013
Seaweed	*	*	0.360	0.140	0.389	0.044	0.031	—	0.375

\* Statistical analyses conducted; see Section 5.3.3.

<sup>a</sup> —: data not available.

## **Annex III**

### **EXPLORING THE DISTRIBUTION OF $^{210}\text{Po}$ IN MOLLUSCS**

This annex provides information on statistical analyses of  $^{210}\text{Po}$  in molluscs to illustrate how most statistical analyses were conducted for different foods according to food subcategories. Activity concentrations of  $^{210}\text{Po}$  are observed to be enhanced in some marine foods, and therefore  $^{210}\text{Po}$  in molluscs was chosen as the illustrative example for this annex.

#### **III-1. BACKGROUND**

In order to support the provision of guidance related to radionuclides in food, statistical analyses were conducted on observed levels (measurements) of natural radioactivity in food (Section 5). These analyses relied on the premise that the activity concentrations of natural radionuclides in food are log-normally distributed (Section 5.3), and sufficient measurement data (the sample) are available to estimate the desired parameters of the global distribution (the population). The analysis uses a combination of graphical tools, formal hypothesis testing and final validation with empirical data to estimate various parameters of the population (median, arithmetic mean, 95th percentile) for each of three food subcategories. Further information on the statistical analysis is presented in Section 5.3.

#### **III-2. ANALYSIS OF $^{210}\text{Po}$ IN SEAFOOD**

In the modern Linnaean taxonomy system, molluscs (phylum Mollusca) comprise ~25% of all named marine organisms and represent the largest marine phylum. The phylum is divided into several taxonomic classes, with bivalves, cephalopods and gastropods being the most important food subcategories for our analyses. These subcategories can themselves be subdivided into collections of food products (Fig. III-1).

As a first step, statistical analysis was conducted for  $^{210}\text{Po}$  in all molluscs combined, that is, all of the bivalve, cephalopod and gastropod measurements. A Q-Q plot of all the log transformed measurements from the merged dataset was examined (Fig. III-2). This plot displays large deviations from the straight line at both the lower and upper ends of the sample distribution, which is a strong indicator that the data do not come from a single homogeneous population but

instead from multiple populations. As a further check on the distribution, a Kolmogorov–Smirnov test (K–S test) was conducted to determine the goodness of fit of the sample data to a log-normal distribution. The K–S test also indicated that the sample data were not representative of a log-normal distribution.

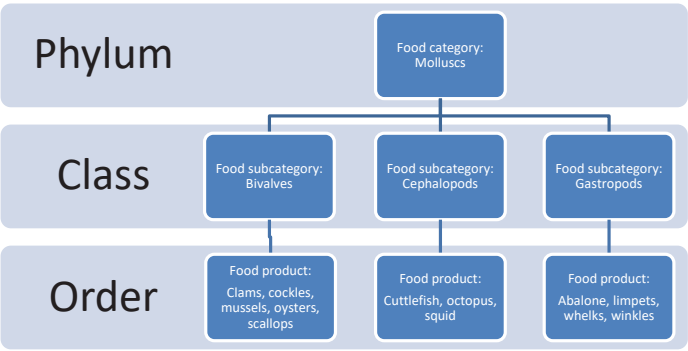


FIG. III-1. Overview of the classification of molluscs as food category–food subcategory–food product.

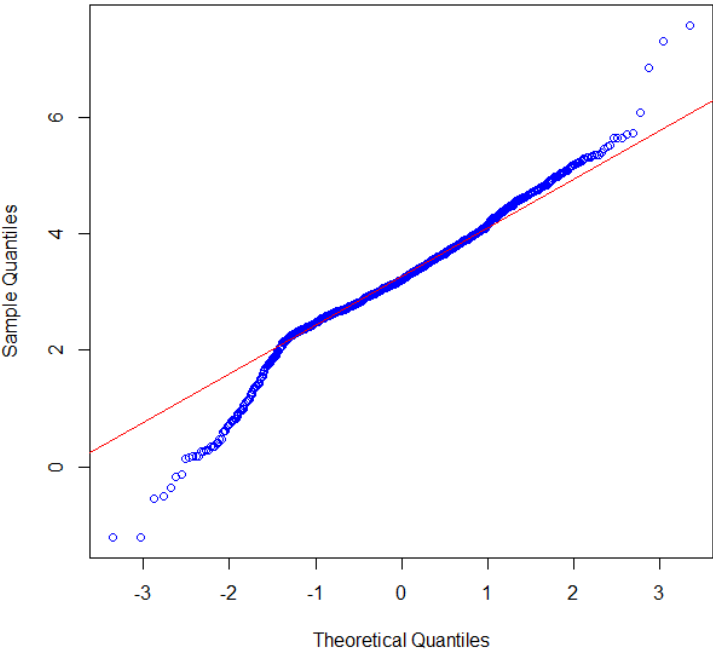


FIG. III-2. Initial Q–Q plot of  $^{210}\text{Po}$  in molluscs (bivalves, cephalopods and gastropods).

Once the merged mollusc dataset was reviewed, it was clear that these data included  $^{210}\text{Po}$  measurements for mollusc parts that were not suitable for human consumption. Further investigation of these measurements indicated that they included measurements of the viscera and shells of molluscs. Therefore, these non-edible parts were removed from the merged dataset, and the data for all molluscs were reanalysed solely for the edible parts (Fig. III-3).

The Q-Q plot indicated that the  $^{210}\text{Po}$  in mollusc measurement data were better behaved with the non-edible parts removed, especially at the upper end of the Q-Q plot. However, the K-S test result indicated a very strong deviation from a log-normal distribution. Therefore, it was concluded that no estimations of levels of  $^{210}\text{Po}$  in molluscs could be derived at the food category level.

The next step was to investigate the behaviour of the measurement data at the food subcategory levels that correspond to the class of mollusc (i.e. bivalves, cephalopods and gastropods). These statistical analyses were conducted solely on the edible parts in the merged dataset. The Q-Q plot in Fig. III-4 displays the  $^{210}\text{Po}$  in bivalve measurement data.

The Q-Q plot in Fig. III-4 indicates that the  $^{210}\text{Po}$  in bivalve measurement data are much better behaved than those outlined for molluscs in Fig. III-3. This Q-Q plot shows a smaller number of observations deviating from the line.

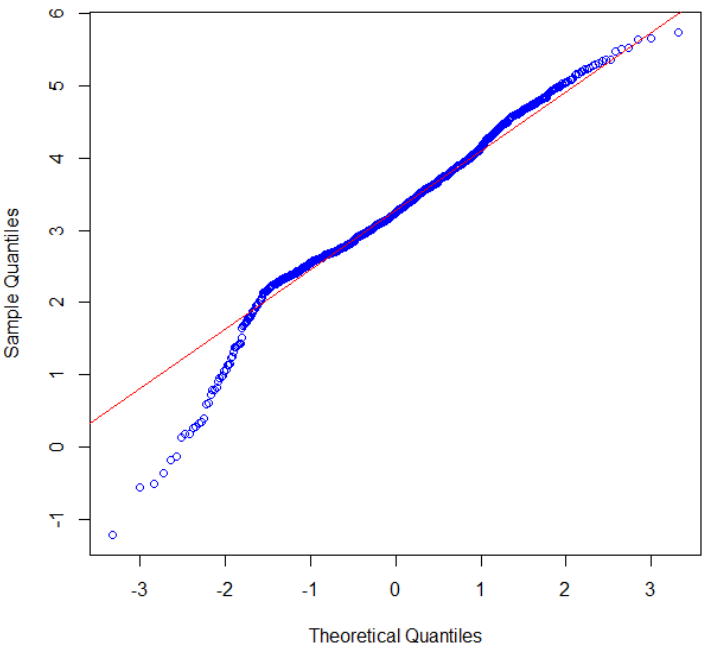


FIG. III-3. Initial Q-Q plot of  $^{210}\text{Po}$  in molluscs (edible parts only).

The goodness of fit K–S test on these bivalve data also indicated a good fit to a log-normal distribution. Therefore, estimates of the global distribution of  $^{210}\text{Po}$  in bivalves were derived using these measurement data (Table III–1).

As a further check on these estimates, the empirical percentage of observations in the sample measurements data above the estimated 95th percentile of 134 Bq/kg was 5.6%, which is in good agreement with the expected 5%.

The Q–Q plot in Fig. III–5 displays the  $^{210}\text{Po}$  in cephalopod measurement data. The Q–Q plot in Fig. III–5 shows that there are only a small number of measurement data available for  $^{210}\text{Po}$  cephalopods ( $n = 25$ ), but the sample data indicate that they are representative of a log-normal distribution, and this was confirmed using the K–S test. The estimates for the global distribution are outlined in Table III–2. The empirical percentage of observations in the sample measurements data above the estimated 95th percentile of 11.6 Bq/kg was 4%, which is in good agreement with the expected 5%, especially given the small sample dataset used to derive these estimates. While the total number of data points is relatively small ( $n = 25$ ), the results obtained from the combination of tests all support the robustness of the results.

Figure III–6 displays a Q–Q plot of the  $^{210}\text{Po}$  in gastropod measurement data. These data comprise  $^{210}\text{Po}$  measurements only in winkles and limpets, all

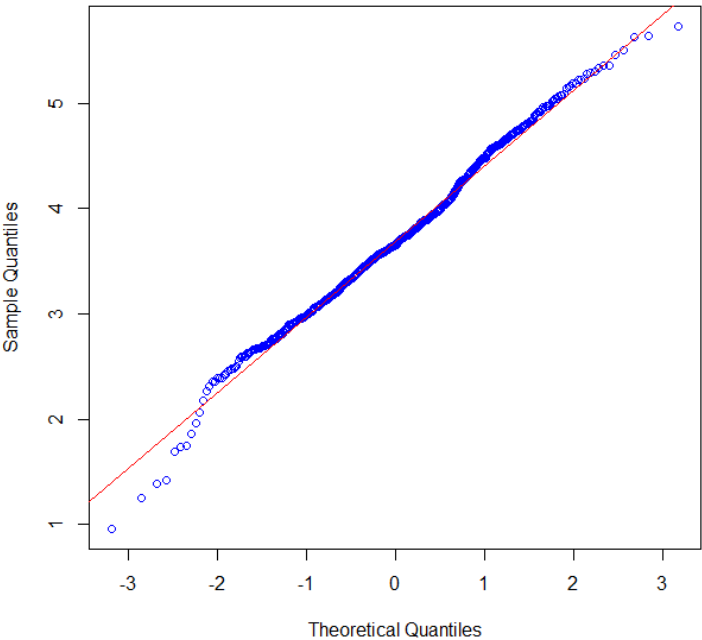


FIG. III–4. Q–Q plot of  $^{210}\text{Po}$  in bivalve molluscs (edible parts only).

TABLE III–1. MEDIAN, ARITHMETIC MEAN CONFIDENCE INTERVAL AND 95TH PERCENTILE VALUES FOR <sup>210</sup>Po IN BIVALVES

Food subcategory	Median (Bq/kg)	Arithmetic mean (Bq/kg)		95th percentile (Bq/kg)
		Lower confidence interval	Upper confidence interval	
Bivalves (clams, mussels, oysters, scallops)	40.37	49.47	55.97	134

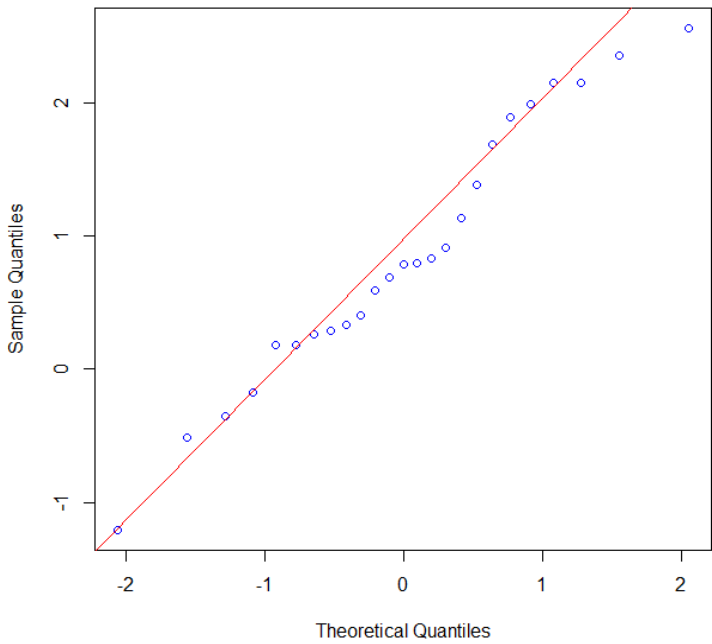


FIG. III–5. Q–Q plot of <sup>210</sup>Po in cephalopod molluscs (edible parts only).

TABLE III–2. MEDIAN, ARITHMETIC MEAN CONFIDENCE INTERVAL AND 95TH PERCENTILE VALUES FOR <sup>210</sup>Po IN CEPHALOPOD MOLLUSCS

Food subcategory	Median (Bq/kg)	Arithmetic mean (Bq/kg)		95th percentile (Bq/kg)	Notes
		Lower confidence interval	Upper confidence interval		
Cephalopods (cuttlefish, octopus, squid)	2.35	2.27	5.76	11.6	Small sample dataset

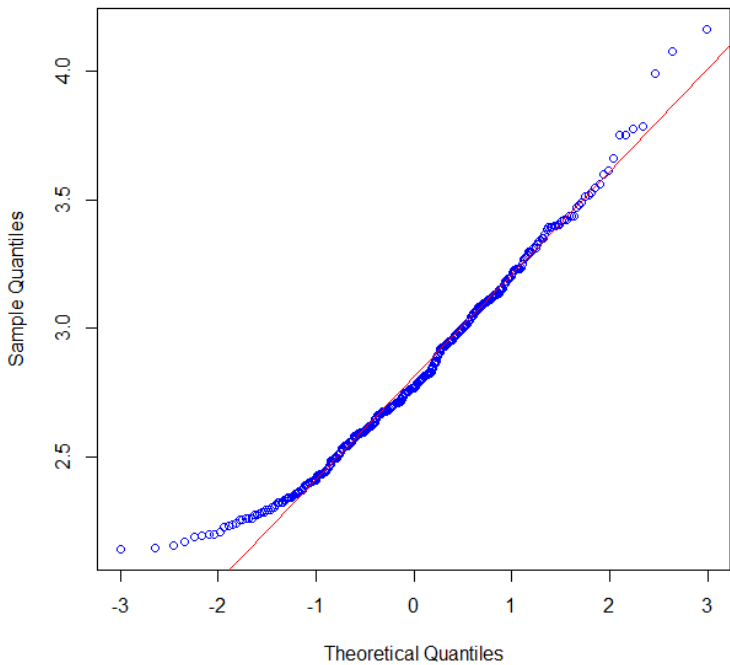


FIG. III–6. *Q–Q plot of <sup>210</sup>Po in gastropod molluscs (edible parts only).*

of which are from the UK (limited data were available from other locations). The plot indicates good agreement with a log-normal distribution. The subsequent goodness of fit K–S test confirms that the sample data are representative of a log-normal distribution. The empirical percentage of observations in the sample measurement data above the estimated 95th percentile of 32 Bq/kg was 5%, which is in agreement with the expected value. The estimates for the global distribution are outlined in Table III–3.

The statistical analyses conducted at the food subcategory level for molluscs demonstrated that the <sup>210</sup>Po measurement data were representative of a log-normal distribution, and estimates related to the global distribution of <sup>210</sup>Po in these food subcategories could be derived on the basis of the data. Therefore, it was not necessary to conduct statistical analyses at the food product level.

TABLE III–3. MEDIAN, ARITHMETIC MEAN CONFIDENCE INTERVAL AND 95TH PERCENTILE VALUES FOR <sup>210</sup>Po IN GASTROPOD MOLLUSCS

Food subcategory	Median (Bq/kg)	Arithmetic mean (Bq/kg)		95th percentile (Bq/kg)	Notes
		Lower confidence interval	Upper confidence interval		
Gastropods (winkles, limpets)	17.48	17.89	19.49	32	Only UK data available



## LIST OF ABBREVIATIONS

<i>CF</i>	concentration factor
CMS	canteen meal study
<i>CR</i>	concentration ratio
DDS	duplicate diet study
FAO	Food and Agriculture Organization of the United Nations
$F_v$	soil to plant transfer factor
GEMS	Global Environment Monitoring System
GPC	gas proportional counter
GSR	general safety requirement
ICP-MS	inductively coupled plasma mass spectrometry
ICRP	International Commission on Radiological Protection
LSC	liquid scintillation counting
MBS	market basket study
NORM	naturally occurring radioactive material
NPP	nuclear power plant
Q–Q plot	quantile–quantile plot
TDS	total diet study
UNEP	United Nations Environment Programme
UNSCEAR	United Nations Scientific Committee on the Effects of Atomic Radiation
WHO	World Health Organization



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Radionuclides of both natural and human-made origin exist throughout the environment. These radionuclides can be transferred to plants and animals that are consumed by humans, thereby resulting in exposure to ionizing radiation and an internal radiation dose. This Safety Report provides information on the observed distributions of concentrations of natural radionuclides in various food products, on the use of 'total diet' and other studies to assess ingestion doses, and on radionuclide concentrations in natural mineral waters. Different dose assessment methodologies are presented, and the advantages and disadvantages of each is discussed, along with approaches used for managing non-radioactive contaminants in food. This publication is jointly sponsored by the IAEA, the Food and Agriculture Organization of the United Nations and the World Health Organization. It is intended to support Member States in the assessment and management of radionuclides in food, and the alignment of national policies with Requirement 51 of IAEA Safety Standards Series No. GSR Part 3, related to radionuclides in food and drinking water.