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Knowledge gaps in relation to radionuclide levels and transfer to wild plants and animals, in the context of environmental impact assessements, and a strategy to fill them

Editor Justin Brown
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Abstract

International activities with regards the development of methods for assessing impacts on the environment from ionising radiation have been substantial in recent years. In developing these methods, there are requirements (i) to determine the transfer of radionuclides within ecosystems and (ii) to determine background dose-rates arising from the presence of naturally occurring radionuclides, in a satisfactory manner. It has quickly become evident that fulfilling these 2 requirements is not entirely straightforward reflecting a lack of data in many cases. This report specifies exactly where these data-gaps lie through analyses of data generated from the most recent studies conducted internationally on this topic. It is evident that information is limited for numerous radionuclides from U-238 and Th-232 decay series and notably, in view of its importance as a contributor to dose-rates in plants and animals, Po-210. The simple way to rectify these data deficiencies is to organise target field campaigns focusing on particular species and radionuclides where information is lacking. To this end, field sampling has been conducted in a semi-natural mountain ecosystem in Norway and freshwater aquatic systems in Finland. It is envisaged that the data derived from the studies briefly described in this report will provide fundamental information for our understanding of the behaviour and fate of natural decay series radionuclides in terrestrial and aquatic systems and provide the basis for more robust way.

Key words

Transfer, non-human biota, impact assessment

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Knowledge gaps in relation to radionuclide levels and transfer to wild plants and animals, in the context of environmental impact assessements, and a strategy to fill them

A report for NKS activity

GAPRAD

Filling knowledge gaps in radiation protection methodologies for non-human biota

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1. Introduction and background

In recent years, there has been significant effort linked in to the development of frameworks to explicitly quantify impacts on the environment arising from exposure by ionising radiation. The former position of the radiological community as paraphrased in the principle "*by protecting man from the effects of ionising radiation, the environment is automatically protected*" (ICRP, 1977; 1991) was arguably untenable (Pentreath, 1998). The issue of how to consider radiation impacts on the environment is, admittedly, not entirely new and publications exist in the open literature dealing with themes related to this topic (IAEA, 1979; IAEA, 1988; NCRP, 1991; UNSCEAR, 1996). Furthermore, the inclusion of references to the protection of the environment in numerous international conventions, principles and statements of intent (e.g. AEPS, 1991; IAEA, 1995; UNCED, 1992; OSPAR, 1998) has augmented pressure for the introduction of an approach that can be used to explicitly assess the impact of ionising radiations on the environment. This background information stimulated some countries to take steps in response to environmental protection legislation by providing guidance on environmental impact assessments for ionising radiation (Copplestone *et al*., 2001; USDoE, 2002). However, there has been considerable divergence in structure and content of these methodologies, for example with respect to transfer data and models incorporated, dosimetric models employed and endpoints of concern. For the sake of clarity, it has become increasingly apparent that a structured, internationally recognised framework for assessing the impacts of radioactivity explicitly for the environment, was required. Indeed, a number of publications within the last decade (Pentreath 1998; Pentreath, 1999; Strand *et al*., 2000; Strand & Larsson, 2001) have called for the development of such a framework. A system of this kind would allow the considerable volume of available data to be organised in a systematic manner.

Within the last few years, the International Commission on Radiological Protection has begun to formulate its thoughts concerning protection of the environment (ICRP, 2003) and it is evident that initial considerations with respect to a framework for environmental protection will be included in the Basic Recommendations of the ICRP following the approval of the report in March 2007. The culmination of recent activities on the development of a protection framework for ionising radiation are the reports of the ICRP as initially considered by ICRP (2003) and subsequent draft documents thereafter. The Commission's framework is being designed so that it is harmonised with its proposed approach for the protection of human beings. To achieve this, an agreed set of quantities and units, a set of reference dose models, reference dose-per-unit-intake data, and reference organisms are in the process of being developed. As a first step, a limited number of reference fauna and flora (RAPs) has been proposed by the Commission. The documents of the ICRP are likely, as is the case with other areas of radiation protection, to from the seminal reference with the view that others can then develop more area-and situation-specific approaches to assess and manage risks to non-human species.

The international Atomic Energy Agency (IAEA) has organised a working group to consider the theme of protection of non-human biota to radiation. The *Biota Working Group* was formed by the IAEA as part of the Environmental Modelling for Radiation Safety (EMRAS) programme in 2004 with the main objective: '*to improve Member State's capabilities for protection of the environment by comparing and validating models being used, or developed, for biota dose assessment (that may be used) as part of regulatory process of licensing and* *compliance monitoring of authorised releases of radionuclides'*. The intercomparison exercises have generally focussed on the transfer component of the assessment, i.e. deriving activity concentrations in selected biota from a given media concentrations and aspects of dosimetry, i.e. deriving absorbed dose-rates to selected species from internal and external distributions of radionuclides. The results of these exercises are considered elsewhere (Vives i Batlle, in press; Beresford et al., in press). The findings from this work and any subsequent activities are likely to form an input to the revision of the Agency's Basic Safety Standards. Nonetheless, the focus of the inter-comparison, especially in relation to the analysis of transfer, has largely pertained to temperate environments reflecting data availability, i.e. transfer data for Arctic environments are less numerous than those for temperate environments and have thus not be considered within case studies. The applicability of the findings from the analyses conducted within the Biota working group to the Arctic is therefore constrained.

At a regional level, methodologies to assess the impact of exposure to ionising radiation on flora and fauna in European temperate and Arctic environments have been developed in two European collaborative projects "FASSET - Framework for Assessment of Environmental Impact" (Larsson *et al.*, 2004) and "EPIC - Environmental Protection from Ionizing Contaminants in the Arctic" (Brown *et al.*, 2003a) respectively. These studies have been superseded by the project "ERICA - Environmental Risk from Ionising Contaminants: Assessment and Management" wherein risk assessment methodologies have been developed and issues relevant to decision making in the context of the management of environmental impacts of radioactivity have been addressed (Beresford et al., 2007a).

1.1. Objectives

The objective of this work was to provide and overview of the coverage of information available in relation to radionuclide levels (for natural radionuclides) and transfer in the environment, within the context of established environmental impact assessment frameworks. In this way, knowledge gaps can be easily identified. Once this initial step had been taken the second objective was to formulate a strategy concerning how these information gaps might be filled, thereby providing a roadmap for a further study within this NKS Research Project.

2. Assessing impacts in the environment

2.1 Components of the environmental impact assessment

The stages in the assessment methodologies developed within the projects FASSET and EPIC are depicted in Figure 1. The initial stage of the assessment requires the selection of radionuclides and of appropriate reference biota and suitable representative organisms (normally defined at the species level) with concomitant ecological information relevant to dose-rate calculation. Following these steps, the exposure assessment is conducted using the basic methodology outlined below. Methods for deriving the transfer and fate of radionuclides in ecosystems are necessary during this procedure as are methods for deriving (weighted or unweighted) dose-rates. Once exposures for reference biota have been derived, they need to be interpreted in terms of biological effects.

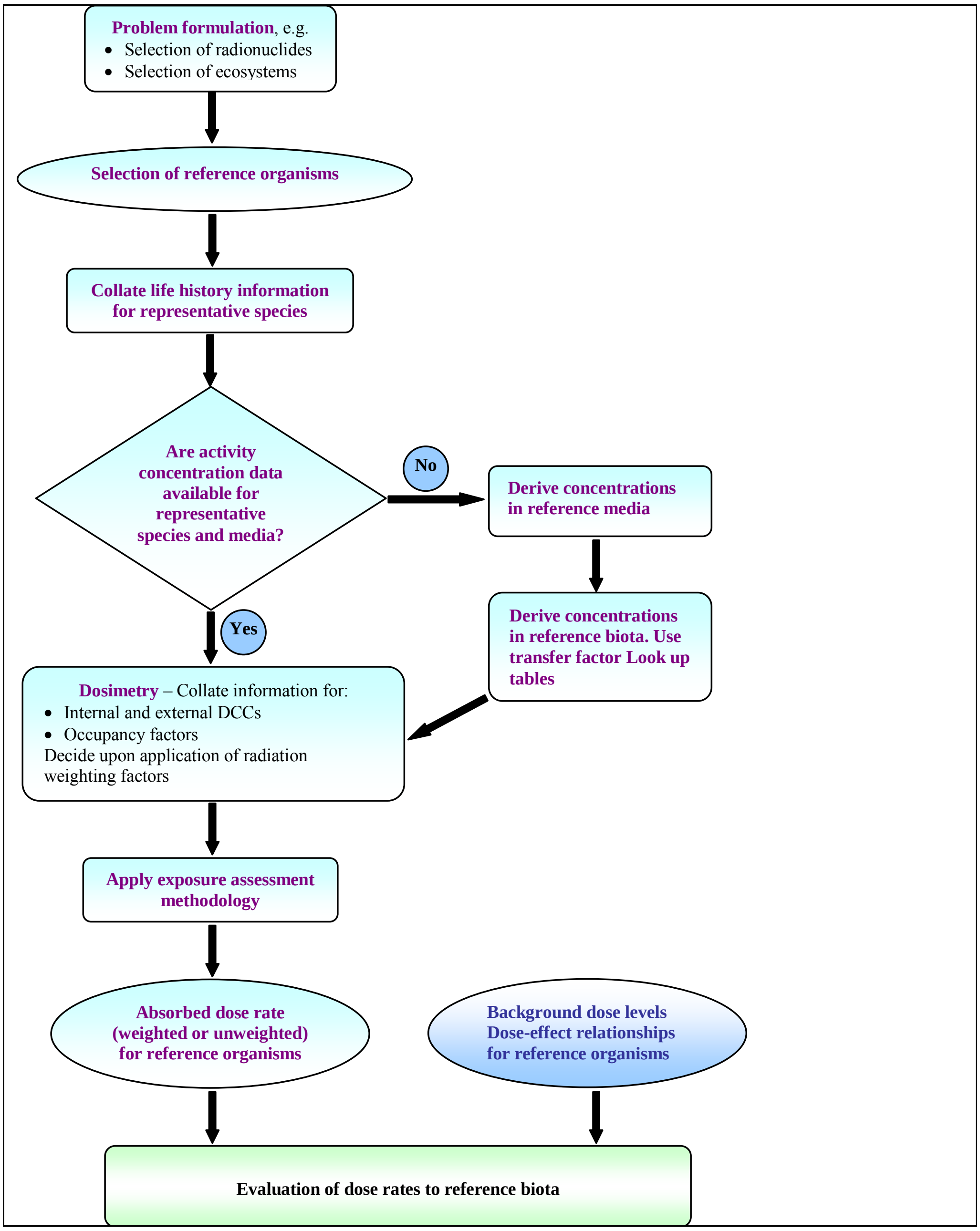

Figure 1. Flow diagram showing stages in the proposed exposure assessment (adapted from Hosseini et al., 2006).

The figure shows that the derivation of transfer is an important component of the assessment as is a reference database characterising background dose-rates.

2.2 Reference organisms

Most, if not all, established methods of assessing impacts of ionising radiation on the environment use points of reference which allow available data to be organised and from which information can be intepolated and/or extrapolated. Central to this type of approach is the concept of reference biota as defined below from the ERICA project :

Reference organisms are a series of entities that provide a basis for the estimation of radiation dose rate. These estimates, in turn, provide the basis for assessing the likelihood and degree of radiation effects to a range of organisms which a re typical, or representative of a contaminated environment.

This can be directly compared with the ICRP's definition for Reference animals and plants:

A **Reference Animal or Plant (RAP)** is a hypothetical entity, with assumed basic characteristics of a specific type of animal or plant, as described to the generality of the taxonomic level of the Family, with precisely defined anatomical, physicological and life history properties that can be used for the purposes of relating exposure to dose and dose to effects for that type of living organism.

The ERICA approach differs slightly in the sense that that ICRP's selected organisms

- are described specifically at the taxonomic level of the family and
- (will) have precisely defined anatomical, physiological and life-history properties.

Nonetheless, it should be noted that there is broad consistency between the reference organism approach adopted by FASSET/ERICA and the RAPs defined by the ICRP. More importantly, the methodology developed within ERICA will be applicable for the ICRP RAPs, i.e. will allow assessments to be conducted for the ICRP RAPs.

Reference organisms provide a means of reducing the assessment to manageable proportions and may allow logical links/associations between sets of data attributed to different organism types to be established. In this way some insight into the potential environmental impacts of ionising radiation may be derived for components of the environment for which data are poor or absent. The reference organism approach has been advocated in a number of earlier publications (Pentreath, 1999; Pentreath & Woodhead, 2000; Strand *et al.* 2000) where it has been argued that (see Pentreath, 1999) an attempt should not be made to model everything but that models should be selected based on an appreciation of the actual and potential data that are likely to become available, and pre-existing information concerning the effects of geometry, the behaviour of radionuclides in the environment and the behaviour of the organisms.

The lists of reference organisms, as considered by ERICA, for freshwater and terrestrial ecosystems are shown in Tables 1 and 2. ICRP RAPs (with definitions in parentheses) are in bold, grey-tone squares:

Table 1. Terrestrial reference organisms

Soil Invertebrate (earthworm)	Detritivorous invertebrate	**Flying insects (Bee)**
Gastropod	Lichen & bryophytes	**Grasses & Herbs (Wild grass)**
Shrub	**Tree (Pine tree)**	**Mammal (Rat)**
Mammal (Deer)	**Bird (Duck)**	**Bird egg (Duck egg)**
Reptile	**Amphibian (Frog)**	

Table 2. Freshwater reference organisms

Phytoplankton	Vascular plant	Zooplankton
Insect larvae	Bivalve mollusc	Gastropod
Crustacean	Benthic fish	**Pelagic fish (Salmonid/trout)**
Bird (Duck)	Mammal	**Amphibian (Frog)**

For utilisation within the impact assessment process, each (ERICA) reference organism has been assigned default attributes relating to radioecology and dosimetry, these being:

- Equilibrium concentration ratios
- Default occupancy factors
- Default ellipoisal geometries (with the 3 primary axes defined) allowing dose-conversion factors to be defined.

2.3 Transfer of radionuclides

Exposure of biota to radiation and transfer of radionuclides in the environment, are intimately linked. Exposure of biota to ionising radiation occurs when radionuclides, present naturally in the environment or released through man's activities, decay releasing radiation of various types and energies. The pathways leading to exposure in aquatic and terrestrial ecosystems can be split into several categories:

(1) Inhalation of (re)suspended contaminated particles or gaseous radionuclides. This pathway is relevant for terrestrial animals and marine birds and mammals.

(2) Contamination of fur, feathers and skin. This has both an external exposure component, e.g. β and γ-emitting radionuclides on or near the epidermis cause irradiation of living cells beneath and an internal exposure component as contaminants are ingested and incorporated into the body of the animal.

(3) Ingestion of lower trophic plants and animals. This leads to direct irradiation of the digestive tract and internal exposure if the radionuclide becomes assimilated and distributed within the animal's body.

(4) Direct uptake from the water column, in the case of truly aquatic organisms (e.g. fish, molluscs, crustaceans), leading to direct irradiation of respiratory system, e.g. gills, and internal exposure if the radionuclide becomes assimilated and distributed within the animal's body.

(5) Intake of water contaminated by radionuclides through the gastrointestinal tract, i.e. the organism drinks water. The same exposure categories as discussed in (3) are relevant here.

(6) External exposure. This essentially occurs from exposure to γ-irradiation and to a much lesser extent β-irradiation, originating from radionuclides present in the organism's habitat. For microscopic organisms, irradiation from α-particles is also

possible. The configuration of the source relative to the target clearly depends on the organism's ecological characteristics and habitat. A benthic dwelling fish will, for example, be exposed to radiation from radionuclides present in the water column and deposited sediments, whereas a pelagic fish may only be exposed to the former.

In the context of European approaches (EPIC, FASSET and ERICA), inhalation and contamination of fur, feathers and skin (exposure pathways (1) and (2) in the above list) have not been considered explicitly in the derivation of transfer parameters or dose-conversion coefficients. The ingestion and direct uptake from water pathways (points (3) and (4) in the above list) have been considered in so far as they relate to internal body burdens of contaminants normally under equilibrium conditions. Irradiation by unassimilated contaminants in the gastrointestinal tract has not been considered nor has exposure occurring due to the consumption of water (point (5) above). Finally, external exposures have been considered in some detail both in terms of contaminant transfer to terrestrial and aquatic habitats and from the dosimetric perspective, the latter having been described elsewhere (Pröhl *et al.*, 2003). An example of how exposure is conceptualised for the aquatic environment is shown in Figure 2.

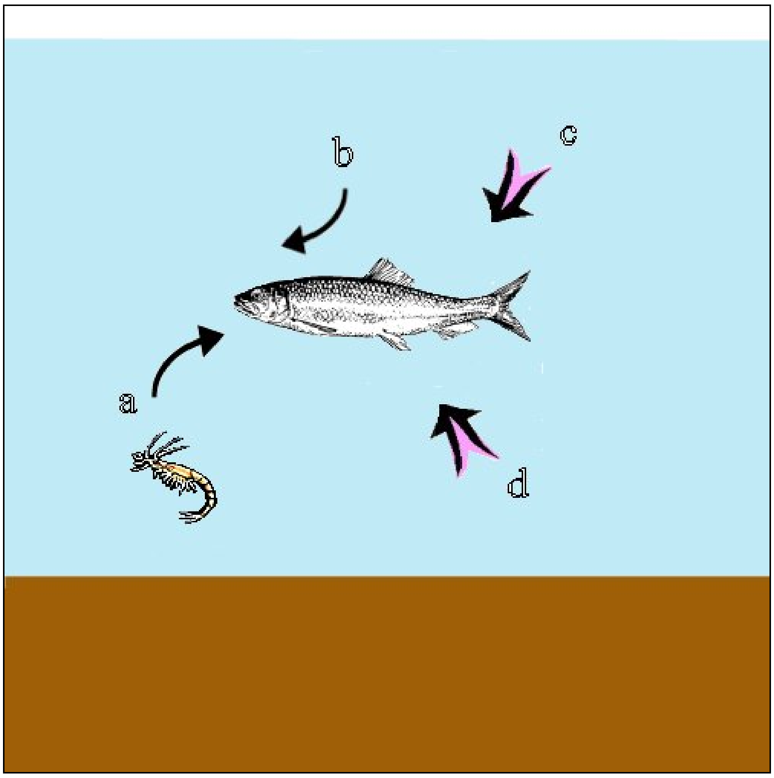

Figure 2. Exposure pathways for aquatic organisms as considered by FASSET. (a) Internal exposure via ingestion of contaminated food and assimilation; (b) internal exposure via direct uptake from the water column; (c) external exposure directly from radionuclides in the water column; (d) external exposure from radionuclides in sediments.

The basis for considering internal burdens of radionuclides (thus allowing internal dose-rates to be derived) is simply derived in many Environmental impact assessment (EIA) approaches by considering integrated parameters that account for all transfer pathways. In this respect, activity concentrations of radionuclides in biota are often predicted from media activity concentrations using equilibrium concentration ratios (CRs). The definitions of CR are:

Terrestrial

$$CR = \frac{Activity\ concentration\ in\ biota\ whole\ body\ (Bq\ kg^{-1}\ \ fresh\ weight)}{Activity\ concentration\ in\ soil\ (Bq\ kg^{-1}\ dry\ weight)}$$

Aquatic

$$CR = \frac{Activity\ concentration\ in\ biota\ whole\ body\ (Bq\ kg^{-1}\ \ fresh\ weight)}{Activity\ \ concentration\ of\ \ filtered\ water\ (Bq\ l^{-1}\ )}$$

Data pertaining to activity concnetrations in organisms and concomitant data on activity concentrations in environmental media are therefore fundamental to the derivation of these parameters.It is the collation of such information that consititutes the basis for this review.

2.4 Placing the calculated dose-rate in context : quantifying Background

Arguably, two points of reference may be used for the purpose of assessing the potential consequences of exposures to radiation on non-human biota. These are (a) natural background dose rates and (b) dose rates known to have specific biological effects on individual organisms (Pentreath, 2002). With respect to the former reference point, a specific task within the EU Fifth Framework “FASSET” project has been to assess doses, arising from naturally-occurring radionuclides, for selected organisms inhabiting European environments.

Bands of derived consideration levels for reference fauna and flora could be compiled by combining information on logarithmic bands of dose rates relative to normal natural background dose rates, simply as a means of presentation, plus information on dose rates that may have an adverse effect on reproductive success, or result in early mortality (or cause morbidity), or are likely to result in scorable DNA damage for such organisms (ICRP, 2003). Such a banding could be essentially on the same basis as previous proposals made for humans (ICRP, 2001b), in that additions of dose rate that were only fractions of their background might be considered to be trivial or of low concern; those within the normal background range might need to be considered carefully; and those that were one, two, three or more orders of magnitude greater than background would be of increasingly serious concern because of their known adverse effects on individual fauna and flora (Pentreath, 2002).

Using natural background as a point of reference is not without controversy. Although the ICRP originally proposed to manoeuvre the radiological protection of man, more specifically when setting levels of individual dose above which a requirement to take all feasible steps to reduce doses would be incumbent, in the direction of using bands of concern based on background dose-rates. However, in the most recent version of the draft recommendations (as of the Summer of 2006) the Commission appear to have distanced themselves from this approach, applying the concept of dose constraints and dose limits but without reference to natural background radiation. This being said the consideration of natural background in implicitly included in the setting of constraints, e.g. in the constraint band under 1 mSv y^{-1},

"*…corresponding doses would represent a marginal increase above the natural background…*". Furthermore, the application of the Linear-non-threshold hypothesis[1], augments the requirement for well defined background dose-rates to be characterised for man.

With respect to protection of the environment, a final decision with regards the application of derived consideration levels was still pending at the time of writing of this report. Irrespective of whether a decision is made against the use of such an approach, the importance of characterising background dose-rates for reference plants and animals is substantial if purely for the sake of allowing incremental dose rates to be defined more precisely.

3. Review of existing information

The method applied for this report has simply been to access databases and reviews conducted recently in the context of the devlopment of environmental impact assessment approaches. The focus of the work has been on natural decay series radionuclides (^{238}U and ^{232}Th decay series radionuclides with half-lives > 10 days[2])

This has entailed :

- Summary of information from the ERICA project: 3 empirical databases have been collated within the ERICA project with respect to terrestrial, freshwater and marine ecosystems. These data have been analysed in this study to provide an overview as to where datagaps exist.
- Analyses of other recently publsihed data, e.g. Beresford et al., (2007b)

It is important to note that there are 2 distinct sets of information embedded in this analyses. The first of these pertain simply to activity concentrations of naturally occuring radionuclides in plants and animals. An improved availability of such data allow background dose-rates to be derived in a more robust manner. The second set of data are concentration ratios, the use of which allows transfer from environmental media (i.e. soil, water) to plants and animals to be modelled. With regard this second information source, there are some pertinent points that warrant discussion.

When reviewing studies reported in the open literature it quickly becomes apparent that data are provided in quite different formats reflecting the *ad hoc* nature of normal research programmes. For example, biota and soil activity concentrations may be presented in relation to ash, dry or wet weight. In the process of deriving representative values, all data must be normalised using appropriate conversion factors (see Beresford et al., submitted; Hosseini et al., submitted). For example when data are presented as activity per unit dry weight of the organism either direct empircal data from the study or assumption concerning the dry:wet weight ratio need to be applied in order to convert all measruements to an activity per unit wet

[1] A hypothesis which is based on the concept that, in the low dose range, above background, radiation doses greater than zero will increase the risk of excess cancer and/or heritable disease in a simple proportionate manner,

[2] This half-life cutoff has been selected owing to the fact that radionuclides with half-lives < 10 days have been included in the dose-conversion coefficients (DCC) of their parent radionuclides. In other words secular equilibrium with the parent is assumed and no explicit transfer or DCC for these particular radionuclides are required.

weight. In some cases within the terrestrial environment, aggregated transfer data are presented wherein activity concentrations in organisms are related to the deposition (in units of Bq per unit area). In this case a conversion needs to be performed accounting for the density of soil in order to derive a corresponding CR. Finally, activity concentration data for fauna are often presented in relation to a particular organ or body part. Once more data need to be normalised converting to an equivalent activity concentration per whole body. This involves the use of information concerning the percentage by mass of selected organs within the body and the distribution of the studied radioelement within the body. As such information is not always readily available performing this "whole-body" conversion is not always practicable.

4. Results + discussion

The CR empirical data coverage for selected radionuclides provided by the ERICA project for terrestrial and feshwater environments is presented in Table 3 and 4 respectively.

For the terrestrial environment, Caesium represents a radioelement for which data coverage is comprehensive. Most reference organisms are characterised by >20 sample values and where this is not the case, at least some empirical information is available. The coverage for Pb is reasonable presumably reflecting the large number of stable element studies that have been conducted on this element. Other radioelements are more poorly characterised with empirical data sets. In the case of Polonioum, some information is available for flora but only for the fauna group mammals. In the latter case it should be noted that although 36 data are available these represent "all mammals" from a single geographical area - the UK. The number of values associated with Thorium is low. In all cases the number of available empirical values is below 20 and for 7 reference categories no information is available at all. A similar situation esists for Uranium although arguably floral reference organisms are endowed with reasonable CR information. For radium there are severe data deficiencies for invertebrates, insects, amphibia and reptiles.

Table 3. CR data coverage for terrestrial reference organisms adapted from Beresford et al. (submitted). The grey cells denote – no data.

Element	Cs	Pu	Po	Ra	U	Pb	Th
Soil Invertebrate (Worm)	N ≤ 20	N ≤ 20			N = ?	N > 20	
Detritivorous invertebrate	N > 20	N > 20		N = ?		N > 20	
Flying insects	N > 20	N > 20				N ≤ 20	
Gastropod	N ≤ 20	N ≤ 20		N ≤ 20		N > 20	
Lichen & bryophytes	N > 20		N ≤ 20	N ≤ 20	N = ?	N > 20	N ≤ 20
Grasses & Herbs	N > 20	N > 20	N > 20	N > 20	N > 20	N > 20	N ≤ 20
Shrub	N > 20		N ≤ 20	N ≤ 20	N > 20	N > 20	N = ?
Tree	N > 20		N ≤ 20	N ≤ 20	N > 20	N > 20	N > 20
Mammal (Rat)	N > 20	N > 20	N > 20	N > 20	N ≤ 20	N > 20	N ≤ 20
Mammal (Deer)	N > 20	N > 20	N > 20	N > 20	N ≤ 20	N > 20	N ≤ 20
Bird	N > 20			N > 20	N = ?	N > 20	N = ?
Bird egg	N = ?						
Reptile	N ≤ 20						
Amphibian	N > 20					N > 20	

In the freshwater environment, radiocaesium again represents a radioelement for which data coverage is fairly extensive. Even in this most favourable case there are data gaps notably for insect larvae and aquatic mammal. The data coverage for Pb is extremely poor – no data are available for any reference organism although it may be assumed that an extended review of stable element data might lead to the extraction of at least some information to mitigate this situation. CR values for Th are limited to a small number of data for fish and vascular plant. Although coverage of U and Po is slightly improved on this there are conspicuous data gaps including one for aquatic birds, mammals and insect larvae. Furthermore, there are no reported data for Po in benthic fish or U in bivalve mollusc. Data coverage for Ra and Pu is of a similar magnitude. Pu CR values in fish appear to be well characterised but in all other cases the number of samples associated with an empirically derived representative value is less than 20.

Table 4. CR data coverage for freshwater reference organisms adapted from Hosseini et al. (submitted). The grey cells denote – no data.

Element	Cs	Pu	Ra	Po	U	Th	Pb
Benthic fish	N≥20	N≥20	10<N<20		10<N<20	N<10	
Pelagic fish	10<N<20	N≥20	10<N<20	10<N<20	10<N<20	N<10	
Vascular plant	N≥20	N<10	10<N<20	N<10	N<10	N<10	
Bivalve mollusc	10<N<20		N<10	N<10			
Insect larvae							
Phytoplankton	10<N<20	N<10	N<10	N<10	N<10		
Amphibian	N<10						
Crustacean	N<10	N<10	N<10	N<10	N<10		
Gastropod	N<10		N<10	N<10			
Zooplankton	N<10	N<10			N<10		
Bird	N<10						
Mammal							

The Environment Agency of England and Wales recently commissioned work to develop databases to underpin environmental impact assessment (Beresford et al., 2007b) using reference animals and plants. The overall aim of the work was to determine the activity concentration ranges of naturally occurring radionuclides in non-human species and to estimate the background radiation dose rates in terrestrial and freshwater ecosystems across England and Wales. The specific aims were to:

- review and collate data from existing literature to establish the activity concentrations of naturally occurring radionuclides from the ^{238}U and ^{232}Th series and ^{40}K in environmental samples including soils, sediments, waters, and particularly non-human species (wild animals and plants) for terrestrial (including coastline) and freshwater ecosystems.
- identify gaps in the collated data, particularly for non-human species, and to design and carry out a sampling and analytical campaign to partially fill these data gaps.

Data relevant to the activities planned in GAPRAD were presented in this report as exemplified in Table 5.

Table 5. Numbers of samples for which ^{40}K, ^{232}Th series and ^{238}U series radionuclide activity concentrations in UK biota were compiled from the literature/in-house sources. Available total U data are also indicated. Note the numbers include measurements reported as below detection limits (reproduced from Beresford et al., 2007b).

RAP	^{40}K	^{210}Po	^{210}Pb	^{226}Ra	^{228}Th	^{230}Th	^{232}Th	^{234}U	^{235}U	^{238}U	Total U
Duck	27										
Frog											
Bee											
Earthworm											
Pine tree	2	5	5	5		5	5	9	8	9	
Wild grass	218	117	147	123		4	151	160	157	160	163
Brown seaweed	13	92	35		10	10	23	19	15	58	
Trout	44										
Total mammal*	141	28	31			2	32	4	4	3	3
Deer	72	1									
Rat											

*Total mammal includes available deer and rat data in addition to that for other mammalian species.

As can be seen from this overview of data, there are no data for some RAPS, notably frog, bee earthworm and rat and very few data for some other groups, notably duck (^{40}K only) and deer (^{40}K, 1 data point for ^{210}Po). In order to address this numerous samples were measured predominately for U and Th. New data were generated for, inter alia, ducks, trout and insects thus providing some new information to fill data gaps albeit specifically for the UK environment. No new measurement of ^{210}Po were made in the study.

Brown et al. (2004) reported background dose rates for aquatic environments (Table 6). Although this was possible for a comprehensive suite of reference organisms in the freshwater environment, it was noted that "*published data on natural series radionuclides in freshwater organisms are sparse and no reference citing data specific for Europe have been identified*." With this being the circumstance for the assessment, it was necessary to revert to transfer data published for other parts of the world, notably India.

A two stage process was adopted whereby initially activity concentration s of naturally occurring radionuclides in freshwater environments were collated (Table 6) and thereafter CR data were applied (Table 7) to derive activity concentrations of naturally occurring radionuclides in biota. The need to adopt such and approach clearly demonstrated the lack of available information for European freshwater environments.

Table 6. Radionuclide concentrations in freshwater (reproduced from Brown *et al*., 2004)

	Concentration (Bq m^{-3})				
Nuclide	Global range[a]	Global average[b]	UK Lake district[c]	Spain Rio Ebro[d]	India R. Kaveri[e]
^{40}K	4–240	26		119 (42–336)	
^{210}Po	0.3–9	2			1.2
^{222}Rn	7–7000	800	285 (75–1040)		
^{226}Ra	0.5–100	5	5.6 (2.1–15)	29 (20–43)	0.93
^{228}Ra	—	0.4			
^{230}Th	—	2.6			
^{232}Th	0.04–0.4	0.11			
^{238}U	0.2–63	11	8.4 (4.0–17.4)	13 (1.9–89)	

[a] IAEA (1976).
[b] Santschi and Honeyman (1989).
[c] Al-Masri and Blackburn (1999).
[d] Pujol and Sánchez-Cabeza (2000).
[e] Shaheed *et al* (1997), Hameed *et al* (1997).

Table 7. Concentration factors for freshwater organisms (reproduced from Brown *et al*., 2004).

	Concentration factor ($m^3\ kg^{-1}$) (wet weight)					
Nuclide	Phytoplankton	Macrophyte	Mollusc	Crustacean	Pelagic fish	Benthic fish
^{40}K	4.0E+00	4.0E+00	2.0E+00	2.0E+00	6.0E−01	6.0E−01
^{210}Po	2.5E+01	1.5E+00	5.0E+01	1.0E+01	3.0E−01	1.0E+01
^{222}Rn	8.0E−04	8.0E−04	8.0E−04	8.0E−04	8.0E−04	8.0E−04
^{226}Ra	1.0E+00	2.0E+00	3.0E−01	1.0E+00	3.0E−01	1.2E+00
^{228}Ra	1.0E+00	2.0E+00	3.0E−01	1.0E+00	3.0E−01	1.2E+00
^{230}Th	1.0E+00	1.0E+00	5.0E−01	2.5E−01	2.0E−02	4.0E−02
^{232}Th	1.0E+00	1.0E+00	5.0E−01	2.5E−01	2.0E−02	4.0E−02
^{238}U	1.0E−01	4.0E+00	1.0E−01	1.5E−01	1.5E−02	2.5E−01

In view of the numerous data gaps that are evident with regards the levels and transfer of radionuclides in environmental systems, approaches to circumvent these deficiencies have been developed by some authors. The method used to fill knowledge gaps recommended by Bereford et al. (submitted) uses available related information, categorised into 3 approaches ranging from most preferred to least preferred options. The options used to provide default CR values, when values could not be derived from the literature, were:

1. Use an available CR value for an organism of similar taxonomy within that ecosystem for the radionuclide under assessment (preferred option).
2. Use an available CR value for a similar reference organism (preferred option).
3. Use CR values recommended in previous reviews or derive them from previously published reviews (preferred option).
4. Use specific activity models for ^{3}H and ^{14}C (preferred option).
5. Use an available CR value for the given reference organism for an element of similar biogeochemistry.
6. Use an available CR value for biogeochemically similar elements for organisms of similar taxonomy.
7. Use an available CR value for biogeochemically similar elements available for a similar reference organism.
8. Use allometric relationships, or other modelling approaches, to derive appropriate CRs.
9. Assume the highest available CR (least preferred option).

Furthermore, combinations of these options can be applied.

Clearly this approach has some merit especially if elucidated through the consideration of applicable specific cases. For example, information on activity concentrations or transfer of natural radionuclides for benthic fish might be approximated using information on pelagic fish. Although the validity of applying such approaches may be reasonably argued in some cases, their applicability in others may be open to question. In view of the behaviour of ^{210}Pb (and its granddaughter Po-210), excess levels deposited via dry and wet deposition following decay via a number of intermediate short-lived radionuclides from gaseous ^{222}Rn, using an element of similar biogeochemistry (e.g. Te Gp VIb for Po-210) might not provide valuable insight to the actual environmental distributions owing to the particular input pathway for this radioelement. Care might also be required in applying a taxonomic analogue approach. For example, it would be erroneous to use CR or activity concentration data pertaining to ^{210}Po for reindeer as a surrogate for a related member of the family cervidae, e.g. deer, because the strong dependence on lichen as a foodstuff for reindeer leads to unusually elevated activity concentrations of polonium in these animals (MacDonald et al., 1996). This in turn reflects the capacity of lichen to adsorb atmospherically derived contaminants. A taxonomic similarity may therefore not be enough to justify application of the method when other factors such as diet more strongly affect the final biotic distribution.

5. Strategy to fill knowledge gaps

It is evident that some of these data deficiencies could be easily mitigated with limited, but focussed, effort involving field-work and analysis. The following activities have been identified where a small effort will reap great dividends, these involve determination of

- Po-210 in soil fauna, small mammals and soil
- Natural radionuclides (U-238, U-234, Ra-226, Ra-228, Po-210, Pb-210) in fish brackish waters and sediments

Linking the sampling work to national monitoring campaigns should limit associated costs.

In view of the fact that it is also important to understand the underlying mechanisms influencing the environmental transfer of natural radionuclide because this will allow us to interpret the data in a more meaningful way, studies will be tailored to fit with this aspiration as far as practicable. For example, it is planned that work will also be conducted to understand Po cycling in a lake system with elevated humic acid content.

The role of skeletal ^{210}Pb decay on subsequent soft tissue ^{210}Po concentrations in mammals is still a matter for some debate (Skuterud *et al*., 2006). In order to elucidate some of the points arising from this contention, studies will be performed to attain information on gastrointestinal update and residence time in mammals (using "man" as the reference species). Such data should add to our understanding of how ^{210}Pb and ^{210}Po are transferred in environmental systems.

A number of possible problems will need to be addressed in fulfilling the overall aims of the strategy these include:

- Time required for analyses: in particular the requirement to recount samples after a period of 6 months in order to establish the level of Pb-210 in samples. Without this information it is not possible to accurately determine the level of unsupported Po-210 originally in the sample.
- Sample digestion – the perchloric acid – wet digestion method is probably the most effective means of totally digesting environmental samples but the procedure is unfortunately hazardous.
- Low activity levels in samples, Beresford et al. (2007b) reported levels of approximately 0.084 Bq kg^{-1} fresh weight for the group mammals (excluding reindeer). These low activity levels mean that quantification of Po-210 in single biological samples that may, for example in the case of mice, weigh a little as a few 10s of grams may be problematic

- Po-210 is known to accumulate in the liver of mammals such as reindeer (Skuterud *et al*., 2006) and the hepatopancreas or liver of other organisms (see Hosseini et al., submitted). This means that some case is required in selecting tissues for analyses. In the context of environmental impact assessments, the whole-body activity concentration is normally the variable that requires consideration. If activity concentrations in muscle are determined it may therefore be necessary to convert to a whole body value using an appropriate conversion factor. Some consideration must also be afforded the fact that some organs may be exposed to much higher dose rates than the body as a whole.
- The applicability of using CRs in the process of calculating activity concentrations in environmental compartments should be explored. Since Pb-210 is delivered via the atmosphere, activity concentrations on the surface of plants are likely to be enhanced and the overall activity concentration (including truly incorporated Po-210) is likely to exhibit a complex relationship with activity concentrations in the underlying soil.

A terrestrial and freshwater sampling campaign was planned and executed in 2007 for the purpose of collecting suitable samples for subsequent analyses of natural radionucldies, details are provided below.

5.1 Terrestrial field study to collect samples and derive levels and CRs

A field study was planned and implemented at Dovre, Central part of Norway (62°17' N, 9°36' E) during the period 17-20th June 2007. This study site was selected primarily on the basis that it forms part of the network for Monitoring programme for Terrestrial Ecosystems (TOV) in Norway, led by the Norwegian Institute for Nature research (NINA), and concerning, *inter alia,* effects of pollution on plants and animals and chemical and biological monitoring. In this way a large dataset of ancillary information would be available facilitating any subsequent interpretation of results. Furthermore, by connecting this field programme to ongoing studies, associated costs could be reduced. The sample site is shown in Figure xxx with a selection of pictures.

The field study was conducted within a designated Landscape-protected area near to Kongsvold adjacent to Dovrefjell-Sunndalsfjella National Park.

Figure 3. Study area for the terrestrial part of the GAPRAD project .

Eight soil profiles were collected during the field expedition. These profiles were spilt into an overlying humus layer and thereafter 3 cm (predominantely mineral soil) increments to a depth of 9 cm using a custom-designed soil corer. This was undertaken with a view to enabling an analyses of the activity distribution of radionuclides with depth. In this way insights into the amount of unsupported Po-210 in the surface soil could be established and information in relation to the migration, if any, of Pb and Po following deposition might be ascertained.

Baited traps were used in the collection of various rodents including Tundra vole (*Microtus oeconomus)* and Grey Red-Backed Vole (*Clethrionomys rufocanus*). Plant samples including samples of bilberry (*Vaccinium myrtillus*) and 2 species of lichens (e.g. *Cladonia stellaris and Cladonia*) were collected by hand. Finally, samples of two earthworm species (*Lumbricus rubellus* and *Aportectodea caliginosa*) were collected in areas of brown earth using a spade.

Samples have been determined for ^{137}Cs. The plan is now to measure all samples for activity concentrations of ^{210}Po and ^{210}Pb.

In view of point made above (section 5 concerning the applicability of Po-210, CRs , the work of Pietrzak-Flis & Skowrońska-Smolak (1995) are of particular relevance. The study presents data concerning the above ground interception and soil to plant uptake of ^{210}Po and ^{210}Pb. This was achieved by harvesting different crop types (including grass) with and without a tented shelter. In this way the contribution from aerial deposition could be assessed. The authors discovered that for those crops with large surface area over ground parts, the contribution from atmospheric deposition could be large. The contribution of the above ground interception of ^{210}Pb and ^{210}Po for grass was in excess of 90 %. Furthermore, the proportion of adsorbed radionuclides compared to truly incorporated, determined by measuring the rinse water from samples, could be significant – several 10s of % in some cases although the actual amount was highly variable. The CR values the authors derived pertained to the tented conditions – i.e. transfer less the direct atmospheric deposition, although notably some degree of surface adsorption even appears to occur without wet aerial deposition.

The paper of Pietrzak-Flis & Skowrońska-Smolak (1995) has implications for interpretation of the GAPRAD results. For bilberry samples, the activity concentrations are likely to reflect predominately the atmospheric deposition of ^{210}Pb and ^{210}Po. If we define the CR strictly as the activity concentrations in the plant arising from the soil to plant transfer of the studied radionuclides, the results from GAPRAD might be considered invalid. This is clearly a point for discussion – in the context of an EIA one would apply a CR in the event that activity concentration in soil had been measured but no direct observation data were available for flora and fauna. The application of the CR would then depend on the contamination source – for sub-surface contamination e.g. arising from infiltration of contaminated ground water, mill tailings etc., a CR based on the underling assumption of soil to plant transfer only would be most appropriate. However, in other cases – those reflecting natural conditions or input from an atmospheric source, transfer data that incorporate the atmospheric deposition of radionuclides may also have relevance. In this latter case, owing to the indirect association with soil concentrations it may be more appropriate to express the data as an aggregated transfer factor thereby relating the activity concentrations in vegetation to deposition.

5.2 Freshwater field study to collect samples and derive levels and CRs

STUK has taken samples for the GAPRAD project from lakes, Baltic Sea and from the environments of the two nuclear power plants in 2007 (Figure 4).

Samples of lake water and three species of fish from three lakes (Table 8) were taken for the analyses of ^{210}Po and ^{210}Pb. Edible parts of fishes were separated and taken for the analyses. Besides edible parts, the 'non-edible' parts of one sample of each species will be analysed for ^{210}Po and ^{210}Pb in order to get the ratio needed to change the results for edible parts to whole-body activity concentrations. Additionally, lake water samples, taken in 2003 from two lakes (Vehkajärvi and Siikajärvi), were analysed for $^{239,240}Pu$. Aquatic plants, as e.g. yellow water lily or water lily from those lakes, taken also in 2003, are planned to be analysed for Pu to get the concentration ratios.

From the environments of the nuclear power plants in Loviisa and Olkiluoto samples representing birds (a swan), reptiles (a snake) and amphibians (a frog) were taken for the analyses. To get the concentration ratios for the terrestrial organisms, samples of surface soil (0-10 cm) were taken (Table 8).

Table 8. Biota, water and soil samples from lakes and sea areas taken for ^{210}Po and ^{210}Pb or ^{239,240}Pu analyses. Edible parts of all the fish samples will be analysed, and from four samples also the non-edible parts (a). Samples marked with (b) are planned to be analysed for ^{239,240}Pu and those with (c) are also planned to be included in the project if there are analytical resources available.

Sample	Species	Latin name	Lake / Sea area	Sampling year
Amphibian	frog	*Rana temporaria*	Olkiluoto	2007
Aquatic plants	yellow water lily (b)	*Nuphar lutea*	Siikajärvi	2003
Aquatic plants	yellow water lily (b)	*Nuphar lutea*	Vehkajärvi	2003
Benthic animals	-	*Saduria entomon*	Loviisa	2007
Benthic animals	lake mussel	*Anodonta sp.*	Mänttä, Keurusselkä	2007
Bird	mute swan	*Cygnus olor*	Loviisa	2007
Fish	bream	*Abramis brama*	Iso-Ahvenainen	2007
Fish	white fish	*Coregonus lavaretus*	Iso-Ahvenainen	2007
Fish	pike	*Esox lucius*	Iso-Ahvenainen	2007
Fish	pike	*Esox lucius*	Myllyjärvi	2007
Fish	bream	*Abramis brama*	Myllyjärvi	2007
Fish	bream (a)	*Abramis brama*	Myllyjärvi	2007
Fish	vendace	*Coregonus albula*	Vesijako	2007
Fish	bream	*Abramis brama*	Vesijako	2007
Fish	pike	*Esox lucius*	Vesijako	2007
Fish	pike-perch	*Stizostedion lucioper-ca*	Vesijako	2007
Fish	pike-perch (a)	*Stizostedion lucioper-ca*	Vesijako	2007
Fish	perch	*Perca fluviatilis*	Vesijako	2007
Fish	perch (a)	*Perca fluviatilis*	Vesijako	2007
Fish	pike	*Esox lucius*	Vesijako	2007
Fish	pike (a)	*Esox lucius*	Vesijako	2007
Fish*	several species (c)	-	several areas (c)	2005
Reptile	viper	*Vipera berus*	Olkiluoto	2007
Soil	0-10 cm	-	Olkiluoto	2007
Water	sea water	-	Baltic Sea	2007
Water	lake water	-	Iso-Ahvenainen	2007
Water	lake water	-	Myllyjärvi	2007
Water	lake water	-	Mänttä, Keurusselkä	2007
Water	lake water		Vesijako	2007
Water	lake water (c)	-	some lakes (c)	2008

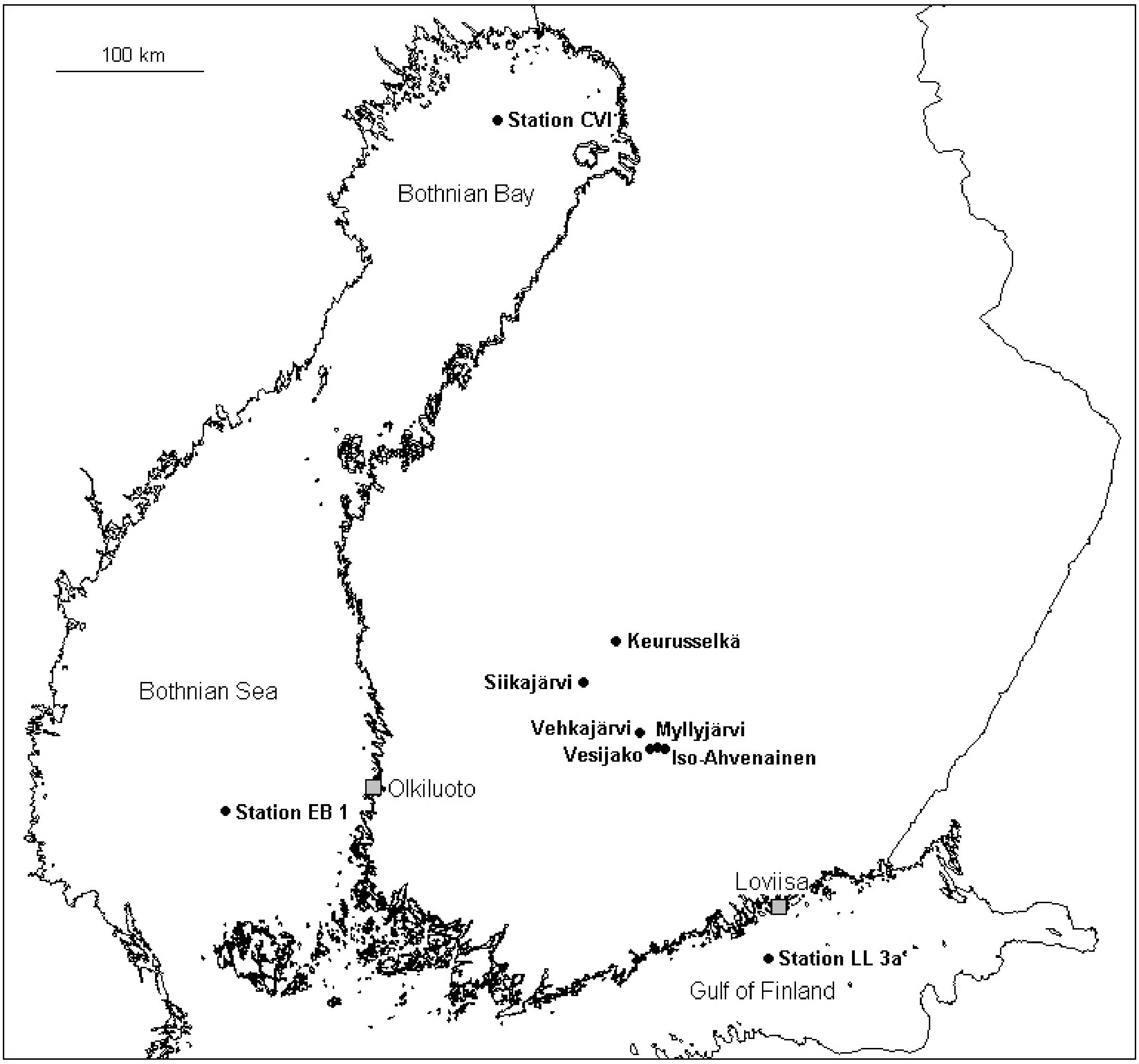

Figure 4. Lakes, land areas, sea areas and sampling stations used in the Finnish contribution to the GAPRAD project. Several types of samples (table 1) were taken for analyses to fulfil the aims of the project as planned.

6. Conclusion (and recommendations)

International activities with regards the development of methods for assessing impacts on the environment from ionising radiation have been substantial in recent years. In developing these methods, there are requirements (i) to determine the transfer of radionuclides within ecosystems and (ii) to determine background dose-rates arising from the presence of naturally occurring radionuclides, in a satisfactory manner. It has quickly become evident that fulfilling these 2 requirements is not entirely straightforward reflecting a lack of data in many cases. This report specifies exactly where these data-gaps lie through analyses of data generated from the most recent studies conducted internationally on this topic. It is evident that information is limited for numerous radionuclides from ^{238}U and ^{232}Th decay series and notably, in view of its importance as a contributor to dose-rates in plants and animals, ^{210}Po. The simple way to rectify these data deficiencies is to organise target field campaigns focusing on particular species and radionuclides where information is lacking. To this end, field sampling has been conducted in a semi-natural mountain ecosystem in Norway and freshwater aquatic systems in Finland. It is envisaged that the data derived from the studies briefly described in this report will provide fundamental information for our understanding of the behaviour and fate of natural decay series radionuclides in terrestrial and aquatic systems and provide the basis for more robust way.

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Key words	Transfer, non-human biota, impact assessment

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