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

Final summary report

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# Filling knowledge gaps in radiation protection methodologies for non-human biota. Final summary report

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## Abstract

The activities of the GAPRAD project are summarised in this report. The background and rationale to GAPRAD are presented and explained. Most notably this relates to a lack of information on naturally occurring radionuclides in terrestrial and aquatic systems that have direct applicability for use in environmental impact assessments. Results from field activities are presented from the Dovrefjell area in Norway (terrestrial study) and selected lake and brackish water systems in Finland. The data mainly concern activity concentrations of Po-210 in environmental media and selected biota allowing concentration ratios to be derived where appropriate. Furthermore, details in relation to Po-210 uptake and biokinetics in humans based on experimental work conducted within the project are presented.

## Key words

Po-210, environmental impact assessment, levels, transfer, concentration ratios, human biokinetics

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**GAPRAD**

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

**Final summary report**

A report for the NKS-B activity October 2008

**Edited by Justin Brown and Runhild Gjelsvik**

Norwegian Radiation Protection Authority, Norway

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# 1 Introduction

The NKS B-programme GAPRAD - Filling knowledge gaps in radiation protection methodologies for non-human biota project started in May 2007. The project has been conducted as a collaborative effort between the Norwegian Radiation Protection Authority, University of Lund in Sweden, RISØ national laboratory in Denmark and Nuclear Safety Authority (STUK) in Finland.

The aim of the project was to identify data on activity concentrations of Po-210 in soil, plants, invertebrate and small mammals. In addition, there were plans to measure concentration of natural radionuclides like U-238, U-234, Ra-226, Ra-228, Po-210, Pb-210 in fish, brackish waters and sediments where practicable.

A kickoff meeting was held at STUK Helsinki, 14<sup>th</sup> June 2007. The minutes from this first meeting were included in the first status report submitted in 2007.

The GAPRAD Activity was presented at the NKS seminar held at the NRPA 28 to 29<sup>th</sup> August 2007.

The deliverable report entitled “Knowledge gaps in relation to radionuclide levels and transfer to wild plants and animals, in the context of environmental impact assessments, and a strategy to fill them” was submitted to the NKS in December 2007.

A status report summarizing the activities in the first 6 months of the GAPRAD project was submitted to the NKS in late October 2007.

Studies conducted as part of GAPRAD have been presented at an international Conference held in Bergen in 2008 (see Brown et al., 2008). The paper presented at this symposium has subsequently been accepted for publication in the Journal *Radioprotection* following peer review.

A Year 2 meeting was held at the NRPA, Norway on 18<sup>th</sup> September 2008. The minutes from this latest meeting are included in this report as Appendix 1.

The Deliverable report entitled “Po-210 and other radionuclides in terrestrial and freshwater environments” was submitted with a slight delay in November 2008. Following comments from the NKS-B Programme Manager a final amended version of the report was submitted on 28.11.2008. Elements from this deliverable and the deliverable submitted in 2007 on “Knowledge gaps...” have been included in the Final report in the process of drawing out some of the main achievements and conclusions resulting from the GAPRAD project.

**This report constitutes the Final Milestone in the project as agreed under contract {(NKS Order [AFT/B(07)8]}. The report aims to summarise the main activities that have been performed in the period covered by the contract.**

Deliverables under the NKS-B GAPRAD activity

[NKS-181](#) Runhild Gjelsvik and Justin Brown (editors): Po-210 and other radionuclides in terrestrial and freshwater environments

[NKS-182](#) J.E. Brown (ed.): Knowledge gaps in relation to radionuclide levels and transfer to wild plants and animals, in the context of environmental impact assessments, and a strategy to fill them

## 2 Key Results from Year 1 of the GAPRAD project

### 2.1 Setting the scene

The deliverable report “Knowledge gaps in relation to radionuclide levels and transfer to wild plants and animals, in the context of environmental impact assessments, and a strategy to fill them.” sets the scene for the work conducted in the GAPRAD project by discussing how the protection of the environment from ionising radiation has become a broadly discussed topic over the last decade or so and by identifying where there have been and currently are problems in developing a framework for environmental impact assessment. The objective of the deliverable report was to provide an 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.

The method applied for the deliverable report was simply to access databases and reviews conducted recently in the context of the development of environmental impact assessment approaches. The focus of the work was on natural decay series radionuclides ( $^{238}\text{U}$  and  $^{232}\text{Th}$  decay series radionuclides with half-lives  $> 10$  days<sup>1</sup>)

This entailed:

- Summarising information from the ERICA project (see Larsson, 2008): 3 empirical databases have been collated within the ERICA project with respect to terrestrial, freshwater and marine ecosystems. These data were analysed in the study to provide an overview as to where data gaps exist.
- Analyses of other recently published data

### 2.2 An overview of transfer (and for natural activity concentration) knowledge gaps for environmental impact assessment

The CR empirical data coverage for selected radionuclides provided by the ERICA project for terrestrial and freshwater environments have been presented in the deliverable report “Knowledge gaps in relation to radionuclide levels and transfer to wild plants and animals, ...” Since the publication of the NKS GAPRAD report, comprehensive overviews of the ERICA transfer data sets have been published in Hosseini et al. (2008) and Beresford et al. (2008a). A synopsis of the key results from the deliverable report are given below.

For the terrestrial environment, 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 Polonium, some information is available for flora but only for the fauna group mammals. In the latter case it should be noted that

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<sup>1</sup> 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.

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 exists 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.

In the freshwater environment, 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 plants. 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.

A recent report by (Beresford et al., 2007) that draws on the concept of reference animals and plants (RAPs) has considered some of the databases that underpin environmental impact assessment with emphasis on the activity concentration ranges of naturally occurring radionuclides in non-human species. An overview of these data show that there are no data for some RAPS, notably frog, bee earthworm and rat and very few data for some other groups, notably duck ( $^{40}\text{K}$  only) and deer ( $^{40}\text{K}$ , 1 data point for  $^{210}\text{Po}$ ). 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 measurements of  $^{210}\text{Po}$  were made in the study.

Brown et al. (2004) reported background dose rates for aquatic environments. 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.

### 2.3 Devising a strategy to mitigate the lack of transfer and activity concentration data

In its simplest sense the strategy to mitigate the lack of transfer and activity concentration data involved the focussed experimental studies including field-work and analyses. The following activities were identified as areas where a small effort will reap great dividends :

- Determination of Po-210 in soil fauna, small mammals and soil
- Characterisation of 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.

The role of skeletal  $^{210}\text{Pb}$  decay on subsequent soft tissue  $^{210}\text{Po}$  concentrations in mammals is still a matter for some debate (Skuterud et al., 2005). In order to elucidate some of the points arising from this contention, studies will be performed to attain information on gastrointestinal uptake and residence time in mammals (using "man" as the reference species). Such data should add to our understanding of how  $^{210}\text{Pb}$  and  $^{210}\text{Po}$  are transferred in environmental systems.



Field work was conducted in at Dovre, Central Norway (62°17' N, 9°36' E) during the period 17-20th June 2007. Numerous samples were collected from freshwater lakes, Baltic Sea and from the environments of the two nuclear power plants by STUK in 2007. Experiments on ingestion and uptake of polonium involving human subjects were conducted in the period 2007 and 2008. In view of the time required to process samples, perform analyses and collate data, the results from all of these studies were not reported until 2008, i.e. Year 2 of GAPRAD as presented below.

## 3 Key Results from Year 2 of the GAPRAD project

Much of the following has been extracted from the Deliverable report entitled “Po-210 and other radionuclides in terrestrial and freshwater environments”. Where this is not the case, this has been specified.

### 3.1 Aquatic environments

#### 3.1.1 $^{210}\text{Po}$ and $^{210}\text{Pb}$ in Freshwater and Brackish water environments

Lake water and fish for  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  analyses were sampled in 2007 from four lakes: Iso-Ahvenainen, Myllyjärvi, Vesijako and Miestämä. Five fish species were studied: perch (*Perca fluviatilis*), pike (*Esox lucius*), bream (*Abramis brama*), white fish (*Coregonus lavaretus*) and vendace (*Coregonus albula*). Lake mussel (*Anodonta sp*) and water samples were collected from lake Keuruselkä in 2007. Additionally, fish samples from various parts of the Baltic Sea and from lakes, belonging to the monitoring programme of STUK in 2005, were analysed for Po and Pb. Furthermore, a benthic isopod (*Saduria entomon*) and a bird, swan (*Cygnus olor*), were collected from the environments of Finnish nuclear power plants in Loviisa.

The average activity concentration of  $^{210}\text{Po}$  in lake waters was 0.0019 Bq/kg. Variation between the lakes was rather low, from 0.0016 to 0.0020 Bq/kg. Activity concentrations of  $^{210}\text{Pb}$  were somewhat higher, on average 0.0031 Bq/kg. Activity concentrations of  $^{210}\text{Po}$  in whole fish varied more than  $^{210}\text{Po}$  in lake water from the same lakes, from 1.0 Bq/kg f.w. to 6.5 Bq/kg f.w. The lowest values for  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  were found in pike-perch and the highest in bream. Contents of  $^{210}\text{Pb}$  in fishes were much lower (5-15 times lower) than those of  $^{210}\text{Po}$ ,  $^{210}\text{Pb}$  activity concentration varying from 0.09 to 1.3 Bq/kg f.w. In edible part of the fish, highest concentrations for both isotopes were measured in vendace. Activity concentration of  $^{210}\text{Po}$  and especially that of  $^{210}\text{Pb}$  in freshwater mussel, *Anodonta sp.*, were somewhat higher than that in fishes (with an exception of bream).

Both  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  concentrations in water from various parts of the Baltic Sea were lower than in lake waters, although values for  $^{210}\text{Po}$  were in most cases below the detection limit, which was estimated to be 0.002 Bq/kg water.

Two parallel analyses were also carried out in various parts of the swan: breast muscle, liver and bones.  $^{210}\text{Po}$  in liver was ten times higher and  $^{210}\text{Pb}$  six times higher than in breast muscle. Activity concentrations of  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  in whole swan were estimated to be 1.0 and 0.4 Bq/kg f.w. In *Saduria entomon* from the Gulf of Finland, activity concentration of  $^{210}\text{Po}$  was four times higher and  $^{210}\text{Pb}$  almost the same (a little lower) than that in the freshwater mussel (*Anodonta sp.*). Among the organisms studied, the highest activity concentration of  $^{210}\text{Po}$  was found in the crustacean *Saduria entomon*.

Further information on the sample preparation, analyses methods and detailed results is provided in the aforementioned deliverable report.

The data presented above and in the deliverable report were used to derive Concentration Ratios, i.e. activity concentration in organism divided by the activity concentration in water for the main organism categories. A comparison was thereafter made with the values underpinning the ERICA database as presented in Hosseini et al. (2008). These data are presented in Tables 3.1 and 3.2.

**Table 3.1.** Concentration ratios (fresh weight) for brackish water biota (whole organism).

Organism	CR, <sup>210</sup> Po		CR <sup>210</sup> Pb	
	GAPRAD study	ERICA	GAPRAD study	ERICA
fish	<b>670 - 5300</b>	44000±120000* 240**	<b>13-84</b>	4400±14000* 300**
crustacean	<b>&gt;9300</b>	56000±66000* 9900 ±1400**	<b>770</b>	7500±2100* 7500**
bird	<b>&gt;510</b>	30000* 240**	<b>190</b>	19000* 300**

\* marine

\*\* freshwater

**Table 3.2** Concentration ratios (fresh weight) for freshwater biota (whole organism).

Organism	CR, <sup>210</sup> Po		CR <sup>210</sup> Pb	
	GAPRAD study	ERICA	GAPRAD study	ERICA
fish benthic	<b>630-9250</b>	240	<b>16-340</b>	300
mollusc bivalve	<b>1740</b>	38000±49000	<b>530</b>	1400

The data from this study constitute an important additional source of material to the data already collated from earlier review. It appears that some of the values in the underpinning ERICA databases may need revision following this work. For example, freshwater CRs for <sup>210</sup>Po in the GAPRAD study appear to be somewhat higher for benthic fish and somewhat lower for bivalve mollusc than those contained within the ERICA databases.

### 3.1.2 Isotopes of plutonium in freshwater environments

STUK also conducted a number of analyses on Pu isotopes. These data were not included in the deliverable report owing to a decision to restrict the scope of the report to a consideration of radioisotopes of polonium and lead only. These additional data are reported below.

Activity concentration of <sup>239,240</sup>Pu in lake waters varied from 4.9 to 39.5µBq/kg and that of <sup>238</sup>Pu between 1.2 and 2.1 µBq/kg. Activity concentrations of <sup>239,240</sup>Pu and <sup>238</sup>Pu were very low and in many cases below detection limit which was estimated to be about 1µBq/kg. In aquatic plants <sup>239,240</sup>Pu varied between 0.6 and 3.4 mBq/kg f.w., concentrations being highest in water horsetail from lake Siikajärvi and lowest in yellow water lily from lake Vehkajärvi. <sup>238</sup>Pu activity concentrations were up to ten times lower than those of <sup>239,240</sup>Pu.

These measurement data were used to derive CR values. As for Po and Pb, the data constitute an important new source of information that may be used to amend existing environmental impact assessment databases pertaining to transfer. A summary of the plutonium CR from the STUK study are presented in Table 3.3.

**Table 3.3.** CRs of  $^{238}\text{Pu}$  and  $^{239,240}\text{Pu}$  for aquatic plants and fish on dry weight and on fresh weight basis.

Lake	Species	CR, $^{238}\text{Pu}$ (d.w.)	CR, $^{239,240}\text{Pu}$ (d.w.)	CR $\pm$ unc %, $^{238}\text{Pu}$ (f.w.)	CR $\pm$ unc %, $^{239,240}\text{Pu}$ (f.w.)
	<b>Aquatic plants</b>				
Vehkajärvi	Yellow water lily		770	-	120 $\pm$ 35
Vehkajärvi	Water lily		< 810	-	< 70
Vehkajärvi	Water horsetail		447	-	130 $\pm$ 42
Siikajärvi	Yellow water lily	1020	200	200 $\pm$ 45	39 $\pm$ 34
Siikajärvi	Water lily	< 2140	476	< 400	89 $\pm$ 22
Siikajärvi	Water horsetail	730	578	190 $\pm$ 53	150 $\pm$ 18
	<b>Fish</b>				
Ontojärvi	Perch	< 970	< 57	< 260	< 15
Vehkajärvi	Perch		590	-	150 $\pm$ 45
Siikajärvi	Perch	< 1890	< 76	< 480	< 20

## 3.2 Terrestrial environments

### 3.2.1 $^{210}\text{Po}$ and $^{210}\text{Pb}$ in small mammals and other fauna

Samples collected during the field work in Dovrefjell, Central Norway, were dried, pulverised and homogenised prior to gamma spectrometric analyses (these data, primarily relating to activity concentrations of  $^{137}\text{Cs}$ , will form part of the terrestrial monitoring programme at the NRPA).

Following this preliminary sample preparation and gamma analyses, small (whole body including hair) mammal samples were analysed at RISØ national laboratories according to Chen et al. (2001). All other terrestrial samples, i.e. soil, plant, lichen, earthworm and wild bird, were analysed at Lund University Hospital (Sweden) according to Flynn (1968).

Po-210 activity concentrations in small mammals have been reported earlier in the aforementioned GAPRAD deliverable report and conference paper (see Brown et al., 2008). In summary, activity concentrations were 41 – 88 Bq kg<sup>-1</sup> d.w.  $^{210}\text{Po}$  for bank vole (n= 8) and 22 – 84 Bq kg<sup>-1</sup> d.w.  $^{210}\text{Po}$  for the common shrew (n=9). The non-parametric Mann-Whitney test has been applied in order to determine whether the shrew and vole data are statistically different. It was found that the null hypothesis (that the 2 dataset have been taken from a common population) can be rejected at the p < 0.008 level . It appears that the primarily herbivorous bank vole is accumulating higher concentrations of these natural radionuclides compared to the insectivorous shrew. The  $^{210}\text{Po}$  activity concentrations for the whole body of the bank vole and shrew are similar in magnitude to activity concentrations determined for the muscle of reindeer, sampled at a site <100 km distant at Vågå in Norway. At this

location activity concentrations of 36 Bq kg<sup>-1</sup> d.w. in female reindeer muscle and several hundred Bq kg<sup>-1</sup> d.w. in liver were determined (Skuterud et al., 2005). The dry mass to fresh mass ratio was on average 0.3. This suggests median activity concentrations by fresh mass of approximately 22 Bq kg<sup>-1</sup> f.w. on average for Bank Vole and 9 Bq kg<sup>-1</sup> f.w. for the Common shrew, albeit that these are preliminary determinations. These activity concentrations are considerably higher than the levels reported in Beresford et al. (2007), where an activity concentration of 0.09 Bq kg<sup>-1</sup> f.w. was reported for a category consigned the title “All mammals” and comprising of 32 assorted samples. Whether this discrepancy reflects the preliminary nature of the results presented in this paper, differences (physiology, diet, habitat) between the mammals considered in the aforementioned study and the present study, differences in what is being measured (i.e. muscle versus whole body etc.) or differences in deposition of <sup>210</sup>Pb between the study areas, remains a subject for further investigation.

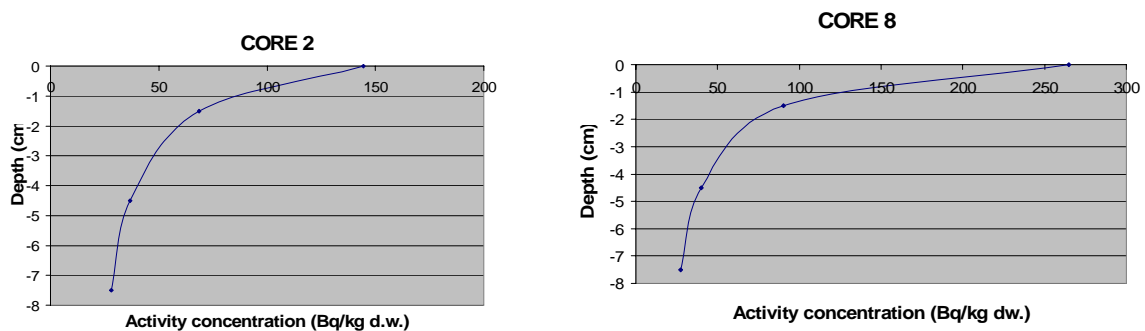
A preliminary data set on the concentrations of <sup>210</sup>Pb in small mammal samples indicates that the correction required for this will not be great (i.e.<10%). The preliminary data reported on <sup>210</sup>Po in the deliverable report may therefore be considered to be a reasonable indication of the activity concentrations of <sup>210</sup>Po at the time of sampling.

Samples of viper and frog, taken from the Olkiluoto area in 2007, were dried, homogenized, digested and analyzed as a whole animal at the STUK laboratories in Finland. Total amounts of <sup>210</sup>Po and <sup>210</sup>Pb in the viper were 5.3 and 3.1 Bq kg<sup>-1</sup> f.w. respectively and those in the frog were 1.68 and 0.93 Bq kg<sup>-1</sup> f.w. respectively. More details concerning the analytical procedures, uncertainties and derivation of concentration ratios are provided in the aforementioned Deliverable report.

The following data, resulting from the Dovrefjell study were not included in the Deliverable report.

### 3.2.2 <sup>210</sup>Po in soil.

Typical activity concentration versus depth profiles in soil are shown in Figure 3.1.



**Figure 3.1.** <sup>210</sup>Po Activity concentrations with depth for 2 cores taken in the Dovrefjell study showing typical activity versus depth profiles..

Several observations can be made from a consideration of these data :

- All profiles exhibit an approximately exponential fall in activity concentrations from elevated levels in humus/surface soils to a “supported” level at depth. This undoubtedly reflects the atmospheric input of <sup>210</sup>Pb followed by the redistribution of the radionuclide to lower soil strata by burial, physical migration and bioturbation. The unsupported activity of <sup>210</sup>Po from atmospheric input has decayed within a few centimetres of topsoil.
- The Supported activity concentration of <sup>210</sup>Po at a level of tens of Bq/kg d.w. is evident at depths below approximately 5 cm.

- Surface activity concentrations (in the range of 84-338 Bq kg<sup>-1</sup> d.w. for the humus layer for the 8 cores analysed) seem relatively high for natural site although direct comparison is difficult because activity concentrations are often quoted for different depths of surface soil.
  - Bunzl & Kracke (1984) measured activity concentrations in the range of 11-28 Bq/kg d.w. <sup>210</sup>Po in soil samples from a heathland in Western Germany. The soil samples consisted of the upper 15 cm of soil.
  - Lapham et al. (1989) determined levels of 70 ± 10 Bq kg<sup>-1</sup> d.w. <sup>210</sup>Po For a control location. Soil samples consisted of the top 5 cm of soil in this study.

Atmospheric <sup>210</sup>Pb concentrations are positively correlated with the size of the underlying landmasses, whereas terrestrial areas covered by ice and snow and marine areas including islands have reduced atmospheric concentrations of <sup>210</sup>Pb (El-Daoushy, 1988). Furthermore, the deposition of <sup>210</sup>Pb is directly correlated with the level of precipitation (Hill, 1960).

### 3.2.3 <sup>210</sup>Po in soil invertebrates, bilberry and wild birds

Earthworm samples comprising of 2 species (Grey worm – *A. caliginosa* and the Red Earthworm - *L. rubellus*) were analysed. Preliminary data (not corrected for ingrowth of <sup>210</sup>Po via <sup>210</sup>Pb between the time of sampling and the time of analysis) show activity concentrations in the range 29-77 Bq/kg d.w. (n=7). These levels are comparable with those determined for small mammals in this same study. It should be noted that these activity concentrations represent situations where the gastrointestinal tract had not been evacuated and therefore it might be expected that the determination may, to some extent, reflect the activity concentrations of the decaying organic material that the worms feed upon. No data were found in the literature to make a direct comparison with the present study.

Samples of Bilberry (*Vaccinium myrtillus*) leaves expressed a range of 14-22 Bq/kg d.w <sup>210</sup>Po. This is commensurate with the level of 8.1 Bq/kg d.w. <sup>210</sup>Po in the leaves of bilberry for a heathland area in west Germany Bunzl & Kracke (1984). These concentrations are furthermore within range (apply a d.w. to f.w. conversion factor of 0.25) for wild grass in the data presented in Beresford et al. (2008b).

Activity concentrations of <sup>210</sup>Po for the breast muscle of Willow Grouse (*Lagopus lagopus*) were in the range 1.4 – 3.4 Bq/kg d.w. (or using a conversion factor of 0.3 to convert between fresh and dry masses → 0.35 – 0.85 Bq/kg f.w.). This falls within the range of 0.05 Bq kg<sup>-1</sup> to 2.30 Bq kg<sup>-1</sup> f.w. for Baltic seabirds determined recently (Skwarzec & Fabisiak (2007). Not surprising the values reported in the Dovrefjell study appear to fall below the range of < 5 mBq g<sup>-1</sup> to 7 ± 5 Bq kg<sup>-1</sup> d.w. determined for ruffed grouse (*Bonasa umbellus*) from a study area with technologically enhanced naturally occurring radioactive material in Canada (Clulow et al., 1992).

### 3.2.4 Concentration ratios (CRs)

There are numerous issues that should be addressed concerning the validity of applying <sup>210</sup>Po CR values in terrestrial environments. The concept of CRs lends itself most suitably to long term post depositional equilibrium conditions when the main transfer pathway is from soil to plant or more indirectly from soil to plant to animal. In the case of <sup>210</sup>Po, the continual atmospheric deposition of this radionuclide results in conditions that can be considered to deviate substantially from this ideal for applicability, with a substantial component of contamination arising from interception of the radionuclide on plant surfaces. Furthermore, the rapid fall in activity concentrations of <sup>210</sup>Po with depth and considerations related to the bioavailability of supported compared to unsupported <sup>210</sup>Po render the interpretation (and thereby appropriateness of applicability) of CRs values problematic.

Nonetheless, CR values have been derived from Po-210 in the open literature and there appears to be some value in making a cursory comparison between the present study and these earlier works.

Concentration ratios have been derived based upon the uncorrected  $^{210}\text{Po}$  data presented above. The number of data points “n” reflects the number of plant and animal samples used in the derivation of average CRs. Correction factors to transform all biota data to fresh weight have been taken from Beresford et al. (2008a). The soil activity concentrations used in the calculations are based on all 8 cores (averaged to a depth of 9 cm and including the humus layer) sampled during the investigations. The standard deviation associated with the soil data have been used in the derivation of the combined uncertainty of the CR values. The summarised data are presented in Table 3.4.

**Table 3.4.  $^{210}\text{Po}$  Concentration ratios ( $\text{Bq kg}^{-1}$  f.w. biota per  $\text{Bq kg}^{-1}$  d.w. soil) for different biota types.**

	CR average	std	n
Bilberry	2,23E-02	8,66E-03	3
Grey worm	1,02E-01	4,33E-02	5
Red worm	1,03E-01	3,78E-02	2
Grouse	9,96E-03	2,91E-03	5
Bank vole	2,55E-01	9,86E-02	8
Shrew	1,42E-01	7,91E-02	9

The review on CR values for a large suite of radionuclides performed by Beresford et al. (2008a) included no empirical data for  $^{210}\text{Po}$  transfer to soil invertebrates or birds. In both cases, the  $^{210}\text{Po}$  CR values were equated to those for terrestrial mammals. The data presented in Table 3.4 are, therefore, useful at very least in providing direct empirical data for such organism categories and suggest that using *in situ* mammal data as surrogate values may have generated conservative transfer values for soil invertebrates and bird in this particular case.

For those cases where direct comparison is possible it can be seen that  $^{210}\text{Po}$  CR values for bilberry are somewhat lower than the generic values provided for shrub (cf. 2.23E-02 with 9.85E-02) whereas data for small mammals from the present study are 2 orders of magnitude higher than the values provided within the general review (cf. circa 2E-01 and 2.78E-3). In speculation, this may reflect the origin of data. Beresford et al. (2008a) used data that appear to have been collated in deriving doses to humans from natural foods and therefore  $^{210}\text{Po}$  activity data probably pertain to edible parts of edible mammals. Although a correction is made to derive an equivalent whole body CR for  $^{210}\text{Po}$  based on association of the nuclide with bone this appears not to characterise adequately the whole body transfer under field conditions.

### 3.3 Polonium in man

Within the framework of GAPRAD a study was set up to establish radiobiological parameters, important in dosimetry, such as fractional uptake parameter gastrointestinal absorption factors  $f_1$  and biological retention times of radioisotopes Po-209 and Po-210 in the body. Gastrointestinal absorption factors have been established in earlier studies with a wide range of results. P.A. Thomas *et al* (2001) showed that a total ingestion of 20 Bq in Caribou meat resulted in a maximum of 3.2 Bq/day in faecal excretion 4-6 days after intake and a maximum of 0.32 Bq/day of urinary excretion 5-10 days after ingestion. From these results a GI factor of  $0.56 \pm 0.04$  was established. ICRP has increased their reported GI factor from 10% (1979) to 50% (1993), which according to other studies seems too small. G.J. Hunt *et al* (1993) reported values of GI factors in the range of 0.6-0.94 with a mean of 0.76 after analyzing a study where 7 volunteers ingested crab meat with a total Po-210 activity intake of up to

44.2 Bq. 10%-25% of daily rate faecal excretion and a maximum urine excretion rate of 0.2%-0.3% was reported. In the 50ies Silberstein *et al* (1950) published GI factor values as low as 0.1-0.3.

In the first part of the study one person was given 50 mBq of Polonium 209 with an oral intake frequency of 24 hours. The goal of this part was to remain the intake frequency until constant radioactive output from urine and feces was maintained, i.e. equilibrium of intake and excretion. 24h urine samples were collected a few times every month until 320 days from the first intake. Then the intake of <sup>209</sup>Po and urine sampling stopped and 24h faeces sampling for a week begun. The results showed clearly a slow decreasing excretion of <sup>209</sup>Po in faeces in the range 0.59%-0.07% of consumed activity. Urine samples analysis showed a fluctuating value of <sup>209</sup>Po excretion with a maximum peak value of about 1 % (ca 17.5 mBq/L) 40 days from the first intake. From this maximum the output activity fluctuates between 0.85 mBq/L-15.91 mBq/L of total intake activity and tends asymptotically against 5.78 Bq/day (5.50 Bq/L).

The next step of the project was to distribute an acute oral intake to two persons of 10 Bq and then study the immediate body burden response by spectrometric analysis of urine and feces. In the acute oral intake study, the maximum daily excretion rates in faeces of 18-50 % can be measured 3 days after intake. Urine activity excretion measures an average of 0.15-1 % of ingested activity after 2 days from intake.

**These results indicate a GI factor of 0.50-0.75. These results correlate well with earlier biokinetic studies of polonium in man.**

## **Acknowledgment**

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## 5 Appendix 1 – Minutes from the Second Annual GAPRAD meeting

### Minutes from GAPRAD meeting

**Venue:** Norwegian Radiation Protection Authority (Staten strålevern), Grini næringpark 13, 1332 Østerås

**Date:** 18th September 2008

**Participants :** Elis Holm (University of Lund), Per Roos (RISØ), Runhild Gjelsvik (NRPA), Justin Brown(NRPA), Lisa Outola (STUK), Ritva Saxen (STUK)

Welcome, update, practical matters - **Justin Brown**

Update (from last year)

NKS-B status seminar 2007: held at Statens strålevern, Oslo, August 28-29.

- GAPRAD was presented at the seminar and (although few results to present) was positively received: copies of presentation are available on request.
- The report "Knowledge gaps in relation to radionuclide levels and transfer to wild plants and animals, in the context of environmental impact assessments, and a strategy to fill them" (show on computer - pdf) submitted to NKS last year on time. No feedback and hasn't been published on the NKS website! Copies of presentation are available on request.
- (Short) Status report sent to NKS in October of last year.

The contract with NKS was discussed.

The group is committed to produce 2 reports: 1 scientific report entitled "Report on Po-210 and other radionuclide transfer in terrestrial and freshwater environments" and 1 final-summary report

The group was also reminded about claiming expenses back from NKS. The participants were encouraged to re-read contract and check that the (economic) administrators for each group had claimed 50 % following signature. The participants will need to wait until we've submitted all reports (and received acceptance of these) before applying for the final payment.

Presentations were given on the work conducted within the project.

- Update on Po studies (Po biokinetics in humans?)- **Elis Holm**
- Sampling and analyses in aquatic environments by STUK - **Ritva Saxen & Lisa Outola**
- Radioecological studies of the terrestrial environment at Thule – **Per Roos**
- Sampling in terrestrial environments in Norway (Dovrefjell study) - **Runhild Gjelsvik and Justin Brown**

A request was made by Justin Brown to receive  $^{210}\text{Pb}$  results for the environmental samples from Lund and RISØ as soon as possible.

The Deliverable report: "Report on Po-210 and other radionuclide transfer in terrestrial and freshwater environments" (deadline October 2008) – **was discussed**.

The following Report outline, responsibilities and deadlines were agreed.

Editors – Runhild and Justin

## 1. Introduction

- Background and rationale - Justin
- Chemistry, decay, physical processes - Elis
- Behaviour of Po-210 in the environment – deposition, biological consideration - Elis
- *Geochemistry, biochemistry – Per*
- Objectives - Justin

## 2. Freshwater and brackish environment (in Finland) –(Po-210 and other relevant – Pb-210) – lisa and Ritva

- Review existing information (e.g. availability of data from ERICA)
- Methodology (sampling area, analyses) – radiochemistry briefly
- Results and discussion

## 3. Terrestrial environment – Justin & Runhild

- Dovrefjell (Norway)
    - i. Review of existing information
    - ii. Methodology (sampling area, analyses)
    - iii. Results and discussion
- STUK – viper and frog – Lisa and Ritva

## 4. $^{210}\text{Po}$ in man - Elis

- **Review of older information – limitations with older work**
- **New study, methods materials, results.**

## 5. Dose-rates in the environment - Justin

- Preliminary dose-rate calculations using newly acquired data and earlier review information : implications for dose-rates in natural systems

## 6. Conclusion and recommendations – Input from Elis (all can contribute?)

Further work! Foodstuffs, Nordic countries

## Acknowledgements

## References

N.B.

References in the main text and the reference list will follow a standard format as used in Journ. Of Environ. Activity.

Timeplan: Submission = End of October

Draft by mid-October

Circa: 20 pages

Plans for continuation of GAPRAD (if appropriate) – A decision was made not to propose a prolongation of the project. New activities concerning Po-210 have been initiated and involve some of the participants from GAPRAD. It was considered necessary to “take stock” of work achieved to date and see how this initial work is received by NKS and the wider community.

The group will stay in contact and consider a follow up to GAPRAD if they consider this to be appropriate.

Po-210 in foodstuffs for Nordic populations could be a theme for a follow-up project.

(Joint) Publications plan – All

- Joint (NRPA, NINA, Lund, RISØ and STUK) publication – Behaviour of Po-210 in terrestrial environments based on Dovrefjell study and terrestrial data from STUK.
  - NRPA to take lead and circulate a first draft in early 2009.
- Po-210 in man (Lund) based on results presented in GAPRAD

Final Summary Report – NRPA to take the lead. This should be a formality following on from the scientific report due in October. Deadline = December 2008. NRPA may contact the GAPRAD participants for additional input (likely to be minimal).

Title	Filling knowledge gaps in radiation protection methodologies for non-human biota - Final summary report.
Author(s)	1. Justin Brown and Runhild Gjelsvik (editors) 2. Elis Holm 3. Per Roos 4. Ritva Saxen and Iisa Outola
Affiliation(s)	1. Norwegian Radiation Protection Authority, Norway 2. University of Lund, Sweden 3. Risø National Laboratory, Denmark 4. STUK - Radiation and Nuclear Safety Authority, Finland
ISBN	978-87-7893-254-9
Date	March 2009
Project	NKS-B / GAPRAD
No. of pages	17
No. of tables	4
No. of illustrations	1
No. of references	19
Abstract	The activities of the GAPRAD project are summarised in this report. The background and rationale to GAPRAD are presented and explained. Most notably this relates to a lack of information on naturally occurring radionuclides in terrestrial and aquatic systems that have direct applicability for use in environmental impact assessments. Results from field activities are presented from the Dovrefjell area in Norway (terrestrial study) and selected lake and brackish water systems in Finland. The data mainly concern activity concentrations of Po-210 in environmental media and selected biota allowing concentration ratios to be derived where appropriate. Furthermore, details in relation to Po-210 uptake and biokinetics in humans based on experimental work conducted within the project are presented.
Key words	Po-210, environmental impact assessment, levels, transfer, concentration ratios, human biokinetics