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Po-210 and other radionuclides in terrestrialand freshwater environments

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Abstract

This report provides new information on Po-210 (and where appropriate its grandparent Pb-210) behaviour in environmental systems including humans. This has primarily been achieved through measurements of Po-210 in aquatic and terrestrial environments that has led to the derivation of information on the levels of this radioisotope in plants, animals and the biotic components of their habitat (i.e. water, soil) providing basic information on transfer where practicable. For freshwater environments, Po-210 concentration ratios derived for freshwater benthic fish and bivalve mollusc were substantially different to values collated from earlier review work. For terrestrial environments, activity concentrations of Po-210 in small mammals (although of a preliminary nature because no correction was made for ingrowth from Pb-210) were considerably higher than values derived from earlier data compilations. It was envisaged that data on levels of naturally occurring radionuclides would render underpinning data sets more comprehensive and would thus allow more robust background dose calculations to be performed subsequently. By way of example, unweighted background dose-rates arising from internal distributions of Po-210 were calculated for small mammals in the terrestrial study. The biokinetics of polonium in humans has been studied following chronic and acute oral intakes of selected Po radioisotopes. This work has provided information on gastrointestinal absorption factors and biological retention times thus improving the database upon which committed effective doses to humans are derived. The information generated in the report, in its entirety, should be of direct relevance for both human and non-human impact assessments

Key words

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

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Po-210 and other radionuclides in terrestrial and freshwater environments

A Deliverable report for the NKS-B activity October 2008

GAPRAD

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

Edited by Runhild Gjelsvik and Justin Brown
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 is 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.

The following milestones have been achieved in the GAPRAD project:

- Milestone - Kickoff meeting at STUK Helsinki, 14th June 2007
- Presenting the GAPRAD activity at the NKS seminar held at the NRPA 28 to 29th August 2007
- Deliverable - Report on knowledge gaps and strategy to fill them, August 2007
- Milestone – October 2007, 1st year field activities completed.
- Milestone - Presentation of data at International Conference on Radioecology and Environmental Radioactivity, June 2008.
- Milestone – Meeting to discuss progress in the project and plan final activities - September 2008
- Deliverable - Report on Po-210 and other radionuclide in terrestrial and freshwater environments, October 2008

This deliverable report "Filling knowledge gaps in radiation protection methodologies for non-human biota" and progress report for 2007 summarises the work performed in the GAPRAD project in 2007.

The project will be completed by December 2008 at which time a final report will be submitted to NKS.

2 Background and rationale

Elis Holm, University of Lund, Sweden.

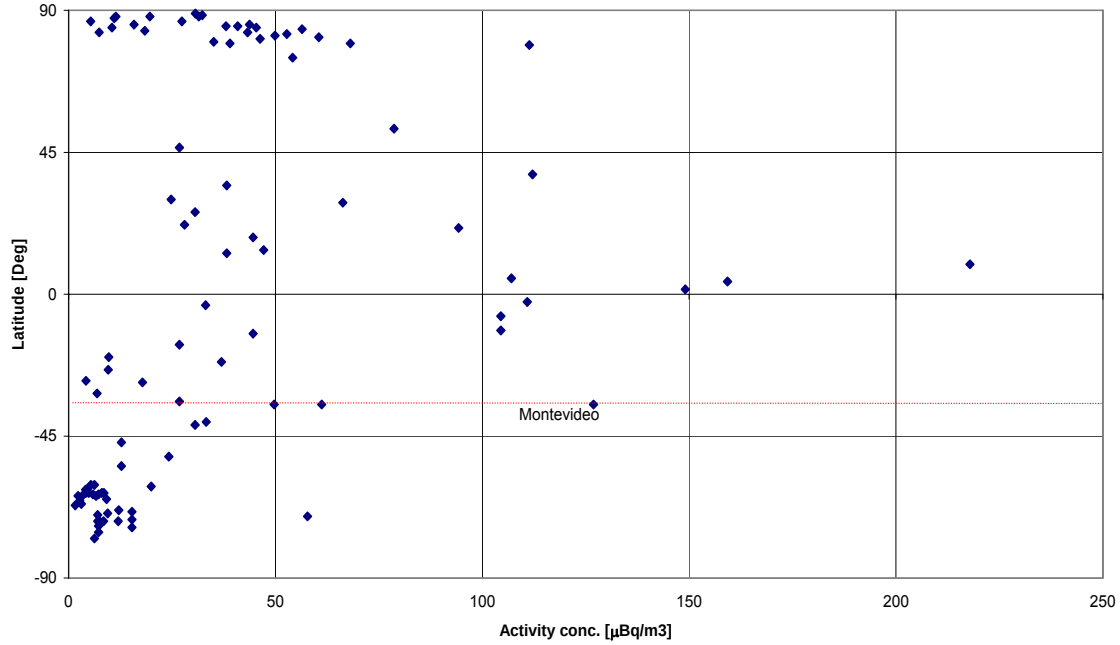
Polonium was discovered by Pierre and Marie Currie in 1898 in the course of research of the radioactivity of uranium and thorium minerals. They found that Po behaved in a similar way to Bi and subsequently were able to separate it from Bi by vacuum sublimation. Polonium-210 is highly radiotoxic with a specific activity of 166 TBq/g. It is a daughter product in the ^{238}U decay chain through ^{210}Pb and ^{210}Bi but can also be produced by neutron activation of ^{209}Bi. It is the alpha emitter which gives the highest dose to humans via food intake.

The main source of ^{210}Pb in the atmosphere is ^{222}Rn which is exhaled from the ground at 18 mBq m^{-2} s^{-1} or 48 EBq per year. This corresponds to an annual production rate of atmospheric ^{210}Pb of 23 PBq (Persson, 1970). This is a surprising figure compared to the amount of ^{137}Cs from the Chernobyl accident and the radiotoxicity of ^{210}Po is much higher than that of ^{137}Cs. Burton and Stuart (1960) found that the concentration gradient of ^{210}Pb increased sharply in the vicinity of the tropopause. The source of ^{210}Pb in the stratosphere is explained by the ascending air at the equator which carries not only ^{210}Pb but also ^{222}Rn and its daughter-products, which ^{210}Pb will be formed from.

The annual precipitation varies from a few Bq per m^2 such as in the Antarctic (Roos *et al*., 1994) to several hundred Bq m^2 (El-Daoushy, 1988). The amount depends on the surrounding land and the possibilities for exhalation of ^{222}Rn. The exhalation over sea is small since the ^{226}Ra concentration in sea water is only about 1mBq l^{-1}. The annual deposition in central Sweden was estimated to about 63 Bq m^{-2} $year^{-1}$ (Persson, 1970).

The figures below (2.1, 2.2 and 2.3) give the ^{210}Pb, ^{210}Po and the activity ratio $^{210}Po/^{210}Pb$ over the North and South Atlantic measured during Swedish Antarctic and Arctic Expeditions. The equilibrium ratio of U to Po is 1.19 $x10^{10}$, so that the Po concentration in uranium ores is less than 0.1 mg/ton. With the advance of nuclear reactors and their intense neutron fluxes, the reaction ^{209}Bi (n, γ) $^{210}Bi \rightarrow$ ^{210}Po became economically feasible. This process is currently used for the production of Po.

210-Po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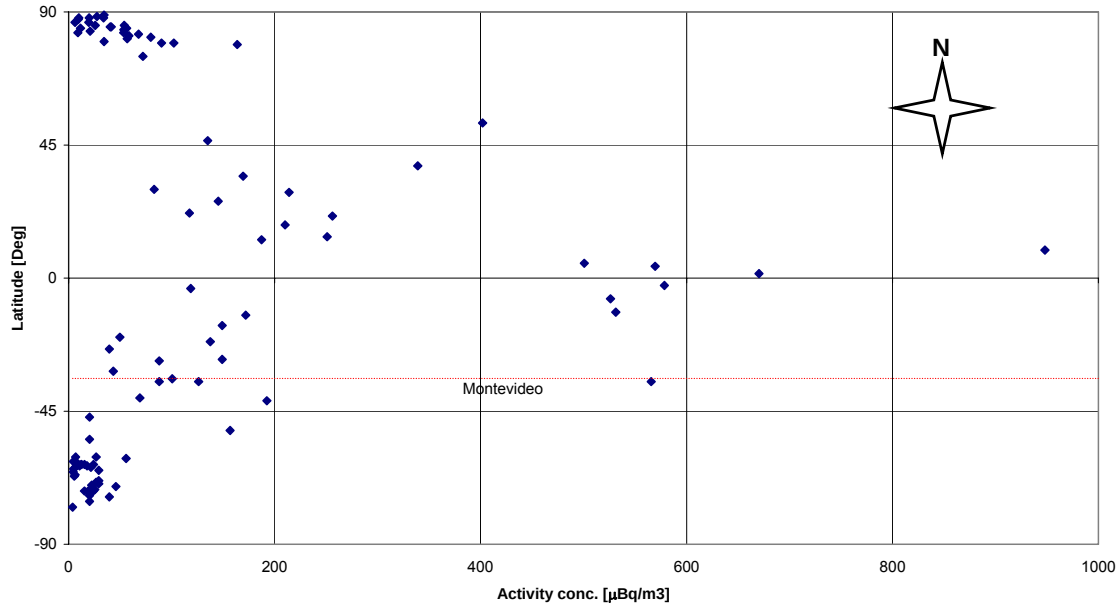
Activity conc. [µBq/m3]

210-Pb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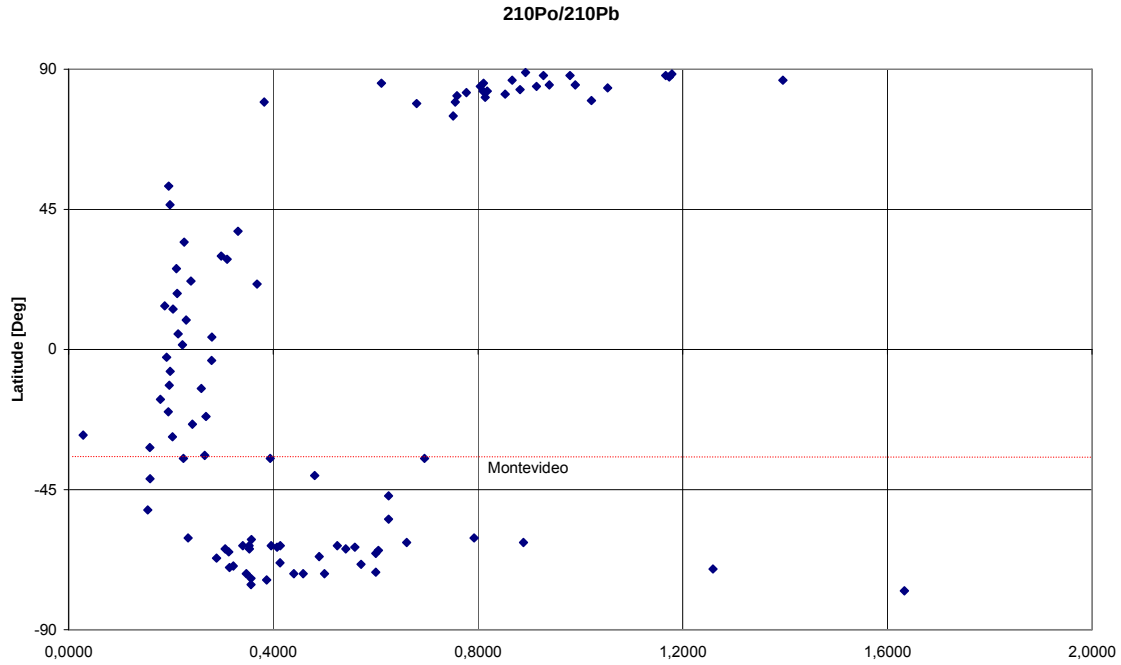

Figure 2.1, 2.2 and 2.3. *Activity concentrations of ^{210}Po, ^{210}Pb and the $^{210}Po/^{210}Pb$ activity ratio as function of latitude over the North and South Atlantic.*

2.1 Physical and chemical characteristics of polonium

A number of other isotopes of Po also appear in naturally occurring decay series. However, they have all short half lives. One difficulty in working with ^{210}Po is its high specific activity (166 TBq/g). The intensive radiation of milligrams quickly decomposes most organic complexing agents and even solvents. Crystal structures of solids are quickly destroyed or altered. Therefore, ^{209}Po is used instead which can be produced by bombarding ^{209}Bi with protons or deuterons in a cyclotron. However, more ^{208}Po is produced than ^{209}Po. Both ^{209}Po and ^{208}Po can be used as radiochemical yield determinants.

Polonium is in the same group as Se and Te in the periodic system with oxidation state +4. It is a silver colored half metal. The melting point is 527 K and the boiling point 1235 K. It is however clear that Po generally evaporates at much lower temperatures. The table below (Table 2.1) shows the decay of the most important Po isotopes.

Polonium is quite soluble in HF, acetic acid and mineral acids. Polonium can be precipitated with hydroxides. However using NaOH our experience is that the precipitation is not complete and according to literature it is better to use NH_3. It is carried almost quantitatively by Bi $(OH)_3$ in ammonical solution. For analytical purpose our experience is that the best precipitation technique at environmental levels is to form a MnO_2 co-precipitate .

Polonium is strongly absorbed in HCl media on anion exchangers. It can be extracted into isopropyl ether or methyl isobutyl ketone from HCl solutions containing KI. Polonium can also be extracted into Tributyl Phosphate (TBP) from 6M HCl and can be recovered with conc. HNO_3. Also Thenoyltrifluroroacetone (TTA) (HCl solution and pH above 1.3) has been used for the extraction of polonium.

Table 2.1. *Physical characteristics of Po isotopes.*

Isotope	Physical half life	Alpha energies	Intensity %	Gamma energies	Intensity %
^{208}Po	2.90 years	4.883 MeV 4.885 MeV	80 20		
^{209}Po	103 years	5,304 MeV	100	260 keV 263 keV 896 keV	0.7 0.23 0.47
^{210}Po	138 days	5.115 MeV	100		

2.2 Deposition and transfer in the terrestrial environment

If a radioactive material is deposited by wet or dry deposition a certain fraction may be intercepted by vegetation and the remainder will reach the ground. This is expressed as:

$$1-f = \exp(-\mu B)$$

where f is the fraction initially retained on vegetation

B is the biomass (kg m^{-2} d.m.)

μ is the interception coefficient (m^2/kg) assuming exponential interception.

Very often the interception fraction, f/B, per unit weight of biomass is used.

There are no specific data for ^{210}Po, but generally values vary from 1-4 depending on precipitation and type of vegetation for ^{137}Cs, ^{7}Be and lower 0.1-1 for ^{131}I (IAEA, 1994)

The fraction retained in mosses and lichens are higher than for grass due to their larger biomass m^{-2}. Persson *et al*, 1974 showed that the radionuclides of fallout radionuclides were retained in lichens in the following order, ^{144}Ce < ^{7}Be < ^{95}Zr > ^{137}Cs > ^{106}Ru ^{155}Eu > ^{210}Pb > ^{125}Sb.

The uptake of radionuclides by plants from soil is described as the transfer factor B_v, the ratio of radionuclide concentration in vegetation and soil (Bq kg^{-1} d.w. plant to Bq kg^{-1} d.w. soil).

Data for polonium are given as 2.3 x 10^{-3} for wheat grain, 1.2 x 10^{-3} for vegetables and 9 x 10^{-2} for grass. Those values are not corrected for aerial contamination and might be a factor of 2-10 lower. In comparison, the values for ^{137}Cs are (1-5) x 10^{-3} for grass and (1-8) x 10^{-2} for cereals. For ^{210}Pb the data are given as 4.7 x 10^{-3} for cereals and 1x10^{-2} for vegetables.

The partition coefficient, K_D to soil is defined as the ratio of radionuclides in the solid and liquid phases. The K_D values for ^{90}Sr and ^{137}Cs range from 20 to 1000 and migration velocities from 1 cm per year to almost zero. Table 1.2 gives dome data for ^{210}Po together with other radionuclides.

***Table 2.2**. Partition coefficients, K_D, of selected radionuclides in soil*

Radionuclide	**Sand** Expected *Range*	**Loam** Expected *Range*	**Clay** Expected *Range*	**Organic** Expected *Range*
^{137}Cs	2.7 x10^{2} *1.8 x10^{0}-4x10^{4}*	4.4x10^{3} *3.3x10^{2}-6x10^{4}*	1.8x10^{3} *7.4x10^{1}-4,4x10^{4}*	2.7x10^{2} *2.0x10^{-1}-3.6x10^{5}*
^{90}Sr	1.1x10^{1} *5.5x10^{-1}-2x10^{-4}*	2x10^{1} *6.7x10^{-1}-6x10^{2}*	1.1x10^{2} *2x10^{0}-6x10^{3}*	1.5x10^{2} *4x10^{0}-5.4x10^{3}*
^{210}Po	1.5x10^{2} *6x10^{0}-3.6x10^{3}*	4x10 *3x10^{1}-5.4x10^{3}*		6.6x10^{3}
^{210}Pb	2.7x10^{2} *2.7x10^{0}-2.7x10^{4}*	1.6x10^{4} *9.9x10^{2}-2.7x10^{5}*	5.4x10^{2}	2.2x10^{4} *8.1x10^{3}-6x10^{4}*

The transfer from feed to animal is described as the transfer coefficients F_m or F_f for milk and other animal products respectively. These coefficients are defined as the amount of an animal's daily intake of a radionuclide that is transferred to 1 kg of the animal product at equilibrium or at the time of slaughter. The coefficients depend on many things such as metabolic homeostasis, effect of chemical and physical form of the radionuclide, influence of age, food habits, variation from year to year of available food etc.

***Table 2.3.** Transfer coefficients, $F_m$for cow milk (d/L), goat milk and F_f (d/kg) for beef.*

Radionuclide	**Cow milk**	**Goat milk**	**Beef**
	Expected *Range*	Expected *Range*	Expected *Range*
^{137}Cs	7.9x10^{-3} *1x10^{-3}-3.5x10^{-2}*	1x10^{-1} *9x10^{-3}-4.7x10^{-1}*	5x10^{-2} *1x10^{-2}-6x10^{-2}*
^{90}Sr	2.8x10^{-3} *1x10^{-3}-3x10^{-3}*	2.8x10^{-2} 6x10^{-3}-3.9x10^{-2}	8x10^{-3} *3x10^{-4}8x10^{-3}*
^{210}Po	3x10^{-4}		5x10^{-3} *6x10^{-4}-5x10^{-3}*
^{210}Pb			4x10^{-4} *1x10^{-4}-7x10^{-4}*

For the incorporation of radioactivity into aquatic fauna is expressed as the concentration factor, C_f, defined as the ratio of the activity concentration in animal tissue to that in water (Bq/kg dw. or ww. organism per Bq/kg or Bq/l water). Our experience is that there is no salinity effect for polonium as for caesium. Table 2.4 gives data for selected radionuclides.

Table 2.4. *Concentration factors for edible portions of freshwater fish. (L/kg).*

Radionuclide	Expected *Range*
^{137}Cs	$2x10^3$ $3x10^1$-$3x10^3$
^{90}Sr	$6x10^1$ 10^0-$1x10^3$
^{210}Po	$5x10^1$ 10^1-$2x10^2$
^{210}Pb	$3x10^2$ $1x10^2$-$3x10^2$

2.3 Biogeochemical behaviour of polonium

Per Roos, Risø national laboratory, Denmark

Studies of the environmental behaviour of polonium has been motivated by the use of ^{210}Pb-^{210}Po in sediment chronology and the ^{210}Po/^{210}Pb ratio in marine scavenging investigations as well as by the toxicity of polonium. In terms of geochemical studies, ^{210}Po and ^{210}Pb remobilization from lake and marine sediments in relation to iron and manganese cycling has been of particular interest. Although dating of sediments using ^{210}Pb does not directly concern ^{210}Po, the problem appears when the analysis of ^{210}Pb is performed using ^{210}Po and assuming equilibrium between the two isotopes. Polonium is generally more reactive towards particulate matter than ^{210}Pb and fluxes to sediments are therefore generally enriched with ^{210}Po relative to ^{210}Pb. If both elements remain immobile in the sediments the problem from a dating point of view then would only concerns the very upper mm to cm of sediments, depending on sedimentation rate and initial disequilibrium ^{210}Po/^{210}Pb. Within some months (equivalent to some mm in most coastal or lake sediments where the technique is most frequently used) equilibrium between the two would be established. However, due to diagenetic processes, redox conditions change with depth in all sediments and at some depth oxides of manganese starts to dissolve when manganese becomes reduced to its divalent state and at a somewhat later (deeper) stage iron similarly becomes more mobile as it becomes reduced to its divalent state. A cyclic behaviour of reduction-oxidation then starts when dissolved ions diffuses upwards and becomes oxidized when approaching more oxidizing conditions, the oxides of manganese and iron are then precipitated and again becomes dissolved once they are buried to a depth where reduction once more occurs and so on. Due to the slow nature of dissolution and diffusion only a minor fraction of iron and manganese undergoes this cyclic behaviour while the rest is more or less permanently buried in the sediments. If the redox zone is situated within the sediment column iron and manganese will move up and down in it and at the redox front an enrichment of both elements occur. In some lakes and poor ventilates coastal seas the redox front moves up a distance in the water column and an effective removal of iron and managanese from the sediments then takes place. Since the amorphous forms of both iron and manganese oxyhydroxides are exceptional effective carriers of transition metals the cyclic behaviour may also affect these elements and in fact many of these elements (e.g., Cu, Co, Ni) show more or less pronounced patterns in some sediments attributed to the Fe-Mn cycling. Since there are several sites for trace metals to attach to in sediments the situation is more complex than just due to the Fe-Mn cycling. The presence of sulphur, in the form of sulphide, similarly is very redox dependent and elements like lead easily forms sulphides (PbS) as do most transition elements. The mobility of lead as a sulphide was studied by Widerlund et.al. (2002)

If lead and/or polonium would significantly be affected by changing redox conditions the ^{210}Pb tool to determine sediment chronology would be seriously hampered. Evidence for remobilisation of both ^{210}Pb and ^{210}Po in anoxic water was found by Benoit and Hemond (1990) while studying a oligotrophic, dimictic lake. They attributed the remobilisation to the redox cycling of Fe and Mn. Furthermore, they could observe a fractionation between ^{210}Pb and ^{210}Po during the process. Polonium

was released at an earlier stage than lead from the sediments and in fact even before the water overlaying the sediments had been anoxic. In a later study, Benoit and Hemond (1991) further showed that the high concentrations of ^{210}Pb and ^{210}Po found in the interstitial water and the diffusion out from sediments matched the elevated concentrations found in the lake water. They also analysed the implications for dating and concluded that the effect from polonium diffusion was small but for ^{210}Pb it could mean significant errors. Results similar to Benoit and Hemond were reported by Balistrieri et.al. (1995) while studying a seasonally anoxic lake. They attributed the behaviour to Fe-Mn cycling and indicated that the cycling of polonium was more closely related to Mn-cycling than Fe-cycling. Polonium having a reputation as sometimes being a volatile element has also been shown to become volatile in both fresh and marine waters by the action of microorganisms (Momoshima et.al., 2001 & 2002) and both ^{210}Pb and ^{210}Po are found at elevated levels in the sea-surface microlayer (Bacon et.al., 1980). The situation is similar to what may appear with methylated heavy metals such as methylmercury although evidence for methylated polonium is lacking.

With concern to biochemistry there are 3 analogue elements in the chalcogen group where polonium belongs – S, Se and Te. They have similar chemical properties but different functions in living organism. Neither tellurium nor polonium have any known biological function while sulphur and selenium are incorporated into several amino acids which due to the powerful redox behaviour of these elements often occur in enzymes. Both S and Se are mostly absorbed in the body as sulphate and selenate and subsequently become incorporated into organic compounds. Even though selenium is an essential element the threshold between essentiality and toxicity can be narrow. Although tellurium has a non-nutrient behaviour in the marine environment it is assimilated by some terrestrial and marine primary producers as well as in fungi in which it was shown to incorporate into amino acids in place of selenium (Ramadan et.al. 1989).

It was earlier suggested (Cherry and Shannon, 1974) that elevated ^{210}Po concentrations found in marine organisms were linked to sulphur uptake. Particularly the visceral organs such as the digestive gland or hepatopancreas of invertebrates and the pyloric caecum of fishes show very high activities. Bustamante et.al. (2002) found levels of ^{210}Po in excess of 4 Bq/g dwt in the digestive gland of the scallop *Chlamys varia* and values in excess of 30 Bq/g dwt in hepatopancreas from the marine penaeid shrimp (Cherry & Heyraud, 1982). Using a median ^{210}Po concentration of about 22 Bq/g dry weight and a wet to dry ratio of 3:1 they calculated a hepatopancreas organ dose of 3.9 Sv/y. The explanation for the very high concentration of polonium in the hepatopancreas has often been attributed to potential binding to sulphur-containing amino acids such as Cysteine and/or to methallothioneins. Cherry et.al. (1983) showed that the enrichment factor for polonium (relative to Al) was exceeded only by Ag, Cd and Se. In a detailed study of the subcellular localization of natural polonium in the hepatopancreas of the Rock Lobster, Heyraud et.al. (1987) provided evidence for exceptional strong binding of polonium to high molecular weight (10^7 dalton or more) proteins although it was not considered a metallothionein. Other investigations of polonium in liver of vertebrates (Aposhian & Bruce, 1991) and fish (Durand et.al., 1999) have on the other hand pointed to the association of polonium to proteins such as metallothionein and ferritine. Potentially polonium can replace selenium in selenocysteine which is present in several enzymes. Also selenium and polonium have similar distribution pattern depending on the internal organs of marine vertebrates and fish (Heyraud and Cherry, 1979).

2.4 Objectives and structure of report

The key aim of this report is to provide new information of ^{210}Po (and where appropriate its grandparent ^{210}Pb) behaviour in environmental systems including humans. The plan was primarily to achieve this through measurements of ^{210}Po in aquatic and terrestrial environments that would lead to the derivation of information on the levels of this radioisotope in plants, animals and the biotic components of their habitat (i.e. water, soil) providing basic information on transfer where practicable. Furthermore, it was envisaged that data on levels would allow more robust background dose calculations to be performed subsequently. A further objective was to study the biokinetics of

polonium in humans thereby augmenting and improving the database upon which committed effective doses to humans are derived. In this way the information generated will be of direct relevance and interest for both human and non-human impact assessments.

The report has been spilt into 5 chapters. Following the introduction, rationale and background to the study (chapters 1 and 2), the report describes the work performed in relation to determining ^{210}Po in freshwater and backish environments (chapter 3), followed by ^{210}Po in terrestrial environments. The final chapter deals with ^{210}Po biokinetics in humans.

3 Polonium-210 in freshwater and brackish environment

Ritva Saxen and Iisa Outola, Radiation and Nuclear Safety Authority (STUK), Finland

3.1 Review of existing information

In the ERICA project, coordinated by EU in 2004-2007 (Larsson, 2008), following radionuclides: Cs, Pu, Co, I, Ra, Sr, Po, Ru, Cm, U, P, Ce, Am, Cl, Sb, Mn, Ag, Th, Cd, Eu, Nb, Ni, Tc, Np, S, Te, Zr, Se, Pb, H, C and following organism groups: benthic and pelagic fish, vascular plants, bivalve molluscs, insect larvae, phytoplankton, amphibian, crustacean, gastropod, zooplankton, birds, mammal, were selected for the study on the transfer of the radionuclides to be used in the ERICA tool. When collecting the transfer data, plenty of data gaps in the transfer of radionuclides into various types of biota in freshwater environment were revealed. The data gaps were addressed and published together with the values found in the open literature (Hosseini et al., 2008). Transfer factors for several radionuclides and several types of biota were based only on a few data. Sparse data and large variations in the activity concentrations of plants and animals in freshwater environment lead to uncertainties when applying the ERICA tool for the estimation of radiation doses to freshwater biota. To fill some of the data gaps and to get more data to improve the uncertainty of the previous estimations, some freshwater and brackish water biota and their habitat were analysed for ^{210}Po and ^{210}Pb.

3.2 Experimental studies

3.2.1 Samples

Lake water and fish for ^{210}Po and ^{210}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 Keurusselkä 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. Reproducibility of the Po and Pb analyses were also tested with these samples. Surface and near-bottom water samples from two sampling stations in the Gulf of Finland and from one station in the Bothnian Sea, collected in 2006, were also analysed for ^{210}Po and ^{210}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 weight of the whole swan was 7.7 kg. The weights of different organs were liver 0.16 kg, muscle and bones 4.54 kg, intestines other than liver 1.12 kg, feather and skin 1,70 kg. Muscle and bones could not be totally separated from each other but assuming that bones account 4.5 % of the total weigh in swan, it was estimated that the weight of muscle would be 4.38 and bones 0.35 kg. Sampling sites are given in Figure 3.1.

Chemical recoveries were 60-80%.

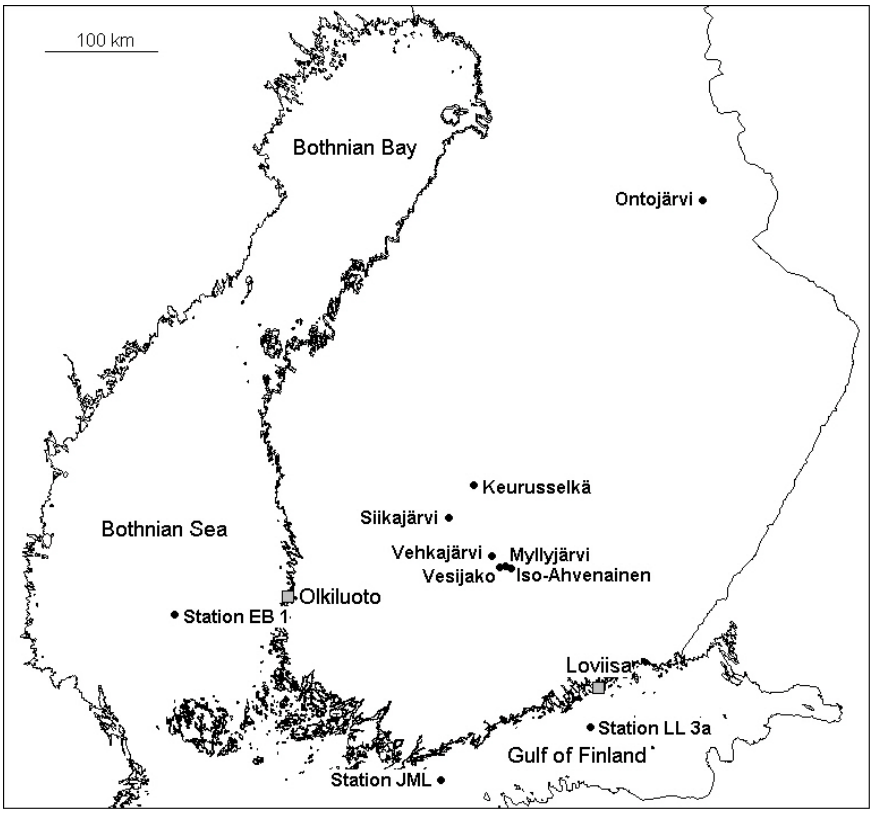

Figure 3.1*. Location of the sampling sites for lake water, Baltic water and biota samples.*

3.2.2 Sample treatment

Fish samples were gutted as usually takes place in the kitchen when preparing fish as food. The edible parts were used for the analyses. The rest of the sample (other than edible parts) were also analysed from one sample of most species to get the correction factor with which the activity concentrations in edible parts can be changed into activity concentrations in the whole organism. Contents of ^{210}Po and ^{210}Pb in swan were analysed separately in breast muscle, in bone plus bone marrow and in liver. Determining the activity concentrations in these organs and using the weights of the organs and the whole organism, the average activity concentration in the whole bird was estimated. Since feathers and skin were not analysed, the activity concentrations of ^{210}Po and ^{210}Pb in whole animal was estimated by making an assumption that the Po and Pb concentration in these parts is similar to that in breast muscle. Admittedly, this assumption may be somewhat tenuous in view of the consideration that hair and possibly feathers are known to accumulate Po, but the implications for the calculations of average concentration in the whole bird are not large because the contribution of feathers to the total mass of the animal is low. For ^{210}Po-^{210}Pb analysis the biota samples were digested using the microwave oven method. One sample had to be divided into several sub samples in the digestion, because the maximum amount of the sample in one cell of the microwave oven is 2.5 g.

3.2.3 Analyses of ^{210}Pb and ^{210}Po

A known amount of ^{209}Po tracer for the yield determination is added to the water sample or to the digested biota sample. Po is separated from the sample by spontaneous deposition to a silver plate in acidic solution. The precipitation vessel with the silver plate and with the sample solution is placed to a water bath and heated to 80^0C. Polonium is spontaneously deposited in 3-4 hours. After the separation, the solution is poured to a glass bottle and the silver plate is rinsed, dried and measured by alpha spectrometry. After a standing period of six months to allow ingrowth of ^{210}Po from ^{210}Pb, a

known amount of ^{208}Po tracer is added to the sample bottle and the sample is mixed well. The solution is transferred to a precipitation vessel. The precipitation vessels are made of teflon. The solution is acidified and spontaneous deposition to a silver plate is carried out. The plate is finally measured with an alpha spectrometer. Activity concentrations of ^{210}Pb and ^{210}Po are calculated from the following equations:

$$A_{Pb} = \frac{e^{\lambda_1 t_{m2}} \times C_{Po}(2.precipitation)}{(1 - e^{(-\lambda_1 (t_2 - t_1))})}$$

$$A_{Po} = e^{\lambda_1 t_1}\left[e^{(\lambda_1 t_{m1})} \times C_{Po}(1.precipitation) - A_{Pb}(1 - e^{(-\lambda_1 t_1)})\right]$$

C_{Po}(2.precipitation) = polonium activity (Bq/l) in 2.precipitation

C_{Po}(1.precipitation) = polonium activity (Bq/l) 1.precipitation

λ_1 = decay correction of polonium / day (λ_1 = Ln2 / T½ = 0,00501)

t_1 = time from sampling to the 1st precipitation

t_2 = time from sampling to the 2nd precipitation

t_{m1} = time from the 1st precipitation to the measurement

t_{m2} = time from the 2nd precipitation to the measurement

3.2.4 Results

Activity concentrations of ^{210}Po and ^{210}Pb

The average activity concentration of ^{210}Po in lake waters was 0.0019 Bq/kg. Variation between the lakes was rather low, from 0.0016 to 0.0020 Bq/kg (Table 3.1a) Activity concentrations of ^{210}Pb were somewhat higher, on average 0.0031 Bq/kg (Table 3.1a). Activity concentrations of ^{210}Po in whole fish varied more than ^{210}Po in lake water from the same lakes, from 1.0 Bq/kg f.w. to 6.5 Bq/kg f.w. (Table 3.1b). The lowest values for ^{210}Po and ^{210}Pb were found in pike-perch and the highest in bream. Contents of ^{210}Pb in fishes were much lower (5-15 times lower) than those of ^{210}Po, ^{210}Pb activity concentration varying from 0.09 to 1.3 Bq/kg f.w. (Table 3.1b). In edible part of the fish, highest concentrations for both isotopes were measured in vendace.

Activity concentration of ^{210}Po and especially that of ^{210}Pb in freshwater mussel, *Anodonta sp*., were somewhat higher than that in fishes (with an exception of bream) (Table 3.2). Both ^{210}Po and ^{210}Pb concentrations in water from various parts of the Baltic Sea were lower than in lake waters, although values for ^{210}Po were in most cases below the detection limit, which was estimated to be 0.002 Bq/kg water (Table 3.3). The standard deviation of parallel determinations of ^{210}Po was from 2% to 15% and that of ^{210}Pb clearly higher (Table 3.4). The origin of some of the fish presented in Table 3.4 is uncertain.

Two parallel analyses were also carried out in various parts of the swan: breast muscle, liver and bones. ^{210}Po in liver was ten times higher and ^{210}Pb six times higher than in breast muscle. Activity concentrations of ^{210}Po and ^{210}Pb in whole swan were estimated to be 1.0 and 0.4 Bq/kg f.w. (Table 3.5). The estimation was made assuming that feather, skin and muscles, which were not analysed, have same activity concentrations than breast muscle. In *Saduria entomon* from the Gulf of Finland, activity concentration of ^{210}Po was four time higher and ^{210}Pb almost the same (a little lower) than that in the freshwater mussel *(Anodonta sp.).* Among the organisms studied by us, the highest activity concentration of ^{210}Po was found in crustacean *Saduria entomon.*

Table 3.1a. *Activity concentrations of ^{210}Po and ^{210}Pb in lake water samples.*

Lake	Ref. date	^{210}Po Bq/kg f.w. ± unc %	^{210}Pb Bq/kg f.w. ± unc %
Myllyjärvi	27.06.2007	0,0019 ± 17 0,0021 ± 17 **0,002**	0,0031 ± 17 0,0034 ± 17 **0,0033**
Vesijakojärvi	27.06.2007	0,0015 ± 17 0,0017 ± 17 **0,0016**	0,0031 ± 17 0,0028 ± 17 **0,003**
Iso-Ahvenainen	26.06.2007	0,0020 ± 17 0,0019 ± 17 **0,002**	0,0032 ± 17 0,0031 ± 17 **0,0032**
Miestämö	26.06.2007	0,0019 ± 17 0,0019 ± 17 **0,0019**	0,0033 ± 17 0,0029 ± 17 **0,0031**

Table 3.1b. *Activity concentrations of ^{210}Po and ^{210}Pb in freshwater fish in 2007.*

Fish	Lake	Parts analyzed	dry matter %	^{210}Po Bq/kg f.w. ± unc %	^{210}Pb Bq/kg f.w. ± unc %
Perch	Vesijakojärvi	edible parts other parts **whole fish**	27,67 33,72	0,139 ± 19 3,632 ± 18 **1,345**	0,057 ± 17 0,161 ±17 **0,093**
Pike	Myllyjärvi	edible parts	22,22	0,939 ± 18	0,075 ± 18
Pike	Iso-Ahvenainen	edible parts	22,29	0,428 ± 18	0,045 ± 18
Pike	Vesijako	edible parts other parts **whole fish**	21,70 25,45	0,664 ± 18 2,905 ± 18 **2,152**	0,115 ± 18 0,140 ± 17 **0,132**
Pike	Vesijako	edible parts	23,59	1,157 ± 18	0,056 ± 18
Pike-perch	Vesijako	edible parts other parts **whole fish**	23,49 33,27	0,079 ± 22 1,492 ± 18 **1,015**	0,014 ± 19 0,123 ± 17 **0,086**
Bream	Iso-Ahvenainen	edible part	22,14	0,138 ± 19	0,053 ± 19
Bream	Myllyjärvi	edible parts other parts **whole fish**	20,01 28,69	0,380 ± 18 8,950 ± 18 **6,532**	0,130 ± 18 1,507 ± 16 **1,119**
Bream	Vesijako	edible part	21,68	0,860 ± 19	0,047 ± 17
Vendace	Vesijako	edible part	24,8	1,863 ± 19	0,697 ± 16
Whitefish	Iso-Ahvenainen	edible part	25,31	0,157 ± 20	0,030 ± 18

Table 3. 2. *^{210}Po and ^{210}Pb in mussel and water collected from Lake Keurusselkä in 2007.*

Sample	Organs	dry matter %	^{210}Po Bq/kg d.w.	^{210}Pb Bq/kg d.w.	^{210}Po Bq/kg f.w. ± unc %	^{210}Pb Bq/kg f.w. ± unc %
Mussel, *Anodonta*	Soft tissue	7,83	73,2	18,0	**5,73** ± 16	**1,41** ± 16
	Shell mean	93,54	2,63 2,78	3,44 2,22	2,46 ± 17 2,60 ± 17 **2,53**	3,21 ± 17 2,07 ± 17 **2,64**
	Whole mussel				**4,69**	**1,81**
Water					**0,0027** ± 17	**0,0034** ± 17

Table 3.3. *^{210}Po and ^{210}Pb in Baltic Sea water.*

Sampling place	**Depth**	**Ref. date**	**^{210}Po Bq/kg ± unc %**	**^{210}Pb Bq/kg ± unc %**
Gulf of Finland (LL3A)	surface	31.7.2006	<0,002	0,0019 ± 18
Gulf of Finland (LL3A)	near bottom	31.7.2006	<0,002	0,0019 ± 18
Gulf of Finland (JML)	surface	1.8.2006	<0,002	0,0015 ± 18
Gulf of Finland (JML)	near bottom	1.8.2006	<0,002	0,0019 ± 18
Bothnian Sea (EB1)	surface	2.8.2006	<0,002	0,0013 ± 19
Bothnian Sea (EB1)	near bottom	2.8.2006	0,0006 ± 21	0,0020 ± 18

Table 3.4. *^{210}Po and ^{210}Pb in fish in 2005, 3-8 parallel determinations on each sample. The arithmetic mean and standard deviation are highlighted.*

Fish	**Sampling Site**	**Ref. Date**	**Dry matter %**	**^{210}Po Bq/kg f.w. ± unc %**	**^{210}Pb Bq/kg f.w. ± unc %**
Perch	Bothnian Sea	30.10.2005	28,13	0,338 ± 14	0,0844 ± 14
				0,281 ± 14	0,0844 ± 14
				0,366 ± 14	0,0844 ± 14
				0,366 ± 14	0,0844 ± 14
				0,281 ± 14	0,0844 ± 14
				0,366 ± 14	0,0844 ± 14
				0,309 ± 14	0,1125 ± 14
				0,309 ± 14	0,0844 ± 14
				0,327 ± 3	**0,0879 ± 11**
Perch	Bothnian Sea	3.10.2005	19,54	0,0481 ± 21	0,0104 ± 21
				0,0389 ± 23	0,0119 ± 23
				0,0379 ± 34	0,0176 ± 34
				0,0416 ± 24	**0,0133 ± 30**
Vendace	Pyhäjärvi, Säkylä	30.10.2005	23,58	1,190 ± 17	0,0465 ± 17
				1,117 ± 14	0,0707 ± 14
				0,886 ± 17	0,0880 ± 17
				1,065 ± 15	**0,068 ± 31**
Whitefish	sold in store	30.6.2005	20,90	0,457 ± 17	0,0878 ± 17
				0,494 ± 17	0,0660 ± 17
				0,489 ± 17	0,1012 ± 17
				0,492 ± 17	0,0815 ± 17
				0,436 ± 17	0,1058 ± 17
				0,474 ± 5	**0,0884 ± 18**
Roach	Päijänne	13.6.2005	25,88	1,190 ± 17	0,285 ± 17
				1,510 ± 17	0,221 ± 17
				1,518 ± 17	0,208 ± 17
				1,496 ± 17	0,245 ± 17
				1,493 ± 16	0,249 ± 16
				1,498 ± 17	0,241 ± 17
				1,451 ± 9	**0,241 ± 11**

Table 3.5. *^{210}Po and ^{210}Pb in swan (Cygnus olor)and a crustacean Saduria entomon collected from Loviisa in 2007.*

Sample	Organs	dry matter %	^{210}Po Bq/kg d.w.	^{210}Pb Bq/kg d.w.	^{210}Po Bq/kg f.w. ± unc %	^{210}Pb Bq/kg f.w. ± unc %
Swan	Breast muscle	25,40	2,62	0,257	0,66 ± 18	0,065 ± 18
			2,37	0,300	0,60 ± 18	0,076 ± 18
	mean				**0,63**	**0,07**
	Liver	28,44	27,7	1,399	7,89 ± 17	0,40 ± 17
			27,5	1,613	7,82 ± 17	0,46 ± 17
	mean				**7,85**	**0,43**
	Bones	82,82	12,0	6,94	9,92 ± 16	5,75 ± 16
			2,28	8,38	1,89 ± 16	6,94 ± 16
	mean				**5,90**	**6,34**
	Whole swan*				**1,03**	**0,37**
Saduria Entomon		22,93	81,2	6,72	18,6 ± 16	1,54 ± 16
			81,1	6,02	18,6 ± 16	1,38 ± 16
	mean				**18,6**	**1,5**
Water					**<0,002**	**0,0019**± 18

* estimated assuming that feather, skin and muscles have same concentration than breast muscle (pectoral).

Concentration ratios, CR

Concentration ratios CR (CR = activity concentration of a radionuclide in organism Bq/kg f.w. / activity concentration of the radionuclide in water Bq/kg) for ^{210}Po and ^{210}Pb in various organisms are given in Tables 3.6a, 3.6b, 3.6c, 3.6d. CRs for ^{210}Po in freshwater fishes ranged from 634 to 11252 and those for ^{210}Pb from 16 to 386 (Table 3.6a). The ratio of ^{210}Po contents in whole fish to that in edible parts of fish varied from 3.2 to 17.2 and that of ^{210}Pb from 1.1 to 8.6. Average CRs of ^{210}Po and ^{210}Pb for various species of freshwater fishes (whole fish) from Table 3.6a are summarized in Table 3.6.

Only edible parts of the fish were analyzed for vendace and whitefish. The results were converted to activity in whole fish using 'whole fish/edible part' ratio obtained from perch. This may cause overestimation of the corresponding CR values for vendace and whitefish. For perch from the Baltic Sea CR of ^{210}Po was 3.5 times and that for ^{210}Pb 1.5 times higher than that for freshwater perch (Tables 3.6a and 3.6d). For freshwater mussel, CR of ^{210}Po in soft tissue was about twice that in shell, while CR of ^{210}Pb in shell was about twice that in soft tissue. Estimated for the whole organism, CR of ^{210}Po was about three times that of ^{210}Pb (Table 3.6b). In swan ^{210}Pb was found to accumulate into bones and ^{210}Po into both liver and bones (Table 3.6c). The bottom animal, *Saduria entomon*, was estimated to have the highest CRs for both ^{210}Po and ^{210}Pb among the organism studied here.

Table 3.6a. *CRs of ^{210}Po and ^{210}Pb for freshwater fishes on fresh weight basis.*

Lake	**Fish**	**Parts analyzed**	**CR± unc %, ^{210}Po**	**CR ± unc %, ^{210}Pb**	**^{210}Po ratio** whole fish/ edible parts	**^{210}Pb ratio** whole fish/ edible parts
Vesijako	Perch	edible parts other parts **whole fish**	87,0 ± 25 2270 ± 25 **841**	19,2 ± 24 54,4 ± 24 **31**	9,67	1,63
Vesijako	Pike	edible parts other parts **whole fish**	415 ± 25 1820 ± 25 **1345**	39,1 ± 25 47,5 ± 25 **45**	3,24	1,14
Vesijako	Pike	edible parts **whole fish***	723 ± 25 **2345**	19,0 ± 25 **22**		
Myllyjärvi	Pike	edible parts **whole fish***	469 ± 25 **1520**	23,0 ± 25 **26**		
Iso-Ahvenainen	Pike	edible parts **whole fish***	 220 ± 25 **712**	 14,2 ± 25 **16**		
Vesijako	Pike-perch	edible parts other parts **whole fish**	 50 ± 28 932 ± 25 **634**	 4,9 ± 25 41,7 ± 24 **29**	12,78	6,03
Iso-Ahvenainen	Bream	edible parts **whole fish ****	71 ± 25 **1220**	16,9 ± 25 **145**		
Myllyjärvi	Bream	edible parts other parts **whole fish**	190 ± 25 4470 ± 25 **3270**	40,0 ± 25 464 ± 23 **344**	17,20	8,61
Vesijako	Bream	edible parts **whole fish****	538 ± 25 **9250**	15,9 ± 24 **137**		
Vesijako	Vendace	edible parts **whole fish#**	1160 ± 25 **11300**	236 ± 24 **386**		
Iso-Ahvenainen	Whitefish	edible parts **whole fish#**	778 ± 26 **7520**	9,6 ± 25 **16**		

* values are estimated using the ratios of whole fish to edible part for pike
** values are estimated using the ratios of whole fish to edible part for bream
# values are estimated using the ratios of whole fish to edible part for perch

Table 3.6b*. CRs of Po and Pb for freshwater mussel (Anodonta sp.) on fresh weight basis.*

Organism	**CR ± unc %, ^{210}Po**	**CR ± unc %, ^{210}Pb**
Mussel, soft tissue	2120 ± 23	414 ± 23
Mussel, shell	937 ± 24	777 ± 24
Mussel, whole	1735	533

Table 3.6c. *CRs for ^{210}Po and ^{210}Pb for swan and Saduria Entomon on fresh weight basis.*

Sample	**Parts analyzed**	**CR± unc %, ^{210}Po**	**CR± unc %, ^{210}Pb**
Swan	Pectoral Liver Bones Whole swan*	>320 >3900 >2950 >510	37 ± 25 225 ± 25 3340 ± 24 192
Saduria Entomon		>9300	769 ± 24

* estimated assuming that feather, skin and muscles have same concentration than pectoral.

Table 3.6d. *CRs of ^{210}Po and ^{210}Pb for brackish water fishes on fresh weight basis.*

Sampling place	Fish	Parts analyzed	CR± unc %, ^{210}Po	CR± unc %, ^{210}Pb
Bothnian Sea	Perch	edible parts **whole fish***	545 ± 25 **5270**	52 ± 23 **84**
Bothnian Sea	Perch	edible parts **whole fish***	69 ± 31 **670**	7,8 ± 29 **13**

* values are estimated using the ratios of whole fish to edible part in freshwater perch

Table 3.6. *Average CRs for ^{210}Po in freshwater fishes (whole body)*

Species	^{210}Po	^{210}Pb
Pike	1481	27
Perch	841	31
Pike-perch	634	29
Bream	4577	209
Vendace	11252	386
Whitefish	7518	16

3.2.5 Discussion

In the ERICA project concentration ratios (CR) in marine and freshwater environment for selected biota types were collated by literature review and missing data were estimated (Hosseini et al., 2008). Concentration ratios calculated in this study are compared with the ones derived in ERICA in Table 3.7a and 3.7b. Both freshwater and marine CRs derived in ERICA are shown in Table 3.7a. Marine CRs were, in most cases, significantly larger than freshwater CRs. Our CRs for ^{210}Po in brackish water (Table 3.7a) were generally between these two values. Concentration ratios of ^{210}Pb in brackish water were 10 times smaller than the values estimated in ERICA for freshwater environment. As shown in Table 3.7b, CRs of ^{210}Po for freshwater fish were higher than previously estimated, while CRs for ^{210}Pb in freshwater environment agreed relatively well with the values from ERICA.

Table 3.7a. *Concentration ratios (fresh weight) for brackish water biota (whole organism).*

	CR, ^{210}Po		CR ^{210}Pb	
Organism	**this study**	**ERICA**	**this study**	**ERICA**
fish	**670 - 5300**	44000±12000* 240**	**13-84**	4400±14000* 300**
crustacean	**>9300**	56000±66000* 9900 ±1400**	**770**	7500±2100* 7500**
bird	**>510**	30000* 240**	**190**	19000* 300**

* marine
** freshwater

Table 3.7b.*Concentration ratios (fresh weight) for freshwater biota (whole organism).*

	CR, ^{210}Po		CR ^{210}Pb	
Organism	**this study**	**ERICA**	**this study**	**ERICA**
fish benthic	**630-9250**	240	**16-340**	300
mollusc bivalve	**1740**	38000±49000	**530**	1400

There has been an increasing interest on behaviour of polonium in aquatic environment (Ryan et al., 2008; Connan et al., 2007; Skwarzec and Fabisiak, 2007; Suriyanarayanan et al., 2008). Connan et al.,

(2007) studied the distribution of polonium between different fish organs and noticed, similarly to us, that highest accumulation of ^{210}Po in fish is in the non-edible parts of the fish e.g. skin, intestines, liver. Higher accumulation of Po in soft tissues of molluscs than in shells has also been noticed by Suriyanarayanan et al (2008) who, similarly to us, also noticed that Pb was much less accumulated in molluscs and mostly in shell.

4 Po-210 in the terrestrial environment

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4.1 Review of existing information

In recent years, there have been intensive activities on a global basis in relation to the development of methodologies for assessing impacts of ionising radiation on the environment. At a European level, the EURATOM-funded project ERICA, Environmental Risk from Ionising Contaminants: Assessment and Management [Larsson, 2008) has proven to be a driving force in this respect. A key activity in the ERICA project was to consider the transfer of radionuclides through food-chains, irrespective of whether humans constitute a component of the food-chain or not, with special focus on provision of data for reference organisms. These organisms, representing the broader ecosystem, provide a basis for radiation dose rates estimations from a contaminated environment.

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). 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 in combination with 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 would be interpreted on the basis 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).

Numerous data deficiencies have been uncovered in addressing issues related to the characterisation of background dose rates to reference organisms. However, on further inspection it becomes evident that some of these data deficiencies could be easily mitigated with limited effort involving field-work and analysis. The objective of this work was therefore to identify data gaps in relation to the levels and transfer of naturally occurring radionuclides in terrestrial ecosystems (with a focus on the important dose-forming radionuclides ^{210}Po and ^{210}Pb) and to plan and conduct a terrestrial field campaign with the purpose of filling some of these information gaps. This would facilitate the calculation of more robust background dose-rates for selected terrestrial system hitherto not studied.

Lead-210 and its granddaughter ^{210}Po are members of the ^{238}U decay series. They are formed in the atmosphere following decay via a number of intermediate short-lived radionuclides from gaseous ^{222}Rn. Deposition of ^{210}Pb, associated with aerosols in the atmosphere, occurs via meso-scale transportation process, sedimentation and precipitation. In Scandinavia, the radionuclide is deposited continuously to earth at a rate of approximately 55 Bq m^{-2} per annum (El-Daoushy, 1988). Early models concerning the atmospheric ^{210}Pb transport were based on the vertical movement of the radioisotope into the troposphere at the equator followed by lateral movement to mid-latitudes and deposition, whereas more recent, refined models have included regional sources of ^{222}Rn to account more robustly for the ^{210}Pb deposition rates observed across the major continents (see Macdonald et al., 1996). Atmospheric ^{210}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 ^{210}Pb (El-Daoushy, 1988). Furthermore, the deposition of ^{210}Pb is directly correlated with the level of precipitation (Hill, 1960).

For mountainous areas dominated by Alpine vegetation, lichen appears to play a key role in the introduction of ^{210}Pb into the foodchain. Lichens are slow growing perennials that have high interception potentials for aerosols in precipitation, and therefore contain significantly higher ^{210}Pb concentrations than vascular plants (Skuterud et al., 2005). Animals feeding on lichens, notably reindeer have been shown to have relatively high muscle and organ ^{210}Po and ^{210}Pb activity concentrations (Skuterud et al., 2005). For animals not feeding on lichen, entry of ^{210}Pb and ^{210}Po into the foodchain presumably primarily occurs through the ingestion of vegetation (with relatively low concomitant activity concentrations relative to lichen), dust and soil.

4.2 Identification of knowledge gaps

The empirical data coverage (Concentration ratios, CRs, and thereby activity concentrations in plants and animals) for selected radionuclides provided by the ERICA project for terrestrial environments is presented by Beresford et al. (2008). The coverage for Pb was found to be reasonable, presumably reflecting the large number of stable element studies that have been conducted on this element. Other radioelements were more poorly characterised with empirical data sets. In the case of Polonioum, some information was available for flora but only for the fauna group mammals. In the latter case it should be noted that although tens of data values are available these represent "all mammals" from a single geographical area - the UK. Although numerous data exist for reindeer, these data were excluded in the work of Beresford et al. (2008) as the air–lichen–reindeer pathway was considered unlikely to be representative of contamination routes for other terrestrial mammals and was therefore likely to result in over predictions for the mammal reference organism category. The number of values associated with Thorium was found to be low. In all cases the number of available empirical values was below 20 and for 7 reference categories no information was available at all. A similar situation existed for Uranium although, arguably, floral reference organisms were characterised by reasonable CR information. For radium there were severe data deficiencies for invertebrates, insects, amphibia and reptiles.

The Environment Agency of England and Wales recently commissioned work to develop databases to underpin environmental impact assessment using reference animals and plants (Beresford et al., 2007). ICRPs Reference Animal or Plant (RAP) are defined as "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, physiological and life history properties that can be used for the purposes of relating exposure to dose and dose to effects for that type of living organism" (ICRP, 2007). In considering an overview of these data, there were no data for some Reference animals and plants, 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. However, no new measurements of ^{210}Po were made in the study. An overview of data availability for UK biota based on a literature review (Beresford et al., 2007) is presented in Table 4.1.

Table 4.1. *Observations of ^{232}Th and ^{238}U decay series radionuclides from Beresford et al., 2007.*

	Po-210	Pb-210	Ra-226	Th-230	Th-232	U-234	U-238
Duck	no data	no data	no data	N ≤ 10	N ≤ 10	no data	no data
Pine tree	N ≤ 10	N ≤ 10	N ≤ 10	N ≤ 10	N ≤ 10	N ≤ 10	N ≤ 10
Wild grass	N > 10	N > 10	N > 10	N ≤ 10	N > 10	N > 10	N > 10
All mammals	N > 10	N > 10	no data	N ≤ 10	N > 10	N ≤ 10	N ≤ 10
Deer	N ≤ 10	no data	no data	no data	no data	no data	no data

4.3 Experimental studies of ^{210}Po in small rodents

4.3.1 Sampling area

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 (Figure 4.1). The field study was conducted within a designated Landscape-protected area near to Kongsvold adjacent to Dovrefjell-Sunndalsfjella National Park. 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.

Eight soil profiles were collected during the field expedition. These profiles were split into an overlying humus layer and thereafter 3 cm (predominantly mineral soil) increments to a depth of 9 cm using a custom-designed soil corer. This was undertaken with a view to enabling analyses of the activity distribution of radionuclides with depth. Baited traps were used in the collection of various small mammals including Bank Vole (*Clethrionomys glareolus*) and the Common Shrew (*Sorex araneus*). Plant samples including samples of bilberry (*Vaccinium myrtillus*) and 2 species of lichens (e.g. *Cladonia stellaris* and *Cladonia arbuscula*) 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.

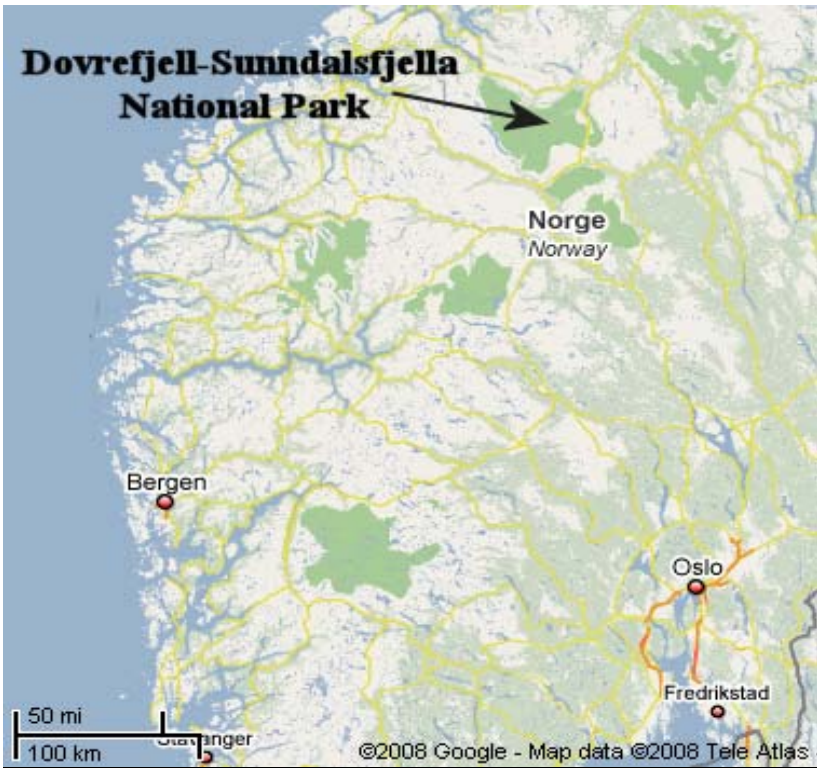

Figure 4.1. *Sampling location – Dovrefjell-Sunndalsfjella National Park*

4.3.2 Analyses of ^{10}Po and ^{210}Pb

A detailed description of the analysis of polonium in the samples is given in Chen *et al*., 2001. Samples were measured at the RISØ National Laboratories in Denmark. Briefly, the freeze-dried material (2-10g) was added to a semi-closed glass flask, ^{208}Po and a known amount of stable Pb (5-10mg) added as yield determinants for ^{210}Po and ^{210}Pb respectively. The sample was completely dissolved using a mixture of HNO_3, HCl and H_2O_2, evaporated to near dryness and polonium plated onto silver discs in a weak hydrochloric solution. The discs were then analysed without delay using solid state PIPS-detectors. The solution remaining after plating onto the Ag-discs were rinsed from remaining traces of polonium using TIOA-extraction in 10M HCl. The aqueous phase containing Pb was set aside for ^{210}Po ingrowth. Yield recoveries were in the range of 63 to 88 %. The overall uncertainty in the small mammal measurements are estimated to be in the range 10 to 20 %.

4.3.3 Results

Only preliminary data for small mammals were available at the time of writing of this paper. When deriving activity concentrations of ^{210}Po for the time of sampling, a simple decay correction procedure cannot be used because the ^{210}Po attributable to the whole body of the samples at the time of analysis is likely to be comprised of activity remaining from the decay of unsupported ^{210}Po and a component arising from ingrowth via ^{210}Pb (between the time of sampling and the time of analysis). Essentially, we can assume that some ^{210}Pb is present in the samples (organs within the body such as the liver are known to accumulate stable and therefore radioactive lead) and that the *in situ* decay of this radioisotope is contributing to the activity measured at the time of analysis.

Since only ^{210}Po results were available at the time of writing of this chapter, the results reported here (provisionally decay corrected to date of sampling) are of a preliminary nature only. Once the level of ^{210}Pb has been determined for the samples, the unsupported activity concentration for ^{210}Po at the time of sampling can be determined precisely.

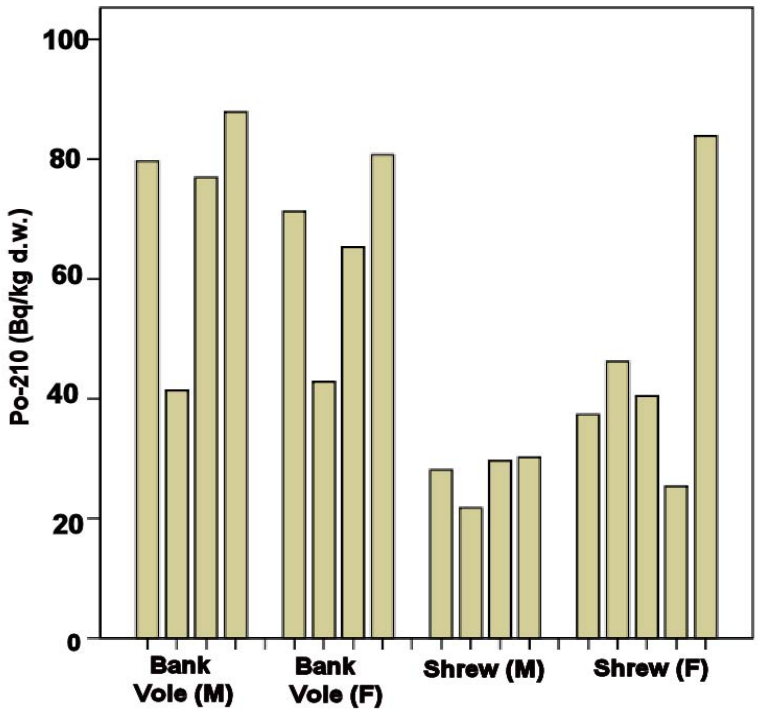

Figure 4.2. *Po-210 activity concentrations in bank vole and common shrew. M= male and F =Female*

The preliminary data exhibit activity concentrations in the range 41 – 88 Bq kg^{-1} d.w. for bank vole ^{210}Po and 22 – 84 Bq kg^{-1} d.w. ^{210}Po for the common shrew (Figure 4.2). The non-parametric Mann-Whitney test has been applied in order to determine whether data ^{210}Po activity concentrations in shrew are statistically different to corresponding data for bank vole. The null hypothesis is that these 2 samples have been taken from a common population so that there is no consistent difference between the 2 sets of data. Since there is no hypothesis concerning whether the mean rank of one population is greater or less than the other, a two tailed test was considered appropriate.

Median activity concentrations of ^{210}Po in bank vole (74 Bq kg^{-1} d.w.) appear to be significantly different to those determined for shrew (30 Bq kg^{-1} d.w.). This is confirmed by the fact that the null hypothesis can be rejected at the $p < 0.008$ level, i.e. the probability that the 2 sets of data come from the same population is extremely low and it is reasonable to conclude that bank vole and shrew ^{210}Po data constitute different populations with different mean ranks.

4.3.4 Discussion

Bank voles have a broad diet, which is mainly herbivorous, including fruit, soft seeds, leaves, fungi, roots, grass, buds and moss. They may also occasionally take invertebrate food such as snails, worms and insects may be eaten. The common shrew feeds on most terrestrial insects, but will also take worms, slugs and snails. The statistical tests undertaken in this study appear to highlight the importance of diet in terms of influence on body burdens of ^{210}Po. It appears that the primarily herbivorous bank vole is accumulating higher concentrations of these natural radionuclides compared to the insectivorous shrew. Because the metabolic rate of these small mammals is expected to be fairly similar, relative to poikilotherms of similar size or homeotherms of different sizes, it may be hypothesized that the biological half-lives for ^{210}Po in shrew and bank vole are also quite similar. If this holds true, the activity concentration of ^{210}Po in the food consumed by these small mammals would appear to be driving the differences observed.

The ^{210}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^{-1} d.w. in female reindeer muscle and several hundred Bq kg^{-1} d.w. in liver were determined (Skuterud et al., 2007).

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^{-1} f.w. on average for Bank Vole and 9 Bq kg^{-1} 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^{-1} 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 or differences in deposition of ^{210}Pb between the study areas, remains a subject for further investigation.

Unweighted dose-rates have been derived using the dose assessment methodology described by Brown et al. (2008). No alpha radiation weighting factor has been applied because international consensus on the magnitude of this value has not been attained despite the consideration of such values in the open literature (see Chambers et al.,, 2006). Background radiation dose rates to Bank Vole (whole-body) attributable to the presence of internally distributed ^{210}Po were calculated to be 0.07 μGy h^{-1}.

4.4 Experimental studies of ^{210}Po and ^{210}Pb in viper and frog

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4.4.1 Samples

Some terrestrial biota samples were collected from the environments of Finnish nuclear power plants. A sample representing amphibians, a frog *(Rana temporaria)* and a sample representing reptiles, a viper *(Vipera Berus)*, were taken from Olkiluoto.

4.4.2 Sample treatment

Samples of viper and frog were dried, homogenized, digested and analyzed as a whole animal. Analytical procedure used in STUK to determine ^{210}Po and ^{210}Pb as described in the studies for the freshwater environment - Chapter 3, above.

4.4.3 Results

Two parallel analyses were carried out on viper and frog samples (Table 4.2). Activity concentration of ^{210}Po was higher than ^{210}Pb in both organisms. Total amounts of ^{210} Po and ^{210}Pb in the viper were 0.43 and 0.25 Bq and those in the frog were 0.12 and 0.065 Bq, respectively. Activity concentrations of ^{210}Po and ^{210}Pb in soil were 38 and 41 Bq/kg f.w. Concentration ratios for viper and frog (Table 4.3) were calculated using the soil concentration as the denominator in the CR calculations.

Table 4.2 *Activity concentrations of ^{210}Po and ^{210}Pb in viper, frog and soil from Olkiluoto in 2007.*

Sample type	**Latin name**	**Dry matter %**	**^{210}Po Bq/kg d.w.**	**^{210}Pb Bq/kg d.w.**	**^{210}Po Bq/kg f.w. ± unc %**	**^{210}Pb Bq/kg f.w. ± unc %**
Viper	*Vipera Berus*	24,75 24,75	20,90 21,84	13,05 12,09	5,17 ± 17 5,41 ± 17 **5,29**	3,23 ± 17 2,99 ± 17 **3,11**
Frog	*Rana temporaria*	29,15 29,15	5,65 5,88	3,39 2,98	1,65 ± 17 1,71 ± 17 **1,68**	0,99 ± 17 0,87 ± 17 **0,93**
Soil		92,75 92,75	42,87 39,73	42,48 46,05	39,8 ± 16 36,9 ± 16 **38,31**	39,4 ± 16 42,7 ± 16 **41,06**

Table 4.3*. CRs of Po and Pb for viper and frog (whole organism). CR = Bq/kg in organism / Bq/kg. in soil. Fresh weight masses are used for both organism and soil when CRs are calculated as f.w. and dry weights are used for both organism and soil respectively when CRs are calculated as d.w.*

Organism	**CR ± unc %, (f.w.)**		**CR ± unc %, (d.w.)**	
	^{210}Po	**^{210}Pb**	**^{210}Po**	**^{210}Pb**
Viper, *Vipera Berus*	0,138 ± 23	0,076 ± 23	0,517	0,284
Frog, *Rana temporaria*	0,044 ± 23	0,023 ± 23	0,140	0,072

5 Polonium in man

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5.1 Human data

Most data were produced during the 1960-170ies. We must the take into account that seldom semi conductor detectors were used, i.e. the background was higher. Yield determinants such as ^{208}Po or ^{209}Po were not used. One assumed almost 100 % recovery after 2 platings which is not true and underestimates the concentrations with a factor of 10-30%

Results for ^{210}Pb are complicated by two factors, build up of ^{210}Po from ^{210}Pb during storage of samples and *in vivo* build up from ^{226}Ra and ^{210}Pb.

The food-chain Lichen- Reindeer-Man has been investigated. The average concentrations for lapps of ^{210}Po in lungs were about 200 mBq per kg wet weight, 670 mBq/kg wet weight in liver and 1100 mBq/kg wet weight in bone (Mussalo et.al, 1978). For non-lapps the concentrations were 114 mBq/kg in lungs and 300 mBq/kg in liver. This can be compared with that (1965) found 440 - 550 mBq/kg in liver from non-smokers and 550 – 740 mBq/kg for smokers.

Hill(1966) measured ^{210}Po and ^{210}Pb in human placenta. The concentrations in persons eating large amounts of reindeer products had 80 times higher concentrations (7,4 Bq/kg w.w.) than those who did not consume reindeer products. The activity ratio ^{210}Po/^{210}Pb was around 4. He also reported data for for residents in UK. such as 600 mBq/kg in liver, 650 mBq/kg in kidney 180 mBq/kg in lungs, 150mBq/kg in testis and 750 mBq/kg in bone. Specific attention was given to radium dial painters. Such workers had 10-280 times higher concentrations than non-exposed people with the highest for hair (900 Bq/kg). Most investigations show that the main source for ^{210}Po in the body is food intake and not by inhalation or cigarette smoking. However, Holtzman (1964) estimated that inhalation is a significant source for ^{210}Pb in human bone.

The activity ratio in human bone ^{210}Po/^{210}Pb is about unity (Holtzman, 1972) and he found activity concentrations of about 400 mBq/kg w.w. in liver, 220 mBq/kg in muscle and 7000 mBq/kg (ash weight) in bone. Cortical bone had lower concentrations than trabecular bone.

5.2 Intake and excretion of ^{210}Po

The intake of ^{210}Po and ^{210}Pb was estimated to about 150 mBq/d (Beowulf *et al*., 1966). Hill (1965) gave a range of 40-400 mBq/d with an average of 120 mBq/d for residents of UK. Interestingly he also suggested that Po is associated with proteins especially those with the highest content of –SH groups. The renal excretion of ^{210}Po from Northern Canadians varied from 15 to 440 mBq per day (Holtzman and Ilcewicz, 1971) depending on if they were Indian, Caucasian or Eskimos. The excretion rates of ^{210}Pb were generally a factor of 2-5 lower.

Holtzman (1970) estimated the biological halftime of ^{210}Po in radium-dial painters to 7 years which is extremely long compared to other estimations. In this case Polonium and Radiolead were supported by the ^{226}Ra in the body

5.3 Experimental studies of polonium in man

The aim of the project was 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. If we could establish these radiobiological parameters it would be possible to calculate or estimate the biological effect of Polonium to the human body. Several studies have been made on polonium-210 in human body. Important parameters as 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±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 ^{209}Po and urine sampling stopped and 24h faeces sampling for a week begun. The results showed clearly a slow decreasing excretion of ^{209}Po in faeces in the range 0.59%-0.07% of consumed activity. Urine samples analysis showed a fluctuating value of ^{209}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.

All results from the studies are presented in figure 5.1 to 5.8.

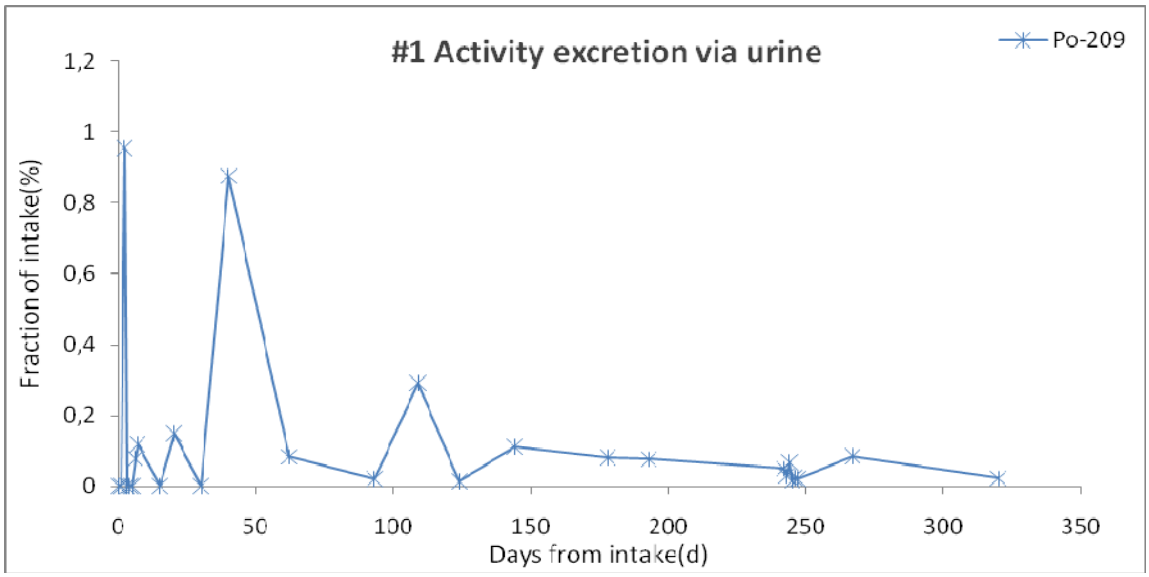

***Figure 5.1**. Excretion of Polonium 209 via urine from person #1 administrated 50 mBq per day.*

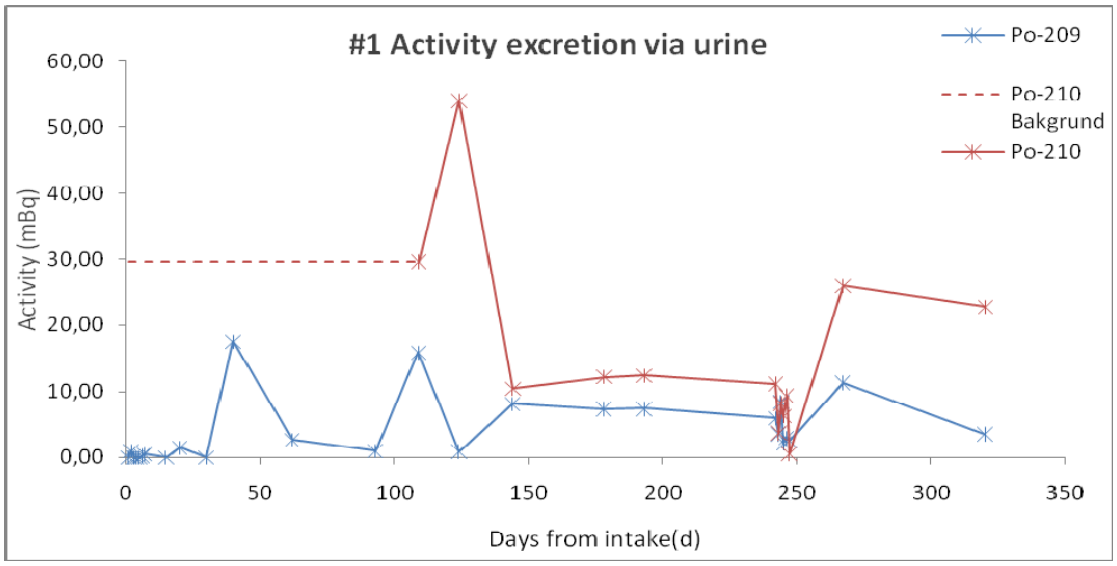

***Figure5. 2**. Excretion of Polonium 209 and Po 210 via urine from person #1 administrated 50 mBq per day.*

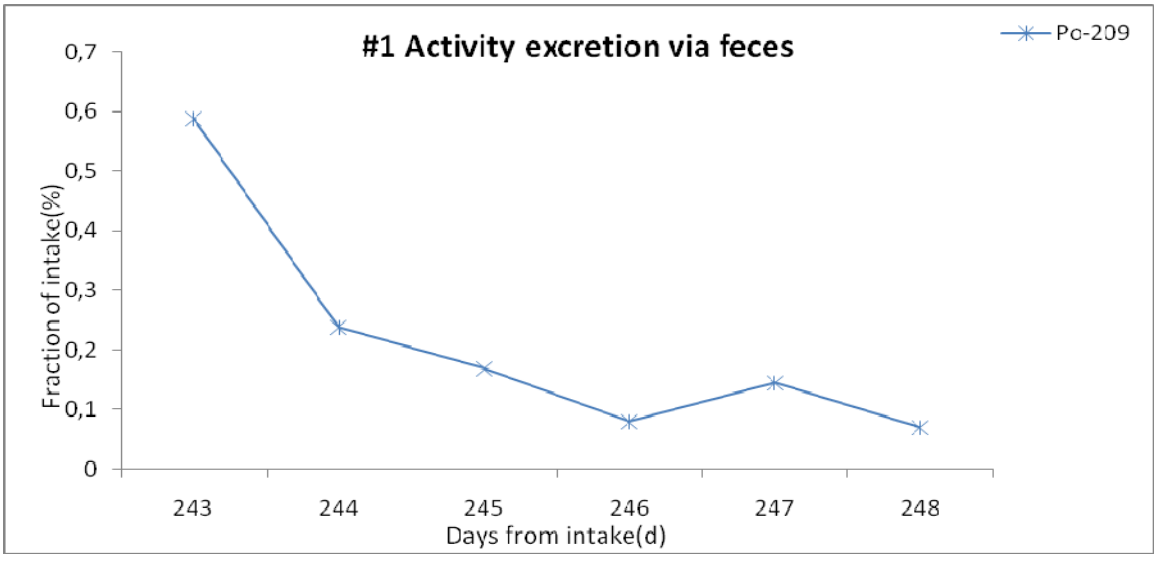

***Figure 5.3**. Excretion of Polonium 209 via faeces from person #1 administrated 50 mBq per day.*

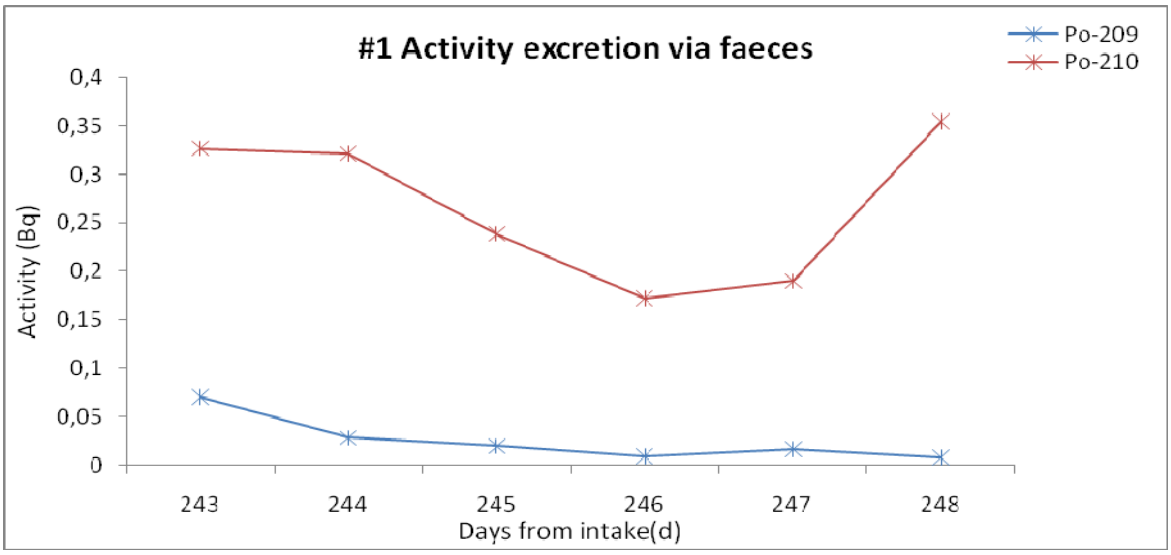

Figure 5.4. *Excretion in Bq of Polonium 209 and Polonium 210 via faeces from person #1 administrated 50 mBq per day.*

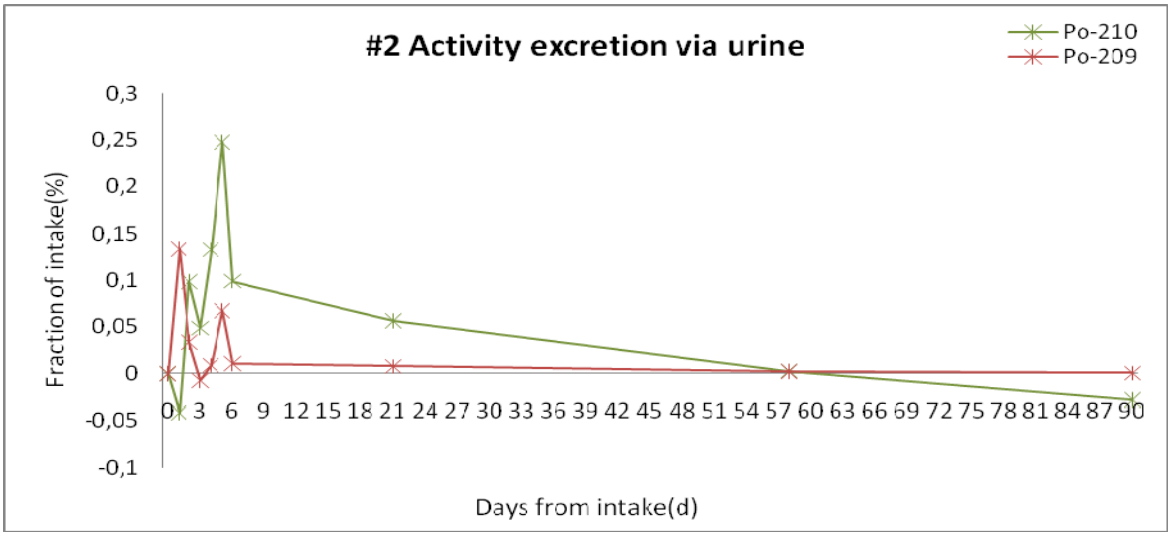

Figure 5.5*. Excretion of Polonium 209 and Polonium 210 via urine from person #2 administrated 10 Bq.*

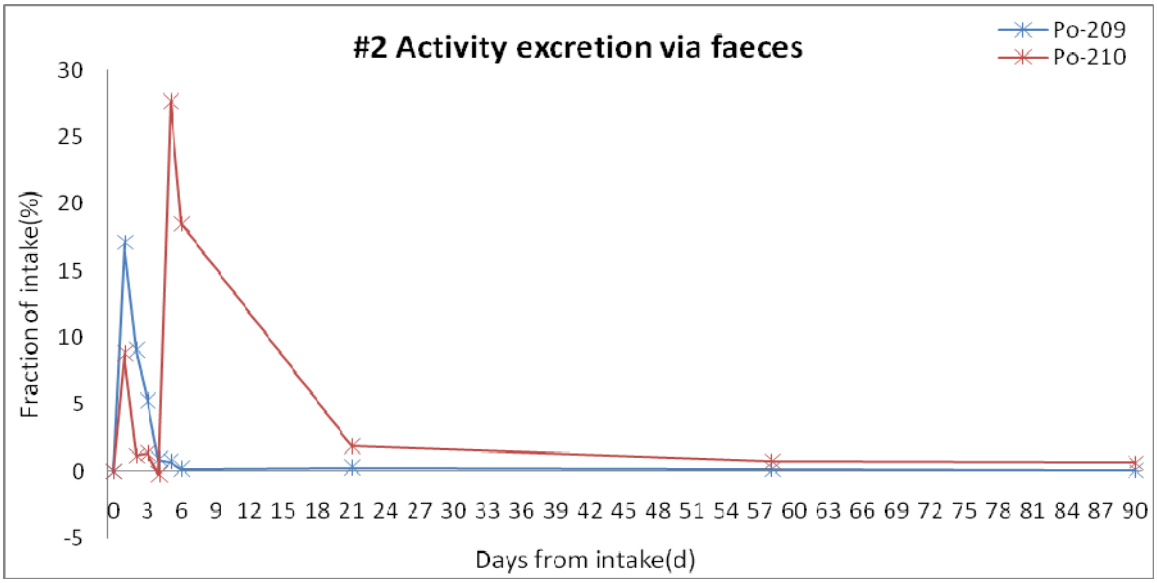

Figure 5.6. *Excretion of Polonium 209 and Polonium 210 via faeces from person #2 administrated 10 Bq.*

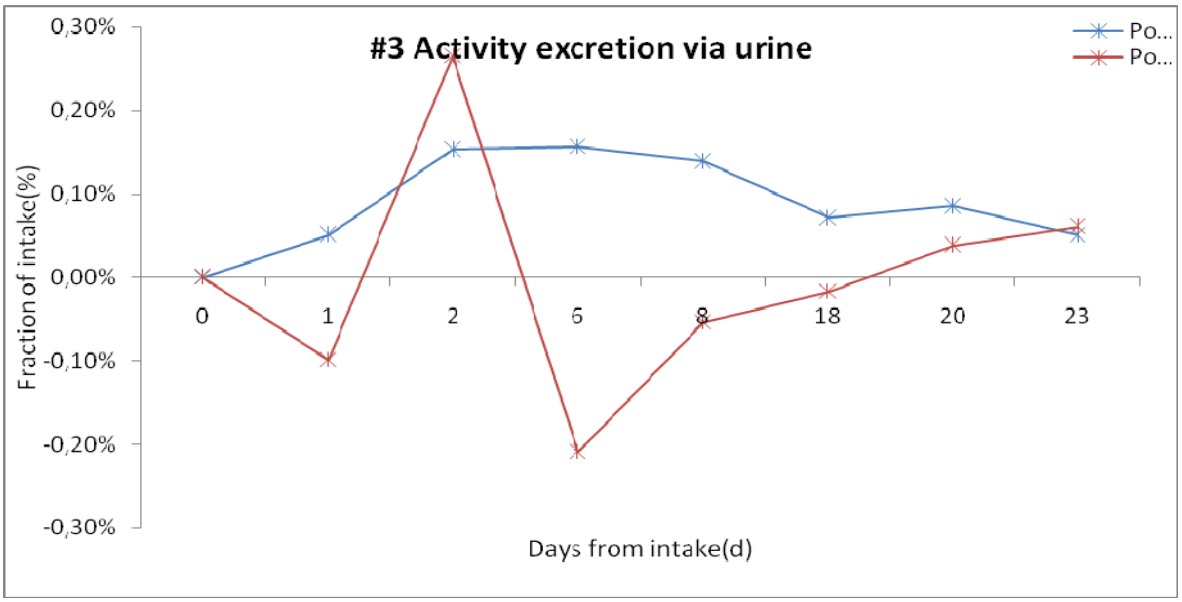

***Figure 5.7**. Excretion of Polonium 209 and Polonium 210 via urine from person #3 administrated 10 Bq.*

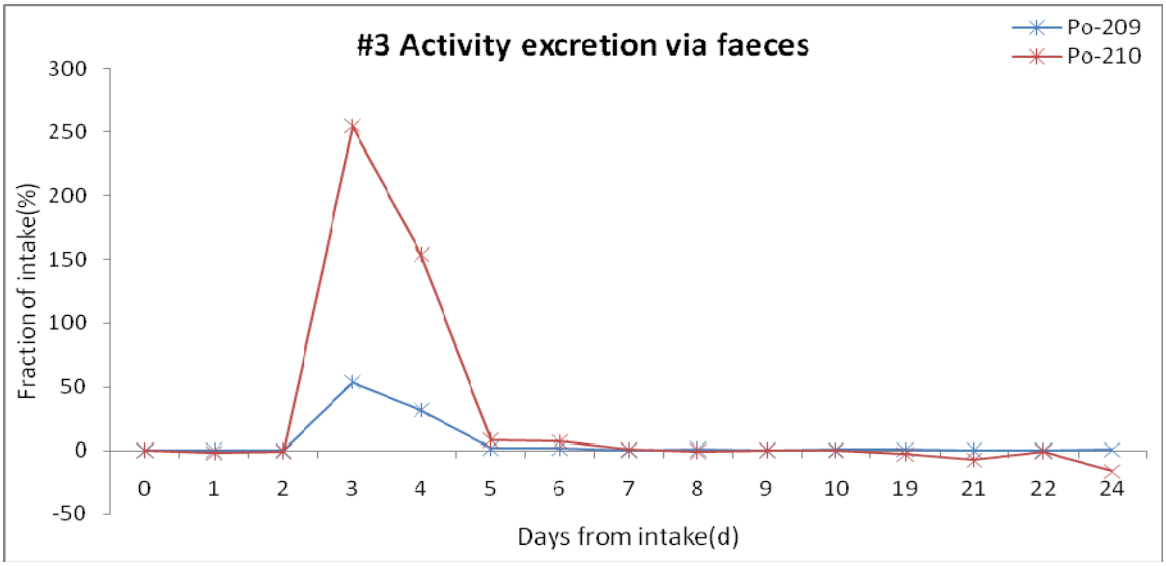

***Figure 5.8**. Excretion of Polonium 209 and Polonium 210 via faeces from person #3 administrated 10 Bq.*

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Abstract	This report provides new information on Po-210 (and where appropriate its grandparent Pb-210) behaviour in environmental systems including humans. This has primarily been achieved through measurements of Po-210 in aquatic and terrestrial environments that has led to the derivation of information on the levels of this radioisotope in plants, animals and the biotic components of their habitat (i.e. water, soil) providing basic information on transfer where practicable. For freshwater environments, Po-210 concentration ratios derived for freshwater benthic fish and bivalve mollusc were substantially different to values collated from earlier review work. For terrestrial environments, activity concentrations of Po-210 in small mammals (although of a preliminary nature because no correction was made for ingrowth from Pb-210) were considerably higher than values derived from earlier data compilations. It was envisaged that data on levels of naturally occurring radionuclides would render underpinning data sets more comprehensive and would thus allow more robust background dose calculations to be performed subsequently. By way of example, unweighted background dose-rates arising from internal distributions of Po-210 were calculated for small mammals in the terrestrial study. The biokinetics of polonium in humans has been studied following chronic and acute oral intakes of selected Po radioisotopes. This work has provided information on gastrointestinal absorption factors and biological retention times thus improving the database upon which committed effective doses to humans are derived. The information generated in the report, in its entirety, should be of direct relevance for both human and non-human impact assessments.
Key words	Po-210, environmental impact assessment, levels, transfer, concentration ratios, human biokinetics

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