



## Distribution of $^{210}\text{Pb}$ and $^{210}\text{Po}$ in boreal forest soil

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### ABSTRACT

Vertical distribution and activity contents of  $^{210}\text{Pb}$  and  $^{210}\text{Po}$  were investigated in forest soils of Scots pine-dominated (*Pinus sylvestris* L.) stands from seven different locations in Finland. The mean total inventory in the soil profile, up to 20 cm, of  $^{210}\text{Pb}$  was  $4.0 \text{ kBq m}^{-2}$  (range  $3.1\text{--}5.0 \text{ kBq m}^{-2}$ ) and  $^{210}\text{Po}$   $5.5 \text{ kBq m}^{-2}$  (range  $4.0\text{--}7.4 \text{ kBq m}^{-2}$ ), the organic soil layer containing 45% of the total inventory of both nuclides. In both the organic and the mineral layers the  $^{210}\text{Po}/^{210}\text{Pb}$  ratio was close to unity indicating a radioactive equilibrium between them. In the litter layer there was, however, a clear excess of  $^{210}\text{Po}$  suggesting that polonium is recycled via root uptake from the root zone to the ground surface. The activity concentration ( $\text{Bq kg}^{-1}$ ) of  $^{210}\text{Pb}$  clearly correlated with organic matter and the Fe, Al and Mn concentrations in soil indicating that radioactive lead is associated both with humic substances and the oxides of iron, aluminium and manganese. Radioactive lead was also seen to follow the behavior of stable lead. No systematic correlation between polonium and soil properties was seen.

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### 1. Introduction

Naturally occurring  $^{210}\text{Pb}$  (half-life 22.3 years) and  $^{210}\text{Po}$  (half-life 138 days) are present in the atmosphere as decay products of  $^{222}\text{Rn}$  (half-life 3.8 days). Both  $^{210}\text{Pb}$  and  $^{210}\text{Po}$  attach to aerosol particles in the atmosphere and deposit onto the ground as dry and wet deposition, which is their main source in surface soil (Avadhani et al., 2005). Soil, in turn, is a major source of these radionuclides entering the food chains (Gerzabek et al., 2003; Kirchner and Dailliant, 1998); thus studies on behavior and distribution of radionuclides in soil are of great significance.

Forest soils are typically podzols in the northern boreal zone. Podzolic soil has a characteristic layered profile: beneath the upper humus layer (O) is a base cation leached eluvial horizon (E) supported by an enriched illuvial horizon (B) and below the B horizon is the undisturbed C horizon and the bedrock (Lundström et al., 2000). In soil, radionuclides originating from atmospheric deposition migrate downward, both in soluble and colloidal forms, within the soil profile and sorb onto organic matter, clay minerals, and metal oxides. The top soil horizons, humus and mineral soil layers, are penetrated by living roots of plants and trees, which may absorb radionuclides (Kirchner and Dailliant, 1998).

Traditionally, the main emphasis in radioecological research has been on pollution radionuclides that have originated from events such

as nuclear weapons tests and the Chernobyl accident. Both  $^{210}\text{Pb}$  and  $^{210}\text{Po}$  have received limited attention, although in general their contribution to the human radiation exposure is comparatively high (Kirchner and Dailliant, 1998; Nielsen and Joensen, 2008; Aarkrog et al., 1997). In an earlier paper we investigated  $^{210}\text{Pb}$  and  $^{210}\text{Po}$  in wild berries and mushrooms at the same sites as in this study and found an excess of  $^{210}\text{Po}$  compared to its grandparent  $^{210}\text{Pb}$  in these plants (Vaaramaa et al., 2009). The present study focuses on vertical distribution and activity contents of  $^{210}\text{Pb}$  and  $^{210}\text{Po}$  in boreal forest soils at a number of different locations in Finland, and considers soil property correlation to the activity concentrations of  $^{210}\text{Pb}$  and  $^{210}\text{Po}$ .

### 2. Material and methods

#### 2.1. Study areas

Soil samples were taken from seven study areas in Finland (Fig. 1). The sites were in Scots pine-dominated (*Pinus sylvestris* L.) forests. Two of them were the main study areas; one in the Southern Finland (Parkano,  $62^{\circ}09.376'\text{N}$ ,  $22^{\circ}52.416'\text{E}$ ) and the other in the Northern Finland (Rovaniemi,  $66^{\circ}21.775'\text{N}$ ,  $26^{\circ}44.148'\text{E}$ ). Ten soil cores were taken from these sites, while only three or four soil cores were sampled from the other five sites located in Lammi, Joroinen, Viitasaari, Lieksa and Kuhmo. In addition, detailed soil characterization was done only for the soils taken from the two main sites. The soil type was sandy till in Parkano and sand in Rovaniemi and the bedrock on both sites is mostly granitic. The average tree age was approximately 80 years in Parkano and 160 in Rovaniemi and the dominating ground vegetation on both sites consist of *Vaccinium myrtillus* L. and *Vaccinium vitis-idaea* L.

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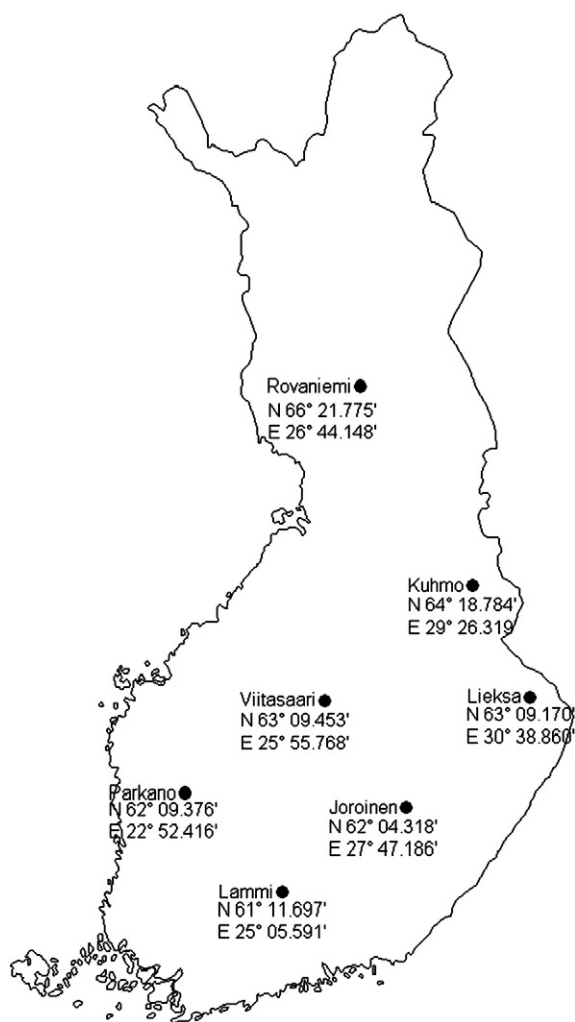


Fig. 1. Map depicting locations of the seven study sites.

## 2.2. Sampling and pre-treatment

Soil samples were collected from Parkano in June 2006, from Rovaniemi in September 2006 and 2007, and from the remaining sites (Lammi, Lieksa, Viitasaari, Joroinen, and Kuhmo) in September 2007. The soil profiles were gathered with a corer used to make holes on golf courses ( $\varnothing$  10.4 cm, length 22 cm) and separated into different soil horizons (litter L, organic O, and mineral soil layers E and B). The litter samples of Lammi, Lieksa, Viitasaari, Joroinen, and Kuhmo included moss. Within the soil profiles at all sites there was an ash-grey, eluvial horizon (E) below the organic layer (O) with the exception of Lammi where the E layer was undeveloped. Below the E layers was the reddish-brown illuvial horizon (B) which was divided into upper (B1) and lower parts (B2). Depth of the soil layers is presented in Table 1.

Table 1

Depth of soil layers (arithmetic mean) at each of the seven study sites (number of soil profiles are in parentheses).

Soil layer, cm	Lammi (4)	Joroinen (3)	Parkano (10)	Viitasaari (3)	Lieksa (3)	Kuhmo (3)	Rovaniemi (10)
O	0–3.5	0–3.3	0–3.8	0–4.7	0–3.3	0–2.7	0–2.3
E	3.5–7.3 <sup>a</sup>	3.3–12	3.8–10	4.7–12	3.3–12	2.7–9.5	2.3–7.9
B1	7.3–15	12–15	10–15	12–15	12–15	9.5–16	7.9–13
B2	15–19	15–20	15–19	15–18	15–19	16–20	13–20

<sup>a</sup> The E layer was unformed.

The soil profiles were classified haplic podzols of which two examples are given in Fig. 2.

Samples were dried (mineral soil at 40 °C and organic layers at 60 °C) and homogenized. The organic soil samples were pulverized in a Vita-Mix® blender, and the mineral soil samples were sieved with a 2 mm sieve to remove the gravel fraction; all analyses were performed on the <2 mm fraction. Dry matter contents were determined from subsamples of soil by drying in an oven at 105 °C for about 24 h. After drying, the samples were cooled in a desiccator and weighed. The results given later are expressed on a dry matter basis.

## 2.3. Analysis of soil properties

The  $^{210}\text{Pb}$  and  $^{210}\text{Po}$  levels were determined for all soil samples, while soil properties were only determined for the main study areas, Parkano and Rovaniemi. The total element concentrations (P, K, S, Ca, Mg, B, Cu, Zn, Mn, Na, Fe, Al, Cd, Cr, Ni, and Pb) were determined by wet digestion ( $\text{H}_2\text{O}_2 + \text{HNO}_3$ ) in a microwave oven and measurement by inductively coupled plasma atomic emission spectrometry (ICP-AES) in the Central Laboratory of the Finnish Forest Research Institute. Exchangeable cations (Ca, Mg, K, Al, Fe, Mn) in the mineral soil samples were determined by extraction with a 1 M  $\text{NH}_4\text{OAc}$  at pH 4.65 and measured by ICP-AES. Soil pH was measured in a 0.01 M  $\text{CaCl}_2$  suspension using a soil to solution ratio of 1:2.5 (v/v). Organic matter content of subsamples was determined by dry ashing for 5 h at 550 °C in a muffle furnace.

Cation exchange capacity (CEC) in the mineral soil was calculated as the sum of the concentrations of base cations and the exchangeable acidity (EA):  $\text{CEC (mmol/kg)} = \text{Ca}^{2+} + \text{K}^+ + \text{Mg}^{2+} + \text{Na}^+ + \text{EA}$ . Exchangeable acidity is the  $\text{H}^+$  concentration in the filtrate from  $\text{NH}_4\text{OAc}$  extraction and normally it is determined by titrating to pH 7 with 0.05 M NaOH. In this case, the titration was not carried and the EA values are estimates based on unpublished material collected during the Finnish National Forest Inventory (NF18). The proportion of Na in CEC (1%) was also based on these earlier data.

## 2.4. Determination of $^{210}\text{Po}$ and $^{210}\text{Pb}$

The determination of  $^{210}\text{Po}$  was based on separation by spontaneous deposition onto a silver disk (Flynn, 1968). After about six months' ingrowth time,  $^{210}\text{Pb}$  was similarly determined from the solution left from the  $^{210}\text{Po}$  deposition.  $^{210}\text{Po}$  is a granddaughter of  $^{210}\text{Pb}$  and in equilibrium their activities are identical. Thus, the  $^{210}\text{Pb}$  activity can be determined from the ingrown  $^{210}\text{Po}$  activity after removal of the initial  $^{210}\text{Po}$ .

For  $^{210}\text{Po}$  separation, 2–4 g soil samples were spiked with  $^{209}\text{Po}$  as a yield determinant tracer and digested first with concentrated  $\text{HNO}_3$ , then with a mixture concentrated  $\text{HNO}_3$  and  $\text{HCl}$  (1:3) and finally with concentrated  $\text{HCl}$ . Organic matter was decomposed with  $\text{H}_2\text{O}_2$ . Before deposition, hydrazine monohydrochloride and ascorbic acid were added to the solution (Vesterbacka and Ikäheimonen, 2005). Polonium was left to spontaneously deposit on a silver disc from 0.5 M  $\text{HCl}$  solution at 70–80 °C. The activity was then measured by alpha spectrometry (Alpha Analyst Genie2000, Canberra). The polonium yield was typically 60–80%.

The solution from the first deposition was stored for approximately six months to allow the ingrowth of  $^{210}\text{Po}$  from  $^{210}\text{Pb}$ , after which the second deposition was performed and  $^{210}\text{Po}$  activity measured. Some  $^{210}\text{Po}$  remains in the solution after the first precipitation, thus a  $^{208}\text{Po}$  tracer was used to determine the yield of the second deposition. All results are decay-corrected to the sampling date (for full calculation details see Vaaramaa et al., 2009).

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