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Aerosol particle size distribution of atmospheric lead-210 in northern Finland

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ABSTRACT

Size-segregated aerosol samples were collected with a high-volume 6-stage cascade impactor at Sodankylä, Finland, 100 km north of the Arctic Circle. The ^{210}Pb content of the samples were determined with radiochemical separation of in-grown ^{210}Po followed by alpha spectrometry. Most of the ^{210}Pb activity was incorporated in accumulation mode aerosol particles. The activity median aerodynamic diameter (AMAD) ranged from 0.53 μm to 0.98 μm 38–61 per cent of ^{210}Pb activity was found to be associated with aerosol particles smaller than 0.69 μm. A slight downward tendency of ^{210}Pb activity median aerodynamic diameter was observed as a function of increasing ^{210}Pb activity concentration. This is related to the continental origin of airborne ^{210}Pb on one hand, and various aerosol particle growth processes on the other hand. Also a clear tendency towards a higher ^{7}Be/^{210}Pb activity ratio as a function of increasing aerosol particle diameter was observed. This, in turn, reflects the different origin of ^{210}Pb, exhalation of ^{222}Rn from the soil into the air, and ^{7}Be, formation by cosmic radiation in the upper troposphere and the stratosphere.

1. Introduction

Lead-210 (^{210}Pb) is formed in the atmosphere from the radioactive noble gas ^{222}Rn emanating from the Earth's crust. 99% of the airborne ^{222}Rn originates from land and only 1% from the sea (Baskaran et al., 1993) and consequently high ^{210}Pb concentrations are found in continental air masses. Owing to the relatively long half-life (22.3 y) of ^{210}Pb, its removal from the atmosphere is governed by the wet and dry deposition processes affecting the aerosol particles carrying it rather than radioactive decay. Lead-210 decays to polonium-210 (^{210}Po, t½ = 138.4 d) via the short-lived beta emitter bismuth-210 (^{210}Bi, t½ = 5.013 d). Being an alpha emitter ^{210}Po is highly radiotoxic, especially when inhaled. It is also enriched in food-chains. Polonium and sulphur belong to the VIA group in the periodic table of the elements. Due to this chemical similarity polonium can replace sulphur in metabolic pathways (Brown et al., 2011; Persson and Holm, 2011). Based on the activity ratio of ^{210}Pb and its progeny (^{210}Bi and ^{210}Po), mean aerosol residence times of one to two weeks have been obtained (Mattsson 1975; Samuelsson et al., 1986; Papastefanou and Bondietti, 1991). No increases of ^{210}Pb concentrations in surface air due to anthropogenic sources have been found (Hötzl and Winkler, 1987). The reports of atmospheric ^{210}Pb concentrations have been reviewed by Preiss et al. (1996) and Baskaran (2011). The vertical distribution of ^{210}Pb in the atmosphere has been studied by e.g. Peirson et al. (1966) and Kownacka et al. (1990).

The size distribution of aerosol particles carrying ^{210}Pb is crucial for two reasons, atmospheric behavior and radiation protection. The efficiency of atmospheric removal processes, especially wet and dry deposition, depends on the size of aerosol particles carrying ^{210}Pb. In some cases the aerosol particle size also affects its atmospheric transport properties. Further, a variety of coagulation, condensation and evaporation processes in the atmosphere can change the size of aerosol particles where ^{210}Pb atoms are attached (Seinfeld and Pandis, 2016). From the radiation protection point of view the size distribution of aerosol particles carrying ^{210}Pb determines the ability of these particles to penetrate into the human respiratory system.

The objective of this work was to determine the aerodynamic size distribution of aerosol particles carrying ^{210}Pb in the subarctic

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atmosphere of northern Finland and to study the related meteorological effects on their concentration.

2. Experimental

Size-segregated aerosol samples were collected in open air 2 m above the ground at the Finnish Meteorological Institute's Arctic Research Centre at Sodankylä, Finland. The site is situated 100 km north of the Arctic Circle at 67.368°N, 26.633°E, and 179 m above mean sea level.

The aerodynamic size distribution of aerosol particles carrying ^{210}Pb was obtained by using a high-volume 6-stage cascade impactor with a regulated air flow rate of about 0.57 m^3 min^{-1} (20 cfm) and Efficient Cutoff Diameters (ECDs) of 0.39, 0.69, 1.3, 2.1, 4.2 and 10.2 μm. Two collection periods were chosen, the first one from 26-Mar-2010 to 10-Apr-2010 and the second one from 27-Jul to 9-Sept-2010. The length of each collection period was 48 h. Glass fiber filters were used as impaction substrates (slotted sheets 14.3 cm × 13.7 cm) for collection of the six coarser size fractions and regular back-up filters 20 cm × 25 cm for the finest size fraction. The bouncing of particles from the impaction surface was assumed to be negligible and therefore no additional coating (oil or grease) was used. After the collection procedure, the filters were folded and compressed to a form of a cylinder. The filters were measured for ^{7}Be activity using HPGe spectrometry at the Aristotle University of Thessaloniki, Greece (Ioannidou and Paatero, 2014).

After the gamma spectrometric analyses the samples were shipped to the Laboratory of Radiochemistry, University of Helsinki. The samples were analysed for ^{210}Pb with the classical method of spontaneous deposition of in-grown ^{210}Po onto silver disks and subsequent alpha spectrometry (Flynn, 1968).

Analysis of backward trajectories was used to confirm the arrival of radionuclides following atmospheric transport and processing and to interpret the activity variations of measured radionuclides. Five days long air mass back-trajectories were calculated with the Flextra model (Stohl et al., 1995), using the meteorological data from the European Centre for Medium-Range Weather Forecasts at Reading, UK (www.ecmwf.int). Eight back trajectories per day were calculated with arrival times of 00, 03, 06, 09, 12, 15, 18, and 21 UTC and an arrival level of 925 hPa.

3. Results and discussion

3.1. Average size distribution of ^{210}Pb-bearing aerosol particles

The activity concentration of ^{210}Pb in different particle size bins shows that most of the ^{210}Pb is attached to accumulation mode aerosol particles with an aerodynamic diameter below 1 μm (Fig. 1 and Table 1). In summer ^{210}Pb tends to be in 25 per cent smaller (by diameter) particles than in spring. This may be related to the residence time of aerosol particles carrying ^{210}Pb. In spring aged aerosols ("Arctic haze") are found in the atmosphere after the winter. In winter the residence time of aerosol particles is longer than in summer due to winter-time small amount of precipitation, reduced rate of chemical reactions in the atmosphere, and stagnant mixing conditions in the troposphere during the Arctic night. In summer, in turn, the aerosols are relatively fresh and have not had as much time to grow to bigger size classes as in the winter-time.

In an earlier work it was observed that most of the atmospheric deposition of ^{210}Pb in Finland occurs via wet removal processes (Paatero et al., 2015). This is confirmed by the results of this study as most of the ^{210}Pb activity is carried by accumulation mode aerosol particles. These particles are too small to be removed by gravitation or inertial impaction but too big to be removed by diffusion. Therefore they are removed mainly by wet deposition (Hinds, 1999).

3.2. ^{7}Be/^{210}Pb activity ratio in the aerosol particles

Activity ratios between ^{7}Be and ^{210}Pb in the aerosol samples were calculated using the ^{7}Be data from the same samples and reported by Ioannidou and Paatero (2014), see Table 2 and Fig. 2. There is a clear tendency towards a higher ^{7}Be/^{210}Pb activity ratio as the aerosol particle diameter increases. Beryllium-7 is formed in the upper troposphere and the lower stratosphere with very few coarse mode aerosol particles. However, the transport of aerosol particles carrying ^{7}Be from the higher altitude to the ground-level air takes time during which these aerosol particles can grow via various atmospheric condensation and coagulation processes. Lead-210, on the other hand, is attached to relatively new particles that have not had time to grow to bigger size classes. The slightly lower ^{7}Be/^{210}Pb activity ratio in the particles larger than 2.1 μm may be due to resuspension of soil particles.

3.3. ^{210}Pb AMAD and activity concentration

Cascade impactors separate aerosol particles into aerodynamic size fractions which are collected individually in different air filters. The filters are counted individually for activity in order to determine activity size distributions and hence activity median aerodynamic diameters (AMAD). Lead-210 AMAD values were calculated from the observed particle size distributions by calculating the percentage distribution in each size fraction and plotting the cumulative frequency distribution on log probability mode.

A slight descending tendency of AMAD can be seen as a function of increasing ^{210}Pb activity concentration (Fig. 3). Often low ^{210}Pb activity concentrations are observed in connection with maritime air masses (Paatero and Hatakka, 2000). These air masses are usually humid and therefore the aerosol particles have grown to a larger size as water vapour has condensed onto them. High ^{210}Pb activity concentrations, on the other hand, are found in dry continental air masses. The absence of moisture in these cases has hindered the growth of the aerosol particles and thus also the AMAD values remain relatively low. The average value of AMAD in our study was 0.75 μm with a range of 0.53–0.98 μm, in agreement with earlier results. Sanak et al. (1981) found values ranging from <0.6 μm to 0.77 μm over the Indian Ocean. In Japan 71–83 per cent of the ^{210}Pb activity was found to be attached to aerosol particles with an aerodynamic diameter below 0.7 μm (Suzuki et al., 1999). Gründel and Porstendörfer (2004) reported that in Germany 96 per

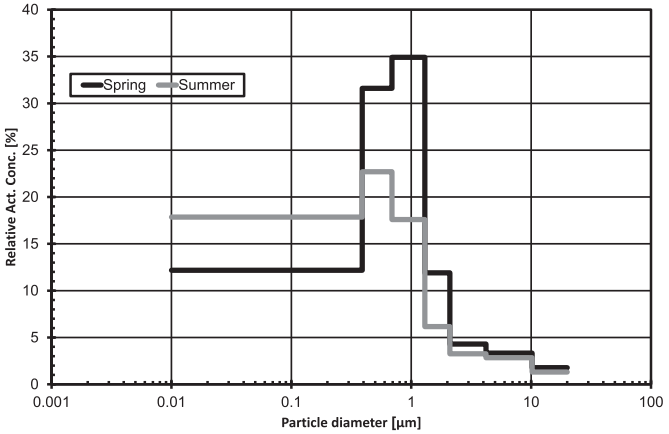

Fig. 1. The average relative activity concentration of ^{210}Pb in different particle size classes in spring and summer. The lower ends of the lines are arbitrary.