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Weekly variation of the ^{210}Pb air concentration in North Estonia

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We performed a gammaspectrometric analysis to determine the content of ^{210}Pb in aerosol filter samples collected weekly in Harku-Tallinn, North Estonia, in 2001–2005. Activity concentrations varied from 65 μBq m^{-3} to 2020 μBq m^{-3} with an arithmetic mean value of 366 μBq m^{-3} and followed a log-normal distribution with a geometric mean value of 308 μBq m^{-3} and dispersion factor of 0.52. The mean values were slightly higher than those expected at a location of comparable latitude and longitude. The analysis of monthly averaged concentrations revealed a dominant seasonal variation governed mainly by the origin of intruding air masses: high ^{210}Pb air concentrations in winter and low values in spring and summer. A multiple linear regression analysis confirms that significant correlations between the ^{210}Pb concentrations in the surface air and local meteorological parameters are observed only in winter.

Introduction

Minor concentrations of the natural radionuclide ^{210}Pb (half life $t_{1/2}$ = 22.3 years) formed from the radioactive inert gas parent ^{222}Rn ($t_{1/2}$ = 3.8 days) are always found in the atmosphere. The exhalation of radon, the ^{226}Ra ($t_{1/2}$= 1600 years) alpha-decay product found mainly in the soil, is a relatively constant source of ^{210}Pb, while the dry and wet deposition of lead-loaded aerosols from the atmosphere forms a sink.

The subgroup ^{210}Pb-^{210}Bi-^{210}Po is responsible for a significant contribution to the internal exposure to population. As a favourable tracer, long-lived ^{210}Pb is widely used in environmental research — e.g. in the dating of lake sediments and ice cores, for the determination of sediment accumulation rates, in atmospheric aerosol and pollutant research, in the modelling of atmospheric transport, and removal processes. As the continental and marine radon source rates differ by about two orders of magnitude, global as well as regional meteorological conditions strongly influence the ^{210}Pb surface air concentration. In marine-to-continental, polar-to-temperate and temperate-to-equatorial interface regions, these influences are particularly significant.

Multiple studies all over the world have been conducted to determine the ^{210}Pb concentration and its variations in the surface air. Presently, data about different locations worldwide are accumulated in the ^{210}Pb database (Preiss *et al*. 1996).

Until now, relatively short-period data on the ^{210}Pb concentration in the surface air in Estonia have been available (Realo *et al.* 2004). The present study reports the results drawn from our data collected during 4.5 years in Harku-Tallinn, Estonia, located near the northern boundary (60°N) of the temperate latitudinal band and

in a transition area from maritime to continental climate, is characterised by a high temporal and spatial variability of weather. Continental air masses from Eurasia are about as frequent as the northern Atlantic maritime air intrusion. The arctic air invasions occur mainly in winter and spring. The climate is relatively humid, as precipitation prevails over evapotranspiration. On average, snow cover persists about 100 days with significant geographical and temporal variations.

Material and methods

For said ^{210}Pb analysis, we used archived aerosol filter samples collected weekly over the period 2001–2005 by the Estonian Radiation Protection Centre (ERPC) in the framework of the national radiation surveillance program. The sampling site at the Harku-Tallinn meteorological station (59°23´54´´N, 24°36´15´´E, 33 m above sea level) is located in North Estonia about eight km east of the centre of Tallinn and five km south of the Gulf of Finland. The long-term average meteorological parameters of the site were the following: relative humidity = 81%, air temperature = 5.1 °C, precipitation = 668 mm and wind speed = 4.4 m s^{-1} with prevailing southwestern winds.

During a week-long sampling period, the mean air volume of 180 000 m^3 passed through a Petrianov FPP filter of a high-volume air sampler. By means of a hydraulic press and specific anvils, the filters were compressed to form a cylinder with the diameter of 41 mm, which were placed in congruous diameter beakers. We analysed the ^{210}Pb content by means of its 46.5 keV line on the HPGe planar detector gamma spectrometer (MULTISPECTRUM, BSI, Latvia). As the sample heights varied from sample to sample, we determined the attenuation coefficient by means of separate measurements with a ^{210}Pb source and measured the height of each compressed sample. Based on the measurement data of the IAEA RGU reference samples of various heights and Monte Carlo modelling (GESPECOR, GEANT4), we adopted a simple procedure for the correction of peak efficiency on the attenuation coefficient and sample height. We tested the quality of our results, including the correction procedure, by participation in the Nordic and IAEA intercomparison exercises.

The daily meteorological data collected at the local automatic station: precipitation (rain, snow), relative humidity, air pressure, temperature and wind speed, were provided by the Estonian Meteorological and Hydrological Institute. We calculated the corresponding weekly and monthly averages needed for the statistical analysis of correlations of ^{210}Pb concentrations on meteorological parameters. For multiple linear regression analysis of the determined concentrations and meteorological parameters we applied the Essential Regression add-in spreadsheet (available at http://www.geocities.com/SiliconValley/Network/1900/) for MS Excel.

Results and discussion

The ^{210}Pb activity concentration in the air demonstrates a considerable temporal variation. The weekly data (Fig. 1) varied in the range of 65 μBq m^{-3} to 2020 μBq m^{-3} with the arithmetic mean of 366 μBq m^{-3}. The concentrations followed approximately the log-normal distribution with a geometric mean of 308 μBq m^{-3} and dispersion factor of 0.52 (Fig. 2). Hötzl and Winkler (1987) and Paatero *et al*. (1998) found that the log-normal distribution is a good approximation also on the daily time scale. Mean concentration values in Harku are higher than those found in Helsinki and Nurmijärvi (Paatero *et al*. 1998) as well as those typical of the comparable latitude and longitude region in the ^{210}Pb database (Preiss *et al*. 1996). Data analysis on two other Estonian locations, Narva-Jõesuu and Tõravere, is in progress, but preliminary mean values of these locations are even higher than in Harku (Realo *et al*. 2004). The reason why the ^{210}Pb air concentrations found here were significantly higher as compared with those recorded in South Finland is still unclear. The most straightforward explanations assuming a stronger influence of continental air masses in Harku than in Helsinki or the varying role of the Baltic Sea need further analyses.

The highest ^{210}Pb concentration values occur in winter during periods characterised by long-term high atmospheric pressure and low temperature, i.e. during the governing of anticyclonic

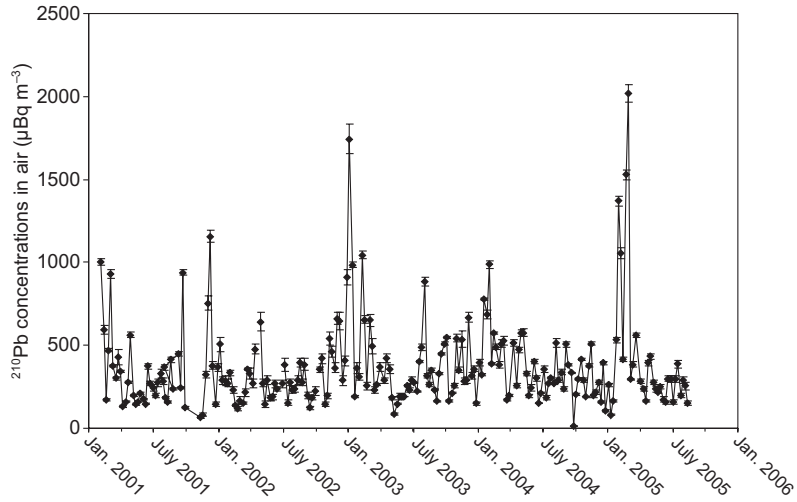

Fig. 1. Weekly variation of ^{210}Pb activity concentrations in the air, Harku-Tallinn, Feb. 2001–Aug. 2005.

conditions. The most pronounced seasonal trend is that winter months are characterised by the highest and the spring months by the lowest concentration values of the year (Figs. 3 and 4). In Finland, Paatero *et al*. (1998) showed that high winter concentrations of ^{210}Pb correlate well with low mixing layer heights, which were typical of the mostly stable tropospheric conditions with short daytime and low-level solar radiation. In spring and summer the concentrations are low because of longer days and strong solar radiation. Many authors (*see* e.g. Preiss *et al*. 1996) consider that low spring and summer concentrations are the result of an efficient vertical mixing generated by intense solar radiation in the troposphere and by large mixing layer heights. This vertical mixing carries ^{210}Pb from the surface layer up to the upper tropospheric layers and brings forth a decrease in the former. The seasonal behaviour of ^{210}Pb concentration in the air in Estonia is very similar to the one registered in Finland by Paatero *et al*. (1998), in Germany by Winkler and Rosner (2000), and in Italy by Vecchi *et al*. (2005). Different seasonal behaviour was observed in some locations in southern Europe: Todorovic *et al*. (2005) and Dueñas *et al*. (2004, 2005) recorded the concentration maxima in summer, while Ioannidou *et al*. (2005) observed these in autumn.

We made an attempt to search for correlations between the measured ^{210}Pb air concentrations and the local meteorological parameters: air temperature, atmospheric pressure, relative humidity, precipitation amount and wind speed. The origin of intruding air masses (continental, marine, arctic) and their variability are the major factors which govern the local meteorological parameters. In the analysis we used the averaged parameter values for weekly (sampling) and monthly periods. The sampling time interval (one week) is long in comparison with the time constants, which are characteristic of the typical time scales of variation in meteorological parameters. For this reason, actual correlations may be partly suppressed or smoothed. In addition, many meteorological parameters are correlated with each other, thus presenting the problem of multicollinearity in their statistical analysis. To

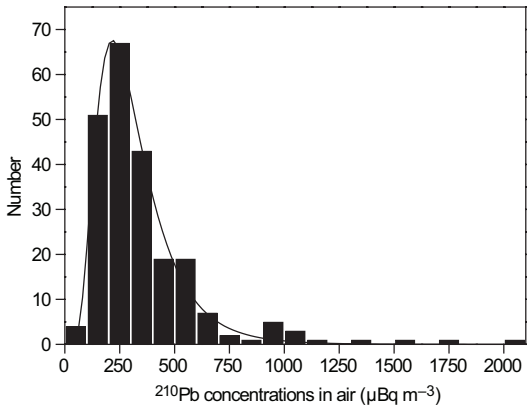

Fig. 2. Log-normal distribution of ^{210}Pb activity concentrations in the air. Solid line is the least-square fit to the histogram with the geometric mean of 308 μBq m^{-3} and the dispersion factor of 0.52.

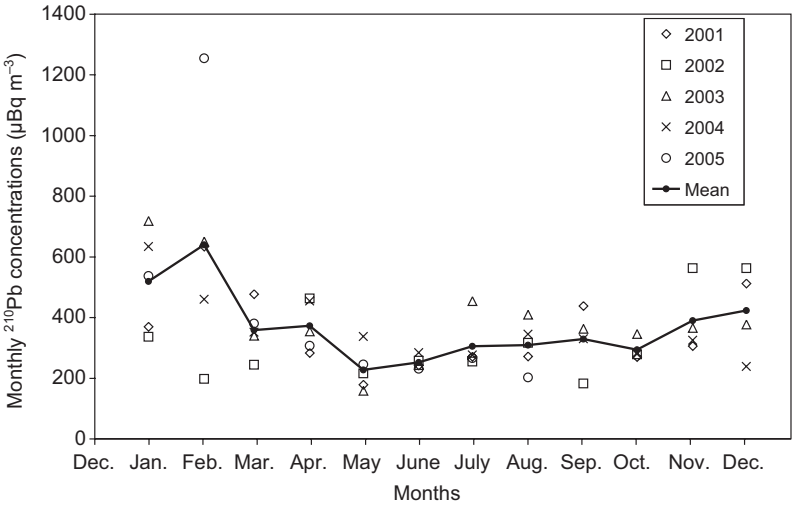

Fig. 3. Monthly averages of ^{210}Pb activity concentrations in air, Harku-Tallinn, Feb. 2001–Aug. 2005. Solid line connects the monthly means over the period.

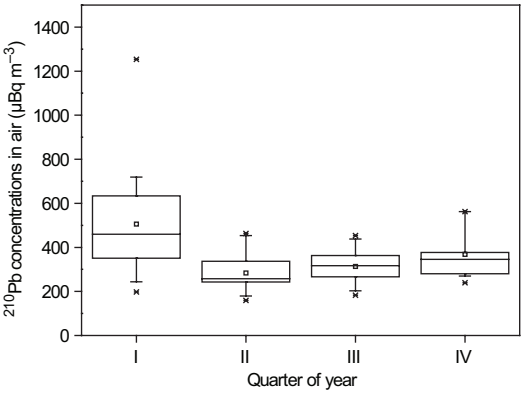

Fig. 4. Seasonal ^{210}Pb air concentrations, $C_{^{210}Pb}$ in 2001–2005 in Harku-Tallinn.

address these issues in the weekly and monthly data, we used the multiple linear regression analysis. The regression analysis of weekly data revealed a significant correlation (R^2 = 0.21, p < 0.01) of the ^{210}Pb air concentration with the air temperature and atmospheric pressure, and a slightly weaker negative correlation (p < 0.05) with the wind speed (Table 1).

As for the winter months (Dec.–Mar.), the multiple correlation with the air temperature and with atmospheric pressure was even better (R^2 = 0.42). In winter, a negative correlation (p < 0.08) between the concentration and the amount of precipitation indicates that the wet deposition by snow might have been effective in removing ^{210}Pb from the air. As for the remaining part of the year (Apr.–Nov.), no meteorological parameter was significant at p < 0.01 (R^2 = 0.04). Rainfall seemed to have no observable effect on ^{210}Pb concentrations in the air.

Monthly-averaged ^{210}Pb activity concentrations varied from year to year as seen in Fig. 3. Monthly concentrations correlated significantly (p < 0.001) with the air temperature: lower air temperatures facilitated higher ^{210}Pb activity concentrations in the atmosphere. Another significant parameter was the average wind speed. In Estonia, strong winds are usually associated with cyclonic activity carrying maritime air masses with a low concentration of ^{210}Pb from the North Atlantic. At the same time there was no significant correlation between the monthly ^{210}Pb concentrations and the North Atlantic Oscillation (NAO) index, the ground-level difference in barometric pressure between Iceland and Portugal (available at http://www.cru.uea.ac.uk/~timo/projpages/nao_update.htm).

It follows from the regression analysis that air temperature and wind speed were the most important regressors to the ^{210}Pb air concentration on weekly as well as on monthly time scale. Their negative regression coefficients are relatively similar on either time scale. Atmospheric pressure was a significant regressor only for measured weekly concentrations in winter (Table 1).

Conclusions

We analysed the weekly concentrations of ^{210}Pb in the surface air of Harku, North Estonia, in

Table 1. Multiple regression model of ^{210}Pb activity concentrations, C_{210Pb} (μBq m^{-3}), at Harku-Tallinn, Estonia in 2001–2005. T = air temperature (K), P = atmospheric pressure (hPa), pr = average daily amount of precipitation (mm), v = wind speed (m s^{-1}), R^2 = coefficients of multiple determination, p = significance levels of multiple linear regression analysis. W = weekly values, M = monthly averages.

Period	Multiple regression model for C_{210Pb}	R^2	p
W, all	$-(10.2 \pm 1.7)T + (4.7 \pm 1.6)P - (41 \pm 20)v - 1440$	0.21	< 0.001 (T, P), < 0.05 (v)
W, Dec.–Mar.	$-(29.0 \pm 4.8)T + (7.3 \pm 1.9)P - (29 \pm 16)$pr + 870	0.42	< 0.01 (T, P), < 0.1 (pr)
M, all	$-(12.7 \pm 2.3)T - (85 \pm 38)v + 4187$	0.40	< 0.001 (T), < 0.05 (v)
M, Dec.–Mar.	$-(35 \pm 7)T + 10044$	0.56	< 0.001 (T)

2001–2005. Weekly concentration values characterized by log-normal distribution suggested a considerable variability. The mean values were higher than the data found at sites of similar geographical longitude and latitude by other authors. Additional analyses are needed to explicate this divergence in the data. The origin of intruding air masses, e.g., continental, marine or arctic, as well as their variability, were the major factors governing the ^{210}Pb concentrations in the surface air. The analysis of monthly-averaged concentrations revealed a dominant seasonal variation trend with high ^{210}Pb air concentrations in winter and low values in spring and summer. We carried out a multiple linear regression analysis to determine the correlation between the ^{210}Pb air concentrations and the local meteorological parameters. Significant correlations occurred only in the winter data. Activity concentrations of ^{210}Pb showed negative correlation with the air temperature and wind speed for both weekly- and monthly-averaged samples. A positive correlation of activity concentrations with atmospheric pressure was witnessed only in the weekly samples. In winter, a negative correlation between the air concentration and the amount of precipitation might confirm the efficiency of wet deposition in removing ^{210}Pb from the air.

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