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Wet deposition efficiency of short-lived radon-222 progeny in central Finland

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The washout efficiency of ^{214}Pb, a short-lived daughter nuclide of ^{222}Rn, was studied by measuring external gamma radiation at Tikkakoski, Central Finland, in 1995. A mean washout ratio (volume per volume) of 1.2×10^6 was obtained in this study. According to the results, rain removes airborne ^{214}Pb more efficiently than snow, which is in agreement with earlier studies. Higher washout ratio values were encountered during afternoon hours than during early morning hours, a result which is associated with the better vertical mixing of the lower troposphere during afternoon hours. The observed inverse correlation between washout ratio and rain intensity supports the earlier results that below-cloud scavenging processes are less efficient removal mechanisms of airborne ^{214}Pb compared to in-cloud processes. The highest washout ratios were measured during westerly winds, which are usually associated with cyclones travelling from the North Atlantic Ocean in an easterly direction. The results obtained in this study can also be applied to fallout estimates of wet-depositing chemical substances, e.g. airborne sulphate or heavy metals, for which equivalent remote-sensing methods, such as that involving gamma radiation in this case, may not be available.

Introduction

In Finland the external dose rate in the surface air varies usually between 0.04 and 0.30 µSv/h (Ristonmaa 1998). Short-term variations in external radiation are mainly caused by precipitation, which scavenges airborne radionuclides to the ground. During the period of the present study, the year 1995, this wet-deposited activity was almost entirely of natural origin, i.e. consisting of the short-lived daughter nuclides of radon-222.

The efficiency with which precipitation scavenges airborne radioactivity to the ground can be described by the washout ratio w_r. This is the ratio of activity concentration in surface-level precipitation to that in surface-level air (Seinfeld and Pandis 1998). These washout ratios take into account all the in-cloud and below-cloud processes determining the destiny of the radionuclide suspended in the air. Because the short-lived ^{222}Rn progeny is mostly attached to aerosol particles, the in-cloud processes involved are nucleation scavenging (growth of a cloud condensation nucleus including the radionuclide into a cloud drop-

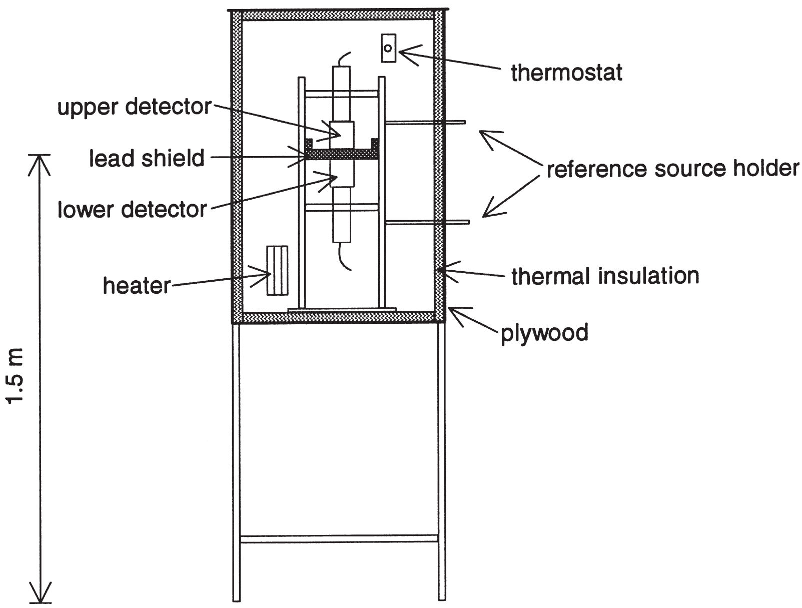

Fig. 1. Gamma detector arrangement. The upper NaI(Tl) scintillation detector is more sensitive to suspended activity and the lower detector to deposited activity.

let) and interstitial aerosol collection by cloud or rain droplets. The other process, below-cloud scavenging, results from the collision of falling hydrometeors with aerosol particles. Two definitions for the washout ratio, based on concentrations per unit volumes and concentrations per unit masses, are used in the literature. The values differ by a factor of about 10^3 (Engelmann 1971). In this paper we use the washout ratios based on the concentrations per unit volumes. As a rule of thumb, for particle-bound lead-210 and sulphate, the washout ratios are of the order of 10^6, in other words one millilitre of rain water contains as much substance as one cubic metre of air (Paatero and Hatakka 1997).

Following the Chernobyl accident in 1986, the Finnish Meteorological Institute (FMI) equipped some of its radioactivity monitoring stations with instruments for measuring external radiation (Paatero *et al*. 1994). In this paper we report, from one station in central Finland, the activity concentration of short-lived radon-222 progeny in rain water based on the increase of external radiation during rainfall. The dependence of the corresponding washout ratios on various meteorological parameters is discussed. The year 1995 was selected for the study because the annual amount of precipitation, 599 mm, was closest to the average amount, 614 mm, for the years 1992–1996 with available data.

Experiment

The Tikkakoski weather sounding station (62°24´N, 25°40´E) is located beside the airport at Jyväskylä. The elevation of the station is 140 m above sea level. The terrain near the station is practically flat and consists of gravel and sand. Surface weather observations are made at the aviation weather station 1 km north of the sounding station.

External gamma radiation is measured with a dual 76 mm × 76 mm NaI(Tl) scintillation detector system (Fig. 1) described in detail earlier (Paatero *et al*. 1994). The detector shelter is on the ground 15 metres from the sounding station building. The pulses exceeding the lower level discriminators (50 keV) are counted with a data logger at 10-minute intervals without pulse height analysis. The count rate varies between 40 000 counts per minute (cpm) in summer and 6 000 cpm in winter, due to the attenuation of gamma radiation by a snow cover. The stability of the gamma measurement system is monitored with a ^{137}Cs source, which is installed once a month with a fixed geometry near the detectors for half an hour.

Only the count rates of the lower detector were used in this study. In order to get a conversion factor between count rate and deposited activity, several assumptions were made. The count rate increase during precipitation was presumed to be due to the deposited short-lived ^{222}Rn daughter nuclides ^{214}Pb and ^{214}Bi only, thus neglecting the effect of e.g. ^{220}Rn progeny and artificial activity on the count rate increase. The fraction of pulses caused by ^{214}Pb (coefficient F below) was assumed to be 52% of all pulses based on the photon energies and gamma emission probabilities of ^{214}Pb and ^{214}Bi and the intrinsic detection efficiency of NaI(Tl) (Knoll 1989).

The calculation of total deposited activity was started by integrating the net counts from the start of the precipitation event to four hours after the end of the precipitation, at which time essentially all the short-lived ^{222}Rn progeny has decayed. The sum of the net counts (X_{TOT}) was converted to the initial count rate (R_0) by multiplying it with the fraction of pulses caused by ^{214}Pb (F) and then with the decay constant λ of ^{214}Pb, because

$$FX_{\mathrm{TOT}} = \int R_0 e^{-\lambda t}\mathrm{d}t \Rightarrow R_0 = \lambda F X_{\mathrm{TOT}} \qquad (1)$$

where t is time. The value used for F was 0.52. Two methods were used for background subtraction (Fig. 2). In the usual case of a decreasing background, the decrease was assumed to be proportional to the rain intensity due to the gamma radiation attenuation in the water layer collecting on the ground or snow surface. In the opposite case the background was assumed to increase linearly. The latter cases usually occurred in spring because of the rapid melting of the snow (Hatakka *et al.* 1998). The decrease of background count rate due to precipitation was more prominent in summer than in winter because of the greater relative contribution of cosmic radiation to the background count rate in winter. For example, during the heaviest precipitation in February, 8.4 mm, the background count rate decreased 0.2%. The corresponding decrease in August, with a precipitation amount of 27.8 mm, was 4%. Because of the need to get a background count value after the almost complete decay of the short-lived ^{222}Rn progeny, subsequent precipitation events were considered as one incident if the time between the two actual events was less than four hours. In some cases the behaviour of the background was too diffuse or the count rate increase too small for a net count calculation. The amount of precipitation due to such incidents represented 13% of the annual amount of precipitation, the number of these incidents being 13 out of a total number of 226. The average number of net counts per precipitation incident was 128 000.

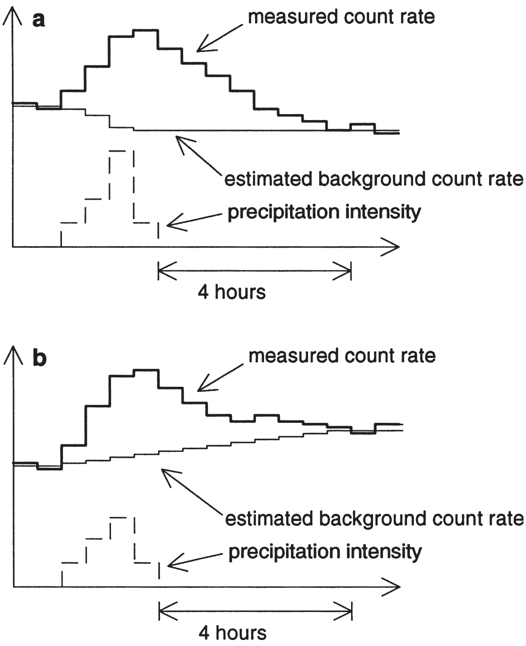

Fig. 2. Background subtraction methods; a = decreasing background, b = increasing background.

Next, the initial count rate so obtained was divided by the overall counting efficiency in order to get the amount of deposited ^{214}Pb activity. Overall counting efficiency depends on three factors: the counting geometry, the intrinsic efficiency of the detector and the gamma emission probability of the particular nuclide.

When assessing the photon flux through the detector, only the side and the bottom surfaces were taken into consideration, and the photon flux was assumed to be equal through both surfaces. The flux φ at a height d of 1.5 m above the ground caused by a uniform unit strength plane source S_A with a radius R of 100 m was calculated with the formula (Dörschel *et al.* 1996)

$$\varphi = \frac{S_{\mathrm{A}}}{2}\ln\left[\frac{\sqrt{(d^2 + R^2)}}{d}\right] \Leftrightarrow \frac{\varphi}{S_{\mathrm{A}}} = 2.1\mathrm{s}^{-1}\mathrm{m}^{-2}\mathrm{Bq}^{-1}\mathrm{m}^2 \quad (2)$$

This calculation was combined with data on gamma emission probability and the intrinsic

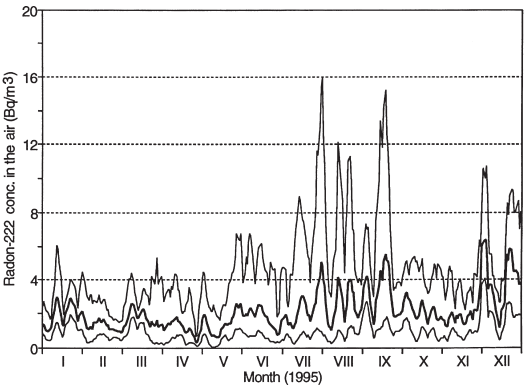

Fig. 3. Five-day moving averages of ^{222}Rn activity concentration in surface air (Bq m^{-3}), Tikkakoski 1995. The lowermost and uppermost thin curves represent daily minimum and maximum hourly median concentrations and the thick line daily average concentration calculated from the hourly medians.

efficiency of the sodium iodide crystal for ^{214}Pb (Knoll 1989):

$$A_{\mathrm{A}}\left[\mathrm{Bq\,m^{-2}}\right] = \frac{S_{\mathrm{A}}R_0}{\varphi A \varepsilon_i \eta} \quad (3)$$

where A_A is the amount of deposited ^{214}Pb, φS_A^{-1} the photon flux per unit strength plane source (2.1 s^{-1} m^{-2} Bq^{-1} m^2), A the sensitive area of the detector (0.0228 m^2), ε_i the intrinsic detection efficiency of NaI(Tl) (0.88) and η the gamma emission probability of ^{214}Pb (0.56). The overall counting efficiency so obtained was further reduced by 20% to counterbalance the absorption of photons in the ground, snow, air and detector shelter, and the reduced detection efficiency in the corners of the detector. The associated error sources have been discussed by e.g. Tyler *et al.* (1996) and Laedermann *et al.* (1998).

Finally the ^{214}Pb activity deposited during the precipitation event was divided by the amount of precipitation to get its activity concentration. The amount of precipitation in each precipitation event was estimated with rain sensor data and the daily precipitation observations of the aviation weather station. If the precipitation observation was 0 mm, a value of 0.1 mm was used instead.

The ^{222}Rn activity concentration in the air was measured with an instrumentation in which air is drawn alternately through one of two fixed filters wrapped around GM tubes (Paatero *et al.* 1994). The air was sampled 7 m above ground and two meters above the roof of the sounding station. The obtained 10-minute values of ^{222}Rn concentrations were processed to hourly medians. Radon-222 was assumed to be in equilibrium with its short-lived progeny.

The washout ratios were calculated by dividing the coincident $^{222}Rn/^{214}Pb$ concentration values in the air and in the precipitation. One to three washout ratio values per precipitation incident were calculated depending on the duration of the precipitation: for the starting hour, for the ending hour, and for the mean point hour of the rainfall event. The obtained data set consists of 213 values for the ^{214}Pb concentration in precipitation and 557 values for the washout ratio.

Results and discussion

The measured amount of deposited ^{214}Pb during the year 1995 was 660 kBq m^{-2}. Taking into consideration the omitted precipitation events, the total deposition was ca. 700 kBq m^{-2}. This amount of ^{214}Pb will produce 1.6 Bq m^{-2} of ^{210}Pb in the ground. This value can be compared to the annual total ^{210}Pb deposition, which varied in 1995 between 80 Bq m^{-2} in Helsinki and 60 Bq m^{-2} in Ivalo, northern Finland (Leinonen 1997). The amount of ^{214}Pb deposition was correlated with the amount of precipitation, the Pearson correlation coefficient R being 0.69. The activity concentration of ^{214}Pb in precipitation varied between 0.02 and 42.7 kBq L^{-1}, the average being 2.87 kBq L^{-1}. In Japan Nishikawa *et al.* (1986) reported radon progeny activity concentrations in precipitation varying between 1 and 11 kBq L^{-1}. The measured radon concentrations in the surface air are lowest in spring when the mixing of the boundary layer is efficient and the exhalation of ^{222}Rn is reduced due to the snow cover and frozen ground (Fig. 3). In late summer and autumn the diurnal variation of concentrations is high due to the simultaneous strong radon exhalation and frequent nocturnal surface inversions. In winter the concentrations are relatively high because of the low mixing height caused by the short daylight duration. From these measurements, an arithmetic mean washout ratio of 1.2×10^6 was obtained.

The ^{214}Pb content of precipitation does not dis-

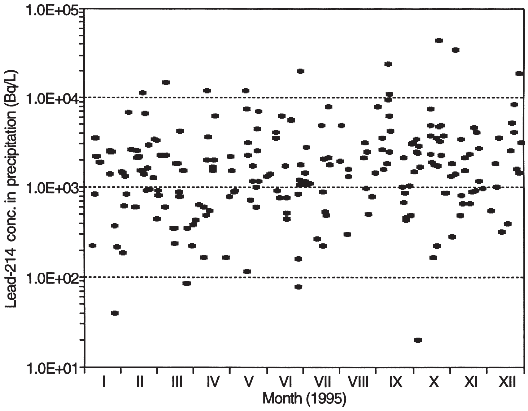

Fig. 4. ^{214}Pb activity concentration of precipitation (Bq L^{-1}), Tikkakoski 1995 (213 cases).

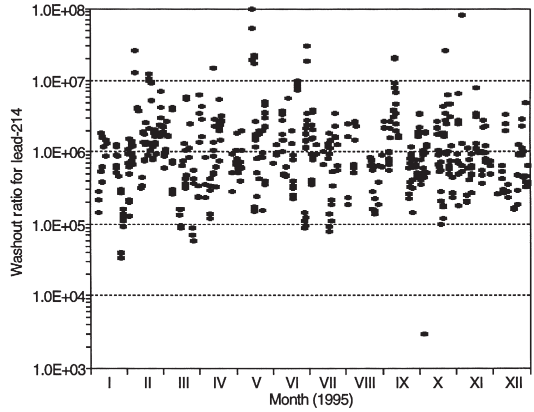

Fig. 5. Washout ratio for ^{214}Pb, Tikkakoski 1995 (557 cases).

play any clear seasonal variation, nor does the washout ratio (Figs. 4 and 5). This is in agreement with results obtained in Helsinki in the case of long-lived beta activity (Paatero and Hatakka 1997). Contradictionary results, with high radon progeny concentrations in precipitation in winter and low concentrations in summer, were reported by Hayakawa (1985). The washout ratios show high variability from one incidence of precipitation to another. However, the washout ratios are higher in the case of rain than in that of snow, which can be seen from Fig. 6 showing the washout ratio as a function of surface air temperature. Similar results for airborne sulphate have been obtained by Nordlund and Tuomenvirta (1998). They reported washout ratios to be lowest below –5 °C, increasing to a maximum at +7.5 °C and then decreasing slowly. Scott (1978) explained the deviation as being due to the differences in precipitation formation mechanisms. Barrie (1985) has also reported lower sulphate washout ratios in the case of snow than in the case of rain. He, however, attributed the difference to variations in the oxidation rate of SO_2. There is no correlation between relative humidity of the surface air and ^{214}Pb concentration in precipitation or washout ratio although one might expect the ^{214}Pb concentration in precipitation to increase due to the evaporation of water in case of low relative humidity.

The washout ratios obtained in this study also exhibit a diurnal variation (Fig. 7). The highest values are found during the afternoon hours, when strong vertical mixing of the lower troposphere

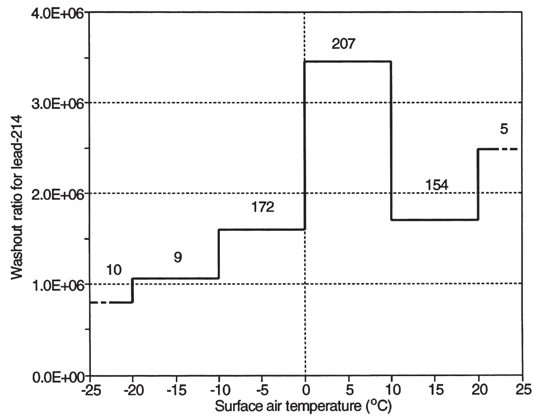

Fig. 6. Washout ratio for ^{214}Pb as a function of surface air temperature, Tikkakoski 1995. The figures indicate the number of washout ratio values within the corresponding temperature range.

can transport ^{222}Rn and its daughter nuclides upwards. Most of the radon progeny are attached to sub-micron aerosol particles which follow the movements of the air parcel and are not sensitive to gravitational settling (Porstendörfer and Reineking 1999). The contribution of this local ^{222}Rn progeny to the total ^{222}Rn progeny concentration in the rain-forming layers of the troposphere varies depending on the source areas of the air masses. The contribution is highest in air masses originating from maritime or Arctic areas with practically no sources of ^{222}Rn. The washout ratios are low during the early morning hours when the lower troposphere is often stratified.

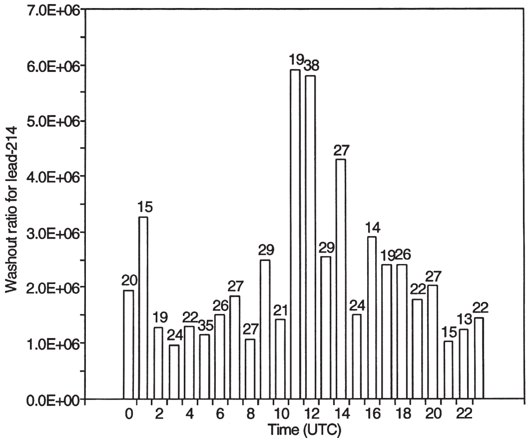

Fig. 7. Diurnal variation of washout ratio for ^{214}Pb, Tikkakoski 1995. The figures indicate the number of washout ratio values during the corresponding hour.

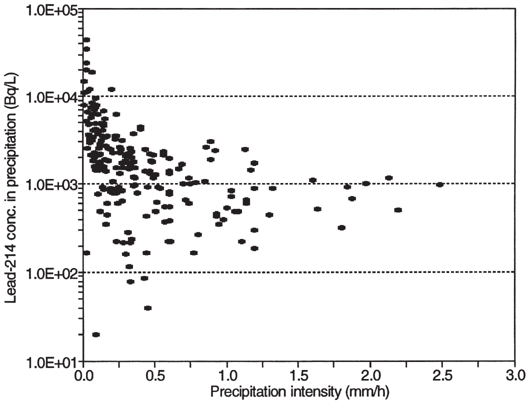

Fig. 8. Concentration of ^{214}Pb in precipitation (Bq L^{-1}) as a function of precipitation intensity, Tikkakoski 1995 (213 cases).

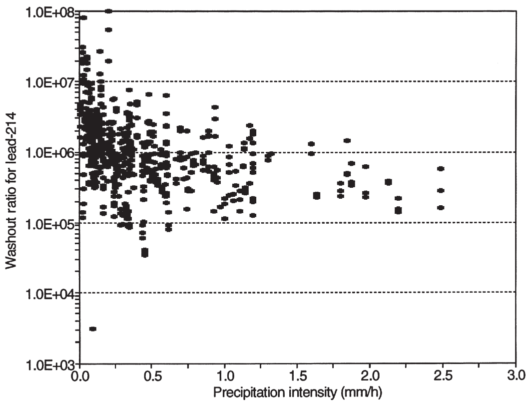

Fig. 9. Washout ratio for ^{214}Pb as a function of precipitation intensity, Tikkakoski 1995 (557 cases).

The relative inefficiency of below-cloud processes in radon progeny removal is related to the aerosol particle size distribution. As mentioned above, most of the radon progeny are attached to accumulation mode aerosol particles with a diameter of a few hundred nanometres. These particles are not subject to impaction by falling hydrometeors because they follow the aerodynamic streamlines around the hydrometeors. These particles are, however, prone to nucleation scavenging and other in-cloud processes (Warneck 1988). This is supported by the behaviour of the ^{214}Pb concentration in the precipitation and the corresponding washout ratio which are both exponentially dependent on the precipitation intensity (Figs. 8 and 9). Apparently even the smallest amounts of precipitation are capable of depositing most of the airborne ^{214}Pb and any "excess" precipitation merely dilutes the activity in a larger water volume. If below-cloud processes were important in this context the washout ratio should increase as a function of precipitation intensity due to the increasing number of collisions between falling hydrometeors and aerosol particles. Scott (1981) reported that for sulphate scavenging the in-cloud processes were 10–50 times more efficient than below-cloud removal. Our results are in agreement with the sulphate washout data reported by Scott (1978). Analogous results for airborne sulphate have been obtained by Lindberg (1982), Barrie (1985), and Nordlund and Tuomenvirta (1998), who discovered that the washout ratio was inversely dependent on the amount of precipitation. Decreasing radon progeny concentrations in precipitation as a function of increasing precipitation intensity have been observed also by e.g. Hayakawa (1985), Fujinami *et al.* (1993) and Fujinami (1996).

The concentration of ^{214}Pb and ^{222}Rn in the precipitation and in the surface air, and the corresponding washout ratio as a function of wind direction are presented in Fig. 10. The highest concentrations in the precipitation and the highest washout ratios are associated with westerly winds. In an earlier study, the highest washout ratios for long-lived beta activity (constituting mostly of

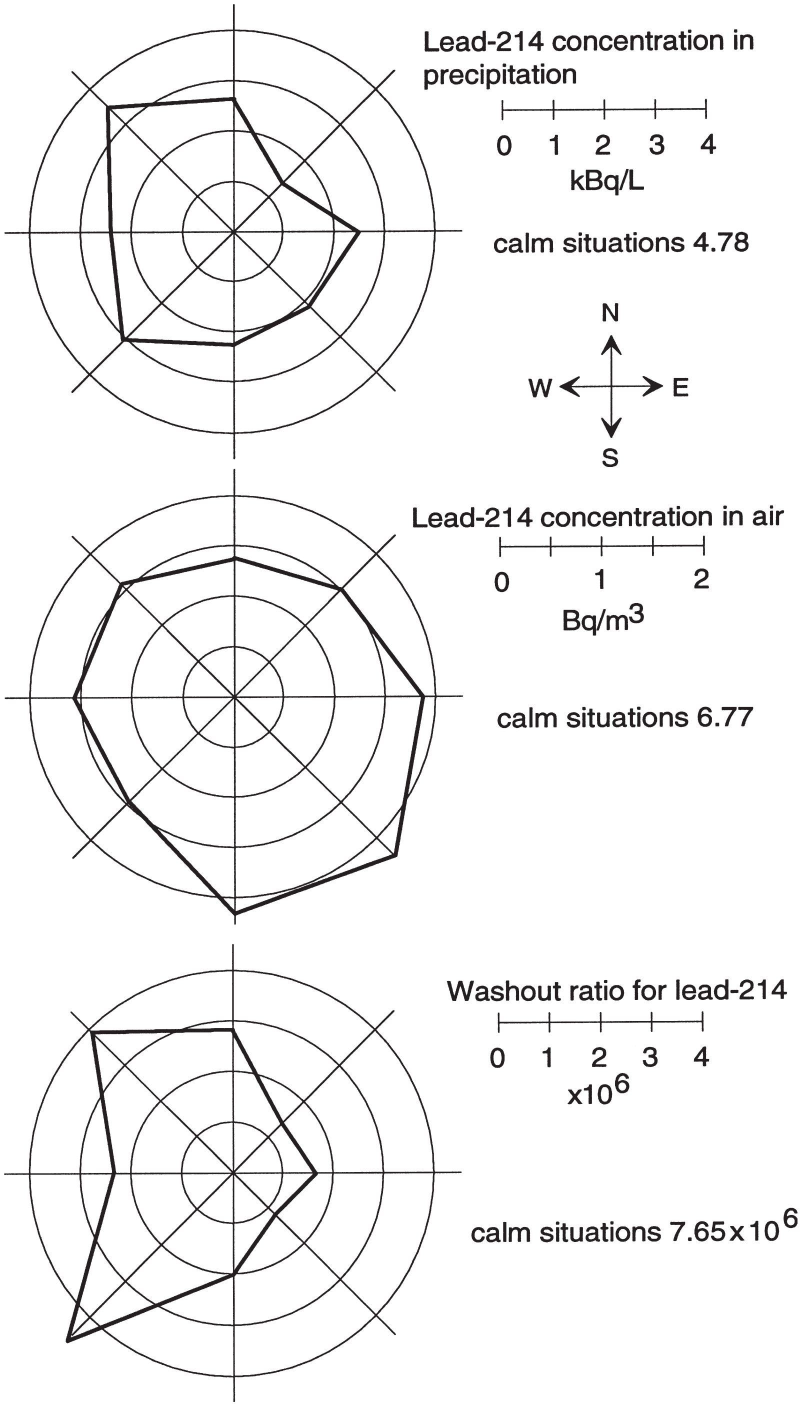

Fig. 10. Concentration of ^{214}Pb in precipitation (kBq L^{-1}) and concentration of ^{222}Rn in surface air (Bq m^{-3}), and the corresponding washout ratio as a function of wind direction, Tikkakoski 1995. The washout ratios in each wind direction classes are averages of the observed washout ratios, not the ratios between average concentrations in precipitation and in surface air within these wind direction classes.

^{210}Pb, the long-lived daughter nuclide of ^{222}Rn) were associated with westerly winds but the highest concentrations in precipitation with easterly winds (Paatero and Hatakka 1997). In Finland, winds from a westerly direction are often connected with low-pressure areas moving eastwards from the North Atlantic Ocean. The general upward movement of the air within the low-pressure areas promotes the transfer of ^{222}Rn progeny from the surface air to the rain-forming layers of the troposphere. On the other hand, during the relatively short period the air masses from the North Atlantic Ocean spent over the continent, the available time for the in-growth of ^{210}Pb is short compared to that of ^{214}Pb. This explains the difference in the relative concentrations of ^{214}Pb and long-lived beta activity in precipitation during westerly winds. During calm situations, locally-exhaled ^{222}Rn increases the ^{214}Pb concentrations both in the air and in precipitation.

Summary and conclusions

The scavenging efficiency of ^{214}Pb, a short-lived daughter nuclide of ^{222}Rn, was studied using recordings of external gamma radiation in Central Finland. A mean washout ratio of 1.2×10^6 was obtained in this study. The washout ratios were higher in the case of rain than in that of snow. Higher values were also encountered during the afternoon hours than during the early morning hours, a result which is associated with the vertical mixing of the lower troposphere. The observed rain intensity dependence of the washout ratios supports the earlier results that below-cloud scavenging processes are less efficient removal mechanisms of airborne ^{214}Pb compared to in-cloud processes. The highest washout ratios were measured during westerly winds, which are usually associated with cyclones travelling from the North Atlantic Ocean in an easterly direction.

As stated by e.g. Barrie (1992), the use of washout ratios involves several possible error sources. Still, the results obtained in this study should be applicable in long-term deposition estimates that smooth the variabilities connected with individual occurrences of precipitation. Additionally, their use may be justified in situations requiring fast fallout assessments. These results can also be applied to wet-depositing chemical substances, e.g. airborne sulphate or heavy metals, which can be difficult if not impossible to measure using real-time remote sensing such as the measurement of gamma radiation in our case.

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