THE APPLICATION OF ENVIRONMENTAL RADIOACTIVE TRACER INVENTORIES IN SOIL TO STUDY DEPOSITION OF POLLUTANTS

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ABSTRACT

The natural radionuclide ^{210}Pb and the anthropogenic one, ^{137}Cs, are present in particle form in the atmosphere attached to the same aerosols which contain the bulk of pollutant sulphur and nitrogen. When scavenged from the atmosphere by precipitation, they are readily filtered by the surface horizons of soil, and their inventories can be used to measure the spatial variation in aerosol and wet deposition within a region due to orography or land use averaged over decades (physical half-life of ^{210}Pb and ^{137}Cs are 22 and 30 years, respectively). Measurement of such variations by conventional methods is very difficult in many cases especially at remote sites. Application of the radioactive tracer technique to deposition studies in several sites in Scotland, Sweden and Northern Iran has lead to results which found to be consistent with deposition estimates obtained from long-term continuous record of cloud frequency and meteorological variables, and also in good agreement with model deposition estimates. The technique is operationally easier to carry out than the direct measurement by artificial collectors over extended period of time, and is particularly valuable in quantifying aerosol and wet deposition processes at sites where conventional methods are not applicable.

INTRODUCTION

The rates of both dry deposition of particles and occult deposition are largely dependent on surface properties and in the case of deposition to forests are rather uncertain [1]. Neither dry deposition nor cloud droplet interception is easily quantified and many studies have been devoted to an assessment of their contribution to total deposition to forests. However, there is a lack of large-scale, long-term measurements in this regard and there is a large gap between the results from field experiments, wind tunnel studies, and model estimates [2].

In the process of establishing critical loads, soil acidification in forests as well as the influence of upland forests on pollutant inputs to catchments needed to be taken into account. Thus, it is important to be able to quantify the atmospheric input to forests in a reasonable way. The quantification is necessary on a local level to make a linkage between modeled deposition estimates and soil loads. Moreover, if the intent is to help protect sensitive areas then there is no substitute for good on-site data [3].

Wet deposition is known to be rather evenly distributed over large areas and its measurement is more straightforward than the measurement of dry and occult deposition by using precipitation collectors. The exception is in upland areas where orographic

enhancement of rainfall occurs and an assessment of the extent of seeder-feeder effect has to be performed.

Several methods exist to estimate the dry and occult deposition to forests on small spatial scales. However, direct measurement of aerosols and cloud droplet deposition is difficult except by micrometeorological methods that suffer mainly from their limitation to certain terrain situations.

In this paper, we report measurements of the soil inventory of atmospherically derived naturally occurring radioisotope ^{210}Pb and the anthropogenic one, ^{137}Cs, to quantify the effects of aerodynamic roughness and orography on the deposition of atmospheric aerosols as particles and/or droplets. This independent technique may enable improved estimates of the effects of land use on long-term inputs of pollutants in precipitation, cloud droplets or as aerosol.

EXPERIMENTAL METHODS

The Sites and Sampling Method

Initial sampling was conducted at Dunslair Heights beneath a Norway spruce canopy and in the adjacent open grassland at an elevation of 450 m asl in the Scottish Southern Uplands. The second site was an old plantation of Scots pine at 420 m asl near the summit of Dun Coillich in the Scottish Highlands. The method was then applied to measure the pattern of total aerosol deposition at ten sites in southwest Sweden, along a transect from Hallands Vadero Island at the coast to the southern Swedish uplands. Afterwards, a series of samples were collected at four sites in the Transtrand Mountains in Central Sweden. Recently, the technique has been applied to several sites in Northern Iran, from -20 m asl to 2050 m asl, in conjunction with a survey of natural radioactivity in the North Alborz ecosystem.

Split-level sampling technique were applied to determine the depth profiles of ^{210}Pb and ^{137}Cs from the surface to 20-30 cm at selected locations from within the canopy as well as the adjacent open land, using hand-driven brass pipe corers and metal surface frames.

The Radioactive Tracer Technique

Natural and artificial radionuclides present in the atmosphere (radon and its daughters, cosmogenic origin beryllium-7, fission products) have long been widely used as atmospheric tracers. These radionuclides are associated with nonradioactive aerosols and hence can serve to trace the fluxes of aerosols to various surfaces. Lead-210 and caesium-137 have been shown to be particularly useful because they are associated mainly with submicron-sized aerosols, which contain the bulk of the pollutant sulfur and nitrogen [4 - 7]. The ^{210}Pb isotope (half-life: 22 y) is the decay product of ^{222}Rn that belongs to the ^{238}U decay series and readily diffuses from soil into the atmosphere. When scavenged from the atmosphere along with carrier aerosols, ^{210}Pb and the stratospheric origin ^{137}Cs (half-life: 30 y) are retained by the organic-rich surface horizons and clay layers of soil that acts as an efficient integrating collector. The residence time of ^{210}Pb in soils is far longer than its half-life [8], so in soils that are not physically disturbed for several decades, its inventory is at steady state. However, an

important correction has to be made to the inventory to compensate for any ^{210}Pb that is produced by the *in situ* decay of ^{222}Rn (termed the *supported* fraction).

While the absolute value for the ^{210}Pb inventory is influenced by larger-scale processes, and by the sources of air scavenged by precipitation, however, at a local scale, and to some extent over small regions, the long-term average ^{210}Pb concentration in the atmosphere may be considered reasonably constant. Thus, variability in the soil inventory of ^{210}Pb provides a direct measure of the local variability in deposition of aerosols (by wet, dry and cloud deposition). In the case of ^{137}Cs, the area of study should be much more limited.

When a steady state between atmospheric supply and radioactive decay exists, the flux of ^{210}Pb from the atmosphere (F_{Pb}) may be obtained as the product of the decay constant (λ) for ^{210}Pb and the inventory of unsupported (atmospheric) ^{210}Pb in the soil profile (I):

F_{Pb}	=	λ	x	I
Bq m^{-2} y^{-1}		y^{-1}		Bq m^{-2}

Its unsupported inventory in undisturbed soils may then be used as a measure of total aerosol deposition averaged over about 30 years, approximately the mean nuclear lifetime.

The specific activities of ^{210}Pb and ^{137}Cs in dried soil samples were determined by non-destructive γ-spectrometry using high resolution HPGe detectors fitted with Compton suppression systems. For the efficiency calibration and sample self-attenuation correction of 46.5 keV gammas from ^{210}Pb, the conventional spiked matrix method has been applied in this study. Measurement of ^{214}Pb inventories were also conducted for each sub-section of a core using the 351.9 keV gamma line from the decay of ^{214}Pb, in order to make corrections for the *supported* ^{210}Pb.

RESULTS AND DISCUSSION

Measurement of radionuclides inventories at the first site, Dunslair Heights, revealed an average canopy enhancement in deposition of approximately 37% (see Table 1), which is found to be consistent with deposition estimates obtained from a long-term continuous record of cloud frequency and meteorological variables, and is also in good agreement with the UK model deposition estimates for the site [9].

The magnitude of enhancement also compares very well with the canopy enhancement found in ^{137}Cs inventories at the site and those measured by Schreiber & Woerner and Bunzl & Kracke [10] in deposition of ^{90}Sr and ^{137}Cs in soils under spruce canopies relative to the adjacent open grasslands in Germany. Under the reasonable assumption of a steady state condition between the atmospheric supply of ^{210}Pb and its radioactive decay in soil, its mean flux to the open moorland is estimated to be 72 Bq m^{-2} y^{-1}. Considering the average annual rainfall of approximately 1200 mm for the site [9], this flux corresponds to a mean concentration of ^{210}Pb in rainfall of 60 mBq l^{-1} which compares very closely to the mean UK ^{210}Pb concentrations in rainfall of 77±14 mBq l^{-1} estimated by Smith et al. [11] and 74 mBq l^{-1} estimated by Eakins & Morison [12], and is consistent with the derived concentration of 62 mBq l^{-1} based on measured atmospheric flux by Clifton [13].

The average canopy enhancement in deposition measured at the second site, Dun Coillich, is approximately 36% relative to adjacent heath land in close correspondence with independent estimates of cloud droplet deposition in total sulfur deposition for the relevant National Grid square, derived from the recent 5 km x 5 km cloud deposition map of UK

	Open moorland	Inside the forest canopy		
Sample location	DH1	DH2	DH3	DH4
^{210}Pb inventory (± σ) Bq m^{-2}	2325 (114)	3121 (557)	3252 (436)	3211 (44)
Increase under canopy		34%	40%	38%
Average increase		37%		

Table 1: ^{210}Pb inventories at Dunslair Heights.

	Open heath land	Inside the forest canopy		
Sample location	DC1	DC2, at the edge	DC3, 20 m inside	DC4, 50 m inside
^{210}Pb inventory (± σ) Bq m^{-2}	3175(440)	4939 (313)	4354 (452)	3634 (187)
Increase under canopy		56%	38%	15%
Average increase		36%		

Table 2: ^{210}Pb inventories at Dun Coillich.

Deposition at the exposed edge of stand, however, is 56% larger than that in the open heath land and 36% larger than the inventory deep inside the canopy, which shows a pronounced *edge effect* in ^{210}Pb deposition (see Table 2). The measured value of canopy enhancement is entirely consistent with the one found for ^{137}Cs inventories at the site and also in close agreement with values reported by Bunzl & Kracke [11]. The mean ^{210}Pb flux to the open heath land is estimated to be 98 Bq m^{-2} y^{-1}, which corresponds to a mean concentration of ^{210}Pb in rain of 75 mBq l^{-1}. The derived concentration is consistent with the mean UK ^{210}Pb concentrations in rainfall mentioned above.

The measured ^{210}Pb inventories at ten sites along a SW to NE transect in Hallands Province of southwest Sweden (Figure 1) show that the deposition increases quite markedly with distance inland to a maximum in upland sites, (G, H & I) roughly 20-30 km from the coast and decreases by almost 30% relative to the maximum, at a site about 60 km from the west coast (site J). This follows the trend in long-term precipitation variation along the transect (see Figure 2). The trend in sulfur wet-deposition calculated from data on its average concentration in rainfall [14 & 15] and sulfur deposition estimates by a regional atmospheric dispersion model developed by Langner et al. [16] show a broadly similar overall pattern. The canopy enhancement in deposition was found to be relatively high for quite modest elevations

of few hundred meters at 20-30 km from the coast, with no enhancement in the coastal region, suggesting that the significant enhancements is due to cloud/fog deposition at elevated sites in-land. The increased input to exposed forests on elevated hills in-land is up to 100% relative to that in open fields covered by short vegetation in the coastal area. The mean ^{210}Pb flux would be approximately 81 and 119 Bq m^{-2} y^{-1} for the open fields in the coastal and upland regions, respectively. These values and their average of 100 Bq m^{-2} y^{-1} compare very well with measurements reported by Al-Doushy [17] and Preiss & Genthon [18], and the estimated value from model-calculated contours by Lee & Feichter [19].

Four sites on the two adjacent mountains, Gammalsaters and Skaftasen, from the Transtrands range in Central Sweden were the other sites sampled in this study. Measurements revealed a significant increase in ^{210}Pb inventory on the summit of Gammalsaters, 870 m asl (site A), relative to the valley at 540 m ASL (site C), (see Figure 3). A higher rate of increase with elevation was observed for atmospheric ^{210}Pb inventories under forest canopies sampled in these mountains relative to the open fields, due to enhanced cloud droplet and aerosol deposition over forest caused by enhanced turbulence and greater cloud frequency combined with greater wind speed. Same increases were observed in an independent study of the nitrogen content of *Calluna vulgaris* (as an indication of the magnitude of N deposition) at these sites by Pitcairn et al. [20]. The mean inventory at the exposed edge of a forest canopy at 620 m asl (site D) exceeds that of open grassland at an elevation of 540 m asl (site C) by approximately 90%.

The mean flux to the open fields situated at 540, 740, and 870 m asl are approximately 86, 100 and 113 Bq m^{-2} y^{-1}, respectively. Attributing the increase in inventories at higher elevation sites to most likely orographic effect and cloud deposition, the measured flux of 86 Bq m^{-2} y^{-1} at the lower elevation site is in good agreement with measurements reported by Al-Doushy [17]. The average flux at Gammalsaters is approximately 100 Bq m^{-2} y^{-1}, which is consistent with the calculated mean value for the latitude band 52.5^{o}-62.5^{o} N based on the reported fluxes of ^{210}Pb in Europe by Preiss & Genthon [19], and the value derived from model-calculated contours by Lee & Feichter [19].

The measured ^{137}Cs inventories at six sites along a NE to SW transect in Guilan Province of Northern Iran show that the deposition decreases with distance from Caspian Sea shore (-20 m asl) up to 20 km inland at an elevation of about 1000 m asl (locations A - D in Figure 4), then a quite markedly increase of around 70% is observed roughly 30 km from the coast at an elevation of 2050 m asl (location E), and decreases by almost 25% relative to the maximum, at a site about 35 km from the coast at an elevation of 1950 m asl (location F). This follows the trend in long-term precipitation variation along the transect up to location D. Considering the topography and meteorological condition of the site, the increase in deposition at locations E & F could be mainly due to seeder-feeder effect and occult deposition enhancement. Location E is a highly exposed site close to the summit looking towards sea, whereas F is located on the opposite front of the mountain, which could have been less affected by occult deposition. Lead-210 inventory measurements of these soil samples are underway.

The depth profiles of ^{210}Pb in more than 500 collected soil samples demonstrate the efficient immobilization of the radionuclide in the surface horizons of soil and show no evidence of a significant accumulation of atmospheric ^{210}Pb below 10 cm depth. The soil inventories of ^{210}Pb provide a measure of long-term variation in deposition rate of atmospheric aerosols as particles and/or droplets that is operationally easier to carry out than the direct measurement by artificial collectors over extended period of time. It appears to be

particularly valuable in quantifying aerosol and wet deposition processes at sites where conventional methods are not applicable.

Based on measurements carried out in this study, it may be concluded that ^{137}Cs has the potential of comparisons within a limited region, but definitely not in-between sites largely as a consequence of the patchy deposition of this radionuclide in convective precipitation associated with the Chernobyl ^{137}Cs deposited in 1986. Moreover, it is found that it has a high mobility in highly organic and acidic soils.

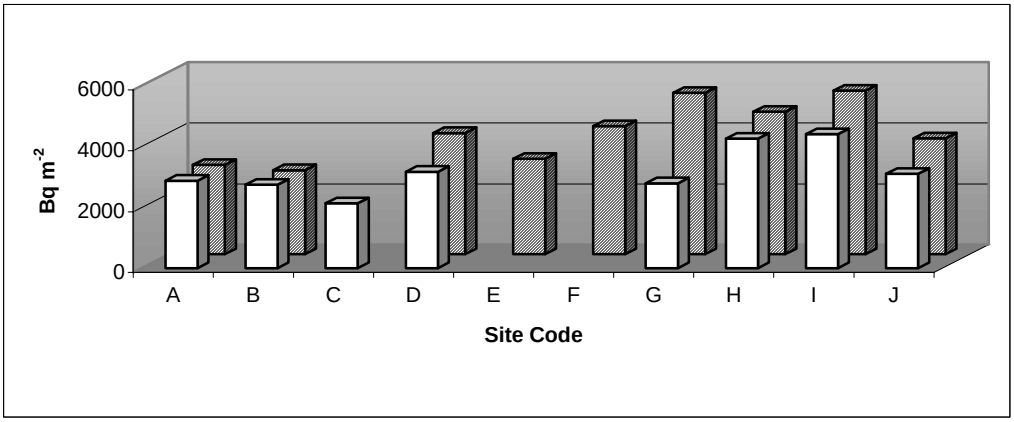

Fig. 1. Mean atmospheric ^{210}Pb soil inventories in the open field (front row) and inside forest canopies (back row) along the SW Sweden transect.

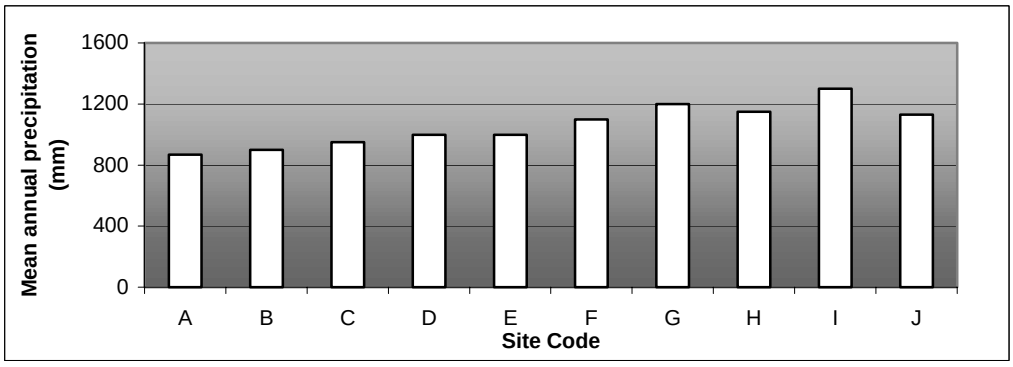

Fig. 2. Mean annual precipitation along the SW Sweden transect derived from precipitation map based on records in the period of 1983-90 [adapted from ref. 14].

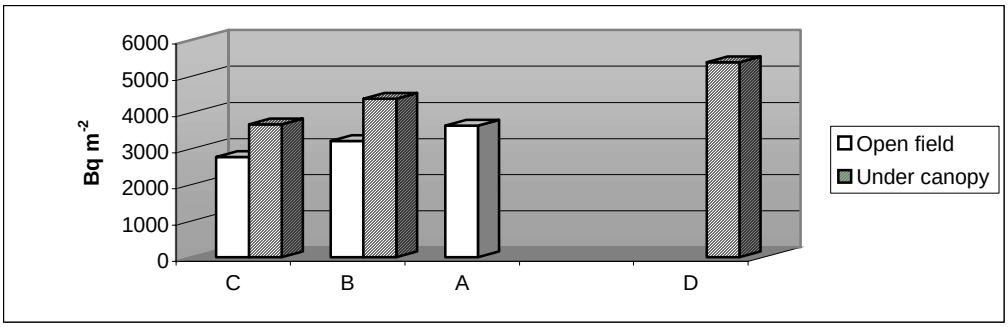

Fig. 3. Mean atmospheric ^{210}Pb inventories in the open fields and inside the forest canopies at Transtrand range in Central Sweden.

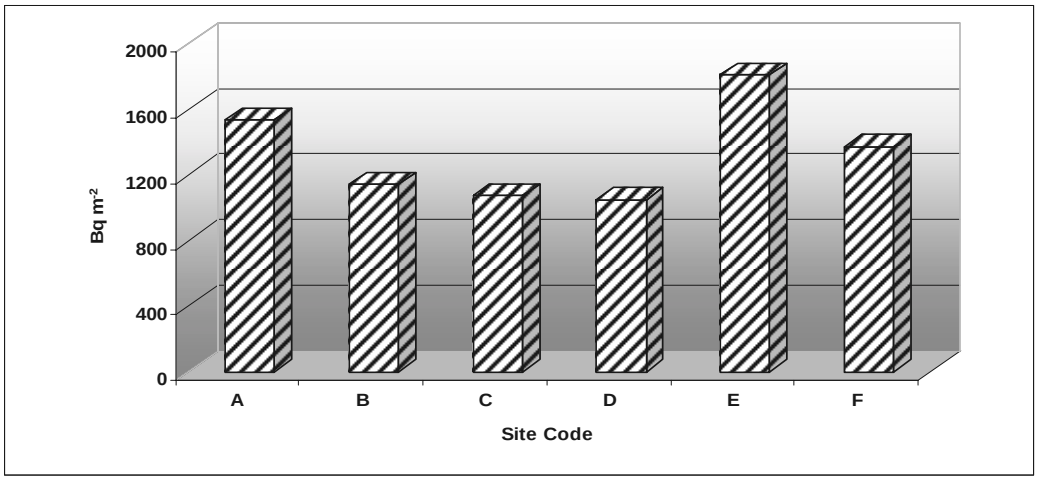

Fig. 4. Mean ^{137}Cs inventories along Asalem-Khalkhal transect in Northern Iran.

REFERENCES

[1] J. W. Erisman et al., "Particle deposition to forests", In: G J Heij and J W Erisman (eds), *Acid Rain Research: Do we have enough answers?* Elsevier Science, Amsterdam, pp 115-126, 1995.

[2] W. Ruijgrok et al., "Dry deposition of particles: Implications and recommendation for mapping of deposition over Europe", *Tellus*, 47B, pp 587-601, 1995.

[3] B. B. Hicks, "On the determination of total deposition to remote areas", in: G J Heij and J W Erisman (eds), *Acid Rain Research: Do we have enough answers?* Elsevier Science, Amsterdam, pp 163-173,1995.

[4] W. C. Graustein and K.K. Turekian, "^{210}Pb and ^{137}Cs in air and soils measure the rate and vertical profile of aerosol scavenging", *J. Geophys. Res.*, 91, No. D13, pp 14355 -14366, 1986.

[5] D. Fowler et al., “Quantifying fine-scale variability in pollutant deposition in complex terrain using ^{210}Pb inventories in soil”, *Water, Air and Soil Pollution*, 105, pp 459-470, 1995.
[6] D. Branford et al., “Spatial variation of wet deposition rates in an extended region of complex topography deduced from measurements of ^{210}Pb soil inventories”, *J. Environ. Radioactivity*, 41, No. 2, pp 111-125, 1998.
[7] M. Vahabi-Moghaddam et al., “Study of aerosol deposition at a wind exposed forest edge using ^{210}Pb soil inventories”, *6th International Conference on Acidic Deposition*, Tsukuba, Japan, 10-16 December 2000.
[8] L. K. Benninger et al., “The use of natural ^{210}Pb as a heavy metal tracer in river estuarine systems”, in: T M Church (ed), *Marine Chemistry in the Coastal Environment*, ACS Symp. Vol. 18, American Chemical Society, New York, pp 201-210, 1975.
[9] A. Crossley, “Particles in orographic cloud and the implication of their transfer to plantsurfaces”, in:M H Unsworth and D Fowler (eds), *Acid Deposition at High Elevation Sites*, Kluwer, Dordrecht, pp 553-66, 1988.
[10] K. Bunzl and W. Kracke, “Cumulative deposition of ^{137}Cs, ^{238}Pu, $^{239+240}$ Pu and ^{241}Am from global fallout in soils from forest, grassland and arable land in Bavaria (FRG)”, *J. Environ. Radioactivity*, 8, pp 1-14, 1988.
[11] J. T. Smith et al., “Inventories and fluxes of ^{210}Pb, ^{137}Cs and ^{241}Am determined from the soils of three small catchments in Cambria, UK”, *J. Environ. Radioactivity*, 37, pp 127-142, 1997.
[12] J. D. Eakins and R. T. Morrison, *A new procedure for the determination of lead-210 in lake and marine sediments*, AERE, R-8475, HMSO, London, 1976.
[13] R. J. Clifton, “The use of radionuclides (unsupported ^{210}Pb, ^{7}Be and ^{137}Cs) in describing the mixing characteristics of estuarine sediments”, In: P J Kershaw and D S Woodhead (eds.), *Radionuclides in the study of marine processes*, Elsevier Applied Science, Essex, England, pp 255-264, 1991.
[14] L. Granat, *Luft-Och Nederbordskemiska Stationsnatet inom PMK NaturvardsverketRapport 3942*,SERI, Sweden, 1990.
[15] O. Westling et al., *Air pollution in southern and central Sweden: Deposition and Effects*, IVL Report B-1079, SERI, Sweden, 1992.
[16] J. Langner et al., “Concentration and deposition of acidifying air pollutants over Sweden”, *Water, Air and Soil Pollution*, 85, pp 2021-2026, 1995.
[17] F. Al-Doushy, “The value of ^{210}Pb in dating Scandinavian aquatic and peat deposits”, *Radiocarbon*, 28, pp 1031-1040, 1986.
[18] N. Preiss and C. Genthon, “Use of a new database of ^{210}Pb for global aerosol model validation”, *J. Geophys. Res.*, 102, No. D21, pp 25347-25357, 1997.
[19] H. N. Lee and J. Feichter, “An intercomparison of wet precipitation scavenging schemes and the emission rates of ^{222}Rn for the simulation of global transport and deposition of ^{210}Pb”, *J. Geophys. Res.*,100-D11, pp 23253-23270, 1995.
[20] C. E. R. Pitcairn et al., “Foliar nitrogen as an indicator of nitrogen deposition and critical loads Exceedance on a European scale”, *Water, Air and Soil Pollution*, 130, pp1037-1043, 2001.