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Wet deposition of ^{210}Pb aerosols over two areas of contrasting topography

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Abstract
Deposition fluxes of ^{210}Pb on low and moderately high-elevation sites of Edinburgh (Scotland) and mid-Wales, respectively, have been measured. The excess ^{210}Pb fluxes in moorland Edinburgh soils did not vary significantly and ranged from 71 to 92 Bq m^{-2} y^{-1} with a mean value of 78 ± 9 Bq m^{-2} y^{-1}, for all the measured sites where both altitude and the mean annual rainfall are similar. On the other hand, the excess ^{210}Pb measured in moorland soils of mid-Wales sites increased by a factor of 2.4 at or near the summit (741 m asl) relative to the coast (∼15 m asl), whereas rainfall increased by a factor of 1.8 over the same height range. On average, the summit to valley ratio of ^{210}Pb concentration in rainfall was a factor of 1.3 due to scavenging of the feeder clouds by the seeder rain. These results are consistent with results for both modelled and field studies on the wet deposition of pollutants in complex terrain reported by several researchers. The long-term ^{210}Pb wet deposition field data will provide an important input parameter for the modelling of wet deposition of aerosols throughout the uplands of the UK and elsewhere where the seeder–feeder process is of common occurrence.

Keywords: ^{210}Pb, wet deposition, orography, enhancement, seeder–feeder

1. Introduction

The fallout of naturally occurring ^{210}Pb provides an important tool for studying pollutant transport processes and deposition fluxes to terrestrial ecosystems. Although there is a large body of data on ^{210}Pb worldwide, this is mainly limited to relatively lowland areas. Aerosol deposition flux measurements over mountain terrain is complicated in several ways by the meteorological conditions that are typical of high elevations. High elevation sites induce additional deposition processes that are not major contributors to deposition in lowland sites. These processes affect the efficiency with which aerosols are scavenged from the atmosphere.

Earlier studies involving measurements of major ionic concentrations as a function of elevation have been conducted to improve the estimates of acid quantities deposited in elevated sites. Results presented by Fowler *et al* [1] from precipitation events measured at levels between 244 and 847 m above sea level (asl), on the slopes of Great Dun Fell (Cumbria, UK) indicate an increased ionic concentration of 2.2 to 3.1 between the valley (∼250 m asl) and the summit (∼847 m asl). The increased concentrations of ions observed were interpreted as resulting from the seeder–feeder process; a mechanism first put forward by Bergeron [2], who proposed that rain falling from the high altitude (seeder) clouds wash out small droplets within the low-level cap (feeder) clouds formed by ascent over the hills and consequently grow by accreting cloud drops. The seeder–feeder mechanism was also supported by theoretical studies of Storebø [3], Bader and Roach [4]. Hill *et al* [5] presented detailed case studies of orographic rain falling over the hills of south Wales using data obtained from scanning radar combined with a network of autographic raingauges. They found that the orographic rainfall enhancement depended on the low-level wind speed, and that over 80% of the total orographic enhancement occurred in the lowest 1500 m above the hill. Model studies of the seeder–feeder effect by Jones and Choularton [6] support this interpretation and also show

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Table 1. Details of sampling sites at mid-Wales location.

Sites	Elevation (m asl)	Rainfall (mm y^{-1})	Notes
YH	15	1300	Five mineral-rich soil samples excavated beneath ∼15 m tall oak woodland canopy and five more from an open grassland covered with ∼10 cm tall grass
BM	430	1800	Seven organic-rich soil samples obtained beneath a Sitka spruce (*Picea sitchensis*) woodland canopy and another six from an open grassland covered with approximately 10 cm tall grass
PL	741	2400	Five samples obtained from an open grassland that was at the summit of Plynlimon mountain
TN	350	2000	Five organic-rich samples obtained from ∼20 cm tall grass and five more samples obtained from the adjacent (approximately 200 m away) 20 m tall Sitka spruce plantation

that the rainfall enhancement and deposition are a function of topography and spatial scale. These results are valuable to improve the wet deposition map of the UK, particularly over the Pennines, the Lake District and Snowdonia where the seeder–feeder effect constitutes a large fraction of annual deposition [7].

Following the observations described above, a technique pointed out by Graustein and Turekian [8], which involves the use of fallout ^{210}Pb and ^{137}Cs accumulated in undisturbed soils to measure the rate of aerosol deposition, was employed to make inferences regarding the above mechanisms. The widely used method for determining the atmospherically derived ^{210}Pb fluxes in soils, given in equation (1), was employed [9–13]:

$$\Phi = \lambda(^{210}\mathrm{Pb}) \times I \quad (1)$$

where Φ is the total flux, including wet and occult deposition processes, (in Bq m^{-2} y^{-1}) of the radionuclide to the Earth's surface, λ is the ^{210}Pb decay constant (0.0311 y^{-1}) and I is the ^{210}Pb inventory in soil, measured in Bq m^{-2}. Equation (1), however, is used based on the assumption that the soils have not been disturbed for at least 100 years to be able to distinguish between the fallout and the *in situ* component of ^{210}Pb in the soil profile, and that the ^{210}Pb inventory is in a steady state, with the rate of ^{210}Pb atoms equalling the mean deposition rate.

The use of soil samples as long-term collectors of atmospheric deposition has advantages over direct collection of precipitation in that: (1) there is a greatly reduced time and expense required for sampling and (2) the deposition rates are relatively free of artefacts introduced by artificial collection apparatus. Therefore, the ^{210}Pb measurement in soils provides practical studies of the effect of local variables such as topography, cloud and vegetation, on the inventories of aerosols on the land surfaces.

This letter reports ^{210}Pb deposition fluxes measured in moorland and woodland soils collected from the UK lowland and moderately high-elevated sites of Scotland and Wales, respectively. The measurements were carried out for the following reasons.

(i) To determine whether high elevation sites have an influence on the long-term annual average wet deposition of ^{210}Pb isotopes.

(ii) To determine whether the presence of clouds frequently shrouding hilltops have a significant influence on the deposition of ^{210}Pb, assuming a constant concentration of the isotope in rainfall and negligible contributions from dry deposition processes.

2. Methods

Two areas of contrasting topography: lowland (<300 m asl) sampling sites located in Edinburgh, Scotland and moderately high-elevated sites in mid-Wales were selected for this study.

2.1. Edinburgh sites

Soils were obtained from five different sites, each with relatively insignificant undulations, and four of which were well-established golf courses. The sites were selected on the reasonable assumption that the soils have not been subjected to erosion or other forms of disturbance in recent decades. From the four golf courses, the rough was chosen since intense perturbation from human activity was expected to be minimal. Furthermore, according to the information gathered from the greenkeepers, grass that was cut on the rough was always left on the site. It is therefore expected that the radioactivity that had deposited onto the rough was not transported elsewhere. Detailed site description and sampling techniques are given elsewhere [14].

2.2. Mid-Wales sites

The four sites selected for this study are: Ynys Hir (YH), grid reference number SN 682 963; Bryn Mawr (BM), grid ref. SN 727 902; Plynlimon Mountain (PL), grid ref. SN 819 883 and Tanllwyth valley (TN), grid ref. SN 855 868. Soils were sampled in June 2002. The sites are situated 7.5, 15, 24, and 28 km, respectively from the west coast lying approximately along a transect leading ESE of the grid north. The same sampling and analytical procedures as for the Edinburgh sites [14] were followed. Rainfall data was obtained from the UK Meteorological Office Map [15]. Information on site history was obtained from the Centre for Ecology and Hydrology staff whereas that of soil characteristics was by

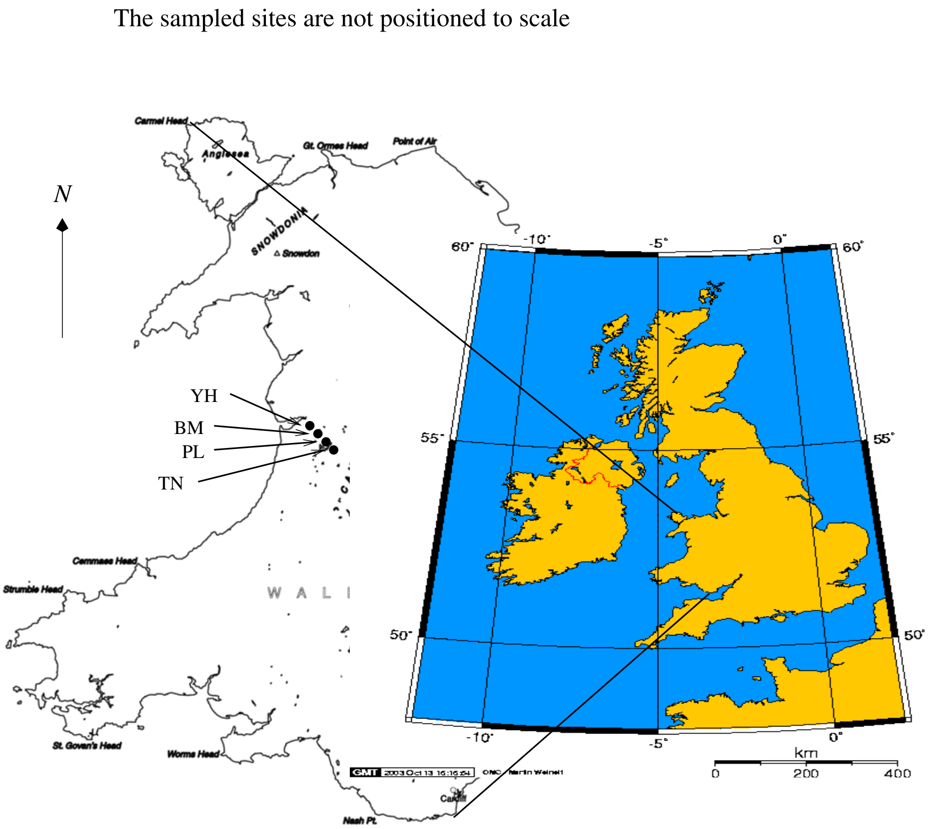

Figure 1. A map of Wales showing approximate positions of the sites sampled: Ynys Hir (YH), Bryn Mawr (BM), Plynlimon (PL) and Tanllwyth Valley (TN).

visual inspection. Figure 1 shows approximate locations of sites sampled in mid-Wales. Table 1 gives a detailed summary of information about the sampled sites.

2.2.1. YH. Ynys Hir, situated ~7.5 km from the coast, was selected on the basis that it will provide the ^{210}Pb flux representative of low altitude sites. Soil samples were collected from both the open grassland and beneath the adjacent moderately dense 10–15 m high oak woodland canopy. The soils were rich in mineral content and contained rocks (mostly slate).

2.2.2. BM. The choice for Bryn Mawr was based on its enhanced rainfall due to the extent of land mass from the coast and rise in altitude relative to YH. Thus, this site was expected to provide information on the effect of increased annual average rainfall on the deposition of ^{210}Pb relative to that at the coast.

2.2.3. PL. The summit of Plynlimon was selected for sampling with the aim of investigating the effect of meteorological conditions on the deposition of aerosols by comparing the measured ^{210}Pb flux at the site with those of relatively low elevation sites. The soil at this site was mainly organic matter at the topmost and mineral matter at depths >10 cm. The grass at the top was relatively short with features suggesting that the area was almost certainly grazed. Adjacent to the site, approximately 200 m eastward, was a Sitka spruce plantation. The samples obtained under the Sitka canopy were disregarded since the soils looked heavily disturbed.

2.2.4. TN. Tanllwyth was selected because of its position on a leeward side of Plynlimon mountain, so as to provide information on the 'shadow effect' in the deposition of ^{210}Pb aerosols. The open grassland soils were poorly drained.

3. Results and discussion

3.1. ^{210}Pb deposition—Edinburgh

Fluxes of excess ^{210}Pb measured from the five (I–V) Edinburgh sites are given in table 2. Since more than one sample was obtained from each site, statistical means and sample standard deviations relative to the mean values were calculated to estimate the deposition fluxes.

Deposition fluxes of ^{210}Pb measured in moorland soils of Edinburgh varied from 71 to 92 Bq m^{-2} y^{-1}, with a mean value of 78 ± 9 Bq m^{-2} y^{-1}. Although the sample size is small, the trends in terms of the mean values are such that there is no significant spatial variability in the deposition of ^{210}Pb. These

Table 2. Mean fluxes of excess ^{210}Pb in Edinburgh moorland and woodland soils. The letter n denotes the number of cores analysed for each population. The dashes (—) indicate that no woodland data were available for the sites.

	^{210}Pb (Bq m^{-2} y^{-1})		
	Moorland (M)	Woodland (W)	Ratio (W/M)
Site: I [$n = 9$] [rain = 700 mm y^{-1}; elevation = 180 m asl]			
Range	72–115	—	
Mean	92 ± 12	—	—
Site: II [$n = 4$ (M); $n = 4$ (W)] [rain = 700 mm y^{-1}; elevation = 50 m asl]			
Range	60–86	94–127	
Mean	73 ± 11	113 ± 16	1.55
Site: III [$n = 4$ (M); $n = 5$ (W)] [rain = 850 mm y^{-1}; elevation = 230 m asl]			
Range	69–88	87–109	
Mean	77 ± 9	101 ± 8	1.31
Site: IV [$n = 4$ (M); $n = 5$ (W)] [rain = 1000 mm y^{-1}; elevation = 260 m asl]			
Range	74–83	113–144	
Mean	80 ± 5	124 ± 12	1.55
Site: V [$n = 8$] [rain = 700 mm y^{-1}; elevation = 40 m asl]			
Range	40–114	—	
Mean	71 ± 23	—	—
Mean	78 ± 9	113 ± 12	1.47

Table 3. Mean fluxes of excess ^{210}Pb in moorland and woodland soils in Wales. The letter n denotes the number of cores analysed for each population. The dashes (—) indicate that no woodland data were available for the sites.

	^{210}Pb (Bq m^{-2} y^{-1})		
	Moorland (M)	Woodland (W)	Ratio (W/M)
Site: YH [$n = 5$ (M); $n = 5$ (W)] [rain = 1300 mm y^{-1}; elevation = 15 m asl]			
Range	99–128	157–214	
Mean	119 ± 12	181 ± 21	1.52
Site: BM [$n = 6$ (M); $n = 7$ (W)] [rain = 1800 mm y^{-1}; elevation = 430 m asl]			
Range	94–254	168–386	
Mean	182 ± 70	244 ± 70	1.34
Site: PL [$n = 4$] [rain = 2400 mm y^{-1}; elevation = 741 m asl]			
Range	239–321	—	
Mean	288 ± 40	—	—
Site: TN [$n = 5$ (M); $n = 5$ (W)] [rain = 2000 mm y^{-1}; elevation = 350 m asl]			
Range	189–277	253–254	
Mean	216 ± 35	264 ± 9	1.22

results compare well with ^{210}Pb deposition fluxes reported by several investigators in other UK sites where both altitude and the mean annual rainfall are quite similar, for example; Fenwick, Scotland and Devon, England; with deposition fluxes of 86 and 95 Bq m^{-2} y^{-1}, respectively [16, 17]. At sites (II–IV) where both moorland and woodland data were available, the woodland mean ^{210}Pb deposition flux was (47 ± 7)% over that of moorland. Assuming the same precipitation input at all sites, enhanced ^{210}Pb input in woodland soils may be attributed to occult deposition. Furthermore, because forests possess structures that promote capture of atmospheric particulates, retained ^{210}Pb aerosols subsequently fall beneath woodland canopies by mechanisms such as wash-off, gravitational settling and as senescent leaves. Fowler *et al* [18] reported soil inventories of excess ^{210}Pb under woodland canopy exceeding those under grass by between 22% and 60%. Similarly, independent studies by Branford *et al* [19] revealed an average enhancement factor of 36% for ^{210}Pb deposited inside forest canopies relative to open grassland soils in the Highlands of Scotland.

3.2. ^{210}*Pb deposition—mid-Wales*

Fluxes of excess ^{210}Pb measured from the four sites in mid-Wales (YH, BM, PL and TN) are given in table 3. It is evident from the table that the ^{210}Pb deposited in moorland increases significantly ($r^2 = 0.90$; $p < 0.05$) with increase in altitude. Because the removal of atmospheric ^{210}Pb occurs primarily through wet deposition processes and the fluxes of the isotope in soils are expected to be closely related to the rainfall pattern, it is useful to first discuss the mean annual rainfall at the sites.

3.2.1. Rainfall increase with altitude. Taking the mean annual rainfall at the site of YH to represent the non-enhanced UK west coast rainfall, the enhancement factors in rain falling over BM, PL and TN due to orography are 1.38, 1.85 and 1.54, respectively. According to Pedgley [20], there are two principal reasons for heavy rains over mountain areas: (1) mountains act as barriers to moist airstreams. The moist air is forced to rise, producing clouds by the process of expanding and cooling of air; and (2) when the days are sunny,

Table 4. The ^{210}Pb concentrations and the input of liquid water content at the summit of Plynlimon relative to that in the rainfall at the coast. The dashes (—) indicate that the enhancement factors were insignificant.

Sites	^{210}Pb concentration (mBq m^{-3})	Ratio Summit/valley
YH	92	—
BM	101	—
PL	120	1.3
TN	95	—

mountains act as high level heat sources. Thus, convective cloud formation may result, even when the effect in (1) is negligible. Enhanced rainfall over moderately sized hills of western Britain has been attributed to the effect of orography on frontal systems coming from the Atlantic Ocean, mainly westerly and/or south-westerly directions [21]. Considering the enhancement factors above, it seems that the rainfall is already significantly enhanced at BM due to uplifting of the airmass by the extent of the 15 km land from the coast.

3.2.2. Profile of ^{210}Pb deposition. The distribution of the deposited ^{210}Pb is characterized by increasing values with the increase in altitude. For moorland data, the ^{210}Pb flux increased by factors of 1.53, 2.42 and 1.82 for BM, PL and TN, respectively, relative to YN. The decrease in enhancement factors from 2.42 (for PL) to 1.82 (for TN) indicates that the seeder–feeder process becomes less effective as the altitude decreases, moving eastward of the hill summit. In this region downwind of the hill, the water vapour content of air is reduced due to scavenging by rain as it passes through the feeder cloud. Thus, the zone of maximum ^{210}Pb deposition is assumed to be at or near the summit for the sampled sites along the transect.

The average ^{210}Pb concentration in rain, C_{rain} (in mBq m^{-3}) can be deduced from the long-term ^{210}Pb atmospheric deposition flux, Φ, given in table 3 and the average rainfall, R (in mm y^{-1}) using the equation:

$$\Phi = C_{\text{rain}} \times R. \qquad (2)$$

The ^{210}Pb concentration obtained this way, however, assumes that all the ^{210}Pb atoms reaching the surface of the earth are delivered by precipitation alone. Thus, these values may be an overestimate, considering that a proportion of the ^{210}Pb atoms will be delivered by processes of dry and occult deposition. Considering the ^{210}Pb concentrations in rain, deduced from equation (2), it is possible to determine whether or not ^{210}Pb concentration in rain water is enhanced at the summit of Plynlimon as would be expected. These results are given in table 4.

The average increase in ^{210}Pb fluxes from the west coast to the summit of Plynlimon was 2.4 whereas rainfall increased over the same height range by a factor of 1.8. This provides a 30% increase in the ^{210}Pb summit/valley concentration ratio in rainfall. For the purpose of comparison, table 5 presents enhancement factors (summit/valley) for ^{210}Pb conducted at Great Dun Fell [22].

Table 5. Characteristics of Plynlimon ^{210}Pb wet deposition data compared to that reported for Great Dun Fell [22].

Description	Reference [22]	This study
Altitude range (m)	160–840	15–741
Rainfall increase from valley to summit	2.0	1.8
^{210}Pb increase between valley to summit	3.3	2.4
^{210}Pb concentration in precipitation (summit/valley)	1.7	1.3

Results obtained from this study when compared with other results obtained from both field measurements of SO_4^{-2}, NO_3^-, NH_4^+ [1, 7], ^{210}Pb [22] and model results [23, 24, 6] show a similar trend in increased wet deposition on hills due to the seeder–feeder scavenging process. Comparing these results with those obtained for the relatively low elevation sites of Edinburgh discussed in section 2.1, it is evident that high elevation sites have a significant influence on the deposition of aerosol-borne ^{210}Pb due to the presence of clouds frequently shrouding the hilltops.

4. Conclusion

Fluxes of excess ^{210}Pb were measured in presumably undisturbed soils obtained from low (<300 m asl) and moderately high-elevated sites of Scotland and mid-Wales, respectively. The results are such that the deposition flux of ^{210}Pb does not vary for the five sites where both rainfall and altitude are quite similar. On the other hand, excess ^{210}Pb measured at the summit of Plynlimon (741 m asl) increased by a factor of 2.4 greater than that of the coast (∼15 m asl) whereas rainfall increased by a factor of 1.8 over the same height range. The measurement showed that, on average, the summit/valley ratio of ^{210}Pb concentration in precipitation was 1.3 due to the seeder–feeder scavenging process. Results obtained from this study, together with other results reported elsewhere, are valuable to improve wet deposition maps in the uplands of the UK. This work needs to be extended, particularly over the Plynlimon hill profile with improved resolution to validate these results.

References

[1] Fowler D, Cape J N, Leith I D, Choularton T W, Gay M J and Jones A 1988 The influence of altitude on rainfall composition at Great Dun Fell *Atmos. Environ.* **22** 1355–62

[2] Begeron T 1965 On the low-level redistribution of atmospheric water caused by orography *Suppl. Proc. Int. Conf. Cloud Phys. (Tokyo)* pp 96–100

[3] Storebø P B 1976 Small scale topographical influences on precipitation *Tellus* **28** 45–59

[4] Bader M J and Roach W T 1977 Orographic rainfall in warm sectors of depressions *Q. J. R. Meteorol. Soc.* **103** 269–80

[5] Hill F F, Browning K A and Bader M J 1981 Radar and raingauge observations of orographic rain over south Wales *Q. J. R. Meteorol. Soc.* **107** 643–70

[6] Jones A and Choularton T W 1988 A model of wet deposition on complex terrain *Atmos. Environ.* **22** 2419–30

[7] Dore A J, Choularton T W, Fowler D and Storton-West R 1990 Field measurements of wet deposition in an extended region of complex topography *Q. J. R. Meteorol. Soc.* **116** 1193–212

[8] Graustein W C and Turekian K K 1989 The effects of forests and topography on the deposition of sub-micrometer aerosols measured by Lead-210 and Cesium-137 in soils *Agricultural Forest Meteorol.* **47** 199–220

[9] Benninger L K, Lewis D M and Turekian K K 1975 The use of natural lead-210 as a heavy metal tracer in the river-estuarine system *Marine Chemistry in Coastal Environment* vol 18 ed T M Church (Washington, DC: American Chemical Society) pp 202–10

[10] Moore H E and Poet S E 1976 ^{210}Pb fluxes determined from ^{210}Pb and ^{226}Ra soil profiles *J. Geophys. Res.* **81** 1056–8

[11] Nozaki Y, DeMaster D J, Lewis D M and Turekian K K 1978 Atmospheric ^{210}Pb fluxes determined from soil profiles *J. Geophys. Res.* **83** 4047–51

[12] Graustein W C and Turekian K K 1983 ^{210}Pb as a tracer of the deposition of sub-micrometer aerosols *Precipitation Scavenging, Dry Deposition, and Resuspension* (New York: Elsevier) pp 1315–24

[13] Graustein W C and Turekian K K 1986 ^{210}Pb and ^{137}Cs in air and soils measure the rate and vertical profile of aerosol scavenging *J. Geophys. Res.* **91** 14355–66

[14] Likuku A S, Branford D, Fowler D and Weston K J 2006 Inventories of fallout ^{210}Pb and ^{137}Cs radionuclides in moorland and woodland soils around Edinburgh urban area (UK) *J. Environ. Radioact.* **90** 37–47

[15] Meteorological Office 1977 *Annual Average Rainfall for (Northern and Southern) Britain: International Standard Period 1941–1970* Meteorological Office: Met.0.886 (NB & SB), London Road, Bracknell, Berkshire RG12 2SZ

[16] MacKenzie A B, Logan E, Cook G T and Pulford I D 1995 Radionuclide and metal distributions in peat deposition in Scotland *Heavy Metals in the Environment,* Hamburg vol 1 ed R D Wilken, U Forester and A Knockel (Edinburgh: CEP Consultants) pp 113–6

[17] He Q and Walling D E 1997 The distribution of fallout ^{137}Cs and ^{210}Pb in undisturbed and cultivated soils *Appl. Radiat. Isot.* **48** 677–90

[18] Fowler D, Skiba U, Nemitz E, Choubedar F, Branford D, Donovan R and Rowland P 2004 Measuring aerosol and heavy metal deposition on urban woodland and grass using inventories of ^{210}Pb and metal concentrations in soils *Water Air Soil Pollut.* **4** 483–99

[19] Branford D, Fowler D and Moghaddam M V 2004 Study of aerosol deposition at a wind exposed forest edge using ^{210}Pb and ^{137}Cs soil inventories *Water Air Soil Pollut.* **157** 107–16

[20] Pedgley D E 1970 Heavy rainfalls over Snowdonia *Weather* **25** 340–50

[21] Sawyer J S 1952 A study of rainfall of two synoptic situations *Q. J. R. Meteorol. Soc.* **78** 231–46

[22] Fowler D, Mourne R and Branford D 1995 The application of ^{210}Pb inventories in soil to measure long-term average wet deposition of pollutants in complex terrain *Water Air Soil Pollut.* **85** 2113–8

[23] Carruthers D J and Choularton T W 1983 A model of the seeder–feeder mechanism of orographic rain including stratification and wind-drift effects *Q. J. R. Meteorol. Soc.* **109** 575–88

[24] Hill T A, Jones A and Choularton T W 1987 Modelling sulphate deposition onto hills by washout and turbulence *Q. J. R. Meteorol. Soc.* **113** 1217–36