

Chapter (non-refereed)

Choubedar, F.; Branford, D.; Weston, K. J.; Fowler, D.; Nemitz, E.; Goulding, K.. 2001 Direct measurements of the long-term enhancement of aerosol deposition onto woodland using a 210Pb tracer method. In: Fowler, D.; Pitcairn, C., (eds.) *Air-surface exchange of gases and particles: poster proceedings*. Edinburgh, CEH, 80-83.

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Direct Measurements of the Long-Term Enhancement of Aerosol Deposition onto Woodland using a ^{210}Pb Tracer Method

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Abstract

The dry deposition flux of atmospheric aerosols has been measured using a ^{210}Pb tracer method. Inventories of ^{210}Pb in soil which have been undisturbed for in excess of 100 years at Rothamsted provide long term coverage dry deposition fluxes in the range $34 \text{ Bq m}^{-2} \text{ y}^{-1}$ for grassland, $60 \text{ Bq m}^{-2} \text{ s}^{-1}$ for deciduous woodland and $68 \text{ Bq m}^{-2} \text{ s}^{-1}$ for a deciduous shelter belt. These fluxes are equivalent to long-term average deposition velocities respectively of 4.6 mm s^{-1} , 8.3 mm s^{-1} and 9.4 mm s^{-1} for aerosols in the size range $0.2 - 0.5 \mu\text{m}$ diameter. The deposition velocities for woodland are substantially larger than those obtained in wind tunnel studies but are similar to field measurements by eddy co-variance methods.

Introduction

The earth's crust contains the radioactive element ^{238}U which decays through ^{226}Ra to ^{222}Rn . This element emanates out of the earth's crust into the atmosphere. ^{222}Rn (mean radioactive lifetime of 5.52 days) is a water-insoluble inert gas which is lost from the atmosphere by radioactive decay to ^{210}Pb . The decay series of ^{238}U is shown in Figure 1. ^{210}Pb atoms (mean radioactive lifetime of 32.3 years) become quickly attached to aerosol particles, (the same size aerosols as SO_2 and NO_x) and are removed from the atmosphere primarily by dry and wet deposition. The mass median diameter of ^{210}Pb -containing particles both in continental and marine environments is in the range of 0.3-0.4 micrometres; typically, 90% or more is present on particles less than 1.0 micrometre (e.g. Turekian *et al.*, 1989, Knuth *et al.*, 1983).

A small proportion of ^{222}Rn emanated from the earth's surface is redeposited as short-lived decay products, but most decay in the atmosphere into ^{210}Pb . The mean residence time of ^{210}Pb is short (a few days) compared with its radioactive half-life (22.3 years), so the downwards flux of ^{210}Pb , in $\text{atoms m}^{-2} \text{ s}^{-1}$, should equal the upwards flux of ^{222}Rn .

Sampling methods

When scavenged from the atmosphere along with carrier aerosols, ^{210}Pb is retained by the organic-rich surface horizontal soil which acts as an efficient integrating collector (Lewis, 1977). Forest soils are generally well suited for ^{210}Pb deposition rate measurements (Graustein and Turekian, 1983). The excess ^{210}Pb (^{210}Pb - ^{214}Pb) in each section of the core were measured. Non-destructive gamma-spectroscopy was used to measure ^{210}Pb in each core. ^{210}Pb total inventory in undisturbed soils was used as a measure of total aerosol deposition averaged over about 30 years, approximately the mean nuclear life time (30.2 years).

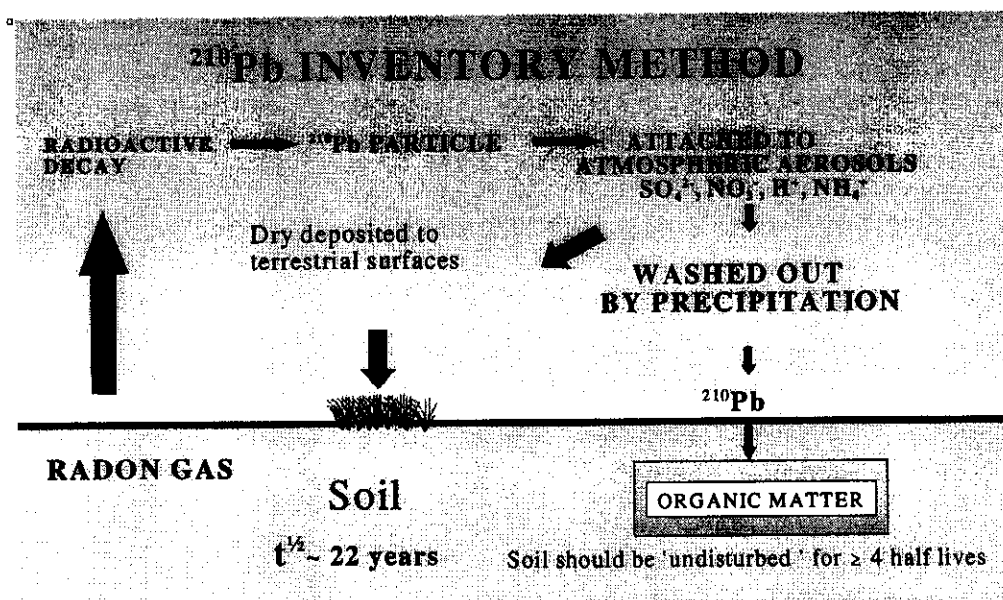
On the bases of the above criteria, soil samples were collected from locations in Rothamsted experimental station in Harpenden, Hertfordshire, England during February 1998.

In total from 10 sites, 50 samples were collected and divided to 150 sub-samples in 3 open areas and 2 woodlands. All of the samples were taken using a corer 10 cm diameter and 25 cm long.

Results and discussions

The ^{210}Pb isotope in the atmosphere is removed by precipitation and dry deposition and therefore provides a valuable tracer to study deposition processes. At a site which experiences little fog or orographic cloud, and where wet deposition is constant over different land uses, the measured inventory of ^{210}Pb in upper horizons of soil may be used to derive a mean annual deposition flux and to study the effect of the vegetation height and roughness length on the deposition flux and deposition velocity (V_{dd}).

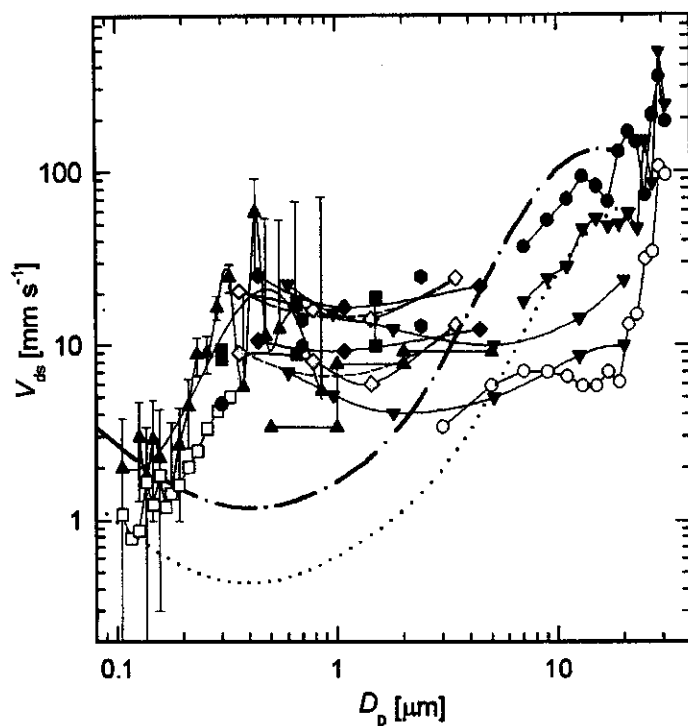
The atmospherically derived ^{210}Pb inventory into the canopies exceed those in adjacent open areas, indicating that forest sites receive more atmospheric inputs. The mean ^{210}Pb inventories are $3753 \pm 139 \text{ Bq m}^{-2}$ for open grassland, $4607 \pm 135 \text{ Bq m}^{-2}$ for Geescroft canopy and $4844 \pm 124 \text{ Bq m}^{-2}$ for Broadbalk canopy, shown in figure 1.



The results of these calculations together with the results from different studies and the wind tunnel results are plotted in Figure 2.

The inventories may be used to calculate the annual deposition fluxes directly. These fluxes may then be partitioned into dry and wet deposition by subtracting the much more readily measured wet deposition (which for this site was provided by measurements at Harwell). The dry deposition flux may be used to calculate deposition velocity using ambient aerosol ^{210}Pb concentration measurements (0.23 mBq m^{-3}).

The results of calculations from this study showed substantially larger dry deposition velocities (V_{dd}) in size range of ^{210}Pb carrier aerosols (sub-micron) than the wind tunnel prediction of dry deposition. The values of V_{dd} for this study were $4.7 \nabla 0.6 \text{ mm s}^{-1}$ for grassland, $8.4 \nabla 0.6 \text{ mm s}^{-1}$ for Geescroft woodland and $9.3 \nabla 0.5 \text{ mm s}^{-1}$ for Broadbalk woodland. These values are an order of magnitude larger than the wind tunnel results which imply that the wind tunnel measurements do not simulate the full range processes occurring in the field.



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|-------------------------------------------------------------------------------------------------------------------|--------------------------------------------------------------------------|
| ◇ Höfken and Gravenhorst (1983) | ▲ Lorenz and Murphy (1989) |
| ◆ Grosch and Schmitt (1988) | Gradient Loblolly Pine, $h_c = 9 \text{ m}$ |
| ■ Brückman (1988) | ● Höfken <i>et al.</i> (1983) TF Beech |
| □ Joutsenoja (1992) $u^* = 0.68 \text{ m s}^{-1}$,
$u = 5.6 \text{ m s}^{-1}$, $z_0 = 0.08 - 0.13 \text{ m}$ | ● Höfken <i>et al.</i> (1983) TF Spruce |
| ▼ Waraghai and Gravenhorst (1989) | ⋯ Slinn (1982), $u^* = 0.65 \text{ m s}^{-1}$ |
| ▼ Gallagher <i>et al.</i> (1992) | Unmodified Model Result |
| $u_* = 0.71 \text{ m s}^{-1}$, $z_0 = 15.7 \text{ cm}$ | — Slinn (1982), $u^* = 1.30 \text{ m s}^{-1}$ |
| ● Gallagher <i>et al.</i> (1992) | Unmodified Model Result |
| $u_* = 0.75 \text{ m s}^{-1}$, $z_0 = 18.6 \text{ cm}$ | ● Choubedar and Fowler (2000) |
| ○ Beswick <i>et al.</i> (1991) | ^{210}Pb grassland Rothamsted ($u_* = 0.14 \text{ m s}^{-1}$) |
| $u_* = 0.37 \text{ m s}^{-1}$, $z_0 = 30.1 \text{ cm}$ | ■ Choubedar and Fowler (2000) |
| ▲ Gallagher <i>et al.</i> (1997a) | ^{210}Pb forests Rothamsted ($u_* = 0.36 \text{ m s}^{-1}$) |
| $u_* = 0.5 \text{ m s}^{-1}$, $z_0 = 3.5 \text{ m}$ | |

Furthermore, the measurements from this study show that V_{dd} may be measured directly by knowing the soil inventory, rain flux and air concentration of the isotope.

Conclusions

Long-term average aerosol dry deposition fluxes and deposition velocities onto terrestrial

surfaces may be obtained from ^{210}Pb inventories in soils. Measurements of ^{210}Pb inventories in soils within the long-term Rothamsted experiments show annual dry deposition inputs into woodland larger than those into grassland by approximately a factor of 2. Deposition velocities for ^{210}Pb carrier aerosols in the size range 0.2 to 0.5 μm diameter were 4.7 mm s^{-1} for grass, 8.4 mm s^{-1} for deciduous woodland and 9.3 mm s^{-1} for a shelter belt deciduous woodland.

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