

Deposition Rates of Atmospheric Particulates Determined from ^{210}Pb Measurements in Soils and Air

Alfred S. Likuku^{1*}, Derek Branford²

¹Botswana College of Agriculture, Private Bag 0027, Gaborone, Botswana

²School of Physics and Astronomy, University of Edinburgh, James Clerk Maxwell Building,
Mayfield Road, Edinburgh EH9 3JZ, United Kingdom

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Abstract

Deposition rates of atmospheric particles were determined using previously published ^{210}Pb data in soils and air. The dry deposition velocities for moorland and woodland soils were 2.2 ± 1.8 and 9 ± 2 $\text{mm}\cdot\text{s}^{-1}$, respectively. The ^{210}Pb concentration in rain was calculated to be 94 ± 10 $\text{mBq}\cdot\text{L}^{-1}$. The large (~4 times) deposition velocities in woodland relative to moorland soils is an indication of the degree of accumulation of particles, and most possibly contaminants within woodland soils, which is of practical importance in the mitigation of pollutant concentrations in urban areas by planting trees.

Keywords: ^{210}Pb carrier aerosols, aerosol deposition velocity, ^{210}Pb concentration

Introduction

The study of deposition of aerosol-borne radioisotopes to the ground provides an important input parameter in the radio-ecological models [1, 2]. In the atmosphere, the fate of radionuclides are determined by two principal processes: precipitation scavenging (the process which involves incorporation of radioactivity into various types of precipitation and subsequent deposition onto the surface of the earth) and dry deposition. Removal of aerosol-borne radioactive substances from the atmosphere is poorly understood in that it depends on complicated microphysical and microchemical processes that are functions of both within and outside natural cloud-bearing layers [3]. Thus, it is usual to simulate these processes by means of a deposition velocity that relates the total deposition flux to the atmospheric concentration [4-6]. Particles from the atmosphere are either removed by hydrometeors or by dry deposition. The latter

process has been studied for 5 or more decades and the mechanisms that regulate transfer to and interception by terrestrial surfaces have been described [7] and reviewed [8]. The mass median diameter for particle ranges for typical aerosols are generally $0.6\text{-}0.8$ μm and is associated with deposition velocities of $0.5\text{-}3$ $\text{mm}\cdot\text{s}^{-1}$ for short vegetation. Field investigations by Fowler et al. [9], however, have revealed that woodlands 'collect' ambient aerosols at approximately three times the rate of grasslands. The capture of significant quantities of particulates by tree species has been recognized for its impact on air quality [10].

Inventories of ^{210}Pb in Edinburgh soils [11] in conjunction with surface level air concentrations [12] were used to calculate the deposition flux and deposition velocity of ^{210}Pb carrier aerosols. The naturally occurring ^{210}Pb ($t_{1/2}=22.3$ years) is a daughter of a radioactive water-soluble inert gas, ^{222}Rn ($t_{1/2}=3.8$ days), belonging to the ^{238}U decay series. Since ^{238}U and ^{226}Ra are ubiquitous in rocks and soils, ^{222}Rn is constantly being produced. Because ^{222}Rn is not chemically bound or attached to other materials, its atoms

*e-mail: alikuku@temo.bca.bw

can outgas from the ice-free continental crust of the earth and decay through a series of short-lived daughters to ^{210}Pb in the atmosphere. Once produced, the ^{210}Pb ions instantaneously (~ 30 s) and irreversibly attach to aerosol particles, and are distributed by the general circulation model [13, 14]. The activities of ^{210}Pb are expected to vary in time and location; influenced mainly by the ^{210}Pb source and meteorological conditions such as transport and deposition mechanisms. Because the mean residence time of ^{210}Pb with respect to the rate of removal in undisturbed soils is in the order of a century; about 5-half-lives of ^{210}Pb [15], the atmospherically derived ^{210}Pb inventories in soils may be used to determine the deposition rate of the radionuclide, provided that the flux has been constant over that period. The widely used method [15-19] of determining the atmospherically derived ^{210}Pb is given by:

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

...where Φ is the total flux (in $\text{Bq}\cdot\text{m}^{-2}\cdot\text{y}^{-1}$) 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 $\text{Bq}\cdot\text{m}^{-2}$. Equation 1, however, is used based on the assumption that the soils have not been disturbed for at least a decade and that the ^{210}Pb inventory is in a steady state, with the rate of ^{210}Pb atoms equaling the mean deposition rate.

This paper reports deposition velocities and concentration in rain of aerosol-borne substances and concentration in rain calculated from ^{210}Pb measurements in soils and surface level air of Edinburgh (55.9°N , 03.2°W). The measurements were carried out for the following reasons:

- to calculate the total deposition flux of ^{210}Pb carrier aerosols,
- to determine the total and dry deposition velocities of ^{210}Pb aerosols, and
- to estimate the ^{210}Pb concentrations in precipitation.

The results presented here may serve as additional and desirable data for the UK coastal sampling sites, which is of importance in a number of tracer studies pertaining earth sciences.

Materials and Methods

Air Sample Collection and Measurement

Air sample data used in this paper, sample collection procedure, measurement and analysis is published elsewhere by Likuku [12].

Soil Sample Collection and Measurement

Soils were collected from five different sites, each with relatively insignificant undulations. Selection of sites was according to:

- the sites are situated at relatively low altitudes (<300 m asl.) with no complex topography between them;
- the mean rainfall at the sites is similar ($\sim 830 \text{ mm}\cdot\text{y}^{-1}$); and

- they are confined to a small region to eliminate the effect of regional changes in the ambient ^{210}Pb concentrations.

The soils were collected from moorland and beneath woodland canopies. The sites were selected on a reasonable assumption that the soils have not been subjected to erosion or other forms of disturbance in recent decades. Samples were counted for activity using a Canberra HPGe (Be-window) detector fitted with an annular NaI(Tl) anti-Compton shield detector. Detailed description of the sampling sites and methods, sample preparations, detector calibration, and analytical procedures are given by Likuku et al. [11].

Results and Discussions

Inventories of ^{210}Pb in both moorland and woodland canopy soils from the five different Edinburgh sites (site I-V) were used to calculate the total (wet and dry) long-term atmospheric deposition fluxes. Their total and dry deposition velocities were also calculated using data given in Table 1, and the results are given in Table 2.

The total ^{210}Pb deposition fluxes in moorland soils varied from $72\text{-}92 \text{ Bq}\cdot\text{m}^{-2}\cdot\text{y}^{-1}$, with a mean value of $78\pm 8 \text{ Bq}\cdot\text{m}^{-2}\cdot\text{y}^{-1}$. The long-term ^{210}Pb deposition flux is comparable with

Table 1. Atmospheric deposition fluxes of ^{210}Pb aerosols at the five Edinburgh sites, for both moorland and woodland canopy soils.

Φ_T ($\text{Bq}\cdot\text{m}^{-2}\cdot\text{y}^{-1}$) Moorland				
Site I	Site II	Site III	Site IV	Site V
114.6	86.5	80.1	82.8	72.1
82.8	70.7	87.9	85.2	94.4
86.0	74.8	69.1	78.3	64.1
96.2	59.5	69.4	73.6	61.6
71.2	–	–	–	59.2
97.5	–	–	–	114.1
98.2	–	–	–	62.2
89.1	–	–	–	38.8
94.9	–	–	–	–
Φ_T ($\text{Bq}\cdot\text{m}^{-2}\cdot\text{y}^{-1}$) Woodland				
Site I	Site II	Site III	Site IV	Site V
–	126.7	100.4	113.0	–
–	108.2	105.4	126.3	–
–	124.7	108.9	117.5	–
–	93.6	101.8	144.2	–
–	–	87.0	120.5	–

Φ_T – total deposition flux. The dashes (–) indicate that no data were available for the sites

Table 2. Atmospheric deposition fluxes and deposition velocities of ^{210}Pb aerosols at the five Edinburgh sites, for both moorland and woodland canopy soils.

Moorland	Site I	Site II	Site III	Site IV	Site V	Average
Φ_T ($\text{Bq}\cdot\text{m}^{-2}\cdot\text{y}^{-1}$)	92 (n = 9)	73 (n = 4)	77 (n = 4)	80 (n = 4)	71 (n = 8)	78±8
Φ_D ($\text{Bq}\cdot\text{m}^{-2}\cdot\text{y}^{-1}$)	24	5	9	12	3	11±8
V_d ($\text{mm}\cdot\text{s}^{-1}$)	19	15	16	17	15	16±2
V_{dd} ($\text{mm}\cdot\text{s}^{-1}$)	5.1	1.1	1.9	2.5	0.6	2.2±1.8
Woodland	Site I	Site II	Site III	Site IV	Site V	Average
Φ_T ($\text{Bq}\cdot\text{m}^{-2}\cdot\text{y}^{-1}$)	–	113 (n = 4)	101 (n = 5)	124 (n = 5)	–	113±12
Φ_D ($\text{Bq}\cdot\text{m}^{-2}\cdot\text{y}^{-1}$)	–	45	33	56	–	45±12
V_d ($\text{mm}\cdot\text{s}^{-1}$)	–	24	21	26	–	24±3
V_{dd} ($\text{mm}\cdot\text{s}^{-1}$)	–	9.5	7.0	11.8	–	9±2

Φ_T – total deposition flux, Φ_D – dry deposition flux, V_d – total deposition velocity, V_{dd} – dry deposition velocity. The dashes (–) indicate that no woodland data were available for the sites.

The letter n denotes the number of samples analyzed.

deposition fluxes reported by several investigators in other UK sites. For example, $86 \text{ Bq}\cdot\text{m}^{-2}\cdot\text{y}^{-1}$ for Fenwick, Scotland [20], $>116 \text{ Bq}\cdot\text{m}^{-2}\cdot\text{y}^{-1}$ for Inverness, Scotland [21], and $95 \text{ Bq}\cdot\text{m}^{-2}\cdot\text{y}^{-1}$ for Jackmoor Brook, Devon, England [22], where both altitude and mean annual rainfall are quite similar.

The mean deposition flux for woodland canopy soils was $113\pm 12 \text{ Bq}\cdot\text{m}^{-2}\cdot\text{y}^{-1}$. This figure is significantly large ($P<0.05$) with a factor of ~ 1.45 over that of the mean deposition flux for moorland soils. The 45% enhancement is due, in part, to the turbulence of air above and within the forest canopy, promoting deposition by impaction. Furthermore, because forests possess structures that promote interception of atmospheric pollutants, the particles intercepted are retained by foliar surfaces and subsequently fall beneath woodland canopies. The mechanisms may either be through wash-off, gravitational settling, and as senescent leaves, contributing to enhanced particle matter in canopy soils. Mechanisms governing deposition of particles onto forests and their efficiencies at which these particles are retained by foliar surfaces have been examined [23, 24] and effects of forest vegetation on the deposition of radioactive material have been successfully demonstrated [9, 25-27]. All these authors report a similar degree of enhancement in the deposition of radioactive material in soils under forest canopies relative to adjacent open grassland soils.

Deposition rates are usually described by the deposition velocities V_d and V_{dd} (in $\text{mm}\cdot\text{s}^{-1}$) for total (wet and dry) and dry deposition velocities, respectively, as follows [4-6]:

$$V_d = \Phi / C_{\text{air}} \quad (2)$$

...where C_{air} is the surface level concentration of a radionuclide at the sampling site.

The ^{210}Pb total (wet and dry) deposition velocities were calculated using both moorland and woodland soil and ^{210}Pb

air concentration data reported by Likuku [12]. To separate the total and dry deposition velocities, the wet deposition flux of $68\pm 10 \text{ Bq}\cdot\text{m}^{-2}\cdot\text{y}^{-1}$ for Edinburgh reported by Fowler et al. [9] (and references therein) was used. This was based on the assumption that since both sites were in close proximity and are of similar rainfall and altitude, with no complex topography in between, both woodland and moorland soils receive the same ^{210}Pb concentrations. The ^{210}Pb deposition flux in precipitation allows calculation of dry deposition velocities (V_{dd}) of aerosols to moorland and woodland surfaces. The dry deposition velocity varied widely from $1.1\text{-}5.1 \text{ mm}\cdot\text{s}^{-1}$ (mean= $2.2\pm 1.8 \text{ mm}\cdot\text{s}^{-1}$) and the mean V_{dd} for woodland was $9\pm 2 \text{ mm}\cdot\text{s}^{-1}$. The moorland deposition velocity is $\sim 50\%$ lower than the mean ^{210}Pb deposition velocity of $3.3 \text{ mm}\cdot\text{s}^{-1}$ reported by Fowler et al. [9]. The difference, however, may be accounted for by possible differences in site characteristics. The mean deposition velocity for woodland is very large when compared to the $<0.5 \text{ mm}\cdot\text{s}^{-1}$ suggested by the Slinn [28] model. These results suggest that woodland captures aerosols 4 times the rate of moorland surfaces. The measurements reported here, however, have advantages over model results in that they average a wide range of meteorological conditions and a range of different site characteristics such as canopy height and structure.

The mean total deposition velocities (V_d) for moorland and woodland soils were $16\pm 2 \text{ mm}\cdot\text{s}^{-1}$ and $24\pm 3 \text{ mm}\cdot\text{s}^{-1}$, respectively. For the sake of comparison, total deposition velocities using moorland soils or bulk precipitation data reported by previous researchers are listed in Table 3. It should be noted, however, that to effectively make comparisons of deposition velocities, one must take into account the different field conditions under which these experiments were conducted. For example, deposition via atmospheric transport processes is non-uniform depending on meteorological conditions and surface roughness effect. In the absence of other data, it was difficult to compare total deposition velocities for aerosols on woodland soils.

Table 3. ^{210}Pb total deposition velocities, V_d reported by various researchers.

Location	Coordinates	V_d ($\text{mm}\cdot\text{s}^{-1}$)	References
New Haven, Connecticut (USA)	41°N, 73°W	9.5	[4]
Bermuda	33°N, 64°W	10	[4]
Eastern and Midwestern USA	33-44°N, 72-105°W	6.0	[19]
Norfolk, Virginia (USA)	33°N, 84°W	7.0	[5]
Western North Atlantic	31-42°N, 60-74°W	19	[29]
Munich (Germany)	48°N, 11°E	10	[30]
Detroit, Michigan (USA)	42°N, 83°W	16	[31]
Málaga (Spain)	36°N, 04°W	15	[3]
Edinburgh (UK)	56°N, 03°W	16±2	Present Study

Data presented in Table 3 shows that the ^{210}Pb deposition velocity calculated from this study compares with a few other deposition velocities, but is either lower or higher than the other reported values. A simple, but plausible, explanation could be that, in most cases, deposition via atmospheric transport processes is not uniform. Thus, a large sample size of ^{210}Pb concentration in air and/or precipitation is required to effectively characterize the average deposition over an area. Furthermore, for the purpose of comparisons, various factors such as sampling techniques must be taken into consideration [3].

It is advantageous to determine total aerosol deposition velocities using ^{210}Pb because: ^{210}Pb concentrations in precipitation and air can be easily measured, the ^{210}Pb production rate at any given time is constant over long periods of time [32], and the size distribution of ^{210}Pb in aerosols are quite similar to the particulate contaminants of interest. Therefore, it can be used to determine the fluxes of these contaminants to the earth's surface, from the knowledge of the ^{210}Pb deposition velocities and the concentration of these contaminants in air [31].

The average ^{210}Pb concentration in rain, C_{rain} (in $\text{mBq}\cdot\text{L}^{-1}$), can be deduced from the long-term ^{210}Pb deposition flux given in Table 2 and the average rainfall data, R (in $\text{mm}\cdot\text{y}^{-1}$) using the equation:

$$\Phi = C_{\text{rain}} \times R \quad (3)$$

The ^{210}Pb concentration obtained in this way, however, assumes that all the ^{210}Pb atoms reaching the earth's surface are delivered by precipitation alone. Thus, the ^{210}Pb concentration value obtained may be an overestimate considering that a proportion of the ^{210}Pb atoms will be delivered by the process of dry deposition.

Using equation 3 and an average rainfall value of $830 \text{ mm}\cdot\text{y}^{-1}$ for Edinburgh, as given on the Meteorological Office Annual Rainfall Map (1977), the ^{210}Pb concentration in rain was found to be $94\pm 10 \text{ mBq}\cdot\text{L}^{-1}$. This value compares well with the reported 80 and 97 $\text{mBq}\cdot\text{L}^{-1}$ UK direct

measurements of ^{210}Pb concentrations in rain made during 1962 and 1963, respectively, at Milton Haven [33], and is very close to the calculated value of $77\pm 14 \text{ mBq}\cdot\text{L}^{-1}$ for the UK as stated by Smith et al. [34].

Conclusions

The ^{210}Pb deposition flux and deposition velocity have been calculated using soil inventory and previously published air concentration data for Edinburgh, UK [12], and the following are the main results:

- The average long-term atmospheric deposition flux of ^{210}Pb in moorland soils was $78\pm 8 \text{ Bq}\cdot\text{m}^{-2}\cdot\text{y}^{-1}$. This value compares well with most ^{210}Pb fluxes in soils reported by several independent investigators. The mean flux in woodland soils ($113\pm 12 \text{ Bq}\cdot\text{m}^{-2}\cdot\text{y}^{-1}$) indicates that forests contribute to significantly ($P<0.05$) higher deposition of air-borne particulates as compared to moorland surfaces.
- The mean deposition velocity for moorland surfaces was $2.2\pm 1.8 \text{ mm}\cdot\text{s}^{-1}$. This value is $\sim 50\%$ lower than the mean value for the west Midlands conurbation reported by Fowler et al. [9]. However, these values may reflect different characteristics of the sites in terms of aerosol composition and surface roughness, and are thus site-specific.
- The estimated ^{210}Pb concentration in precipitation was $94\pm 10 \text{ mBq}\cdot\text{L}^{-1}$. This value is close to both derived and direct ^{210}Pb concentration measurements reported in literature.

The influence of woodland in the deposition of atmospheric aerosols observed from this study may be of practical importance in the current development regarding the beneficial effects of trees in urban air quality because although the application of urban woodland may improve the local air quality by increasing the uptake rates of gaseous, particulate, and aerosol pollutants from the atmosphere, the application must be considered together with the long-term effects of the deposited pollutants that accumu-

late in the woodland soils. These results are also valuable as input parameters in modeling of pollutant deposition maps in relation to land cover.

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