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Cosmogenic ^{7}Be deposition in North Wales: ^{7}Be concentrations in sheep faeces in relation to altitude and precipitation

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Abstract

Beryllium-7 (^{7}Be) is a cosmogenic radionuclide with a half-life of 53.3 days produced mostly in the stratosphere by cosmic ray spallation of nitrogen and oxygen and entering the lower troposphere by atmospheric circulation processes. Most of the nuclide is removed by rainout during precipitation, so given that rainfall generally increases with altitude, it was considered probable that ^{7}Be deposition would also be greater at higher altitudes. The aim of this study was to determine if there is any relationship between ^{7}Be concentrations, altitude and precipitation by measuring ^{7}Be activity within sheep faeces. An area of North Wales and Northwest England was selected for study, extending from the Dee Estuary at sea level to Snowdon at 1065 m. The results obtained showed a significant increase in ^{7}Be activity with increasing altitude and precipitation consistent with predictions based on the existing literature.

Keywords: Beryllium-7; Deposition; Precipitation; Altitude; North Wales; Sheep faeces

1. Introduction

Beryllium-7 (^{7}Be) is a cosmogenic radionuclide with a half-life of 53.3 days, produced in the upper atmosphere by cosmic ray bombardment and transmutation (spallation) of nitrogen and oxygen (Arnold and Al-Salih, 1955; Peters, 1955, 1959).

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About 75% of ^{7}Be is produced in the stratosphere and 25% produced in the upper troposphere (Johnson and Viezee, 1981). ^{7}Be is removed from the troposphere by radioactive decay and wet and dry deposition. Dry deposition accounts for only 3–8% of the total fallout, the remainder being accounted for by rainout (raindrop formation around ^{7}Be carrier particles) and washout (^{7}Be carriers gathered by falling rain) (Murray et al., 1992; Ishikawa et al., 1995; Wallbrink and Murray, 1994). The mean tropospheric residence time of ^{7}Be has been estimated at between 22 and 48 days (Bleichrodt, 1978; Durana et al., 1996). Given the high percentage of wet deposition of Be-7 it would therefore seem probable that ^{7}Be concentrations on the ground will be greater in areas of high rainfall. Since precipitation is generally greater in upland regions, an increase in ^{7}Be with altitude is to be expected. Beryllium isotopes are readily absorbed by plants that are, in turn, eaten by sheep and it is expected that concentrations of ^{7}Be in the faeces of sheep grazing in upland areas will be higher than in the faeces of lowland sheep. Since sheep graze over a relatively limited home area in relation to digestive transit time for grass (Milne et al., 1976) they effectively integrate small-scale spatial heterogeneity in ^{7}Be deposition for a given altitude.

Most ^{7}Be occurs in the environment mainly as the highly soluble Be^{2+} ion (Knies et al., 1994). This species accounts for about 70% of all the hydrolysed beryllium species, which explains its significant foliar interception by plants (Bettoli et al., 1995). Foliar absorption, both by wet and dry deposition, has been acknowledged as the dominant mechanism of plant uptake of atmospherically derived isotopes with relatively short half-lives (Bettoli et al., 1995). This depends mainly on the presence of anionic organic groups in the plant structure (Bettoli et al., 1995). Papastefanou et al. (1999) showed that ^{7}Be was not taken up by the roots of plants because of its short half-life and slow transport time through the soil profile, however, ^{7}Be and ^{10}Be do accumulate in the structural tissue of plants (Bettoli et al., 1995). Foliar absorption is the dominant uptake mechanism by grasses since ^{7}Be is confined to the upper 20 mm of the soil profile, independently of season and precipitation (Wallbrink and Murray, 1996; Bettoli et al., 1995). The ^{7}Be isotope is strongly bound to the internal structure of the leaves, so activity depletion simply derives from radioactive decay and it is not released by leaching or evaporation (Bettoli et al., 1995; Papastefanou et al., 1999). As a result, grazing animals ingest ^{7}Be with their food although it has been shown that animals reject beryllium in the food supply and therefore excrete most of the beryllium consumed. Measurements of the bioaccumulation factor for ^{7}Be for animals relative to plants tend to be of the order of 0.06 (International Joint Commission, 1997). Although high faecal concentrations in themselves do not suggest low absorption (since elements may be excreted from the circulatory system to the gastrointestinal tract) this low bioaccumulation factor probably explains the relatively high concentrations of ^{7}Be in sheep faeces.

The estimated global inventory of ^{7}Be ranges from 20 g (Eisenbud and Gesell, 1997) to 3.2 kg (International Joint Commission, 1997). Seasonal variations in atmospheric deposition do occur, mostly as a result of washout caused by increased precipitation, at the study site latitudes, in the winter season (Herbert et al., 1989). Despite existing in such minute quantities, it can be detected in sheep faeces and in grass by gamma ray spectrometry. Previous measurements concerning ^{7}Be and altitude have used either aerosol sampling from aircraft in the stratosphere or troposphere (for example, Young and Silker, 1980). Other measurements include the ^{7}Be activity of individual rainfall events (Knies et al., 1994; Caillet et al., 2001), snowfall events (Ishikawa et al., 1995) and surface deposition in the United States (Koch and Mann, 1996). Most of these studies indicate that ^{7}Be deposition is dependent on five factors: latitude, season, altitude, precipitation and solar activity.

The object of this study was to investigate the relationship between ^{7}Be concentrations in ovine faecal samples, altitude and precipitation by collecting sheep faeces from various locations and altitudes and correlating with meteorological data. Previous studies in sheep have mainly investigated post-Chernobyl caesium (^{134}Cs and ^{137}Cs) depositions in Snowdonia, the English Lake District and continental Northern Europe (Bonnett et al., 1989). A convenient proxy method for measuring radio-caesium concentration in sheep meat involved the testing of sheep faeces for radionuclides (McGee et al., 1994). A similar method to measure ^{7}Be concentration by determining its 447 keV gamma peak was used in this study.

2. Methods

Samples of sheep faeces were collected from a variety of locations, given here in UK Ordnance Survey format. One location in Cheshire, N.W. England: Burton (SJ 315745) at sea level on the Dee Estuary and five in North Wales: Halkyn Mountain (SJ 215705, 250 m, a.m.s.l); Moel-y-Crio (SJ 193698, 263 m, a.m.s.l); Moel Findeg (SJ 207613, 370 m, a.m.s.l); Moel Llys-y-Coed (SJ 154655, 465 m, a.m.s.l) and Snowdon (SH 609543, 1065 m, a.m.s.l). The locations were chosen for their differing altitudes, proximity to a Meteorological Office weather monitoring station and regional geographical separation. However, Halkyn and Moel-y-Crio, were selected as a control group since as well as their proximity to a weather station, their different locations (about 2 km apart) on the same upland region with similar rainfall patterns where expected to give similar ^{7}Be concentrations. The weather stations are at the following respective locations: Ness Gardens (SJ 303754, 38 m, a.m.s.l); Moel-y-Crio (SJ 193698, 263 m, a.m.s.l); Loggerheads, Colomendy Centre (SJ 200622, 215 m, a.m.s.l); Nannerch (SJ 147684, 210 m, a.m.s.l) and Snowdon Summit (SH 609543, 1065 m, a.m.s.l). Daily rainfall data obtained from the Metrological Office for each weather station were processed to give monthly averages. This was chosen because of the 53.3 day half-life of ^{7}Be and to provide a convenient time period for comparison of precipitation throughout the year.

Sampling of sheep faeces was carried out over the winter months, between December 2000 and February 2002. Only fresh faecal samples were collected because the concentration of ^{7}Be would diminish fairly rapidly in older samples due to the relatively short half-life of ^{7}Be. A fresh sample was deemed to be one with mucus present on the sample surface giving it a wet appearance. Care was taken to ensure that no supplementary sheep feeds were in use in the study areas, since these would clearly affect the ^{7}Be concentrations. The grass from one location, Moel-y-Crio, was also subjected to analysis for ^{7}Be activity to enable a coarse comparison between grass and faecal ^{7}Be concentrations to be made. Whenever possible, samples were taken only a day or two before analysis. If this was not possible and storage of the sample was necessary, then the ^{7}Be activities determined in sheep faeces were decay corrected to the day of sampling.

Samples were dried at 110 °C to constant weight to ensure complete removal of water and ground to <1 mm particle size prior to analysis. A gamma ray spectrum (>24 h runtime, detector efficiency: 0.025 at 477 keV) for each sample was obtained and analysed for ^{7}Be activity. The detector was a 2-inch NaI detector with integral photomultiplier tube (Model S88-I/75, EG and G Instruments) housed in 50.8 mm aged lead shielding, connected to an Ace Mate amplifier (NS 925 SCINT, EG and G Instruments). The multi-channel analyser was a 2K Ace card with Maestro II emulation software. The apparatus was calibrated using cobalt-60 and caesium-137 standards of known energies, 1332 keV and 662 keV, respectively, and the efficiency of the detector at various energies determined by calibration with caesium-137 and potassium-40 of certified energies. Few other significant gamma peaks were observed in the 447 keV regions and so the risk of interference was estimated as insignificant. In addition, previous measurements on this system allowing samples to decay and calculating the half-life of the measured ^{7}Be gave readings in good agreement with the accepted half-life of this element giving confidence in these results.

The results were analysed using Pearson's Product Moment Correlation to explore relationships between rainfall, altitude and ^{7}Be concentrations.

3. Results

There is a clear trend of increasing activity with altitude, with an obvious anomaly at Moel Llys-y-Coed (Fig. 1). The activity of the samples is lowest at the Dee Estuary and greatest on Snowdon. This trend is also clear in relation to rainfall (Fig. 2). The strong correlation ($r = +0.96$, $p < 0.01$) between altitude and ^{7}Be activity suggests that ^{7}Be deposition is greater at increased altitude (Fig. 4). However, it must be remembered that the rainfall patterns for the area under study are largely altitude dependent and there is a slightly stronger correlation between rainfall and ^{7}Be activity (Fig. 3, $r = +0.98$, $p < 0.01$). The apparent anomaly at Moel Llys-y-Coed (Figs. 1 and 3) may be explained by the fact that the rainfall data were not collected at the exact same site, the weather station being at a slightly lower altitude and 2 km distant.

The grass samples from Moel-y-Crio were found to have a mean ^{7}Be activity of 87.7 Bq kg^{-1}, which is of a similar order to that in found by Papastefanou et al. (1999), in Greece, of between 2.1 Bq kg^{-1} and 348.0 Bq kg^{-1} depending on season and precipitation, with a mean of 54.4 Bq kg^{-1}.

The mean activity of sheep faeces from the same location at the same time was 205.8 Bq kg^{-1}. In this case, the bio-concentration factor from grass to sheep faeces is 2.35, but further investigation from other sites and seasons would be needed to

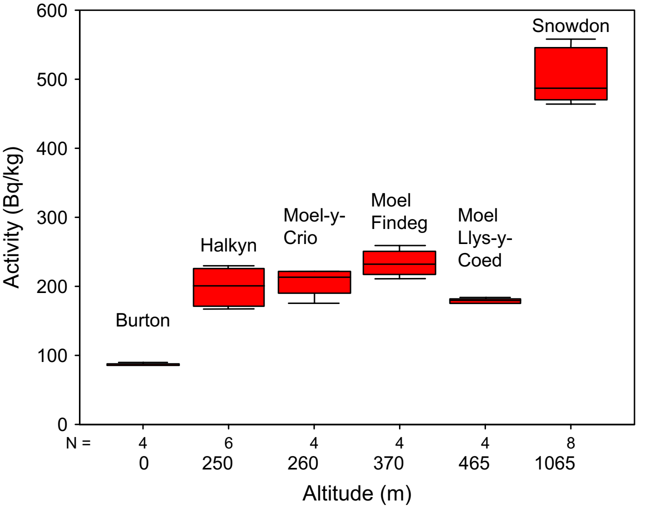

Fig. 1. Box plot showing the spread around the mean ^{7}Be activity and altitude (the box represents the interquartile range, the dividing line the mean, and the whiskers are the largest values within 1.5 interquartile ranges).

validate this figure. In any case, given the low bioaccumulation of ^{7}Be into animal tissue, the factor is a reflection of the digestibility of dry matter.

4. Discussion

This study has shown a significant positive correlation between ^{7}Be deposition and increasing altitude. However, this variation in activity would appear to be related to increased precipitation rather than altitude per se, the two factors being closely related in the area under study (Meteorological Office, 2001). The ^{7}Be activity increase with altitude may also be enhanced by the effect of greater snowfall at higher altitudes, about 5 cm of snowfall was observed only on Snowdon during the period of this study. This follows from the work carried out in Japan by Ishikawa et al. (1995), which demonstrated that ^{7}Be-bearing aerosols are scavenged more effectively from the atmosphere through rainout processes due to snowfall than those due to rainfall. If this is the case, ^{7}Be deposition will be greater in areas of higher snowfall, as suggested by the activity data collected on Snowdon. At this location it can be seen that the mean winter activity is more than 500 Bq kg^{-1}, which is more than twice that noted at any other location (Fig. 2). However, it was not possible from the meteorological data to discriminate between rainfall and snowfall events for this study. There is scope for further work here although the problem remains that sheep may move to lower altitudes following snowfall. Studies of this type confined to the summer months, where there is virtually no snowfall in this region, will help resolve this issue.

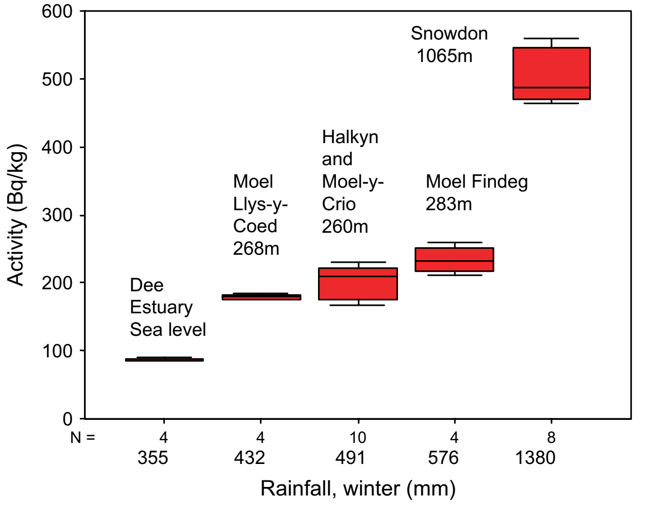

Fig. 2. Box plot showing the spread around the mean between ^{7}Be activity and winter rainfall (the box represents the interquartile range, the dividing line the mean, and the whiskers are the largest values within 1.5 interquartile ranges). (Halkyn and Moel-y-Crio data are combined as these locations have the same rainfall data).

Faecal samples collected from the Dee Estuary at Burton were found to have greater dry mass than samples collected elsewhere. This was due to the presence of silt and sand that had previously been ingested by the sheep, confirmed by microscopic examination of the dried samples. This increased mass due to the inorganic content of the faeces will have necessarily depressed the apparent ^{7}Be

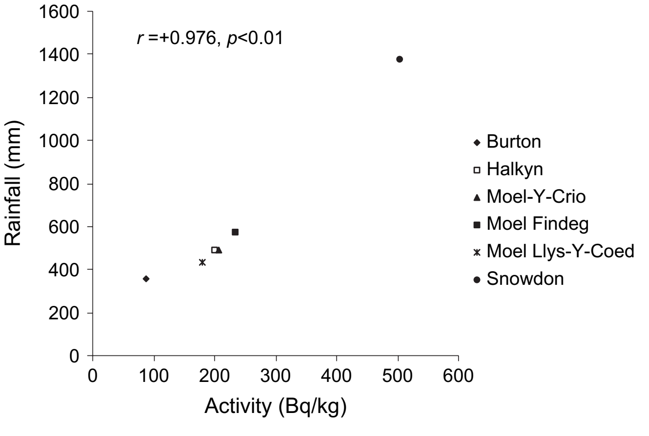

Fig. 3. Scatter plot showing the relationship between beryllium-7 activity and location (winter rainfall). Note the similar activity at Halkyn and Moel-y-Crio due to the similar rainfall data ($r = +0.976$, $p < 0.01$).

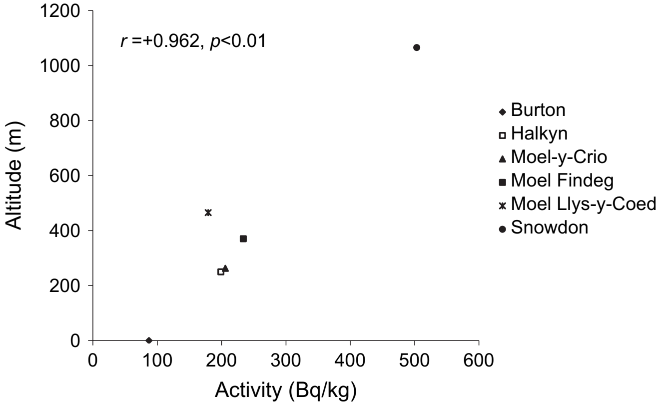

Fig. 4. Scatter plot showing the relationship between beryllium-7 activity and location (altitude). Note the anomaly for Moel Llys-y-Coed. ($r = +0.962$, $p < 0.01$).

activity levels found at this site. Although the activity at this site was the lowest as would be expected from the low rainfall, this result must be treated with caution.

Most workers agree that there are strong correlations between precipitation and ^{7}Be deposition, which has been found to be the case in this study. However, some workers, for example Brown et al. (1989) and Koch and Mann (1996) have found either zero or negative correlation with precipitation. However, Bettoli et al. (1995) have recognised the seasonal variation in ^{7}Be deposition being related to the seasonal pattern of rainfall frequency. Nevertheless, they have found no linear correlation between ^{7}Be deposition and precipitation, as expected and estimated by some workers, for example Knies et al. (1994). These discrepancies would appear to result from different sampling regimes. Those finding positive correlation with rainfall have taken samples over long periods, for example monthly rainfall totals, whereas those studies that found negative or zero correlation involved individual rainfall events (Brown et al., 1989). These discrepancies would appear to be due to the timing of the collection of single event samples, and are likely to be greater if the sample is taken before or after a storm event, during which full washout occurs (Caillet et al., 2001). It has been calculated that tropospheric reload of ^{7}Be takes approximately two days following a major rainfall event (Caillet et al., 2001). Rainout algorithms describing the washout process (scavenging) of ^{7}Be with time have been calculated (Sakashita et al., 1999). These may prove useful in future investigations, particularly concerning ^{7}Be deposition during single rainfall events. Given these short-term fluctuations the advantages of using faecal samples (which effectively buffer against rapid fluctuations and integrate concentrations over a large grazing area) are obvious.

Further work is clearly required to determine the absolute relationship between ^{7}Be deposition and altitude/precipitation, over long term and short-term periods. The addition of further sampling sites at intermediate altitudes and locations between Snowdon and Moel Llys-y-Coed would be advantageous, since the geographical remoteness of Snowdon from the other locations may have introduced other variables.

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