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Temporal variability of beryllium-7 fallout in southwest UK

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ABSTRACT

Cosmogenic beryllium-7 has been widely employed as a sediment tracing tool and continued development of its use as a soil erosion tracer requires knowledge of fallout temporal dynamics. Data regarding beryllium-7 fallout in the UK are scarce and here the authors provide a record of beryllium-7 fallout in southwest England spanning a two-year period. A monthly fallout record was developed for Plymouth, UK using regular rainfall sampling to determine beryllium-7 rainfall activity concentration (Bq L^{-1}) and deposition flux (Bq m^{-2}). Data showed a general tendency for higher activity during the spring/summer months and lower activity in the autumn/winter months. Comparison with data for other UK sites (Chilton and Aberporth) for the same period found significant differences in ^{7}Be activity in rainwater and lower variability in Plymouth than Chilton and Aberporth. Total deposition was largely controlled by rainfall in Plymouth although regression coefficients suggested greater importance of other atmospheric controls at the Chilton and Aberporth sites. Use of a deposition proportion to rainfall proportion ratio identified periods when deposition was influenced by varying ^{7}Be activity in rainfall. Broad ranges in ratios were found for Chilton and Aberporth and this has implications for sediment tracer studies requiring estimates of ^{7}Be deposition flux across months or seasons.

1. Introduction

Beryllium-7 (^{7}Be) ($t_{1/2}$ 53.3 days) is a cosmogenic radionuclide produced in the upper atmosphere by cosmic ray spallation of nitrogen and oxygen. Upon fallout, its affinity with sediment (Kaste et al., 2002) has led to its widespread use as a sediment tracer in terrestrial and fluvial-marine environments (Taylor et al., 2013; Walling, 2012). With continued development of ^{7}Be application as a tracing tool it is necessary to improve our knowledge regarding the temporal variability of fallout (Walling et al., 2009). A number of factors affect fallout temporal variability by influencing ^{7}Be availability in surface air. For example, rates of ^{7}Be production are dependent upon cosmic ray activity and can, therefore, be influenced by solar cycles with ^{7}Be concentrations in surface air demonstrating a negative correlation with sunspot number (Ioannidou et al., 2005). On a seasonal basis, concentrations of ^{7}Be in surface air are influenced by mixing of stratospheric air with the upper troposphere (stratosphere-troposphere exchange (STE)). Higher rates of production in the stratosphere and longer residence time of ^{7}Be-bearing aerosols creates a concentration gradient relative to the troposphere (Doering and Akber, 2008a). This concentration gradient can be reduced during folding of the tropopause, which encourages mixing of the troposphere with stratospheric air and this commonly occurs during spring at mid-latitudes (Feely et al., 1989). Convective circulation during warmer months can also influence seasonal variation in ^{7}Be concentrations in surface air by driving the downward transport of ^{7}Be-enriched air from the upper troposphere (Doering and Akber, 2008a; Feely et al., 1989; Ioannidou et al., 2005). Because ^{7}Be-bearing aerosols are readily scavenged by precipitation (Ioannidou and Papastefanou, 2006), seasonal patterns in rainfall can also affect surface air concentrations by removing available ^{7}Be (washout) (Doering and Akber, 2008a; Feely et al., 1989). Wet deposition is the dominant pathway of ^{7}Be flux to the Earth's surface with dry deposition accounting for <10% (Ioannidou and Papastefanou, 2006; Wallbrink and Murray, 1994). ^{7}Be deposition (Bq m^{-2}) is, therefore, well correlated with rainfall (mm) (Ayub et al., 2009; Caillet et al., 2001; Doering and Akber, 2008b; Gonzalez-Gomez et al., 2006; Mabit et al., 2014) although some variation in depositional flux can be attributed to changes in ^{7}Be activity in rainwater (Bq L^{-1}). This is influenced by atmospheric

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processes such as washout and circulation of enriched air (Baskaran, 1995; Caillet et al., 2001; Gonzalez-Gomez et al., 2006). There is also some evidence to suggest that ^{7}Be activity in rainwater can be influenced by rainfall intensity with more effective scavenging of aerosols by fine rainfall droplets (Ioannidou and Papastefanou, 2006).

The availability of ^{7}Be in surface air and its subsequent fallout is, thus, influenced by a complex combination of factors, which are likely to contribute to temporal variability of ^{7}Be activity in rainwater and deposition. In the UK, data regarding the temporal variability of ^{7}Be fallout are scarce and, to the best of the authors' knowledge, no published, peer-reviewed data exist for southwest England. This contribution provides a record of monthly ^{7}Be fallout for Plymouth, southwest England (average annual rainfall 1007 mm (1981–2010) (Met Office)) spanning a two-year period (2009–2011). Potential controls upon temporal variability of ^{7}Be rainfall activity and deposition are discussed and, in addition, data are compared to secondary available records obtained for other areas of the UK during the same study period.

2. Method

A total of 30 rainfall samples were collected from a flat roof area located ~50 m a.m.s.l (~15 m above ground level) on the Plymouth University campus (50° 22′ 55″ N, 04° 08′ 34″ W), over a 25 month period from February 2009 to March 2011. A tipping-bucket rain gauge with 0.2 mm resolution and 10 min logging intervals was located adjacent to the sample site. Total (wet and dry) fallout was sampled in plastic containers (3 L) using 0.05 m^{2} area funnels. Prior to deployment, 10 mL HCl (2.5 M) was added to each container to prevent adsorption of ^{7}Be to vessel walls during the sample period. Containers were exposed for periods of between 3 and 30 days depending upon the frequency of rainfall events. At the point of sampling funnels were rinsed with a known volume of HCl (3%) and the containers replaced with acid-cleaned vessels (soaked in 10% HCl for a minimum of 48 h).

Each sample was pH checked to ensure < pH 2 and then filtered to remove any coarse debris (Whatman grade number 41 filter papers). ^{7}Be was then preconcentrated from solution by coprecipitation with MnO_2 following the method detailed by Short et al. (2007). For this method, 1 mL of 0.2 M $KMnO_4$ was added per litre of rainwater sample and then pH adjusted to 8–10 using concentrated NH_4OH. Once at the desired pH, 1 mL of 0.3 M $MnCl_2$ was added to the sample whilst stirring. MnO_2 precipitate was then allowed to settle for 24 h prior to removal by vacuum filtration using 0.45 μm cellulose nitrate filter paper. The filter paper was then air-dried, fixed with cellophane and sealed in a 50 mm Petri dish prior to analysis by gamma spectrometry.

All ^{7}Be analyses were carried out by gamma-ray spectrometry using high purity germanium detectors (HPGe) (Ortec) in the Plymouth University Consolidated Radioisotope Facility (CoRiF) (which operates within ISO 9001:2008 certification). Efficiency calibration was carried out using a filter source of the same geometry as the extract samples (0.45 μm cellulose nitrate), prepared from a QCY58b multi-nuclide standard solution (GE Healthcare Life Sciences, Amersham, UK). GammaVision-32 software was used for the gamma spectra evaluation. All ^{7}Be activity values were reported in Bq L^{-1} (±standard uncertainty, 2σ confidence level). Samples were decay corrected in accordance with the rainfall period and where more than one sample was taken in any particular month, a mean value was reported. Laboratory analytical quality control procedures were carried out following Wallbrink et al. (2002).

Reproducibility of the method was tested by separate analyses of subsamples from one batch and relative standard deviation (RSD) was found to be 10%. ^{7}Be recovery from solution using the coprecipitation method was tested by reprecipitating the filtrate from 3 samples. In each case the ^{7}Be activity in the filtrate was below Minimum Detectable Activity (MDA). MDA values for these samples were below 10% of the total activity, suggesting that ^{7}Be recovery was greater than 90%, in agreement with Short et al. (2007).

^{7}Be deposition (or inventory) (Bq m^{-2}) was estimated by multiplying the decay corrected rainfall activity (Bq L^{-1}) with rainfall volume (L m^{-2}) for the sample period.

Monthly ^{7}Be rainfall activities for 2009 and 2010 have been reported for Chilton, England (51° 34′ 02″ N, 1° 17′ 43″ W; 114 m a.m.s.l.; average annual rainfall c. 612 mm (1981–2010) (Met Office)) and Aberporth, Wales (52° 07′ 49″ N, 4° 32′ 22″ W; 55 m a.m.s.l.; average annual rainfall (1981–2010) 888 mm (Met Office)) (Fig. 1) as part of a monitoring programme by the Environment Agency on behalf of the Department of Energy and Climate Change (Mitchell, 2009, 2010). Analyses were undertaken on the raw data provided by Mitchell (2009, 2010) to provide a comparison with Plymouth data.

3. Results

3.1. Rainfall activity concentrations and temporal variability

Data for monthly ^{7}Be activity concentrations in rainfall and mean monthly rainfall are shown in Table 1. The range of ^{7}Be activity concentrations over the study period in Plymouth was 1.56 (±0.13) to 2.67 (±0.20) Bq L^{-1} which agrees well with the values reported for other areas of the UK (Short et al., 2007). Fig. 2 shows a general trend of higher activity during the spring/summer months and lower activity during the autumn/winter periods. The mean activity concentration for February to December 2009 was 1.95 (±0.23) Bq L^{-1} and for January to December 2010, 1.97 (±0.34 Bq L^{-1}). The annual mean activity concentrations in Chilton and Aberporth were 0.75 (±0.6) Bq L^{-1} and 1.27 (±0.78) Bq L^{-1} in 2009 and 0.77 (±0.38) Bq L^{-1} and 1.67 (±0.84) Bq L^{-1} in 2010, respectively. The Kruskal Wallis test showed that there is a significant difference between the median concentrations of all three locations ($H(2) = 37.56$; $p < 0.001$).

To quantify the temporal variability of ^{7}Be activity concentration in rainfall within a seasonal context, mean seasonal values and standard deviations were calculated (Fig. 3). In Plymouth, the relative standard deviation (RSD) within a season was generally 10% with the exception of 15% in summer, 2010 and 0.7% in autumn, 2010. The larger variation in summer 2010 can be attributed to the low concentration value returned for August (1.56 Bq L^{-1}) in comparison to that of June (2.19 Bq L^{-1}). Variability highlighted in Plymouth was much lower than the seasonal variability for Chilton and Aberporth. RSD for Chilton ranged from 39% in summer, 2010 to 85% in autumn, 2010 with winter 2009–2010 showing 40% variability. Variability in Aberporth ranged from 10% in summer, 2010 to 60% in spring, 2009 with a winter value of 34%.

3.2. ^{7}Be deposition dynamics

Total rainfall for the whole study period in Plymouth (February 2009–February 2011) was 1789 mm with rainfall in 2010 below the 30 year average (1971–2000, 1000 mm (Met Office)) at 841 mm. Deposition for the 2009–2010 period is outlined in Table 2. Deposition in Plymouth was largely explained by rainfall ($r^2 = 0.94$; $p < 0.001$) and showed a stronger relationship with rainfall than in Chilton ($r^2 = 0.22$; $p < 0.05$) or Aberporth ($r^2 = 0.58$; $p < 0.001$) (Fig. 4).