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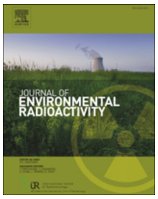

Downward migration of radiocaesium in organic soils across a transect in Scotland

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ABSTRACT

Following the accident at the Chernobyl nuclear power plant in 1986, radioactive materials including ^{137}Cs were distributed over large parts of the former Soviet Union and Europe. Due to the relatively long physical half-life of ^{137}Cs (30.1 y) measurable activity concentrations can still be observed in the natural environment. We have determined the depth distribution of ^{137}Cs from the testing of nuclear weapons in the atmosphere and Chernobyl, in eight highly-organic soils along an approximately east to west transect across the Central Highlands of Scotland. The ^{137}Cs activity concentrations in the soil profiles, corrected for radioactive decay to May 1986, were between 2.8 and 14.4 kBq m^{-2}. There were differences in the pattern of distribution of ^{137}Cs in the soil profiles. The mean migration depth of ^{137}Cs in the soil profiles was 12.2 cm with a range between 8.2 and 17.4 cm. Quantitative mineralogical analysis of the ash obtained after heating the soil to 400 °C indicated that clay minerals alone did not have a prime role in controlling the migration of ^{137}Cs down the profile.

1. Introduction

Radioactive ^{137}Cs does not occur naturally and is derived from anthropogenic activities. The main burdens of ^{137}Cs in soils are those from the atmospheric testing of nuclear weapons in the 1950's and in the beginning of the 1960's (Cambray et al., 1985; Carter and Moghissi, 1977), known as bomb derived radiocaesium, the nuclear power plant accident at Chernobyl in 1986 (ApSimon et al., 1989; Cambray et al., 1987; De Cort et al., 1998) and in 2011 from the tsunami damaged Fukushima Dai-ichi reactors (Masson et al., 2011). The accident at Chernobyl, which occurred on 26th of April 1986, led to an explosion and fires that burned for several days. Not only did the explosion contaminate the immediate surroundings of the power plant but also large areas of Europe, causing health and environmental problems far from the source (Smith and Beresford, 2005; Yablokov et al., 2010). For the general public, exposure to ^{131}I (half-life 8.0 d), ^{90}Sr (half-life 28.8 y) and ^{137}Cs (half-life 30.1 y) all of which were dispersed by wind were of most concern. In Europe contamination by ^{137}Cs created the greatest long-term problem, leading to restrictions on the distribution and marketing of contaminated food products. In Scotland much of the radioactivity was deposited on upland areas and has had long lasting effects on the rural economy. In 1986, following emergency orders, restrictions on the movement of sheep were imposed on 2144 farms in Scotland involving around 1.4 million sheep (FSA, 2007). In 2011 the ^{137}Cs activity in sheep on all farms in Scotland were below acceptable limits and the last restrictions on marketing sheep were lifted (FSA, 2010) but restrictions continued to be in force in other parts of the UK, especially in the northern part of Wales (FSA, 2011). All of the restrictions in the UK were finally lifted in June 2012 (FSA, 2012).

Less than one half-life of ^{137}Cs has elapsed since the accident at Chernobyl and therefore substantial ^{137}Cs physically remains in the environment and will do so for several decades to come. Once in the soil, ^{137}Cs can be fixed to clay minerals but forms reversible bonds with organic matter. Hence, the clay and organic matter content of soils affect the downward movement and potential availability of ^{137}Cs for uptake by plants. Partly to improve countermeasures in case of another accident, many studies have been conducted to enhance the knowledge of how Cs behaves in soil and how it is distributed within and between different plants. Immediately following the accident at Chernobyl it was believed that radiocaesium in mineral soils would be rapidly fixed by clay minerals and that in organic soils and peats it would be readily leached. For organic soils this belief was unfounded and ^{137}Cs remained near to the surface and available to plants (Cheshire and Shand, 1991).

Caesium forms a univalent cation with a small hydration energy and is easily adsorbed to negatively charged constituents in

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the soil, especially to illites. It is believed that the strong affinity for illites comes from the presence of frayed edge sites, which account for a minor part of the cation exchange capacity of illites (Cremers et al., 1988; Dumat et al., 1997). The fixation of Cs increases with time as the cations penetrate the lattice (Krouglov et al., 1998) and it has become evident that even if illite only makes up a small part of the clay fraction, its adsorption capacity for Cs is important (Cremers et al., 1988). Rosén et al. (2006) have shown that clay added to a peaty soil followed by repeated wetting and drying cycles, reduced the amount of ammonium-acetate exchangeable Cs. In a study of sorption behaviour of ^{134}Cs in 19 soils with a range of pH values, cation exchange capacities, extractable K, clay and organic matter contents, Shenber and Eriksson (1993) concluded that the distribution coefficient (K_d) values had the strongest correlation with the clay contents.

Soil organic matter has little selectivity for Cs since it is unlikely to form complexes. The adsorption of Cs to organic matter is determined by the concentration of Cs in relation to other competing cations and to the cation exchange capacity of the specific organic substance. Dumat et al. (1997) suggested that clay minerals prevent Cs from binding to organic matter or that the binding to organic matter is so weak that it is easily reversible. This was also the opinion of Maguire et al. (1992) who studied three different clays, namely illite, kaolinite, and smectite (from bentonite), and their role in the sorption of Cs to organic matter at different pH-values. Maguire et al. (1992) found that humic acids added to the clays depressed the sorption of Cs by the clays, especially for illite. The existence of Cs on exchange sites is just an intermediate state to being fixed to minerals (Shand et al., 1994). In some organic soils, there might not be sufficient illitic minerals to fix all the radiocaesium and radiocaesium left will compete with the other cations for occupancy of exchange sites. Hence the radiocaesium concentration in the soil solution will depend on other ionic components being present (Kudelsky et al., 1996). Organo-mineral soils cover a large part of the UK, especially Scotland. Therefore, knowledge of the composition of the amounts of mineral matter in peaty soils and how it changes with depth is important for assessing the movement and extractability of radiocaesium. Cheshire et al. (1998) found significant correlations between the amounts of mica together with vermiculite and chlorite, and the extractability of radiocaesium. Small amounts of clay minerals and zeolites in peaty soils are important in controlling the movement and plant availability of radiocaesium (Dumat et al., 1997; Rigol et al., 1998; Shenber and Johanson, 1992). Differences between the enrichment of Chernobyl and bomb derived radiocaesium in clay fractions have been observed (Spezzano, 2005) and attributed to ageing, i.e. the slow process of fixation in specific sites in illites. Rosén et al. (1999) found that migration rates were slow in clay soils and fast in organic soils.

The distribution of ^{137}Cs over the UK is variable. An airborne survey of northern Britain and Ireland found some clustering of high levels of ^{137}Cs activity on high ground, and directional banding on the ground, representative of rainfall during the passage of the Chernobyl derived plume (Scheib and Beamish, 2010). The vertical distribution of ^{137}Cs in soil affects the exposure of man and animals to ionising radiation. Gamma rays from ^{137}Cs in soil travel in air and the intensity of the radiation from the decay of ^{137}Cs experienced by man or animals on the land is dependent on the depth distribution and activity of the ^{137}Cs in the soil profile and the attenuation factors related to the elemental composition, bulk density, structure and moisture content of the soil (Demir et al., 2008). The amount and distribution of ^{137}Cs in the plant rooting zone also influences the transfer of radioactivity into the food chain, and the transport of radioactivity in the terrestrial environment by erosion and runoff (Smith and Beresford, 2005).

Apart from the survey carried out by Cawse and Horrill (1986) in 1977 prior to the Chernobyl accident, there is little information about the location depth and downward movement of radiocaesium in the highly organic soils of Scotland. Our hypothesis is that ^{137}Cs movement is largely controlled by the amount and forms of soil minerals, especially by those clay minerals with a capacity to fix interlayer cations. To test this hypothesis we have determined the distribution of ^{137}Cs in eight soil profiles at 1- to 2.5-cm depth increments for a transect across Scotland and investigated relationships with bulk soil mineral composition measured quantitatively by recently developed full-pattern fitting X-ray diffraction (XRD) methods.

2. Materials and methods

2.1. Sites and soils

In 1990 a series of four approximately E–W transects across Scotland were established (Bracewell et al., 1993) to monitor the trends in pollution of Scottish soils (TIPSS), see Fig. 1. Our study involved Transect 2 (T2), which comprised eight undisturbed, organo-mineral soils (Table 1). During the autumn of both 1990 and 1999 soil samples were taken using a cylinder (diameter 25 mm, height 50 mm) pushed into the ground. In 1990, only one sample was taken at each site. In 1999, four samples were taken at each site, in a north, south, east and west directions from a central point at a distance of 10 m, except for soil from site 2.1 where the distance was 20 m. During autumn 2008, soil samples were taken at sites 2.7 and 2.8 using the method described by Rosén et al. (1999) for Swedish agricultural soils. Briefly, at each location, three circles (A, B, C) of 5 m radius, centres 20 m apart, were mapped out in a south–north direction. Within each 5-m circle, 5 soil cores were collected. For the uppermost 10-cm of the soil, a cylindrical corer with a diameter of 57 mm was used for soil sampling and the core cut horizontally into 10-mm slices. In the middle of the first core pit, a core sampler with a diameter of 22 mm was used to core down from 10 to 50 cm. For some sites we were unable to core to 50-cm depth because of the occurrence of rock or hard, mineral horizons. This deeper core was cut into 25-mm slices. All subsamples from the same depth in each 5-m circle were put together to form a composite sample. The soil samples were air-dried (30 °C), sieved (2 mm) and any larger particles discarded. Soils from the other sites (2.1–2.6, Table 2) were sampled in spring 2010 by the protocols of Rosén et al. (1999) already described.

2.2. Radioactivity

The homogenised soil samples were placed in plastic containers with a volume of 35 or 60 ml depending on the amount of sample available. The gamma emissions from ^{137}Cs activity in the soil samples were measured using a computer aided germanium detector system (HPGe) in a low background laboratory for periods between 3 and 24 h depending on expected activity concentrations to obtain a statistical counting error due to the random process of decay ranging between 5 and 10%. Activities determined at the time of counting were corrected for radioactive decay back to the fallout day of 3rd May 1986. The mean migration depth X for ^{137}Cs in the soil was calculated using Eq. (1) (Arapis et al., 1997), where X_i is the centre of each layer in the soil profile, q_i is the relative activity concentration in each layer calculated using Eq. (2). In Eq. (2), A_i is the activity concentration in each layer, and A_{total} is the total activity concentration in the profile.