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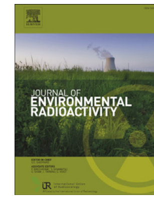

Long-term behavior of ^{90}Sr and ^{137}Cs in the environment: Case studies in Switzerland

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We present long-term records of the ^{137}Cs and ^{90}Sr activity concentrations in soil, grass and milk from two lowland and two alpine pastures of Switzerland. The data is used for better understanding the long-term behavior of these radionuclides in the environment. Transfer factors between compartments are used as qualitative indicators of the magnitude of transfer and as a way to compare different elements (e.g. Cs and Sr) in similar conditions. The long-term behavior was quantified by means of the effective half-life which integrates all processes that cause a decrease of activity in a given medium such as leaching, fixation, erosion and radioactive decay. Our study shows that ^{90}Sr is more likely transferred from alpine soil to grass than ^{137}Cs. This is explained by a stronger fixation of Cs in the soils. We observed higher transfers of ^{90}Sr to grass in soils with lower Ca concentrations, and vice versa. In contrast, the transfer of ^{137}Cs to grass was not affected by the variations of the K content in the soil. We provide evidence that shows that ^{137}Cs, after intake by dairy cattle, is more likely transferred to milk than ^{90}Sr. However, as the ^{90}Sr and Ca transfers to milk are influenced by parameters/processes that were not taken into account in our study, our result cannot be entirely validated. The effective half-lives of ^{137}Cs and ^{90}Sr in soil, grass and milk corresponded with previous estimates in alpine soils. We have found that processes other than radioactive decay are responsible for a major decrease of the ^{90}Sr activity in soil. For ^{137}Cs, on the other hand, radioactive decay is among the most relevant process. Our data shows to be of interest in studying the trends of behavior of radionuclides in alpine regions.

1. Introduction

The atmospheric nuclear weapons testing (NWT) in the last century led to radioactive pollution worldwide (UNSCEAR, 2000; Corcho Alvarado et al., 2014; Shaw, 2007). Accidents in nuclear facilities, such as the Chernobyl accident in 1986 and the Fukushima accident in 2011, further injected radioactive pollutants into the environment, although at a smaller scale (UNSCEAR, 2000; Shaw, 2007; IAEA, 2006). These injections of radioactive contaminants have prompted high concern about their potential impact on human health (UNSCEAR, 2000; ICRP, 1990). Assessing the resulting long-term radiation doses that humans might receive, either by exposure from radionuclides on the ground or by ingestion (via food and water), requires a good understanding of the long-term behavior of radionuclides in the relevant environments (IAEA-TECDOC-1616, 2009). Substantial amounts of data have been collected worldwide since the beginning of the NWT. This data includes the distribution and accumulation of radioactive materials in different parts of the biosphere and the main exposure pathways and mechanisms of radioactive contamination (IAEA-TECDOC-1616, 2009; Gastberger et al., 2001; Lettner et al., 2006, 2007; Pourcelot et al., 2007; Guillaume et al., 2012). The anthropogenic radionuclides ^{137}Cs, ^{90}Sr, ^{238}Pu, ^{239}Pu, ^{240}Pu, and ^{241}Am are among the most investigated contaminants as they have the potential to be radiotoxic and have long physical and ecological half-life (UNSCEAR, 2000; IAEA-TECDOC-1616, 2009). Due to their rapid incorporation into the food chain, we have focused our study on ^{90}Sr and ^{137}Cs.

The radioactive fallout originating from the accident in the nuclear power plant of Chernobyl led to a very heterogeneous spatial distribution of the radioactive contamination in Europe (Pourcelot et al., 2007; Lettner et al., 2009; Chawla et al., 2010). This heterogeneity was caused by changing meteorological conditions during

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the passage of the radioactive plume (Pourcelot et al., 2003). Generally, higher atmospheric deposition rates have occurred in regions with higher rainfall rates (Pourcelot et al., 2007; Hölgye and Filgas, 1995). This is why some alpine mountainous regions, which received higher amounts of precipitation than lowland regions, shows higher levels of contamination. This regional variability of precipitation has led to a strong correlation of the contents of anthropogenic radionuclides like ^{137}Cs and ^{90}Sr in soil, grass and milk at altitude (Lettner et al., 2006; Pourcelot et al., 2007; Guillaume et al., 2012; Lettner et al., 2009).

After atmospheric deposition, radionuclides are affected by different chemical, physical and biological processes in the soil-water-plant compartments (Ehlken and Kirchner, 2002). Cs mobility in soils has shown to be strongly affected by soil properties such as clay content, organic matter content, pH, ammonium and potassium content (IAEA-TECDOC-1616, 2009; Strebl et al., 2007; Riesen et al., 1999). In clay rich soils, for example, Cs shows a very low mobility and low availability for plant uptake (IAEA-TECDOC-1616, 2009). Sr, on the other hand, shows higher mobility than Cs. A faster downward migration of ^{90}Sr with respect to ^{137}Cs and other radionuclides (e.g. ^{241}Am, ^{239,240}Pu), have been demonstrated in mineral (sandy) and organic soils of France (Solovitch-Vella et al., 2007), and in alpine pastures in Austria (Gastberger et al., 2000) and Switzerland (Chawla et al., 2010). Some studies have shown that Sr competes with Ca for exchange sites in minerals (Strebl et al., 2007).

The radionuclide uptake by plants is highly influenced by site and plant specific parameters rather than the bulk radionuclide concentration in soil (Bunzl et al., 2000). Recent studies have focused on investigating radionuclide speciation in soil in order to determine their potential bioavailabilities and mobility. Substantial changes in soil temperature and moisture content have shown to have a significant impact on the geochemical forms of the radionuclides (Kovacheva et al., 2014). For example, periods of cooling, freezing and drought resulted in a significant increase of the exchangeable ^{137}Cs in fluvisol and cambisol soils (Kovacheva et al., 2014). This process may increase the bioavailability of this element.

A recent study in undisturbed alpine soils of Switzerland demonstrated that exchangeable K and Ca are good indicators of the extent of ^{137}Cs and ^{90}Sr transfer from soil to plant in silty-sandy soils and for plant species with short growing periods (Guillaume et al., 2012). This study also found that the uptake of ^{137}Cs by some plant species is less sensitive to variations in the amount of exchangeable K. In the case of ^{90}Sr, no significant differences were observed in the sensitivity of the plants to variations of the amount of exchangeable Ca. However, the levels of ^{90}Sr accumulation were highly dependent on the plant species (Guillaume et al., 2012).

Enhanced plant uptake of ^{137}Cs and ^{90}Sr have been reported for alpine pastures compared to intensively managed lowland pastures (Lettner et al., 2006, 2007; Pourcelot et al., 2007; Gastberger et al., 2000; Strebl et al., 2002). For ^{137}Cs this has been explained by the high retention of this radionuclide by the Alpine biomass and by the low amounts of exchangeable potassium in the soil solution of the Alpine soils (Gastberger et al., 2000). In the case of ^{90}Sr, this is related to the low amounts of exchangeable Ca in Alpine soils. However, other parameters/processes such as an extended snow cover and fixation by the biomass may likely impact the plant uptake (Gastberger et al., 2000).

Elevated ^{90}Sr and ^{137}Cs activities have been observed in milk samples from cows who grazed in Alpine pastures (Pourcelot et al., 2007). The contents of ^{90}Sr and ^{137}Cs in milk has generally shown lower variabilities than in soil and grass due to the grazing path of the animals over large areas (Pourcelot et al., 2007; Mück and Gerzabek, 1995). Milk has therefore been proposed as a more robust indicator of large-scale contamination in western Europe (and Switzerland) than soil and grass samples (Pourcelot et al., 2007).

This research is aimed at understanding the long-term transport processes of the long half-lives radionuclides ^{90}Sr and ^{137}Cs in alpine regions. This study is based on interpreting two decades of collected ^{90}Sr and ^{137}Cs data from the environmental compartments soil – grass – milk from two lowland and two alpine pastures from Switzerland. The study focuses on radionuclides that have been present in the soil for long-term periods and therefore in potential equilibrium within the different soil compartments (e.g. minerals, organic matter, soil water and living organisms). These radionuclides in the root zone are consequently available for plant uptake for long periods. It is assumed that the decrease of the radionuclide in a compartment of the environment can be described by an exponential model. The study also aims a better understanding of the competing pairs Ca/Sr and Cs/K in their transfer from soil to plants.

2. Materials and methods

2.1. Study sites

The Swiss territory has been contaminated by the global radioactive fallout originated from the atmospheric NWTs and by the fallout originated from the accident in the nuclear power plant of Chernobyl (Chawla et al., 2010; Huber et al., 1996). The global fallout from the NWTs produced a roughly equal contamination of ^{90}Sr and ^{137}Cs in the Swiss territory (Huber et al., 1996). In contrast, after the Chernobyl accident, the ^{90}Sr deposition was insignificant compared to the large fraction of ^{137}Cs deposited in the soils (Chawla et al., 2010; Huber et al., 1996; Jost et al., 2009). Aerosol measurements showed that after the Chernobyl accident the activity of ^{90}Sr in the Swiss atmosphere was about 1% of the value of ^{137}Cs (Huber et al., 1996).

The four investigated sites typify a variety of Swiss environments, namely two lowland and two alpine grazing pastures. The sites are located in the Swiss Mittelland (Fig. 1) and they are included in the long-term monitoring programme of Switzerland. The alpine pastures are of a few square kilometers size and they are used for extensive grazing between mid-June and end-September. The small parcels of Fahrni and Diesse are located at altitudes of about 850 m.a.s.l., and the sites of Gimmelwald and Mürren/Allmendhubel at higher altitudes of 1380 and 1900 m, respectively (Table 1). The main characteristics of the sites are given in Table 1. Alpine pastures are characterized by very heterogeneous plant composition and harbor high species richness (Meisseret al, 2014; Wohlgemuth, 2002). Studies have shown that small parcels can have more than 150 vascular plant species. Cattle animals have shown to have preferences with respect to some of these species (Meisseret al, 2014; Wohlgemuth, 2002). We are interested in the behavior of the radionuclide in the group of species, but not in any particular species. The Red Holstein and the Holstein are the two type of cattle breed in the investigated sites. These animals are feed mainly with grass, but additional supplements like corn, feedgrain and concentrates are also given. The origin and composition of the supplements was not investigated in this study.

2.2. Sampling

Soil and grass sampling are focused on selected small grazing parcels in Switzerland. Fahrni (845 m.a.s.l) and Diesse (852 m.a.s.l) are classified as lowland pastures. Gimmelwald (1380 m.a.s.l) is an alpine pasture. Mürren/Allmendhubel (1900 m.a.s.l) is an alpine pasture for summer grazing of the dairy cattle from Gimmelwald. At each site, composite soil samples were collected once a year