



Influence of chemical composition of precipitation on migration of radioactive caesium in natural soils



H. Thørring^{a,*}, L. Skuterud^a, E. Steinnes^b

^aNorwegian Radiation Protection Authority, P.O. Box 55, NO-1332 Østerås, Norway

^bDepartment of Chemistry, Norwegian University of Science and Technology, NO-7491 Trondheim, Norway

ARTICLE INFO

Article history:

Received 15 November 2013

Received in revised form

7 March 2014

Accepted 9 March 2014

Available online 3 April 2014

Keywords:

Chemical climate

Acid rain

Natural soil

Column study

Vertical distribution

Chernobyl fall-out

ABSTRACT

The aim of the present work was to study the impact of the chemical composition of precipitation on radiocaesium mobility in natural soil. This was done through column studies. Three types of precipitation regimes were studied, representing a natural range found in Norway: Acidic precipitation (southernmost part of the country); precipitation rich in marine cations (highly oceanic coastal areas); and low concentrations of sea salts (slightly continental inland areas). After 50 weeks and a total precipitation supply of $\sim 10\,000\text{ L m}^{-2}$ per column, results indicate that acidic precipitation increased the mobility of ^{134}Cs added during the experiment. However, depth distribution of already present Chernobyl fallout ^{137}Cs was not significantly affected by the chemical composition of precipitation.

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1. Introduction

In a previous field study we investigated regional differences in soil mobility and plant uptake of ^{137}Cs from natural soils in Norway, focusing on the possible impacts of regional differences in ionic composition of precipitation (Thørring et al., 2012). The present work was performed as a supplement to this field study.

Natural soils in Norway are typically podzols with a distinct top layer of decomposed vegetation materials where most plants take up their nutrients. Generally, the thickness of this humus layer range between 3 and 15 cm, and the organic matter content is 60–90% – often with a slightly higher mineral content in the lower part of the layer due to partial mixing with the underlying eluvial (E) horizon (Thørring et al., 2012; Gjelsvik and Steinnes, 2013).

In most types of soils, podzols included, specific binding to frayed edge sites (FES) of micaceous clay minerals such as illite is very important for radiocaesium retention (Cremers et al., 1988; Hird et al., 1996; Valcke and Cremers, 1994). The presence of organic matter, however, may reduce the affinity of caesium to clay minerals by e.g. blocking specific sites or forming clay–humus complexes (Dumat and Staunton, 1999; Staunton et al., 2002).

Adsorption of radiocaesium to humus sites can be important in highly organic soil (Valcke and Cremers, 1994), but will dominate only in cases where micaeous clays are largely absent – such as in soils with organic matter approaching 100% (Rigol et al., 2002). In such cases radiocaesium is – at least in principle – generally mobile, since caesium binds reversibly to humus. However, plant uptake (Thørring et al., 2012) and/or retention by microorganisms (Avery, 1996; Sanchez et al., 2000) may to some extent limit vertical migration in soil.

Several studies have shown that NH_4^+ and K^+ compete with Cs^+ for FES (e.g. Sanchez et al., 1999; Rigol et al., 2002), and it is well known that bivalent ions such as Mg^{2+} and Ca^{2+} effectively mobilise Cs^+ bound to humus sites. In many areas, the local precipitation can be a major source of such competing ions in natural soils. E.g. precipitation from the southern part of Norway contains rather high levels of H^+ and NH_4^+ from long-range atmospheric transport of pollutants (Semb, 1976; Tørseth and Semb, 1995), and areas near the open ocean receive precipitation enriched in marine cation such as Na^+ and Mg^{2+} (Låg, 1968; Varskog, 1995).

The aim of the present controlled laboratory experiment was to study the impact of various ionic composition of precipitation on vertical migration of aged and freshly added radiocaesium in soil from one of the areas in Norway where the Chernobyl fallout still causes problems for animal husbandry (e.g. Skuterud et al., 2005). Undisturbed soil columns were used in the experiment (similarly to

* Corresponding author. Tel.: +47 67162662; fax: +47 67147407.

E-mail address: havard.thorring@nrpa.no (H. Thørring).

e.g. Bachuber et al., 1982; Schimmack and Bunzl, 1992; Schimmack et al., 1994), and since conducted in parallel with Thørring et al. (2012), three comparable precipitation regimes were considered. Based on results from earlier field studies on natural surface soils in Norway (Gjelsvik and Steinnes, 2013), our working hypothesis was that the mobility of radiocaesium in soil would be higher using acidic precipitation and precipitation rich in marine cations in comparison to ion poor precipitation.

2. Materials and methods

2.1. Site description and field work

Sampling was performed in late September 2007 in the mountainous areas of the central part of southern Norway, which received high deposition of radiocaesium from Chernobyl. Caesium-137 remaining from the atmospheric nuclear weapons tests is of minor quantitative importance compared with the Chernobyl fallout in this area (Backe et al., 1986, 1987). The sampling site is located in the Northern boreal vegetation zone at 825 m above sea level in an area with pine forest (UTM 32V 0503098, 6839151). This area is shielded by high mountains to the west and the annual precipitation is rather low (500–800 mm), with low concentrations of marine ions and acid components (Thørring et al., 2012). The bedrock is from the Caledonian period with rock types such as gabbro, diorite, anorthosite, mica schist and phyllite. Crowberry (*Empetrum nigrum*) and cowberry (*Vaccinium vitis idaea*) were dominating understorey species at the sampling site; other abundant species being mosses (*Hylocomium splendens*, *Pleurozium schreberi*), lichens (*Cladonia stellaris*), and grass (*Deschampsia flexuosa*). The soil type was podzol with humus thickness of 2–3 cm in average.

Undisturbed soil profiles were sampled in the following manner: High-density polyethylene (PEHD) plastic tubes 40 cm long with an outer diameter of 20 cm and material thickness 0.75 cm were forced vertically through the soil using a rubber mallet. To ease the work, the outside of the down pointing end of the tubes was sharpened to 0.2–0.3 mm, and the surrounding soil was gradually removed using a spade. The soil profiles were cut using a sharp metal plate and a lid was attached to the bottom of the tubes. Ten soil profiles were taken using this method and will be referred to as “columns” in the following. In addition, five reference profiles (0–40 cm) were sampled using a soil auger of diameter 5 cm. All soil samples were collected within an area of approximately 2×10 m.

The five reference profiles were temporarily stored in a fridge until they were cut in 2 cm slices down to 10 cm depth, and thicker slices below that depth (generally 5 cm). These reference soil samples were subsequently transferred to polyethylene bags and put on cold storage (-20 °C). The 10 whole columns were also stored at -20 °C for a couple of months (awaiting the lab set-up).

2.2. Column study

After thawing, about 5 cm of the bottom soil was removed, and a piece of filtering cloth and a perforated steel plate with 3 mm holes were fastened in the bottom part of each column. Grouting was used to seal the small gap between the perforated plate and tube. Understorey species such as grass, lichen and mosses were removed from the top of the column and stored at -20 °C for subsequent analyses.

Three types of artificial precipitation – with composition based on monitoring data from the European Monitoring and Evaluation program (EMEP) (<http://www.nilu.no/projects/ccc/>), and representing a natural range found in Norway – were prepared for the

study. The composition of type A precipitation is representative of the long-range polluted, acidic precipitation from the southernmost part of the country, whereas type B is rich in marine cations such as Na^+ and Mg^{2+} typical of areas very close to the open ocean. In order to make the composition of precipitation B more distinctly different from precipitation A it was decided to not add any additional strong acid, ammonium or nitrate. Precipitation type C is characteristic of slightly continental precipitation, containing low concentrations of sea salts. The compositions of the three types of artificial precipitation used are shown in Table 1.

All dilutions in the study were performed using purified water (conductivity: $0.055 \mu\text{S cm}^{-1}$). For control/monitoring purposes, conductivity and pH was measured in all series of freshly made precipitation ($n = 35$) during the project period. No significant deviations were observed for these parameters during the study.

Nine of the ten columns were used in a specially made set-up, with groups of three columns receiving artificial precipitation of type A, B or C, respectively (Fig. 1). The 10th column was left at room temperature (with lid on) without addition of artificial precipitation. Besides that, it was treated as the other nine columns, and regarded a 6th reference profile in the following. The surface area of the columns was 269 cm^2 .

A peristaltic pump (Watson-Marlow 323S) with 2×5 channel micro pumpheads (314 MC) was used to supply the columns with artificial precipitation. Pump flow was 0.55 ± 0.02 mL per minute. A filter paper (VWR 415, 180 mm) was placed on the soil surface to enhance dispersion of artificial precipitation on the soil surface. The upper part of the column was thereafter covered with plastic film to minimise evaporation loss. The study was performed at room temperature (19 ± 2 °C). During the project period 272 ± 5 L of artificial precipitation was added to each column of which $98 \pm 2\%$ were recovered in the discharge water. The added amount of artificial precipitation corresponds to approximately $10\,000 \text{ L m}^{-2}$. Total amount of cations supplemented during the project period corresponds to 4400, 12000 and 180 mmol c m^{-2} for precipitation type A, B and C, respectively. Following an equilibrium period of 83 days of continuous precipitation, 7000 Bq of ^{134}Cs was added to all columns in order to investigate differences in mobility between “freshly added” ^{134}Cs and “older” ^{137}Cs mainly from the Chernobyl accident, presumably more strongly fixed in the soil. Only insignificant quantities of ^{134}Cs were left from the Chernobyl fallout in the sampling area by 2008–2009 and are thus unlikely to have had any impact on the results.

Effluent samples were taken at regular intervals (about 3 times a month). Conductivity and pH were measured in all samples, whereas radiocaesium (^{134}Cs , ^{137}Cs) was determined in selected samples using gamma spectrometry (NaI detector). To determine the water retention time in the columns a tracer test using tritium was performed. A portion of 14 kBq tritium was added to each of the 9 columns, and samples of effluent were gathered twice a day until break-through was observed. After that samples were taken once a day (or less). Tritium concentrations were determined using liquid scintillation counting (Quantulus Rack Beta). The recovery of added tritium in the discharge water was $98 \pm 1\%$.

After 50 weeks the column set-up was taken down. The soil was gradually pushed out of the plastic tubes, while the profiles were carefully cut in slices – 2 cm thick for the uppermost 12 cm, and 3–5 cm further down. These samples, together with the references described above, were dried (105 °C, ~ 48 h) and homogenised using hand force, and only larger stones were removed. All samples were analysed using gamma spectrometry (HPGe detector). During sample preparation it was observed that the thickness of the humus layer differed between soil columns, ranging from approximately 1 cm in A2 and C2 to about 7 cm in C3. Mass depths for each of the nine treated column are therefore shown in Table 2.

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