Applied Radiation and Isotopes xxx (xxxx) xxx–xxx

Contents lists available at ScienceDirect

Applied Radiation and Isotopes

journal homepage: www.elsevier.com/locate/apradiso

Environmental radioactivity study of Austrian and Bavarian forest ecosystems: Long- term behaviour of contamination of soil, vegetation and wild boar and its radioecological coherences

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ARTICLE INFO

Keywords:
Chernobyl
Radiostrontium
Radiocaesium
Wild boar
Vegetation
Soil
Environmental analysis
Liquid scintillation counting
Gamma-ray spectrometry

ABSTRACT

^{137}Cs and ^{40}K in soil, vegetation and flesh of wild boar samples from Austrian and Bavarian regions were investigated by gamma-ray spectrometry and ^{90}Sr in bones of wild boar with Liquid Scintillation Counting (LSC) after radiochemical separation. The soil core profiles revealed that 70–97% of the soil caesium content is still accumulated in the 0–10 cm soil depth. From all vegetation samples the mushrooms, particularly the bay boletus showed the highest ^{137}Cs contamination. The activity concentration of ^{137}Cs in muscle tissue of boar ranged from 14.9 ± 1.5 Bq/kg (Bavaria) to 4711 ± 377 Bq/kg (Lower Austria). In the bones of wild boars, ^{90}Sr activity concentration ranged from 1.4 ± 0.2 Bq/kg (Bavaria) to 70.3 ± 10.5 Bq/kg (Upper Austria).

1. Introduction

Thirty years after the Chernobyl accident on 26th of April 1986 the consequences are still noticeable and remain an important topic in the media and for society. Following the Chernobyl nuclear accident, enormous efforts have been undertaken to monitor the released radionuclides, both in Austria and Bavaria (Fieltz, 2005). Since forests are almost closed ecosystems, pollutants and therefore radionuclides, return to the ecosystem when the organism, whether flora or fauna, dies. Plants and animals can, therefore, take up these radionuclides again. Research showed a cycle for ^{137}Cs in the spruce needles and the surrounding earth in the Weinsberger Forest in Austria (Strebl et al., 1999).

The objective of this project is to study the behaviour of radionuclides in forest ecosystems 30 years after the Chernobyl accident and to show the interactions between soil, vegetation and wild boar.

Gamma-emitters offer the advantage of allowing the qualitative and quantitative determination of the activity in a straightforward radioanalytical measurement, without major requirements with respect to sample preparation. The analysis of other radionuclides, in particular pure β^--emitters, is much more elaborate because it usually requires the separation of the targeted radionuclide from other β^--emitters, which may even occur in excess. In particular, this refers to ^{90}Sr ($T_{1/2}$ = 28.8 years) which is a prominent fission product and biologically relevant. Due to its chemical similarity with calcium, radiostrontium is taken up into the bone and has a long biological half-life. ^{90}Sr (maximum β^--energy 545.9 keV) has a short-lived daughter, ^{90}Y ($T_{1/2}$ = 64 h), which is a very poor γ -emitter as well, but a powerful β^--emitter with a maximum β^--energy of 2278.7 keV. Environmental analysis of ^{90}Sr (and its daughter nuclide ^{90}Y) requires the effective separation from other β -emitters before its measurement using liquid scintillation counting (LSC) or other radioanalytical methods (Steinhauser et al., 2013).

2. Methods

2.1. Sample preparation and gamma-ray spectrometry of vegetation and soil

2.1.1. Sampling

At each location samples of the most abundant vegetation species (mushrooms and plants) were collected at a plot representing the

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http://dx.doi.org/10.1016/j.apradiso.2017.03.008
Received 15 August 2016; Received in revised form 6 March 2017; Accepted 10 March 2017

typical fauna of the location. Samples were drawn from each part of the plot to pick up the diversity of the location and filled into plastic bags and weighed on a scale directly in the field.

In addition a second and third walk through the Weinsberger Forest was done on 6th September and 25th October 2015 to collect all kinds of mushrooms, that could be discovered. This additional sampling was necessary due to a lack of mushroom samples resulting from a too dry summer during the field sampling in July 2015.

Beside vegetation samples two kinds of soil samples were collected at each location. One set of samples were taken a spade for soil parameter analyses of the Ah and B horizon. An adequate amount of sample were put into a plastic bag and weighed with the scale. A second set of samples was obtained with a 60 cm long soil cutter which was hammered into three positions, one meter apart from each other, in the location plot to gain a depth profile of caesium. The layers were divided on the basis of the expected caesium distributions into 5 cm layers for 0–20 cm depth and 10 cm layers for 20–60 cm depth. Each layer was mixed together with the appropriate layer of the three positions respectively and packed into plastic bags and weighed to receive the fresh mass.

2.1.2. Preparation and measurement

All vegetation and soil samples were dried in a drying oven for 24 h at 45 °C and afterwards for 48 h at 105 °C. The 45 °C procedure was done to make sure samples would not stick on the vessels when losing water quickly. After determination of the dry mass the samples were homogenised and ground. The gamma-ray spectrometry measurement of the samples (6 h respectively) was started at the Low -Level Counting Laboratory of the BOKU Vienna (University of Natural Resources and Life Sciences). For evaluation of the efficiency of the geometry, Monte Carlo simulations were done for each sample.

This procedure was done for all vegetation and soil samples except of the Ah and B horizon samples. These were dried and directly tested for soil parameters like pH, conductivity, CEC, carbon and nitrogen content according to the Austrian norms in the laboratory of the BOKU Vienna.

2.2. Sample preparation and gamma-ray spectrometry of wild boar flesh

Samples of wild boar bones and flesh were obtained from various hunters from Austria and Bavaria in the summer of 2015. The muscle samples for the measurement of ^{137}Cs came from the forelegs of the wild boars. Fat and tendons were removed in order to obtain pure muscle tissue.

Due to the sample inhomogeneity, uncertainties of the measuring geometry were estimated to be around 5%. These uncertainties were smaller than the relative measurement uncertainties of about 8-10%. The samples were measured for 20,000 s (5,5 h) and evaluated with the programme GENIE™2000 (Canberra).

2.3. Determination of ^{90}Sr in the bones of wild boar

Since ^{90}Sr is a pure β^- -emitter, it must be separated from other radionuclides to be measured by LSC, therefore radiochemical processing is needed. First the bones were ashed at 600 °C over 38 h and then ground. Approximately 3 g of this powder were weighed and mixed with concentrated HNO_3 (15 ml) and H_2O_2 (3 ml) then dissolved with microwave digestion. Then the solution was filtered into a 50 ml volumetric task and filled up to the mark. The capsule was rinsed several times with water and emptied into the filter to be sure the sample was transferred in its entirety. From the 50 ml volumetric flask, 0.25 ml were pipetted into a second 50 ml volumetric task and diluted to the mark for ICP-MS measurement. The remaining solution was transferred into a beaker and 0.4 ml of a Sr-Carrier solution ($Sr(NO_3)_2$: 5 mg/ml) were added and the resulting solution evaporated to dryness. The residue was taken up in 12 ml 8 M HNO_3 and twice 60 µl was pipetted into two 50 ml volumetric flasks for ICP-MS measurement.

For the separation of the strontium from other radionuclides, strontium-selective resins (Sr-resin) produced by Eichrom®/TrisKem® were used. The Sr-resin was preconditioned with 30 ml 8 M HNO_3, and then the solution with the sample was applied to the resin. The beaker was rinsed with 25 ml of 8 M HNO_3 and the solution was applied to the resin as well. Then the resin was washed with 10 ml of a solution with 3 M HNO_3 and 0.05 M Oxalic Acid to remove calcium. Afterward, the resin was washed with 10 ml 3 M HNO_3 and 10 ml 0.05 M HNO_3.

The strontium-fraction was then eluted into a beaker with 45 ml 0.05 M HNO_3 and evaporated to dryness. To determine the strontium-loss through the intermediate steps, ICP-MS measurements were done to evaluate the amount of strontium in the steps immediately after the filtration, before the resin and after the resin. After the Sr solution has evaporated to dryness, the residue was taken up in 8.12 ml 0,05 M HNO_3 and pipetted into a LSC-vial. From the vial two portions of 60 µl were pipetted into two 50 ml volumetric flasks and diluted for ICP-MS measurement. To the 8 ml potion of Sr-solution, 12 ml of the Scintillation–Cocktail “HiSafe 3” were added and shaken. The ICP-MS measurements revealed a strontium recovery of 90 – 95% through the radiochemical processing before applying to the resin. The resin itself had a recovery of 80–90% depending on how often it was used. For each sample individual recoveries were determined and taken into account.

To validate the measurements, not only were the samples measured, but also a ^{90}Sr standard and a background. To prepare the ^{90}Sr reference standard (Activity: 0.4991 Bq ± 5% (1σ), 12.10.2007), 80 µl of a ^{90}Sr solution (6.28 Bq/ml) and 0,4 ml of the Sr carrier were pipetted into a beaker and evaporated to dryness. The residue was then taken up in 8 × 1 ml 0.05 M HNO_3 and pipetted into a vial. To the solution in the vial, 12 ml of HiSafe3 were added and shaken. The preparation of the background–vial followed the same procedure but only with 0.4 ml Sr-carrier.

The measurements were started a few hours after the radiochemical separation or on the next day and were stored in the liquid scintillation counter to stay dark and cool. The ^{90}Sr standard was measured for 20 min, the background and the samples for 80 min. For each sample 6 cycles were measured and an average outcome was calculated. The relative measurement uncertainties for the ^{90}Sr - measurements were 15–18%.

3. Results

3.1. Results of the soil depth profiles

The bioavailability and the transport of caesium in the soil are influenced by 4 main factors: pH value, clay minerals, humic substances and homologous cations.

According to Frissel et al. (1992) a low pH value leads to higher bioavailability. Clay minerals, which consists of different combinations of SiO_4-Tetraeder and $AlOH_6$-Oktaeder layers, have a high cation exchange capability. The fixation of caesium takes place in the interlayer-edge zones, the so called frayed edge sites (Fraiture, 1990). Humic substances are also able to fixate caesium due to their large quantity of functional groups, but the fixation is not as effective as by clay minerals (Strebl et al., 1996). Another important role is played by homologous cations like K^+ or Rb^+ as the uptake of caesium in plants is reduced by direct competition when increasing the concentration of K^+ and Rb^+(Fraiture, 1990). To involve soil parameters into the study the pH value and the cation exchange capability (CEC) were evaluated for the locations. Garmisch Partenkirchen was the only site with limestone as rock and thus provided different results.

The analysis of the soil core profiles indicate a very slow migration of caesium into deeper soil layers as the majority of the caesium fraction is still stored in the superficial soil layers. At the 13 locations 70–97% of the caesium content in the soil is accumulated in the