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Transfer of natural radionuclides from hay and silage to cow's milk in the vicinity of a former uranium mine

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ABSTRACT

After the closure of the former Žirovski Vrh uranium mine in Slovenia, mining and milling wastes were deposited on two waste piles, which are located close to the mine. These wastes contain elevated levels of natural radionuclides from the uranium decay chain. Due to different migration processes (erosion, aerial deposition, through groundwater), these radionuclides can be transported via fodder into cow's milk, which is an important foodstuff for Slovenian people. Therefore, natural radionuclides were analysed in the transfer food chain from soil to cow's fodder and cow's milk. After sampling, ^{238}U, ^{234}U, ^{230}Th, ^{226}Ra, ^{210}Pb and ^{210}Po were determined using radiochemical separation methods and alpha spectrometry or proportional counting. Hay and silage to milk concentration ratios (kg dry weight L^{-1}) were calculated and were 0.260 for ^{238}U, 0.255 for ^{230}Th, 0.070 for ^{226}Ra, 0.021 for ^{210}Pb and 0.019 for ^{210}Po. The calculated annual ingestion dose due to milk consumption for the natural radionuclides analysed was 9 μSv/year for adults and 389 μSv/year for infants with the highest contribution of ^{210}Po (51% for adults and 63% for infants) and ^{210}Pb (36% for adults and 24% for infants). This study provides new data quantifying the transfer of natural radionuclides to milk, which is a parameter for which there have been very few previously reported values.

1. Introduction

Wastes with enhanced natural radionuclide concentrations, such as uranium mining and milling wastes, are often situated close to agricultural areas. This is the case for the former uranium mine at Žirovski Vrh, in Slovenia, where, amongst other agricultural crops, farmers produce fodder for dairy cows. In addition, phosphate fertilizers used in fodder production usually contain a higher content of natural radionuclides compared to the environmental background (Ghosh et al., 2008). This can additionally enhance activity concentrations in the soil used for producing fodder (Ioannides et al., 1997). Due to the relatively high human milk consumption, especially of infants, it is important (i) to measure the activity concentration of natural radionuclides in milk, and (ii) to quantify the transfer of natural radionuclides through the food chain from soil via fodder to milk. Whereas there are many studies concerning natural radionuclide activity concentrations in milk (Pietrzak-Flis et al., 1997; Ababneh et al., 2010; Al-Masri et al., 2004; Giri et al., 2011), there is a lack of data which enable the calculation of transfer parameter values for natural radionuclides in milk (IAEA, 2010; Fesenko et al., 2007). Therefore, in this study natural radionuclides were analysed in soil from fields used for fodder production, in the fodder produced (hay and silage) and in cow's milk, and the data used to estimate the transfer parameter values.

This study provides new data quantifying the transfer of natural radionuclides to milk, which is a parameter for which there have been very few previously reported values (Howard et al., 2009a, 2009b).

2. Materials and methods

2.1. Sampling and sample preparation

All samples were collected from a farm which lies about 500 m from the former U concentrate production plant (Fig. 1). Soil samples were obtained from three fields where fodder for cows was produced (Fig. 1). A pooled sample covering the whole of each field was obtained by random sampling with a corer designed to sample about 200 g at once at the depth from 0 to 15 cm. This gave about 5 kg in total of each soil sample. Pre-treatment of the soil samples was carried out according to ISO 11464 (1994). The sample was dried at 80 °C to constant weight. Then the larger stones and roots were removed and the sample was sieved through a 2-mm screen sieve. The fraction which passed through the sieve was then

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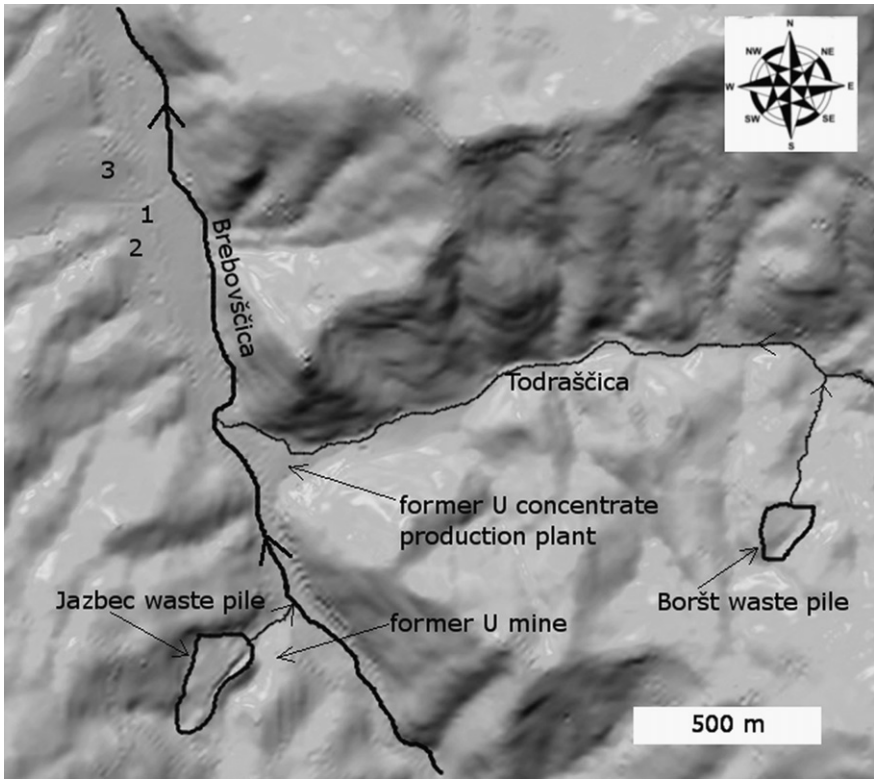

Fig. 1. Soil sampling locations (1–3) and area of the former uranium mine Žirovski Vrh.

homogenised and a subsample for the analysis was taken by hand quartering.

Sampling of fodder took place at the time when the cows were being fed. Due to possible variations in natural radionuclide activity concentrations in hay and silage, samples of hay and silage were randomly taken directly from the fodder bin of each cow. About 5 kg of hay and 5 kg of silage was collected and dried to constant weight. Hay and silage samples were not washed due to the fact that cows ingested hay and silage that is not washed and the transfer of radionuclides could not be assessed properly, if the samples would be washed.

Milk was collected from two cows (each with a weight about 500 kg), which were always kept indoors and were fed with hay and silage. About 2.5 L of milk was collected from each cow during milking and both samples were combined to get one sample of about 5 L of milk. Afterwards, the milk sample was weighed and then dried at 60 °C and reweighed to determine the dry matter content. The dried milk sample was homogenised and stored until analysis.

2.2. Radiochemical separation procedure for ^{238}U, ^{234}U, ^{232}Th, ^{230}Th, ^{226}Ra

The radiochemical separation procedure for ^{238}U, ^{234}U, ^{232}Th, ^{230}Th and ^{226}Ra is outlined below and the detailed procedure can be found in Štrok et al. (2010). All samples were analysed in duplicate. The radiochemical separation procedure was the same for soil, hay, silage and milk samples. Dried sample was first ashed at 650 °C in a muffle furnace to remove organic matter. After that, the sample was fused with Na_2O_2 and Na_2CO_3 at 900 °C. Then ^{232}U, ^{229}Th and ^{133}Ba tracers were added. The fusion cake was then dissolved by the addition of concentrated HNO_3, and transferred to a Teflon beaker. Then, the sample was further digested by the addition of concentrated HCl, HNO_3 and HF acids and H_2O_2. Finally, concentrated H_2SO_4 was added and evaporated until incipient dryness.

After final evaporation, radionuclides were co-precipitated on $Fe(OH)_3$. After washing, the precipitate was dissolved with 3 M HNO_3/1 M $Al(NO_3)_3$ and thorium was separated from uranium and radium on a TEVA separation column.

The uranium and radium-containing eluate obtained from the separation on the TEVA column was transferred to a UTEVA separation column, where uranium was separated from radium.

Radium and barium were co-precipitated from the radium-containing eluate with $PbSO_4$. Then the precipitate was dissolved in 0.1 M EDTA/0.5 M NaOH. Afterwards a $Ba(Ra)SO_4$ precipitate was formed by a consecutive addition of Ba carrier, pH 0–5 indicator, 1:1 acetic acid, a saturated solution of Na_2SO_4 and 0.125 mg/mL $BaSO_4$ substrate. After 30 min, the solution was filtered through a 0.1 μm filter. Finally, the filter was dried and mounted on a stainless steel planchet and dried again.

Thorium was microprecipitated from the thorium-containing eluate with NdF_3. Uranium from the uranium-containing eluate was firstly reduced with 15% $TiCl_3$ and then microprecipitated with NdF_3. Both thorium and uranium microprecipitates were placed in an ice bath for 30 min into an ice bath prior to filtration. Filtration was carried out through a 0.1 μm filter that was previously rinsed twice with 10 μm/mL NdF_3 substrate solution. Finally, the filter was mounted on a stainless steel disc and dried.

2.3. Radiochemical separation procedure for ^{210}Pb

Samples for ^{210}Pb were not ashed. In a similar manner as for uranium, thorium and radium, ^{210}Pb was also extracted using a radiochemical separation procedure adopted from Štrok et al. (2010) and is therefore only briefly summarised here. All samples were analysed in duplicate and the radiochemical separation