Study of plutonium redistribution in a raised peat bog following a fire

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Abstract

Peat lands are archives of past diversity, climate, and other environmental conditions. The redistribution of radionuclides in a peat bog after a fire consists of change in vertical distribution and releases/losses to the atmosphere through evaporation and resuspension. The study of different radionuclides such as plutonium isotopes will be important to understand the radiation exposures to man following fire on contaminated land.

In this work, a fast and sequential procedure for uranium, plutonium and americium determination in soil samples is presented. Together with the study of redistribution of uranium and plutonium isotopes in a raised peat bog located in the nature reserve of Vakö Mire (south of Sweden) following a big fire (1992).

The procedure is based on the four main steps of any radiochemical procedure: pre-treatment, isolation of radionuclides, source preparation and measurement via alpha spectrometry. The sequential separation of the radionuclides is performed by extraction chromatography, employing UTEVA resin in columns and DGA resin in cartridges.

Once the procedure was established, two cores of the Vakö Mire peat bog were studied (one from the burnt area and another from the not affected area). The first 10cm of each core sliced in 1 cm layers were analyzed. The results for plutonium in the burnt core show an increase in the $^{239+240}$Pu activity concentration (Bq/m^2) at the ash layer, while in the not affected area the concentration slightly increases with depth due to the global fallout. Regarding uranium isotopes, their behaviour in the peat bog is different from plutonium. The non-burnt area presents much higher values of activity concentration than the burnt one due to its higher inorganic content. The fire made this element easily movable via weathering conditions and as results the U content in the burnt core is hardly 10% of the total amount accumulated in the non-burnt area. Moreover, ^{234}U and ^{238}U isotopes were found in secular equilibrium.

Introduction

Peat lands are archives of past diversity, climate, and other environmental conditions. The upper layers of a peat land are an archive for recent environmental information, such as atmospheric deposition of natural and man-made radionuclides, and offer an opportunity to understand how environmental conditions have been recorded in peat profiles by relating the deposition to the peat to recent historical records.

The redistribution of radionuclides in a peat bog after a fire consists of change in vertical distribution and releases/losses to the atmosphere through evaporation and resuspension. Selected radionuclides such as ^{210}Pb, ^{137}Cs and $^{239+240}$Pu will be of major concern (among others). This is of importance to understand the radiation exposures to man following fire on contaminated land.

Materials and Methods

Samples

The nature reserve of Vakö Mire is located in the south of Sweden. The area of the peat bog is 13 km^2 and is one of the largest raised peatbogs in Sweden. Most of it was affected by the fire in 1992 but some areas remained intact (Hökönsintresseförening, 2012). In the summer of 2015, through an expedition in this area, different peat bog samples were taken (Figure 1). The view of the reserve in 2015 is shown in Figure 2.

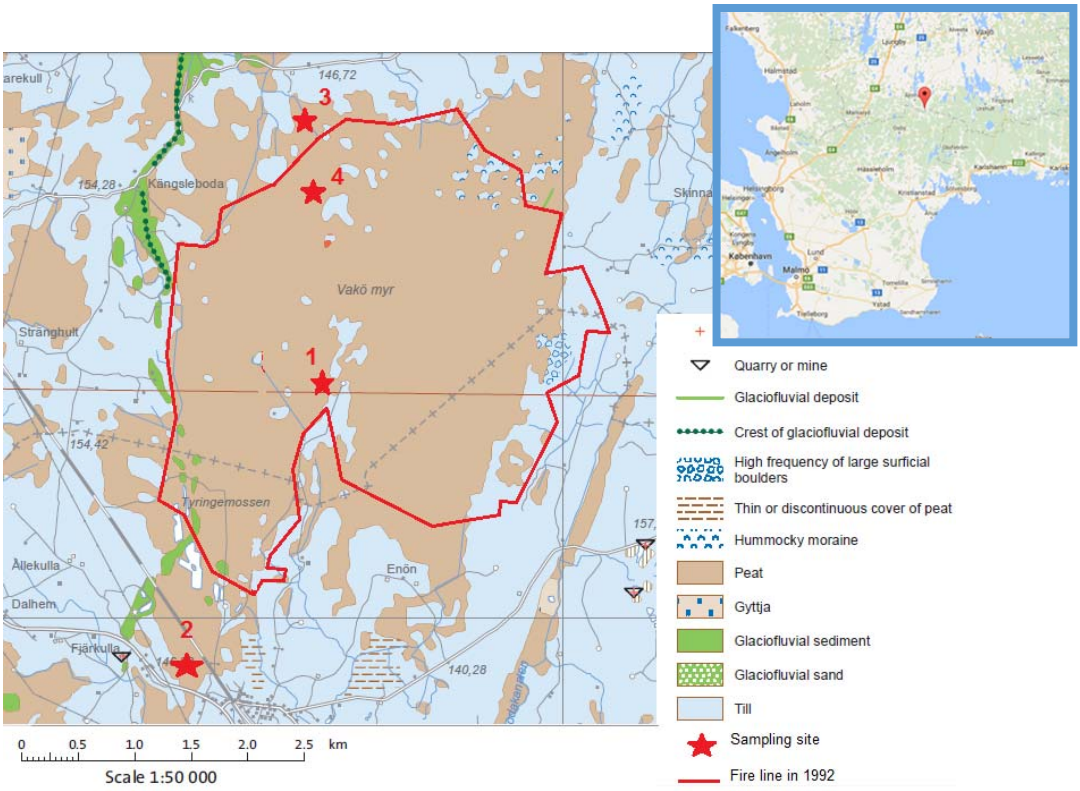

Figure 1. *Nature reserve of Vakö Mire, sampling points and situation in Sweden.*

Figure 2. *View of Väko Mire in 2015.*

In this expedition vertical cores were taken by a steel corer, with a diameter of 7.7 cm. Several cores were taken in the peat bog both in the area of the fire and outside the affected area (Figure 3). The cores were transported to the laboratory for further treatment.

The cores were sliced in 1 cm layers. Assuming a growth rate of 2 mm/year, the growth in the peat bog 24 years after the fire has been about 4-5 cm. Below this depth, we expect to find the "ash layer", as it is seen in the core from the burnt area (Figure 3).

Figure 3. *Cores from the peat bog in the area subject to the fire (left), and from the unburnt area (right). The ash layer is seen at a depth of about 5 cm in the core from burnt area.*

Radiometric technique

The procedure is based on the four main steps of any radiochemical procedure: pre-treatment, isolation of radionuclides, source preparation and measurement via alpha spectrometry.

Initially each 1cm slice was dried at 60-70 degrees for several days until the weight became stable, removing this way the moisture of the sample. Afterwards the dried fraction was calcined at 450 degrees for 24 hours, removing most of the organic fraction. Then a leaching digestion with aqua regia was applied. In order to carry out the sequential separation of the radionuclides, extraction chromatography employing UTEVA resin in columns and DGA resin in cartridges is performed. The complete procedure is shown in Figure 4.

After isolation, every fraction containing either U isotopes or Pu will be electroplated. The method of electrodeposition applied in this case follows strictly the method detailed in Hallstadius (Hallstadius, 1984).To maximize the chemical recovery of both actinides U and Pu sources are electroplated at 1.2 A during 1 hour in a 20mm diameter steel disk.

The measurement system is an Alpha Analyst from Canberra equipped with 12 PIPS detectors and the software used to analyze spectra was Genie 2000.

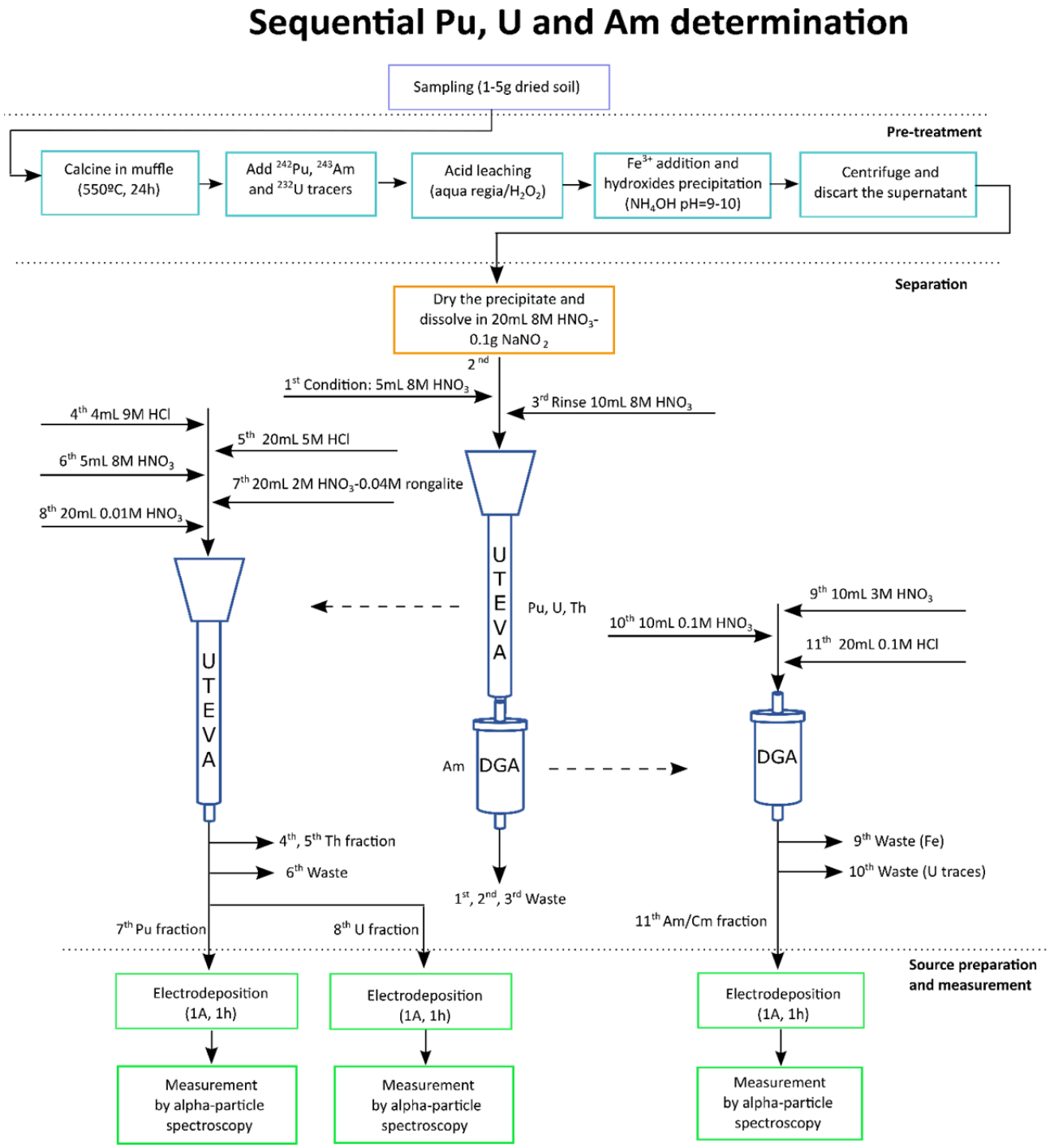

Figure 4. *Fast and sequential procedure for uranium, plutonium and americium determination in soil samples.*

Results and Discussion

Drying and Calcination

Core 12 from the burnt area (Site 4 in Figure 1), and Core 10 from the unburnt area (Site 3 in Figure 1), were analyzed in the laboratory. The 1cm slices of each core were dried and calcined. Figure 5 shows different behavior between the cores: while Core 12 is compound mainly (averaging 97%) by

organic matter, Core 10 (non affected by fire) shows a higher inorganic fraction, specially from 10cm and deeper, where it is compound around 75% by organic matter.

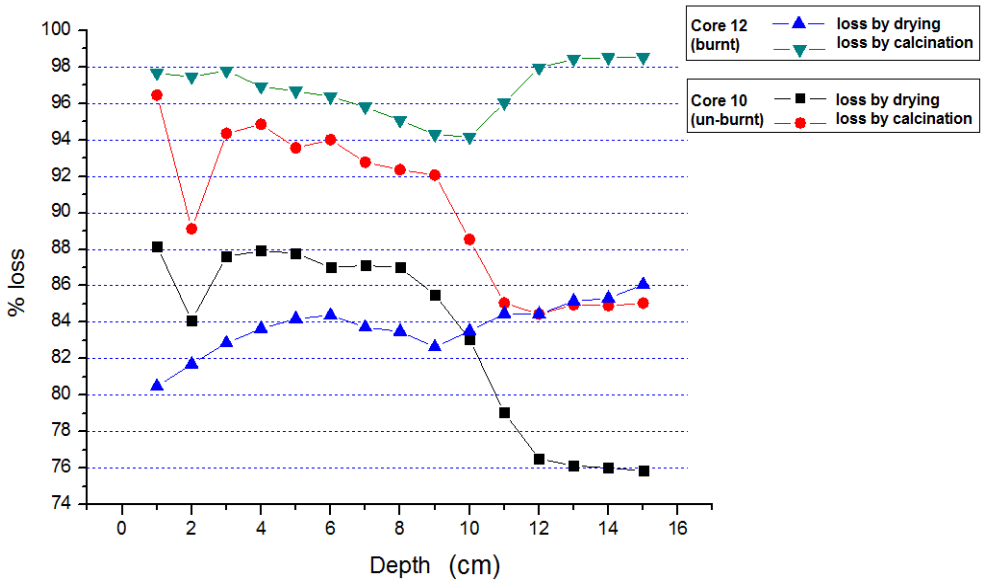

Figure 5. *Loss on every 1cm layer for both cores: after drying and calcination.*

Plutonium Redistribution

The first 10cm of each core sliced in 1cm layers were analyzed for plutonium and uranium determinations. The results for plutonium in the burnt core show an increase in the $^{239+240}$Pu activity concentration (Bq/m^2) at the ash layer (6cm depth), while in the non-affected area the concentration slightly increases. Assuming an annual growth rate of 2mm, we would expect the fallout of $^{239+240}$Pu from nuclear tests to be at 10cm depth in a general peat bog (Core 10). In Väko Mire the fire went deeper (until 20cm) and the organic matter turned into ashes, concentrating plutonium isotopes in the "ash layer".

The ^{238}Pu/$^{239+240}$Pu ratio in 1972 was 0.036 , 36% originated from the satellite failure SNAP 9A and the rest from the global fallout (Holm and Persson, 1975). The fallout from the Chernobyl accident was very patchy with an activity ratio of 0.47 which decreased to 0.37 in 2015. The increase of ^{238}Pu, expressed as Bq·m^{-2}, was only about 9% in Sweden following the Chernobyl accident and probably less in the studied area (Holm, 1991). In Core 12, the ratios are mainly situated around this line.

Uranium Redistribution

The behavior of uranium isotopes in the peat bog is different from plutonium (Figure 6). The non-burnt area presents much higher values of activity concentration than the burnt one due to its higher inorganic content. The fire made this element easily movable via weathering conditions and as results the U content in the burnt core is hardly 10% of the total amount accumulated in the unburnt area. Moreover, ^{234}U and ^{238}U isotopes were found in secular equilibrium.

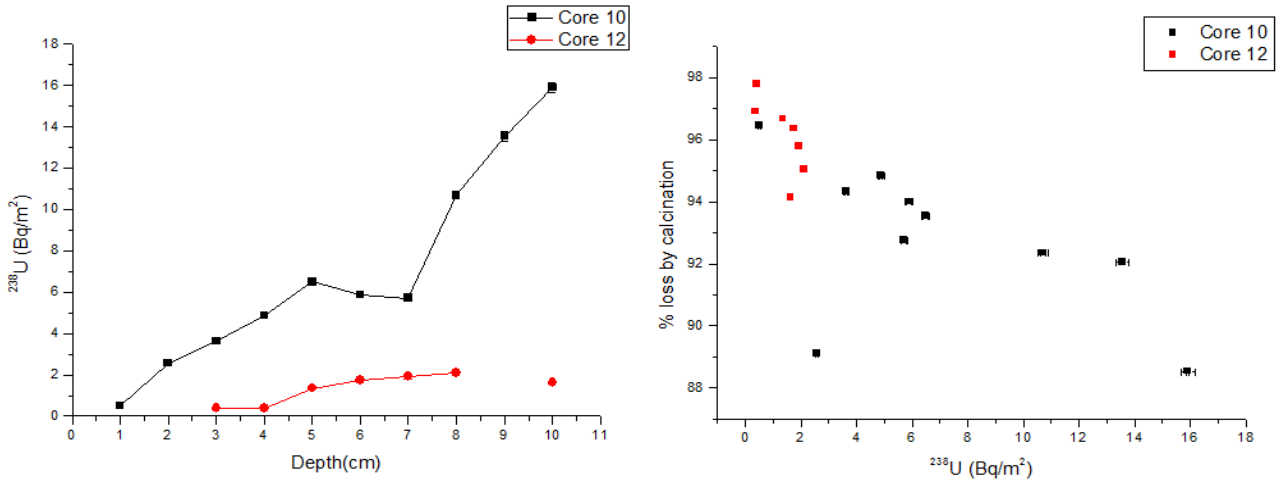

Figure 6. *Activity concentration (Bq/m^2) of ^{238}U as a function of depth (left), and content of organic matter (represented by % of loss by calcination) versus ^{238}U activity concentration (right) in two cores from Vakö mire.*

Conclusions

A fast and sequential procedure for Uranium, Plutonium and Americium determination in soil samples has been developed. The separation step is based on extraction chromatography, employing UTEVA resin in columns and DGA resin in cartridges.

Two peat bog cores of the nature reserve of Vakö Mire (south of Sweden) have been studied. One core of the area affected by the big fire of 1992, and one of the non-affected area.Plutonium isotopes experimented a redistribution in the peat bog due to the fire. The $^{239+240}$Pu activity concentration (Bq/m^2) increased at the ash layer, unlike in the non-affected core. The ^{238}Pu/$^{239+240}$Pu ratio is maintained constant due to the global fallout. The behaviour of Uranium isotopes differ from plutonium element. The fire made U easily movable via weathering conditions, and the activity concentration of the non-burnt area is much higher also due to its higher inorganic content. ^{234}U and ^{238}U isotopes were found in secular equilibrium.

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