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Short Communication

Using thin section autoradiography to detect the Uranium content of Fe/Mn soil nodules

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Abstract. The natural radioactivity of some paleosols from carbonate and siliciclastic rocks was studied using synthetic polycarbonate plates (CR39) in some Central and Southern Italian regions. The results demonstrate that Fe/Mn nodules, which are likely to form commonly in hydromorphic soils, are capturing Uranium together with rare metallic elements which have similar ionic potential.

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INTRODUCTION

Until now very few papers studied natural uranium in soils (Rosholt et al., 1966; Short et al., 1989; Morton et al., 2002). Recently, the concentration of naturally occurring radionuclides (^{238}U, ^{232}Th, K_{nat}) was measured in some "terra-rossa" type soils from carbonate rocks in Spain, Italy and Turkey using gamma-ray spectrometry at the Gran Sasso National Laboratory of INFN (Italy) by Laubenstein and Magaldi (2008). The Uranium content ranges from 1 to 5 ppm, the thorium content - from 3 to 30 ppm, and the potassium content is from 0.13% up to 1.3%. These last results indicate that soils characterised by absence or scarcity of 2:1 clay minerals are poor in uranium, whereas soils with illite–smectite as dominant minerals in the clay fraction, are noticeably richer in uranium. The radioactivity of "terra-rossa" type soils was also measured previously by Tadolini and Spizzico (1998) in Apulia (Italy) and Vaupotic et al. (2007) in south-west Slovenia and west Croatia. Our aim is to identify some micromorphological features which could have the uranium capturing capacity, assuming as negligible the content of scarcely mobile thorium. For this purpose analysis was performed using alpha autoradiography of thin sections of soil which were properly prepared from undisturbed soil samples.

METHODOLOGY

We followed a simple methodology proposed firstly by Ochmann and Solecki (2005) for some coherent rocks. Transparent plates of CR39 poly-carbonate (Pittsburg Plate Glass Co. trademark product supplied by INTERCAST EUROPE S.p.A.) have been placed on the polished thin sections of soil for a period of one year. The plates were etched for 6 hours at 60 °C temperature in a 6% NaOH solution in order to enhance the present alpha tracks. Afterwards, the plates were analysed using the image analysis software ImageJ in order to locate the tracks and to determine the track density. The track density is proportional to the concentration of U (and Th). The calculation of the uranium content assuming absence of Th was carried out as follows:

$$\textit{Alpha activity in Bq kg}^{-1} = D/0.00039$$
$$\textit{Uranium concentration in } 10^{-6}\ \textit{gg}^{-1} = D/0.0049$$

where $D = P*8*1000/S$ is the alpha particle density in the thin section of the soil, P is the number of alpha tracks

on the film/mm^2 and S is the exposure time of the CR39 film in seconds. The full section area is 25 × 40 mm = 1000 mm^2. For the microscopic inspection the field view at 100x magnification is 3 mm^2.

The constant factors for the calculation of the uranium activity was obtained after Ochmann and Solecki (2005) and some experimental data empirically.

Preliminary measurements were carried out on thin sections of B horizons from "terra-rossa" type soils (Calcic and Chromic Luvisols) originating from the carbonate plateaux of Murgia (Bari, Southern Italy) and on thin sections from B horizons of Pleistocene Luvisols of fluvial-marine terraces in the neighbourhoods of Campiglia Marittima, (Livorno Province, Italy). From each of the sets some samples were measured for their uranium content by normal gamma-ray spectrometry.

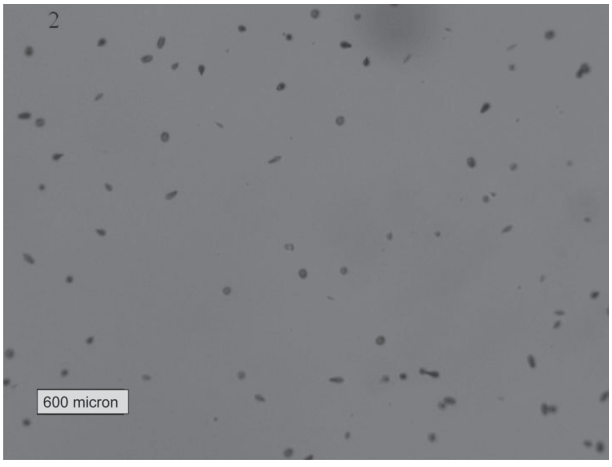

Fig. 1. Alpha track distribution in the clay matrix of a soil sample (Haplic Luvisol) with 2–2.5 × 10^{-6} gg^{-1} of uranium from Campiglia Marittima, Livorno, Italy

Fig. 2. Alpha track cluster caused by a concentric nodule from a sample of "terra rossa" soil (Calcic Luvisols) near Conversano, Murgia plateau, Bari, Italy. The approximate uranium content of the whole nodule is 0.22 Bq kg^{-1}

The measurement of the nodules was performed assuming that the emission area was equal to section area of the whole particle.

RESULTS

The results demonstrate that the method is by all means comparable with the gamma-ray spectrometry.

The exposed CR39 plates revealed three typologies of image: (A) a random track distribution for the red soil clay matrix (groundmass) (Fig. 1); (B) circular-shaped clusters of tracks mainly related to undifferentiated Fe/Mn and impregnative nodules (Fig. 2); (C) highly compacted concentration of tracks produced by small sized radioactive minerals included in larger non radioactive minerals originating from the parent material.

The types A and C were expected, whereas the images of type B should be interpreted as Uranium occurring in high concentration in the nodules present in the thin sections of undisturbed soils. As the nodules are rather rare in all sampled soils, only few nodules were examined.

DISCUSSION AND CONCLUSION

According to Short et al. (1989), indurate nodules of Fe/Mn oxides/hydroxides can irreversibly adsorb uranium despite acid leaching. This is not surprising, because Fe/Mn nodules are mainly formed by amorphous or cryptocrystalline Fe minerals as goethite, hematite, lepidocrocite, ferrihidrite, maghemite, etc., and Mn minerals as birnessite, vernadite, psilomelane, lithiophorite, etc. Moreover, the main minerals commonly include numerous elements such as Al, Ca, Mg, Ba, Li, K, Na, Zn, Cu, Co, Pb, Ni, Cr (Gallaher et al., 1973; Ross et al., 1976; Uzochkwu and Dixon, 1986; Gilkes and McKenzie, 1988; D'Amore et al., 2004; Liu et al., 2002; Negra et al., 2005), as well as Th and U (Short et al., 1989). The adsorption of the above listed elements seems to be controlled by the ionic radius (0.5 to 1.4 Å) and ionic charge (+1 to +4).

A very exhaustive review on the accumulation of microelements in Fe/Mn nodules of soil was recently given in Timofeeva (2008), Timofeeva and Golov (2007, 2010). Nevertheless, radionuclides as uranium and thorium have not been mentioned there.

Our preliminary research is suggesting that a detailed study of the distribution of uranium in soil nodules could be very interesting in order to assess both the potential radon emission from soil and derived material, and the possible uptake of radionuclides in plants. The latter is one of the main vectors for the introduction of radioactive elements into humans by the food chain (Morton et al., 2002).

The alpha track method is not particularly important for assessing the overall natural radioactivity but appears nevertheless to be a valid substitute and less expensive compared with more sophisticated devices (e.g., microprobe analysis) in order to discover the location of U (and perhaps Th and Ra) concentration. Currently, a new research project is carried out, related to collection and

analysis of the nodules of more than 50 paleosoil samples from Tuscany and Abruzzi. The expected result is to establish a possible relationship between U content and soil aspects as, for example, the type of nodules, occurrence of microelements in the matrix, the degree of soil evolution and its presumed age.

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