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A study of contaminated soils near Crucea-Botușana uranium mine (East Carpathians, Romania): metal distribution and partitioning of natural actinides with implications for vegetation uptake

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Between 1962 and 2009, National Company of Uranium - CNU, the former Romanian Rare Metals Mining Company, mined over 1,200,000 tones of pitchblende ore in the East Carpathians (Crucea-Botușana area, Bistrița Mountains). The exploration and mining facilities include 32 adits, situated between 780 and 1040 m above sea level. Radioactive waste resulted from mining are disposed next to the mining facilities. Mine dumps (32) cover an area of 364,000 square meters and consist of waste rock (rocks with sub-economic mineralization) and gangue minerals. Older dumps (18) have been already naturally reclaimed by forest vegetation, which played an important role in stabilizing the waste dump cover and in slowing down the uranium migration processes.
The soils samples have been collected from different mine dumps in the Crucea-Botușana uranium deposit, mainly from 1, 4, 5, 6, 8, 9, 1/30 and 950 mine waste galleries. Soil samples were collected from the upper part and slope at each mine dump, from the vegetation root zones.
Total uranium concentration in soils collected from Crucea-Botușana site ranged from 6.10 to 680.70 ppm, with a mean of 52.48 ppm (dry wt.). Total thorium varies between 7.70 and 115.30 ppm (dry wt.). This indicates that the adsorption of the radioactive elements by the soils is high and variable, influenced by the ore dump – sample relationship.
The sequential extraction has emphasized the fact that the uranium is associated with all the mineral fractions present in the soil samples. A great percentage of U can be found in the carbonate (21.77%), organic (15.04%) and oxides fractions (15.88%) – in accordance with the high absorbed/adsorbed properties of this element. The percentage of uranium detected in the exchangeable fraction is rather small – 2.16%. It is also to be expected that the uranium should be irreversible adsorbed by the organic matter and by the clay minerals due to its ionic radius and to its positive charge. The fact that 21.77% of the total uranium can be found in the specifically absorbed and carbonate bound fraction, indicated the important role played by the carbonates in the retention of U; one the other hand this fraction is liable to release the uranium if the pH should happen to change.
Thorium appear in high-enough concentration in the soil is scarcely available because 70.29% is present in residual fraction, and about 21.78% in the crystalline iron oxides occluded fraction and organically and secondary sulfide bound fraction. This is certainly due to the fact that this naturally occurring radionuclide can be associated with relatively insoluble mineral phases like alumino-silicates and refractory oxides. Its association with the organic matter (10.93%) suggests that it can form soluble organic complexes that can facilitate its removal by the stream waters.
Grounded on these results, we were able to prove that the examined mine dumps can represent an impact on the environment, which constitute an argument in favor of the initiation of a program of remedying the quality of the environment from this mining zone. Although from our research it resulted that the natural actinides does not concentrate in the exchangeable fraction (Th) or it concentrates very little in it (U), the isolation of the mineral fraction of soil rich in U and Th helps us in the future identification of the links between the bioavailability and the pedogenesis, connections which control the cycle of the radioactive metals.