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Distribution of Natural Radionuclides, Rare Earth Elements, Metals and Metalloids in a Phosphogypsum Stockpile

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Abstract

A first detailed study of phosphogypsum (PG) from a stockpile in Barreiro (Portugal) was performed aiming for a better characterization of this industrial waste deposit, considering its enhanced content in natural radionuclides and toxic metals, making it a potential contamination source to the Tejo estuary. Whole samples and aggregates of these wastes resulting from phosphate industries were analysed by neutron activation, gamma-spectrometry, X-ray diffraction and scanning electron microscopy. This work clearly shows that a significant chemical heterogeneity in the PG occurs due to the existence of aggregates with different compositions randomly distributed. Among these aggregates, the dark grey ones have high concentrations of Sc, Cr, Zn, Ga, Ba, REE, Ta, W, Th and U and the highest concentrations of 226Ra and 210Pb. The separation of these dark aggregates prior to any application of the PG would lead to a safer reuse of these wastes. The chemical patterns, including the REE distribution with a significant negative Ce anomaly, found in PG, are certainly related with the geochemical signatures of the phosphate rocks used as raw material. This PG stockpile may play a significant role as a radioactive source in the Tejo estuarine environment.

The environmental impact of massive amounts of phosphogypsum (PG) produced as a waste from the phosphate industry is a worldwide concern due to its content in toxic elements and natural radionuclides. He application of PG, for instance as raw material in building and soil enrichment stuﬀ ,or the alternative decision of letting the PG stockpiles remain in place with some further reasonable remediation, demands a characterization of its properties. He PG, the insoluble by-product resulting from the sulfuric acid attack (wet chemical treatment) of the phosphate rock to produce phosphoric acid, has associated high activities of natural radionuclides from U and H series [1,2] and is designated as a TENORM. His by-product is usually deposited in dumps and stacks of very large volume and surface representing a great danger to the environment [3]. During the phosphoric acid production, the radioactive equilibrium between the 238U and its daughters (226Ra, 210Pb) is broken and each radionuclide is distributed diﬀerently depending on its solubility. Most of the radium is transferred to the PG and the major amounts of the uranium partition into the phosphoric acid [4,5]. PG stockpiles contain two main types of mineral phases: a) the original materials which were not modified during acidification and b) minerals formed during acidification. He majority of minerals (up to 95%) belong to the second group, which is formed mainly of bassanite and gypsum [3,6]. High contents of sodium fluorosilicate have also been reported [7]. In addition, other minerals are usually present such as anhydrite, calcite, quartz, mica, phyllosilicates and phosphate-bearing phases [6,8]. High concentrations of Rare Earth Elements (REE) may be also expected, since some phosphate minerals are usually rich in these elements [9]. 6ite-specific contamination levels of REE and natural radionuclides in an estuarine ecosystem related to phosphate industry were reported by.

For the study-Distribution of Natural Radionuclides, Rare Earth Elements, Metals and Metalloids in a Phosphogypsum Stockpile the PG whole samples from Barreiro stockpile (Portugal) show significant variation of the chemical elements and radionuclides concentrations, probably due to the occurrence of diﬀerent proportions of aggregates. Gypsum is associated mainly to brushite and bassanite. On the surface of gypsum, several porous particles with donut shape were found which correspond to secondary phosphates rich in fluorine (brushite). Among the aggregates found in the PG, the dark grey ones, rich in bassanite with traces of Ti oxide, have high amounts of most of the chemical elements studied, particularly Sc, Cr, Zn, Ga, Ba, REE, Ta, W, H and U. The REE patterns with a significant negative Ce anomaly found in PG are certainly inherited from the parent rock (phosphate rock). He majority of the radioactivity found in the PG whole samples and in the dark grey aggregates is mainly due 226Ra and 210Pb found in the PG stockpile are certainly associated with the presence of these dark grey aggregates. The previous separation of these aggregates before any application of the PG would contribute to a safer reuse of these wastes. The results obtained in this work show how radionuclides and metals can be distributed in fine particles and can be a potential radioactive source in the Tejo estuary area.

Keywords: Gamma spectrometry; Instrumental Neutron Activation Analysis (INNA); Phosphogypsum; Radioactivity; Radionuclides; Rare earth elements; Trace elements; Technologically Enhanced Naturally Occurring Radioactive Materials (TENORM)