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Natural radionuclides in plants, soils and sediments affected by U-rich coal mining activities in Brazil

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ABSTRACT

Mining activities can increase the mobility of metals by accelerating the dissolution and leaching of minerals from the rocks and tailing piles to the environment and, consequently, their availability for plants and subsequent transfer to the food chain. The weathering of minerals and the disposal of coal waste in tailing piles can accelerate the generation of acid mine drainage (AMD), which is responsible for the higher dissolution of metals in mining areas. In this context, the behavior of U, Th and K in soils and sediment, and the transfer factor (TF) of ^{238}U, ^{234}U and ^{210}Po for soybean, wheat, pine and eucalyptus cultivated around a coal mine in southern Brazil was evaluated. Alpha and gamma spectrometry were used for the measurements of the activity concentration of the radioelements. ^{210}Po was the radionuclide that is most accumulated in the plants, especially in the leaves. When comparing the plant species, pine showed the highest TF values for ^{234}U (0.311 ± 0.420) for leaves, while eucalyptus showed the highest TF for ^{238}U (0.344 ± 0.414) for leaves. In general, TF were higher for the leaves of soybean and wheat when compared to the grains, and grains of wheat showed higher TF for ^{210}Po and ^{238}U than grains of soybean. Deviations from the natural U isotopic ratio were recorded at all investigated areas, indicating possible industrial and mining sources of U for the vegetables. A safety assessment of transport routes and accumulation of radionuclides in soils with a potential for cultivation is important, mainly in tropical areas contaminated with solid waste and effluents from mines and industry.

1. Introduction

Natural radioelements are part of the chemistry of soils and waters, resulting in radiological dose for living organisms. Mining activities contribute to increase this dose as they accelerate the leaching of elements and minerals from rocks to the environment (USEPA, 1995; Baik et al., 2003; Flues et al., 2006; Fungaro and Izidoro, 2006).

In coal mines, the disposal of waste in tailing piles and ore exposure to weathering conditions can accelerate the formation of acid mine drainage (AMD). This acid effluent accelerates the leaching of toxic elements in coal mining areas and is generated when sulfide minerals (e.g. pyrite, Fe_2S) are oxidized due to exposition to atmospheric oxygen, promoting a reduction of the pH level and the solubilization of metals into the aqueous medium (Berghorn and Hunzeker, 2001; Mkandawire, 2013). In addition to the release of radionuclides by discharge by tailings seepage waters, emissions of radionuclides by dry or wet deposition of re-suspended radioactive material from tailings piles may lead to elevated levels in the nearby soil, where they can be adsorbed, retained and taken up by plants (Planinsek et al., 2016). Particulate matter and radionuclides released to the atmosphere from coal-fired power plants can also return to the ground as wet or dry deposition, contributing to the radiation dose exposure (Papp et al., 2002).

The occurrence of uranium (U) in coal has been investigated for more than a half of a century and the major concern is related to the use of coal for electricity generation (Baxter, 1993; Arbuzov et al., 2012). In addition to common contaminants related to sulfur- and metal-bearing minerals, the particulate matter and effluents

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generated during the coal fired-power plant and coal mining activities may contain ^{226}Ra, ^{210}Pb and ^{238}U (UNSCEAR, 2010). As example, coal contains approximately 12–24 Bq kg^{-1} of ^{238}U and 12–17 Bq kg^{-1} of ^{232}Th (UNSCEAR, 2010).

In solutions, adsorption is an important mechanism that controls U activity concentration (Prikryl et al., 2001). The chemical behavior of U depends on several characteristics, e.g. pH, Eh, concentration and type of organic and inorganic ligands, presence of chelating agents, speciation, mixing rate and movement of the water, reactions of complexation, precipitation, adsorption and desorption. However, since to obtain and interpret all this information is complex, pH and Eh are the most common parameters used in investigations about U mobilization processes and explain most variation of concentration of U in waters (Langmuir, 1978; Giblin et al., 1981).

In this context, to evaluate radiological impacts due to extraction, processing and burning of coal, some plants have been used for the assessment of soil contamination. The most commonly index used to estimate the radionuclides transport and their accumulation in vegetables is the transfer factor (TF), defined as the ratio between the concentration of the element in plants (A_m = activity concentration of the radionuclide in plant dry matter) and in the soil (A_s = activity concentration of the investigated element in dry soil). This is alternatively known as the concentration ratio (CR). The TF describes the concentration of the element that can be absorbed by a plant from its substrate under equilibrium conditions, assuming that the accumulation is directly proportional to the concentration of the element in the soil (Sheppard and Sheppard, 1985).

An understanding of the mobility of natural radionuclides in soils and their TFs to different plants requires knowing their interactions with soils (Shtangeeva, 2010). The uptake of trace elements by biota can be affected by the electrical conductivity (EC), cation exchange capacity (CEC), total organic carbon (TOC), nutrients, mineralogy and climate, in addition to (bio)chemical processes such as complexation, precipitation and redox reactions (Sohlenius et al., 2013; Gupta et al., 2014; Zhao et al., 2016). However, in about 50% of the investigations focusing on the TF, these characteristics were not considered (Vandenhove et al., 2009), which limits our understanding on the metal uptake by the biota. An assessment of the radionuclide distributions in the soil–plant system may be also rather complicated because there is little information related to the rate of radionuclides uptake and storage by different parts of plant species (Shtangeeva, 2010).

Until the present, most of the studies of TF were in temperate areas, where climatic conditions, weathering processes, nutrients cycling and metal uptake by living organisms differ significantly from those in tropical areas (IAEA, 2010). Few systematic investigations have been done in cultivated tropical soils affected by coal mining, especially in Brazil. According to the Food and Agricultural Organization (FAO, 2012), Brazil is the 10th largest vegetable producer in the world and the 2nd largest soybean producer. Paraná State is the 2nd largest producer of this legume in Brazil, and also has the 3rd largest coal reserve in the country that is associated with a U deposit.

Previous studies indicated high levels of radionuclides and trace elements in soils from Southern Brazil associated with AMD (Flues et al., 2006; Campaner et al., 2014). This is the case of Figueira city, Paraná State, Brazil, where the coal mined there exhibits high content of pyrite (MINEROPAR, 2001) that may accelerate the environmental problems associated with AMD generation. The coal has also elevated contents of U and thorium (Th) (Fernandes et al., 1997). Flues et al. (2006) reported ^{238}U and ^{232}Th concentrations ranging from 813 Bq kg^{-1} to 2609 Bq kg^{-1} and from 22 Bq kg^{-1} to 40 Bq kg^{-1}, respectively, in coal sampled in that area.

Some researchers in Brazil investigated the absorption of radionuclides by plants from soil (Vasconcellos et al., 1987; Santos et al., 1993, 2002; Lauria et al., 1994; Mazzilli et al., 2012), but no investigation was performed in areas affected by coal mining. Radionuclide analysis in vegetables cropped near coal mining areas in southern Brazil, especially for those plants used for human consumption, could improve the radiological risk assessment. In this context, this paper aims 1) to evaluate the influence of a coal-fired power plant and coal mining activities on the characteristics of nearby soils and sediments and 2) to investigate the uptake of radionuclides by plants (soybean and wheat – leaves and grains, pine and eucalyptus – leaves and bark) in an impacted tropical environment.

2. Material and methods

2.1. General features of the study area

The city of Figueira is located in the northeastern of Paraná State in Brazil (Fig. 1). The climate is divided into a rainy season (October to March) and a dry season (April to September). The predominant soils are shallow, acidic, dystrophic, with a poor drainage and a high saturation of exchangeable aluminum (Morrone and Daemon, 1985). The soils around the mining area are used for agricultural activities. Major crops in the area are soybean and corn, for which the grains are exported mainly to the US, China and Europe. Waters from the Laranjinha river (the main watercourse in the region that flows into the Paranapanema river) and the Pedras stream (its tributary where the mining effluents used to be discharged) are used for agricultural irrigation.

In the region, the outcropping stratigraphic units belong to the Phanerozoic Paraná Sedimentary Basin and evolved along the South American Platform that spreads in seven states in Brazil as well as in Uruguay, Argentina and Paraguay. The units with Permian age belong to Itararé, Guatá (Rio Bonito and Palermo formations) and Passa Dois (Irati, Sierra Alta and Teresina formations) groups (Shuqair, 2002; Bizzi et al., 2003). The coal deposit is hosted in Rio Bonito Formation and U mineralization has also been observed within Permian coal seams from Rio Bonito Formation, occurring along the interface from the coal-bearing horizon, sandstones and carbonaceous siltstones (Medeiros and Thomaz Filho, 1973).

In the investigated area, the coal horizon thickness varies between 0.50 and 0.65 m and it is located at a variable depth of 38–75 m (ANEEL, 2011). Chemical analysis (moisture content = 6%, volatile matter = 28.8%, fixed carbon = 32.5%, ash content = 38.7%, sulfur content = 4–12%, calorific value = 4300 kcal kg^{-1}) classify the coal as high volatile bituminous (Shuqair, 2002).

Coal is nowadays extracted from an underground mine (named 08), in operation since 2014, although mining activities have occurred since the 1950s and left important impact associated with water and soil quality. The water used for coal washing after its crushing has been recycled in a closed system and physically treated in a pond. Before 2008, the effluent had been discharged into a stream near the mine. This pond also receives the acid effluent generated in the tailing piles. However, the effluents may reach the soil, groundwater and surface water, modifying their chemical composition and contributing to the surface water, groundwater and soil contamination (Galhardi and Bonotto, 2016).

The coal waste was disposed in two main tailing piles: one composed of ‘sterile’ waste (older than 50 years) that was not protected with waterproofing and CaO addition to control pH and another that accumulates material with high pyrite concentration, which has waterproofing and CaO was added to control pH. The tailing piles are located in an outcrop area of Palermo Formation, composed by clays and siltstones, interbedded with sandy layers