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The influence of particle size on radionuclide activity concentrations in Tejo River sediments

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ABSTRACT

Sediment samples from Tejo River were analyzed for ^{228}Ra, ^{226}Ra, ^{137}Cs and ^{40}K by HPGe gamma spectrometry. The activity concentration data were statistically analyzed. The activity concentrations values were in the range of about two orders of magnitude for each radionuclide. The influence of the particle size on the radionuclide concentrations was observed. The different environmental origins of the radionuclides ^{228}Ra, ^{226}Ra, ^{137}Cs and ^{40}K, in the sediments were demonstrated through correlation analysis. Cluster analysis showed a close relationship between ^{228}Ra and ^{226}Ra and a different behavior for ^{40}K. The data obtained in this study provides useful information on the background radioactivity of the studied area and can be further used for radiological mapping of the Tejo River.

1. Introduction

Radioactivity occurs naturally in the environment. The radionuclide content of a river depends on mineralogical features in the catchment area and the chemistry of the river as a whole. The major sources of natural radionuclides in sediments have different possible origins. These include weathering and recycling of terrestrial minerals and rocks (igneous or metamorphic) containing ^{40}K and radionuclides of the uranium and thorium radioactive series, rainfall and other depositional phenomena such as gravitational settling and precipitation. In stream sediments U and Th may be found incorporated into the existing minerals or they may be adsorbed directly from river water onto clay minerals or organic debris. Occasionally, U may be removed from river water to sediments directly if reducing bottom conditions exist (El-Gamal et al., 2007).

Artificial radionuclides can be introduced into rivers for direct and indirect inputs. Directly through the aqueous discharges from nuclear installations and indirectly from wash-off of land deposited activity within the river catchment following nuclear weapon testing or nuclear accidents. Most of the radioactivity deposited on surface soils, depending on the radionuclide's geochemistry, is washed out by rains and drained in to the rivers. After reaching the river ecosystem, radionuclides may be transferred through the water-sediment-biota pathways (Ajayi and Kuforiji, 2001; Ajayi, 2008) to humans, by using the river water as drinking water or for irrigation and by the consumption of contaminated fish.

River sediments, consisting of mineral particles with different sizes, are considered long term reliable indicators of river pollution by radionuclides because water pollution components are deposited in the sediments. Long-term radioactive pollution may accumulate and whenever the sediments are re-suspended the radionuclides re-enter the sediment-biota chain (Bikit et al., 2006). Therefore, the knowledge of the concentrations and distribution of the radionuclides in the river sediments are of great interest since it provides useful information on the background and on the temporal changes in radionuclide activity concentrations within the river. In the framework of the environmental radioactivity survey performed in Portugal (Madruga, 2008) several components of the Tejo River, including sediments, have been monitored (Madruga et al., 2009). The Tejo River originating in Spain runs through Portugal on its way into the Atlantic Ocean. The Tejo River receives discharges from three Spanish Nuclear Power Plants (NPP) with the Almaraz NPP located nearest the Portuguese border (about 120 km). In Portugal, the sedimentation load and natural distribution of sediments are affected by two dams (Fratel and Belver) (Carreiro and Sequeira, 1998).

The main objective of this study is to assess, through statistical analysis, the activity distribution of natural (^{228}Ra, ^{226}Ra, ^{40}K) and anthropogenic (^{137}Cs) radionuclides in sediments collected in Tejo

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River and the influence of the sediment particle sizes on radionuclides concentrations. Besides, the knowledge of radionuclides concentrations and distribution in Tejo River sediments can be useful for the radiological mapping of the studied area.

2. Material and methods

2.1. Sampling and sample preparation

Surface sediment samples, around 4 kg each, were collected monthly (from years 2001–2009) in Tejo River at two selected locations: Vila Velha de Ródão (VVR) near the Spanish border and upstream of Fratel and Belver dams, and Valada (V) downstream of these dams (Fig. 1).

The samples were collected with a metal sampler (Berthois cone) and stored into labeled plastic boxes. At the laboratory the samples were homogenized, dried at room temperature, crushed, sieved and fractions lower than 1000 μm (*A*-bulk sediment), between 250 μm and 63 μm (*B*-sand) and lower than 63 μm (*C*-silt/clay) were used for analysis. Representative aliquots of each sieved sample were transferred to 180 cm^3 or 10 cm^3 plastic containers (depending on the amount available), closed, sealed and left aside for about one month to ensure that secular equilibrium is reached between radium isotopes and its short lived progeny in uranium and thorium radionuclide series.

2.2. Gamma spectrometry analysis

Samples were counted for 15 h and analyzed for ^{228}Ra (determined through ^{228}Ac), ^{226}Ra (determined through ^{214}Pb and ^{214}Bi daughters), ^{137}Cs and ^{40}K by gamma spectrometry.

The measurement system is composed by HPGe detectors and standard analog electronics. The detectors were shielded from environmental background, by using lead shields with copper and tin lining. The efficiency calibration was performed using NIST-traceable multinuclide radioactive standards, covering the energy range 46.5–1836 keV, customized to reproduce the exact geometries of the samples in a water-equivalent epoxy resin matrix by Analytics, Incorporated. The density correction was carried out through GESPECOR version 4.2 software® (Sima et al., 2001) and Genie 2000® (Canberra) was used for data acquisition and analysis. The stability of the system (activity, FWHM, centroid) was

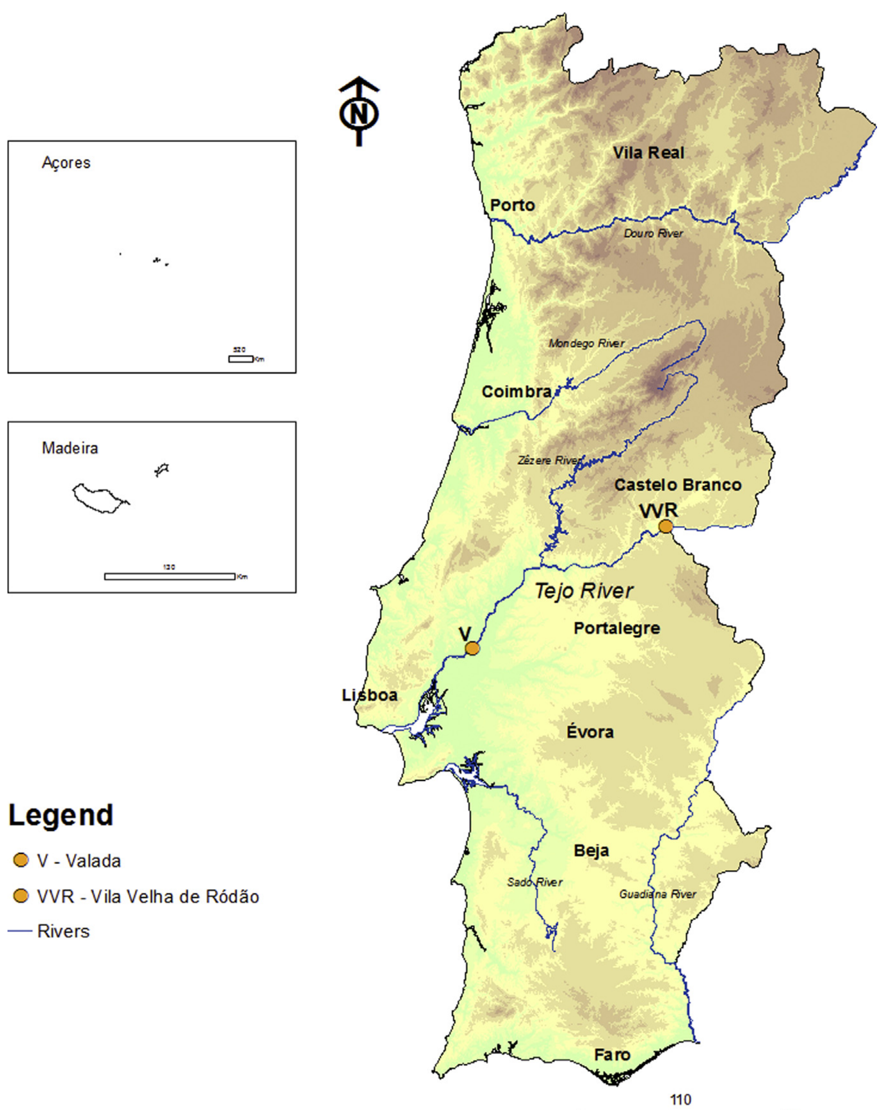

Fig. 1. Sampling locations in Tejo River (Portugal).