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Activity concentration and spatial distribution of radionuclides in marine sediments close to the estuary of Shatt al-Arab/Arvand Rud River, the Gulf

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ABSTRACT

Tigris and Euphrates rivers both emerge in eastern Turkey and cross Syria and Iraq. They unite to Shatt al-Arab/Arvand Rud River and discharge in Arabic/Persian Gulf. The activity concentration of natural and anthropogenic radionuclides was measured during the August of 2011 in a number of surficial sediment samples collected from the seabed along an almost straight line beginning near the estuary mouth and extending seaward. The results exhibited low activity concentration levels and an almost homogeneous spatial distribution except locations where sediment of biogenic origin, poor in radionuclides, dilute their concentrations. Dose rates absorbed by reference marine biota were calculated by the ERICA Assessment Tool considering the contribution of 40 K. The results revealed a relatively low impact of 40 K mainly to species living in, on and close to the seabed. Also, statistical association of radionuclides with selected stable elements (Ca, Ba and Sr) did not indicate presence of by-products related with oil and gas exploitation and transportation activities. Moreover, a semi-empirical sedimentology model applied to reproduce seabed granulometric facies based entirely on radionuclides activity concentrations.

1. Introduction

Shatt al-Arab in Arabic (meaning “Stream of the Arabs”) or Arvand Rud in Persian (meaning “Swift River”) is the river formed by the confluence of Tigris and Euphrates, the two main rivers of Mesopotamia. Both Tigris and Euphrates rivers emerge in eastern Turkey and cross Syria and Iraq before they unite near the Iraqi town of al-Qurnah. The river's length is approximately 200 km and it discharges into the Arabic or Persian Gulf (referred also as the Gulf). The river is navigable for ocean going vessels as far as Basra -Iraq's main port- and it constitutes the border between Iraq and Iran. The Karun River is the main tributary that joins the waterway from the side of Iran.

The estuary covers approximately (10–20) $\times$ 10^3 km^2 containing sandbars, marshes, lagoons, ox-bow lakes and tidal flats. This environment accumulates the impact of two full-scale wars and other non-military anthropogenic pressures. Before the Gulf war (1991), Iran and Iraq disputes for the control of the waterway ended by an eight-year war (1980–88) between the two countries, including extensive coastal attacks and use of chemical weapons. During the Gulf war, about 11 million barrels of oil were deliberately spilled directly into the sea. A six-month fire of 700 Kuwait oil wells resulted in widespread airborne fallout affecting the water by oil combustion products. Weapons containing depleted uranium were used resulting to the release of fine aerosol containing poorly soluble uranium oxides. On the other hand, non-military activities still pose threats for the environment as the area is exposed to the production and transportation of one-third of the world's oil consumption, the river water is exploited for continuous irrigation purposes and also receives wastes from urban centres, power plants and industries. Along with the anthropogenic impact, the natural stresses of high seawater temperature and salinity lead to a steady deterioration of the conditions of the marine environment of the Gulf (Sheppard et al., 2010).

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In coastal areas, marine sediment may accumulate several potential hazardous substances (toxic metals, organic pollutants, radionuclides etc.) due to deliberate or accidental anthropogenic releases and natural input processes as well. Potential accumulation of radionuclides in the sediment of Shatt al-Arab estuary could be a result of several anthropogenic activities mainly related with oil and gas exploration and extraction, oil-treatment and oil transportation activities (Al-Saad and Salman, 2012). Also, physical processes and function may further contribute to their transportation and accumulation. Natural radionuclides participate as trace elements into the lattice of minerals, so the abrasion and stream erosion of riverbank rocks become a physical process of their transportation into the estuary and subsequent accumulation in coastal sediment. Both anthropogenic and natural accumulation processes under several circumstances (e.g. deposition in coastal regions of tailings rich in natural radionuclides, coastal areas with high level of natural radioactivity) may result in enhanced levels of activity concentrations of natural radionuclides in the coastal sediment (Bahari et al., 2007; Mohanty et al., 2004).

Radionuclide activity concentration measurements in the marine sediment mainly have the purpose to estimate the radioecological condition of a marine area. Additionally, they have been proved as valuable tracing techniques for investigation of several oceanographic and geophysical phenomena. For instance, activity concentrations of thorium ^{232}Th, radium ^{226}Ra, potassium ^{40}K, lead ^{210}Pb and caesium ^{137}Cs in marine sediment have been widely used worldwide to reproduce granulometric facies maps of bays (Ligero et al., 2001, 2009) and lagoons (Tsabaris et al., 2007), to examine the results of high ship traffic in sedimentological processes of harbours (Papaefthymiou et al., 2007), to radiometric date lakes sediment cores (Arnaud et al., 2006), to estimate the rate of sedimentation (Tsabaris et al., 2012) and the migration of radionuclides in the sediment column (Tsabaris et al., 2015), to investigate sediment mobilization and evaluate the sediment budget of river estuaries (Walling, 2003) and more (Jones, 2001).

The aim of this work is to provide baseline information regarding the activity concentration levels of natural (^{226}Ra, ^{224}Ra, ^{228}Ra, ^{40}K and ^{235}U) and artificial (^{137}Cs) gamma-ray emitter radionuclides in the marine sediment of the estuary as well as to estimate their radiological impact on reference marine species living in the estuary. The significance of these tasks is considered high due to the extensive activities related with oil and gas exploitation and transportation that are held in this region, leading to by-products (e.g. production waters, scale, and sludge) containing enhanced activity concentrations (OGP, 2008) that may have been released in the environment. The significance of this baseline information relies also on the fact that in the near future more countries in the area will construct and operate nuclear energy facilities. So, an extensive database created prior to their operation may play an important role in future radiological evaluations. Another objective of the work is an assessment regarding the presence of by-products from the activities related with oil and gas industry in the sediment of the estuary. This task is based on data of selected major and minor elements and statistical analysis. Moreover, the spatial distribution of the radionuclides is examined and its behaviour is explained considering the origin of the sediment. Also, a semi-empirical model proposed by Ligero et al. (2001, 2009) was applied to reproduce the granulometric facies of the seabed based exclusively on activity concentrations.

2. Study area and field work

During the period 25–30 August of 2011, in the framework of *MARITECH* marine project, 52 surficial sediment samples were collected by divers' team in the coastal area of the estuary south of the Iraqi port of Al-Faw. The sediment sampling was realized by simple grabbing techniques so each sample was a disturbed mixture of sediment belonging to the upper seabed layers (approximately 3–5 cm). The sampling locations are depicted in Fig. 1 and their coordinates are presented in Table 1. The sampling was accomplished along an almost strait line starting close to the mouth of the estuary towards to the sea. The total distance between the two edges was approximately 18 km. The sampling locations depths varied from 3 to 15 m. On board, the samples were recorded and characterized according their colour, texture and shell fragments content. In most of the samples, many shell fragments were found and were removed. Finally, the samples were water tightly packed and transported to the laboratory for further treatment.

3. Materials and methods

The samples were appropriately prepared for activity concentration measurements of radionuclides by gamma-ray spectroscopy and measurements of major/minor elements concentration by X-ray fluorescence analysis. In a subgroup of 18 selected samples particle size analysis was also performed.

3.1. Radioactivity measurements

For gamma-ray spectrometry, the sediment samples were pretreated according to a standard preparation methodology following IAEA guidelines (IAEA, 2003). Stones, shells, algae and grains greater than 2 mm were removed from the sample material and the weight of the remaining sediment was recorded. Then, the material was dried until constant weight (in total for 48 h) and the weight of the dry sample was recorded. The dry material was homogenized and pulverized in a twin swinging motorized mill with agate mortar and balls. After this process the sediment material, dried and homogenized, was closed in small plastic containers of cylindrical shape with volume 64 cm^3. The containers were kept airtightly sealed to prevent any radon exchange with the atmospheric air and the measurements commenced after a period of 21 days so secular radioactive equilibrium between radium ^{226}Ra and radon ^{222}Rn progenies as well as thorium ^{228}Th and radium ^{224}Ra (and its progenies) was achieved.

The activity concentration measurements were accomplished with a High Purity Germanium Detector (Coaxial n-type HPGe Detector System by ORTEC) with nominal relative efficiency of 50% and resolution of 1.85 keV at 1.33 MeV. The followed methodology for energy and full energy photopeak efficiency (fepe) calibration procedures according to the voluminous geometry of the sediment samples is described in detail elsewhere (Tsabaris et al., 2007, 2012). Briefly, a diluted liquid radioactive source of know activity concentration, comprising ^{152}Eu (93%) and ^{154}Eu (7%), is dripped into inert material (talcum powder) enclosed in the same type of plastic container that used for the measurements. This calibration process reproduces the voluminous geometry of the samples and the use of talcum powder imitates the density and porosity of the pulverized sediment. Also, true coincidence summing corrections were obtained by the EFFTRAN application (Vidmar et al., 2011). This Monte Carlo based application, takes into account gamma – gamma and gamma – X-ray coincidences regarding the geometry and the characteristics of the source and the detector. It provides correction factors for each gamma-ray with values greater than 1 in case of summing out and less than 1 in case of summing in (crossover transition) effects (Gilmore, 2008). Using the described methodology a number of experimentally derived efficiency coefficient values were calculated in the energy range of 121.8–1408 keV. The estimation of efficiency coefficient in the whole energy range 121.8–1460.8 keV was achieved by fitting the