Determination of natural radioactivity in Euphrates river

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Levels of naturally occurring radionuclides (radium isotopes, U isotopes, $^{210}$Po and $^{210}$Pb) in water, sediments and biota samples collected from Euphrates river during the 1999–2000 period have been determined. Results have shown that the water contained relatively high levels of $^{226}$Ra; the largest value of 1150 mBq$^{-1}$ was observed. These relatively high levels of $^{226}$Ra, which is one of the main radioactive contaminants in the oil industry, may be due to past discharges of production water from the oil fields situated near the river banks. $^{226}$Ra/$^{238}$U activity ratio was found to be more than unity in all water samples varying between 13 and 242. In addition, the results of sediment analyses have also shown lower values for $^{226}$Ra/$^{238}$Ra activity ratio than unity in those samples collected nearby the oil fields. Moreover, concentrations of other naturally occurring radionuclides such as uranium isotopes, $^{210}$Po and $^{210}$Pb for most samples (water, sediments and biota) were found to be within the natural levels and in agreement with those values reported for other local and international studies. Only mussel species were found to contain high levels of $^{210}$Po, about 1335 Bq$^{-1}$ dry mass was observed in Anodonta sp species. However, the results of this study can be considered a baseline for monitoring of future changes. A regional research project (including Turkey, Syria and Iraq) to study this river (from the Anatolia Mountains to the Arabian Gulf) is necessary to determine the impact of all potential sources of contaminants.

Introduction

In 1990, a National Program for Monitoring Radioactivity in the Syrian Environment has been initiated. The main objectives of the program were to provide an early warning system for an emergency abatement system, record the environmental radioactivity data in all Syrian land, so to establish the required background data in the country, monitor radionuclides pathways following a release of radioactive materials due to accidents, cross/or in the boarders and to ensure that national and international regulations, related to discharges of radionuclides into the environment being adopted. Therefore, gamma-radiation exposures and natural radioactivity measurements in different types of water, soil, air, food and crops from different parts of the country have been evaluated. According to these surveys, most of the potential sources of natural and artificial radionuclides were identified. The impact of phosphate industry, which is considered to be the potential source of natural radionuclides, on the Syrian environment has been evaluated by performing several studies in most important sites including mines, phosphoric acid production plants and the export platform at Tartous port on the Mediterranean sea. Another identified potential source for enhanced natural radioactivity in the Syrian environment is the oil industry. Naturally occurring radioactive materials (NORM) resulting from the $^{232}$Th and $^{238}$U-series can be concentrated and accumulated in tubing and surface equipment in the form of scale and sludge. One of the main radioactive wastes containing NORM is the production water, which contains mainly radium isotopes and their daughters. Uncontrolled disposal of this water could lead to environmental pollution. This type of pollution has been seen in the Syrian oil fields, which are located at several sites situated approximately 700 km to the northeast of Damascus city near to Euphrates, Tigris and Al-Khabour rivers basin. In the past, unlined evaporation large lagoons have been used to hold water production, which have been lost by evaporation. Some run-off channels (one of these is about 50 km long) had been created to allow the water to run-off into the surrounding environment, and a significant degree of sub-surface contamination has occurred.

The aim of the present work was to determine the natural radioactivity in Euphrates river for establishing the background base line levels for future monitoring.

Experimental

Area of study

Euphrates river is one of the most famous rivers in the world. It rises in the high mountain of northern Anatolia and flows down through Turkey, Syria and Iraq. Euphrates river (860 km long in the Syrian land) is the main source for water supply in the northern and eastern areas of Syria for drinking, domestic uses and irrigation.

Sampling

Sediments of 1 kg, river water of 30 liter, fish, aquatic plants and other mussel samples were collected during the 1999 and 2000 period from eight sampling locations (Fig. 1). Biota samples were collected by hand, placed in plastic bags and transported to the laboratory in an ice box. Biota samples were then washed with

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distilled water several times to remove all sand and dried in the oven at 90 °C for 24 to 48 hours to reach a constant mass. The samples were ground, homogenized and sieved to pass through an 560 μm sieve. The water samples were filtered through filter with a pore size of 45 μm and acidified by adding concentrated hydrochloric acid (5 ml per one liter of water). The pH value was determined to be less than 2.

Radiochemical analysis

The samples were subjected to radiochemical analysis of 210Po. 0.5 g of each sample was spiked with 0.2 Bq of 208Po as a yield tracer and the sample was digested using concentrated nitric acid for at least 24 hours. When the solution became clear, concentrated hydrochloric acid was added and the solution was gently evaporated near to dryness. The sample was then dissolved in 100 ml of 0.5 mol·l⁻¹ hydrochloric acid and the resulting solution was heated to 80 °C. 210Po was then spontaneously plated onto a rotating silver disc after reduction of iron with ascorbic acid. Alpha-counting of 208Po (5.15 MeV) and 210Po (5.3 MeV) was done using an alpha-spectrometer (OASIS, Oxford) with passive ion implanted silicon detector (with an active area of 300 mm², background counts per day 3.6 and the minimum depletion thickness 100 μm). 210Pb activity was obtained by measuring the growth of 210Po activity after storage of the solution for at least six months.

For water samples, 210Pb and 210Po were precipitated from 4 liter volume by MnO₂. The precipitate was dissolved in 1.5 mol·l⁻¹ HCl and processed as described for solid samples.

The determination of uranium isotopes (234U and 238U) in all samples was performed using standard alpha-spectrometry where uranium was electroplated on a stainless steel disc from the solution after chemical purification using anion-exchanger Bio Rad AG 1 x4. The electroplated source was measured using the above mentioned alpha-spectrometer and 236U was used as radioactive tracer for the determination chemical recovery.

A liquid scintillation counter (Packard Tri-carb 1500) was used to determine 226Ra level in water samples using the method described by BLACKBURN and AL-MASRI. Radium was separated as BaRa chloride which was dissolved in 1 ml of distilled water and mixed with the liquid scintillation cocktail (Ultima Gold) for counting.

Sediment and biota samples were measured by gamma-ray spectrometry (Eurysis Systems) using a high resolution (1.85 keV at 1.33 MeV), high relative efficiency (80%) low background HPGe detector to determine natural gamma-ray emitters, 40K and radium isotopes (224Ra, 226Ra, 228Ra). About 40 g of each solid sample was filled in a special container and stored for one month for counting and 226Ra, 224Ra and 228Ra were then determined by measuring their gamma-emitting daughters, 214Pb (351.9 keV), 214Bi (609 keV), 212Pb (238 keV), 212Bi (727.3 keV) and 228Ac (911.1 keV).

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Fig. 1. Sampling points