Investigation of the radiological impact on the coastal environment surrounding a fertilizer plant

O. El Samad [a,*], M. Aoun [a], B. Nsouli [a], G. Khalaf [b], M. Hamze [c]

[a] Lebanese Atomic Energy Commission, National Council for Scientific Research, P.O. Box: 11-8281, Beirut, Lebanon
[b] Marine Research Centre, National Council for Scientific Research, Lebanon
[c] National Council for Scientific Research – CNRS, Lebanon

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ABSTRACT

This investigation was carried out in order to assess the marine environmental radioactive pollution and the radiological impact caused by a large production plant of phosphate fertilizer, located in the Lebanese coastal zone. Natural radionuclides (^{238}U, ^{235}U, ^{232}Th, ^{226}Ra, ^{210}Po, ^{210}Pb, ^{40}K) and anthropogenic ^{137}Cs were measured by alpha and gamma spectrometry in seawater, sediment, biota and coastal soil samples collected from the area impacted by this industry. The limited environmental monitoring program within 2 km of the plant indicates localized contamination with radionuclides of the uranium decay chain mainly due to the transport, the storage of raw materials and the free release of phosphogypsum waste.

1. Introduction

Phosphate industries are considered an important potential source of natural radionuclide contamination of the ^{238}U and ^{232}Th series in the environment. These elements may pose long term hazard due to their radio-toxicity and are considered the main sources of human radiation exposure (IAEA, 1994a; Euratom, 1999). The main raw materials used for fertilizer production are phosphate ores. Besides phosphate minerals, phosphate ores contain a wide range of contaminants including natural radionuclides of the ^{238}U decay chain such as radium, polonium, thorium and lead radioisotopes (Al-Masri et al., 2002; Othman et al., 1994). Radionuclide activity concentrations in phosphate ores are in the range of less than 1 up to several Bq g^{-1} (Berish, 1990; Rutherford, 1996; Mahjoubi, 2000). During production processes, amounts of contaminants are transferred from the ores to the produced fertilizers, phosphoric acids and effluents mainly as solid waste known as phosphogypsum (Rutherford, 1996; Carvalho, 1995). Several studies on the radiological impact of these industrial sites have

been carried out and in many countries the exposure of workers and public to radiation from phosphate plants has resulted in the regulation of these plants (Carvalho, 1995; Brigden et al., 2002; Righi et al., 2005; Paridaens and Vanmarcke, 2001).

In the Mediterranean countries, most of the industries are located at the seaside due to the need for cooling and transportation (Fakhri et al., 2008). Studies have shown that discharge of phosphogypsum and effluent waste into water results in significantly elevated levels of radionuclides such as polonium, lead and uranium in seawater, sediments and biota (especially fish, shellfish and crustaceans) (Brigden et al., 2002). Ingestion of contaminated biota will result in an increased radiation dose to the public. The main goal of this study was to assess the marine environmental radioactive pollution and the radiological impact caused by a large production plant of phosphate fertilizer located in the eastern coast of the Mediterranean Sea, 40 km north of the capital of Lebanon Beirut.

This industry produces since 1957 a large range of phosphate based fertilizers: simple, triple superphosphate and phosphoric acid (MoE/Envirotech, 1999) with a reported capacity exceeding 765,000 tons per year of various products (Yager, 2005). Phosphate fertilizers produced are derived from sedimentary phosphate ores, usually imported by rail from Syria (Al-Hajj and Muscat, 2000).

The UNSCEAR indicates an average of 1500 Bq kg^{-1} for ^{238}U in sedimentary phosphate ore deposits (UNSCEAR, 1988, 1993). Approximately 95% of the phosphogypsum residue is discharged as waste directly to the sea (MoE/Envirotech, 1999). In order to evaluate the radiological impact on the coastal environment surrounding the fertilizer plant, concentrations of radionuclides were measured in the different matrices and the radiation exposure of the population was estimated.

2. Materials and methods

2.1. Sampling and sample preparation

During the year 2009, two sampling campaigns were carried out by the Lebanese Marine Research Center in the marine area subjected to the impact of the fertilizers plant. Samples of seawater (10 samples), marine sediments (21 samples) and biota (6 samples) were collected from several locations distributed at around 1.2 km from the south outlet of the industry to 1.2 km from north-west outlet (Fig. 1 and Table 1). Coastal soil samples (6 samples) were taken from the south (175 and 500 m) east (600 and 900 m) and north (1100 and 1700 m) of the industry (Table 2).

Radionuclide activity concentration was analyzed in the laboratories of the Environmental Radiation Control Department of the Lebanese Atomic Energy Commission. Alpha spectrometry was used for the determination of ^{238}U, ^{234}U in seawater and ^{210}Po in biota samples. ^{210}Pb in biota and ^{238}U, ^{235}U, ^{232}Th, ^{226}Ra, ^{137}Cs, ^{40}K in sediments and soil samples were analyzed by gamma spectrometry.

Before analysis, seawater samples were acidified by adding concentrated hydrochloric acid, while solid samples were oven-dried at 85 °C for 48 h, ground and homogenized.

2.2. Alpha spectrometry analysis

2.2.1. Polonium-210 analysis in biota

Polonium-210 was measured using the standard silver disc techniques (Harley, 1978; Al-Masri et al., 2002). 7 g dry weight of biota samples were spiked with a known amount of ^{208}Po (0.6 Bq) as a yield tracer and samples were completely digested using a combination of mineral acids (nitric and hydrochloric acids). Residuals were dissolved in 100 mL of 0.5 mol/L hydrochloric acid and ascorbic acid was added to reduce the iron content. ^{210}Po was spontaneously self-plated from the solution onto a prewashed silver disc.

2.2.2. Uranium isotopes analysis in seawater

Four liters of seawater samples, acidified by hydrochloric acid until pH 2–3, were spiked with 0.2 Bq of ^{232}U as a yield tracer. pH was adjusted to 8–9 by NH$_4$OH after adding KMnO$_4$. By addition of around 20 mL of 6 MnCl$_2$ H$_2$O (0.2 mol/L) a brown color solution with a precipitate was obtained. This solution was kept for 1 or 2 days to settle and the precipitate was dissolved with a mixture of (HCl, distilled water and H$_2$O$_2$), heated to dryness and taken in 10 mL hydrochloric acid (7 mol/L).

For the extraction of uranium isotopes an anion exchange resin (Bio Rad AG 1X4, 100–200 mesh) was used. After elution, uranium isotopes were electroplated on a silver disk.

An alpha spectrometer (Alpha Analyst TM, Canberra, USA) with a passive ion implanted silicon detector (active area 300 mm^2 background counts per day of 3.6 and a minimum depletion thickness of 100 μm) was used for the counting.

Radionuclide activities were corrected for recovery by comparison with measured activity of the yield tracer and for radioactive decay to the time of sampling.

2.3. Gamma spectrometry analysis

A gamma spectrometer from Canberra equipped with extended range low-level coaxial High Purity Germanium detector (HPGe) with beryllium window, 50% relative efficiency and high resolution (2.1 keV at 1332 keV) was used in this study. The detector was surrounded with a 10-cm-thick lead shield in order to reduce the background radiation and by a 0.5 cm copper layer to attenuate the X-rays emitted by the lead shield. The detector was connected to standard integrated data processor DSA 1000

Fig. 1. Geographic distribution of the sampling locations.