Journal of Marine Systems 177 (2018) 21–27

Contents lists available at ScienceDirect

Journal of Marine Systems

journal homepage: www.elsevier.com/locate/jmarsys

An overview of the ecological half-life of the ^{137}Cs radioisotope and a determination of radioactivity levels in sediment samples after Chernobyl in the Eastern Black Sea, Turkey

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ARTICLE INFO

Keywords:
Chernobyl
^{137}Cs
Ecological half-life
Sediment
Lifetime cancer risk
Black Sea

ABSTRACT

A study which determined the activity concentration of ^{137}Cs in sediments contaminated by effluents from the Chernobyl accident which had collected along the coast of the Eastern Black Sea region in Turkey was carried out in 1993. Marine sediment samples were collected in 2015 from the same fifteen sampling points, and the activity concentrations of ^{226}Ra, ^{232}Th, ^{40}K and ^{137}Cs were determined for the sediment samples. The activity concentrations ranged from 10.94–25.95, 12.14–33.05, 265.74–459.89 and 2.08–37.45 Bq kg^{-1} for ^{226}Ra, ^{232}Th, ^{40}K and ^{137}Cs respectively. The results showed that there was a steep decline in ^{137}Cs within the sediment at most of the sampling sites from the Eastern Black Sea region during the 22-year period, except for two sites at which the measured levels were much higher. This may be the result of the combined effects of radioactive contaminant entry into this area from rivers, environmental changes and nuclear testing between 1993 and 2015. Furthermore, the ecological half-life (EHL) of the ^{137}Cs radionuclide was estimated for the sediment samples, and radiological hazard parameters such as the absorbed dose rate in air (D), the annual effective dose equivalent (AEDE) and the excess lifetime cancer risk (ELCR) were calculated and compared with the international recommended values. It was shown that these sediments do not present any significant health risk for humans in this area.

1. Introduction

Radioactive pollutants in marine environment have been recognized as a serious environmental concern (Pappa et al., 2016). Anthropogenic radionuclides resulting from accidents in nuclear reactors, nuclear tests, industrial activities and medical wastes are sources of pollution that can cause serious impacts on marine environment. Natural and anthropogenic radionuclides can be transported by different media over short or long distances and finally deposited in marine sediments (Otansev et al., 2016; Papaefthymiou et al., 2013). It is well known that many anthropogenic radionuclides entered the Black Sea after the Chernobyl power plant accident (Topcuoglu et al., 2003). Turkey, especially the northern part of it, is one of the countries which have been significantly contaminated by the Chernobyl accident. After the accident, the radioactive cloud transported to Turkey by meteorological conditions contaminated the soil surface and flora in the coastal areas of the Black Sea region (Cevik et al., 2006; Celik et al., 2009; Cevik and Celik, 2009). In addition, radionuclides mainly originated from the drainage basins of the Danube, Dnieper and Don rivers entered marine environment in the Black Sea region (Ergül et al., 2013). The primary radionuclide affecting the environment from a long-term perspective is ^{137}Cs, with a physical half-life of 30 years; this is the nuclear fission product responsible for the major share of the pollution of the Black Sea marine. The concentrations of ^{137}Cs vary from region to region according to the different sources of contamination (IAEA, 1995). In recent years, increasing anthropogenic pressures have produced significant changes in the ecosystems of the Black Sea (Delfanti et al., 2014). As a result of many factors, such as technological development and population growth, the amount of radioactive materials, petroleum products and the variety of metals in the aquatic environment of the Black Sea have increased the pollution. Therefore, many living organisms in the aquatic environment are under risk of extinction due to the

Abbreviations: EHL, ecological half-life; D, absorbed dose rate in air; AEDE, annual effective dose equivalent; ELCR, excess lifetime cancer risk; IAEA, International Commission on Radiological Protection; HPGe, High purity germanium; UNSCEAR, United Nations Scientific Committee on the Effects of Atomic Radiation; MDA, Minimum detectable activity; DW, dry weight

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http://dx.doi.org/10.1016/j.jmarsys.2017.09.005
Received 14 March 2017; Received in revised form 24 September 2017; Accepted 26 September 2017
Available online 28 September 2017

great majority of pollutants that accumulate in water resources (Koçbaş and Güner, 2008).

Natural and artificial radionuclides have been used in research into aquatic environments. Radionuclide concentrations are the key factors in any assessment of the radiological hazard to marine organisms and humans (Nielsen et al., 1999). A knowledge of the concentrations and distributions of these radionuclides in sediment is of interest, since this is a useful measure for understanding the decline of ^{137}Cs and for the monitoring of environmental contamination using radioactivity (Ravisankar et al., 2015). EHL is used to monitor the decreasing trend of ^{137}Cs in natural systems. EHL describes the decrease in the concentration of a contaminant within an ecosystem over time. In the case of radionuclides, such as ^{137}Cs, EHL may be influenced by a variety of physical, chemical, and biological processes (Peles et al., 2000).

A large number of studies of the behavior of radionuclides in sediments in different parts of the world have been carried out over recent decades (Mohanty et al., 2004; Putyrskaya et al., 2009; Desideri et al., 2002; Pappa et al., 2016; El-Saharty, 2013; Sharif et al., 1994; Oni et al., 2011; Ramasamy et al., 2009). However, there has been no comprehensive study on the change of natural and artificial radioactivity in the bottom sediment specimens in the marine environment on the Eastern Black Sea coast of Turkey for many years. Therefore, this study provides updated information and an overview of the ^{137}Cs radionuclide, which is relevant to the Eastern Black Sea region.

The main purpose of this study was to determine the EHL of ^{137}Cs and the distribution patterns and trends in activity concentrations of anthropogenic ^{137}Cs and the naturally existing radionuclides such as ^{226}Ra, ^{232}Th and ^{40}K, as recorded in various regions of the Black Sea coastal environment in sediments, by comparing ^{137}Cs activity concentrations measured in 2015 with those measured in 1993 (Varinlioğlu et al., 1995) after the Chernobyl accident. Moreover, the radiological hazard parameters such as D, AEDE, and ELCR were calculated and compared with recommended international values.

2. Materials and methods

2.1. Study area and sampling

The Black Sea is surrounded by six countries located in Europe and Asia: Bulgaria, Georgia, Romania, Russia, Turkey and Ukraine. The Eastern Black Sea region is located in the north-east of Turkey. The shoreline length of the region is nearly 450 km (Yüksek et al., 1995). The radioactivity levels of sediment samples were determined at certain coordinates in the marine environment of the Eastern Black Sea by Varinlioglu et al. in 1993. In this study, our sampling points were selected from the same points as the study done by Varinlioglu et al. in 1993. The selected sampling points were the provinces of Giresun, Trabzon and Rize, which are located on the eastern part of the coast in the Black Sea region in Turkey.

Seven marine sediment samples (39–205 m depth) for each site were collected from the fifteen different sites of the Eastern Black Sea coastal region of Turkey in August 2015. The sampling sites are given in Fig. 1. After 2000, the sampling depths at selected sampling points along the Eastern Black Sea coast changed somewhat, since the coastline in this region had been strongly impacted by the highway construction (Ergül et al., 2008). Therefore, details of some of the information including recorded new depths related to the sampling points are given in Table 1. All sampling locations were identified using a GPS.

2.2. Sample preparation

Each marine sediment sample of 0.5–1 kg was collected from the sea coast using a Van Veen grab sampler at the predetermined locations; these were packed in a nylon made of non-radioactive material, and were sealed and labelled to avoid mixing and contamination. Each sediment sample was homogenised, dried in a temperature-controlled furnace at 105 °C for 24 h to remove moisture and sieved through a 400 mesh. About 120 g of each sample was sealed in gas-tight, radon-impermeable, cylindrical polyethylene plastic containers (5.5 cm diameter and 5 cm height) for gamma activity analysis (Baltas et al., 2016).

2.3. Gamma spectrometry measurement

Gamma spectrometry measurements of all samples were conducted with a coaxial HPGe detector of 55% relative efficiency and a resolution of 1.9 keV at the 1332 keV gamma of ^{60}Co (Ortec, GEM55P4-95 model). The detector was shielded in a 10 cm thick lead well internally lined with 2 mm Cu foils. The spectrum analysis was performed using computer software Genie 2000 obtained from CANBERRA. As an example, a gamma-ray spectrum of a sample taken from one of the sediment samples belonging to the T1 station recorded with the HPGe detector is presented in Fig. 2 showing the gamma lines from various daughter radionuclides of ^{226}Ra and ^{232}Th series, ^{40}K and ^{137}Cs. The efficiency of the detector was determined with a ^{152}Eu source (Amersham Company, UK) of known activity. The counting time for each sample was selected to be 50,000 s to obtain the gamma-spectrum with good statistics. To determine the background distribution in the environment around the detector, an empty container was counted in the same manner and in the same geometry as the samples. The background spectra were used to correct the net peak area of γ-rays of the measured isotopes (Baltas et al., 2014; Kucukomeroglu et al., 2010).

The specific activities of these samples were in accordance with their certified values within errors in the order of 3%–7%. The gamma-energy lines of 351.9 keV (^{214}Pb) and 609.3 keV (^{214}Bi) were used to represent the ^{226}Ra series, while 911.1 keV (^{228}Ac) and 583.1 keV (^{208}Tl) were used to represent the ^{232}Th series. ^{40}K was investigated using gamma lines at 1460.8 keV (Cevik et al., 2010). The activity values were given in Bq kg^{-1} in dry weight for sediment samples. The activity concentrations for the ^{226}Ra, ^{232}Th, ^{40}K and ^{137}Cs in the measured samples were computed using the following equation:

$$C = \frac{N}{\varepsilon \cdot P_\gamma \cdot t \cdot m} (\text{Bq kg}^{-1}) \quad (1)$$

where N is the net counting rate of the gamma ray, ε is the photo peak efficiency of the detector used, P_γ is the absolute transition of gamma decay, t is the counting time in seconds and m is the mass of the sample in kilograms.

The MDA of the present measurement system was calculated as follows (Currie, 1968):

$$MDA = \frac{\sigma\sqrt{B}}{\varepsilon \cdot I_\gamma \cdot t \cdot w} \quad (2)$$

where MDA is in Bq kg^{-1}, σ is the statistical coverage factor, which is equal to 1.645 (confidence level 95%), and B is the background for the region of interest of a certain radionuclide. The MDA for the radionuclides of interest was calculated as 0.16, 0.24, 1.69 and 0.02 Bq kg^{-1} for ^{226}Ra, ^{232}Th, ^{40}K and ^{137}Cs, respectively. The activity concentrations are expressed in Bq kg^{-1} of dry weight sediment. The data quality was checked using the certified reference material for the moss-soil (IAEA-447). Accuracy, expressed as recovery of reference material, was 95%–98% for all of the radionuclides.

2.4. Ecological half-life calculation

The ecological half-life (EHL) of a radionuclide includes the physical decay half-life and the ecological (environmental dispersion/dilution) half-life. This is the time required for the radionuclide to decrease by 50% in the environment as a result of physical, chemical and biological factors. The EHL is determined using the following equation:

$$EHL = \log_e 2/\lambda_e \quad (3)$$