Assessment of natural radioactivity in the marine environment in Croatia

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Abstract

Investigations of natural radioactivity, particularly radium and uranium, in the Adriatic Sea water and biota have already been performed on selected locations along the Croatian coast, as a part of an extensive monitoring program of the Croatian environment, conducted by the Radiation Protection Unit, Institute for Medical Research and Occupational Health, Zagreb. The paper deals with increased levels of natural radioactivity in a bay at the Croatian cost, which is due to geographic characteristics exposed to any kind of pollution including radioactivity originating from the coal fired power plant situated about 5 km from the seaside. Slag and ash are accumulating continuously, consequently to regular operation of the power plant. Previous investigations of used coal resulting slag, showed increased activity concentrations of natural radioactivity. In order to assess the sensitivity of the bay to the radioactive pollution, natural radioactivity concentrations were studied in the zone between the plant and the seaside, as well as in sea water. ^{226}Ra concentrations in the sea water were found to be increased in comparison to ^{226}Ra levels measured at the selected open sea locations.

1 Introduction

The central role of energy in economic and social development has long been recognized and a great deal of effort has been devoted to developing technologies for the extraction, production, and use of all types of energy. The aim has always been to reduce costs, make systems more efficient, and provide access to previously untapped energy sources. Since electrical energy production is always associated with the emission of several pollutants into the atmosphere, reducing the adverse environmental and health effects have become the questions of public and scientific priority in recent decades. [1,2]

The natural radioactive series of uranium and thorium with their decay products in secular equilibrium are always present in coal. By burning coal, the activities have been redistributed, taking them from underground where the impact on living environment was none and liberating them into air and over the surface of the ground where it can modify ambient radiation fields and population radiation exposure. Several coal mines in Croatia have more or less elevated concentrations of natural radioactivity. The presence of uranium varies from mine to mine and even from one coal-layer to another in the same mine.[3]

The paper deals with increased levels of radioactivity in the bay at the Croatian cost of the Adriatic Sea, which is due to its geographic location exposed to any kind of pollution including the radioactivity emitted from the coal fired power plant situated about 5 km from the seaside. The coal used for the regular plant operation originates from a domestic coal mine, which in addition to higher levels of natural radioactivity (uranium) has an elevated concentration of sulphur, up to 10%.[4] Coal has been mined mostly at adjacent coal mine area, about 10 km from the plant site. Over the past few years the coal used in the plant includes about one half of imported coal characterized by different radioactivity characteristics. The aim of this study was to determine

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radioactivity level at the ash and slag deposits and to assess the risk to the Adriatic Sea water and biota. Investigations of natural radioactivity have already been performed at selected locations along the Croatian coast, as a part of an extensive monitoring program of the Croatian environment conducted by the Radiation Protection Unit, Institute for Medical Research and Occupational Health, Zagreb.

2 Description of the Area

The plant is situated in the coastal area, about 5 km from the seaside, and 10 km from the more populated urban area in the continental part of the region. The plant is surrounded by the hills with sparsely dissipated villages. A more populated urban center is situated at the seaside. The direct combustion of coal, or its conversion to other fuels, results in the concentration of the noncombustible mineral matter, including most of the radionuclides, in the ash or gaseous residues. Besides the immediate surrounding area of the plant, radioactively polluted material affects also the zone between the plant and the seaside, as well as submarine area of the bay. In view of this, the location of the deposit sites, i.e. the storage of large quantities of ash and slag, presents a remarkable environmental problem.

The coal ash and slag deposit site is situated close to the power plant site. Ash and slag are accumulating continuously, consequently to regular operation of the power plant. There have been performed some measurements of natural radiation in marine in vicinity of coal mine and power plant, as well as in ashes generated at power plant. After incineration the initial radioactivity becomes 7-10 times more concentrated than in coal. The particulates and the solid incombustible ash containing uranium and ^{226}Ra together with sulphur bound to the mineral substance, are here the main problem.

The slag and ash pile is situated on low permeable Eocene flysh. However, this sediment could be very easy weathered and affected by proluvial process. Geological setting of the broader area is characterized by Mesozoic carbonates (limestone, dolomites) which are due to irregular circulation of groundwater environmentally very sensitive and, thus, the monitoring of radionuclides which would possibly migrate in the groundwater from this pile, is not easy.

3 Material and Methods

Investigations of radioactive contamination were based on measurements performed in the field and in the laboratory.

In situ gammaspectrometrical measurements were carried out using HPGe Ortec detector (resolution 1.74 keV on 1.33 MeV ^{60}Co, relative efficiency 21.6%), and included several measuring points at the protected and still operating part of the deposit and the seaside control points.

Determination of radioactive contamination required analyses of different types of samples: the samples of sea water, waste water from the plant at the point of mixing with sea water, and sea bioindicators The collected samples of sea bioindicators were gammaspectrometrically analysed in the laboratory using Ge(Li) Ortec detector (resolution 1.78 keV on 1.33 MeV ^{60}Co, relative efficiency 16.8%) with a 4,096 channel analyser and a personal computer. All samples were measured in Marinelli beaker, volume 1 L. Measurement time was 80,000 sec.[5]

The liquid samples were radiochemically separated, after which ^{226}Ra was determined by alpha spectrometric measurements using Ortec Si(Li) surface barrier detector. The counting time for each measurement was 60,000 sec or longer.

4 Results and Discussion

In situ gammaspectrometrical measurements involved several measuring points at the deposit site (still operating part and covered part of the deposit site) and at several control points at the seaside. At the still operating part of the deposit site the measurements showed the presence of natural radionuclides of uranium and thorium decay series, ^{40}K and ^{137}Cs. Corresponding contribution of measured radionuclides to the absorbed dose rate was calculated for each measuring point. The results of measurements at 20 measuring sites indicated a great dispersion of the obtained absorbed dose rates. Over the recent years was used the mixture of domestic and imported types of coal, characterized by the different radioactivity levels that resulted in varied radioactivity values obtained at the operating part of the ash and slag deposit site.

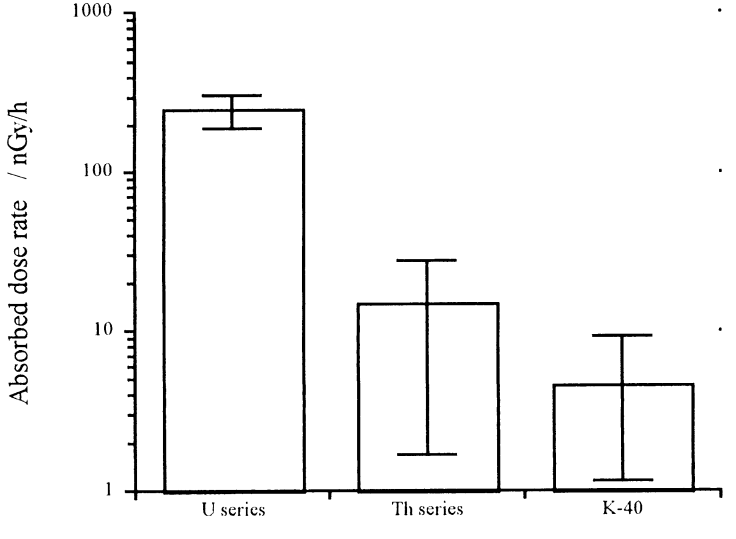

Figure 1: Contributions of radionuclides of uranium and thorium decay series and ^{40}K to the absorbed dose rate at the operating part of the ash and slag site

Figure 1 shows contributions of radionuclides of uranium and thorium decay series and ^{40}K to the absorbed dose rate of 20 measuring points at the operating-deposited ash and slag site.

In order to determine the efficacy of the protective layer at the soil-covered part of the deposit site, in situ gammaspectrometrical measurements were carried out at several measuring points. For each measurement was calculated corresponding contribution of measured radionuclides to the absorbed dose rate. The calculated absorbed dose rates at some measuring points of the operating ash and slag deposit were up to five times higher than the average calculated absorbed dose rate at the covered part of the deposit site (177±40 $nGyh^{-1}$). The statistical analysis of the difference between the absorbed dose rate at the operating pile and protected part of the deposit site showed statistically significant difference of $P(t)<0,01$ which indicates the satisfying efficacy of the protective covering of the deposit site.

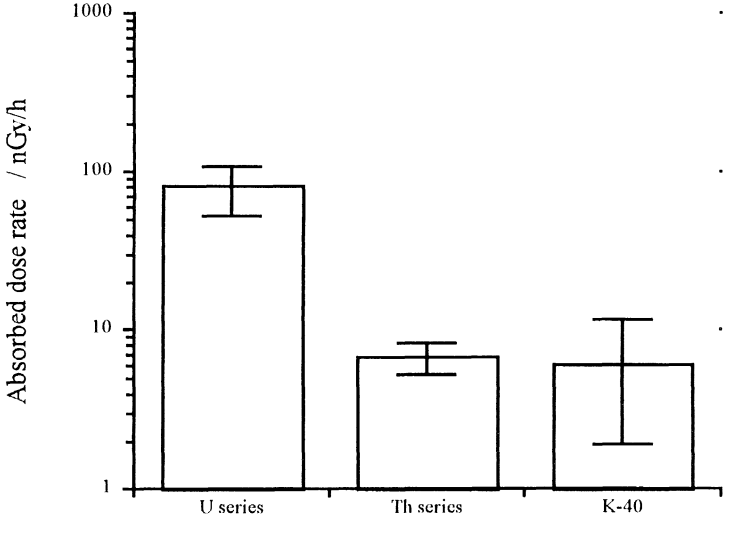

Figure 2: Contributions of radionuclides of uranium and thorium decay series and ^{40}K to the absorbed dose rate for several location at the seaside

Figure 2 shows the average absorbed dose rates for several locations at the seaside. There is a noticeable difference between the still active part of the deposit site and the results obtained at the seaside which correspond to the absorbed dose rate measured in other parts of Croatia.

In the samples of bay water, the mean ^{226}Ra concentration of 5.2±0.9 Bqm^{-3} was within the same order of magnitude with ^{226}Ra concentration in the Mediterranean Sea (3.7 Bqm^{-3})[6], and as well as in the open Adriatic Sea far from the plant deposit bay.[7]

Figure 3 shows ^{226}Ra specific activities determined in the samples of sea water (in the bay) and waste water from the plant at the point of mixing with the bay water. ^{226}Ra activity concentrations decrease with increasing distance from the mixing point.

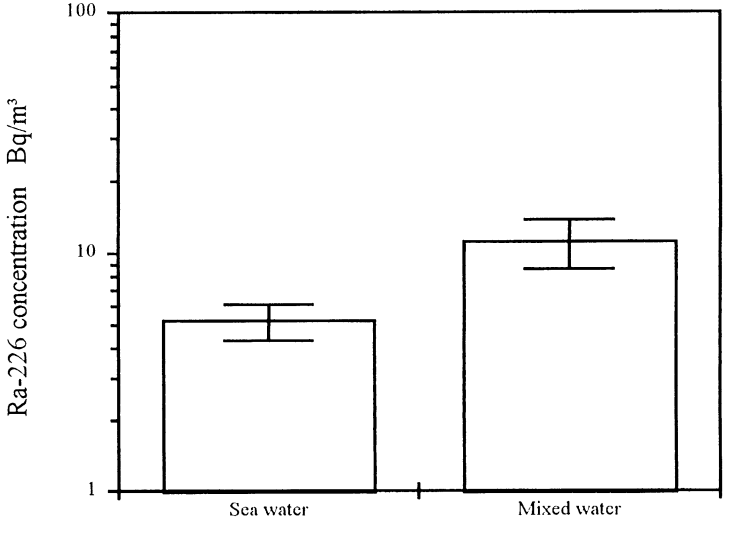

Figure 3: ^{226}Ra activity concentrations in the samples of sea water and waste water at the mixing point with the sea water

The samples of mussels were collected at the seaside as most suitable indicators of a sea biota. Gammaspectrometrical analyses carried out to detect possible accumulation of natural radionuclides in sea organisms showed that all values except ^{40}K were below the detection limit of the instrument.

5 Conclusion

On basis of our study and the obtained data it can be concluded that the investigated power plant and its deposit site present no significant risk to the inhabitants and the environment of the region. This is due to the geographical location of the plant in a sparsely inhabited area, relatively distant from more populated and a tourist region of the seaside.

The results of the measurements also confirm that the ash and slag deposit site of both the protected and still operating part of the deposit site are well monitored and involve all the necessary protective measures. All obtained data can be used as a valuable database for future estimations and modeling of the impact of radioactive pollution to the marine environment and developmental prospects of the region.

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