Natural Radionuclides in Coal and Waste Material Originating from Coal Fired Power Plant

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Abstract. This paper presents long-term investigations of natural radioactivity in coal, used for power production in the coal-fired power plant (CFPP) situated on the Adriatic coast, and resulting slag and ash. Activity concentrations of ^{40}K, ^{232}Th, ^{226}Ra and ^{238}U in used coal and resulting waste material have been measured for 25 years. As expected, it was demonstrated that the content of radionuclides in deposited bottom and filter ash material are closely related with radionuclide activity concentrations and mineral matter fraction in used coals. The external hazard index has been calculated and discussed for the slag and ash depository. During the first decade of operation of the CFPP has been used domestic coal produced in nearby area characterized by higher background radiation compared with the rest of Croatia. Therefore, the coal itself had relatively high ^{226}Ra and ^{238}U activity concentrations while potassium and thorium content was very low, ^{40}K activity concentrations being 2-9% and those of ^{232}Th 1-3% of total activity. As, in addition, the sulphur concentrations in coal were very high use of domestic coal was gradually abandoned till it was completely substituted by imported coal originated from various sources and of low natural radioactivity. Upon this, activity concentrations of uranium series radionuclides in deposited waste materials decreased significantly. Consequently, waste material i.e., slag and ash, generated in the last several years of coal fired power plant operation could be readily used in cement industry and as additive to other building materials, without any special restrictions according to the Croatian regulations dealing with building materials and European directives.

KEYWORDS: Natural Radioactivity, Coal, Slag, Ash, and Building Material

Introduction

The coal fired power plants (CFPP) are one of the sources of the technologically enhanced natural occurring materials (TENORM). The radiation levels can exceed background several times [1]. The investigated plant is situated about 5 km from the seaside in the Northern Adriatic. This area has been included in an extensive monitoring program of the Croatian environment [2]. This program has been conducted by the Radiation Protection Unit of the Institute for Medical Research and Occupational Health, Zagreb. The pile of slag and ash resulting from operation of CFPP is accumulated continuously on the well monitored deposit site. The pile contains about one million tons of different waste material. The levels of natural radioactivity in the slag and ash pile depend on different kinds of coal used for the plant operating [3].

The results obtained in the study point to a possibility that slag and ash pile accumulated over the past few years of the plant operation contain material that could be presently used in cement industry without any special restriction. It should be mentioned that since local coal-mines had been recently closed, only good quality imported coals are used for the plant operation [4].

Material and Methods

The samples of coal were collected at coal deposit in CFPP or on vessel for coal transport. Bottom and fly ash were collected at electro-filters inside CFPP while gypsum, slag and ash were collected at the operating ash and slag pile deposit. The samples of building material (cement, gypsum, concrete etc.) were collected at a factory of building material.

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All the samples were gamma-spectrometrically analyzed in the laboratory using HP Ge and/or Ge(Li) detector (resolution 1.78 keV on 1.33 MeV ^{60}Co, relative efficiency 16.8%; resolution 1.56 keV on 1.33 MeV ^{60}Co, relative efficiency 18.7%) with electronic units and software for analysis of gamma spectra on a personal computer. All samples were measured in Marinelli beakers of appropriate volume. Measurement time was 80,000 sec or higher.

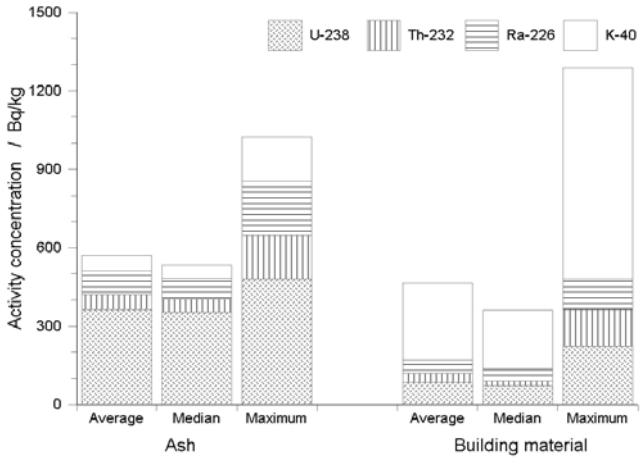
Results and discussion

Till the beginning of 1990's the majority of coal used for CFPP operation originated in local mines. In that period, deposited waste material was characterized with relatively high ^{226}Ra and ^{238}U activity concentrations, while potassium and thorium content was very low (^{40}K activity was 2-9% and ^{232}Th 1-3% of total activity). When domestic coal has been completely substituted with imported coal of much better radiological quality, activity concentrations of uranium series radionuclides in deposited waste materials decreased significantly. However, potassium and thorium activity concentrations in slag and ash pile material increased.

Since no more than 22% of ash for composing building material is needed [5], slag and ash pile material generated in the last several years of coal fired power plant operation could be generally used in cement industry without any special restriction.

Fig.1 shows the activity concentration of some natural radionuclides (in Bqkg^{-1}) measured in ash and slag samples and building materials.

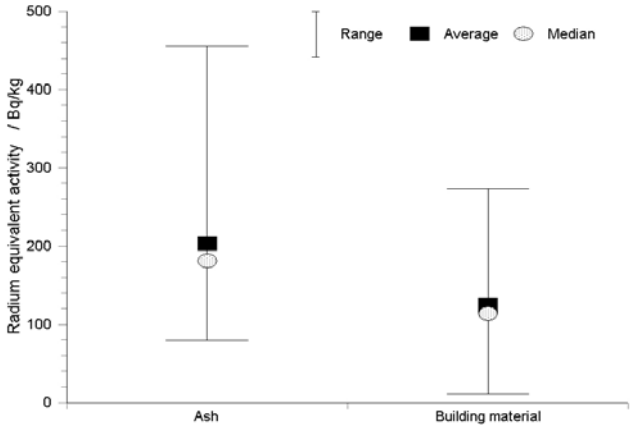
Figure 1: Activity concentration of ^{238}U, ^{232}Th, ^{226}Ra and ^{40}K (average value, median value and maximum) in ash and slag samples and in building material samples in 2000s



The natural radioactivity of building materials is usually determined from ^{226}Ra, ^{232}Th and ^{40}K contents. As 98.5% of the radiological effects of the uranium series are produced by radium and its daughter products, the contribution from the ^{238}U has been replaced with the decay product ^{226}Ra. Radium equivalent activity, Ra_{eq}, is an index that has been introduced to represent the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K by a single quantity, which takes into account the radiation hazards associated with them. For safe use of materials for building constructions, the maximum value of Ra_{eq} must be less than 370 Bqkg^{-1} [6].

Fig.2 shows the radium equivalent activity for ash samples and samples of building material.

Figure 2: Radium equivalent activity, Ra_{eq}, of ash and building material samples



The external hazard index, H_{ex}, is obtained from Ra_{eq} expression through the supposition that its maximum value allowed (equal to unity) corresponds to the upper limit of Ra_{eq} (370 $Bqkg^{-1}$). This index value must be less than unity in order to keep the radiation hazard insignificant; i.e. the radiation exposure due to the radioactivity from construction materials is limited to 1.0 $mSvy^{-1}$. Then, the external hazard index, H_{ex}, can be defined as:

$$H_{ex} = \frac{A(Ra)}{370} + \frac{A(Th)}{259} + \frac{A(K)}{4810} \leq 1 \qquad (1)$$

where *A(Ra)*, *A(Th)* and *A(K)* are ^{226}Ra, ^{232}Th and ^{40}K activity concentrations in $Bqkg^{-1}$ respectively. The safety criterion for materials used in building construction should be less or equal to unit. Fig. 3 shows frequency histogram of external hazard index, H_{ex}, calculated for each ash and slag sample (minimum H_{ex} value was 0.214 and maximum 1.228). Fig. 4 shows frequency histogram of H_{ex} calculated for each sample of building material (minimum H_{ex} value was 0.030 and maximum 0.736).

Figure 3: Frequency histogram of external hazard index, H_{ex}, of ash samples in 2000s

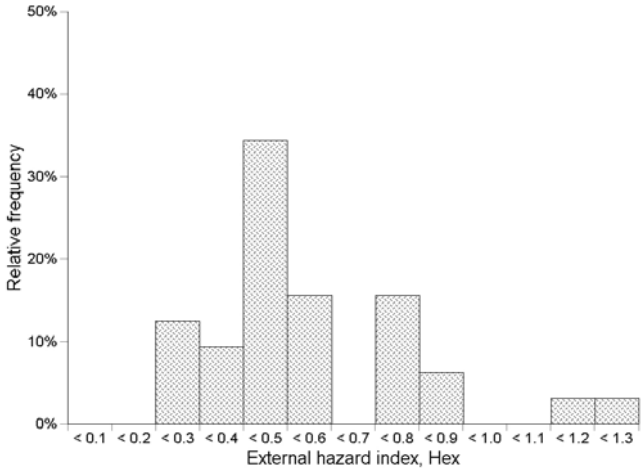
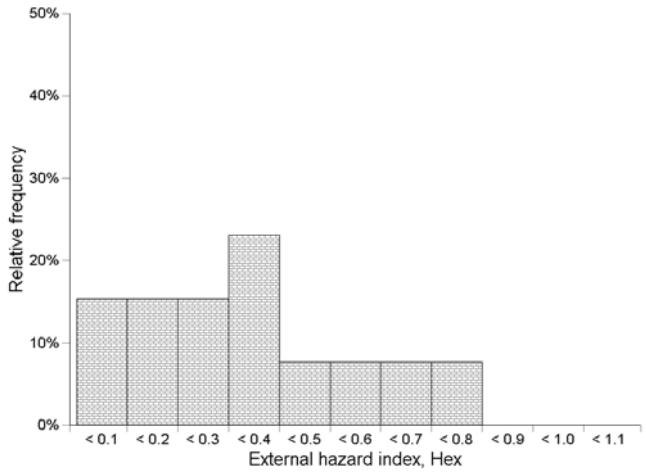


Figure 4: Frequency histogram of external hazard index, H_{ex}, of building material in 2000s

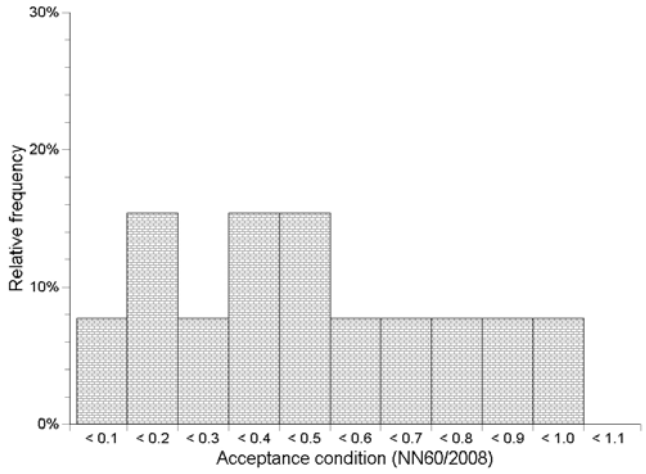


According to the Croatian laws, which deal with the radiation protection [7,8] and radiological protection principles concerning the natural radioactivity of building materials proposed by European Commission (EC) [9], activity concentrations in $Bqkg^{-1}$ of ^{226}Ra (A(Ra)), ^{232}Th (A(Th)) and ^{40}K (A(K)) in building construction materials must satisfy the following criterion:

$$\frac{A(Ra)}{300}+\frac{A(Th)}{200}+\frac{A(K)}{3000}\leq 1 \qquad (2)$$

Fig. 5 shows frequency histogram of calculated acceptance criterion for all measured building materials in 2000s.

Figure 5: Frequency histogram of acceptance criterion calculated for building material in 2000s [8].



Conclusion

The waste material resulting from the normal operation of the investigated CFPP plant has been generally used in cement industry but in a percentage determined by its level of radioactivity. Introduction of imported coal in the plant operation resulted with possibility of an unrestricted use of the waste material in accordance with the Croatian and European regulations for building materials.

Although recent radioactivity measurements showed an improved environmental situation, due to proximity of the seaside and developmental prospects of the region regular monitoring still remains necessary.

On the 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.

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.

Acknowledgements

This study is a part of a research projects *Environmental Radioactivity and Radiation Protection* and *Radioecology of the Adriatic Sea and Coastal Areas* supported by the Ministry of Science, Education and Sports of the Republic of Croatia.

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