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Full Length Research Paper

An investigation on natural radioactivity from mining industry#

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The environmental problem of naturally occurring radioactive materials (NORM) is omnipresent on earth and their radioactivity may become concentrated as a result of human activities. Various industries produce concentrated radioactivity in their by-products. Mining originating industries such as the coal industries, petroleum extraction and processing and natural gas, mining enrichment waste, phosphate, etc have been well known and widely investigated. The Environmental Protection Agency (EPA) describes NORM wastes from the mining and processing of three categories of metals: Rare earth metals, special application metals and metals produced in bulk quantities by industrial extraction processes. Moreover, NORM has a lot of negative effects on the natural resources (water supplies, soils, air, etc.) and living organisms (human, animals, plants, microorganisms, etc.). In this study, we investigated NORM levels that originated from mining industry and the concentration of NORM in drinking water supplies. NORM parameter of gross alpha and gross beta were also in this study, seasonal changes in gross alpha and gross beta were investigated. The obtained results showed that, natural activity concentrations of α- and β-emitting radionuclides in all water samples did not exceed World Health Organisation (WHO) and Turkish Standards of Drinking Water (TS 266) recommended levels (Table 1). Concentrations ranging from 0.0062 Bq/l to 0.79 Bq/l and from 0.004 to 0.18 Bq/l were observed for the gross α and gross β activities, respectively. For all samples, the gross β activities were higher than the corresponding gross α activities.

Key words: Natural radioactivity, mining industries, gross alpha, gross beta.

INTRODUCTION

Radiation of natural origin at the earth's surface consists of two components namely cosmic rays and radiation from the radioactive nuclides in the earth's crust. The latter component is the terrestrial radiation, which mainly originates from the so-called primordial radioactive nuclides that were made in the early stage of the formation of the solar system. Uranium, thorium and potassium are, however, the main elements contributing to natural terrestrial radioactivity (UNSCEAR, 2000).

Naturally-occurring radioactive material (NORM) is the term used to describe materials that contain radionuclides that exist in the natural environment. Long-lived radioactive elements of interest include uranium, thorium and potassium and any of their radioactive decay products, such as radium and radon. These elements have always been present in the earth's crust and within the tissues of all living species. Studies of terrestrial natural radiation are of great importance for various reasons. They serve as useful tracers for atmospheric variation studies. It is usually realized that natural environmental radiation mainly depends on geological and geographical conditions. Also, an understanding of natural radioactivity was used to determine the age of the earth using the radioactive decay of ^{238}U to ^{206}Pb and ^{235}U to ^{207}Pb. Each isotope has, however, a unique decay rate so that the ratio of the number of daughter atoms to parent isotopes in rocks and minerals depends on the time which has elapsed since the system became closed (Jamal, 2002). Many of the mining and processing

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Figure 1. A map of the study area and location of the sampling points.

(physical, chemical and termal): uranium, thorium, gold, silver, copper, nickel, iron, aluminium, molybdenum, tin, titanium, vanadium, etc. industries such as afore-mentioned cause natural radiation.

Extraction of minerals

This was undertaken by conventional underground or open pit mining techniques or acid leaching. Mineral processing can involve dry techniques including electrostatic or magnetic separation or wet techniques such as acid or alkaline leaching and chemical flotation or electrical or furnace smelting. All of these processes can affect the concentrations of radionuclides in both waste and product streams.

Mineral sands production

Mineral sands production (including ilmenite, leucoxene, rutile, zircon, monazite and xenotime) leads to several waste materials from primary processing of ore. Some of these waste materials are returned to the mined out pit for disposal.

In copper production

Wastes containing NORM arise in tailings from the flotation stage and furnace slags from the smelting stage.

In tin and tantalum production, dry and wet separation stages produce tailings slurry that is further treated and disposed off in a tailings dam close to the mine site. Tantalum products also contain low levels of NORM. Iron and steel production wastes include blast furnace slags and dusts and fumes from the sinter plant and off gas cleaning in the blast furnace operation.

In phosphate ore processing

NORM is found in tailings from fertiliser production that are normally used as backfill at the mine site and also in fertilizer product at levels which do not cause any significant increase in the uranium and thorium levels in soil treated with fertilizer. In phosphoric acid production, the majority of the NORM is left in the phosphogypsum, which can be stockpiled on site or disposed of as landfill. Phosphogypsum may also be used in fertilizer, soil conditioner, building material (plasterboard, cement aggregate) and in road construction (ARPANSA, 2004).
A lot of studies can be found on natural radioactivity in earth soil and water samples. In this study, natural radioactivities in water samples were investigated from mining origin.

MATERIAL AND METHODS

Site description

The study area at the Konya city in Turkey is shown in Figure 1. It is

Table 1. Drinking water standards of gross alpha and gross beta (WHO, 2006; TS266, 2005).

Radionuclide	WHO (WHO,2006)	Institute of Turkish Standards, (ITS 266, 2005)
Gross alpha	0.01 Bq/l (2.7 pCi/l)	0.0037 Bq/l (0.1 pCi/l)
Gross beta	0.1 Bq/l (27 pCi/l)	0.037 Bq/l (1 PCi/l)

in the Turkey's central Anatolia region. Konya has a lot of mine resources. These are barite, bentonite, mercury, porcelain clay, clay, limestone, chrome, meerschaum, marble, talcum, asbestos, lignite, etc. (MTA, 2010). Naturally, these mining resources and their processing activity lead to natural radiation on nearest water supplies and soil.

Sampling

In this study, gross alpha and gross beta activities were measured for the samples which have been taken from eight different drinking water resource (1 surface water, 6 well water, 1 tap water) in Konya city for two different seasons (winter and summer). Sampling points are shown in Figure 1. The values of measurements of the samples which were taken in the Centrum of Konya show that there is no natural radiation problem in water resources. Although, some measurements result to higher gross alpha parameter according to TS 266 (Turkish Standards of drinking water), but are still lower than WHO standards, so this causes no problem. Furthermore, gross beta activity values are fairly low according to TS 266 also, WHO standards, so beta radioactivity causes no danger (TS 266, 2005). Drinking water standards of gross alpha and gross beta are shown in Table 1.

Determination of natural radioactivity

Primarily, the aim of this study was to determine the level of natural radioactivity in different drinking water supplies in Konya city. Eight sampling sites for drinking water supplies have been selected in Konya city. Figure 1 gives a detailed description of all sampling sites. Sample bottles were rinsend with pure water, then filled completely to minimize headspace. The sample bottles were cleaned using a modified procedure before collection. (Laxen and Harrison, 1981). Whole drinking water samples were collected by 1 L capacity of polypropylene bottles and acidified pH 2 with pure HNO_3 until analysis. Measurements of radioactivity level in all water samples were performed by the method of EPA using the gross alpha and gross beta counting system Epa method 900 (EPA, 1980). An aliquot of a preserved drinking water sample is evaporated to a small volume and transferred quantitatively to a tared 2-inch stainless steel counting planchet. The sample residue is dried to constant weight, re-weighed to determine dry residue weight, then, counted for alpha and/or beta radioactivity. Counting efficiencies for both alpha and beta particle activities were selected according to the amount of sample solids from counting efficiency vs sample solids standard curves (EPA, 1980). Gross α and β-activities were determined by means of a Tennelec LB 1000 low background proportional counter subsequently (Tennelec, 1989). The counter was calibrated with ^{241}Am and ^{90}Sr–^{90}Y standards by using potentials of 1100 and 1600 V corresponding to the α and β-plateaus, respectively. The total yields obtained for the α and β-countings were 16 and 34%, respectively.

Calculations

Equation 1: The following equation was used to calculate the alpha radioactivity:

$$Alpha\ (pCi/L) = \frac{A\ x\ 1000}{2.22\ x\ C\ x\ V} \quad (1)$$

Where, A is the net alpha count rate (gross alpha count rate minus the background count rate) at the alpha voltage plateau; C is the alpha efficiency factor read from the graph of efficiency versus mg of water solids per cm^2 of planchet area, (cpm/dpm); V is the volume of sample aliquot, (ml) and 2.22 is the conversion factor from dpm/pCi (EPA, 1980).
Also, equations that were used to calculate beta radioactivity are given as:

Equation 2: Is used if there are no significant alpha counts when the sample is counted at the alpha voltage plateau, the beta activity can be determined from the following equation:

$$Beta\ (pCi/L) = \frac{B\ x\ 1000}{2.22\ x\ D\ x\ V} \quad (2)$$

Where, B is the net beta count rate (gross count rate minus the background count rate at the beta voltage plateau); D is the beta efficiency factor read from the graph of efficiency versus mg of water solids per cm^2 of planchet area, (cpm/dpm); V is the volume of sample aliquot, (ml) and 2.22 is the conversion factor from dpm/pCi (EPA, 1980).

Equation 3: Is used when counting beta radioactivity in the presence of alpha radioactivity by gas-flow proportional counting systems (at the beta plateau), alpha particles are also counted. Since alpha particles are more readily absorbed by increasing sample thickness than beta particles, the alpha/beta count ratios vary with increasing sample thickness.

Therefore, it is necessary to prepare a calibration curve by counting standards containing americium-241 with increasing thickness of solids on the alpha plateau and then, on the beta plateau, plotting the ratios of the two counts versus density thickness.

The alpha amplification factor (E) from that curve is used to correct the amplified alpha count on the beta plateau. When significant alpha activity is indicated by the sample count at the alpha voltage plateau, the beta activity of the sample can be determined by counting the sample at the beta voltage plateau and calculating the activity from the following equation:

$$Beta\ (pCi/L) = \frac{(B - AE)\ x\ 1000}{2.22\ x\ D\ x\ V} \quad (3)$$

Where, B (as defined in equation 2); D = (as defined equation 2); A = (as defined in equation 2); E is the alpha amplification factor read from the graph of the ratio of alpha counted at the beta voltage/alpha counted at the alpha voltage versus sample density

Table 2. Winter season, activity concentrations of gross alpha and gross beta in water supplies samples in Konya city. (Esmeray and Aydın,2006)

Sample location	α Radioactivity (Bq/l)	β Radioactivity (Bq/l)
Apa Dam	0.072 ± 0.0023	0.20 ± 0.004
Anit	**0.079 ± 0.0026**	0.08 ± 0.004
Kongaz II	0.007 ± 0.0021	0.06 ± 0.004
Kas III	0.0062 ± 0.0026	0.05 ± 0.004
Forestry directorate	**0.052 ± 0.0025**	0.10 ± 0.004
City center (KOSKI)	**0.007 ± 0.0020**	0.01 ± 0.003
Marangozlar	**0.014 ± 0.0022**	0.09 ± 0.004
Meram II	0.0077 ± 0.0011	0.18 ± 0.004

Table 3. Spring Season, Activity concentrations of gross alpha and gross beta in water supplies samples in Konya City. (Esmeray and Aydın,2006)

Sample location	α Radioactivity (Bq/l)	β Radioactivity (Bq/l)
Apa Dam	0.018 ± 0.0019	0.11 ± 0.004
Anit	0.035 ± 0.0023	0.07 ± 0.003
Kongaz II	0.006 ± 0.0019	0.09 ± 0.004
Kas III	0.0012 ± 0.0018	0.04 ± 0.003
Forestry directorate	0.008 ± 0.0023	0.13 ± 0.004
City center (KOSKI)	0.006 ± 0.0017	0.04 ± 0.003
Marangozlar	0.065 ± 0.0028	0.18 ± 0.004
Meram II	0.007 ± 0.0021	0.18 ± 0.004

thickness; V is the volume of sample aliquot, (ml) and 2.22 is the conversion factor from dpm/pCi (EPA, 1980).

Finally, all results were converted between units (pCi/l) to (Bq/l) and used for experimental results. (Esmeray, 2005)

RESULTS AND DISCUSSION

Activity concentrations of gross alpha and gross beta in drinking water in Konya are shown in Tables 2 and 3. As shown in Table 2, Apa Dam, Anit and Forestry Directorate's gross alpha activity concentrations are over than the standards of ITS and nevertheless, all results of gross beta activity concentrations are permissible limits for both WHO and ITS regulations for drinking water. Only Marangozlar's gross alpha activity concentration is more than ITS standards, as shown in Table 3, but all other gross alpha activity concentrations are permissible limits for both WHO and ITS regulations for drinking water standards. In addition, all gross beta and gross alpha activity concentrations are generally good and acceptable for WHO and ITS's recommended levels of drinking water (Table 1).

All experimental results show that, the gross alpha activity ranges between (0.0062 (Meram II) and 0.79 (Anıt) $Bq.l^{-1}$-winter season) (0.0012 (Kas III) and 0.065 (Koski) $Bq.l^{-1}$-spring season). Besides, gross beta activity lies between (0.001(Koski) and 0.2 (Apa Dam) $Bq.l^{-1}$-winter season) (0.004 (Koski, Kas III) and 0.18 (Meram II, Marangozlar) $Bq.l^{-1}$-spring season). Activity concentrations of gross alpha and gross-beta in water supplies in Konya are shown in Table 3 (winter and spring season).

As shown in Tables 2 and 3, the lowest average were 0.0062 ± 0.0026 Bq/l (gross alpha) and 0.01 ± 0.003 Bq/l (gross beta) in winter and the highest seasonal average were 0.079 ± 0.0026 Bq/l (gross alpha) and 0.20 ± 0.004 Bq/l (gross beta) in winter. Also, the lowest average were 0.0012 ± 0.0018 Bq/l (gross alpha) and 0.04 ± 0.003 Bq/l (gross beta) in spring and the highest seasonal average were 0.065 ± 0.0028 Bq/l (gross alpha) and 0.18 ± 0.004 Bq/l (gross beta) in spring. Evaporation conditions, temperature changes, solubility of radioactive materials can be causes of seasonal changes in gross alpha and gross beta activities in Konya. Besides, the concentration of radioactivity increases in spring and summer due to high evaporation rates and the increase of solubility of salts due to the higher temperature of the water (Hosseini, 2007). Additionally, high levels of natural radioactivity in drinking water are accompanied with potential health risks for the population by increasing the radiation dose. Hence, water must be purified before using it. There are several methods known to remove radioactivity from water

such as aeration to remove granular activated carbon (GAC), ion exchange methods (IX), reverse osmosis (RO) to remove “Gross alpha and gross beta, uranium etc.” and various adsorption methods to remove other natural radioactivity sources (Esmeray and Aydin, 2008).

Conclusions

The levels of gross alpha and gross beta activity in Konya region located in central anatolia of Turkey have been studied. Experimental results observed show that, gross alpha and gross beta activity concentrations are lower than standards of WHO and ITS (WHO, 2006; ITS, 2001). This kind of the study must be continued for other parts of Konya and Turkey. The results and data obtained in this work are a baseline which can be used to evaluate possible future changes.

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