The Analyst

Concentrations of radon and decay products in various underground mines in western Turkey and total effective dose equivalents†

Güngör Yener and Eşref Küçüktaş

Ege University, Institute of Nuclear Sciences, 35100 Bornova, Izmir, Turkey

In the present work radon concentration measurements were performed for one year in 12 different boron, chromium and coal underground mines in Western Turkey. Lucas cells and nuclear track detectors were used for the measurements of radon and its decay products. The effects of parameters, such as type of mine, gallery depth and ventilation rate, on the radon concentration in mine air were examined. The radiation exposure doses of miners due to the inhalation of radon and radon daughters were determined. Gamma survey measurements were also realized together with radon measurements and the total effective dose equivalents in mSv y^{-1} were estimated.

Keywords: *Radon; underground mines; effective dose*

Exposure to radon and its decay products is the most significant component of natural radiation exposure of the general population. Among the radon isotopes ^{222}Rn, with the longest half life (3.85 d), is the most important one since it is formed from alpha decay of ^{226}Ra in the decay chain of ^{238}U that is widely distributed throughout the earth's crust. Radon emanates from soil, rock and water and becomes dispersed in air. Being a noble gas it migrates by diffusion and convection without any significant interaction with the constituents of air or any airborn particulates.[1]

Human exposure to radon progeny occurs out of several sources. Underground mining is one of the most important technologically enhanced causes that highly contributes to occupational health risk since the ore dust containing the members of the uranium and thorium decay series are transported to the galleries through water or air circulation during mining operations. Epidemiological studies have indicated that the presence of radon and its decay products in inhaled air causes a health risk for lung cancer.[2,3] Although there exist large uncertainties associated with risk estimates, studies, especially on uranium miners, have shown that the relative risk for lung cancer increases almost linearly[4,5] with working level month (WLM).‡

The first evidence of a health risk associated with exposure to radon and its decay products dates back to the sixteenth century when it was noted that the mining population in Scheeberg (Germany) and Bohemia were suffering from a widespread fatal lung disease known as 'Schneeberger Krankheit').[6] In the 1950s the theoretical grounds together with experimental investigations were used to establish the direct relation between radon progeny and increased lung cancer observed in Europe and in the United States.[6] Intensive epidemiological investigations have been realized on occupational health risk but analyses related to the non-mining population did not start until late 1970s and they are comparatively rare.

In recent years, substantial epidemiological and dosimetric information has been collected by the International Commission on Radiological Protection (ICRP) and in 1990, the Committee decided that exposure of miners should be classified as occupational exposure. In 1991, the commission accepted that the dose limit for workers would be 20 mSv y^{-1}, it was assumed that the exposure limit for radon progeny would be 2 WLM y^{-1}.[7]

Most of the epidemiological studies done on underground miners dealt with uranium and phosphate mines.[8,9] However, high radon concentrations are not confined to uranium mines and mills. Since uranium minerals occur widely dispersed in the earth's crust they are found to accompany many other minerals that are being mined commercially.[1]

Exposure of miners around the world from data of variable quality for 750 mines in 12 countries have been summarized by the ICRP. Human exposure levels have an average value of 1 WLM y^{-1} with perhaps 10% or so exceeding 2 WLM y^{-1} in non-uranium mines. Average exposures are 0.2 WLM y^{-1} and 1 WLM y^{-1} for coal miners and uranium miners, respectively.[7]

The radon concentration in mine air depends primarily on the uranium content of the mineral and also on other parameters like geological structure, porosity, ventilation rate, moisture and activity type in the mine.[10] On the other hand the severity of exposure to radon progeny depends on their concentration in air, the probability of attachment to aerosols and the particular portion of the respiratory system where they end up.[11] Therefore, in the calculations for dose estimation the physical parameters that effect the radon progeny concentrations in air, aerosol attachment fractions, the accumulation in different sections of respiratory system and the risk factors for these cells must be taken into consideration.[12]

In this work radon concentrations in 12 different underground mines in Western Turkey have been measured monthly using two different methods. The annual exposure doses were estimated using a calculation programme that has been developed.

The scope of this work excludes the treatment of other hazards, mechanically, and toxic air contaminants that are characteristics of all mining operations. However, the protective measures developed to control radiation exposure may decrease other hazards. The high ventillation rates reduce both radon, radon progeny and air toxic contaminant concentrations.

Experimental

Radon concentration measurements

Mean ^{222}Rn concentrations in 5 boron, 5 coal and 2 chromium underground mines were measured for monthly intervals from October 1994 to October 1995 using Lucas cell and track etching methods. The Lucas cell used in the measurements is a 160 ml cylindrical shell with 53 mm diameter and 73 mm height. The inner surface is coated with scintillation material

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‡ WLM is a traditional unit used to describe potential alpha energy exposure.[1] One working level (WL) is 1.3×10^5 MeV of potential alpha energy per liter of air and it corresponds to an activity concentration of 100 pCi l^{-1} = 3700 Bq m^{-3} for ^{222}Rn. One WLM is exposure of 1 WL during 170 h per month or 3.5×10^{-3} Jh m^{-3}.

made of silver-activated ZnS sensitive to alpha particles. It has a quartz window and is optically coupled to a photomultiplier tube (PM) tube. The cell has two inlets, one connected to a vacuum pump to make the air with radon enter the cell after passing through a filter located at another inlet to prevent the decay products from entering the cell. Sampling and counting periods were taken as 10 min. The alpha counts in cpm were converted to radon concentrations in Bq m^{-3} using the calibration factor of 71 Bq m^{-3}/cpm obtained from a series of experiments done by a radium standard.

The average radon concentrations in the air of underground mines were also measured using nuclear track detectors. CR-39 films cut in 1 cm^2 pieces were attached to the bottom of the plastic cups, the front of them were covered with a filter to prevent the dust from entering the cups. The films in the cups were hung at different points in the galleries; after 3 months exposure they were collected and chemically processed to turn the alpha tracks in to visible etch pits. The process solution was 20% NaOH, the bath temperature 70 °C and the developing time 12 h. The calibration constant obtained was 5 kBq m^{-3}/track h^{-1} in a series of experiments using a 174.26 l tank with known radon concentrations maintained by a Ra standard.

Gamma survey

It is necessary to emphasize that underground miners are subject to the radiation of not only the radon progeny inhaled but also to the external gamma radiation from long lived radon daughters, ^{214}Bi and ^{214}Pb. Two gamma survey meters, one Scintrex-B GS-4 (Scintrex, Concord, Ont., Canada) and the other Ludlum micro-R meter (Ludlum Measurements, Sweetwater, TX, USA) were used in gamma measurements. The background counts with these survey meters were registered as 45–50 cps and 4–5 μR h^{-1}, respectively. The effective dose equivalents were calculated using the conversion factor,[13] 1 μR h^{-1} = 0.04 mSv y^{-1}.

Natural radionuclides in the ores

Since the radiation doses are closely related to the radionuclide content of the ore in the mine, the ore samples taken from the mines were analysed for their uranium, thorium and potassium concentrations. In geological samples it is generally assumed that ^{238}U and ^{232}Th are in radioactive equilibrium with ^{226}Ra and ^{228}Ra, respectively. Therefore, the concentrations determined through the activity of the decay products are named as equivalent concentrations and denoted by eU and eTh.

An ore sample of 100 g from each mine was ground, dried and then sealed in a 5.7 mm diameter cylindrical polyethylene box and left for about one month to attain equilibrium between radium and radon. Gamma spectra were taken with a 3 in × 3 in detector, 4096 channel Ortec 7010 analyser (EG & G Ortec, Oak Ridge, TN, USA) and related electronic accessories. The eU, eTh and %K contents were determined from 1.76 MeV ^{214}Bi, 2.62 MeV ^{208}Tl and 1.46 MeV ^{40}K gamma lines, respectively, with a method given elsewhere.[14,15]

Dose calculations

Definitions

Absorbed dose, *D*, is a measure of the average energy absorbed by a cell. The conventional unit is the rad and the SI unit is the gray (Gy) where 1 Gy = 100 rad.

The dose equivalent, *H*, is the product of the absorbed dose, *D*, by the quality factor *Q*: $H = D \times Q$. The conventional unit is the rem and the SI unit is the sievert (Sv) where 1 Sv = 100 rem.

Since the influence of radiation on different organs and on different individuals is not same the idea of 'tissue dose equivalent' H_T was introduced by the ICRP. It is obtained by correcting the dose equivalent using parameters such as deposition coefficient, tissue mass, working period and breathing rate explained below. Another concept in relation to total dose of individuals 'effective dose equivalent' H_E has been developed by the ICRP to place limits on the total exposure by adding all H_T values. It is obtained as the sum of the mean tissue dose equivalents multiplied by a tissue weighting factor, W_T, which accounts for the radiosensitivity of an organ or tissue, namely[7]

$$H_E = \Sigma W_T H_T + H_E(\gamma) \quad (1)$$

Calculation procedure

From 1956 a significant amount of work was realized on developing mathematical models for dose calculations. The historical development for the published dose calculations is given for ^{222}Rn progeny by James.[6] The calculations are based on the radiation delivered to lung, since it is the most sensitive organ as far as radon and its progeny are concerned.

^{222}Rn, its parent ^{226}Ra and its decay products are members of the ^{238}U decay chain. A segment of this chain that has the products of prime radiological interest owing to their potential for retention in the lung is given below.

$$\rightarrow {}^{222}\text{Rn} \xrightarrow[3.82\ \text{d}]{\alpha} {}^{218}\text{Po} \xrightarrow[3.11\ \text{m}]{\alpha} {}^{214}\text{Pb} \xrightarrow[3.82\ \text{d}]{\beta} {}^{214}\text{Bi} \xrightarrow[19.9\ \text{m}]{\beta} {}^{214}\text{Po} \rightarrow$$

Decay constants of these products and other physical parameters are used in the calculations as explained below. Three steps are followed in the mathematical computations:

Step 1. Calculations related to the medium. The calculation in this section is based on the steady state Jacobi model.[6] In this, attached and unattached fraction, potential alpha energy concentration (PAEC) in the mine air and equilibrium factors are calculated in relation to the physical parameters of the medium. These parameters are: λ_v, the probability for removal of attached and unattached product by ventilation; λ_a, the probability for attachment to aerosols or for radioactive decay to transform to the next product; λ_I, the probability for disappearance of a product with its own decay constant; $\lambda_{d,f}$ and $\lambda_{d,a}$, the removal of free or attached products by plate out on fixed surfaces; and *p*, the appearance of a free product from decay of an attached product.

The first four mechanisms here have decreasing effects on the nuclide concentrations.

The basic idea in the calculations is to set up the differential equations of daughter nuclides in attached (a) and free (f) forms and to solve them to obtain the concentrations. The general form of the differential equation is

$$dN_{i,x}/dt = (\text{rate of production})_{i,x} - (\text{rate of removal})_{i,x} \quad (2)$$

here N_i refers to product nuclei and x indicates the free or attached form of it.

Considering the incremental or decremental effect of the above parameters on the product concentrations the differential equation for the first product ^{218}Po in the free form would be written as follows:[7]

$$dN_{2,f}/dt = \lambda_1 N_1 - \lambda_r N_{2,f} \quad (3)$$

Solution of this equation gives the number of free ^{218}Po nuclei as

$$N_{2,f} = (\lambda_1 N_1/\lambda_r)(1 - e^{-\lambda r t}) \quad (4)$$

where N_1 and N_2 refer to ^{222}Rn and ^{218}Po, respectively, and

$$\lambda_r = \lambda_2 + \lambda_v + \lambda_a + \lambda_{d,f}$$

Similarly, the number of other products are also calculated.

A great deal of research has been done for the numerical values of the physical parameters used above, the results of these works have been collected and reported in ref. 6. The results show a wide range. Since there is no possibility to measure these parameters except ventillation rate in mines, for the numerical values in our calculations we used geometric means of the reported values.[6] These parameters are, in fact, closely related to the mining activities and it is not possible to measure them separately, therefore, observation of radon progeny concentrations with respect to mining activity is not done.

Step 2. Calculations related to the respiratory system. In this part of the computation procedure the fractions of the radon progeny concentrations accumulated and PAEC in different sections of the respiratory system were calculated. The respiratory system is divided into three regions,[16] the naso-pharnyx (N), bronchial trache (B) and pulmonary cells (P). The energy $DPAEC^J$ deposited in each portion of the system is computed by making use of the deposition coefficients[16] and decay constants by the relation[6]

$$DPAEC^J = (5.79\ \lambda_2\ DN_{2j} + 28.5\ \lambda_3\ DN_{3j} + 21\ \lambda_4\ DN_{4j})\ 10^{-10}\ \text{joule} \quad (5)$$

where j refers to the parts of the respiratory system, DN_{2j}, DN_{3j} and DN_{4j} are the doses from the product nuclides ^{218}Po, ^{214}Pb and ^{214}Bi deposited respectively in the regions j and λ_2, λ_3, λ_4 are the decay constants of these nuclides.

Step 3. Dose equivalents. The tissue doses were calculated using the relation[6]

$$H_T = \frac{(DPAEC)^J}{m^J} V_S T_S Q \quad (6)$$

where V_S is the breathing rate (1.2 $m^3\ h^{-1}$), T_S is the annual working period (2000 h), Q is the quality factor, which is 20 for α radiation. The mass, m^J, of tissue j is taken as 0.04 and 0.07 kg for regions B and P, respectively. The dose deposited in region N (nose, mouth and trache) is small and ignored in the calculations.

The annual effective dose equivalents from radon progeny were calculated by eqn. (1)

The weight factors, W_T, of regions B and P were accepted as 0.06.[1]

Radon itself also contributes a small amount to total dose, it is evaluated using the relation[17]

$$H_E\ (Rn) = 1.8 \times 10^{-10} A_1 \quad (7)$$

where A_1 is the radon concentration in Bq m^{-3}. H_E (γ) (for gamma exposure is calculated using the conversion factor 0.04 mSv y^{-1} = 1 μR h^{-1} given before.

Results and discussion

Radionuclide concentrations of the minerals taken from the underground mines investigated are given in Table 1.[18] As expected, U and Th concentrations are higher in coal minerals than in boron and chromium minerals. In fact, the natural radionuclide content of these latter two minerals are lower than the mean concentrations found in rocks.

Table 2 gives the range of all experimental data obtained from the measurements and of the dose equivalents calculated using the model summarised in the previous paragraphs.[18]

The parameters used in the calculations are the measured radon concentrations at different depths of each underground mine, decay properties of radon daughters, attachment fractions calculated, measured ventilation rates and weight factors for the tissues. The exposure doses in WLM y^{-1}, the tissue doses, annual effective dose equivalents due to radon progeny and due to gamma radiation were calculated at each measurement station. Overall results are summarised in Table 2.

Effect of ventilation

Ventilation rate is the most effective parameter used in the calculation of lung doses. Radon progeny concentration as well as the doses exhibit large variations with ventilation rate even in the same mine from code to code. A typical set of experimental data that shows the effect of ventilation on radon concentrations is given for coal mine II in Table 3. The measurements have shown, as expected, that increased ventilation causes a decrease

Table 1 Natural radionuclide contents of the ores

Mine	eU (ppm)	eTh (ppm)	K (%)
Coal I	4.93	3.72	0.23
Coal II	5.08	3.95	0.23
Coal III	6.43	3.98	0.28
Coal IV	6.02	4.07	0.25
Coal V	5.02	6.02	0.28
Boron I	0.12	4.88	0.17
Boron II	0.11	4.11	0.15
Chromium I	0.15	1.66	0.19
Chromium II	0.12	1.73	0.13
Chromium III	0.18	1.42	0.17
Chromium IV	0.10	1.02	0.16
Chromium V	0.15	0.72	0.18

Table 2 Ranges of average Rn concentrations, the exposure doses and the annual effective dose equivalents in mSv y^{-1}

Mine	Radon/Bq m^{-3}	Exposure dose/ WLM y^{-1}	Lung dose, H_T	Radon progeny, H_E	Gamma, H_E	Total, H_E
Boron I	63–112	0.41–0.78	26.2–50	1.57–3	0.12–0.44	1.75–3.12
Boron II	51–117	0.31–0.66	18.9–43.2	1.14–2.6	0.2–0.24	1.38–2.8
Coal I	51–96	0.21–0.67	14.2–42.3	0.86–2.5	0.2–0.36	1.06–2.7
Coal II	42–185	0.13–0.86	9.2–57.4	0.63–3.44	0.44–0.88	0.99–4.16
Coal III	33–74	0.22–0.55	13.9–34.6	0.83–2.08	0.4–0.56	1.35–2.48
Coal IV	31–156	0.2–0.95	14–60.7	0.84–3.64	0.32–0.56	1.22–4.16
Coal V	74–96	0.49–0.62	31.2–39.2	1.77–2.36	0.32–1.4	1.97–3.57
Chromium I	10–34	0.06–0.23	4.2–14.4	0.31–0.86	0.12–0.16	0.37–0.98
Chromium II	13–35	0.09–0.25	7.1–15.6	0.35–0.94	0.08–0.16	0.51–0.97
Chromium III	10–15	0.09–0.11	4.5–6.7	0.27–0.37	0.08–0.16	0.43–0.56
Chromium IV	10–20	0.11–0.78	5–10.7	0.3–0.64	0.08–0.16	0.46–0.8
Chromium V	12–34	0.08–0.24	5.3–15.2	0.32–0.91	0.08–0.16	0.4–0.99

Table 3 Average doses from coal mine II

Mine	Radon/ Bq m^{-3}	H_T/mSv y^{-1} B	P	T	Rn, H_E/ mSv y^{-1}	Gamma, H_E/ mSv y^{-1}	Total H_E/ mSv y^{-1}	WLM y^{-1}	Ventillation rate/h^{-1}
Code +32	42	7.5	1.7	9.2	0.552	0.44	0.99	0.13	2.24
Code −18	50	9.6	2.3	11.9	0.71	0.88	1.59	0.17	1.87
Code −14	56	10.8	2.6	13.4	0.8	0.48	1.28	0.19	1.87
Code +5	185	45.4	12	57.4	3.44	0.72	4.16	0.86	1.0
Code +1	60	12.3	3.1	15.4	0.92	0.68	1.6	0.22	1.60
Code +24	48	8.5	2.0	10.5	0.63	0.56	1.19	0.15	2.27

in aerosol concentration and residence time of progeny in mine air. This leads to a reduction in dose due to a large decrease in the potential alpha-emitter concentration available for deposition in the respiratory system. The reduction in radon exposure due to improved ventilation is documented for New Mexico miners as 5.40 WLM in 1967 to 0.5 WLM in 1980 and subsequently stayed at this level.[19]

Effect of mine type

In spite of the fact that there exists only natural ventilation in the galleries, relatively low average radon concentrations were observed in chromium mines. It is because, firstly, the natural radionuclide content of these ores are lower (as seen in Table 1). Secondly, the geological structure is in the form of massive rocks with low porosity which resist radon migration and emanation. As a result the lowest total annual effective dose equivalents are also low in these mines.

The lowest and the highest values for total annual effective doses are 0.37 mSv y^{-1} and 4.16 mSv y^{-1} in chromium I and coal II mines, respectively. The maximum lung dose observed was 60.7 mSv y^{-1} in, again, coal mine II. Although the data obtained for coal mines are the highest, as far as the ranges and the average values are concerned the data for boron mines are higher than the others.

Little investigation is reported in the literature for concentrations of U and Th in boron and chromium mines. In coal mines, the average activity concentration is given as 1.6 ppm for both U and Th in the Unscear report[20] based on the analysis of samples from 15 countries.

In the underground mines studied in this work the highest exposure dose, 0.95 WLM y^{-1}, again was observed in coal mine II. This is lower than the limiting value of 2 WLM y^{-1} given by the ICRP,[17] but it is higher than the average exposure dose, 0.2 WLM y^{-1} for underground coal miners obtained from studies done in different countries. In the EPA report[1] the mean annual radon decay product exposure is estimated as 0.3 WLM y^{-1} for the non-uranium miners. Our results for boron and for some coal mines are higher than this value.

This is the first work done on radiation exposure of underground miners in Turkey. The work will continue extending the study area and monitoring duration.

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