Original Scientific Paper

PUBLIC EXPOSURE TO ^{226}Ra IN DRINKING WATER*

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Received in June 2005
Accepted in February 2006

This paper presents a new method for calculating the effective dose of ^{226}Ra regularly ingested with drinking water over a long period of time. The method is based on the assessment of cumulated ^{226}Ra activity in the fraction retained in the whole body at time t (in days) after intake [so called m(t) value].
For modelling, simulation, and visualisation of the continuous intake of ^{226}Ra by drinking water, we used the Simulink® program package integrated with the Matlab®. The dose assessment was performed for ^{226}Ra activities of 5 mBq L^{-1}, 50 mBq L^{-1}, 1000 mBq L^{-1} and 5000 mBq L^{-1}. The results suggest that ^{226}Ra activities above 1000 mBq L^{-1} produce effective doses which are below the recommended maximum. However, the potential effect of ^{226}Ra activities of this extent is still unknown in children.

KEY WORDS: *effective dose, population exposure*

It is well known that water can be a source of radiation, as it contains certain amounts of naturally occurring radionuclides. Their levels in drinking water may be increased through a number of human activities such as nuclear fuel cycle and medical or other uses of radionuclides (1, 2). In addition to the control of radionuclide concentrations for radiation protection, it is very important to assess the effective dose in order to predict possible biological damage to the organism.

The effective dose received from drinking water is less than 5 % (0.1 mSv) of the total effective dose received from all natural sources (2.4 mSv). Below this reference dose level drinking water is acceptable for human consumption (1). While the World Health Organization (WHO) defines safe water consumption using the annual effective dose, other authors limit themselves to comparing radionuclide activity concentrations with reference concentrations described by the WHO or state legislation (4-13).

However, effective dose assessment is very important because it includes both metabolic and dosimetric considerations. Therefore, the aim of this paper is to show the importance of calculating time-dependent body activity concentration and the effective dose as a consequence of everyday intake of certain amount of radionuclides. This calculation was compared with the standard dose calculation recommended by the WHO on the example of ^{226}Ra, since it is one of the most common natural radionuclide in water could cause adverse effects on the human organism such as bone sarcoma and head carcinoma (3).

METHODS

The effective dose assessment proposed by the WHO

The WHO effective dose assessment is based on an assumed consumption of 2 L of water per day for one year, which equals 730 L a year. They recommended Equation 1 for the calculation of the

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*Preliminary report presented at the 6th Symposium of the Croatian Radiation Protection Association with international participation, Stubičke Toplice, Croatia, 18-20 April 2005 reference concentration of radionuclide in drinking water [Bq L^{-1}] regarding the annual effective dose of 0.1 mSv (1). On the other hand, this equation offers the assessment of the effective dose for determined radionuclide concentration in water.

$$reference\_concentration = \frac{10^{-4}[Sv/year]}{730[L/year] \cdot dose\_conversion\_factor[Sv/Bq]} \quad [1]$$

where dose conversion factor is the committed effective dose per unit intake [Sv Bq^{-1}].

The dose conversion factor for ^{226}Ra is $2.8 \cdot 10^{-7}$ Sv Bq^{-1}.

In this paper we used the Equation 1 to calculate the annual ^{226}Ra effective doses from the consumption of drinking water with ^{226}Ra concentrations of 5 mBq L^{-1}, 50 mBq L^{-1}, 1000 mBq L^{-1} and 5000 mBq L^{-1}. This calculation was also used by some other authors (4, 9, 11, 12, 14, 15).

The effective dose assessment using the calculation of cumulated activity

This method assumes that the activity present in the body is the function of time. Furthermore, the dose to the target organ depends on the activity in the source organ and on the length of time for which the activity is present. The product of time and activity is called cumulated activity, Ă [Bq s] (Equation 2), and essentially, it is the measure of total number of radioactive disintegration occurring during the time that radioactivity is present in the source organ (16).

$$\widetilde{A} \approx \int A(t)dt \quad [2]$$

Although both the WHO method and the method by calculation of the cumulated activity include the same amount of water, the second method takes into account the intake frequency which has a great effect on dose received in a determined time. For this reason, the body activity is calculated as the sum of activity from each single water intake including changes in the body activity caused by metabolism. These changes in the body activity are described by m(t), a value given by the IAEA (17). This m(t) is the fraction of a unit intake retained in the body. Its values for 20000 days are given in Figure 1.

In this paper, we calculated the body activity for 20000 days (about 55 years) according to the Equation 3:

$$A_{body}(t) = m(t) \cdot A_{water} \quad [3]$$

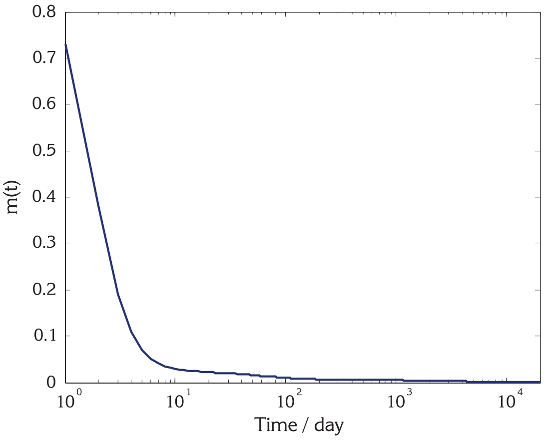

Figure 1 *m(t) values for ^{226}Ra for 20000 days*

where $A_{body}(t)$ is ^{226}Ra body activity [Bq]; and A_{water} is ^{226}Ra activity concentration of 2 L of water [Bq].

The simulation of water consumption for 20000 days and the calculation of cumulated activity were performed using a Simulink® 6.1. program package, integrated with the Matlab® 7.0.1..

The radiation dose delivered by the activity in a source organ is proportional to its cumulated activity. To calculate the radiation dose it is necessary to determine the energy emitted by cumulated activity. This energy emitted per unit of cumulated activity is described as the equilibrium absorbed-dose constant, Δ [Gy kg Bq^{-1}s^{-1}] (16). ^{226}Ra is mainly alpha emitter (Table 1) even though it does produce gamma-energies during decay, but these have a negligible influence on the equilibrium absorbed-dose constant, in the order of 10^{-16}. The product of cumulated activity and equilibrium absorbed-dose constant is the energy emitted during radioactivity is present in a source organ.

Table 1 *Energies and branching ratios of ^{226}Ra alphas and gammas (18)*

Alpha / keV	Relative frequency of ith emission / %
4314.6	0.0078
4601.9	5.5500
4784.5	94.5500
Gamma / keV	**Relative frequency of ith emission / %**
81.07	0.1700
83.78	0.3100
94.70	0.1100
97.60	0.0300
185.99	3.2800
262.27	0.0054
414.60	0.0004
449.37	0.0003
600.66	0.0006

The equilibrium absorbed-dose constant for ^{226}Ra in this study was calculated using the Equation 4 and values presented in Table 1.

$$\Delta = 1.6 \cdot 10^{-13} \sum_i N_i \cdot E_i \qquad [4]$$

where N_i is the relative frequency of ith emission; and E_i is the average energy of ith emission [MeV].

To assess the effective dose it is necessary to determine the fraction of energy emitted by the source organ that has been absorbed by the target organ. This fraction is called the absorbed fraction, Φ_i, and for ^{226}Ra it equaly 1 because ^{226}Ra is alpha emitter and consequently the emitted energy is believed to absorbed within the same organ (16). In other words the target organ and the source organ could be considered the same organ, which simplifies dose calculation.

Since m(t) refers to the total body activity, the cumulated activity calculated in this paper is the total body cumulated activity. The effective dose of ^{226}Ra can be calculated using the following equation:

$$E = \frac{\tilde{A} \cdot \Delta \cdot \Phi \cdot Q}{m} \sum_i W(E)_i \cdot k_i \qquad [5]$$

where Ă is the total body cumulated activity [Bq s]; Δ is the equilibrium absorbed-dose constant for ^{226}Ra (7.649 10^{-13} Gy kg Bq^{-1} s^{-1}); Φ is the absorbed fraction; Q is the quality factor (equalling 20 for alpha particles); m is the total body weight of the Reference Man (19) (70 kg); $W(E)_i$ is the weighting factor for the effective-dose calculation for organ i; and k_i is a fraction of the total body equivalent dose that stands for the organ i.

The coefficient k_i was calculated from equivalent dose coefficients for different organs (20), assuming an intake of 1 Bq. The effective dose was calculated using Matlab®.

RESULTS AND DISCUSSION

In this study, we calculated the effective doses of ^{226}Ra from water consumption, based on ^{226}Ra concentrations of 5 mBq L^{-1}, 50 mBq L^{-1}, 1000 mBq L^{-1} and 5000 mBq L^{-1}. Table 2 shows annual effective doses of ^{226}Ra calculated according to the Equation 1.

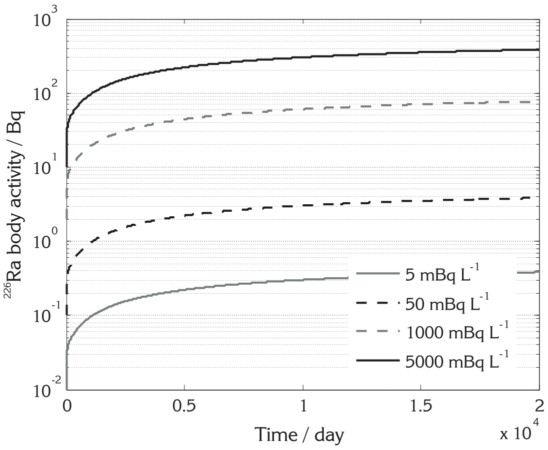

Figure 2 *Total body activity of ^{226}Ra for 20000 days following consumption of 2 L of water per day*

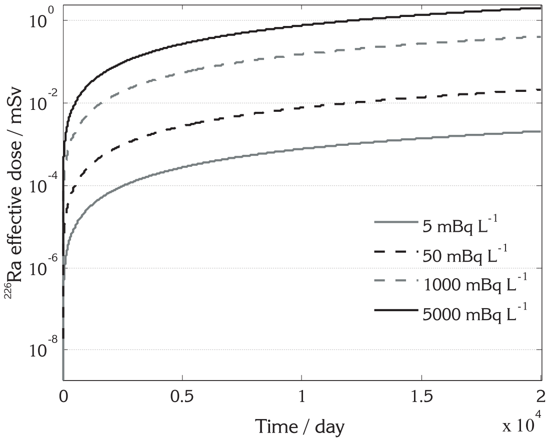

Figure 3 *^{226}Ra effective doses following consumption of 2 L of water per day*

Figures 2 and 3 show the respective total body activity and effective dose of ^{226}Ra, calculated according to the second method (Equations 3 and 5, respectively). The results are based on the intake of 2 L of water a day over 20000 days. In order to determine the annual dose, we calculated the difference between the total body effective dose at time t and the total body effective dose at time *t-365* (Figure 4).

Table 2 *Comparison between annual ^{226}Ra effective doses calculated using the WHO method and the method based on cumulated activity*

^{226}Ra concentration / mBq L^{-1}	Annual ^{226}Ra effective dose / mSv	
	1st method (WHO)	2nd method (this study) (after the first 365 days)
5	1.02x10^{-3}	6.3x10^{-6}
50	1.02x10^{-2}	6.3x10^{-5}
1000	2.04x10^{-1}	1.3x10^{-3}
5000	1.02	6.3x10^{-3}

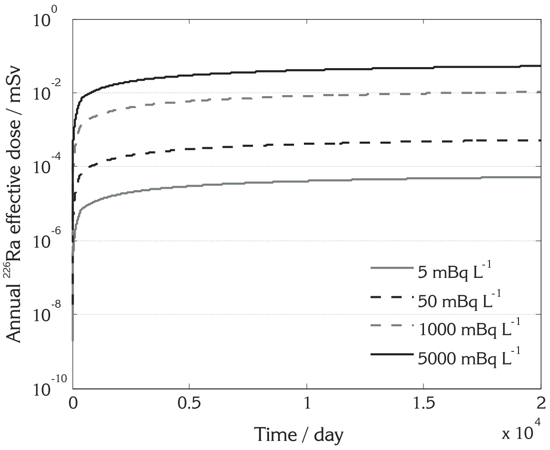

Figure 4 *Annual total body ^{226}Ra effective doses following consumption of 2 L of water per day*

The retention of ^{226}Ra in the body depends on the fractional absorption in the gastrointestinal tract (f_1), the fraction of an element entering the gastrointestinal tract which reaches body fluids. The f_1 values reported by the International Commission for Radiation Protection (ICRP) for each age group are: 0.2 for adults, 0.6 for infants and 0.3 for children between 1 and 15 years (20). The calculation of the m(t) values used in this study was based on the f_1 value for adults and the assumed half-life in body fluids of 0.25 days (17).

The calculated m(t) values suggest that a great amount of ^{226}Ra, above 70 %, is excreted from the body on the first day after the ingestion while the rest of ^{226}Ra follows the metabolic path of calcium. This means that it is largely deposited into the bone. Every following intake has the same distribution in the body. In other words, the great amount of ingested ^{226}Ra will be excreted from the body (by faeces and urine) and the rest will be distributed among organs according to the metabolic parameters.

Both metabolism and radioactive decay determine the ^{226}Ra activity concentration in a tissue (organism) (3), influencing the total body effective dose. Therefore, to assess the total body effective dose at time t it is necessary to know the exact amount of activity in the body at time t. The WHO method of calculation (Equation 1) does not specify either the frequency of everyday consumption or the changes in the body concentration in the first few days after intake. The most important factor in Equation 1 is the dose coefficient. The committed effective dose per unit intake is used for a rough assessment of received dose. It does not show dose distribution over a period of time.

In contrast, our calculation method based on cumulated activity offers a good insight into the distribution of ^{226}Ra body activity concentration over time and its effective dose. Furthermore, our method shows significantly lower values of ^{226}Ra effective dose than the first, as it takes into account the continuity of water consumption (Table 2).

Although Equation 1 provides a very simple estimation of the effective dose of ^{226}Ra, it is not recommended for the estimation of long-term exposure. Instead, the second method would be more appropriate.

Figure 2 shows that in 55 years of water consumption the total body ^{226}Ra activity rises continuously, causing a constant increase in the ^{226}Ra effective dose. However, the annual ^{226}Ra effective dose does not exceed the recommended 0.1 mSv. In other words, long-term consumption of water, even with the concentration of ^{226}Ra above 1000 mBq L^{-1} results in the allowed level of annual effective dose.

There is evidence that the retention of ^{226}Ra is greater in growing than mature organisms and that much of the variation with age is due to the elevated uptake of radium by the immature skeleton (3, 20). Consequently, it is possible that ^{226}Ra activity concentration above 1000 mBq L^{-1} will not result in an acceptable effective dose in children.

CONCLUSIONS

This paper points out a difference between two approaches to the calculation of the effective radiation dose: the WHO method and a method which includes everyday water intake of 2 L.

Since ^{226}Ra body activity drops significantly in the first few days after intake, it is very important to take into account not only the ingested activity, but also the length of time for which the activity is present in the body.

Our results show that in adults, regular consumption of ^{226}Ra through drinking water, even at activity concentrations few times over the maximum recommended value, will not lead to an effective dose higher than recommended 0.1 mSv. However, this conclusion does not apply for a child organism, since this model includes metabolic and dosimetric parameters for adult metabolism only.

REFERENCES

1. World Health Organization. Radiological quality of drinking water. In Guidelines for drinking water quality. 3rd ed. China: Sun Fung; 2004. p. 197-209 [cited 2005 May 2]. Available from: http://www.who.int/water_sanitation_health/dwq/GDWQ2004.web.pdf
2. United Nations Scientific Committee on the Effects of Atomic Radiation. Exposures from natural sources of radiation. In: Sources and effects of ionizing radiation. Report to the General Assembly with Scientific Annexes. New York: United Nations; 1993. p. 33-89.
3. Schlenker RA, Keane AT, Holtzman RB. The retention of ^{226}Ra in human soft tissue and body; implications for the ICRP 20 alkaline earth model. Health.Phys. 1982;42(5):671-93.
4. Kuo Y-C, Lai S-Y, Huang C-C, Lin Y-M. Activity concentrations and population dose from radium-226 in food and drinking water in Taiwan. Appl Radiat Isot.1997;48(9):1245-49.
5. Dávila Rangel JI, López del Río H, Mireles Garcia F, Quirino Torres LL, Villalba ML, Colmenero Sujo L, Montero Cabrera ME. Radioactivity in bottled waters sold in Mexico. Appl Radiat Isot. 2002;56:931-6.
6. Bortoli Mde**,** Gaglione P. Radium-226 in environmental materials and foods. Health Phys. 1972;22:43-8.
7. Kriege LB, Hahne RMA. ^{226}Ra and ^{228}Ra in Iowa drinking water. Health Phys. 1982;43(4):543-59.
8. Sajo-Bohus L, Gómez J, Capote T, Greaves ED, Herrera O, Salazar V, Smith A. Gross alpha radioactivity of drinking water in Venezuela. J Environ Radioactiv. 1997;35(3):305-12.
9. Martín Sănchez A, Rubio Montero MP, Gómez Escobar V, Jurado Vargas M. Radioactivity in bottled mineral waters. Appl Raiat Isot. 1999;50:1049-55.
10. Baradács E, Hunyadi I, Desző Z, Csige I, Szerbin P. ^{226}Ra in geothermal and bottled mineral waters of Hungary. Radiat Meas. 2001;34:385-90.
11. Somlai J, Horváth G, Kanyár B, Kovács T, Bodrogi E, Kávási NJ. Concentration of ^{226}Ra in Hungarian bottled mineral water. J Environ Radioactiv. 2002;62:235-40.
12. Kralik C, Friedrich M, Vojir F. Natural radionuclides in bottled water in Austria. J Environ Radioactiv. 2003;65: 233-41.
13. Dueńas C, Fernăndez MC, Liger E, Carretero J. Natural radioactivity levels in bottled water in Spain. Wat Res. 1997;31(8):1919-24.
14. Godoy MJ, Amaral ECS, Luiza M, Godoy DP. Natural radionuclides in Brazilian mineral water and consequent doses to the population. J Environ Radioactiv. 2001;53:175-82.
15. Oliveira Jde, Mazzilli BP, Oliveira Sampa MHde, Bambalas E. Natural radionuclides in drinking water supplies of São Paulo State, Brazil and consequent population dose. J Environ Radioactiv. 2001;53:99-109.
16. Cherry SR, Sorenson JA, Phelps ME. Internal radiation dosimetry. In: Physics in nuclear medicine. 3rd ed. Philadelphia: Saunders; 2003. p. 405-25.
17. International Atomic Energy Agency. Methods for assessing occupational radiation doses due to intakes of radionuclides. Safety reports series. No.: 37. Vienna: IAEA; 2004.
18. Battelle memorial institute: Software NuclideNavigator. Version 3.4. 2000.
19. International Commission on Radiological Protection. Report of the Task Group of Reference Man. Oxford, New York, Toronto, Sydney: Pergamon Press; ICRP Pub 23. 1974.
20. International Commission on Radiological Protection. Age-dependent doses to members of the public from intake of radionuclides: part 2 ingestion dose coefficients. Oxford, New York, Tokyo: Pergamon Press; ICRP Pub 67. 1993;23(3/4).

Sažetak

IZLOŽENOST STANOVNIŠTVA ^{226}Ra U PITKOJ VODI

U ovome radu opisana je metoda izračunavanja ^{226}Ra efektivne doze kao posljedice svakodnevnog pijenja vode. Procjena efektivne doze bazira se na procjeni kumulativne aktivnosti koja je izračunana s pomoću m(t) vrijednosti (udjela aktivnosti u vremenu t nakon unosa u organizam). Proces kontinuiranoga višegodišnjeg pijenja vode (20 000 dana) simuliran je u programu Simulink (programski paket Matlab). Rezultati se odnose na vode koncentracija ^{226}Ra od 5 mBq L^{-1}, 50 mBq L^{-1}, 1000 mBq L^{-1} i 5000 mBq L^{-1}. Rezultati ove metode uspoređeni su s rezultatima metode koju predlaže Svjetska zdravstvena organizacija. Pokazalo se da kontinuiranim unosom ^{226}Ra u organizam odraslog čovjeka kontinuirano raste koncentracija ^{226}Ra u organizmu. Također time raste i ^{226}Ra efektivna doza koja nakon 20000 dana doseže od 0.002 mSv (5 mBq) do 2.1 mSv (5000 mBq). Međutim, odgovarajuće godišnje ^{226}Ra efektivne doze čak i pri pijenju vode koncentracije ^{226}Ra od 5000 mBq L^{-1} ne prelaze vrijednost od 0,1 mSv, kako to predlaže Svjetska zdravstvena organizacija. Važno je naglasiti da se opisana metoda bazirala na parametrima svojstvenim odraslom organizmu te da je za procjenu doze na dječji organizam potrebno izraditi poseban model.

KEY WORDS: *efektivna doza, izloženost stanovništva*

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