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Radon Concentration in Air and Evaluation of the Radiation Dose in Villages near Shizhuyuan, Southern Hunan, China

Wanyu Tan * and Yixun Nie

School of Civil Engineering, Hunan City University, Yiyang 413000, China; nieyixun@hncu.edu.cn
* Correspondence: tanwanyu@hncu.edu.cn

Abstract: Radon is one of the important natural sources of radiation and pollutants. When radon and its progeny are inhaled by the human body, they can cause radiation damage to the respiratory system and can lead to lung cancer. Indoor and outdoor radon concentrations were measured in five villages near Shizhuyuan W-polymetallic deposit using a RAD7 detector; moreover, the corresponding radiation dose and lifetime risk probability were evaluated. The results show that the average value of indoor radon concentration was 216.6 ± 121.1 Bq m^{-3}, which is above the worldwide average indoor radon level of 40 Bq m^{-3}, and the average outdoor value was 34.6 ± 13.4 Bq m^{-3}, which is higher than the worldwide outdoor average of 10 Bq m^{-3}. A total of 42% of the dwellings investigated in our study had a higher radon level than the Chinese permissible indoor radon level of 200 Bq m^{-3}. The total annual effective dose ranged from 5.21 mSv y^{-1} to 49.38 mSv y^{-1}, with an average value of 14.63 mSv y^{-1}, which is higher than the ICRP recommended value of 3–10 mSv y^{-1}. This average total dose value corresponds to an average lifetime risk probability of 5.8% for residents in the whole study area.

Keywords: radon concentration; inhalation; annual effective dose; lifetime risk probability

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1. Introduction

Radon, which is colorless, odorless and tasteless, is a naturally occurring inert radioactive gas produced by alpha decay of radium in the uranium and thorium decay series. There are three main isotopes of radon (abbreviated to Rn), namely, ^{222}Rn ($T_{1/2}$ = 3.8 d), ^{219}Rn ($T_{1/2}$ = 3.92 s) and ^{220}Rn ($T_{1/2}$ = 54.5 s). Due to the short half-life of ^{219}Rn and ^{220}Rn, ^{222}Rn is the most abundant and hazardous radon isotope in the environment (in this paper, we only study ^{222}Rn). A part of radon generated by soil and rock containing the parent nuclide radium can transport to the atmosphere via pores or cracks, and become a significant factor influencing the health risk associated with radiation exposure from natural sources. Radon and its progeny are ubiquitous in the environment due to the wide spread of uranium, thorium and radium in the earth's crust, and are the major contributors to human exposure from natural radiation sources. Public exposure to radon and its short-lived decay products accounts for more than 50% of the dose exposure from all natural radiation sources [1].

Epidemiological studies have confirmed that exposure to high concentrations of radon and its progeny for a long time injures the extrathoracic and bronchial epithelial cells and even causes lung cancer. The International Agency for Research on Cancer categorized radon as a carcinogenic factor of type I, and 8–25% of the total lung cancer deaths worldwide are due to inhalation of radon and its decay products in air [2–4]. In addition, a certain part of the inhaled radon can dissolve in blood and then circulate in the whole human body with the blood, posing a risk of radiation exposure to other organs/tissues (e.g., bone surface, liver, kidney, small intestine and muscle) and even fetuses, which may result in other diseases such as leukemia, bone cancer, kidney cancer and fetal abnormalities [5]. Therefore, radon in the environment, especially radon in indoor air, and its radiological hazards have attracted great attention. Some international organizations have implemented radon projects worldwide and proposed permissible radon levels, e.g., USEPA [6] recommended an action level of 148 Bq m^{-3}. In addition, many scientists have studied the radon level in air as well as the potential radiation hazards in the world [2,7–18]; for example, Kudo et al. (2015) studied the internal exposure caused by the inhalation of radon and thoron progeny in Yangjiang [13], a high background radiation area in China, demonstrating that the total dose caused by radon and thoron was about two times higher than the global average. Kaur et al. (2017) estimated the annual inhalation dose due to exposure of indoor radon [2], thoron and their progeny in some villages located in sub-mountainous regions in India, showing that the dose value was below the limit recommended by ICRP. In addition, Dong (2015) measured the indoor radon concentration of typical residential areas in Urumqi [8], China, and calculated the corresponding annual effective dose to an individual and the absorbed dose to some key tissues of the individual.

Shizhuyuan W-polymetallic deposit, located in Chenzhou, Hunan Province, China, is characterized by its large-scale accumulation of W–Sn–Mo–Bi–Pb–Zn–Ag [19]. Recently, part of this region has been found to be associated with uranium mineralization and present a relatively high radon concentration in soil [20]. Some studies [21,22] have shown that the main source of indoor radon is the subjacent soil, building material, outdoor air and water, and the outdoor radon mainly originates from soil and rock containing uranium and radium. In this sense, radon levels in residential areas surrounding Shizhuyuan deposit is a significant issue; however, there is no report on the radon concentration in air in this area and the corresponding radiation risk. We studied the radon concentration in underground drinking water in the area and found that the groundwater has an abnormally high radon concentration [23]. In this study, we measured the outdoor and indoor radon concentrations in five villages near Shizhuyuan. In addition, we estimated the annual effective dose from inhalation of radon and the doses to a number of organs and tissues and to fetuses to consider the health impact on the residents.

2. Study Area

This study was carried out in five villages nearby Shizhuyuan W-polymetallic deposit (N 25°44′17″, E 113°10′7″), which is approximately 15 km from Chenzhou City in southern Hunan Province, China. A geological map of the study area is shown in Figure 1. Shizhuyuan deposit is closely related to the Qianlishan granitic complex, with strong greisen and skarn alteration. The main lithologies outcropped in this area include Precambrian gray metamorphic sandstone, Middle-Upper Devonian Shetianqiao and Qiziqiao formations, which mainly consist of moderate-to-thick-layered micritic limestone, Middle Devonian Tiaomajian Formation sandstone, Qizuqiao formation dolomous limestone, Upper Devonian poly rock and Muddy strip limestone [19]. Moreover, the uranium content in these rocks is relatively high [24]. For example, the average U content of granite is 26.4×10^{-6} g/g, which is about 10 times the average value of the continental upper crust (2.7×10^{-6} g/g). Especially in the hydrothermal Pb–Zn ore deposit, uranium mineralization is also associated locally, with a uranium average grade of 0.1% in the Jinshiling lead–zinc deposit.

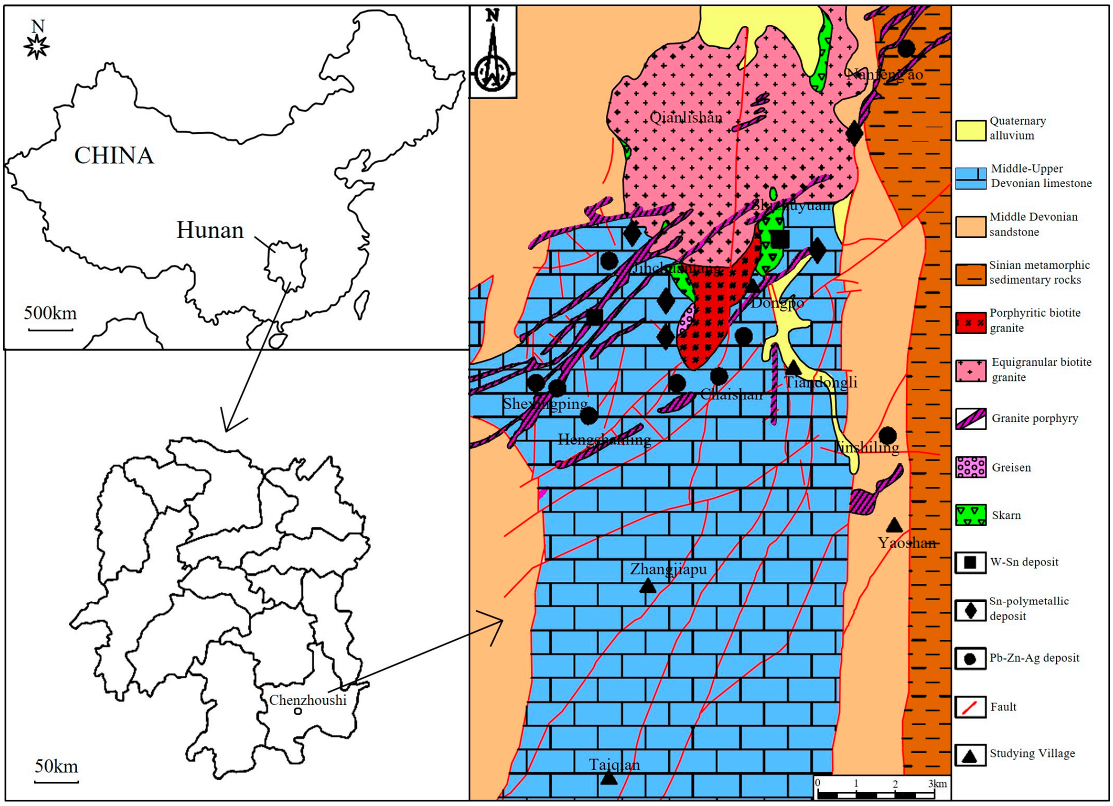

Figure 1. Geological map of the study area showing sample locations.

3. Methodology

3.1. Sampling Strategy

A total number of 77 dwellings were selected in 5 villages (i.e., Yaoshan, Tiandongli, Taiqian, Zhangjiapu and Dongpo) near Shizhuyuan deposit for radon surveys. The numbers of selected houses were 26, 7, 14, 9 and 18, respectively. The measured houses were basically selected according to the population distributions of the five villages and the residents' willingness to participate in the study. Indoor radon measurements were conducted in the center of rooms, about 1 m above the floor and away from windows. In addition, we chose 284 locations, including 80 in Yaoshan, 28 in Tiandongli, 63 in Taiqian, 55 in Zhangjiapu and 58 points in Dongpo, around these dwellings to measure the outdoor radon concentration.

3.2. Radon Concentration Measurement

Radon concentrations were measured using a RAD-7 detector (Durridge Company, Billerica, MA, USA), which was calibrated in Hunan Province Key Laboratory of Radon, China. The RAD7 detector was attached to the filter and connected to the desiccant tube, as shown in Figure 2, and then these were placed on a little table of 1 m height. Before radon measurement, the RAD7 detector was opened and purged until the humidity was lower than 10%. Sniff mode was selected, and the cycle time of the RAD7 was set to 15 min. The detailed measurement procedure is available in the user manual for the RAD7 radon detector. Considering the seasonal variation of the radon concentration in the air, each house was measured once a month for 12 months. It was measured continuously for 1 h each time, obtaining a concentration result every 15 min, resulting in 4 radon concentration data samples. A total of 48 radon concentration data samples were obtained for each house, and then the average radon concentration was calculated for each house. The measurement method for the radon concentration in outdoor air was the same as for indoor air.

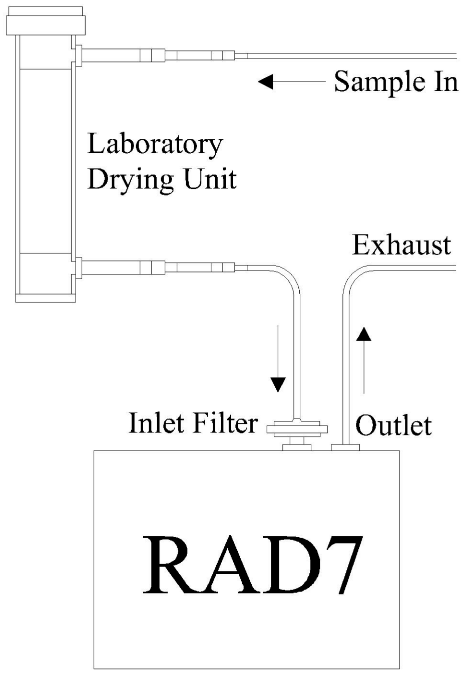

Figure 2. Diagram of radon measurement apparatus.

3.3. Estimation of Radiation Dose and Lifetime Risk Probability

The annual effective dose resulting from inhalation of radon in air can be calculated by Equation (1):

$$AED_{inh} = C \times EF \times t \times OF \times DCF_{inh} \quad (1)$$

where AED_{inh} is the annual effective dose due to radon inhalation, in mSv y^{-1}; C is the average radon concentration in indoor or outdoor air, in Bq L^{-1}; EF is the equilibrium factor between radon and its progeny; t is the total time of a year, i.e., 8760 h; OF is the occupancy factor; and DCF_{inh} is the dose conversion factor for radon inhalation, in Sv (Bq h $m^{-3})^{-1}$.

Considering that the residential characteristics (e.g., house construction and volume, ventilation mode and rate) and lifestyle of people in China are different from those of other countries, an indoor EF of 0.49 [25] and outdoor EF of 0.6 [26] were used for the estimation of AED_{inh}. As people in rural areas usually work outdoors for a longer time than those in cities, an indoor OF of 0.7 and outdoor OF of 0.3 were used in this study. In addition, the latest dose conversion convention of 13 mSv WLM^{-1} (1 WLM = 6.37 × 10^5 Bq h m^{-3}) recommended by ICRP [27] for adults was used.

In the respiratory process, a small amount of inhaled radon (indoor or outdoor) would dissolve in blood and may result in a certain radiation dose, and the annual effective dose of this part can be evaluated by the Equation (2):

$$AED_{bl} = C \times t \times OF \times DCF_{bl} \quad (2)$$

where AED_{bl} is the annual effective dose due to radon dissolved in blood via inhalation, in mSv y^{-1}; C is the radon concentration in air, Bq L^{-1}; t is the total time of a year in hour, 8760 h; OF is the occupancy factor, 0.7 for indoor and 0.3 for outdoor; DCF_{bl} is dose conversion factor for radon dissolved in blood via inhalation, and a value of 0.17 × 10^{-6} mSv (Bq h $m^{-3})^{-1}$ recommended in the UNSCEAR Report 2000 [1] was used for evaluation of AED_{bl}.

In addition, the inhalation of radon and its progeny can result in a great absorbed dose to some organs and tissues, especially lung and bronchial epithelial cells. Therefore, we calculated the annual absorbed dose (AD_i) to organs/tissues (e.g., lung, stomach, small intestine and bone surface) and to fetuses, according to the method proposed by Kendall and Smith [28]. The evaluation equation can be expressed as:

$$AD_{\rm i} = (C_{\rm in} \times H_{in} \times OF_{in} + C_{out} \times H_{out} \times OF_{out}) \times t \times q_i \quad (3)$$

where AD_i is the annual absorbed dose to organ/tissue i, in mSv y^{-1}; C_{in} and C_{out} are indoor and outdoor radon concentrations, respectively, in Bq L^{-1}; H_{in} and H_{out} are the indoor and outdoor respiratory rates, assumed to be 0.78 m^3 h^{-1} and 1.2 m^3 h^{-1}, respectively; t is the total time of a year in hours, 8760 h; OF_{in} is the indoor occupancy factor (0.7) and OF_{out} is the outdoor occupancy factor (0.3); q_i is the dose coefficient from inhaled radon decay product for organ/tissue i, in mSv Bq^{-1}, and the value of q_i is available from Kendall and Smith [28] (2002); here we selected the q_i of F type.

Based on the linear no-threshold dose–response curve hypothesis and detriment adjusted nominal risk coefficients, the lifetime risk due to inhalation of radon can be estimated by the following equation:

$$LRE = AED \times LE \times NRC \quad (4)$$

where LRE is the evaluated lifetime risk probability; LE is the average lifetime expectancy (70 years in China); AED is the annual effective dose due to inhalation of radon and its daughters, in mSv y^{-1}; NRC is the nominal risk coefficient [29], and the values for cancer and heritable effects are 5.5×10^{-2} Sv^{-1} and 0.2×10^{-2} Sv^{-1}, the sum being 5.7×10^{-2} Sv^{-1} (ICRP, 2007) [29].

4. Results and Discussion

4.1. Radon Concentration in Air

The measured indoor and outdoor radon concentrations are listed in Table 1. The data show that the indoor radon concentration ranges from 72.3 Bq m^{-3} to 705.6 Bq m^{-3}, with an average value of 216.6 $\pm$ 121.1 Bq m^{-3}, which is higher than the Chinese and worldwide indoor average levels of 43.8 Bq m^{-3} and 40 Bq m^{-3}, respectively. This average is higher than the Chinese permissible indoor radon level of 200 Bq m^{-3} (CEPA, 2010 [30]) and the USEPA recommended action level of 148 Bq m^{-3} (USEPA, 2003 [6]). In fact, 42% of the dwellings investigated in our work have a radon level higher than 200 Bq m^{-3}. Compared with the indoor radon concentration of other Chinese regions, the average radon level of the whole study area is higher than those measured in Yangjiang [13], Guangzhou [31] and Urumqi [8]. Moreover, it is significantly higher than the average indoor levels of 42.8 Bq m^{-3} in Hunan Province [26] and 64.7 Bq m^{-3} in Chenzhou city [32].

Table 1. Radon concentrations in air of residential areas near Shizhuyuan (Bq m^{-3}).

Location	Indoor					Outdoor				
	Houses	Min	Max	Average	σ	Points	Min	Max	Average	σ
Yaoshan	26	72.3	705.6	254.1	168.9	80	21.6	167	43.2	20.0
Tiandongli	7	103.6	285.7	183.2	59.9	28	16.9	53.5	30.2	8.1
Taiqian	14	89.6	418.2	207.8	97.0	63	20.7	44.5	32.1	5.8
Zhangjiapu	9	81.9	212	157.2	44.4	55	17.1	41.2	29.4	5.8
Dongpo	18	78.7	331.1	212.1	84.2	58	16.7	83.5	32.8	9.5
Study area	74	72.3	705.6	216.6	121.1	284	16.7	167.0	34.6	13.4

It can be seen from Table 1 that the outdoor radon concentration varies from 16.9 Bq m^{-3} to 167.0 Bq m^{-3} with a mean value of 34.6 $\pm$ 13.4 Bq m^{-3}. This value is more than twice the Chinese average outdoor radon concentration [26] of 14 Bq m^{-3} and more than three times the worldwide average level of 10 Bq m^{-3} but is still low compared with the USEPA action level. The outdoor average concentration in our work is higher than the mean level (26.3 Bq m^{-3}) of Hunan Province reported by Pan [26].

Radon in air mainly comes from the exhalation of radon in rocks and soils. The concentration of ^{222}Rn in air is mainly affected by the abundance and geochemical properties of parent radionuclides (i.e., ^{238}U and ^{226}Ra) of the surrounding rocks and soils. The magmatic activity and magmatic–hydrothermal system in Shizhuyuan area brought a large number of metals such as W, Sn, Bi, Mo, Pb, Zn and Ag, which form some large-scale polymetallic deposits. They also brought the radioactive element U, which produces a relatively high uranium background and a relatively high radon concentration in indoor and outdoor air. Especially in Yaoshan Village, which is located near the Jinshiling Pb–Zn–Ag–(U) deposit, with obvious enrichment and mineralization of U, the average indoor radon concentration is significantly higher than that in other villages. The highest radon concentration of indoor and outdoor air also occurs in Yaoshan Village and is near the ore vein of Pb–Zn–Ag–(U) with about 0.1% U.

4.2. Radiation Dose and Lifetime Risk Probability

The annual effective dose (AED_{inh}) from inhalation of indoor and outdoor radon for residents of the study area are presented in Table 2. The value of AED_{inh} due to indoor radon is in the range of 4.43 mSv y^{-1} to 43.25 mSv y^{-1}, with an average value of 13.28 mSv y^{-1}, which is much higher than the worldwide average annual dose of 2.45 mSv y^{-1}, calculated using Equation (1) with the worldwide indoor radon level of 40 Bq m^{-3}. This mean value is also above the limit of 3–10 mSv recommended by ICRP [29]. In comparison, the value of AED_{inh} due to outdoor radon is much lower than that due to indoor radon, ranging from 0.54 mSv y^{-1} to 5.37 mSv y^{-1}, with a mean of 1.11 mSv y^{-1}. This is above the worldwide average annual effective dose of 0.32 mSv y^{-1} corresponding to the average global outdoor radon concentration of 10 Bq m^{-3}. The annual effective dose due to radon dissolved in blood via inhalation was evaluated and is summarized in Table 2. The value of AED_{bl} ranges from 0.08 to 0.74 mSv y^{-1}, with an average of 0.23 mSv y^{-1}, and from 0.01 to 0.02 mSv y^{-1}, with a mean of 0.01 mSv y^{-1}, for indoor and outdoor radon, respectively. These results also show that the AED_{bl} value due to outdoor radon is much lower than that due to indoor radon and can be ignored.

Table 2. The annual effective dose due to inhalation of radon and its progeny in air (mSv y^{-1}).

Location		**AED_{inh}**		**AED_{bl}**		**Total**
		Indoor	**Outdoor**	**Indoor**	**Outdoor**	
Yaoshan	Min	4.43	0.69	0.08	0.01	5.21
	Max	43.25	5.37	0.74	0.02	49.38
	average	15.58	1.39	0.25	0.01	17.23
Tiandongli	Min	6.35	0.54	0.10	0.01	7.00
	Max	17.51	1.72	0.31	0.02	19.56
	average	11.23	0.97	0.19	0.01	12.40
Taiqian	Min	5.49	0.67	0.09	0.01	6.26
	Max	25.63	1.43	0.44	0.02	27.52
	average	12.74	1.03	0.22	0.01	14.00
Zhangjiapu	Min	5.02	0.55	0.09	0.01	5.67
	Max	12.99	1.33	0.22	0.01	14.55
	average	9.64	0.95	0.16	0.01	10.76
Dongpo	Min	4.82	0.54	0.08	0.01	5.45
	Max	20.29	2.69	0.35	0.02	23.35
	average	13.00	1.06	0.22	0.01	14.29
Study area	average	13.28	1.11	0.23	0.01	14.63
Worldwide average		2.45	0.32	0.04	0.004	2.81

Note: the worldwide average is evaluated using Equation (2) with the average worldwide indoor radon concentration of 40 Bq m^{-3} and outdoor of 10 Bq m^{-3}.

Table 2 also shows that the total annual effective doses from indoor and outdoor radon for an inhabitant who spends 70% of his/her time indoors and 30% of his/her time outdoors in the five villages are very large. The average values of total annual effective doses are 17.23 mSv y^{-1}, 12.40 mSv y^{-1}, 14.00 mSv y^{-1}, 10.76 mSv $y^{-1}$and 14.29 mSv y^{-1} for resident in Yaoshan, Tiandongli, Taiqian, Zhangjiapu and Dongpo, respectively, which are all above the ICRP recommended action level of 3–10 mSv y^{-1} (ICRP, 2007) [29].

For better assessment of the radiological risk to the residents, we also calculated the doses from radon and its progeny to some organs/tissues and to fetuses. The estimated doses are summarized in Table 3. The data show that the lung, kidney and bone surface receive a higher dose than other organs/tissues selected in our study, with lung > kidney > bone surface. Except for the above three organs, the rest of the organs/tissues receive a dose about 1 to 2 orders of magnitude lower than that to lung, e.g., the average dose to fetuses is very low (0.04 mSv y^{-1}) and can nearly be ignored.

Table 3. Summary of annual doses from radon and its progeny to various organs/tissues and fetuses (mSv y^{-1}).

Location		Lung	Stomach	Small Intestine	Colon	RBM [a]	Bone Surface	Liver	Breast	Kidney	Gonads	Brain	Bladder	Muscle	Fetus
Yaoshan	Min	10.35	0.05	0.05	0.05	0.08	0.41	0.12	0.04	1.49	0.04	0.05	0.06	0.04	0.02
	Max	97.54	0.51	0.47	0.43	0.74	3.90	1.13	0.39	14.05	0.39	0.43	0.55	0.39	0.16
	average	33.79	0.18	0.16	0.15	0.26	1.35	0.39	0.14	4.87	0.14	0.15	0.19	0.14	0.06
Tiandongli	Min	13.72	0.07	0.07	0.06	0.10	0.55	0.16	0.05	1.98	0.05	0.06	0.08	0.05	0.02
	Max	38.38	0.20	0.18	0.17	0.29	1.54	0.45	0.15	5.53	0.15	0.17	0.21	0.15	0.06
	average	24.29	0.13	0.12	0.11	0.18	0.97	0.28	0.10	3.50	0.10	0.11	0.14	0.10	0.04
Taiqian	Min	12.35	0.06	0.06	0.05	0.09	0.49	0.14	0.05	1.78	0.05	0.05	0.07	0.05	0.02
	Max	53.51	0.28	0.26	0.24	0.41	2.14	0.62	0.21	7.71	0.21	0.24	0.30	0.21	0.09
	average	27.38	0.14	0.13	0.12	0.21	1.10	0.32	0.11	3.94	0.11	0.12	0.15	0.11	0.04
Zhangjiapu	Min	11.14	0.06	0.05	0.05	0.08	0.45	0.13	0.04	1.60	0.04	0.05	0.06	0.04	0.02
	Max	28.60	0.15	0.14	0.13	0.22	1.14	0.33	0.11	4.12	0.11	0.13	0.16	0.11	0.05
	average	21.11	0.11	0.10	0.09	0.16	0.84	0.24	0.08	3.04	0.08	0.09	0.12	0.08	0.03
Dongpo	Min	10.73	0.06	0.05	0.05	0.08	0.43	0.12	0.04	1.54	0.04	0.05	0.06	0.04	0.02
	Max	46.17	0.24	0.22	0.20	0.35	1.85	0.54	0.18	6.65	0.18	0.20	0.26	0.18	0.08
	average	27.95	0.15	0.13	0.12	0.21	1.12	0.32	0.11	4.02	0.11	0.12	0.16	0.11	0.05
Study area	average	28.63	0.15	0.14	0.13	0.22	1.15	0.33	0.10	4.12	0.11	0.13	0.16	0.11	0.05
Global average [b]		5.57	0.03	0.03	0.02	0.04	0.22	0.06	0.02	0.80	0.02	0.02	0.03	0.02	0.01

Note: (a) RBM is the red bone marrow; (b) the global average dose is calculated using Equation (3) with the average worldwide indoor radon concentration of 40 Bq m^{-3} and outdoor of 10 Bq m^{-3}.

The analysis above demonstrates that the exposure to indoor radon due to inhalation is a substantial part of radiological hazards in the study area. Thus it is better to take measures to reduce exposure to radon, such as increasing indoor ventilation, removing radon progeny in air by filters and using radon-proof material to decorate houses, especially the floor.

Based on the average total annual effective doses listed in Table 2, the lifetime risk for residents in the five villages was calculated, and the results are summarized in Table 4. The lifetime cancer risks for residents in Yaoshan, Tiandongli, Taiqian, Zhangjiapu and Dongpo are 6.6%, 4.8%, 5.4%, 3.1% and 5.5%, respectively. In comparison, the lifetime risk for heritable effects is relatively low, ranging from 0.1% to 0.2%. The average lifetime risk corresponding to the average total annual effective doses for residents in the whole study area is 5.8%, including 5.6% for cancer and 0.2% for heritable effects.

Table 4. The mean total effective dose and lifetime risk probability for residents in the study area due to inhalation of radon and its decay products.

Location	Mean Total Effective Dose (mSv y^{-1})	Lifetime Risk Probability ($\times 10^{-2}$)		
		Cancer	Heritable Effects	Total
Yaoshan	17.23	6.6	0.2	6.8
Tiandongli	12.40	4.8	0.2	5.0
Taiqian	14.00	5.4	0.2	5.6
Zhangjiapu	8.06	3.1	0.1	3.2
Dongpo	14.29	5.5	0.2	5.7
Study area	14.63	5.6	0.2	5.8

5. Summary and Conclusions

In this study, measurement of radon concentrations in indoor and outdoor air was carried out in the villages nearby Shizhuyuan W-polymetallic deposit to evaluate the health risk from radon to the residents. The results display that the average indoor and outdoor radon levels are much higher than the Chinese and global average indoor and outdoor radon levels, and 42% of the dwellings investigated in our work have a higher radon level than the Chinese permissible indoor radon level of 200 Bq m^{-3}. The average value of the total annual effective dose from indoor and outdoor radon is above the ICRP's recommended value, and the corresponding lifetime risk for residents in the whole study area is 5.8%. Moreover, the data analysis demonstrates that the exposure to indoor radon due to inhalation is a substantial part of radiological hazards in the study area. Therefore, it is better to take measures to reduce exposure to radon, such as increasing indoor ventilation, removing radon progeny in air by filters and using radon-proof material to decorate the houses, especially the floor. There are a large number of non-ferrous metal deposits similar to Shizhuyuan in Chenzhou, Hunan Province. Previously, these deposit areas were used to focus only on the environmental issues of heavy metals. This study shows that the radiation environment of radon in these non-ferrous metal mineralization areas should also be given attention.

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