Rapid estimation of environmental radioactivity surrounding Xiangshan uranium deposits, Jiangxi province, Eastern China

Abstract. The surveys of terrestrial gamma dose rate, radon concentration indoor and in water and specific activity of radionuclides of soil were carried out in 14 villages and a town in Xiangshan uranium deposit and surrounding area, Jiangxi province, Eastern China, in 2017–2018, using a scintillator dosemeter, an ionization chamber and a high-purity germanium gamma spectrometer to study radiation status in these places after remediation. A radioactive hot spot was discovered in a village near the mining of a waste rock heap where specific activity of $^{238}$U, $^{226}$Ra, $^{232}$Th and $^{137}$Cs of soil was as high as $1433 \pm 76$ Bq/kg, $1210 \pm 62$ Bq/kg, $236 \pm 13$ Bq/kg and $17 \pm 1.1$ Bq/kg, respectively. The dose rate on a waste rock heap was about 2423 nGy/h. Approximately 50% of the houses in a village near the uranium mining site had radon concentrations that exceeded 160 Bq/m$^3$. There was a significant positive correlation between indoor radon concentration and outdoor gamma dose rate ($R^2 = 0.7876$). Abnormal radon concentration was observed in a rising spring sample providing residents with tap water up to 127.1 Bq/l. Four tap water samples and three of five well water samples exceeded the limit of radon concentration of drinking water in China (11.1 Bq/l). The mean annual effective doses from gamma dose rate data were 0.86 mSv/y and 1.13 mSv/y for indoor radon. The study shows that there are some radioactively contaminated places surrounding the Xiangshan uranium mine. The local outdoor dose rate averages may be used to estimate local indoor radon concentrations.

Keywords: gamma dose rate • indoor radon • water radon • cesium • annual effective dose • Xiangshan uranium deposit

Introduction

The issue of high-level exposure to natural radiation caused by uranium mining has been focused by governments and public organizations all over the world. Studies of miners exposed to radon and its decay products provide a direct basis for assessing lung cancer risk, and epidemiological studies of miners and residential exposure to radon and its decay products are carried out [1–3]. In recent years, detailed radioactive assessments of uranium mines and treatment plants have been conducted in Kazakhstan, Kyrgyzstan, Uzbekistan and Tajikistan, including surveys of gamma dose rate and indoor $^{222}$Rn/$^{220}$Rn concentration. The effective dose in the vicinity of uranium mine tailings and nuclear facilities reached to several tens of mSv/y [4], and annual radiation doses of several hundred mSv could be received as a consequence of using abandoned radioactive filtration materials for insulation in the houses in Minkush [5–7]. Although Portugal’s survey of 60 uranium mining areas displayed that most mining sites did not have radionuclide enrichment, radioactive anomalies
found that near mining facilities and tailings were 200 times higher than those in unaffected areas [8]. The gamma dose rate on the surface of uranium mine tailings in Jaduguda, India, and nearby areas varied from 0.8 to 3.3 μGy/h with an average of 1.4 μGy/h, while the local background level was 0.1–0.15 μGy/h [9]. Serbia found that the terrestrial gamma dose around an old uranium mine was twice that of the background area [10]. The uranium content in the soil of the uranium deposit in western Namibia was up to 1865.5 ± 56 Bq/kg [11]. The Brazilian uranium deposit in Paraíba had a high gamma radiation dose level, and the annual effective dose range was 0.01–19.11 mSv, with an average of 2.64 mSv [12]. These results displayed that further environmental remediation is needed in some places. In China, there are few studies and reports on radioactive status surrounding uranium mines, although environmental remediation works have been performed in some uranium mines since the later of 1980s [13]. In Yunnan province, outdoor radon concentration on a tailings dam was greater than 173 Bq/m² and the depth of soil contaminated, due to 232U and 226Ra enrichment in stream sediments, was about 90 cm with the maximum distance of 800 cm [14]. The maximum activity concentration of 232U in the topsoil in Xiaozhuang uranium deposit was about 442 Bq/kg [15]. However, there are few reports on levels of terrestrial gamma dose rate and indoor radon in residential buildings surrounding uranium mines other than tailing dams and nuclear facilities in China.

Uranium prospecting and uranium mining started between 1955 and 1958 in China [16]. Xiangshan uranium deposit, one of the largest uranium deposits in China, was discovered in 1957 by airborne radioactive survey, and it began open-pit mining in 1966, while underground mining started in 1960s [16, 17]. Xiangshan uranium ore field (XUOF) is located in Xiangshan volcanic basin, which is famous for volcanic hydrothermal uranium deposits in China. There are more than 20 uranium deposits hosted in XUOF. The area of Xiangshan basin is about 300 square kilometres with east-west length of about 26 km and north-south width of 12 km [18–20]. Therefore, it is beneficial to carry out an environmental radioactive survey and assessment of the level of gamma dose rate, radon concentration indoor and in water and annual effective dose in XUOF.

A rapid estimation method and radiological dose for environment radiation are presented based on the measured data of indoor and outdoor gamma dose rates, radon concentration indoor and in water and specific activity of radionuclide of soil in 14 villages and a town surrounding Xiangshan uranium deposit in 2017–2018.

Materials and methods

Sample collection and pretreatment

All soil samples were collected from 0 cm to 5 cm topsoil, and sampling sites were located using handheld GPS precisely positioning in this survey. Soil samples were put into plastic bags numbered in the field after removing plants and stones in them, to be sure that the weight of each sample was not less than 1 kg.

Soil samples were ground to less than 80 mesh powder, and then they were dried at Celsius temperature of 105 degree for more than 8 hours in the laboratory. Dried soil powder was packaged in a sealed plastic box for one month to reach the radioactive equilibrium between radium and radon. Finally, the specific activity of uranium, radium, thorium, potassium and cesium in the soil sample was determined by a high-purity germanium (HPGe) gamma spectrometer, described in detail in the section ‘Specific activity of radionuclides of soil’.

Gamma dose rate

A portable plastic scintillator dosemeter was used for indoor and outdoor gamma dose rate measurements (CKL3120, made in China). The dose rate at every site was the arithmetic mean of three readings multiplied by calibration factor, and each reading was the average of ten measurements after subtracting the dose rate caused by cosmic ray. The dosemeter was calibrated at the National Institute of Metrology of China, where the dose rate range of the calibration device (environment level) was from $1 \times 10^{-8}$ Gy/h to $1 \times 10^{-4}$ Gy/h. The dosemeter used in this survey has a calibration factor of 1.14 (the expansion uncertainty of the calibration factor is 15%). The measured gamma dose rate ranged from 76.3 nGy/h to 2423 nGy/h in the Xiangshan area of Jiangxi province.

Indoor radon concentration

Indoor radon concentration was measured using an ionization chamber radon analyser (AlphaGUARD, Germany) with continuously flow mode and sampling time of 10 minutes. The AlphaGUARD was placed on the top of a wooden table with the height of 90 cm. Radon measurement was generally performed in the living room of a residential house in the first floor continuously for 3 hours. The AlphaGUARD was calibrated in Germany and in the reference radon chamber at the National Institute of Metrology of China. Meanwhile, indoor radon concentration and equilibrium equivalent radon concentration were detected using a semiconductor radon monitor ERS-RDM-2S (Tracerlab CLd., Germany).

The ERS/RDM-2S is a dual-channel radon/thoron and progeny monitor. This device for measuring radon decay products is used to measure potential alpha energy concentration (Cp) and activity concentration of single nuclides (Ci; Po-218, Pb-214, Bi/Po-214). Measurements are carried out in sampling of decay products on filters using air flow through the filters driven by a pump and measurement of alpha radiation of sampled nuclides. So the results of equilibrium equivalent radon concentration (EECRn) are given in accordance to the setting up of counting interval [21].
Specific activity of radionuclies of soil

Specific activity of $^{238}$U, $^{226}$Ra, $^{232}$Th, $^{40}$K and $^{137}$Cs in soil samples was determined by an HPGe gamma spectrometer, which consists of a broad energy germanium detector (BE3830, Canberra), a digital multichannel spectrometer (Inspector 2000), a low background lead chamber and a cryogenic liquid nitrogen refrigeration system. The energy resolution of the spectrometer is 1.67 keV at 1332 keV @ Co-60. Genie 2000 software package was used for spectra data processing.

The specific radioactivity concentrations of $^{238}$U, $^{226}$Ra, $^{232}$Th and $^{40}$K in soil samples were evaluated from the counts of the photo peaks of $^{234}$Th (63.3 keV), $^{214}$Pb (351.9 keV) and $^{214}$Bi (609 keV), $^{208}$Tl (583.2 keV) and $^{228}$Ac (911.2 keV), and $^{40}$K (1460 keV), respectively. The activity is calculated by formula (1) [22]:

$$Q_j = \frac{A_{ji} - A_{ib}}{P_j \eta_i W D_j}$$

where $Q_j$ is activity concentration [Bq/kg], $A_{ji}$ is the net area of the $i$th photo peak of $j$th nuclide [counts/s], $A_{ib}$ is the background net area of the $i$th photo peak of the $j$th nuclide [counts/s], $P$ is the emission probability of the $i$th gamma ray emitted by the $j$th nuclide, $\eta_i$ is the detection efficiency value corresponding to the $i$th gamma ray full energy peak, $W$ is the sample mass [kg] and $D_j$ is decay correction coefficient of the $j$th nuclide corrected to sampling time.

Water sample collection and water radon measurement

Water samples for radon survey in water were collected from tap water and well water of the resident houses. The water samples were collected following the procedure described in the ‘Radon in Water Sampling Program’ [23]. The bottle for tap water sample was filled with fresh water after the pipe valve was turned on for at least 2 min, while well water sample was taken from the deep groundwater after pumping several times. Every plastic bottles for sampling had a volume of 250 ml. Radon concentration in water was measured using the radon monitor AlphaGUARD PQ2000 PRO (Genitron, Frankfurt, Germany) by continuous measurement mode with 10 minutes sampling time; the procedure and radon calculation equation are presented in detail in the article of Li et al. [24].

Results and discussion

Overview of the study area

The study area is located in the central part of Xiangshan basin, and Xiangshan Mountain is the highest peak in the study area. The outcrops mainly consist of the upper Jurassic dyke, the rhyolite dyke, the dark purple red sandstone and the conglomerate.

<table>
<thead>
<tr>
<th>Location</th>
<th>Mean</th>
<th>Standard deviation</th>
<th>Range</th>
</tr>
</thead>
<tbody>
<tr>
<td>Villages and town</td>
<td>Outdoor</td>
<td>123</td>
<td>39</td>
</tr>
<tr>
<td>J</td>
<td>Indoor</td>
<td>145</td>
<td>23</td>
</tr>
<tr>
<td>FS</td>
<td>Outdoor</td>
<td>251</td>
<td>12</td>
</tr>
<tr>
<td>YK</td>
<td>Outdoor</td>
<td>1592</td>
<td>1174</td>
</tr>
<tr>
<td>F</td>
<td>Outdoor</td>
<td>662</td>
<td>4.3</td>
</tr>
<tr>
<td></td>
<td>Outdoor</td>
<td>449</td>
<td>199</td>
</tr>
</tbody>
</table>
site YK with dose rate 662 nGy/h, which was near Village 14. After investigation, there were some scattered uranium ores.

Radon concentration in the dwellings

Radon and the equilibrium equivalent radon concentration

All indoor radon measurements were performed in the living room on the first floor in good air exchange condition, which was called a normal living environment. The doors and windows were not closed during radon measurement.

The mean indoor radon concentration of 14 villages and a town is listed in Table 2. There are two sites with high indoor radon concentration. The arithmetic mean in site YKB and Village 14 was 190 ± 9 Bq/m³ and 131 ± 90 Bq/m³, respectively. According to multiple measurements in Village 14, about 50% of the houses had an indoor radon concentration greater than 160 Bq/m³. The average radon concentration of 8 houses in Village 4 was 72 ± 56 Bq/m³, ranging from 25 Bq/m³ to 191 Bq/m³, where about 25% of

Table 2. Indoor radon concentration in the Xiangshan area

<table>
<thead>
<tr>
<th>Map location</th>
<th>AlphaGUARD [Bq/m³]</th>
<th>ERS-RDM-2S [Bq/m³]</th>
<th>Relative error [%]</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>n</td>
<td>Mean/C₁</td>
<td>SD</td>
</tr>
<tr>
<td>1</td>
<td>3</td>
<td>29</td>
<td>3.7</td>
</tr>
<tr>
<td>2</td>
<td>2</td>
<td>40</td>
<td>4.9</td>
</tr>
<tr>
<td>3</td>
<td>3</td>
<td>32</td>
<td>12.5</td>
</tr>
<tr>
<td>4</td>
<td>8</td>
<td>72</td>
<td>55.5</td>
</tr>
<tr>
<td>5</td>
<td>2</td>
<td>29</td>
<td>12.4</td>
</tr>
<tr>
<td>8</td>
<td>2</td>
<td>24</td>
<td>1.7</td>
</tr>
<tr>
<td>9</td>
<td>2</td>
<td>54</td>
<td>20.4</td>
</tr>
<tr>
<td>10</td>
<td>2</td>
<td>23</td>
<td>2.6</td>
</tr>
<tr>
<td>12</td>
<td>2</td>
<td>40</td>
<td>2.4</td>
</tr>
<tr>
<td>13</td>
<td>2</td>
<td>34</td>
<td>6.9</td>
</tr>
<tr>
<td>14</td>
<td>4</td>
<td>131</td>
<td>89.7</td>
</tr>
</tbody>
</table>

Notes: n is number of samples at the map location. In every building, the measurements were taken at least 8 times by AlphaGUARD with 10 minutes sampling time. Mean is the arithmetic mean, and SD is the standard deviation of the samples. SD* is standard deviation values displayed by the ERS-DOM-2S.
the dwellings were recorded to have radon concentrations higher than 100 Bq/m³. Village 4 was close to the ore processing plant, YKB was a uranium mining site and Village 14 was the nearest small village close to the mining area.

Table 2 displays that results measured by ionization chamber and semiconductor radon monitor are in good agreement. About 83% radon measurements have relative error less than ±30%, except those in Village 5.

After comprehensively analysing the measured data, it was found that indoor radon concentration in the Xiangshan area will be greatly increased because indoor radon concentration in each village obtained in this survey was measured under the normal ventilation conditions of normal life.

As we know, if the doors and windows are closed, the indoor radon concentration will generally increase, because the radon released from the soil and building materials will accumulate in the confined space and eventually reach a dynamic equilibrium. The research results showed that indoor air exchange rate has an effect on indoor radon and its daughter concentration [25]. The radon concentration in a room with closed doors and windows is significantly higher than that in the state of opening the doors and windows. If the new building is well sealed, resulting in low indoor air exchange rate, the indoor radon concentration will increase. Therefore, it will receive a higher dose exposure of radon in a closed room. According to articles [26, 27], indoor radon concentration in China increased significantly at present compared with that in 1980s.

However, people living in a village in southern China, due to the high air temperature and humidity, are at least in a good air exchange conditions of adequate convection and natural ventilation in three seasons (spring, summer and autumn) every year. Therefore, it is more representative to measure the indoor radon concentration in this normal living style.

The average radon concentrations and equilibrium equivalent radon concentrations in the same room are shown in Fig. 3. The equilibrium radon concentration varies greatly from house to house, and its variation trend is inconsistent with that of the radon concentration measured by the AlphaGUARD and ERS-RDM-2S. The inconsistency might be caused by convection of the rooms and air humidity. Of course, the number of samples we surveyed in each village is not enough, so the equilibrium equivalent radon concentration is a reference.

**Indoor radon concentration and outdoor dose rate**

It is found that a significant positive correlation existed between the average indoor radon concentration (in good air exchange condition) and average outdoor dose rate in each village, with the goodness of fit $R^2 = 0.7876$. The results are shown in Fig. 4.

There are some articles that discussed the possible relationship between indoor radon concentration and outdoor gamma dose rates. Bossew et al. proposed in his article [28]: ‘A further use could be found in terrestrial dose rate as a proxy of the geogenic radon potential, as both quantities are related by partly the same source, namely uranium content of the ground’. Stojanovska et al. [29] analysed the simultaneous measurement of indoor and outdoor gamma dose rates and radon concentrations in the Republic of Macedonia, suggesting that ‘the possibility to predict $R_{min}$ with a model of a linear functional dependence of $H_{out}$ has proved possible, but with a low coefficient of determination ($\rho^2 = 0.13$)’.

Figure 4 shows us an example of positive relationship between the average indoor radon level in a high ventilation rate and the outdoor dose rate.
A total of 29 soil samples were collected from the Xiangshan area. Specific activity (radioactivity) of natural radionuclides $^{238}\text{U}$, $^{226}\text{Ra}$, $^{232}\text{Th}$ and $^{40}\text{K}$ and artificial radioactive nuclide $^{137}\text{Cs}$ in soil were determined by an HPGe gamma spectrometer in the laboratory. The results are listed in Table 3.

Table 3 shows that specific activities of $^{238}\text{U}$ series elements are significantly high. The numbers of samples with high concentration of $^{238}\text{U}$ and $^{226}\text{Ra}$, more than 100 Bq/kg, account for about 41% and 28% of all samples, respectively. The mean values of $^{238}\text{U}$ and $^{226}\text{Ra}$ were 119 ± 97 Bq/kg and 86 ± 56 Bq/kg, ranging from 30 Bq/kg to 511 Bq/kg and 31 Bq/kg to 321 Bq/kg, respectively, not including the value of site 21.

Site 21, a radioactivity hot spot found in the top layer soil in our investigation, was located near the uranium mine and Village 14, where the concentration of $^{238}\text{U}$ and $^{226}\text{Ra}$ of the soil sample was 1433 ± 76 Bq/kg and 1210 ± 62 Bq/kg, respectively. Compared with the world’s average of 30 Bq/kg for uranium and thorium, the content of uranium and thorium of soil in site 21 was 48 times and 40 times of the world average value.

The specific activity of thorium of soil was also higher than that of soil in the background area. The average activity of thorium was 93 ± 28 Bq/kg, ranging from 34 Bq/kg to 153 Bq/kg, except for the data of hot spot site 21.

Cesium-137

Cesium-137 is an artificial radionuclide, derived from atmospheric nuclear explosions and nuclear tests, and its half-life is 30 years. As it can be deduced from Table 3, approximately 31% of the samples (9 soil samples) were contaminated by $^{137}\text{Cs}$ element. The specific activities of $^{137}\text{Cs}$ of soil samples, collected from village LPZ and site YK, were 16.8 Bq/kg and 17.0 Bq/kg, respectively, and they were much higher than those of other soil samples. The others with a higher specific activity of $^{137}\text{Cs}$ were collected from forest or grassland. The results display that erosion and migration of top soil are very weak in some places with a high specific activity of $^{137}\text{Cs}$.

Radon concentration in water

A total of 29 tap water samples and six well water samples were collected from the Xiangshan area. The radon concentrations in all samples in the Xiangshan area are listed in Table 4. As can be seen from Table 4, the radon concentrations in tap water are drastically varied, and the arithmetic mean of radon concentration is 3.29 Bq/l with the standard deviation of...
Table 4. Radon concentration in water surrounding Xiangshan uranium deposits [kBq/m³]

<table>
<thead>
<tr>
<th>Water type</th>
<th>Numbers of samples</th>
<th>Mean</th>
<th>Standard deviation</th>
<th>Minimum</th>
<th>Maximum</th>
</tr>
</thead>
<tbody>
<tr>
<td>Tap water</td>
<td>28</td>
<td>3.29</td>
<td>6.14</td>
<td>0.06</td>
<td>24.87</td>
</tr>
<tr>
<td>Well water</td>
<td>6</td>
<td>28.90</td>
<td>44.56</td>
<td>2.35</td>
<td>118.04</td>
</tr>
</tbody>
</table>

6.14 Bq/L, ranging from 0.06 Bq/L to 24.87 Bq/L. The radon concentrations in well water are generally high, with an average of 28.9 Bq/L, and the minimum and maximum values are 2.37 Bq/L and 118.04 Bq/L, respectively. In addition, we found a rising spring in Village 2, which was the drinking water source of the local villagers. The radon concentration in two spring water samples, collected in April 2018 and May 2018, was 127.1 Bq/L and 182.1 Bq/L, respectively (these values are not counted in Table 4).

The tap water for the residents in this area mainly comes from the accumulation of surface water on the mountain, while the well water originates from groundwater of several meters to ten meters underground. From the data in Table 4, it can be found that the average radon concentration in groundwater is relatively high, which has exceeded the radon concentration standard of 11.1 Bq/L of drinking water in China [22].

Annual effective dose estimate

Based on the measured dose rate per hour, the annual effective dose was computed using the indoor occupancy factor of 0.8 and outdoor occupancy factor of 0.2 [30], covering the air dose rate to an effective dose equivalent using 0.7 Sv/Gy. From the data listed in Table 1, the annual effective dose for the public in the Xiangshan area was 0.71 mSv/y and 0.15 mSv/y, respectively, for indoor and outdoor. So the mean of the total gamma effective dose (indoor+outdoor) was 0.86 mSv/y, ranging from 0.59 mSv/y to 1.36 mSv/y.

Exposure due to radon comes mainly from the inhalation of the decay products of radon. If the parameters of the equilibrium factor of radon and its progenies of 0.4, occupancy time indoor of 7000 h and the effective dose convert coefficient of 9 nSv per Bq·h/m² were taken for the annual effective dose calculation, the mean effective dose caused by indoor radon in the survey area would be 1.13 mSv/y.

The annual effective dose caused by radon in drinking water can be calculated by the following formula:

\[
D_{eff} = k \cdot G \cdot C \cdot t
\]

(2)

where \(D_{eff}\) is the effective dose from ingestion [Sv], \(k\) is the ingesting dose conversion factor of Rn [Sv/Bq], \(G\) is the water consumption [/d], \(C\) is the concentration of Rn [Bq/L] and \(t\) is the duration of consumption, where \(t\) equates to 365 days.

When the average value of the dose conversion factor for adults was accepted as \(3.5 \times 10^{-8}\) Sv/Bq [30] and the average drinking water consumption of 1 l/d per person was assumed in annual effective dose calculation, the mean of the annual effective dose contributed by radon in water was 4.2 μSv/y and 36.9 μSv/y, respectively, for tap water and well water (Table 4).

Conclusion

The surveys of gamma dose rate, radon concentration and soil radioactivity were conducted in 15 villages, including a town, in Xiangshan uranium mine area in Jiangxi province. Based on the measured data and their results, following conclusions were concluded.

The radioactive levels around Xiangshan uranium mine areas are basically at the higher background level, but there are still some hot spots there. The uranium content at site YK near the uranium mining office has reached to 1435 Bq/kg; the gamma dose rate is as high as 2423 nGy/h on the waste rock pile (site FS). Therefore, remediation measurement and work in this region are needed.

High radon concentrations were observed in some dwellings. About 50% of the houses in Village 14 had radon concentrations higher than 160 Bq/m³, with the highest one being 245 Bq/m³. In Village 4, approximately 25% of the dwellings surveyed exceeded the level of 100 Bq/m³. We can try to predict high radon potential based on local outdoor dose rate data because there is a significant positive correlation between the average outdoor dose rate and average indoor radon concentration (\(R^2 = 0.7876\)).

The arithmetic mean of radon concentration was 3.29 ± 6.14 Bq/L and 28.9 ± 44.56 Bq/L, respectively, for tap water and well water. The average radon concentration in well water in this area has exceeded the radon concentration standard of 11.1 Bq/L of drinking water in China.

The mean of the total gamma effective dose (indoor+outdoor) in this survey area was 0.86 mSv/y, and the mean effective dose was 1.13 mSv/y and 4.2 μSv/y due to indoor radon and water radon, respectively. It was noticed that the average effective dose in well water was as high as 36.9 μSv/y.

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