Journal of Radioanalytical and Nuclear Chemistry (2021) 327:789–799
https://doi.org/10.1007/s10967-020-07562-2

Spatial distribution and characteristic of radiological hazard of the paddy field around a decommissioned uranium mine in eastern China

Yan-Jun Huang[1] · Li-Tao Yang[1] · Feng Zhao[1] · Gui-Yin Guo[1] · Lian-Sheng Wu[1]

Received: 28 August 2020 / Accepted: 12 December 2020 / Published online: 25 January 2021

Abstract
In this study, the natural radioactivity in pit-water and paddy soil around a decommissioned uranium mine in eastern China was investigated. The gamma radiation absorbed dose rate, annual effective dose equivalent, radium equivalent activity, and radiation hazard index were calculated, and their spatial distribution was presented. The profile distribution of the activity concentrations of radionuclides was also investigated for the possible deposition effect. The radioactivity accumulation in the paddy-soil due to the irrigation was further discussed. It was shown that the accumulation of radioactivity in the paddy soil was limited from the irrigation.

Keywords Uranium mine · Natural radioactivity · Spatial distribution · Radiological assessment · Paddy soil

Introduction

The rapid development of the atomic energy industry has continuously raised the demand for natural uranium since the mid-1950s [1–3]. The industries of uranium mining and smelting have accordingly developed. The mining of uranium mines would give rise to the increasing of a large number of tailings, waste dumps, and relic, which would change the local environment, and cause profound environmental pollution [4]. Of particular concern is the possible increase of radiological hazard on the local environment and public [3].

China's uranium deposits are mainly distributed in southern and eastern regions, such as Guangxi, Guangdong, and Jiangxi Province. It was reported that 85% of proven uranium deposits are distributed in these regions [5], which are as well as densely populated, and are the most important rice production bases [6, 7]. In the process of uranium mining, radionuclides in the mining area would be transferred in the rice through various pathways (especially irrigation), and increase hazard to the local public [5, 7–10].

In the last 20 years, the early-developed uranium mines have gradually decommissioned [11]. The radiological impact was assessed effectively to meet the requirement of environmental restoration. One of the most important concerns during the assessment is the mine water [1–4, 11]. Years of mining would result in a large number of barren rock and disturbed residual mine ores or soils around uranium mine. Due to the heavy rainfall and high groundwater level, reservoirs would form locally for some open-pit uranium mine, and the pit-water was often used for irrigation of surrounding paddy fields. The use of the pit-water for agricultural purposes (irrigation) would result in the transfer of the natural radionuclides through the terrestrial food chain, and the subsequent internal exposure for the local public individuals due to ingestion of contaminated foodstuffs. The possible environmental impact and radioactive contamination on paddy rice have always attracted local public concern [9]. Therefore, the site release in reality for the uranium mine is far from over, local public complaints and government affairs about the assessment of the radiological hazard still happened occasionally in China [12].

In this paper, we take a decommissioned uranium mine in eastern China as an example to assess the radiological hazard from the paddy field, where the around paddy field was irrigated with the pooled mine pit-water. The spatial

✉ Yan-Jun Huang
hyj1231@163.com

[1] Suzhou Nuclear Power Research Institute Co. Ltd., CGN, Xihuan Road 1788, Suzhou 215004, People's Republic of China

distribution of radionuclides concentration and the health risk indexes in the local environment was presented. The study will provide a reference for effective evaluating the radiological impact and the extent of contamination, and help to solve the public's concerns about the impact of pit-water irrigation around the decommission uranium mine through the scientific assessment method.

Materials and methods

The research area

The uranium mine is located in a province in eastern China (Fig. 1). It had been mined for more than 50 years and was decommissioned in 2010. The area to which the uranium mine belongs is a subtropical greenhouse climate zone. The average annual temperature is 18.4 °C, the average relative humidity is 76%, the average annual rainfall is 1750 mm, and the frost-free period is 267 days.

It was an open-pit mine. During the mining, the ore was just transported to another site for centralized smelting. A pit with an area of about 3 hectares was formed at the mine site with a maximum depth of 80 m through years of catchment after abandoned years ago. The nearest settlement is located about 200 meters north of the pit boundary, and has a population of about 400 people. Since the mining area is mainly surrounded by paddy fields, the residents use the pit-water for irrigation, mainly through gravity irrigation or pumping. The irrigation area is about 6 hectares in the north of the pit. In the rainy season, the water would overflow through a water gate located in the north of the mine pit.

Sampling

Surface water from the mine pit and the contrast site was sampled with a sampling pump for the analysis of total uranium and natural gamma radionuclides. ^{238}U and ^{226}Ra were mainly concerned radionuclides in this study. Before the sampling, the polyethylene containers with volumes of 25 L were washed with the sampling water twice. After filled with the water, 10 ml of concentrated nitric acid was added into each container to acidify the water to avoid the possible adsorption of the radionuclides on the interior.

According to the uniform distribution of paddy fields and shape of each farming block, 30 points were selected for surface soil sampling and analyzing (within 20 cm depths), including 5 contrast sites (C01–C05). The contrast sites are located across a creek to the research area that are unaffected from the mine pit-water. 5 points of the total sampling sites were selected for profile analysis (focus on the sites most likely to be affected, as well as one control site), i.e., S13, S16, S21, S24, and C05. The soils for profile analysis were sampled every 20 cm, and the total sampling depth was

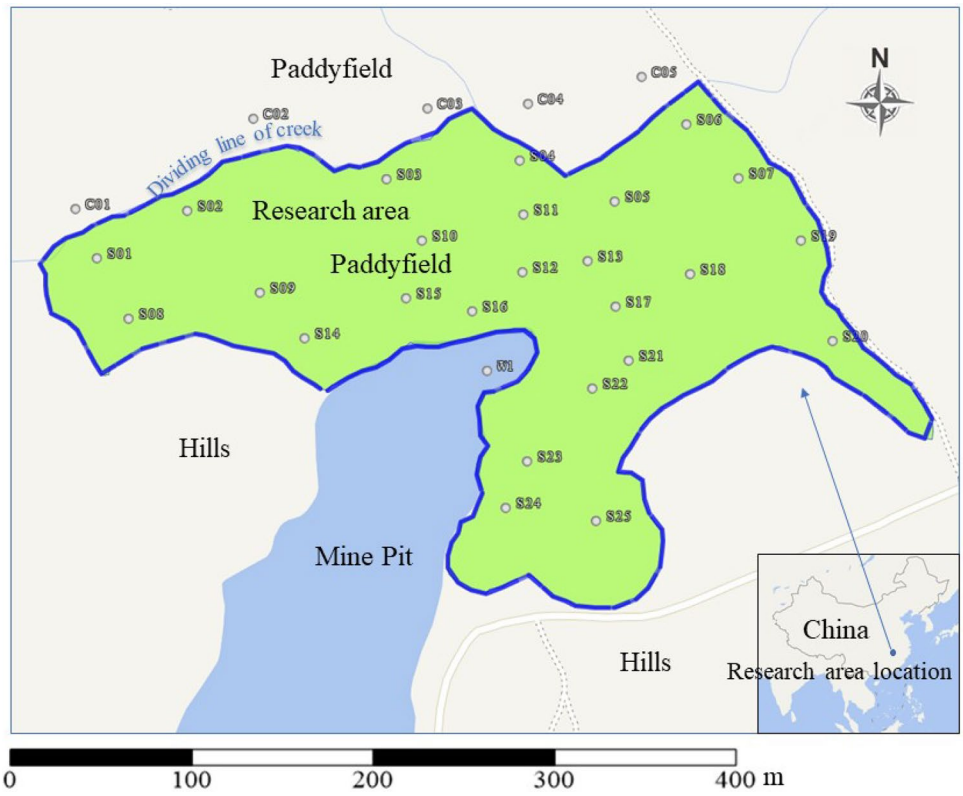

Fig. 1 Locations of the research area and the sampling sites. S1–S25, the sampling sites at the research area; C01–C05, the contrast sites; W1, the sampling site at the mine pit-water (*note*: the contrast site of a small-scale reservoir is situated on the 0.5 km west of the research area (W2), and is not marked on the figure)

about 80 cm. The soils were sampled in the harvest season, and the field was soft without free water. The sampled soil was analyzed for natural gamma nuclides, including ^{238}U, ^{232}Th, ^{226}Ra, ^{40}K.

Sample processing and radioactivity determination

Mine water

1. Laser fluorimetry

Total uranium was analyzed on a laser fluorescence analyzer (WGJ-III Trace uranium analyzer, Hangzhou Daji Electric Instrument Co. Ltd). The analysis principle is based on the fluorescence of the uranium complex with high luminescent yield formed with an enhancer reagent and the uranyl species $(UO_2)^{2+}$ in water [13]. The detection limit for uranium is about 0.05 ng ml^{-1}.

The analysis for each sample includes the following procedures. Take a 5.00 mL water sample in a quartz cuvette, determine the fluorescence intensity as N_0, add 0.5 mL of fluorescent enhancing reagent, mix well, and determine the fluorescence intensity as N_1. Then add 5μL of uranium standard solution to the sample, mix well, and record the fluorescence intensity as N_2. Then total uranium (C_U) can be calculated by the following equation:

$$C_U = \frac{(N_1 - N_0)C_0V_0}{(N_2 - N_1)V} \quad (1)$$

where C_U is the total uranium concentration in the sample (μg L^{-1}); N_0, N_1, and N_2 are the fluorescence intensity recorded according to the procedures mentioned above; C_0 is the uranium concentration in the standard (μg L^{-1}); V_0 and V are the volume of the uranium standard (0.05 mL) and sample (5 ml), respectively.

2. Gamma spectrometry

For the water sample subjected to a high-resolution gamma spectrometer, took a 40 L water sample, evaporated, and concentrated to 1 L with an electric heater. Then it was filled into a 1 L PVC Marinelli beaker, sealed for up to 20 days to ensure the radioactive secular equilibrium among the daughter nuclides of ^{226}Ra, ^{220}Rn, and their short-lived decay products, and then counted on a high-resolution gamma spectrometer (GMX5084-P4 & DSPEC-jr2.0-POSGE, Ortec inc.). It used a GMX (N-type) coaxial radiation detector with a carbon fiber window, and the relative efficiency is about 50%. The ultra-low-background lead shield system was used that features an overall thickness of 4 inches of lead materials and an inner liner of copper and thin layers. A liquid nitrogen dewar was used for cooling the detector.

The spectrometer was calibrated with radionuclides of ^{238}U, ^{232}Th, ^{226}Ra, ^{40}K, ^{60}Co, and ^{137}Cs in freshwater standards in a PVC Marinelli beaker with the same dimensions, which was obtained from the National Institute of Metrology, China (NIM), and were guaranteed within the validity period during the monitoring stage in this study

The activity concentration of ^{238}U was determined from its daughter nuclide of ^{234}Th(63.3 keV), and the activity concentration of ^{226}Ra was determined from its daughter nuclide of ^{214}Pb (351.9 keV). To determine the activity concentration of ^{232}Th, the gamma-ray from its daughter nuclide of ^{212}Pb (238.6 keV) was used. To determine the activity concentrations of ^{40}K, the principal gamma-ray of 1460 keV was used. The total counting time for each sample was 80,000 s, which could make one sample could be measured every 24 h. The activity concentrations of ^{238}U, ^{232}Th, ^{226}Ra, and ^{40}K were calculated using the following equation:

$$C_w = N/\varepsilon\eta V \quad (2)$$

where C_w is the activity concentration of ^{238}U, ^{232}Th, ^{226}Ra, and ^{40}K in water (Bq L^{-1}); N is the net gamma counting rate (cps); ε is the detection efficiency at the corresponding energy; η is the absolute transition probability of gamma-ray; V is the volume of the sample (40 L).

The a priori minimum detection concentrations (MDC) for ^{238}U, ^{232}Th, ^{226}Ra, and ^{40}K in water were about 0.18 Bq L^{-1}, 0.006 Bq L^{-1}, 0.004 Bq L^{-1}, and 0.04 Bq L^{-1}.

Soil

The surface paddy soil was sampled within 20 cm depth with a shovel for all the sites except for the sites of profile analyzing. As the paddy field was cultivated in blocks, a mixture with separate samples of a random number in each block was collected. Firstly, debris such as grassroots was removed for each sample, and then it was mixed thoroughly. For profile sampling, a dynamic sampler was used (core diameter 4 cm), and take profile samples every 20 cm depth down to 80 cm. Then, the samples were dried in an oven for 48 h (110 °C), weighed, and crushed into fine powders. After sieving for 60 mesh-size, each sample was sealed into an airtight PVC container and kept for up to 20 days to allow radioactive secular equilibrium among the decay products of ^{226}Ra and then measured on a high-resolution gamma spectrometer. Simulated soil sample standard with known activity concentrations was used for sample calibration, which was obtained from the National Institute of Metrology, China (NIM), and the spectrometer and gamma-ray energy of each nuclide were the same as that of the water samples. An average 300 g of weight for each sample was used, and the counting time was about 80000 s, which could make one sample could be

measured every 24 h. The activity concentrations of ^{238}U, ^{232}Th, ^{226}Ra, and ^{40}K were calculated using the following equation:

$$C_s = N/\varepsilon\eta m \tag{3}$$

where C_s is the specific activity of ^{238}U, ^{232}Th, ^{226}Ra, and ^{40}K in soil (Bq kg^{-1}); m is the mass of the sample (kg); N, ε, and η have the same meaning as in the previous equation.

The a priori minimum detection concentrations (*MDC*) for ^{238}U, ^{232}Th, ^{226}Ra, and ^{40}K in soil were about 21 Bq kg^{-1}, 1.3 Bq kg $^{-1}$, 1.9 Bq kg $^{-1}$, and 13 Bq kg $^{-1}$.

Radiation risk index

1. Gamma absorbed dose rate (D_r)

The environmental surface gamma absorbed dose rate (D_r) reflects the environmental background gamma radiation level and the changes caused by human practice, and is also used to assess the radiation dose received by the public individuals [10]. In this study, through the specific activity of natural radionuclides in the shallow surface soil, the Beck's formula was used to estimate [14–16]:

$$D_r = 0.462C_{Ra} + 0.604C_{Th} + 0.0417C_K \tag{4}$$

where C_{Ra}, C_{Th} and C_K are the specific activities (Bq kg^{-1}) of ^{226}Ra, ^{232}Th, and ^{40}K, respectively; the coefficients are the conversion factor corresponding to the nuclides.

Although the Beck's formula was established based on the assumption of uniformly distributed of the natural radionuclides within 5 cm depth and the secular equilibrant of U-series and Th-series(Beck et al., 1964), it was used in this study as an approximate method.

2. Annual effective dose equivalent

The annual average effective dose equivalent (A_{ex}) received by an adult individual was calculated using the following equation [14]:

$$A_{ex} = D_r \times t \times 0.7 \times 10^{-6} \tag{5}$$

where 0.7 is the conversion factor from absorbed dose in the air to the effective dose received by an adult individual (Sv Gy^{-1}); t is the working time in the paddy field. According to the characteristics of local farming, it is assumed that the annual arable time is 50 h, planting time is 50 h, weeding and fertilization time is 50 h, and harvest time is 50 h. So, the total working time in the paddy field was estimated as 200 h a^{-1} conservatively.

3. Radium equivalent concentration

The radium equivalent concentration (Ra_{eq}) is widely used to evaluate the radiation hazard of the material [9, 14, 16, 17]. It is assumed that 370 Bq kg^{-1} of ^{226}Ra, 259 Bq kg^{-1} of ^{232}Th, and 4810 Bq kg^{-1} of ^{40}K produce the same gamma dose rate, which gives the annual dose of 1.5 Gy (1 mSv) at a height of 1 m above the ground. It can be calculated by the following equation [14, 16]:

$$Ra_{eq} = C_{Ra} + 1.43C_{Th} + 0.077C_K \tag{6}$$

where C_{Ra}, C_{Th}, and C_K have the same meaning mentioned in Eq. (4).

4. Exposure index

Beretka and Matthew defined two indexes (internal/external exposure index) to limit the radiation dose to 1 mSv a^{-1} [18]. The external exposure index (H_{ex}) is calculated using equation [14, 18]:

$$H_{ex} = C_{Ra}/370 + C_{Th}/259 + C_K/4810 \tag{7}$$

If the calculated value is less than the unity, the radiation hazards caused by radioactive ^{226}Ra, ^{232}Th, and ^{40}K in this area to the human body can be ignored [16, 18].

The internal exposure index (H_{in}) is calculated using equation [14, 18]:

$$H_{in} = C_{Ra}/185 + C_{Th}/259 + C_K/4810 \tag{8}$$

If the value of H_{in} is less than unity, the hazard from radon and its short-lived progeny to the respiratory organs is negligible [16, 18].

Results and discussions

Results

Radioactivity in mine water

The analysis result of the radioactivity in water is shown in Table 1. It is indicated that total uranium in mine pit-water is 10.4 μg L^{-1}, which is higher than the result from the contrast site, i.e., 0.090 μg L^{-1}. It is shown that there is obviously enhancing of uranium content in the mine pit-water, with almost 116 times the result from the contrast site. The result is within the range of uranium concentrations in the freshwater of China (0.02–42.35 μg L^{-1}), but higher than the average (1.66 μg L^{-1}), which was investigated for natural radiation in China during the 1980s [19]. Estimated by the natural abundance of ^{238}U, 1 μg U is equivalent to 12.24 mBq ^{238}U [20], and the total uranium measurement results in the mine

Table 1 The radioactivity concentrations in mine pit-water and contrast reservoir

Element or radionuclides	Mine reservoir water	Contrast reservoir water
Total uranium (μg L^{-1})	10.4 ± 0.8	0.090 ± 0.07
^{238}U(mBq L^{-1})	140 ± 41	<89
^{232}Th(mBq L^{-1})	<2.3	<2.4
^{226}Ra(mBq L^{-1})	6.7 ± 2.2	<4.7
^{40}K(mBq L^{-1})	41 ± 12	113 ± 19

The uncertainties for total uranium were estimated by propagating the uncertainties of each parameter in Eq. (1), i.e., the relative uncertainties from V and V_0 are about 1.5%(k=2) respectively, while from C_0 is about 4.2% (from the certificate of the standard solution, k=2), and from $(N_1\text{-}N_0)/(N_2\text{-}N_1)$ are about 6%(evaluated with 9 times of reading on the laser fluorimetry for each sample, k=2). The total relative uncertainties for uranium analysis in this study is about 7.6%(k=2). The uncertainties for gamma radionuclides were just estimated based on common statistics of counts on the characteristic gamma-ray as 2σ, where σ is the standard deviation

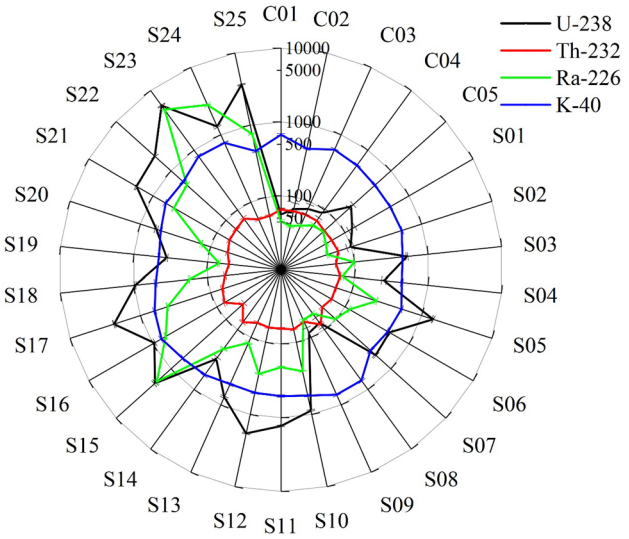

Fig. 2 The radar plots of activity concentrations ^{238}U, ^{232}Th, ^{226}Ra, and ^{40}K in the sample of surface paddy soil (Bq kg^{-1})

pit-water could be converted to the activity concentration of ^{238}U as 127.3 mBq L^{-1}.

For the gamma spectrometric analysis, it was shown that the activity concentration of ^{238}U in mine pit-water is about 140 ± 41 mBq L^{-1}, which is in accordance with the transfer values from total uranium (127.3 mBq L^{-1}). The results of ^{232}Th and ^{40}K are lower than that of ^{238}U, which indicates the principal impact from ^{238}U and its progenies should be concerned. The related results of ^{238}U and ^{226}Ra in the contrast reservoir are lower, while those of ^{232}Th in both water samples are below the *MDC*. For the activity concentration of ^{40}K, the higher value in contrast reservoir is related possibly to the lower depth and capacity, as well as its usage for aquaculture.

Radioactivity in paddy soil

1. Surface paddy soil

Statistics of radionuclides activity concentrations in surface paddy soil are shown in Table 2, the detailed result is displayed in Fig. 2. It is shown that the activity concentrations of ^{238}U in the research area vary from 77.0 to 5600.0 Bq kg^{-1} with an average of 1212.9 Bq kg^{-1} and a standard deviation of 1262.5 Bq kg^{-1}. All the values of activity concentrations of ^{238}U is higher than the national average activity concentration that investigated during the 1980s (with an average of 39.5 Bq kg^{-1} and a standard deviation of 34.4 Bq kg^{-1}) [19]. 40% of the results are higher than 1000 Bq kg^{-1} (ten sites), which indicated further intervention should be carried out according to the IAEA recommendation of 1000 Bq kg^{-1} for radionuclides excluding ^{40}K was derived using the exclusion concept for the natural region [21]. Particularly, higher values occurred in the north shore side of the mine pit and the south-east region of the research area. Sampling site S23 had the largest value of 5600.0 Bq kg^{-1}, which was affected possibly by the residual soil during the mining around the region. The deposition of the activity is also expected as a factor for the high values, which need further identification through detailed retrospective examinations. Meanwhile, it could be observed that higher values occurred near the mine pit, and have a decreasing trend in the distance.

A similar trendency of ^{226}Ra could be observed as a daughter nuclide of ^{238}U. It could be seen that the activity concentrations of ^{226}Ra in the research area vary from

Table 2 A statistics of the activity concentrations in surface paddy soil (Bq kg^{-1})

Radionuclides	Research area				Contrast area			
	AV	SD	Min	Max	AV	SD	Min	Max
^{238}U	1212.9	1262.5	77.0	5600.0	96.6	53.6	56.0	190.0
^{232}Th	62.6	7.6	49.2	81.8	64.5	1.8	61.5	66.1
^{226}Ra	577.6	1073.4	45.9	4791.0	48.8	7.8	40.1	58.7
^{40}K	562.1	107.9	427.0	786.0	564.2	77.3	475.0	673.0

AV means the average; *SD* means the standard deviation

577.6 to 1073.4 Bq kg^{-1}, with an average of 577.6 Bq kg^{-1} and a standard deviation o 1073.4 Bq kg^{-1}, which is higher than the result in the national soil that investigated during the 1980s (with an average of 49.1 Bq kg^{-1} and a standard deviation of 27.6 Bq kg^{-1}) [19]. There are only 3 soil samples with the analysis results that are higher than the derived value of 1000 Bq kg^{-1} from the exclusion concept for the natural region [21], which indicated an influence of the different geochemical characteristics of ^{238}U and ^{226}Ra in the natural environment [22].

The activity concentrations of ^{232}Th are not notable, which are in the range of 48.2–81.8 Bq kg^{-1} with an average of 62.6 Bq kg^{-1} and a standard deviation of 7.6 Bq kg^{-1}. They are lower and more homogenous than that of ^{238}U and ^{226}Ra. The corresponding activity concentrations were in the range of the national investigated values (ranged in 1–437.8 Bq kg^{-1} with an average of 49.1 Bq kg^{-1} and a standard deviation of 27.6 Bq kg^{-1}) [19].

The activity concentrations of ^{40}K are in the range of national activity concentrations and are comparable to the national average (ranged in 11.5–2185.2 Bq kg^{-1} with an average of 580 Bq kg^{-1} and a standard deviation of 202.0 Bq kg^{-1}) [19].

As shown in Table 2 and Fig. 2, the results of ^{238}U and ^{226}Ra in the research area are higher than those in the contrast site, while those of ^{232}Th and ^{40}K in the research area are closed to those in the contrast site, which indicated that the paddy field in the research area was affected from the uranium mining significantly.

To explore further the activity concentrations of ^{238}U, ^{232}Th, ^{226}Ra, and ^{40}K in the surface paddy field of the research area, we plot the contour distribution of the activity concentrations as displayed in Fig. 3. It could be found that the activity concentrations of ^{238}U and ^{226}Ra in the south-east region are higher, and have a decreasing trend with the distance to the mine pit. Especially in the north region, the corresponding results are closed to those in the contrast sites. Also, the paddy field area about 3 hectares for ^{238}U and ^{226}Ra above 1000 Bq kg^{-1} could be identified, mainly in the north and south-east regions.

To explore the correlations between the activity concentrations of ^{238}U, ^{232}Th, ^{226}Ra, and ^{40}K, the Pearson's correlations coefficients are shown in Fig. 4. It could be observed for the strong correlation (r between 0.6 and 0.8) between ^{238}U and ^{226}Ra ($r=0.76$) and that between ^{226}Ra and ^{40}K ($r=0.61$), while weak correlation (r between 0.2 and 0.4) between ^{232}Th and ^{40}K($r=0.47$) and between ^{238}U and ^{40}K ($r=0.31$). None correlation between the activity concentrations between ^{238}U and ^{232}Th, ^{226}Ra and ^{232}Th could be observed as the Pearson's coefficients are both 0.03 respectively. The correlation analysis may provide some meanings of the geochemical of the radionuclides in the local environment. The strong correlation between the activity concentrations of ^{238}U and ^{226}Ra indicates a similar source and the transfer characteristics [10].

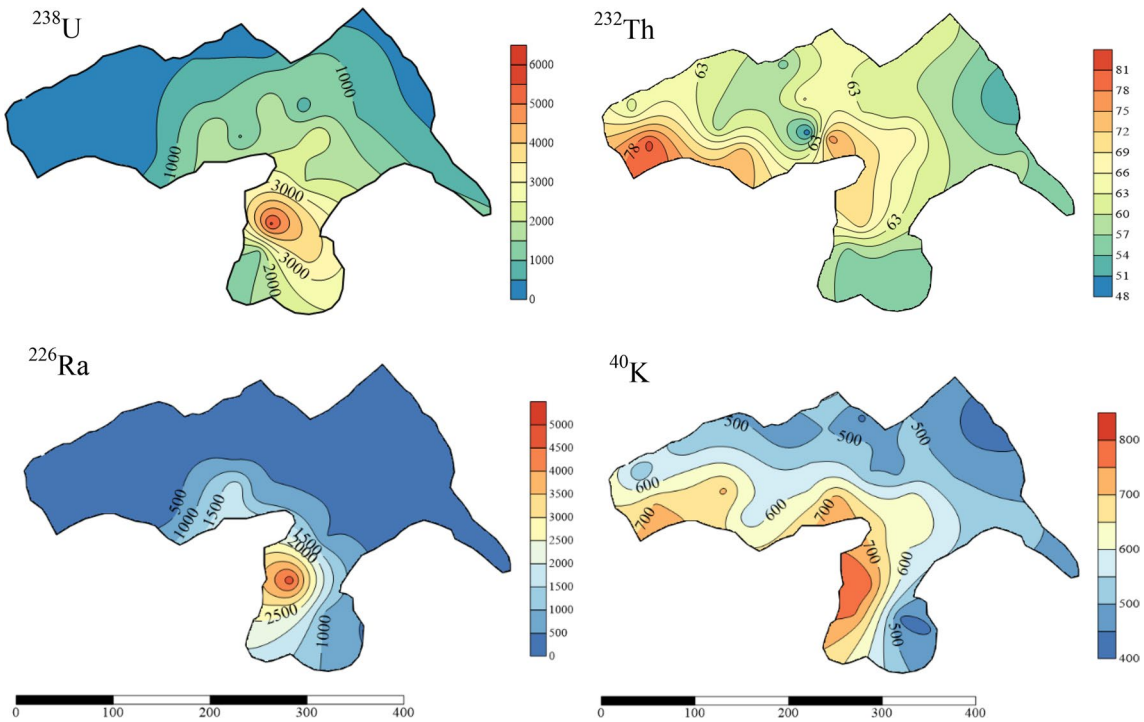

Fig. 3 The contour plots of the distribution of the activity concentrations (Bq kg^{-1})

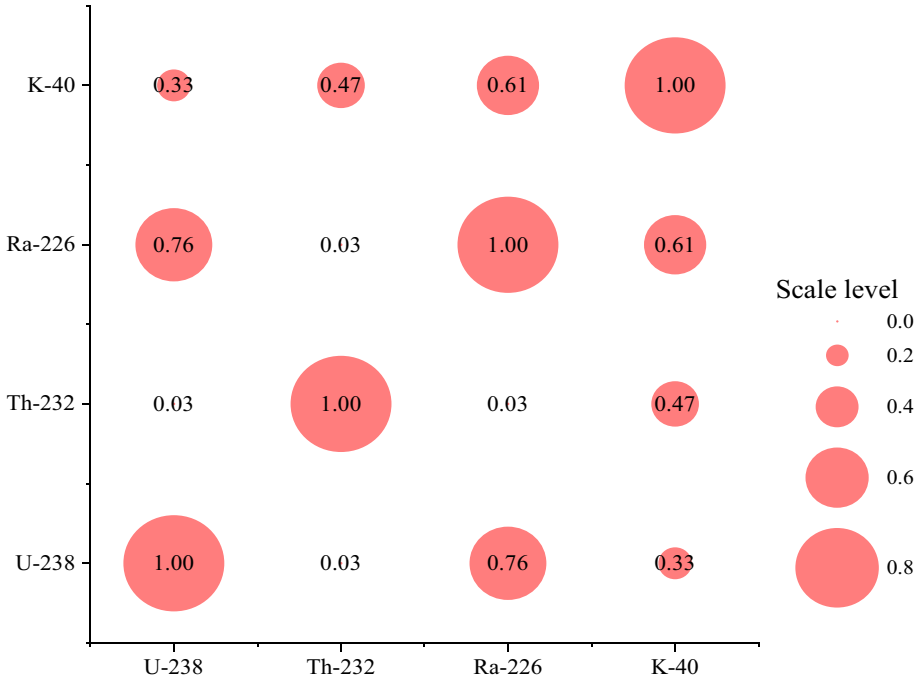

Fig. 4 The matrix illustration of the Pearson's coefficients between the activity concentrations of ^{238}U, ^{232}Th, ^{226}Ra, and ^{40}K

2. Profile distribution

The profile distribution of radioactivity is important to understand the behavior along with the history and give some information such as weathering, erosion, biological function, and cultivation influence [23, 24]. Five paddy soil sampling sites were selected for profile investigation, including a contrast site (C05). The corresponding results of the activity concentrations of ^{238}U, ^{232}Th, ^{226}Ra, and ^{40}K are displayed in Fig. 5.

^{238}U and ^{226}Ra profile from the research area showed a similar pattern, obviously higher values occurred for the surface soil, getting mainly lower values as deeper positions. At the depth of about 60–80 cm, the activity concentrations in the research area decrease to similar levels of that in the contrast site. The phenomenon indicates a deposition process of the anthropogenically introduced activity. The profile distribution of the ^{238}U for site S24 is slightly different. It was beginning with a lower value at the surface (0–20 cm), then getting a higher value at 20–40 cm, and a lower value at a deeper position. The special profile is related to the fact

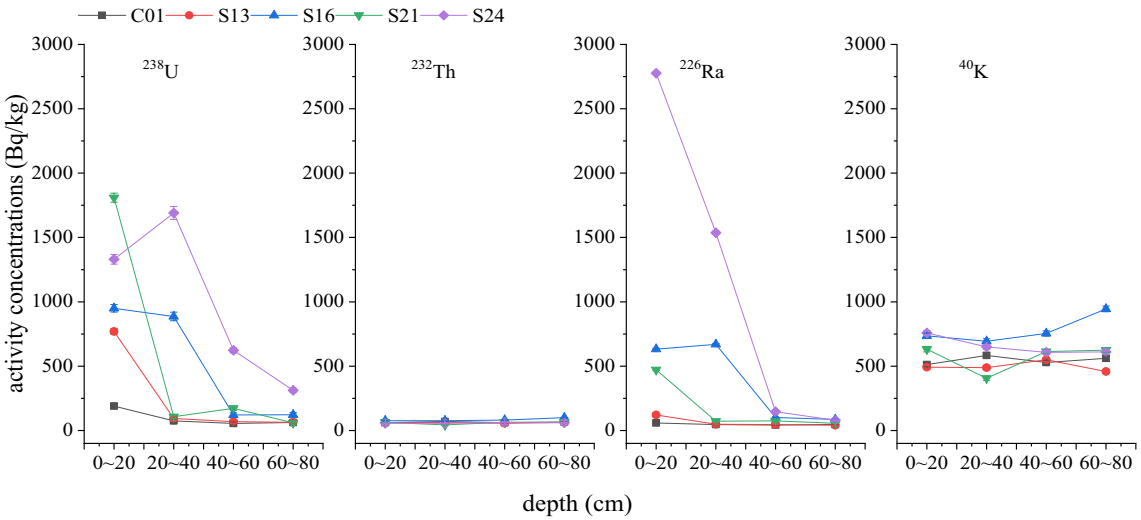

Fig. 5 The profile distribution of the activity concentrations in paddy soils

that it is remediated soil with a coating of other soil materials. The profile distributions of ^{226}Ra are relatively weak for the others sites of the research area, which was ascribed to the possible disequilibrium in the U-series and the difference of the geochemical characteristics, as the ratios of ^{238}U/^{226}Ra are mainly in the range of 1.1–6.7 for the surface paddy soil (excluding S24) with average about 3.9, and the ratio of ^{238}U/^{226}Ra are mainly in the range of 0.5–7.5 for profile soil with an average about 2.5. Since the profile distribution is highly associated with the geochemical characteristics of the radionuclides and the soil, and further investigations should be carried out to give more scientific meaning.

Note the comparison of the profile distributions of activity concentration between the research area and the contrast site (C08), it could be seen that profile variations of the ^{238}U, ^{226}Ra are inconspicuous, which is shown the possible impact from the mining process and the irrigation impact with the mine pit-water.

For the activity concentrations of ^{232}Th and ^{40}K, it presents quite homogeneous profile distributions, and could not give any meaningful conclusions. Although ^{40}K activity always revealed a high variability due to the cultivating and the fertilization, there is none uniformed profile characteristic in our study.

Radiological risk assessment

The corresponding statistics of the radiological risk indexes calculated with the activity concentrations of surface paddy soil are shown in Table 3, and the distribution in the research area are displayed in Fig. 6.

D_r is observed with values range from 82.0 nGy h^{-1} (S02) to 2289.1 nGy h^{-1} (S23) with an average of 328.1nGy h^{-1} and a standard deviation of 498.9nGy h^{-1}. The results are higher than the national average of 62.8nGy h^{-1} [19]. Note D_r in the south-east area is higher, the higher risk in the region should be concerned, which was attributed mainly to the radionuclides of U-series. Take site S23 as an example, 96.7% of the values of D_r was contributed from ^{226}Ra, while only 1.9% and 1.4% of D_r were contributed from ^{232}Th and ^{40}K respectively.

The results for the A_{ex} were calculated with values varied from 0.01 mSv a^{-1} to 0.32 mSv a^{-1} with an average of 0.05 mSv a^{-1} and a standard deviation of 0.07 mSv a^{-1}. The corresponding average is lower than the world average annual effective dose from outdoor terrestrial gamma radiation (0.460 mSv a^{-1}) [14]. It indicates that the radiation hazard is insignificant for the local residual public concluded from the assessment of A_{ex}, as only the 200 h of farming time per year were assumed in this study.

The radium equivalent activity (Ra_{eq}) provides a basis for comparing the activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K in soil to obtain the total radioactivity. The calculated values could be found varying from 177.6 Bq kg^{-1} to 4953.1 Bq kg^{-1} with an average of 710.4 Bq kg^{-1} and a standard deviation of 1079.0 Bq kg^{-1}. 60% of the values in the research area (15 sites) are higher than the maximum permissible values of 370 Bq kg^{-1}, while all the values in the contrast area are lower than the corresponding limit. As the annual effective dose fro Ra_{eq} value of 370 Bq kg^{-1} corresponds to an effective dose of 1.0 mSv for public individuals, the observed maximum value of Ra_{eq} of 4953 Bq kg^{-1} (S23) corresponds to an annual effective dose of about 13.4 mSv a^{-1}, and the soils should be restricted for building materials to avoid radiological health hazards.

The external hazard index (H_{ex})could be observed varying from 0.5 to 1.8 with an average of 0.8 and a standard deviation of 0.4. The acceptable value of 1.0 corresponds to 1.5 mSv a^{-1} for the radiological health hazards for its usage as building raw material. It could be observed that 40% of the values (10 sites) from the research area are higher than 1.0. The internal hazard index (H_{in}) could be observed varying from 0.6 to 3.2 with an average of 1.2 and a standard deviation of 0.8, and 60% of the values (15 sites) could be identified to be higher than the limit of 1.0. It could be observed that all the external exposure and internal exposure indexes in the contrast area are lower than the limit of

Table 3 A statistic of the radiological hazard indexes for the research area and the contrast area

Area	Parameters	D_r	A_{ex}	Ra_{eq}	H_{ex}	H_{in}
Research area	*AV*	328.1	0.046	710.4	1.9	3.5
	SD	498.9	0.070	1079.0	2.9	5.8
	min	82.0	0.011	177.6	0.5	0.6
	max	2289.1	0.320	4953.1	13.4	26.3
Contrast area	*AV*	85.0	0.012	184.5	0.5	0.6
	SD	4.6	0.001	9.3	0.0	0.0
	min	77.3	0.011	168.9	0.5	0.6
	max	88.8	0.012	192.8	0.5	0.7

AV means the average; *SD* means the standard deviation

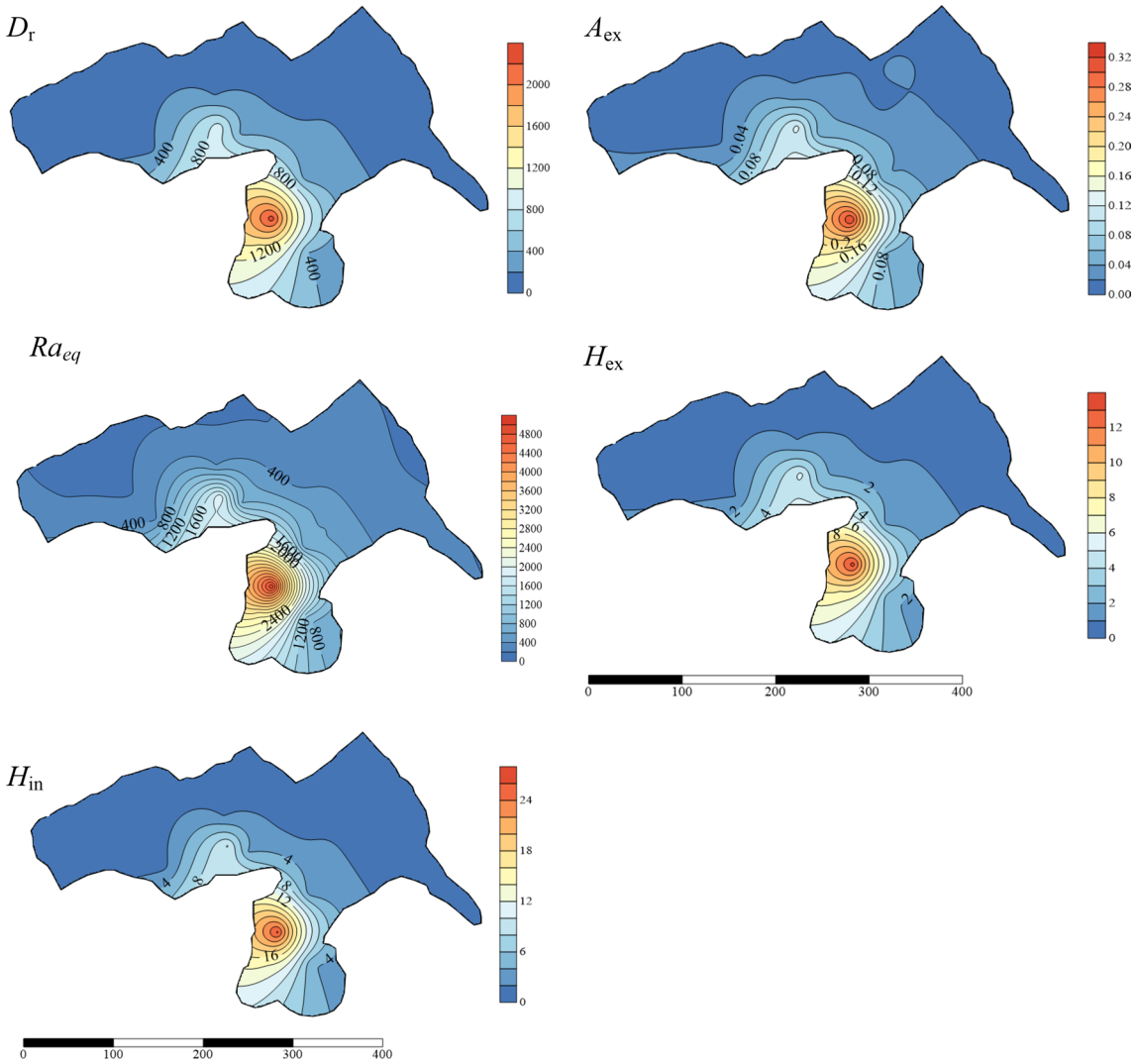

Fig. 6 The contour plots of the radiological hazard indexes

1.0, and the corresponding hazard could be concluded as acceptable.

Overall, the higher radiological hazard in the research area could be identified compared to the contrast, especially in the south-east region of the research area. The area for Ra_{eq} values above 370 Bq kg^{-1}, for H_{ex} and H_{in} values above unity, is all about 3 hectares.

The estimation of the impact of irrigation

According to the previous description, the results of ^{238}U and total uranium in the mine pit-water are in good agreement. A conservative model with the activity concentrations of ^{238}U was used to estimate the cumulative impact of irrigation water on the paddy fields assuming that all the radionuclides in the irrigation water were deposited in the surface soil. Estimating the deposition flux of the radionuclide ^{238}U is based on irrigation water consumption:

$$M = C_U \cdot V/666.7 \quad (9)$$

where M is the deposition flux from the irrigation water (Bq $m^{-2}{\cdot}a^{-1}$); C_U is the activity concentration of ^{238}U in the pit-water (Bq L^{-1}); V is the irrigation water consumption (m^3); 666.7 is the conversion factor between Chinese area unit of mu and square meters. The activity concentration of ^{238}U in the mine pit-water (C_U) is 140 mBq L^{-1} (i.e. 140 Bq m^{-3}). The water consumption of 500 m^3 mu^{-1} in the research area was assumed [25].

According to estimates, the irrigation deposition flux through the pit-water is about 105 Bq $m^{-2}{\cdot}a^{-1}$. Considering that the typical tillage depth of the paddy soil is within

20 cm, the ^{238}U radioactivity deposited in the soil due to the irrigation can be estimated by the following equation:

$$C_s = M/\rho d \quad (10)$$

where C_s is the annual radioactivity level deposited in the soil (Bq kg^{-1}·a^{-1}); ρ is the density of paddy soil (kg m^{-3}) with a typical value of about 2500 kg m^{-3}; d is the tillage depth, assuming about 0.02 m.

It can be estimated that if the mine pit-water was used for irrigation every year, the annual deposited ^{238}U amount in the paddy soil is about 2 Bq kg^{-1}. This level is very small compared to the level of ^{238}U activity concentration monitored in paddy soil. So, the higher values of ^{238}U and ^{226}Ra in the surface paddy soil of the research area maybe are correlated with other significant issues such as the disturbance of the land surface during mining, the temporary storage of ores on-site, dewatering of mine workings, and the other reclamation activities that all have potential to significantly affect the concentrations and loads of dissolved and suspended materials in surface water off-site [4]. So, further investigation for the source of the radioactivity in the research area should be carried out based on retrospective examinations.

Conclusion

In this paper, the natural radionuclides concentrations and the radiological hazards, as well as the distribution characteristics around a decommissioned uranium mine in eastern China were investigated. It was shown that the total uranium concentration in the mine pit-water is about 116 times higher than that in the contrast reservoir, and the activity concentrations of ^{238}U and ^{226}Ra are higher levels in the south-east region of the research area and north shore side of the mine pit. The maximum activity concentrations of ^{238}U and ^{226}Ra were observed as 5600 Bq kg^{-1} and 4791 Bq kg^{-1}, respectively. It was estimated that a paddy field area about 3 hectares for ^{238}U and ^{226}Ra above 1000 Bq kg^{-1} or an area with high radiological hazard in the research area could be identified.

The profile distributions analysis of the selected sites showed that the activity concentration of ^{238}U and ^{226}Ra presents the characteristics of higher at surface layer and lower at the bottom layer, which may be related to the geochemical conditions in the region and the deposition of radioactive materials during uranium mining. Based on conservative estimates, the cumulative radioactive effect of mine water irrigation on the paddy field is considered relatively limited.

The study would provide a scientific reference and decision basis for the concerns about the radiological impact on the local public individuals and local government. Further research on the source of the elevated concentration of ^{238}U and ^{226}Ra in the paddy soil should be carried in the future.

Compliance with ethical standards

Conflict of interest The authors declared that they have no conflicts of interest to this work.

References

1. IAEA (2009) IAEA-TECDOC-1622 Status and trends of nuclear technologies- Report of the international project on innovative nuclear reactors and fuel cycles (INPRO). International Atomic Energy Agency Vienna, Austria
2. IAEA (1997) IAEA-TECDOC-982 Planning for environmental restoration of uranium mining and milling sites in central and eastern Europe. International Atomic Energy Agency Vienna, Austria
3. IAEA (2005) Environmental contamination from uranium production facilities and their remediaiton. In: Proceedings of an International Workshop on Envrironmental Contamination from Uranium Production Facilities and Their Remediation, Lisbon, Portugal, 2005. International Atomic Energy Agency
4. National Academy of Sciences (2012) Uranium mining in Virginia: scientific, technical, environmental, human health and safety, and regulatory aspects of uranium mining and processing in Virginia. The national Academies Press, Washington
5. Zhang ZS, Li MY, Yang YX, Liu YJ, Li XC (2007) Radiation contamination and treatment of some hard-rock-type uranium mines in Gan, Yue and Xiang areas (in Chinese). Uran Min Metall 26(4):191–196
6. Zhang JD, Li YL, Jian XF (2008) Situation and development prospect of uranium resources exploration in China (in Chinese). Strateg Study CAE 10(1):54–60
7. Ouyang J, Liu Z, Ye T, Zhang L (2019) Uranium pollution status and speciation analysis in the farmland-rice system around a uranium tailings mine in southeastern China. J Radioanal Nucl Chem. https://doi.org/10.1007/s10967-019-06783-4
8. Wu YY, Qin ZY, Zhao XC, Tang MJ, Lu XF, Lei JJ, Ma YL, Xie P (2019) Radionuclide contents in food collected from regions surrounding retired uranium mine in Northwest of Guangxi and their committed dose to public. Occup Health 35(23):3214–3216
9. Wang Z, Qin H, Liu X (2019) Health risk assessment of heavy metals in the soil-water-rice system around the Xiazhuang uranium mine, China. Environ Sci Pollut Res 26(6):5904–5912
10. Liu YY, Zhang CY, Wei QL, Guo YZ, Gao B, Zhou WB (2018) Spatial distribution and radiation evaluation of the radionuclides in paddy soil of the uranium tailings area (in Chinese). Asian J Ecotoxicol 13(5):305–312
11. Pan YJ, Li YC, Xue JX (2009) Status and countermeasures for decommissioning of uranium mine and mill facilities in China (In Chinese). Radiat Prot 29(3):167–171
12. Luo PP, Hu CJ (2012) Analysis and countermeasures of emergent environment complaints in uranium mountain (in Chinese). Environ Sci Manag 37(S1):5–6

13. Ministry of Environmental Protection (2017) HJ 840-2017 Technical guidelines for environmental impact assessment Analytical methods for micro-quantity of uranium in environmental samples. China Environmental Science Press, Beijing
14. UNSCEAR (2000) UNSCEAR 2000 Report to the General Assembly, with scientific annexes Volume I: Sournces. United Nations Scientific Committee on the Effects of Atomic Radiation. United Nations Scientific Committee on the Effects of Atomic Radiation, New York, USA
15. Beck HL, Condo WJ, Lowder WM (1964) Spectrometric techniques for measuring environmental gamma radiation. HASL-150, Department of Energy, Washington
16. Huang Y-J, Chen C-F, Huang Y-C, Yue Q-J, Zhong C-M, Tan C-J (2015) Natural radioactivity and radiological hazards assessment of bone-coal from a vanadium mine in central China. Radiat Phys Chem 107:82–88. https://doi.org/10.1016/j.radphyschem.2014.10.001
17. Tufail M (2012) Radium equivalent activity in the light of UNSCEAR report. Environ Monit Assess 184(9):5663–5667. https://doi.org/10.1007/s10661-011-2370-6
18. Beretka J, Matthew PJ (1985) Natural radioactivity of Australian building materials, industrial wastes and by-products. Health Phys 48(1):87–95. https://doi.org/10.1097/00004032-198501000-00007
19. State Bureau of Environmental Protection (1995) Natural environmental radioactivity levels in China (1983–1990). China Atomic Energy Press, Beijing, China
20. IAEA (1989) Technical Report No. 309 Construction and Use of Calibration Facilities for Radiometric Field Equipment. International Atomic Energy Agency, Vienna, Austria
21. IAEA (2004) Safety Guide No. Rs-G-1.7 Application of the concepts of exclusion, exemption and clearance. International Atomic Energy Agency Vienna, Austria
22. Tsikritzis LI, Fotakis M, Tzimkas N, Kolovos N, Tsikritzi R (2008) Distribution and correlation of the natural radionuclides in a coal mine of the West Macedonia Lignite Center (Greece). J Environ Radioact 99(2):230–237. https://doi.org/10.1016/j.jenvrad.2007.07.014
23. Matsuda N, Mikami S, Shimoura S, Takahashi J, Nakano M, Shimada K, Uno K, Hagiwara S, Saito K (2015) Depth profiles of radioactive cesium in soil using a scraper plate over a wide area surrounding the Fukushima Dai-ichi Nuclear Power Plant, Japan. J Environ Radioact 139:427–434. https://doi.org/10.1016/j.jenvrad.2014.10.001
24. Karadeniz Ö, Yaprak G (2008) Vertical distribution and gamma dose rates of 40 K, 232Th, 238U and 137Cs in the selected forest soils in Izmir, Turkey. Radiat Prot Dosimetry 131:346–355. https://doi.org/10.1093/rpd/ncn185
25. Jiangxi Provincial Department of Water Resources (2017) DB 36/T 619-2017 Agricultural water quotas in Jiangxi Province. Jiangxi Provincial Department of Water Resources, Nanchang