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Estimation of Annual Effective Dose Due to Ingestion of ^{210}Pb and ^{210}Po in Crops from A Site of Coal Mining and Processing in Southwest China

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Abstract: Since the exploitation of mineral resources results in the release of radionuclides, and consuming radionuclides affects public health in the short and long term. A case study of the environmental radiation impact from coal mining and germanium processing was carried out in southwest China. The coal mines contain germanium and uranium and have been exploited for more than 40 years. The farmlands around the site of coal mining and germanium processing have been contaminated by the solid waste and mine water in some extend since then. Samples of crops have been collected from contaminated farmlands in research area. The research area covers a radius of 5 km, in which there are 2 coal mines located. ^{210}Pb and ^{210}Po have been analyzed as the key radionuclides during monitoring program. The average activity concentrations of ^{210}Pb and ^{210}Po in the crops were 1.38 and 1.32 Bq/kg in cereals, 4.07 and 2.19 Bq/kg in leafy vegetables and 1.63 and 1.32 Bq/kg in root vegetables. The annual effective doses due to the ingestion of ^{210}Pb and ^{210}Po in consumed crops have been estimated for adult residents living in research area. The average annual effective dose was 0.336mSv/a, while the minimum was 0.171 mSv/a and the maximum was 0.948 mSv/a. The results show that crops grown on contaminated farmland contained an enhanced level of radioactivity concentration. Ingestion doses of local residents in research area were significantly higher than the China average level of 0.112 mSv/a, and the world average level of 0.042 mSv/a through ^{210}Pb and ^{210}Po in crops intake respectively.

Keywords: Natural radioactivity; risk assessment; ^{210}Pb and ^{210}Po; radiological impact; polluted mine site

1. Introduction

Naturally occurring radioactive material (NORM) comprises radionuclides associated with the ^{238}U and ^{232}Th decay chains as well as ^{40}K [1]. These radionuclides are present in many natural resources. During mining and processing, radionuclides in fossil fuels transfer into the wastes, which are usually disposed of in spoil heaps and tailings ponds on the mining site [2]. For some mining enterprises, proper management and adequate control are not taken to minimize the impact on environment [3,21]. The radionuclides release into the air, soil and surface water from the mine wastes as a result, and then public health is affected [4].

China is the biggest producer of coal, but only a few mines are associated or paragenetic with uranium [5-9]. Such coal mines also reserve various resources such as aluminum, iron, silicon, germanium, and gallium richly [10-13]. Some coal mines associated with uranium or other metals cause radioactive impact on surrounding areas by several pathways, such as the emission of dust (gas) during mining and smelting process, the accumulation of coal slag, the discharge of liquid effluents and residues of germanium purification [14-20].

After the radioactive wastes of mineral exploitation are released into environment, the radionuclides transfer and redistribute in different types of receptor environments [21]. In cropland, one of the receptors, exposure pathways associated with cultivation of agricultural plants can be assessed. Radionuclide accumulation in plants is determined based on radionuclide concentrations in top soil layer, and it accounts for root transfer from contaminated soil and interception by plant leaves [22, 30]. Residents living around the mines consume foodstuffs cultivated in the contaminated croplands, as a result, the potential exposure risks of ingesting radionuclides of local residents increase evidently. Therefore, our study aims to (1) investigate the radioactivity of level of crops cultivated in local croplands, (2) estimate the ingestion doses of adult residents living in the research area, and (3) assess the environmental radiological impact caused by coal mines located in southwest China. An area with a radius of 5 km was defined as the research area with 2 coal mines located in. Cereals, root vegetables and leafy vegetables planted in research area were sampled. They are the most common crop types in local dietary. ^{210}Pb and ^{210}Po in crops were selected as key radionuclides that contribute the most of ingestion doses among natural radionuclides in ^{238}U and ^{232}Th decay chains [1].

2. Materials and Methods

2.1. Site description

Research area is located in the southwestern China, as shown in Figure 1. In research area, coal mines contain germanium and uranium. They were mined only for local livelihood and industry combustion at the beginning. Since 1970s, coal mines were exploited for germanium production [2]. The average activity concentration of natural radionuclide ^{238}U in coal ores is 624 Bq/kg, while the maximum is 2. 17 kBq/kg [3]. The research area locates in countryside with a population density of 195 per square kilometer. Climate is mild in this area and is suitable for planting common cereals and vegetables.

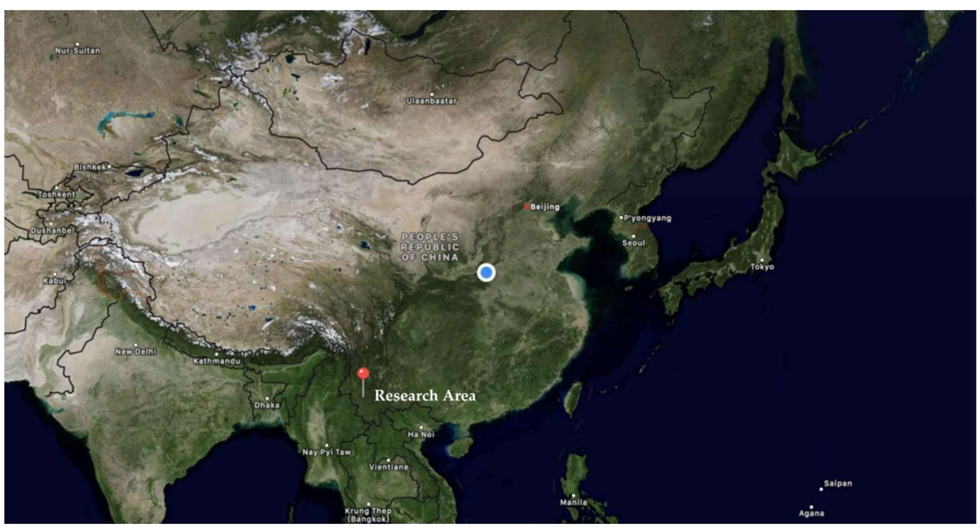

Figure 1. The location of research area.

The monitoring program was carried out in 21 sampling locations. The ingestion doses were estimated for adult residents from 11 villages. The population of the villages ranges from 206 to 4367. The locations of sampling, coal mines and villages are shown in Figure 2.

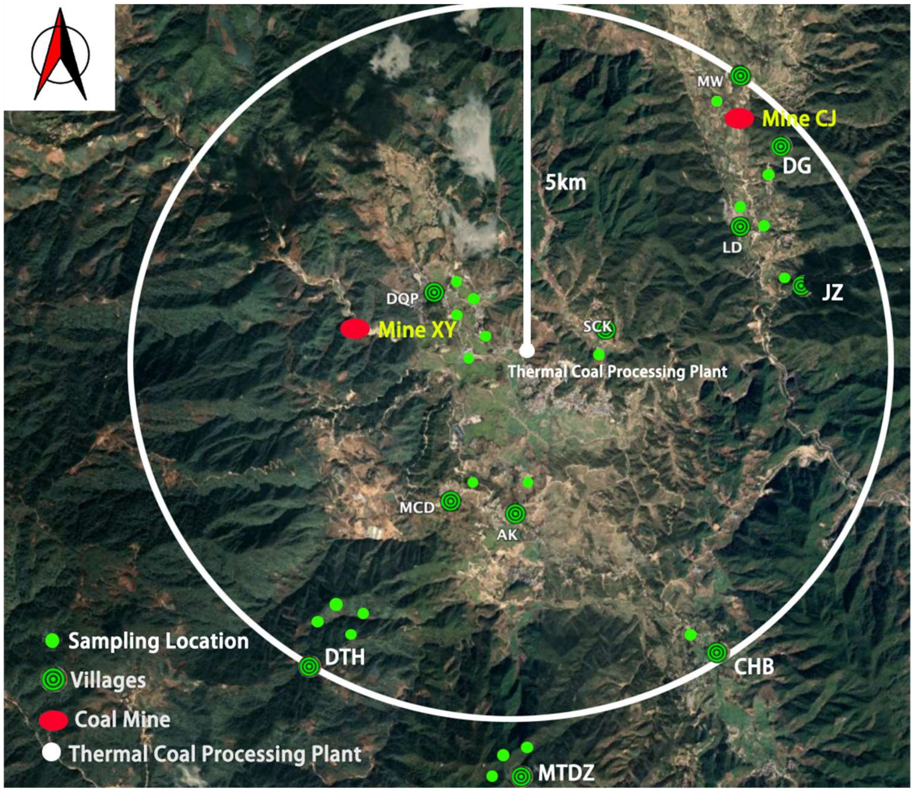

Figure 2. The locations of sampling sites, coal mines and villages.

The research area covers a radius of 5 km, in which there are 2 coal mines located, a thermal coal processing plant is located in the center. One of the 2 coal minse is Mine XY located in the western part and is 2.5 km from the center, while another is Mine CJ located in the northeast part and is about 5 km far away from the center of this area. The annual production of germanium for Mine XY is 50,000 t, while the annual coal production of Mine CJ is about 8000 to 10000 t.

There are 4 villages located in the area surrounding Mine CJ, which are Village MW, Village DG, Village LD and Village JZ. The farmlands in the area are distributed along a small river. Village MW is nearest to Mine CJ and at the upstream, while the other villages are at the downstream of the river from Mine CJ. There are 4 villages near the Mine XY, which are Village DQP, Village SCK, Village MCD and Village AK. Villages and farmlands locate in a flat area along the valley.

In research area, Village CHB and Village DTH are far from two coals mines. It is about 6 km from Village CHB to Mine XY. But farmland around Village CHB at the downstream in the valley may be effected by the river release from Mine XY. Village DTH locates at a higher altitude than Mine XY dose. Farmland around the village may not be affected by Mine XY geographically. However, there are large quantities of mining wastes and ore tailings around this area.

Village MTDZ is outside the research area and on the different side of the mountain. Crops growing in the farmland around the village were not affected by mining and processing. Therefore, concentrations of radionuclides in the samples from Village MTDZ were considered as a reference level, or the background level of this area.

2.2. Analysis Methods

Corns, wheats, rice, greens and plantains grown in the research area were collected as representative samples. In the laboratory, the samples were washed and dried. The ratios of dry weight to fresh weight for cereals, root vegetables and leafy vegetables are 1.0, 0.3 and 0.1 respectively. Then the dry samples were pretreated to analytical samples.

To measure the activity concentrations of ^{210}Pb, the crop samples were dissolved to solutions at first by wet erosion method in the laboratory. The stable Pb carrier was added to the solution samples for the recovery rate in the chemical and physical process. ^{210}Pb and Pb carrier in the solution samples were adsorbed and concentrated by Fe $(OH)_3$ co-precipitation undergoing heating. ^{210}Pb and its carrier were isolated from most interference by an anion exchange. The $PbSO_4$ precipitation had been purified via being precipitated for three times. The $PbSO_4$ precipitation had been stored in the environmental temperature for one month. And then, the beta rays of ^{210}Bi and the daughter of ^{210}Pb were detected to determine the radioactivity concentration of ^{210}Pb [23].

The activity concentrations of ^{210}Po were measured with its 5.30 MeV alpha particle emission. ^{209}Po (4.88 MeV, $t_{1/2}$ = 109 y) was used as the internal tracer. The polonium tracer was added at first, then samples were dissolved by concentrated 20 mL HNO_3 respectively. After an addition of 2 mL HCl, the solutions were evaporated to dryness. Ascorbic acid was added to reduce Fe^{3+} to Fe^{2+}, and then polonium was plated on a silver disc from a dilute HCl medium. PIPS - alpha spectrometer (PIPS: Passivated Implanted Planar Silicon) with 4 detectors range from 450 mm^2 to 1200 mm^2 was used to measure the alpha activities. The detection limit of ^{210}Po in samples is 0.0003 Bq. Sample count times were 864000 s and the maximum counting errors are in the order of ± 10% [24, 25].

2.3. Dose calculations

For public exposure to radionuclides, ingestion of radionuclides is a significant route of intake. Elements incorporated into food maybe more readily absorbed from the gastrointestinal (GI) tract than inorganic forms of these elements, according to ICRP Publication 103 [26].

The ingestion doses for adults are calculated as the following general equation:

$$E_{ing,p}=C_{p,i}DF_{ing}H_p \tag{1}$$

where

$E_{ing,p}$ is the annual effective dose from consumption of nuclide i in foodstuff p (Sv/a),

$C_{p,i}$ is the concentration of radionuclide i in foodstuff p at the time of consumption (Bq/kg),

DF_{ing} is the dose coefficient for ingestion of radionuclide i (Sv/Bq),

H_p is the consumption rate for foodstuff p (kg/a).

Dose coefficients provide numerical links between E_{ing} and measurable quantities, which means the intake of radionuclides by ingestion in this paper. Dose coefficient used for ingestion of ^{210}Pb and ^{210}Po are shown in Table 1 [27].

Table 1. Dose coefficient for ingestion dose.

Radionuclides	Parameters	Value	Unit
^{210}Pb	Dose coefficient	6.9E-7	Sv/Bq
^{210}Po		1.2E-6	Sv/Bq

3. Results

3.1. Monitoring Data

The activity concentrations of ^{210}Pb and ^{210}Po in samples were analyzed in dry weight (DW). Samples included corns, wheats, rice, greens and plantains. The values of average and range of activity concentrations analyzed in each kind of crops are shown in Table 2.

Table 2. Activity concentrations of ^{210}Pb and ^{210}Po in samples (Bq/kg.DW).

Samples	Numbers	^{210}Pb	Range	^{210}Po	Range	Types
Rice	2	7.26	7.25-7.27	5.81	5.80-5.81	Cereals
Wheats	1	1.92	1.92	2.73	2.73	Cereals
Corns	12	0.35	0.16-1.13	0.45	0.16-0.81	Cereals
Greens	3	4.07	2.42-6.42	2.19	1.74-2.59	Leafy vegetables
Plantains	3	1.63	1.19-3.42	1.32	0.80-1.67	Root vegetables

3.2. Ingestion Dose Estimation

Monitoring data of samples were collected from research area and the adults' food consumption rate of local residents were investigated to estimate the ingestion doses. The adults' consumption rates of cereals, root vegetables and leafy vegetables are shown in Table 3 [28].

Table 3. Representative adults' consumption rates of local residents.

Vegetative food	Cereals (kg.DW /a)	Root vegetables (kg.FW /a)	Leafy vegetables (kg.FW /a)
	126.2	29.1	132.3

While estimation, the activity concentrations of ^{210}Pb or ^{210}Po in samples are represented for dry weight, whereas the annual intakes of root vegetables and leafy vegetables are represented for fresh weight, annual intake of cereals is represented for dry weight. Therefore, when radionuclides intake was calculated, the ratios of dry weight to fresh weight were considered.

The ingestion doses of adult villagers living in research area and Village MTDZ are shown as follows. Contributors of different type of crops are shown in Table 4, while contributors of different radionuclides are shown in Table 5.

Table 4. The annual ingestion doses of adult due to intake of ^{210}Pb and ^{210}Po in different crops (mSv/a).

Villages	Cereal intake	Root vegetable intake	Leafy vegetable intake	Total intake dose
MW	0.579	0.023	0.094	0.697
DTH	0.830	0.023	0.094	0.948
SCK	0.053	0.023	0.094	0.171
DQP	0.089	0.030	0.094	0.213
MCD	0.087	0.023	0.094	0.205
DG	0.086	0.023	0.094	0.203
LD	0.087	0.023	0.094	0.204
JZ	0.102	0.023	0.094	0.219
AK	0.136	0.023	0.094	0.254
CHB	0.129	0.023	0.094	0.246
Average	0.218	0.024	0.094	0.336
MTDZ	0.042	0.023	0.060	0.125

Table 5. The annual ingestion doses contributed by ^{210}Pb and ^{210}Po in crops (mSv/a).

Villages	^{210}Pb intake	^{210}Po intake	Total intake dose
MW	0.236	0.461	0.697
DTH	0.419	0.529	0.948
SCK	0.090	0.081	0.171
DQP	0.136	0.076	0.213
MCD	0.083	0.121	0.205
DG	0.088	0.115	0.203
LD	0.099	0.106	0.204
JZ	0.090	0.129	0.219
AK	0.083	0.171	0.254
CHB	0.083	0.164	0.246

Average	0.141	0.195	0.336
MTDZ	0.051	0.074	0.125

4. Discussion

4.1. The Correlation of the Radionuclides in Crops and Soils

In our study, each crop sample corresponding to a soil sample was collected at the same location. The concentrations of ^{210}Pb and ^{210}Po in corn and soil samples are shown in Figure 3. Due to nearly equilibrium in decay chains, the concentrations of ^{210}Pb and ^{210}Po in soil are almost equal.

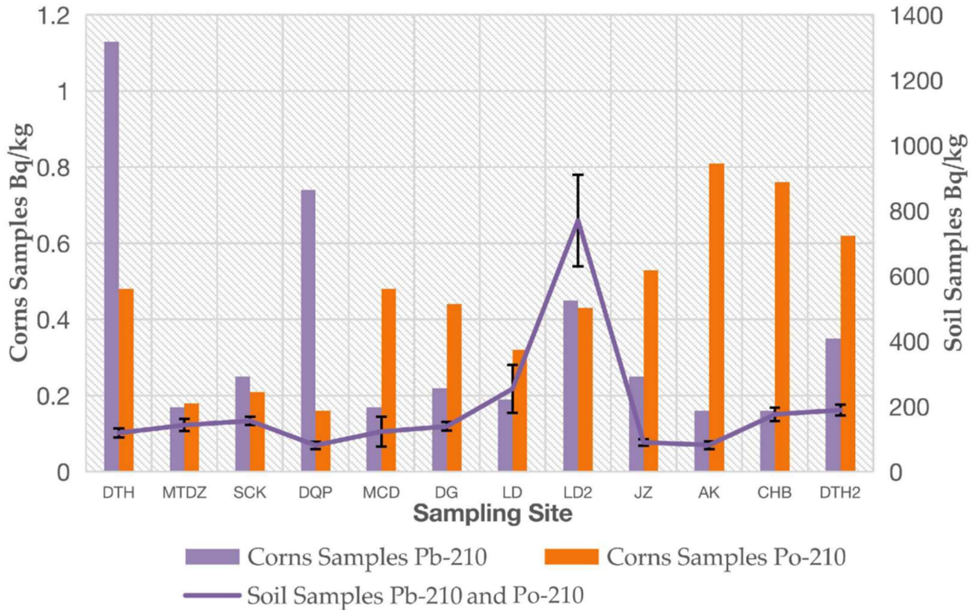

Figure 3. The activity concentrations of ^{210}Pb and ^{210}Po in crop sample and its corresponding farmland soil sample.

In Figure 3, the correlation of the activity concentrations of ^{210}Pb or ^{210}Po in crops and soils is not obvious. This situation indicates that transfer of ^{210}Pb or ^{210}Po from soil to crop may be affected by various factors, such as contaminated soil, soil classification, irrigation water.

Behaviours of plants accumulating radionuclides from the soil and irrigation water depend on biological characteristics and radioecology parameters. Different accumulation of radionuclides in crops can be seen in Figure 4.by comparing the activity concentrations in different types of crops collected in the same position. In Village DTH, 2 corns samples and 2 rice samples were collected, and in Village MTDZ, 1 corn sample and 2 leafy vegetables samples were collected, while in Village DQP, 3 root vegetables samples and 1 leafy vegetable sample were collected. The comparison results of the activity concentrations of ^{210}Pb and ^{210}Po in the samples are shown

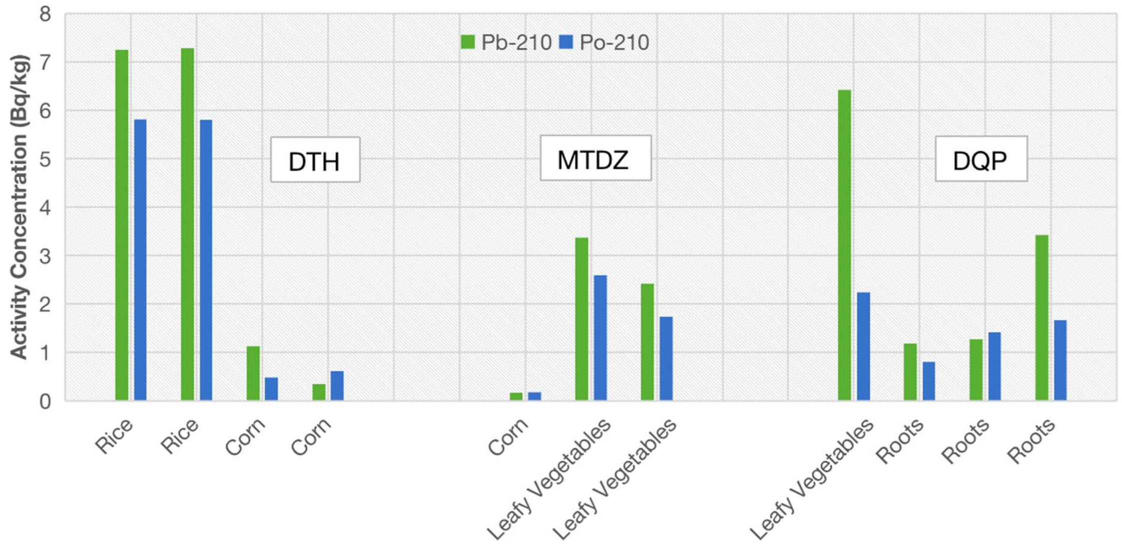

Figure 4. The radionuclides concentrations of different type of crops in 3 villages.

In Village DTH, the activity concentrations of ^{210}Pb or ^{210}Po in rice are much higher than that in corns. Due to the same soil conditions, so that transfer factor of rice is higher than that of corn. This situation is the same as description in the Technical Reports Series No. 472 [29] published by the IAEA, in which transfer factors of ^{210}Pb or ^{210}Po in soil to rice are 8.4E-03 and 1.3E-02, transfer factors of ^{210}Pb or ^{210}Po in soil to corns are 8.5E-04 and 2.4E-04 respectively.

In Village MTDZ, the activity concentrations of ^{210}Pb or ^{210}Po in leafy vegetables are higher than that in corns. The concentration ratios of ^{210}Pb or ^{210}Po for leafy vegetables are 8.2E-02 and 7.4E-03 in soil respectively, which are higher than that for corns.

In Village DQP, the activity concentration of ^{210}Pb or ^{210}Po in leafy vegetables is much higher than root vegetables.

4.2. Ingestion doses

The average annual effective doses of adult in China and the worldwide due to ingestion of ^{210}Pb or ^{210}Po in crops were calculated. The reference levels of ^{210}Pb and ^{210}Po in cereals, root vegetables and leafy vegetables of China and the world are shown in Table 6 [30, 1].

Table 6. The annual ingestion doses contributed by ^{210}Pb and ^{210}Po in foods (mSv/a).

Reference Level	Cereal		Leafy vegetables		Root vegetables		Total intake dose
	^{210}Pb	^{210}Po	^{210}Pb	^{210}Po	^{210}Pb	^{210}Po	mSv/a
	Bq/kg	Bq/kg	Bq/kg	Bq/kg	Bq/kg	Bq/kg	
World	0.05	0.06	0.08	0.10	0.03	0.04	0.042
China	0.03	0.04	0.36	0.43	0.03	0.03	0.112

* For world reference level here, the food consumption rates of cereals, root vegetables and leafy vegetables for adult is 287.6 kg/a as the same food consumption rates of research area used in dose calculation.

Concentrations of naturally occurring radionuclides in foods vary widely with the different background levels, contaminated soil, as well as climate, and agricultural conditions [31]. Meanwhile, Chinese traditional diet is quite different from western countries, which may lead to different ingestion doses.

The average annual effective dose due to ingestion of ^{210}Pb and ^{210}Po in crops for adult living in research area was 0.336 mSv/a, with the range from 0.171 to 0.948 mSv/a. While the dose for adult living in Village MTDZ was 0.125 mSv/a, as a reference level, or the background level of the research area. The background level is higher than the reference level of both China and the world. The estimated ingestion doses of adult living in the villages which are around coal mines are shown in Figure 5.

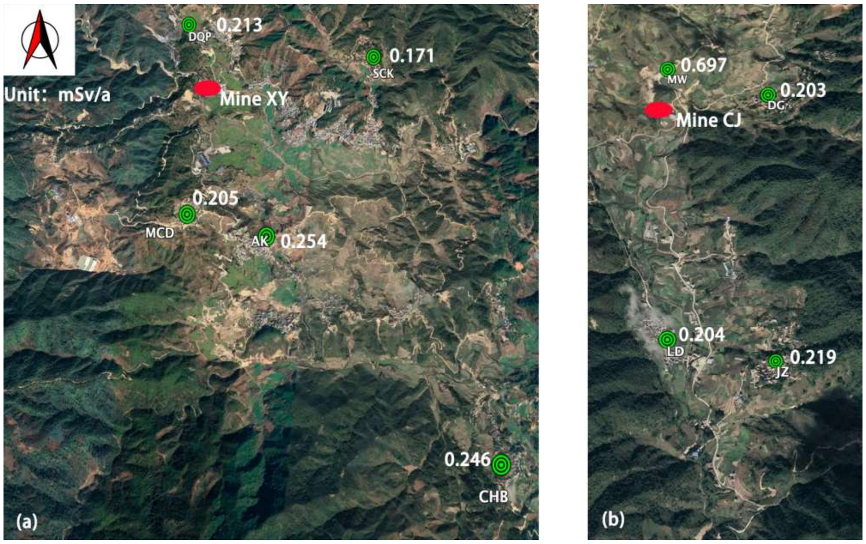

Figure 5. The ingestion doses estimated of adult residents living in research area(a) Villages around Mine XY; (b) Villages around Mine CJ.

The ingestion dose of adult living in Village MW was 0.697 mSv/a. Village MW is close to the Mine CJ. There is a river passing Village MW to Village DG, Village LD and Village JZ. It is a source of irrigation water for farming in this area. the radionuclides released into the downstream of the river from Mine CJ. As a result, the farmlands were affected. The study indicated that the ingestion doses of the adult living in Village DG, Village LD and Village JZ were lower than the value of residents in Village MW, but higher than the reference level. It implies that the radiological impact of mining activity may decrease with the distance. Although Village DQP is close to Mine XY, the ingestion dose of adult living in Village DQP was 0.213 mSv/a. Village DQP is affected less because it locates in upstream of Mine XY.

The ingestion dose of adult living in Village DTH was 0.948 mSv/a, which was the highest in the research area. It implies that farmlands in Village DTH were contaminated more serious than others. The estimation result of residents living in Village MTDZ was 0.125 mSv/a, which is the lowest, and as the background level of this area. It is close to the China reference level of 0.112 mSv/a. Village MTDZ was less polluted by the exploitation of coal mines.

5. Conclusions

In our study, twenty-one samples of cereals, root vegetables and leafy vegetables grown on the farmlands in the research area were collected. The activity concentrations of key radionuclides of ^{210}Pb and ^{210}Po in samples were analyzed. The radioactivity concentrations of the rice were the highest, while that of corns were the lowest. The average activity concentration of ^{210}Pb for rice, wheats, corns, greens and plantains was 7.26 Bq/kg, 1.92 Bq/kg, 0.35 Bq/kg, 4.07 Bq/kg and 1.63 Bq/kg, and value of ^{210}Po for rice, wheats, corns, greens and plantains was 5.81 Bq/kg, 2.73 Bq/kg, 0.45 Bq/kg, 2.19 Bq/kg and 1.32 Bq/kg respectively. According to the China's national standard of the Limited Concentrations of Radioactive Materials in Foods [32], the limited concentration of ^{210}Po for cereals, root vegetables and leafy vegetables is 6.40 Bq/kg, 2.80 Bq/kg and 5.30 Bq/kg respectively. Therefore, activity concentration of ^{210}Po in all crops did not exceed the limited concentrations of China's national standard.

The effective doses due to ingestion of ^{210}Pb and ^{210}Po in crops were estimated for adult living in 10 villages in research area, while the result in Village MTDZ was considered as the reference level. The average ingestion doses of adults living in research area was 0.336 mSv/a, with a range from 0.171 to 0.948 mSv/a. The estimation result for MTDZ's villagers was 0.125 mSv/a. Comparing with the reference levels of China and the world, 0.112 and 0.042 mSv/a respectively, the estimation values of local residents are much higher. The ingestion doses of local residents are about three times as many as the China reference level, and eight times of the world reference level. The results indicated that the mining and processing of coal mines for 40 years in research area resulted in an obviously radiological impact on the environment, and led to a significantly increase in ingestion doses of local residents. The results also revealed that when the farmlands in the villages were close to coal mines and located at downstream of mining activity, the ingestion dose of adult living in villages were higher. In order to reduce potential exposure risks, crops with low concentration ratios are suggested to be cultivated in farmlands contaminated slightly. For the farmlands seriously polluted, arrangements like soil remediation are required.

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