Investigation on natural radionucildes levels in multiple media in bone-coal mine areas of five provinces

KONG Ling-Li[1], ZHANG Liang[2], LI Ying[3], YE Ji-Da[4], JIANG Shan[5], SHI Jin-Hua[1], WU Zong-Mei[4]

([1] *Wuhan Academy of Environmental Science, Wuhan 430015;*

[2] *Environmental Monitoring Center Station of Hunan Province, Changsha 410004;*

[3] *Environmental Monitoring Center Station of Jiangxi Province, Nanchang 330029;*

[4] *Environmental Radiation Monitoring Station of Zhejiang Province, Hangzhou 310012;*

[5] *Environmental Radiation Surveillance Station of Anhui Province, Hefei 230061)*

Abstract This paper reports methods and results of the investigation of natural radionuclides levels in multiple media in the bone-coal mine areas in Hubei, Hunan, Jiangxi, Zhejiang, and Anhui Provinces. In the studied bone-coal mine areas, the specific activities of ^{238}U and ^{226}Ra in the soil samples were 0.37 and 0.24 Bq/g, respectively; the specific activities of ^{226}Ra in the samples of bone-coal, bone-coal cinder and bone-coal cinder brick (BCCB) were 1.3, 1.4 and 0.9 Bq/g, respectively. In the water samples collected from the bone-coal mine areas, the average concentrations of natural uranium and ^{226}Ra were 33μg/L and 58 mBq/L, respectively, while in the water samples collected from outside the bone-coal mine areas, they were 3.4μg/L and 45 mBq/L, respectively. In addition, the specific activities of ^{238}U and ^{226}Ra in air aerosol samples from the bone-coal mine areas were 0.6 and 0.5 mBq/m^3, respectively.

Key words Bone-coal, Bone-coal cinder, Bone-coal cinder brick (BCCB), Radionuclides, Specific activity

CLC number X837

1 Introduction

In five provinces of Hubei, Hunan, Jiangxi, Zhejiang and Anhui, there is a quite rich bone-coal reserve (over 4.54×10^{10} ton) and it is almost 90% of the total reserve of China. In the "National Investigation of Natural Radioactive Level" (abbreviated as "NINRL" hereinafter) from 1983 to 1990, it has been found that the specific activities of natural radionuclides such as ^{238}U and ^{226}Ra are rather high in bone-coal. This high activity is a main effect factor which causes the rise of activity level in bone-coal mine areas and the rise of radon concentration in bone-coal cinder brick (BCCB) houses. This effect has been recognized by the local environment protection administrations. It has been proved that the investigation of radionuclides level is very helpful for environment administrations to estimate and assess the environment impact of mining and utilizing bone-coal in China, and it also provides fundamental data and scientific basis for formulating policies, laws, regulations and standards for mining and utilizing bone-coal.

In 1991, National Environment Protection Agency and Safety Protection Bureau of the Chinese Nuclear Industry Corporation have drawn up the project of "the study of effect of mining and utilizing radioactivity-associated bone-coal on environment". There are five main items have been estimated and assessed in this project, including γ-ray doserate, natural radionuclides, concentration of ^{222}Rn, heavy metal elements, and estimation of additional dose. The investigation of natural radionuclides content in various media (soil, water and air) is one important part in the project. The investigation lasted more than two years, and was ended in 1993.

This paper will report the specific activities of natural radionuclides ^{238}U, ^{226}Ra, ^{232}Th and ^{40}K in the samples of soil, water and air collected from the

Supported by State Environment Protection Bureau and Chinese Nuclear Indnstry Corporation (Project No.90201001)

Received date: 2004-10-22

bone-coal mine areas in the five provinces.

2 Methods and instruments

2.1 Sample collection and preparation

The ambient media of the bone-coal mine areas in the five provinces have been sampled strictly by the method stipulated in Ref.[1]. When collecting the ambient solid samples, one should arrange the sample points as a quincunx in a square area of 10m×10m. Each sample of the five points was at least 1kg and the 3cm surface layer should be scraped off. Finally, one solid sample of about 1~3kg was taken from the mixture of the five samples by quartatering. In laboratory, before measurement, the solid samples were dried in the shade or by fire, ground into particles with the size of 0.016 mm, and sealed in the polythene boxes of ϕ75mm×50mm. The ambient water was sampled twice a year, once in high water season and once in low water season. Each water sample, 50L, was acidified by HNO_3 to pH2~3, then carried to laboratory for radiochemical analysis. The ambient air was sampled by the LXCL-15-1 filter membrane. Every aerosol sample was collected through the air of 10,000m^3 and was analyzed by the HPGe γ spectrometer.

Altogether, in this investigation, we have collected 81 samples of bone-coal, 58 of bone-coal cinder (BCC), 44 of bone-coal cinder brick (BCCB), 39 of soil, 8 of flue dust ash, 3 of cement, 34 of water and 8 of aerosol.

2.2 Measuring methods and instruments

The γ-ray spectrometry was used for radionuclides analysis. The instrument used for measuring specific activity of radionuclides is ADCAM100 HPGe γ spectrometer made by ORTEC Company, USA. For solid samples analyzed by the spectrometer, the lower level of detection (LLD) is 0.9 Bq/kg for ^{238}U, and 0.3 Bq/kg for both ^{40}K and ^{232}Th. For aerosol samples, the LLD is 2.6×10^{-5} Bq/m^3 for ^{238}U and ^{232}Th, and 0.86×10^{-5} Bq/m^3 for ^{226}Ra and ^{40}K. And for water samples, the LLD is 0.05 μg/L for natural uranium, 0.5 mBq/L for ^{226}Ra, 0.07 μg/L for natural thorium, and 0.6 mBq/L for ^{40}K.

3 Results

In this investigation, all the measuring methods and instruments for natural radionuclides in various media of bone-coal mine area can be traced to the national metrology standard.

3.1 Solid

The solid materials collected from the bone-coal mine areas of the five provinces include bone-coal, BBC, BCCB and soil. The natural radionuclides' mean specific activities of various solid materials are shown in Fig.1~ Fig.4, which demonstrate the differences between the natural radionuclides' mean specific activities of the four solid samples from the bone-coal mine areas and the activities of soil samples of the five provinces, which were measured in "NINRL". Except for the BCC and soil in Hubei Province, the specific activities of ^{232}Th and ^{40}K are lower than those of soil of the five provinces, given by "NINRL". The specific activities of ^{226}Ra in bone-coal, BCC, BCCB and soil are 1302, 1437, 963 and 237 Bq/kg, respectively, which are 28, 31, 21 and 5 times, respectively, as much as the ^{226}Ra's specific activities of soil given by "NINRL".[2-6] And the ^{226}Ra's specific activity in BCC of Hubei Province is 3133 Bq/kg, which is 84 times the ^{226}Ra's value in soil given by "NINRL".

The specific activities of ^{226}Ra, ^{232}Th and ^{40}K of Hubei, Hunan, and Zhejiang Provinces in flue dust ash, the total of ash in dust remover, chimney ash and flue ash, produced by bone-coal burning, are listed in Table 1. Table 1 also lists the specific activities of ^{226}Ra, ^{232}Th and ^{40}K for three kinds of cement samples mixed with bone-coal cinder from Hunan Province.

Table 1 shows that, except for the ^{40}K of the flue dust ash in Hubei, the specific activities of ^{232}Th and ^{40}K in flue dust ash of Hubei, Hunan and Zhejiang, are all close to or lower than the values in soil measured in "NINRL", and the mean specific activities of ^{226}Ra of the flue dust ash in Hubei, Hunan, and Zhejiang are 3699, 625 and 1748 Bq/kg, respectively, which are 99, 11 and 40 times, respectively, the values of soil in these three provinces measured by "NINRL". And among the three kinds of Hunan's cement samples mixed with bone-coal cinder, the specific activity of ^{226}Ra in clinkerless cement and in building cement is 210 and 240 Bq/kg, respectively, both of which are higher than 200 Bq/kg, the limit value set in Ref.[7].

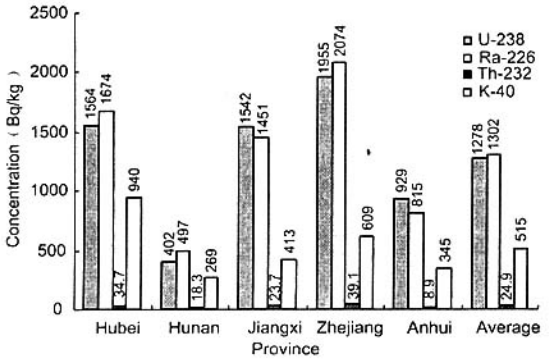

Fig.1 Concentrations of various radionuclides in bone-coal samples of the five provinces.

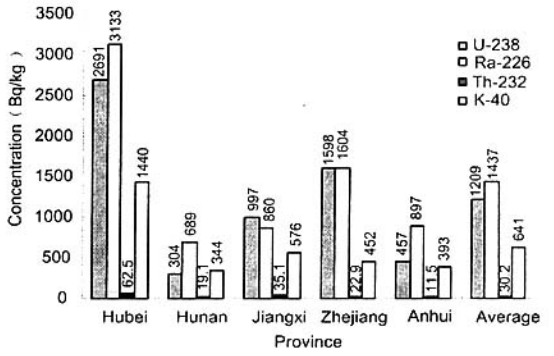

Fig.2 Concentrations of various radionuclides in bone-coal cinder samples of the five provinces.

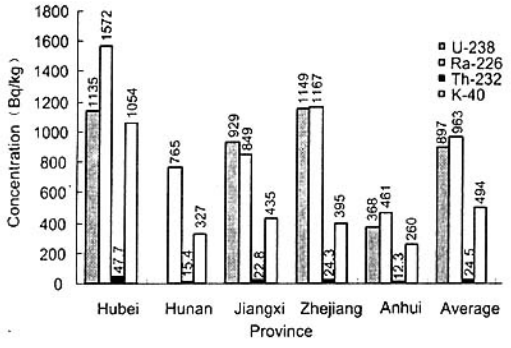

Fig.3 Concentrations of various radionuclides in bone-coal cinder brick samples of the five provinces.

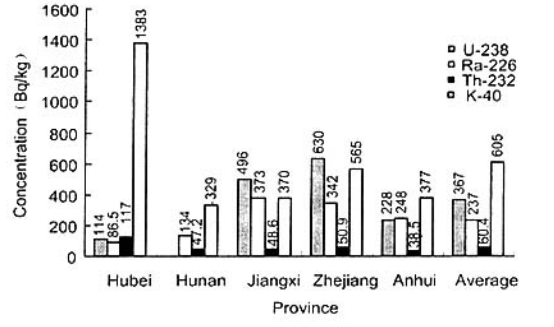

Fig.4 Concentrations of various radionuclides in soil samples from bone-coal areas of the five provinces.

Table 1 Specific activities of radionuclides in flue dust ash and cement from bone-coal electric power plants (Bq/kg)

Province	Item	*n*	^{226}Ra		^{232}Th		^{40}K	
			Specific activities	Mean	Specific activities	Mean	Specific activities	Mean
Hubei	Ash in dust remover		1653～7244	3699	22.4～94.5	47.7	63～2618	1209
Hunan	Ash in dust remover			589		27.9		369
	Chimney ash			600		30.2		292
	Flue ash			685		35.3		556
	Average	3		625		31.1		406
Zhejiang	Ash in dust remover	4	640～2200	1685	29.0～76.0	45.0	700～900	790
	Chimney ash	1	1670～2000	1810	40.3～68	50.0	590～835	747
	Average	2		1748		47.5		769
Average of the three provinces				2024		42.1		795
Hunan	Yiyang cement			52		21.0		160
	Clinkerless cement			210		14.0		240
	Building cement			240		18.0		210
	Average	3	52～240	167	14.0～21.0	17.7	160～240	203

Fig.5 shows the ratios of specific activities of ^{226}Ra, ^{232}Th and ^{40}K in BCC and flue dust ash to those in bone-coal. The radionuclides of ^{226}Ra, ^{232}Th and ^{40}K in bone-coal are concentrated partially in BCC and flue dust ash by burning. The condensation factors of ^{226}Ra, ^{232}Th and ^{40}K from bone-coal to BCC or flue dust ash are about 1.5.

3.2 Water

Table 2 shows the concentrations of natural radionuclides in water samples from galleries, escape canals, ponds, and rivers inside and outside the bone-coal areas in Hunan, Jiangxi, Zhejiang and Anhui Provinces.

In the water samples of bone-coal areas in the

four provinces, the mean specific activities of ^{226}Ra and ^{40}K and the mean concentrations of natural uranium and thorium are 58.1mBq/L, 244 mBq/L, 32.6 μg/L and 2.26 μg/L, respectively, which are 18, 3.5, 48 and 17 times, respectively, as much as the values for river waters of the four provinces, given by "NINRL".[8-11]

For the water samples from outside of bone-coal area in Jiangxi, Zhejiang and Anhui Provinces, the mean specific activities of ^{226}Ra and ^{40}K and the mean concentrations of natural uranium and thorium are 45.4 mBq/L, 964 mBq/L, 3.40 μg/L and 2.87 μg/L, respectively, which are 13, 13, 6.3 and 21 times, respectively, as much as the values measured in "NINRL". All the specific activities of natural uranium and natural thorium, and concentrations of ^{226}Ra and ^{40}K are close to or lower than the values measured in "NINRL" with an exception of values for pool water in Jiangxi Province, which are relatively higher.

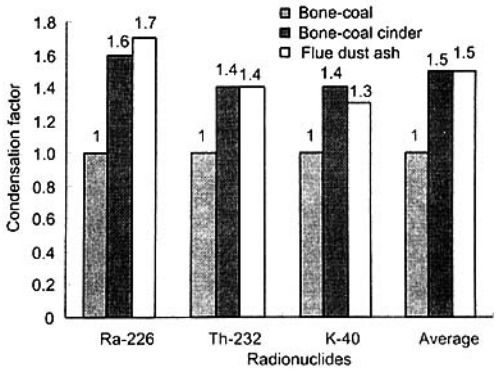

Fig.5 Condensation factors of radionuclides in various media.

Table 2 Concentrations of natural radionuclides in waters from inside and outside of bone-coal areas

	Provinces	Type	Natural uranium (μg/L)		Natural thorium (μg/L)		^{226}Ra(mBq/L)		^{40}K(mBq/L)	
			Range	$\bar{x}$	Range	$\bar{x}$	Range	$\bar{x}$	Range	$\bar{x}$
Inside mine areas	Hunan	Gallery water		59.7		$<L_D$[1]		100		320
		Drain water		15.3		$< L_D$		46		740
		Average		37.5		0.01		73		530
	Jiangxi	Gallery water	13.8–20.2	17.5	1.38–15.3	7.0	135–324	141	10.9–1500	333
	Zhejiang	Drain water	38.4–53.9	46.1	0.25–0.90	0.58	3.1–21.2	12.1	50–63.0	56.5
	Anhui	Middle reaches[2]	2.34–63.3	32.8	1.03–2.32	1.68	3.5–5.7	4.60	20.5–81.6	51.1
		Lower reaches[2]	5.21–46.7	26.0	0.90–1.53	1.22	3.5–13.2	8.35	35.7–92.9	64.3
		Average		29.4		1.45		6.48		57.7
Average of the four provinces				32.6		2.26		58.1		244
Outside mine areas	Jiangxi	Pool water	3.0–12.9	7.95	4.4–9.45	6.90	91–169	130	43.1–5550	2797
	Zhejiang	Up of pour-in-point[3]	0.16–0.30	0.23		$< L_D$	0.7–3.97	2.34	50.0–54.8	52.4
		Down of pour-in-point[4]	0.21–0.30	0.26		$< L_D$	0.8–2.02	1.41	50.0–55.3	52.6
		Average		0.25		0.04		1.88		52.5
	Anhui	Upper reaches[2]	0.50–1.73	1.12	1.46–1.85	1.66	3.1–10.8	6.95	16.9–50.6	33.8
		Water of river[5]	0.73–5.08	2.90	1.32–1.98	1.65	1.4–1.8	1.60	46.7–54.0	50.4
		Average		2.01		1.66		4.28		42.1
Average of the three provinces				3.40		2.87		45.4		964

Notes: 1) If the measured value is $<L_D$, half L_D was used instead for calculation; 2) Upper reaches, middle reaches, lower reaches of two little rivers which flow past the bone-coal areas; 3) 100 m up from the pour-in-point in Fuchunjiang River; 4) 100 m down from the pour-in-point in Fuchunjiang River; 5) The river is connected with little rivers which past the bone-coal areas.

3.3 Gas

Fig.6 shows concentrations of natural radionuclides in aerosol samples collected from Anren bone-coal mine areas of Zhejiang Province.

The results show that the concentrations of natural radionucildes in Anren bone-coal mining area and living quarters are both higher than those in the reference points. Especially the concentration of ^{238}U and ^{226}Ra is 9.1 times and 8.5 times the concentration in the reference points, respectively. The highest concentrations occurred in mining areas, the lowest ones oc-

curred in the reference points, and concentrations in the living quarters were placed in the middle.

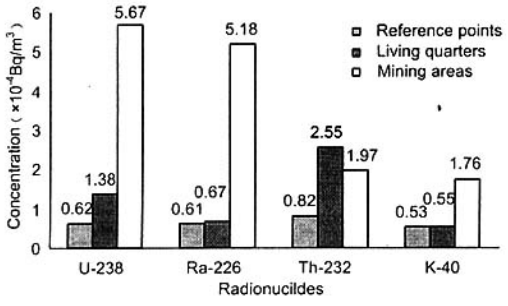

Fig.6 Concentrations of radionuclides in aerosol in Anren bone-coal areas.

3.4 Relocation of radionuclides

Fig.7 shows the transfer quantities of ^{238}U from underground in the five provinces. These magnitudes were estimated based on the specific activities of bone-coal and annual output of bone-coal.

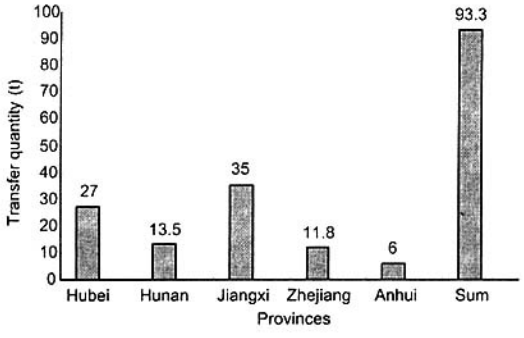

Fig.7 The transfer quantity of natural uranium in bone-coal in the five provinces.

In the bone-coal mines of the five provinces, there is 93.3 ton natural uranium transferred from underground to ground every year, through mining and using of bone-coal. And the influence of radioactivity of natural uranium on the ground on the environment should not be neglected.

4 Conclusions

4.1 Strengthen monitoring

The investigation shows that the bone-coal mining and utilizing was associated with radioactive exposure, and the specific activities of ^{238}U and ^{226}Ra were quite high in some bone-coal areas. In order to protect miners as well as environment itself in the bone-coal areas, we must strengthen the radioactive monitoring during the whole process of bone-coal mining and utilizing. And ^{226}Ra should be the key radionuclide to be monitored.

4.2 Strictly implement relevant standard

In Hubei, Hunan, Jiangxi, Zhejiang and Anhui Provinces, the specific activities of ^{226}Ra in bone-coal cinder are quite high, with the mean values from 689 to 3133 Bq/kg. Especially when the bone-coal cinder is used as building materials, it is important that the Standard GB6566-2001 "Limit of radionuclides in building materials" should be strictly implemented.

Acknowledgement

We would like to express gratitude to Professors of Pan Ziqiang, Chen Zhuzhou, Hu Fengquan, He Zhenyun, Zhang Weiming for their technical support in this investigation.

References

1 National Environment Protection Agency, Regulation of environmental activity level investigation (a restricted publication, in Chinese), 1986

2 Hubei Environmental Monitoring Centre Station. Radiat Prot (in Chinese), 1992, **12**(4): 300

3 Zhuang Nanfu, Huang Yi, Zeng Lichu, *et al.* Radiat Prot (in Chinese), 1991, **11**(2): 145

4 Li Xinde, Zheng Shuihong, Wu Xiangrong, *et al.* Radiat Prot (in Chinese), 1993, **13**(6): 291

5 Zhejiang Environmental Monitoring Centre Station. Radiat Prot (in Chinese), 1986, **6**(4): 287

6 Zhu Jinqiu, Shao Guangnan, Jiang Shan, *et al.* Radiat Prot (in Chinese), 1991, **11**(4): 295

7 State Standard of PRC, Limit of radionuclides in building materials, GB6566-2001 (in Chinese)

8 Hunan Environmental Monitoring Centre Station. Radiat Prot (in Chinese), 1991, **11**(2): 136

9 Zhejiang Environmental Monitoring Centre Station. Radiat Prot (in Chinese), 1986, **6**(4): 320

10 Sun Yiru, Zheng Shuihong, Wu Xiangrong, *et al.* Radiat Prot (in Chinese), 1991, **11**(5): 358

11 Chen Shuping, Zhu Jinqiu, Wang Weining, *et al.* Radiat Prot (in Chinese), 1991, **11**(4): 295