# Natural radioactivity level of associated bone-coal mining area in Zhejiang Province

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**Abstract** The geographic distribution,  $\gamma$ -radiation level and specific activity of radionuclides of the bone-coal mines in Zhejiang Province were reported. The weighted average of  $\gamma$ -radiation dose rate of the bone-coal mines is 566 nGy/h for 107 main bone-coal mines. The weighted mean activity of  $^{238}$ U,  $^{226}$ Ra,  $^{232}$ Th and  $^{40}$ K in the samples are 949, 918, 34 and 554 Bq/kg for 171 samples of bone-coal, respectively.

**Keywords** Bone-coal, Natural radioactivity,  $\gamma$ -radiation level, Radionuclides

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#### 1 Introduction

Bone-coal is a coal resource reserved in ancient strata. About 5 billion years ago, under the shallow sea environment, it was generated by the lower forms of life, for example, algae and mushrooms, when the stratum was moving and suffered the anticyclone, and it is a saprogenic coal of no smoke with high mutation. The bone-coal reserve in Zhejiang Province is relatively rich, and the chronicle of mining and utilizing bone-coal is long-standing. More than one hundred years ago, local people spontaneously started to mine the bone-coal on a small scale. Instead of charcoal, it was used to make lime and then the lime was used to build houses or to improve the acidic soil. By 1970's, the bone-coal had been extensively applied in Zhejiang Province. Because of high content of radionuclides and bad resolvability, mining and utilizing of bone-coal may cause environment radioactive pollution and increase the workers' additional dose. In order to find out the distribution of  $\gamma$  radiation level of bone-coal mines and the impact on surrounding environment, and to take some managerial and protective measures, it is essential to carry out a general investigation about the radiation level of bone-coal in Zhejiang Province.

This paper reported the  $\gamma$  radiation level and the content of radionuclides  $^{238}$ U,  $^{226}$ Ra,  $^{232}$ Th and  $^{40}$ K in

four bone-coal mine belts in Zhejiang Province. And some suggestions are given on the bone-coal's reasonable mining and utilization.

# 2 Characteristics and distribution of bone-coal

The bone-coal mines in Zhejiang Province have some characteristics, such as shallow embedment, high thickness stable stratum and easy exploitation. There are much ash (65%~80%) and sulfur (2%~5%) included in the bone-coal. The content of carbon and heat value are low, i.e. (10%~25%) and (3400~10000) kJ/kg, respectively.

There are three bone-coal strata, which are the Sinian Period, Cambrian Period and Permian Period. The bone-coal of low Cambrian Pystem is the most rich, which came into existence mainly in middle section of Hetang formation. At the present, the reserves of bone-coal proved up are 1.6 billion ton in Zhejiang Province, while the forecast capacity may reach 10.6 billion ton. The bone-coal is mainly distributed in four belts in the west of Zhejiang Province. The aspects of the bone-coal belts are from southwest to northeast with about 400 km long and 50 km wide (Fig.1). The first belt is from Chihuai Village in Kaihua County to Yaocun Village in Anji County, passing Chun'an County, Lin'an County and Hangzhou City. The

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length of the belt is 200 km with 2 to 4 strata of bone-coal and a thickness of 40 to 50 m. The second belt is from Huibu Village in Changshan County to Longmen Village in Fuyang County, passing Quzhou City, Jiande County and Tonglu County. There is 1 to 3 strata of bone-coal, and their thickness is from 15 to 37 m. The third belt is from Dachen Village in Jiangshan County to Xiangyang Village in Shaoxing County, passing Jiande County, Pujiang County and Zhuji County. There is one stratum of bone-coal with a thickness of 2 to 11 m. The fourth belt is from Yinzhu Village in Tonglu County to Moganshan Hill in Deging County, passing Fuyang County, Lin'an County and Hangzhou City. During early 1970's, the utilization of bone-coal in Zhejiang Province has reached a new stage through large-scale exploration and exploitation. The bone-coal is used not only for calcinations traditionally, but also for many purposes such as generating electricity, chemical industry, building materials industry and so on. The annual productivity of bone-coal in Zhejiang Province has reached 2~4 million ton.



**Fig.1** Distribution of bone-coal mine belts in Zhejiang Province.

#### 3 Measurement and instruments

In this investigation, all procedures including the measurement of  $\gamma$  radiation level of bone-coal mine areas, the collection of bone-coal samples and the specific activity analysis of radionuclides in bone-coal were carried out according to 《Regulation of the Investigation of Radioactivity Level of Environment》.

The number of measurement points of  $\gamma$ -ray dose rate is more than 5 on the basis of mining scale at every bone-coal mine. The measurement height is 1 m from ground. The reference spot was located in the field of 2-3 km far away from bone-coal mine area. The γ-ray doserate at the reference spot was regarded as background (including cosmic radiation). One kilogram bone-coal was sampled as a sub-sample after rooting out 3 cm surface layer in one of five sub-sampling points distributed in an area of 10m×10m which shaped like a plum blossom. The 1~3 kg bone-coal was collected as a sample after the five sub-samples were mixed equally. Number of the bone-coal samples collected in the first, second and third belt is 15, 43 and 113, respectively, and no sample was collected in the fourth belt because of shorter bone-coal belt and lower  $\gamma$  radiation level. Five samples of local ordinary coal were collected as the reference samples of bone-coal.

The SG-102 X- $\gamma$  doserate meter (Laoshan Electronic Instrument Research Institute, China) was used for measuring  $\gamma$ -ray doserate. The ADCAM100 HPGe  $\gamma$  spectrometer (ORTEC Company, USA) was used for measuring specific activity of radionuclides. Its lower limit of detection (LLD) is 0.9 Bq/kg for <sup>238</sup>U and <sup>40</sup>K, and 0.3 Bq/kg for <sup>232</sup>Th and <sup>226</sup>Ra.

#### 4 Doserate of the bone-coal mines

The  $\gamma$ -ray doserate of 107 bone-coal mines with larger-scale is listed in Table 1. The  $\gamma$ -ray doserate of 67 bone-coal mines in Zhejiang Province is lower than 500 nGy/h (the background deducted, the same below), accounting for 62.6% of total bone-coal mines; that of 34 mines ranges from 500 to 2000 nGy/h, accounting for 31.8%; while that of 6 mines is more than 2000 nGy/h, accounting for 5.6%. The maximum  $\gamma$ -ray doserate is 3125 nGy/h of Zhuge bone-coal mine at the boundary of Lanxi City and Jiande County in the third belt.

The  $\gamma$ -ray doserates among the four bone-coal belts are obviously different. The mines with  $\gamma$ -ray doserate more than 500 nGy/h in the first, second, third and fourth belt account for 45.5%, 18.8%, 54.8% and 14.8% of the total bone-coal mines, respectively. The average  $\gamma$ -ray doserate for the four belts is 487, 316, 879 and 253 nGy/h, respectively.

**Table 1**  $\gamma$ -ray doserate and its distribution

Belt No.	Number of mines	Doserate (nGy/h)	Average (nGy/h)	<500(	<500(nGy/h)		500~2000(nGy/h)		>2000(nGy/h)	
				$\overline{n}$	(%)	n	(%)	n	(%)	
	22	108 ~ 1213	487	12	(54.5)	10	(45.5)	0		
	16	17 ~ 1004	316	13	(81.2)	3	(18.8)	0		
	42	3 ~ 3125	879	19	(45.2)	17	(40.5)	6	(14.3)	
	27	49 ~ 800	253	23	(85.2)	4	(14.8)	0		
Total	107	3 ~ 3125		67	(62.6)	34	(31.8)	6	(5.6)	

We divided each belt into three sections, i.e. upper section (north of 30°20'N), middle section (30°20'N~ 29°20'N) and bottom section (south of 29°20'N). The doserate in each section in four

bone-coal belts is shown in Table 2. There is a falling trend from southwest to northeast for all the four belts in Zhejiang Province.

**Table 2** γ-ray doserate distribution of each section in the four bone-coal belts in Zhejiang Province

Belt No.	Section	<500 nGy/h		500~	500~2000 nGy/h		>2000 nGy/h	
		n	(%)	n	(%)	n	(%)	
	Upper	1	8.3	1	10			
	Middle	5	41.7	3	30			
	Bottom	6	50.0	6	60			
	Upper							
	Middle	9	69.2	1	33.3			
	Bottom	4	30.8	2	66.7			
	Upper							
	Middle	17	89.5	11	64.7	1	16.7	
	Bottom	2	10.5	6	35.3	5	83.3	
	Upper	4	17.4					
	Middle	19	82.6	4	100			
	Bottom							

Table 3 lists  $\gamma$ -ray doserate values of 107 bone-coal mines in 18 counties or sities of five districts. The average  $\gamma$ -ray doserate values were acquired by the weighted average method of samples (every bone-coal mine as a measurement point). The average  $\gamma$ -ray doserate of five districts is given in a decreasing order as follows: Jinhua (958 nGy/h), Quzhou (751 nGy/h), Shaoxing (607 nGy/h), Hangzhou (348 nGy/h) and Huzhou (314 nGy/h).

#### 5 Content of natural radionuclides

The analysis result of natural radionuclies <sup>238</sup>U, <sup>226</sup>Ra, <sup>232</sup>Th and <sup>40</sup>K in 171 bone-coal samples collected from the first, second and third belt in this investigation is shown in Table 4. The average specific

activity of <sup>238</sup>U, <sup>226</sup>Ra, <sup>232</sup>Th and <sup>40</sup>K in the 171 bone-coal samples is 949, 918, 34 and 554 Bq/kg, respectively. The average specific activity of <sup>238</sup>U in the bone-coal samples is 22 times that in ordinary coal sample (43.6 Bq/kg) of Zhejiang Province. The average specific activity of <sup>226</sup>Ra in the bone-coal samples is 18 times that in ordinary coal sample (50 Bq/kg) of Zhejiang Province and 17 times that in ordinary coal sample (54 Bq/kg), China. <sup>[1]</sup>

The bone-coal sample collected in the third belt has the highest content of <sup>238</sup>U and <sup>226</sup>Ra, and that in the first belt has the lowest. There is a little difference in the content of <sup>238</sup>U and <sup>226</sup>Ra in the first and second belt, but the content of <sup>232</sup>Th and <sup>40</sup>K in the three belts is almost at the same level.

**Table 3** γ-ray dose rate in 18 counties and cities in Zhejiang Province (nGy/h)

District	County or city name	Number of monitoring spots*	Dose rate (nGy/h)	Average (nGy/h)	
Jinhua	Lanxi City	3	2846~3125	3076	
	Yiwu County	2	3~13	8	
	Pujiang County	7	47~705	321	
	Weighted average	12	3~3125	958	
Quzhou	Quzhou City	4	330~2165	820	
	Longyou County	3	622~1274	949	
	Kaihua County	12	127~908	492	
	Jiangshan County	5	649~2785	1410	
	Changshan County	4	135~1004	485	
	Weighted average	28	127~2785	751	
Shaoxing	Shaoxing County	7	109~1371	883	
	Zhuji County	12	172~864	446	
	Weighted average	19	172~1371	607	
Hangzhou	Chun'an County	8	122~1218	524	
	Jiande County	3	355~2785	1071	
	Fuyang County	8	99~800	357	
	Tonglu County	13	17~554	203	
	Yuhang County	5	75~326	189	
	Lin'an County	5	49~187	151	
	Weighted average	42	17~2785	348	
Huzhou	Anji County	2	108~508	308	
	Deqing County	4	150~424	317	
	Weighted average	6	108~508	314	
Total weighted average		107	3~3125	566	

<sup>\*</sup> Every bone-coal mine is regarded as a measurement spot.

Table 4 Specific activities of radionuclides in three bone-coal belts in Zhejiang Province (Bq/kg)

	Sample's number	<sup>238</sup> U		<sup>226</sup> Ra		<sup>232</sup> Th		<sup>40</sup> K	
Belt No.		Specific activity	Mean						
	15	9~984	402	8.2~1056	492	3.8~48	20.4	10.3~136	530
	43	10~1431	646	5.6~1300	694	14~62	37	52~900	571
	113	64~5200	1136	37~4797	1060	11~91	34	24.6~1413	551
Total	171	9~5200	949	5.6~4797	918	3.8~91	34	10.3~1413	554
Ordinary coal A1)	5		43.6		50				
Ordinary coal B <sup>2)</sup>	563				54				

Note: 1) Ordinary coal of Zhejiang Province; 2) Quoted from Ref.[1].

## 6 Discussion

The distribution of bone-coal mines in Zhejiang Province presents four belts. The investigation results

of 107 bone-coal mines are as follows:

(1) The average  $\gamma$ -ray doserate in the first, second, third and fourth bone-coal belts is 487, 316, 879 and 253 nGy/h, respectively. The  $\gamma$ -ray doserate of each

belt has a trend of gradually decreasing from southwest to northeast. The  $\gamma$ -ray doserate of six bone-coal mines in the third belt is more than 2000 nGy/h.

- (2) The average specific activity of  $^{226}$ Ra in the first, second and third bone-coal belts are 492, 694 and 1060 Bq/kg, respectively, and with a weighted average of 918 Bq/kg. The decreasing trend of the  $^{226}$ Ra content in the three bone-coal belts is almost the same as that or  $\gamma$ -ray doserate, which is gradually declined from southwest to northeast.
- (3) Most of radionuclides is still reserved in bone-coal cinder<sup>[2]</sup> after the bone-coal burned. Therefore the content of radionuclides in bone-coal must be

measured when the bone-coal is exploited and utilized. If the bone-coal cinder is adopted as building material, one must comply with the criteria set by GB6566-2001. [3]

## References

- Jiang Xiwen, Liu Qiusheng, Li Ruixiang, et al. Radiation Protection (in Chinese), 1989, 9(3): 181
- 2 Ye Jida, Kong Lingli, Li Yin, *et al*. Radiation Protection (in Chinese), 2004, **24**(1):1
- 3 State Standard of PRC: 《Limit of Radionuclides in Building Materials》, GB6566-2001, 2001 (in Chinese)