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Level, distribution, variation and sources of Pb-210 in atmosphere in North China

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Abstract

96 aerosol samples were collected in north China and analyzed by gamma spectrometry. The activity concentrations of Pb-210 ranged from 97 to 27,955 μBq/m^3, with average of 879, 3389 and 813 μBq/m^3 in Beijing, Changbai and Hunchun, respectively. Annual effective dose for inhabitants due inhalation varied from 0.68 to 5.94 μSv. There was a positive correlation between Pb-210 and $PM_{2.5}$ in Beijing. Seasonal variation of Pb-210 concentration was observed in Changbai and Hunchun. Analysis of their correlation with meteorological parameters could help to assess the effects of present pollutant emission and provide scientific support for prevention strategy.

Keywords Pb-210 · Outdoor air · Gamma spectrometry · Radioactivity

Introduction

Lead is a heavy metal, which mainly accumulates in human bones after uptake through the diet and inhalation. Lead-210, with a half-life of 22.3 years, is an important radioisotope of lead and regards as a high-toxicity radionuclide because of its chemical and radiological effect. Pb-210 in the air mainly originates from the decay of Rn-222 which is emitted from the earth to the atmosphere as gas. The constant releases of Rn-222 from the earth causes constant Pb-210 level in the air. The particle containing Pb-210 deposited on earth was therefore widely used as a chronology tool for dating of sediment. However, increasing human activities, such as mining, fuel burning and coal heating significantly increases the Pb-210 level in the air, because large amount of Pb-210 associated to particles and Rn-222 in gas form were released to the air during combustion in these processes [1, 2]. Lacking of fly ash removal systems in the factories where coal was used as a fuel or the coal is directly for domestic heating further increase the emission of Pb-210 into the air [3]. The Pb-210 in the air would finally enter human body through food chain and inhalation, which cause an exposure of Pb-210. Pb-210 is a beta emitter that decays into toxic alpha emitting Po-210 and beta emitting Bi-210 (1162 keV), which consequently cause a high radiation toxicity and possible health risk to humans. IAEA and WHO have developed operational intervention levels (OILs) and guidance levels regarding the activity concentration of Pb-210 in food and water, which is 2.0 Bq/kg and 0.1 Bq/L, respectively [4, 5]. OILs of Pb-210 in food given by IAEA safety standards is based on assumption of equilibrium radioactive chains with Bi-210, Po-210.

There are many large-scale coal-fired power plants and several smaller local power plants usually with insufficient fly ash removal systems in China [6]. In addition, air pollution including air particles (e.g. $PM_{2.5}$, PM_{10}) has become one of the most serious environmental problems in large

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part of China [7–10]. However, there is limited investigation on level of air Pb-210 in China. Meanwhile, its variation with season, location and $PM_{2.5}$, the major driving and control parameters on the level of air Pb-210 and its variation are still unclear. This work aims to measure Pb-210 concentrations in the air at three locations, Beijing, Changbai and Hunchun, with a different human activities in north China, and to investigate the controlling parameters influcing the Pb-210 level in the air.

Experimental

Equipment and sampling

Aerosol samples were collected at the roof of building in each investigated site through portable Staplex TF1A type (Clover company, USA) high-volume air sampler, the maximum sampling flow rate is 2 m^3/min, the 3M E853:5379 model aerosol membrane with size of BMF 20 g/m^2 (Whatman Company, UK) were used as filter when collect particles in the air. Collection efficiency of the membrane was larger than 90% for size of particles great than 0.2 µm. Angle of each sampler was properly arranged during sampling in order to avoid the gas discharge influence from each other of the sampler [11]. In this work, three sites were selected that stand for different scales of location in north of China, they are Beijing, Changbai and Hunchun. For the size of three cities, it is high connected with population. Beijing, capital of China with populations of more than 20 million and number of cars more than 5 million, is one of the largest cities in China. Changbai is a county, stand for the medium scale, with populations of 86,300 while Hunchun, the sampling site is located in a small town, with populations around 5000 and the number of cars is not much. The locations of sampling sites are showed in Fig. 1.

In each investigated site, aerosol samples were collected at the roof of building. These buildings are around 10–15 m high, usually is a fifth to sixth floor. The sampling site in Beijing is on the roof of sixth floor building, the sampling site in Changbai and Hunchun is on the roof sixth and fifth floor building, respectively. Height of the sampling locations was set similar when possible obtained. During this work, parallel samplings were carried out at the same location.

The meteorological conditions such as temperature, humidity and air pressure were recorded during each sampling procedure. The flow rates of about 1.5–2 m^3/min were also recorded at the beginning and end of the sampling, respectively, then the volume of the air samples were calculated according to recorded data and recommended methods [11]. Sampling usually started at 8:00 am and lasted nearly 24 h, in general the total volume of the air for each sample is 2000–3000 m^3. Two 3M type filters which collected the atmospheric aerosol particles were folded and placed in the mold of stainless steel, then compressed into a 50 mm-in-diameter and 4.5 mm-in-thickness disc by hydraulics, the samples were then transferred to cylindrical polyethylene beakers (dia. 75 mm × ht. 35 mm) and sealed until measurement.

Measurement of radionuclide using gamma spectrometry

A gamma spectrometry consisting of a broad-energy High Purity Germanium (HPGe) type detector named BE5030 (CANBERRA®) shielded in a lead chamber was used for measurement of Pb-210 in aerosol samples, the system has a relative efficiency of 50.5%, and a resolution of 1.88 keV for the 1332 keV Co-60 peak. The measured background in 20–2000 keV is 53 count/min. Software Genie 2000® was used for spectral acquisition and analysis. Activity concentration of Pb-210 in the aerosol was calculated by formula (1):

$$C = \frac{S}{R \times f \times V \times \xi \times \varepsilon} \quad (1)$$

where S means total net peak area counted by 46.5 keV γ-ray derived from Pb-210, expressed in count/s, R the correction coefficient for radioactive decay, f the emission probability of 46.5 keV γ rays derived from Pb-210, which is 4.25% [12], V means the total volume of each collected outdoor air sample, expressed in m^3, ξ the collection efficiency of air filter membrane, amount to 90%, ε means the gamma spectrometry detection efficiency of 46.5 keV γ-ray derived from Pb-210.

The efficiency for the HPGe gamma spectrometry had been previously calibrated by using a set of mixed nuclides standards filter (identification code: X13598) which is traceable to NPL (National Physical Laboratory, UK). Efficiency calibration of the spectrometer was carried out according to standards [11, 13, 14]. Self-attenuation factor of 46.5 keV γ-ray depending on the density and composition of sample has been established in the laboratory, and used for correct the counting efficiency of the samples. Each samples was normally measured for 24 h. Energy drift of the spectrometers was checked every day and found that no significant shift was observed. For quality control, an intercomparison of γ-spectrometry measurement and analysis was organized by the framework of the minutes on technical cooperation of National Institute for Radiological Protection, Chinese Center for Disease Control and Prevention (NIRP, China CDC), Radiation Monitoring Technical Center of Ministry of Environmental Protection (RMTC) and Japan Chemical Analysis Center (JCAC). Our

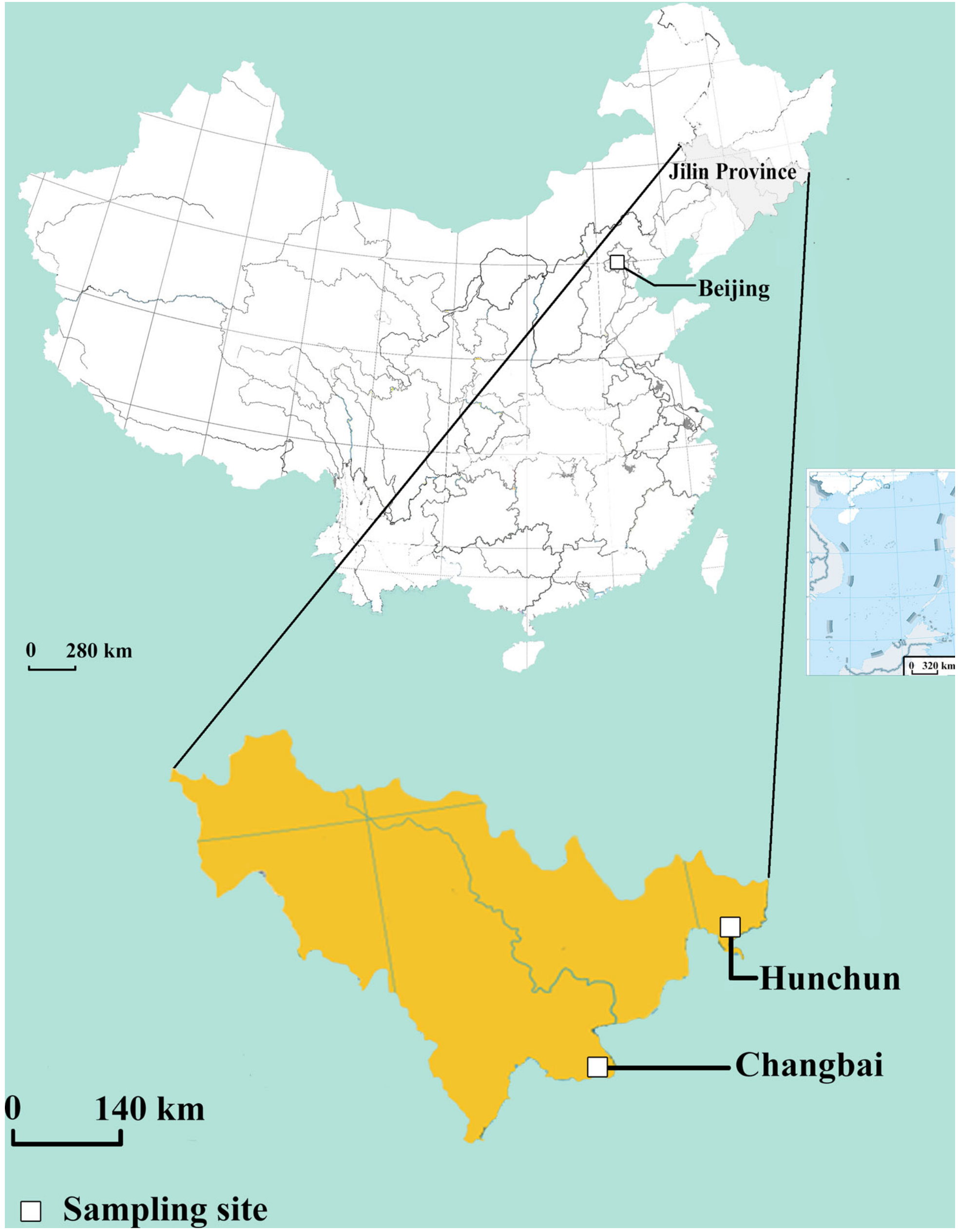

Fig. 1 Locations of the sampling site

laboratory periodically participates in the intercomparison exercises for many times [15, 16]. The results of participation in these intercomparison exercises generally resulted in *Z*-scores less than 2, a *Z*-score equal to or less than 2 indicates that the measurement is satisfactory.

During sampling days, the data of Air Quality Index (AQI), $PM_{2.5}$ and PM_{10} concentration were obtained from local metrological reports [17–19]. Statistical Analysis System (SAS) 9.3 was applied for data analysis, and the correlation was considered to be significant at *P* value less than 0.05.

Results and discussion

The samples from Changbai and Hunchun were collected only at January and September in the period of 2016–2017, which aims to estimate the seasonal and regional difference of the two sites. The samples from Beijing were collected in the period of March to May in 2015, when the haze was the heaviest during the whole year of Beijing.

A total of 96 outdoor air samples were collected. The reported expanded uncertainty of the result was based on a standard uncertainty multiplied by a coverage factor (k = 2), providing a coverage probability of approximately 95%. The uncertainty evaluation was carried out in accordance with the requirements of national standard [11, 13]. Pb-210 concentration changes, correlation between AQI, $PM_{2.5}$ and PM_{10} with Pb-210 concentration are illustrated in Figs. 2, 3 and 4 for Beijing, Changbai and Hunchun, respectively. The data of AQI, $PM_{2.5}$ and PM_{10} was obtained from the local metrological reports. Statistical results for sampling site of Beijing demonstrated the correlation coefficients between Pb-210 activity concentrations and AQI, Pb-210 activity concentrations and $PM_{2.5}$, Pb-210 activity concentrations and PM_{10} were 0.75, 0.80 and 0.64, respectively, and there were statistical significant ($P < 0.0001$). Seasonal concentration changes of Pb-210 from 2016 to 2017 with the trend of air quality index in Changbai has shown a correlation with the consumption of main fossil fuels (coal and oil) in winter (Fig. 3).

The average values and the sample information were summarized in Table 1. The average Pb-210 activity concentrations in Beijing, Changbai and Hunchun were 879, 3389 and 813 μBq/m^3, respectively, with the variation range of 97–27,955 μBq/m^3. The average values in winter were 20,403 and 1456 μBq/m^3 in Changbai and Hunchun, respectively, while in autumn it were much lower with values of 869 and 701 μBq/m^3 in Changbai and Hunchun, respectively. Pb-210 concentrations in Changbai was significantly higher than that in Hunchun in winter (January, $P < 0.01$), but the difference was insignificant in autumn between two sites (September, $P = 0.1498$). As we known, the basic source of Pb-210 in air is the decay of Rn-222 escaping from soil, the release of human activities, and the impact of environmental changes. The major difference in two sites is human activities, as there are more populations in Changbai (∼ 86,300 persons) than in town of Hunchun (∼ 5000 persons). So, difference in winter may be attributed to the more amount use of coal in Changbai for more larger scale of domestic heating in winter because the lowest tempeture is around − 40 °C in winter. Some researchers indicated that the radionuclides concentrations were enhanced by coal combustion [3, 20, 21]. In addition, the sampling sites in Changbai is located in the center of urban district, while the sampling site in Hunchun is located in Jingxin Town, which is approximately 35 km far away from the center urban district,. The air Pb-210 concentrations in winter was significantly higher than that in autumn in both Changbai group ($P < 0.01$) and Hunchun group ($P < 0.01$). The measured results in these work are comparable to those reported in Poland [1], Egypt [22] and Germany [23], while is not consistent with literatures reported by Talbi [24].

Series of simple linear correlation analysis were carried out between different sites. It demonstrated that the correlation coefficient between Pb-210 activity concentrations and AQI, Pb-210 activity concentrations and $PM_{2.5}$, Pb-210 activity concentrations and PM_{10} were 0.73, 0.76 and 0.70, respectively in Hunchun group, and there were statistical significant ($P < 0.01$). But there were strong correlations among AQI, $PM_{2.5}$ and PM_{10}, partial correlation analysis was carried out and the results presented that none of the three pairs had a significant correlation after excluding impact of the other two factors. The same statistical methods were used to analyze the data of Changbai group.

Statistical results of Beijing group demonstrated that the correlation coefficients between Pb-210 activity concentrations and AQI, Pb-210 activity concentrations and $PM_{2.5}$, Pb-210 activity concentrations and PM_{10} were 0.75, 0.80 and 0.64, respectively, and they were statistical

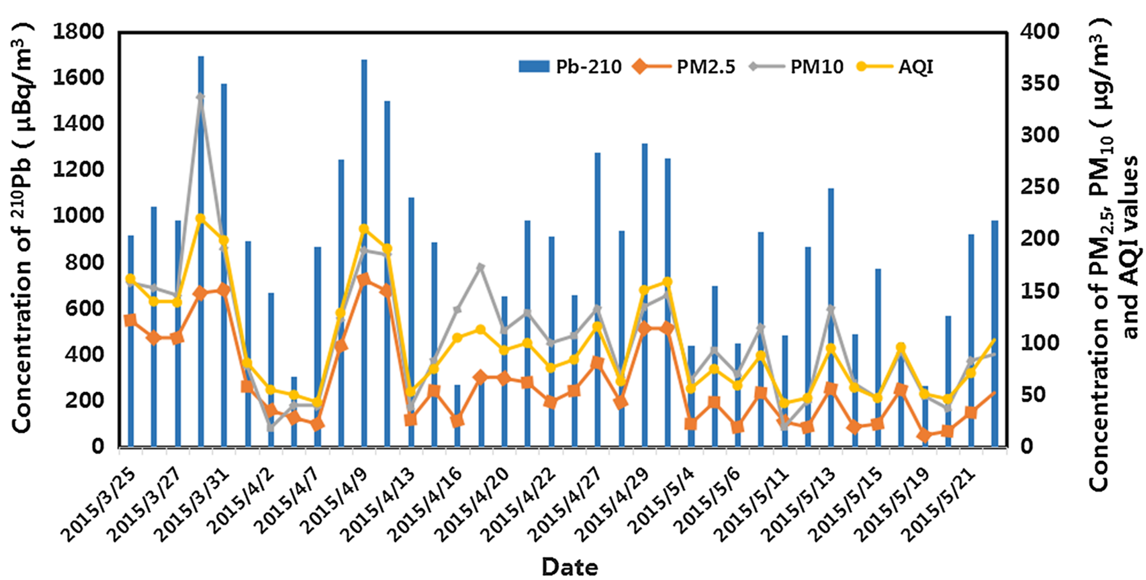

Fig. 2 Pb-210 concentrations in aerosol samples collected in Beijing and corresponding information of air quality index

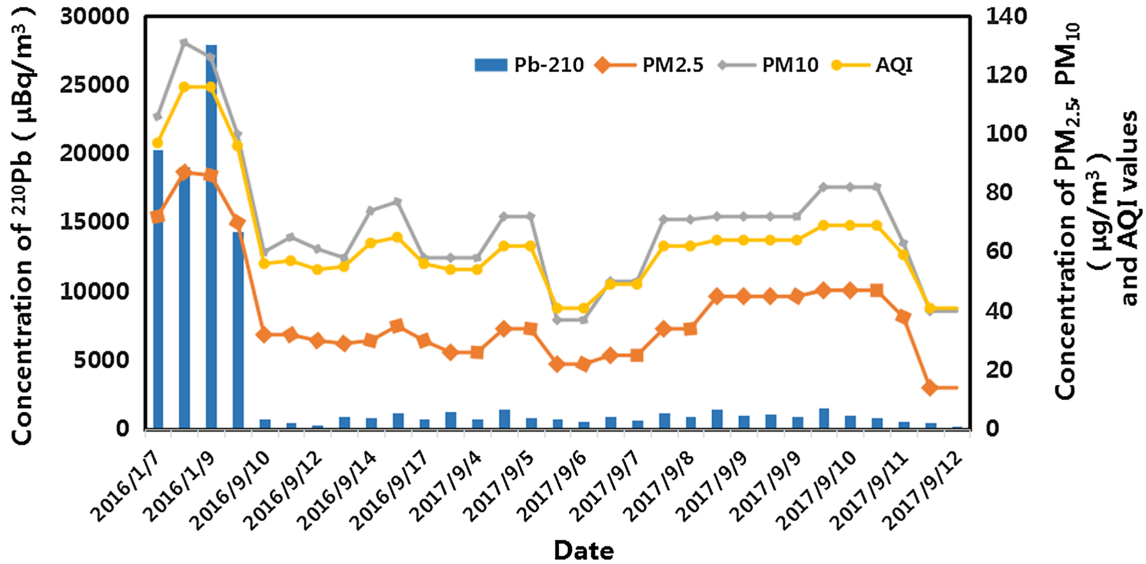

Fig. 3 Pb-210 concentrations in aerosol samples collected in Changbai and corresponding information of air quality index

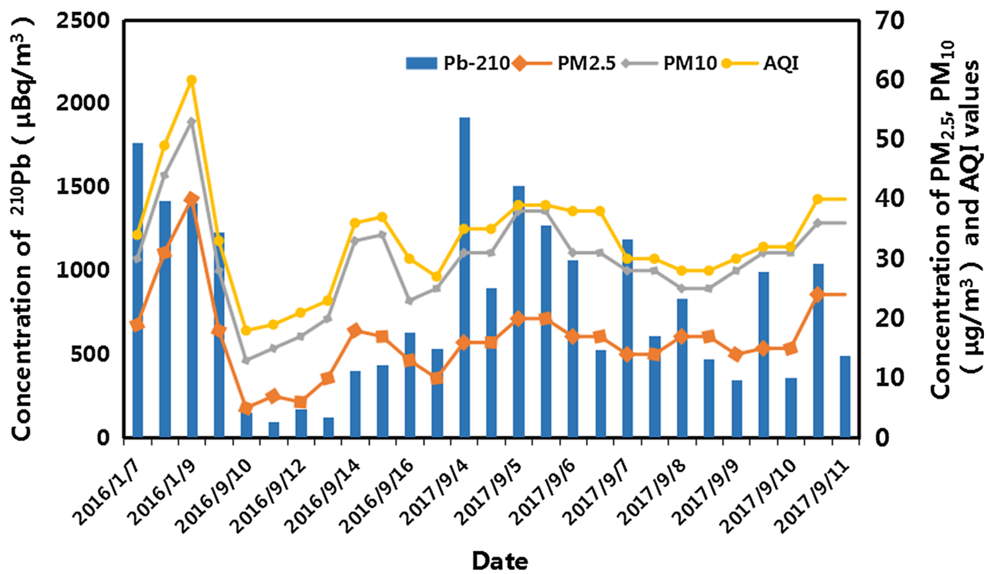

Fig. 4 Pb-210 concentrations in aerosol samples collected in Hunchun and corresponding information of air quality index

significant ($P < 0.0001$). However, partial correlation analysis result presented that only the correlation between Pb-210 activity concentrations and $PM_{2.5}$ was significant ($r^* = 0.43$, $P = 0.0088$) after excluding impact of the other two factors. It can be concluded that there was a positive correlation between Pb-210 activity concentrations and $PM_{2.5}$ ($P < 0.01$) for Beijing group. It has been reported that lead had been the highest concentration within the heavy metal detected in $PM_{2.5}$ and showed particle size dependence with lead metals concentrations, which were high in the particle size range of 1.1–4.7 μm [25, 26], which was partial consistent with this study.

The measured Pb-210 activity concentrations were compared with different studies as listed in Table 2. It showed that most of Pb-210 concentrations in different sites of China were higher than that in districts of other countries, especially in Changbai. Most of these values have exceeded the world average levels that reported by UNSCEAR (500 $\mu Bq/m^3$) [27], indicated there are sources of systemic and regional pollution risks. Therefore, the studies to trace, prevent, and control the source of major pollution must be strengthened.

To evaluate corresponding annual effective dose of Pb-210 due inhalation for inhabitants, the committed effective dose was calculated by formula (2):

$$E = A \times T \times b \times d \quad (2)$$

where A is the mean values of Pb-210 activity concentration in air of investigated sites, expressed in Bq/m^3, T the Pb-210 dose conversion coefficient, expressed in Sv/Bq,

Table 1 Summary of sample information in three sites

Sites	Date	*n*	Item	Pb-210 (μBq/m^3)	$PM_{2.5}$ (μg/m^3)[a]	PM_{10} (μg/m^3)[a]	AQI[a]
Beijing	2015-Mar. to May	38	Range	267–1698	11–161	18–337	42–220
			Median	904	53	97	86
			Mean	879	62	106	98
Changbai	2016-Jan	4	Range	14,310–27955	70–87	100–131	96–116
			Median	19,673	79	116	107
			Mean	20,403	79	116	106
	2016-Sep	7	Range	265–1187	29–35	58–77	54–65
			Median	750	30	61	56
			Mean	733	31	65	58
	2017-Sep	20	Range	237–1556	14–47	37–82	41–69
			Median	883	34	71	62
			Mean	917	33	63	57
	Total	31	Range	237–27,955	14–87	37–131	41–116
			Median	888	34	71	62
			Mean	3389	39	70	64
Hunchun	2016-Jan	4	Range	1229–1765	18–40	28–53	33–60
			Median	1415	25	37	41.5
			Mean	1456	27	38.75	44
	2016-Sep	8	Range	97–635	5–18	13–34	18–37
			Median	288	11	22	25
			Mean	321	10	23	26
	2017-Sep	15	Range	348–1932	14–24	25–38	28–40
			Median	895	17	31	35
			Mean	903	17	31	34
	Total	27	Range	97–1923	5–40	13–53	18–60
			Median	635	17	31	33
			Mean	813	17	30	33

[a]The data of AQI, $PM_{2.5}$ and PM_{10} was obtained from metrological report (http://www.tianqihoubao.com/)

dose conversion coefficient of Pb-210 with ages group of 1–2 years old, 2–7 years old, 7–12 years old, 12–17 years old and above 17 years old were 2.90E−06, 1.50E−06, 1.40E−06, 1.30E−06, 9.00E−07 Sv/Bq, respectively [40]. Where *b* means the respiratory rate, expressed in m^3/d [41], *d* the fraction of time spent outdoors by inhabitants of the considered area corresponds to 0.2, 365 × 0.2 = 73 days, expressed in days [41]. After considering different respiratory rate corresponding different ages group, the committed effective dose attributed to Pb-210 in all sample sites were estimated and presented in Table 3. The results shows that internal dose through inhalation of the above mentioned air was estimated to be 0.68–5.94 μSv. The maximum annual effective dose are nearly 5% of the effective doses from outdoor radon progeny in China, which is 126 μSv [42].

Conclusions

Level of Pb-210 in atmosphere in North China were higher than that a selection of other countries. Based on observation from year of 2015 to 2017, Pb-210 together with comprehensive investigation data of Air Quality Index (AQI), $PM_{2.5}$ and PM_{10} concentration were obtained to analysis the correlation and trend of those factors. The average activity concentrations of Pb-210 during the winter season in Changbai and Hunchun were higher than those in autumn. The seasonal variation was statistically significant (*t* test, 95% CI), which mainly results from the increment of coal combustion in winter. Pb-210 activity concentrations in Changbai were higher than that in Hunchun in winter. The statistical results of the activity concentration in Beijing demonstrated that there was a positive correlation between Pb-210 activity concentrations and $PM_{2.5}$ ($P < 0.01$). The developed method and obtained data can

Table 2 Comparison of Pb-210 activity concentrations in air

Country	Sample sides	Sampling time	Range (μBq/m^3)	Mean (μBq/m^3)	References
Estonia	Toraver	2001–2008	120–2760	570	[28]
	Narva-Joesuu	2001–2008	80–2530	530	[28]
	Harku-Tallinn	2001–2005	65–2020	366	[28]
Spain	Malaga	2009–2011	–	550	[29]
	Caceres	2004–2011	120–1820	600 ± 340	[30]
	Granada	2010–2014	159–1309	617	[31]
Italy	Mt. Cimone	1998–2011	50–2300	460	[32]
Pakistan	Islamabad	2007–2009	56–760	284 ± 150	[33]
Monaco	A museum roof near the Mediterranean	1998–2010	20–2820	1130 ± 440	[34]
Poland	Lodz	2008–2009	167–1847	556	[1]
Norway	Svalbard	2004	80–264	82 ± 59	[35]
England	Oxfordshire	1999–2002	–	201 ± 8	[36]
China	Qianzhong	2002–2003	–	2770 ± 630	[37]
	Hangzhou	2011–2012	220–2730	1510	[38]
	Qingdao	2015–2016	60–1610	700 ± 500	[39]
	Beijing	2015	267–1698	879	This work
	Changbai	2016–2017	237–27,955	3389	This work
	Hunchun	2016–2017	97–1923	813	This work

"–" means values were not given by corresponding references

Table 3 The effective dose attributed to Pb-210 due inhalation

Sampling sites	Annual effective dose (mSv)				
	1–2 years old	2–7 years old	7–12 years old	12–17 years old	> 17 years old
Beijing	1.03E−3	8.55E−4	1.29E−3	1.54E−3	1.15E−3
Changbai	3.96E−3	3.30E−3	4.99E−3	5.94E−3	4.44E−3
Hunchun	8.17E−4	6.80E−4	1.03E−3	1.23E−3	9.15E−4

be used as a baseline to evaluate air quality, corresponding effective dose from inhalation for inhabitants were evaluated, with maximum effective dose of 4.44 μSv for adult. Distribution analysis of the results could help to assess the effects of historical emission control measurement and provide experience and scientific support for the following air pollutant prevention strategy making in Beijing and other north areas of China.

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