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Distribution Characteristics of ^{7}Be and ^{210}Pb in Near-Surface Aerosols in Mianyang Area and Their Significance to O_3 Tracer

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Abstract: In order to understand the quality of radiation environment in the near surface air in Mianyang area, to grasp the trend of its change, to explore the sources of aerosol substances near the surface and their significance in tracking the concentration of O_3 near the ground, a detailed gamma spectrum analysis of aerosol samples from March 2018 to February 2019 in Anzhou District, Jiangyou City, Zitong County and Pingwu County was carried out. In general, the activity concentration of ^{7}Be is higher in spring and autumn, while it is the lowest in summer. The annual average is 1.90-2.13mBq/m^3, which is basically the same as the distribution characteristics of inland, mid-latitude, and low-altitude areas in the world. The activity concentration of ^{210}Pb is a "U" type distribution feature throughout the year. It is the lowest in late spring or early summer and has an annual average of 1.24-1.66mBq/m^3, which is a relatively high value of ^{210}Pb activity concentration on global land. The correlation analysis between ^{7}Be and ^{210}Pb and O_3 in near-surface aerosols showed that ^{7}Be and O_3 were weakly positively correlated, while the ratio of ^{7}Be/^{210}Pb was significantly positively correlated with O_3, and ^{210}Pb and O_3 were significantly negatively correlated; The ^{7}Be/^{210}Pb ratio can be used as a good tracer of O_3 sources in near-surface air. The O_3 source in the near surface air in Mianyang area is greatly affected by the vertical convection activity of the atmosphere.

Keywords: Aerosol, ^{7}Be, ^{210}Pb, Distribution Characteristics, O_3, Tracing

1. Introduction

Natural radionuclide ^{7}Be is a radionuclide produced by cosmic rays bombarding ^{14}N and ^{16}O target nuclei with a half-life of 53.3 days [1-3]. ^{7}Be is mainly produced and stored in the stratosphere. When ^{7}Be is formed, it is quickly adsorbed on the surface of sub-micron aerosol particles. It migrates with the atmospheric dynamic process, enters the surface environment through dry and wet deposition, and finally disappears through radioactive decay. The ^{7}Be produced and enriched in the stratosphere is transported to the troposphere through the Brewer-Dobson circulation on a global scale or exchanged with the troposphere through the stratosphere into the troposphere [4-6]. Natural radionuclide ^{210}Pb is a terrestrial radionuclide with a half-life of 22.3 years. Studies have shown that [7-9]: ^{210}Pb is formed by ^{222}Rn decay of ground (soil or rock) through 5-order decay. It eventually disappears through dry, wet settlement and decay.

In recent years, the Global Atmosphere Watch has proposed ^{222}Rn, ^{7}Be, and ^{210}Pb as one of the regular observations [10]. Since natural radionuclides are the most important source of radiation to humans [11, 12], in China, ^{7}Be and ^{210}Pb are the necessary nuclides for monitoring the environmental quality of atmospheric aerosol radiation, and it is important to observe their long-term changes. In addition, as ^{7}Be and ^{210}Pb have known sources and destinations, they are widely used in tracing environmental material sources [10, 13] and can be used as tracers for environmental material sources.

O_3 is one of the most important atmospheric pollutants in recent years. The main sources are the high tropospheric downward dynamic transport and the near surface photochemical transformation process. The current research is mainly to monitor its status, and few studies have been conducted on its source. Radiation environmental quality monitoring in China is unevenly distributed. In many cities in the East, there are reports on the observation of ^{7}Be and ^{210}Pb in aerosols [14-18], but there is a lack of reports on time and space continuity, and there are fewer applications in the detection of pollutant. Based on the analysis of gamma energy spectrum of near-surface aerosols in Mianyang area in the past year (March 2018 to February 2019), the dynamic trend of the concentration of ^{7}Be and ^{210}Pb in aerosols in Mianyang area in the past year was established, and the radiation environmental quality of near-surface air in Mianyang area was mastered. The application of ^{7}Be and ^{210}Pb as tracer in the source of O_3 near ground was initially explored.

2. Experiments and Data

From March 2018 to February 2019, in Anzhou District (31.53°N, 104.55°E, 511.16m above sea level), Jiangyou City (31.78°N, 104.73°E, 532.98m above sea level), Zitong County (31.64°N, 105.17°E, 491.72m above sea level) and Pingwu County (32.41°N, 104.55°E, 858.49m above sea level) carry out near-ground aerosol sampling and monitoring and analysis work. The four automatic monitoring stations for the radiation environment were built on the roof of the 5th floor of the office building. The air intake of the sampler was about 18 meters from the ground, avoiding interference from the secondary settlement of the ^{7}Be and ^{210}Pb nuclides collected on the ground. Sampling using polypropylene filter film, set sampling flow rate of $1.0m^3min^{-1}$, the total sampling volume is standard condition not less than $10000M^3$. After the sampling is completed, the sample filter film is folded and pressed into a cake shape in a mold with a diameter of 70mm or 50mm. The package is packaged in a special sample box or polyethylene self-sealing bag for testing.

The measuring instrument used in this experiment is the US ORTEC GEM35P4 low-background high-purity germanium gamma spectrometer. The resolution of the 1332keV omnipotent peak is 1.85keV, and the background measurement is less than 1.9CPS within 24 hours. The measuring instrument is verified by the China Institute of Metrology and Testing, and is equipped with aerosol sample standard substances and passive efficiency scale software. The maximum relative deviation of aerosol standard material sample radionuclide activity concentration is 15% through passive efficiency scale, and the measurement results are reliable. In order to reduce the uncertainty of the statistical count in the measurement, the measurement time of single sample and blank sample (base spectrum) in this experiment is 24 hours. The measurement results are corrected by decay to the end of the sampling time.

The near-ground O_3 concentration data comes from the official website of the Mianyang Ecological Environment Bureau (Http://sthjj.my.gov.cn/mygov/150650887172259840/index.html). The location of the four near-ground O_3 monitoring stations is exactly the same as the above four near-ground aerosol monitoring points.

3. Consequence and Discussions

3.1. Distribution Characteristics of ^{7}Be and ^{210}Pb in Near Surface Aerosols in Mianyang Area

The maximum annual variability of the activity concentration of ^{7}Be nuclides in aerosols at four monitoring points in Mianyang region ranges from 0.33-4.20mBq/m^3 (see table 1) and the annual mean variation range from 1.90-2.13mBq/m^3 (see table 1). It is basically the same as the annual average of 2.45mBq/m^3 in the world of ^{7}Be [19], which is far lower than the high altitude areas of Qinghai Waliguan Mountain and Guizhou Guanfeng Mountain [15], and also lower than Xi'an [16], Hangzhou [17], Shenzhen [18] and other places; The maximum annual range of activity concentration of ^{210}Pb nuclides in aerosols is 0.78-3.15mBq/m^3 (see table 1) and the annual mean range is 1.24-1.66mBq/m^3 (see table 1). For the relative high value of the ^{210}Pb value variation range of 0.5-1.5mBq/m^3 on the global land [20], it is basically equivalent to the annual mean of ^{210}Pb of 1.5mBq/m^3 in the Chengdu region of the Sichuan Basin [12], and the higher ^{210}Pb indicates that the input air mass comes from the land boundary layer gas mass. Because of the hysteresis effect of the water body on ^{222}Rn, ^{210}Pb in the maritime boundary methane is much lower than in land [21].

Table 1. *Activity concentration range and annual mean of ^{7}Be and ^{210}Pb in near-surface aerosols in Mianyang area.*

	^{7}Be Activity Concentration (mBq/m^3)		**^{210}Pb Activity Concentration (mBq/m^3)**	
	range	**Average per year**	**range**	**Average per year**
Anzhou District	0.33-4.20	1.90	0.86-3.15	1.66
Jiangyou City	0.61-3.32	2.13	0.98-2.51	1.46
Zitong County	0.88-3.54	2.12	0.81-2.54	1.40
Pingwu County	1.27-2.99	2.13	0.78-1.91	1.24

According to the meteorological methods in the "Climate Season Division", March to May is spring, June to August is summer, September to November is autumn, and December to February is winter. This article uses the season to refer to the above meteorological methods divided. *There was no significant seasonal change in the activity concentration of the ^{7}Be nuclide in the aerosols at the four monitoring points (see Figure 1)*, but there were significant high values in the spring and autumn seasons, reflecting the ^{7}Be "spring leakage" characteristics consistent with the global observation point report [22, 23]. The lowest values appeared in July and August in summer, indicating that the "wet removal" effect of the short half-life of ^{7}Be nuclides during the heavy rainfall season [24], The annual change of ^{7}Be in the near surface aerosols in Mianyang presents a consistent change pattern [25] with the mid-latitudes (30°N-40°N) of the world, reflecting the characteristics of inland, mid-latitude, and low-altitude areas [15]; The concentration of ^{210}Pb activity in aerosols near the ground at the four monitoring points generally showed "U" type variation (see Figure 1). The minimum values were issued in April or May in spring and the maximum in January in winter.

Looking from the whole year, the annual average activity concentration of ^{7}Be in Anzhou District is lower than that of the other three observation points. The annual average of ^{7}Be in Jiangyou City and Pingwu County are basically the same, while the annual average activity concentration of ^{210}Pb is weak regional differences.

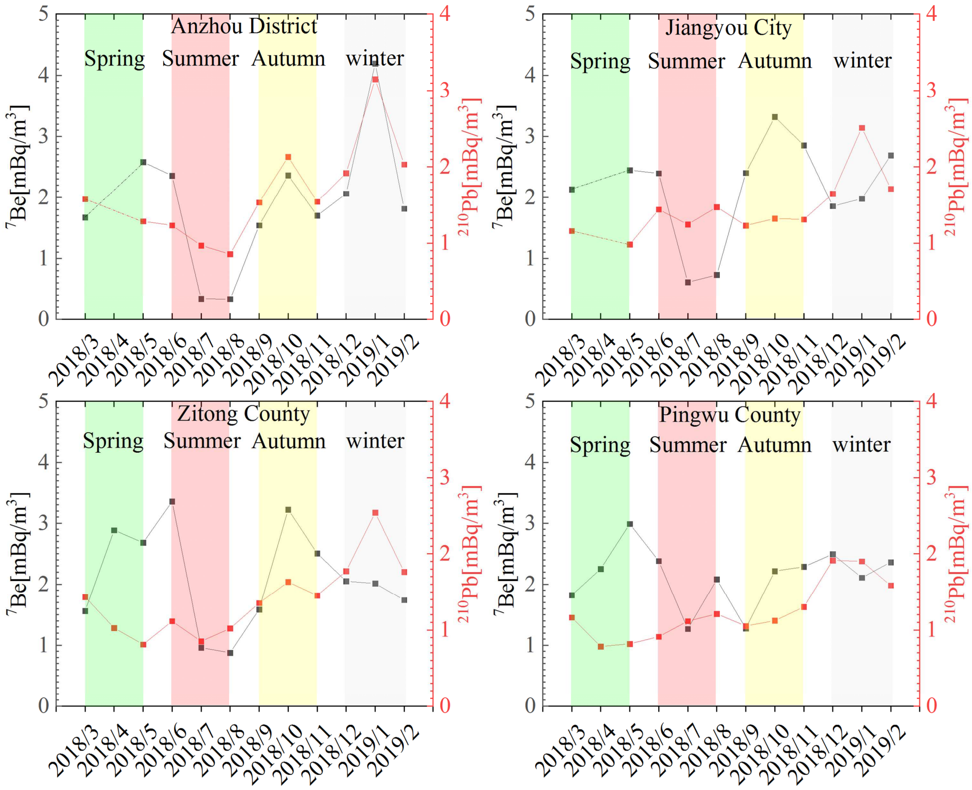

Figure 1. *Annual variation of ^{7}Be and ^{210}Pb activity concentration in near-surface aerosols in Mianyang area (The dotted line in the graph represents the missing intermediate data and the trend line connecting the data at both ends).*

3.2. The Significance of ^{7}Be and ^{210}Pb in Near Surface Aerosols in Mianyang Area on O_3

^{7}Be come from the stratosphere or the upper troposphere, and ^{210}Pb come from the ground. They are adsorbed on aerosols and then participate in the atmospheric circulation through the atmospheric convection process. The higher ^{7}Be and the lower ^{210}Pb represent strong vertical downward convection in the atmosphere, whereas the vertical downward convection in the atmosphere is weaker [2]; The lower ^{7}Be and the lower ^{210}Pb represent the weaker atmospheric convection activity, while the higher ^{7}Be and the higher ^{210}Pb represent the air mass affected by the high-latitude continental air mass (Siberian autumn and winter cold air) [20]. Since ^{7}Be and ^{210}Pb have determined sources and their own physical and chemical characteristics, the ^{7}Be/^{210}Pb ratio is more important in physics than ^{7}Be or ^{210}Pb. ZHENG X D et al believe that the joint tracer effect of ^{7}Be and ^{210}Pb can effectively represent the vertical transport process [21]; ZHU H L et al believe that the high ^{7}Be/^{210}Pb ratio represents the strong vertical convection process in the atmosphere, and the low ^{7}Be/^{210}Pb ratio represents the weak vertical convection process in the atmosphere [2]; The simulation results such as Koch also found that theoretically using the ^{7}Be/^{210}Pb ratio is more effective than a single nuclide to indicate the vertical transport process of aerosols. However, due to the complexity of ^{210}Pb motion in the atmosphere, ^{7}Be and ^{210}Pb values should also be combined analysis [19].

By analyzing the correlation between the concentration of ^{7}Be and ^{210}Pb in the near surface aerosols in Mianyang area and the O_3 concentration in the near surface, the results show that the relationship between ^{7}Be and O_3 is weakly *positive (see Figure 2)*. The $^{7}Be/^{210}Pb$ ratio and O_3 were significantly positively correlated (see Figure 2), *and ^{210}Pb and O_3 were significantly negatively correlated (see Figure 3)*. The $^{7}Be/^{210}Pb$ ratio is an indicator of the vertical convection intensity of the atmosphere, and ^{210}Pb is a typical terrestrial natural radionuclide produced on the Earth's surface, both of which are significantly positive (negative) related to O_3. The results show that O_3 source in the near surface air of Mianyang area is greatly affected by vertical convection.

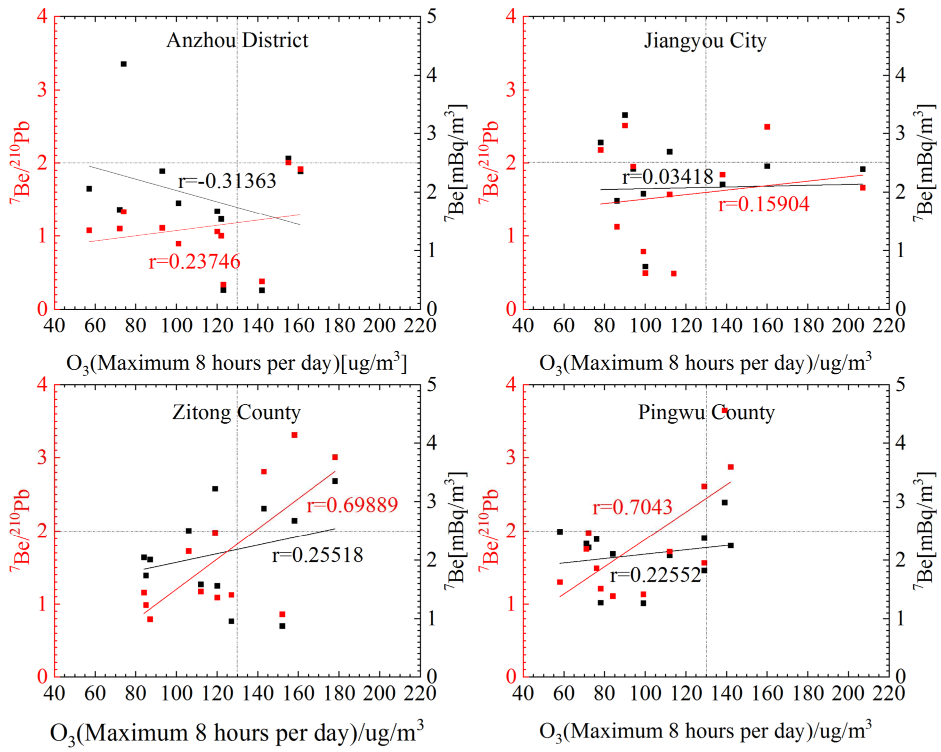

Figure 2. *Correlation between ^{7}Be and $^{7}Be/^{210}Pb$ ratios in near-surface aerosols and near-surface O_3 concentration in Mianyang area.*

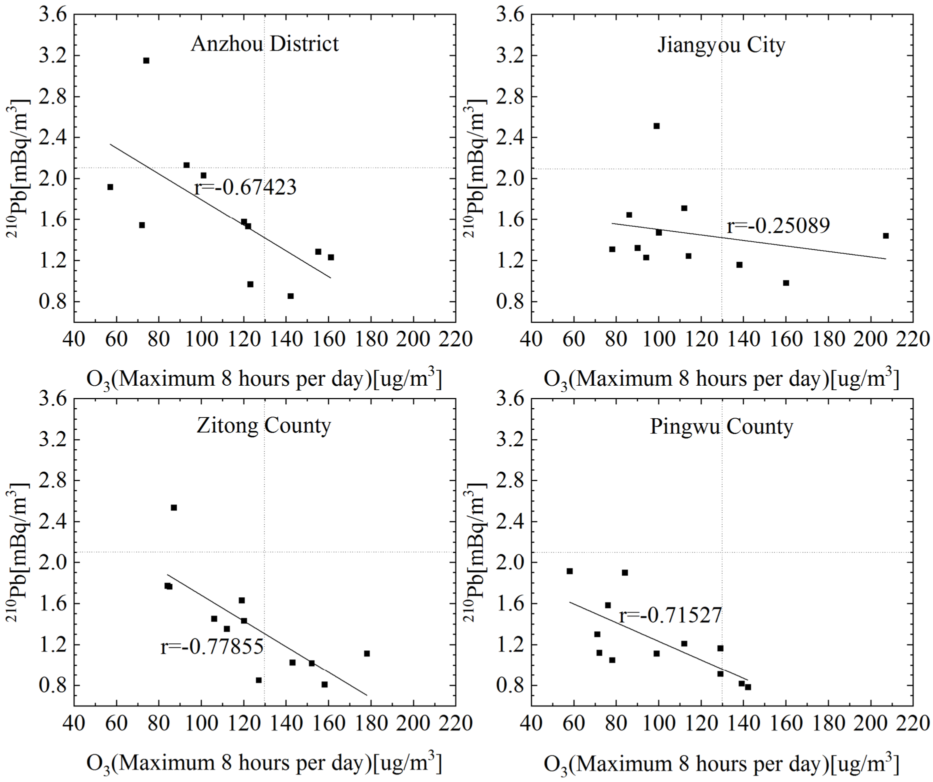

Figure 3. *Correlation between ^{210}Pb and O_3 Concentration in Near-surface Aerosol in Mianyang Area.*

In spring, the four monitoring points ^{7}Be and ^{210}Pb are reversed, showing higher ^{7}Be and lower ^{210}Pb, indicating that spring is a strong vertical downward transport process of the atmosphere, and aerosol material is mainly from the stratosphere or higher troposphere. *Come, In the same period, the ratio of O_3 concentration to $^{7}Be/^{210}Pb$ was a very consistent trend (see Figure 4)*, indicating that O_3 near the ground is mainly a contribution to the strong vertical downward transport process of the atmosphere; In summer, due to the wet cleaning effect of ^{7}Be and ^{210}Pb materials during the heavy rainfall season, and complicated factors such as the difference in the wet cleaning efficiency of the rainfall on the two nuclides, no study is made on the atmospheric summer convection process in this article; In antumn, in general, ^{7}Be and ^{210}Pb are positive changes, and the synchronization becomes larger, indicating that the atmospheric convection activity is strong, and the atmosphere is mainly transported vertically downward. However, the transport of aerosol materials from the stratosphere or the higher troposphere to the lower troposphere is decreasing compared to spring, while the concentration of O_3 near the ground is slowly decreasing during the same period, indicating that O_3 near the ground is mainly a contribution to photochemical processes; At the beginning of winter, the differences between the changes of the four monitoring points ^{7}Be and ^{210}Pb were relatively large. Jiangyou City, Zitong County and Pingwu County were lower ^{7}Be and higher ^{210}Pb, indicating that the vertical downward convection of the atmosphere was weak, and aerosol materials were mainly from the ground. Transmission to the higher troposphere, however, at the end of the winter, Jiangyou City, Zitong County and Pingwu County were higher ^{7}Be and lower ^{210}Pb, indicating that the vertical downward convection process of the atmosphere was increasing, and aerosol materials were mainly transmitted from the stratosphere or the higher troposphere to the lower troposphere. In winter, the O_3 concentration in the near ground is gradually increasing, indicating that the main source of O_3 in the near ground is changing from the photochemical process to the vertical downward transport process of the atmosphere.

Anzhou District Monitoring Point ^{7}Be and ^{210}Pb have a very consistent change relationship in the autumn and winter seasons (see Figure 1), especially in winter with a higher ^{7}Be and a higher ^{210}Pb value. Indicates that the monitoring point is greatly affected by high-latitude continental air mass (Siberian autumn and winter cold air). The aerosol material comes from high-latitude continental air mass. The horizontal convection process in the atmosphere is more obvious. Caution should be used as a tracer material. At this time, the values of ^{7}Be and ^{210}Pb should be used to make specific judgments.

In this paper, the distribution characteristics of ^{7}Be and ^{210}Pb in near-surface aerosols and their tracer significance to O_3 sources in near-surface air are discussed. Moreover, there is only one year of continuous monitoring data, no in-depth study on the tracking mechanism, and there is a lack of auxiliary analysis of meteorological parameters. These aspects should be strengthened in future studies.

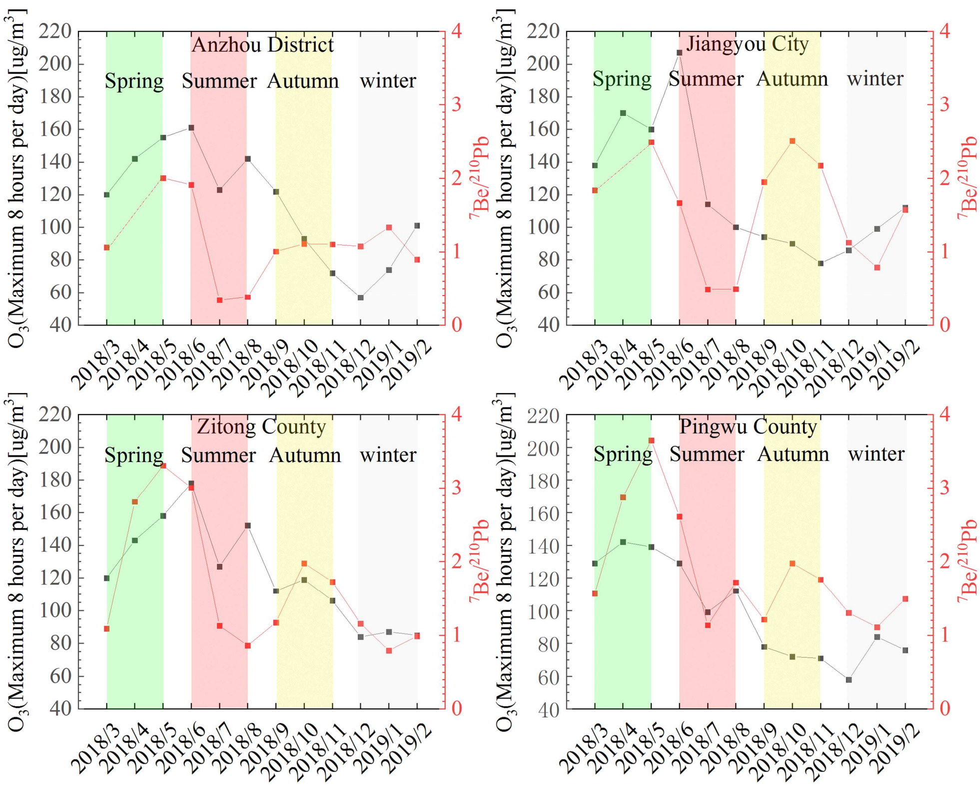

Figure 4. *Annual Variation of $^{7}Be/^{210}Pb$ Ratio and O_3 Concentration in Near-surface Aerosols in Mianyang Area (The dotted line in the graph represents the missing intermediate data and the trend line connecting the data at both ends).*

4. Conclusion

The annual mean of activity concentration of ^{7}Be nuclides in aerosols in Mianyang area is 1.90-2.13mBq/m^3, which is basically the same as the annual average of ^{7}Be in the world, and it is far lower than that of high altitude Mt. Waliguan in Qinhai and Mt. Guanfeng in Guizhou, and slightly lower than Xi'an, Hangzhou and Shenzhen. The annual mean activity concentration of ^{210}Pb nuclides in aerosols is 1.24-1.66mBq/m^3, which is a relatively high value of ^{210}Pb on land in the world, but it is basically equivalent to the annual mean of ^{210}Pb in Chengdu, Sichuan Basin.

There are no obvious seasonal changes in ^{7}Be in the near surface aerosols in Anzhou District, Jiangyou City, Zitong County and Pingwu County. The ^{7}Be in spring and autumn seasons are relatively high, while the in summer is the lowest, and it is showing the same annual distribution characteristics as inland, mid-latitude, and low-altitude areas; The overall concentration of ^{210}Pb activity in aerosols at the four monitoring points showed a "U" type distribution throughout the year, with the lowest in late spring or early summer.

The ratio of $^{7}Be/^{210}Pb$ in aerosols in Mianyang can be a good tracer of O_3 sources in near-surface air. O_3 sources in the near surface air of Mianyang area are greatly affected by vertical convection in the atmosphere.

In this paper, because the sampling time period is longer and the measurement index period is longer than that of ozone, there is a situation that the measurement index is smoothed and some information is covered. In the future, we can study a shorter time scale, such as daily sampling measurement, to explore the indicative significance of the tracer index in different seasons.

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Biography

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