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# Lead isotope trends and sources in the atmosphere at an artificial wetland

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With the rapid development of industry, studies on lead pollution in total suspended particulate matter (TSP) have received extensive attention. This paper analyzed the concentration and pollution sources of lead in the Cuihu Wetland in Beijing during the period of 2016–2017. The results show that the lead contents in TSP in the Cuihu Wetland were approximately equal in summer and spring, greater in winter, and greatest in autumn. The corresponding lead concentrations were 0.052 ng/m<sup>3</sup>, 0.053 ng/m<sup>3</sup>, 0.101 ng/m<sup>3</sup>, and 0.115 ng/m<sup>3</sup>, respectively. We compared the <sup>206</sup>Pb/<sup>207</sup>Pb data with other materials to further understand the potential sources of atmospheric lead. The mean values of <sup>206</sup>Pb/<sup>207</sup>Pb from spring to winter were 1.082, 1.098, 1.092, and 1.078, respectively. We found that the lead sources may be associated with coal burning, brake and tire wear, and vehicle exhaust emissions. We also calculated the enrichment factor values for the four seasons, and the values were all much greater than 10, indicating that the lead pollution is closely related to human activities.

**Lead isotope trends and sources in the atmosphere at the artificial wetland**

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## Abstract

With the rapid development of industry, studies on lead pollution in total suspended particulate matter (TSP) have received extensive attention. This paper analyzed the concentration and pollution sources of lead in the Cuihu Wetland in Beijing during the period of 2016–2017. The results show that the lead contents in TSP in the Cuihu Wetland were approximately equal in summer and spring, greater in winter, and greatest in autumn. The corresponding lead concentrations were 0.052 ng/m<sup>3</sup>, 0.053 ng/m<sup>3</sup>, 0.101 ng/m<sup>3</sup>, and 0.115 ng/m<sup>3</sup>, respectively. We compared the <sup>206</sup>Pb/<sup>207</sup>Pb data with other materials to further understand the potential sources of atmospheric lead. The mean values of <sup>206</sup>Pb/<sup>207</sup>Pb from spring to winter were 1.082, 1.098, 1.092, and 1.078, respectively. We found that the lead sources may be associated with coal burning, brake and tire wear, and vehicle exhaust emissions. We also calculated the enrichment factor values for the four seasons, and the values were all much greater than 10, indicating that the lead pollution is closely related to human activities.

**Keywords:** Total suspended particulate matter · Lead concentration · Lead isotope ratio · EF values · Artificial wetland

## 1 Introduction

Air pollution especially particulate matters' pollution has become an issue in the public eye in China (Florig 1997). Particulate matter pollution not only adversely affects human health, but also acts as a catalyst for climate change (Seaton et al. 1995; Kyotani, Iwatsuki 2002). Studies have shown a positive correlation between air pollution and respiratory system diseases like lung cancer (Dockery et al. 1993). Researchers found that atmospheric aerosols can affect cloud microphysics and indirectly cause changes in light radiation to affect climate (Charlson, Hofmann 1992; Dickerson et al. 1997). Aerosol particles is a mixture of liquid and

solid materials which contains trace metals, ions, and organic compounds and so on (Gang, Jiuhai 2015). Total suspended particulate matters (TSP) played an important role in analyzing aerosols' chemical constitution, studying the spatial and temporal variations, revealing the relationship with meteorological factors, and tracing sources (Cong et al. 2018; Ragosta et al. 2002). Atmospheric input of heavy metal elements has a long-term adverse impact on the geobiochemical cycle of ecosystems (Kelly et al. 1996). Therefore, it is imperative to understand the heavy metals in TSP.

Trace metals such as Pb, Cd, Hg, and Cr are biologically non-functional and are highly toxic (Salt et al. 1995). Lead has been designated as one of the most dangerous environmental pollutants by the United Nations Environment Programme (Morel 2008; Shi et al. 2008). With the rapid development of industry, anthropogenic Pb has become the major source of the lead in the environment. They were widespread in the atmosphere, soil, water, plants and animals (Wang et al. 2013). It is very important to study the geochemical cycle of lead in the environment (Hao et al. 2008; Dawson et al. 2010; Bove et al. 2011; Uzu et al. 2010).

Lead has four stable isotopes which can be used as a tracer of anthropogenic pollution.  $^{206}\text{Pb}$ ,  $^{207}\text{Pb}$  and  $^{208}\text{Pb}$  are three radiogenic isotopes while  $^{204}\text{Pb}$  is non-radiogenic isotope. These four isotopes can be used as a "footprint" for different sources of lead pollution in the environment, especially for the human activities (Grousset et al. 1994)(Komárek et al. 2008). The inductively coupled plasma-mass spectrometry (ICP-MS) was designed for analyzing stable isotopes more precisely, especially for Pb. The development of ICP-MS made it possible to trace the sources and investigation of heavy metals in different materials. It is widely used to identify the natural sources and anthropogenic pollution (Wiederhold 2015). The unique lead isotope ratio ranges make it easier to find out the major sources of lead, even though sometimes it may be overlapped

(Wang et al. 2013)(Bindler et al. 1999; Bollhöfer, Rosman 2001; Veyseyre et al. 2001; Kaste et al. 2003; Zhang et al. 2007). This makes scientists more convenient to identify and quantify the sources of lead in different environmental samples (e.g., atmospheric deposition (Gallon et al. 2005), sediment (Dang et al. 2015), and soil (Huang et al. 2015)), as well as in organisms (Martinez-Haro et al. 2011).

Cuihu wetland in Beijing is one of the most typical artificial wetland, which is constructed to improve the environmental conditions. It is reported that artificial wetland is a long term green technology to remove the heavy metals from the polluted areas (Huang et al. 2017), such as Pb. It can also theoretically influence the heavy metal air pollution by increasing humidity and decreasing temperature. However, there were few studies focused on atmospheric lead pollution in an artificial wetland in Beijing. On the other hand, it is difficult to making a systematic research about the lead pollution in the particles by only knowing their total concentrations. Thus, efforts must be made to identify the possible lead sources of the total suspended particulate matters, thus, to control and reduce the air pollution (Zheng et al. 2004). Therefore, we studied atmospheric lead concentrations and lead isotopic ratios in the Cuihu Wetland in Beijing. We analyzed the temporal variations of lead in TSP in the Cuihu Wetland and compared the differences of lead pollution in atmosphere over different regions and land use types. Another primary target of this study is to determine the sources of lead. We measured lead isotopic ratios in TSP and calculated the enrichment factor (EF) values over a year. Based upon the results, the study attempts to examine the effects of human activity on Pb in the atmosphere and the potential sources of Pb in the total suspended particulate matter in the region. It is helpful for us to have a systematic acknowledgement on the lead pollution in the TSP of the air in an artificial wetland.

## 2 Materials and methods

## 2.1 Sampling site

The Cuihu Wetland is a typical country wetland located north of the Shangzhuang Reservoir in the Haidian District of Beijing. The area of the Cuihu Wetland is 1.57 km<sup>2</sup>, of which approximately 0.09 km<sup>2</sup> is water with an approximate maximum length and width of 1.9 km and 1.2 km, respectively.

The weather is rainy and hot in summer (June–September) and dry and cold in winter (December–March). Spring (March–June) and autumn (September–December) are short. The sampling site was on Crane Island near the center of the Cuihu Wetland (Fig. 1). The island's main vegetation is willow (*Salix babylonica*), with reeds (*Phragmites communis*) growing on the more flat areas of the island.

## 2.2 Sampling process

An intelligent medium-flow total suspended particle sampler (TH-150, Wuhan Tianhong Instruments Co., Ltd) and Teflon filters (Beijing RyderCase Instruments Co., Ltd) were used to collect TSP. The sampling flow rate was fixed at 100 L min<sup>-1</sup>. The filters were put in an open plastic bag and conditioned in a constant temperature (25°C) and humidity (50%) chamber for 24 h before and after sampling (Marcazzan et al. 2001). The filters were transported to and from the sampling site in sealed plastic boxes.

Ambient TSP samples were collected at the sampling site on Crane Island from September 2016 to August 2017. Three samples were collected simultaneously at the site during each of the four seasons during the year (Gang, Jiuhai 2015). The duration per sample was 12 h (from 08:00 to 20:00).

## 2.3 Chemical analysis

The determinations of the lead concentration and the isotopic composition ( $^{206}\text{Pb}$ ,  $^{207}\text{Pb}$ , and  $^{208}\text{Pb}$ ) were performed via inductively coupled plasma-mass spectrometry (Bi et al. 2007; Dai 2015). A quarter of a filter sample was first placed in a Teflon digestion vessel. Then add 8 mL of nitric acid (6%, v/v) and 2 mL of hydrogen peroxide to the vessel. The vessel was covered and placed in a microwave digestive system to dissolve the sample (Gang, Jiu hai 2015). The sample digestion was performed according to Table 1. Then, the sample solution and filter residue mixture were transferred to a Teflon crucible to heat at 150°C until nearly dry; 5 mL of nitric acid (6%, v/v) was then added to the vessel for 15 min to dissolve the filter residue. After cooling, the solution was diluted with nitric acid (1%, v/v) and then used to determine the metal elements. Finally, the solution was measured using an ICP-MS to determine the lead concentration and the isotopic composition.

## 2.4 Statistical analysis

The statistical treatments of the data were performed using SigmaPlot 12.5 and the IBM SPSS Statistics 22 statistical software.

## 2.5 Enrichment factor analysis

We calculated the enrichment factors (EF) to identify the origin of the heavy elements and to calculate the proportions of the anthropogenic sources (Mai, Lee 2010; Yang et al. 2010). In previous studies, these measures have been effective tools to distinguish different sources of heavy metals such as natural sources and anthropogenic sources (Petaloti et al. 2006; Ayrault et al. 2010). The folume of EF is caculated via the following relationship:



$$EF = \left( \frac{[E]}{[R]} \right)_{\text{sample}} / \left( \frac{[E]}{[R]} \right)_{\text{crust}},$$

where E is the considered element, R represents the reference element for crustal material,  $([E]/[R])_{\text{sample}}$  is the concentration ratio of E to R in the aerosol sample, and  $([E]/[R])_{\text{crust}}$  indicates the mean concentration ratio of E to R in the crust (Yongming et al. 2006). Al is abundant in the earth's crust and is frequently used as a reference element (Yongming et al. 2006; Taylor, McLennan 1995; Duan et al. 2012). We calculated the EFs using the value of Al in Chinese soil in 1990, due to the stability and lack of anthropogenic sources. If EF approaches unity, the crustal soil is the predominant source of the element. Operationally, given the local variation in the soil composition, if  $EF > 10$ , it can be assumed that the anthropogenic pollution is the primary source of the elemental abundance (Basha et al. 2010).

### 3 Results and discussion

#### 3.1 Concentration of lead in atmosphere particles

Figure 2 presents the mean concentrations of lead ( $\pm$ SE) in the TSP during the observation period. The concentrations of TSP are also given in the figure. The concentrations of TSP were more than 1000 times greater than the Pb concentrations. The summer season has the lowest concentrations of TSP at 68.867 ng/m<sup>3</sup>. The highest concentrations are seen in winter at 244.213 ng/m<sup>3</sup>. The TSP concentration in spring is higher than that in autumn, with values of 171.528 ng/m<sup>3</sup> and 101.042 ng/m<sup>3</sup>, respectively. However, the seasonal trend is slightly different for TSP and lead. The average concentrations of lead in the four seasons vary from 0.055 ng/m<sup>3</sup> to 0.115 ng/m<sup>3</sup>. The lowest concentration of lead was recorded during summer and spring followed by winter, while the highest concentration was found during autumn at 0.115 ng/m<sup>3</sup>. The concentrations in summer and spring were 0.052 ng/m<sup>3</sup> and 0.053 ng/m<sup>3</sup>, respectively. The

concentration was approximately 0.101 ng/m<sup>3</sup> in winter. Even though the concentrations in autumn and winter are higher than those in spring and summer, the only significant difference is between autumn and summer ( $P < 0.05$ ). There were no significant differences between the other seasons.

The Cuihu Wetland is a typical country wetland in Beijing and is little affected by outside conditions in comparison with some industrial sites, which are influenced by heavy metals related manufacturing processes. The average lead concentrations in cuihu wetland were quiet low which were even below the safe limits of the international agencies. The WHO and USEPA standard for atmospheric lead is 0.500 ng/m<sup>3</sup> (Health 2000). During the present study, the average concentration of lead (0.080 ng/m<sup>3</sup>) was found to be under the limits of the WHO and USEPA standard. The reason for the lower concentration of lead in the atmospheric particulate matter in Cuihu may be due to the self-purification of the wetlands and its distance from pollution sources. In addition, the difference between the lead concentrations in the local atmosphere and the WHO level may be due to the different situations of the climate especially the metrological data during the research. We can refine the experimental data by performing additional repetitions and increasing the number of samples.

Variations in average lead levels showed the following sort during the study period: levels in summer were approximately equal to levels in spring, levels in winter were greater, and levels in autumn were the greatest, which is slightly different from a study in Islamabad during the period of 2004–2005, where the levels in summer were approximately equal to the levels in spring, levels in autumn were greater, and levels in winter were greatest (Shah,Shaheen 2008). The results show that the metal content is inversely to temperature.. Even though the concentration of lead in autumn is higher than that in winter, the difference between them is not significant (Kim et al.

1997; Kim et al. 2002; Mishra et al. 2004). Studies have found positive relationships of lead with relative humidity and negative relationships of lead with the temperature (Jonsson et al. 2004; Kim et al. 1997). Other studies show that the wind speed appreciably affects the spread of trace metals. it is shown that the wind speed affects the dilution of lead in the environment (Ki et al. 2002; Vallius et al. 2005; Fang et al. 2002; Ragosta et al. 2002). Furthermore, studies show that the rainfall scavenging is of great efficiency in removing heavy metals from the atmosphere (Mircea et al. 2000).

Data for lead concentrations in the Cuihu Wetland and other sites are listed in Table 2. We selected nine different types of sampling sites. The lead concentration in the Cuihu Wetland is approximately 4–8 times higher than those in wetlands in Taiwan, with values of 0.010 ng/m<sup>3</sup> and 0.025 ng/m<sup>3</sup>, respectively (Guor-ChengFang et al. 2010; Fang,Chang 2012). The annual concentration of lead in the Cuihu Wetland is similar to that in Haeng Goo Dong, Korea, which was sampled in a grassland (Kim 2004). Another study of lead in TSP in Beijing had a concentration of 0.690 ng/m<sup>3</sup>, which exceeds the limit of the WHO and USEPA standard. This may because the sample site is within a residential area (Okuda et al. 2008). However, efforts must be made to lower the lead concentration. The lead concentrations in forests were very low, followed by grasslands (Wang et al. 2016; Kim 2004; Quiterio et al. 2006). Lead concentrations appeared a little high in industrial areas (Ki et al. 2002; Shaheen et al. 2005; Shah,Shaheen 2007). However, this also depends on the meteorological parameters when the samples were collected and levels are very different in different cities.

### 3.2 Sources of atmospheric lead

The lead isotope compositions in the four seasons are shown in Table 3. In general, the samples show a wide range of lead isotope ratios, ranging from 36.145 to 37.949 for <sup>208</sup>Pb/<sup>204</sup>Pb, from

2.094 to 2.206 for  $^{208}\text{Pb}/^{206}\text{Pb}$ , from 15.129 to 15.773 for  $^{207}\text{Pb}/^{204}\text{Pb}$ , from 16.490 to 18.121 for  $^{206}\text{Pb}/^{204}\text{Pb}$ , and from 1.061 to 1.168 for  $^{206}\text{Pb}/^{207}\text{Pb}$  (Table 3).  $^{206}\text{Pb}/^{207}\text{Pb}$  is relatively important in studying the sources of lead in the environment, as it can be determined precisely. The  $^{206}\text{Pb}/^{207}\text{Pb}$  isotope ratio revealed differences in the behavior in different seasons at the sampling site.

Regardless of the lead sources (lithogenic or anthropogenic), the average  $^{206}\text{Pb}/^{207}\text{Pb}$  ratio in the four seasons followed the order: summer (1.098) > autumn (1.092) > spring (1.082) > winter (1.078).

The lead isotope compositions of the TSP are helpful to further understand the potential sources of atmospheric lead. We compared the  $^{206}\text{Pb}/^{207}\text{Pb}$  and  $^{208}\text{Pb}/^{206}\text{Pb}$  data with that of other materials (Table 4). Due to the Th-rich environment in China, relatively high  $^{208}\text{Pb}$  abundances may interfere with estimations of contributions from alkyl lead additives (Chen et al. 2005). Therefore, in the following discussion, we give priority to the influence of  $^{206}\text{Pb}/^{207}\text{Pb}$ . The results show that the average ratios of TSP in spring are in the range of 1.063–1.098, which is closest to leaded vehicle exhaust (Mukai et al. 1993). In addition, the  $^{206}\text{Pb}/^{207}\text{Pb}$  isotope ratios in autumn are 1.061–1.132, which are similar to those in spring. The  $^{208}\text{Pb}/^{206}\text{Pb}$  analysis results are consistent with those of  $^{206}\text{Pb}/^{207}\text{Pb}$ . The Cuihu Wetland is very close to a road, which is a training route for a driving school that has more students in spring and autumn. This indicates that traffic plays an important role in lead emissions. However, unleaded gasoline has been widely promoted in China, which makes high concentrations of lead controversial. Such concentrations may be due to the high lead emissions that entered the atmosphere over the past decades, resulting in a relatively high concentration of lead in the soil along the roadside. The movement of vehicles can act to re-suspend dust containing lead into the air (Shah et al. 2006;

Ragosta et al. 2002; Kim et al. 2002; Shah,Shaheen 2008). It is thought that the sand mining plant near the Cuihu Wetland also plays an important role in increasing the lead concentration. Lead isotope ratios of Chinese coal is reported varied widely (Mukai et al. 2001). It is interesting that the lead contents in coal are quiet low, while it is quiet high in the coal combustion dust samples. This may because the combustion process has a "concentration effect" on the emission of lead into the atmosphere (Chen et al. 2005). The  $^{206}\text{Pb}/^{207}\text{Pb}$  isotope ratios in summer ranged from 1.069 to 1.168, which reflects several factors, such as leaded vehicle exhaust, unleaded vehicle exhaust, and coal. The  $^{208}\text{Pb}/^{206}\text{Pb}$  ratios indicate that the major source of lead is coal. In winter, the isotope ratios of  $^{206}\text{Pb}/^{207}\text{Pb}$  and  $^{208}\text{Pb}/^{206}\text{Pb}$  were 1.069–1.200 and 2.160–2.202, respectively. The  $^{206}\text{Pb}/^{207}\text{Pb}$  values indicate the contribution of three sources: coal, metallurgic dust, and industrial sources. Meanwhile, the  $^{208}\text{Pb}/^{206}\text{Pb}$  values are very close to those of coal. Increased coal burning in winter is therefore the main source of lead. Trace elements were released into the atmosphere throughout coal combustion via bottom ash, fly ash and gaseous phase. The release of heavy metals depends on the composition of the coal and also on gas temperature and residence time in the flue gas (Mariepierre Pavageau et al. 2002). Studies have also show that more than 50% of lead in coal may be released into the atmosphere during normal coal pyrolysis processes (Zajusz-Zubek,Koniecznyński 2003). This reminds us that it is of great significant to control the combustion and emission process in ways of reducing the lead pollution in the air.

### 3.3 Enrichment factors

Figure 3 shows the EFs of lead for TSP in the four seasons using Al as the reference element. The EFs represent the enrichment or depletion of lead of the samples. If an element's EF value is less than 10, it can be considered to be a crustal (or topsoil) source

that is primary caused by soil- or rock-weathered dust blowing into the atmosphere. If the EF value is much greater than 10, e.g., tens to tens of thousands, the element is likely enriched and reflects not just the contribution of crustal material but may also be related to contributions from different human activities.

The EF values in TSP for each season varied substantially from 213.654 (summer) to 9623.153 (autumn). The average EF value of lead is 805.160 in spring, 557.390 in summer, 5132.950 in autumn, and 3008.090 in winter. It is obvious that the lead in the atmospheric particles came from anthropogenic sources. The highest EF value is found in autumn, which also had the highest lead concentration. It can be seen that the EF variation is similar to the trend in the Pb concentrations, which is autumn > winter > spring > summer. These findings indicate that the variation in lead is closely related to human activities. The lead sources are associated with coal burning, brake and tire wear, vehicle exhaust emissions, and the metal industry (Hieu, Lee 2010; Xu et al. 2013). One possible reason for the high EF values is that the Cuihu Wetland is fairly close to a main road, which is a training route for a driving school. This may increase the opportunity for pollution via brake and tire wear and vehicle exhaust emissions. However, coal burning in autumn and winter also leads to an increase in lead from anthropogenic sources. Other studies have also shown a similar lead enrichment in other places of China (Pan et al. 2015). One study surveyed the EFs in TSP measured at five sites from 2009 to 2010. The results showed that lead was highly enriched in TSP samples in Beijing, Tianjin, Baoding, Tangshan, and Xinglong, with EFs exceeding 100. These high EFs indicate that the lead is of anthropogenic origin, is a key tracer of coal burning (Degen 1963), and is rich in particles emitted from fossil fuels and

biofuel burning (Christian et al. 2009; Wang et al. 2008).

EFs have been widely used to evaluate the anthropogenic/natural contributions of trace elements (Duce et al. 1975; Polidori et al. 2009). However, the size distribution of the particulate matters or soil samples was another important factors that affects the enrichment of lead (Farao et al. 2014; Li et al. 2013). Therefore, more efforts must be done to figure out the effect of the size distribution to the sources of lead.

## 4 Conclusions

This study showed that the lead concentrations in TSP vary from 0.055 ng/m<sup>3</sup> to 0.115 ng/m<sup>3</sup> during a year. The average lead concentrations exhibited the following pattern during the study period: the level in summer was approximately equal to that in spring, levels in winter were greater, and levels in autumn were greatest. The lead isotope ratio proved to be a useful tool to characterize the source of the atmospheric lead contamination. Regardless of the lead source, the average <sup>206</sup>Pb/<sup>207</sup>Pb ratio in the four seasons followed the order: summer (1.098) > autumn (1.092) > spring (1.082) > winter (1.078). We also calculated the EF values in TSP for each season. These findings indicate that the variation in lead is closely related to human activities. The sources of lead may be associated with coal burning, brake and tire wear, vehicle exhaust emissions, and the metal industry. We found several possible ways that human activities affect the lead in the environment. However, further effort is needed to decrease and remove such pollution.

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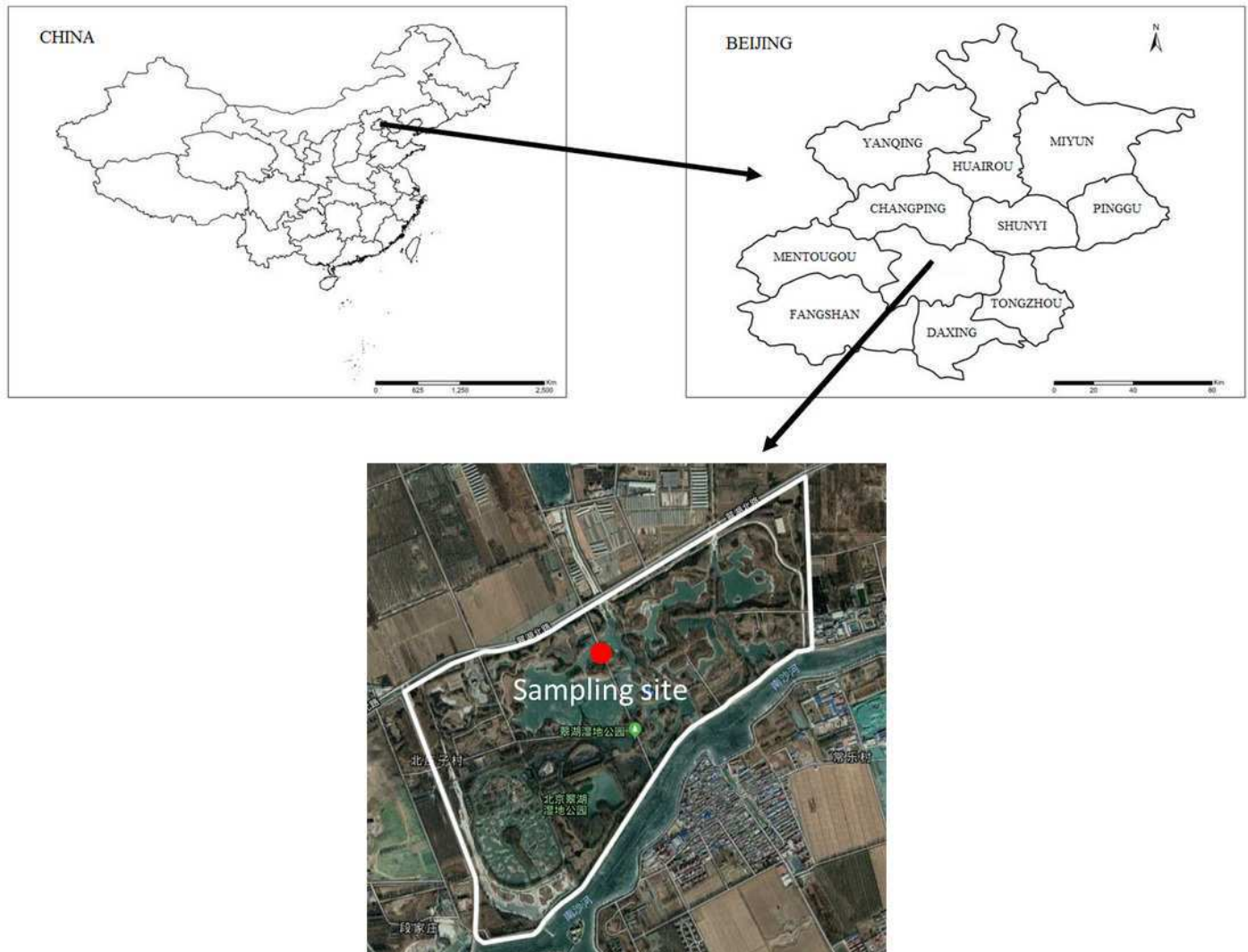
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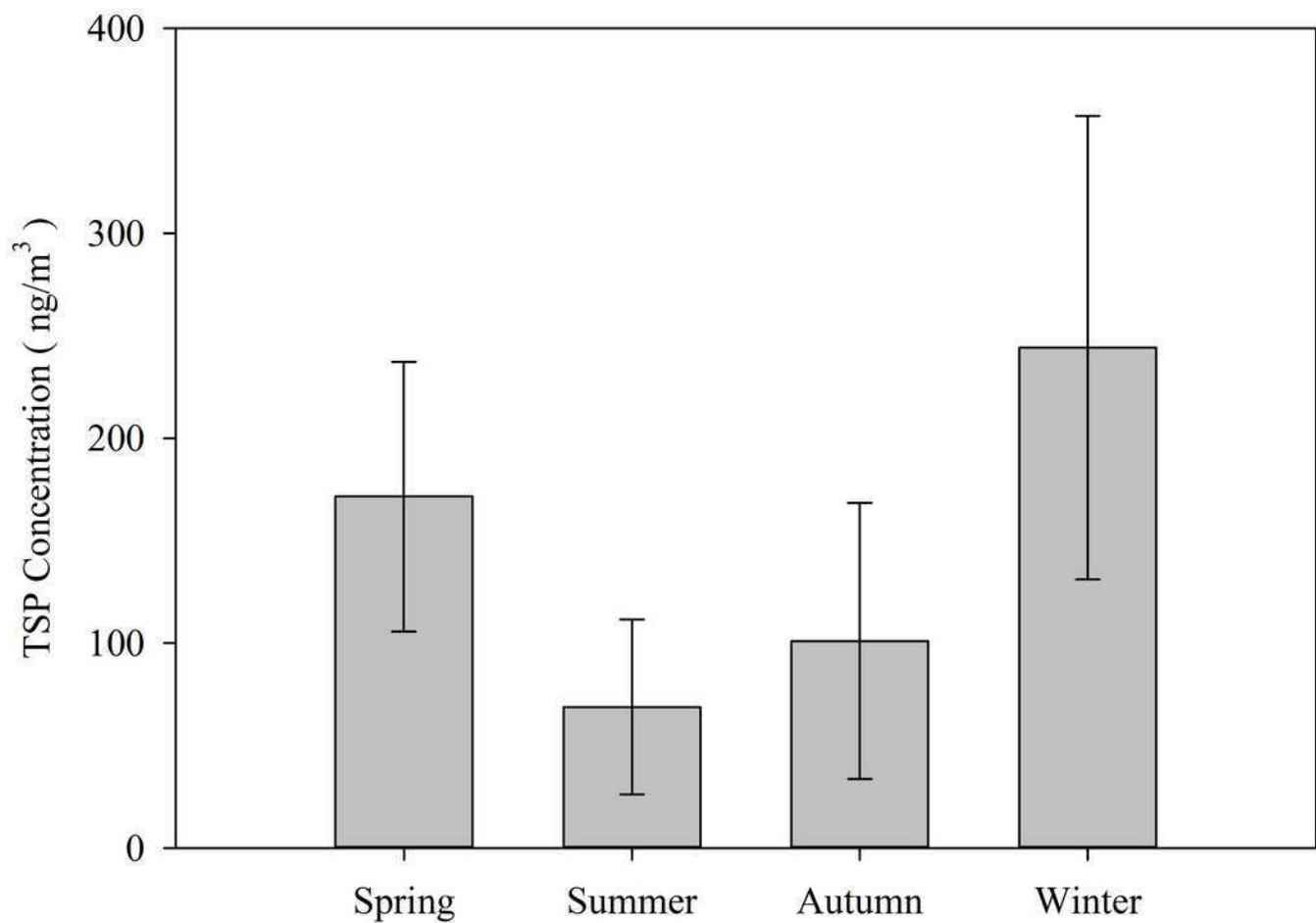
# Figure 1

Position of the sampling site



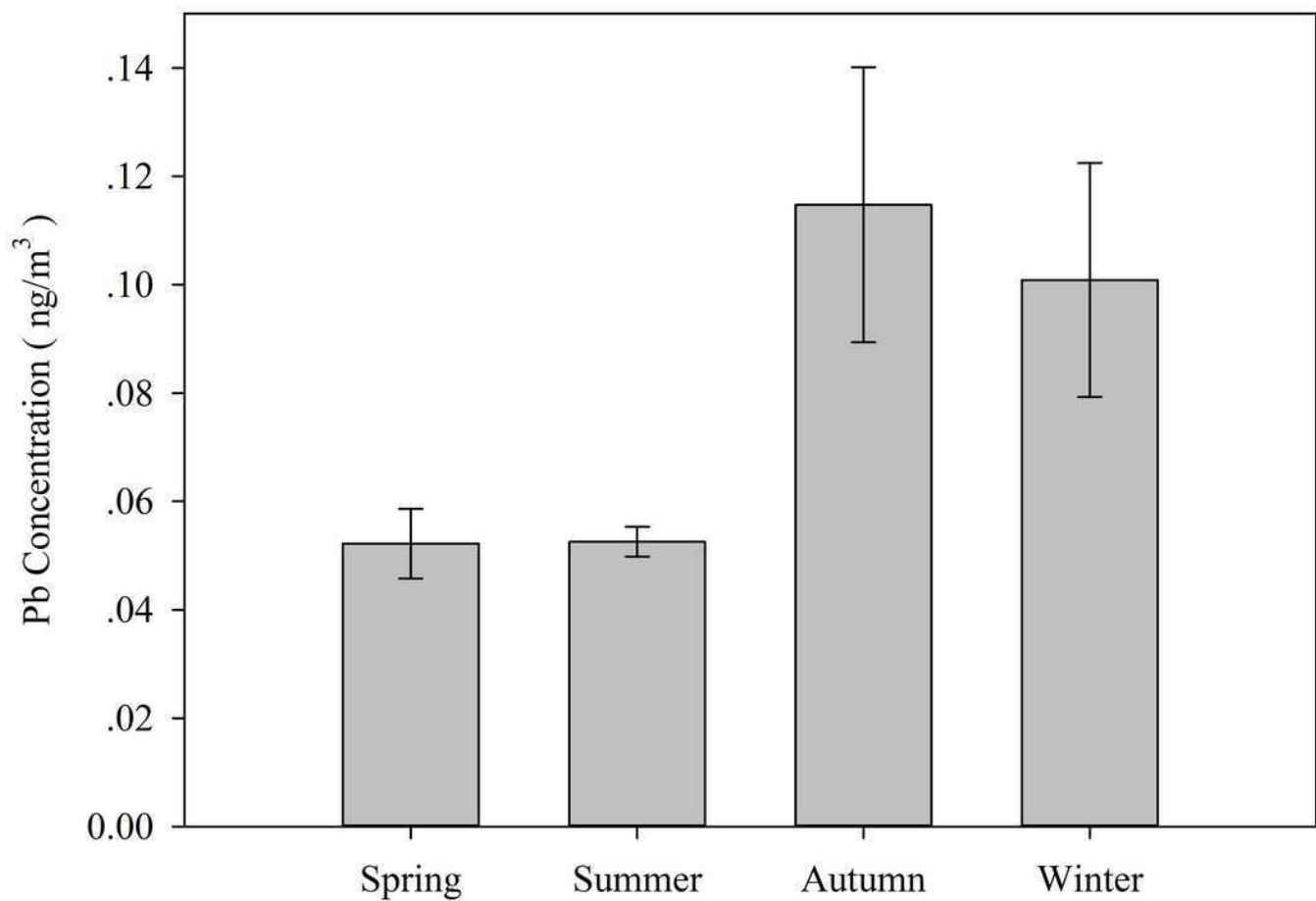
## Figure 2

Seasonal variations in TSP ( $\pm$ SE) expressed in  $\text{ng}/\text{m}^3$  during the study period



## Figure 3

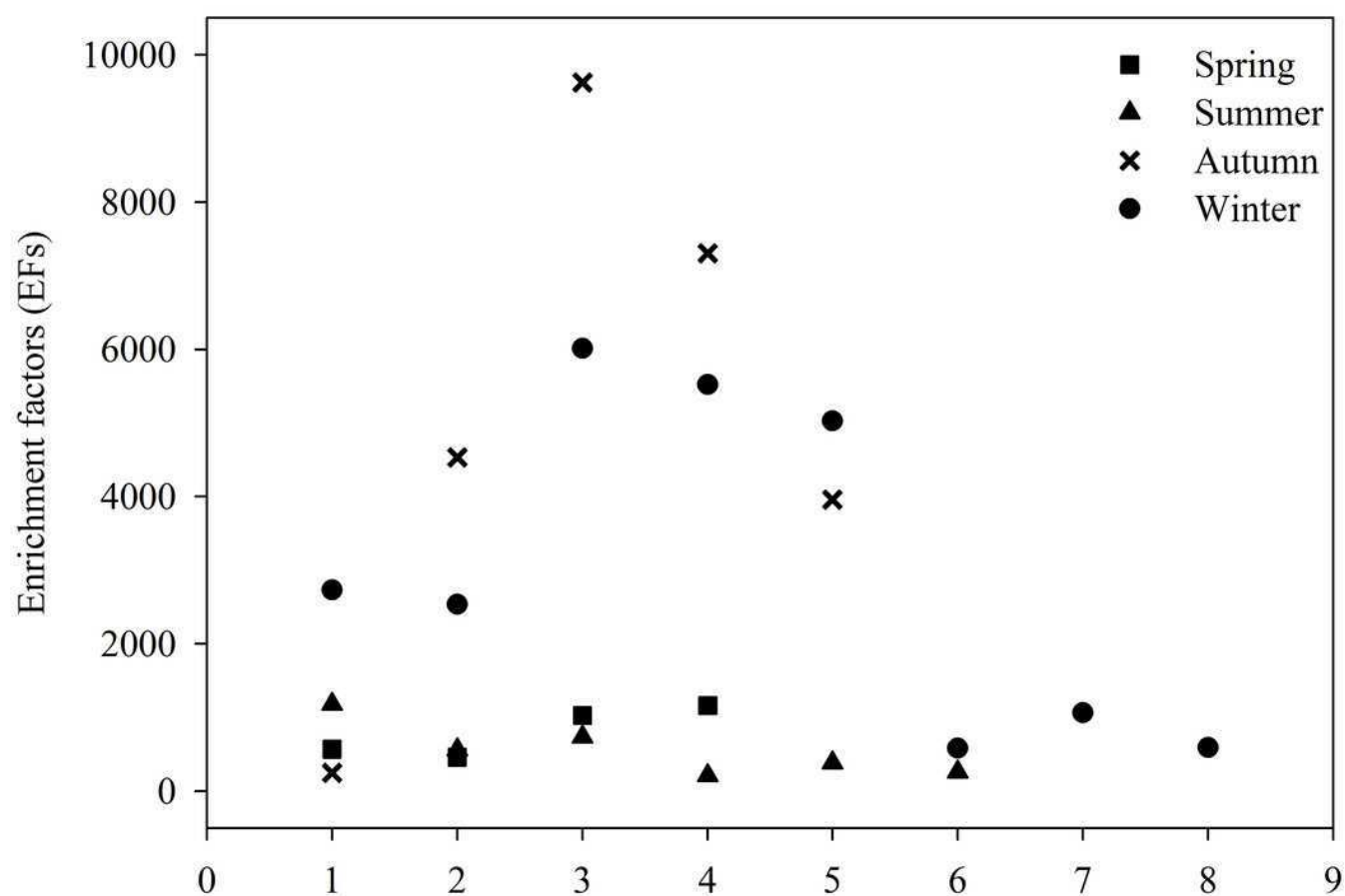
Seasonal variations in the lead concentrations ( $\pm$ SE) expressed in  $\text{ng}/\text{m}^3$  during the study period





# Figure 4

Lead enrichment factors during the study period



# **Table 1**(on next page)

The microwave digestion procedure

Step	Power (W)	Heating time (min)	Temperature (°C)	Processing time (min)
1	1200	10	150	10
2	1200	5	210	20

1

## Table 2 (on next page)

Lead concentrations in TSP in the Cuihu Wetland and other sites worldwide

City	Size	Pb (ng/m <sup>3</sup> )	Season	Character	Reference
Shenyang, China	TSP	0.115	2013–2014	Farmland	(Wang et al. 2016)
Hailun, China	TSP	0.037	2013–2014	Farmland	(Wang et al. 2016)
Taichung, Taiwan	TSP	0.574	2002	Farmland	(Fang et al. 2003)
Tongyu, China	TSP	0.031	2013–2014	Grassland	(Wang et al. 2016)
Haeng Goo Dong, Korea	TSP	0.084	1991–1995	Grassland	(Kim 2004)
Taejon, Korea	TSP	0.260	2002	Industrial	(Ki et al. 2002)
Islamabad	TSP	0.214	2003	Industrial	(Shaheen et al. 2005)
Islamabad	TSP	0.128	2004	Industrial	(Shah,Shaheen 2007)
Quan-xing, Taiwan	TSP	0.015	2010	Industrial	(Guor-ChengFang et al. 2010)
Chang-hua, Taiwan	TSP	0.019	2010	Downtown	(Guor-ChengFang et al. 2010)
Taiwan	TSP	0.180	2004	Downtown	(Fang et al. 2002)
Beijing, China	TSP	0.690	2005	Residential	(Okuda et al. 2008)
He-mei, Taiwan	TSP	0.016	2010	Residential	(Guor-ChengFang et al. 2010)
Islamabad, Pakistan	TSP	0.144	2004–2005	Urban area	(Wang et al. 2016)
Chang-Hua, Taiwan	TSP	0.034	2009–2010	Urban area	(Fang,Chang 2012)
Changbai Mountain, China	TSP	0.018	2013–2014	Forest	(Wang et al. 2016)
Ilha Grande, Brazil	TSP	0.001	2005	Forest	(Quiterio et al. 2006)
Bei-shi, Taiwan	TSP	0.044	2010	Suburban/Coastal	(Guor-ChengFang et al. 2010)
Gao-Mei, Taiwan	TSP	0.025	2009–2010	Wetland	(Fang,Chang 2012)
Gao-mei, Taiwan	TSP	0.010	2010	Wetland	(Guor-ChengFang et al. 2010)
Beijing, China	TSP	0.080	2016–2017	Wetland	This study

# **Table 3**(on next page)

Lead isotope compositions

		Spring	Summer	Autumn	Winter
$^{208}\text{Pb}/^{204}\text{Pb}$	Mean	36.773	37.033	36.795	36.596
	Range	36.710–36.888	36.471–37.949	36.145–37.559	36.233–37.166
$^{208}\text{Pb}/^{206}\text{Pb}$	Mean	2.184	2.162	2.171	2.189
	Range	2.165–2.204	2.094–2.197	2.112–2.206	2.160–2.202
$^{207}\text{Pb}/^{204}\text{Pb}$	Mean	15.568	15.600	15.529	15.517
	Range	15.443–15.688	15.429–15.761	15.129–15.732	15.350–15.773
$^{206}\text{Pb}/^{204}\text{Pb}$	Mean	16.838	17.134	16.957	16.720
	Range	16.678–16.971	16.758–18.121	16.490–17.787	16.505–17.034
$^{206}\text{Pb}/^{207}\text{Pb}$	Mean	1.082	1.098	1.092	1.078
	Range	1.063–1.098	1.069–1.168	1.061–1.132	1.069–1.100

# **Table 4**(on next page)

Isotope ratios and the elemental content of possible additional lead sources



Materials	$^{206}\text{Pb}/^{207}\text{Pb}$	$^{208}\text{Pb}/^{206}\text{Pb}$	Reference
Leaded vehicle exhaust	1.11	2.194	(Mukai et al. 1993)
Unleaded automobile exhaust	1.131–1.164	2.106–2.142	(Tan et al. 2006)
Coal	1.153–1.182	2.090–2.220	(Mukai et al. 2001)
Metallurgic dust	1.161–1.185	2.054–2.100	(Tan et al. 2006)
Industrial sources	1.176	2.1	(Mukai et al. 2001)
TSP (Spring)	1.063–1.098	2.165–2.204	This study
TSP (Summer)	1.069–1.168	2.094–2.197	This study
TSP (Autumn)	1.061–1.132	2.112–2.206	This study
TSP (Winter)	1.069–1.200	2.160–2.202	This study