Environ Monit Assess (2016) 188: 251
DOI 10.1007/s10661-016-5246-y

# The occurrence and sources of heavy metal contamination in peri-urban and smelting contaminated sites in Baoji, China

**Wenbo Deng · Xuxiang Li · Zhisheng An · Liu Yang**

Received: 30 October 2015 / Accepted: 14 March 2016 / Published online: 28 March 2016
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**Abstract** Atmospheric deposition, soil, plant, ore, and coal cinder samples were collected and analyzed to determine heavy metal concentrations in a typical peri-urban industrial area of Baoji. The lead isotope ratio method was employed to trace the source and dispersion of atmospheric heavy metal contamination. Results showed that concentrations of lead, zinc, cadmium, and copper in atmospheric deposition significantly exceed soil background levels and Chinese soil environmental quality standards. The most polluted sites were located in the downwind direction of the smelter, which confirmed this site to be the major pollution source for this area. The other source of heavy metals in this area is a power plant. The investigation into lead isotopes revealed compositions in atmospheric deposition samples were similar to those in ores and coal cinders identifying smelting as the predominant pollution source of lead with the power plant having a minimal effect. Similar isotopic compositions were also found in plants, indicating that the major source of lead in plants was derived from atmospheric deposition, although some evidence was found to suggest uptake from the soil to the roots as an additional contaminant pathway.

W. Deng · X. Li (✉) · L. Yang
School of Human Settlements and Civil Engineering, Xi'an Jiaotong University, Xi'an 710049, China
e-mail: xxli@mail.xjtu.edu.cn

Z. An
Institute of Earth Environment, Chinese Academy of Sciences, Xi'an 710061, China

**Keywords** Atmospheric deposition · Heavy metal · Lead isotope · Atmospheric pollution · Baoji · China

## Introduction

Rapid urban and industrial development in the last few decades has promoted socioeconomic advancement in China. However, it has also caused many environmental problems in peri-urban regions, such as agricultural crop contamination by heavy metals, since these regions provide better market accessibility and higher prices (Smit 1996). Due to the industrial planning of China, an increased number of factories are also located in suburbs leading to a higher risk of heavy metal contamination in soil and plants in peri-urban regions (Hu and Ding 2009). Atmospheric deposition represents a major pathway of anthropogenic inputs of trace metals into the surface environment (Wong et al. 2003) meaning that increased knowledge of processes affecting atmospheric deposition of heavy metals is required.

The moss technique, devised by Ruhling and Tyler (1968, 1971) in the 1970s, is widely used for the estimation of atmospheric trace metal deposition. However, this method is limited to areas without a proliferation of indigenous mosses and is, therefore, not suitable in urban or peri-urban areas. Alternatively, the use of various sampling devices for the collection of atmospheric deposition is a practical approach to monitoring the concentration of heavy metals in the atmosphere. The main instruments for collecting atmospheric deposition include air samplers and passive bulk deposition

samplers (Dye et al. 2000). An air sampler directly draws the air to collect a sample of atmospheric particulates, whereas a passive bulk deposition sampler relies upon pollutants undergoing natural deposition processes. This method of passive sampling can provide valuable information on the influence of air inputs of heavy metals on the surface environments.

Although concentration measurements can provide useful information about enrichments of an element, the source of this element is frequently ambiguous. To solve this uncertainty, isotopic fingerprinting is studied. Lead isotopic analysis has been extensively applied in studies of Pb contamination in different environmental media including soils (Li et al. 2011), sediments (Zhao et al. 2011), plants (Bi et al. 2009), and atmospheric aerosols (Chen et al. 2005). In natural conditions, Pb has four isotopes: $^{204}$Pb, $^{206}$Pb, $^{207}$Pb, and $^{208}$Pb. The last three isotopes are derived from the radioactive decays of $^{238}$U, $^{235}$U, and $^{232}$Th (Fature 1986), as shown in Table 1. $^{204}$Pb has remained stable at 1.4 % in abundance over time (Russell and Farquhar 1960). However, the abundance of the remaining three isotopes significantly covaries and is dependent on the age of the ore body. Different types of ores have distinct isotope ratios and these differences do not disappear in industrial and environment processes. In general, the $^{206}$Pb/$^{207}$Pb ratio of anthropogenic Pb tends to be smaller than the same ratio of naturally derived Pb (Sturges and Barrie 1987). For these reasons, the Pb isotopic analysis represents an important tool for pinpointing the origin of Pb, and in some cases, may be capable of apportioning the contributions of Pb contaminant sources (Mihaljevič et al. 2006).

Baoji, which is located in the western Guanzhong plain, is the second largest city in Shannxi Province and has more than 1000 years of history. Since the 2000s, Baoji has experienced significant industrial and urban development and contamination from heavy metals has become a significant problem. The Dongling Lead-Zinc Smelting Plant (LZSP), which is located in the peri-urban region of Baoji, produces approximately 100,000 t of Pb and Zn each year which has caused the largest lead-zinc deposit in Shannxi Province. Some unprotected smelting wastes are dispersed through various environmental pathways, including the atmospheric deposition of smelting dust, and are an important source of heavy metal contamination in the surrounding area.

The purpose of this study was threefold. The primary purpose was to provide information on heavy metal contamination through atmospheric deposition and the spatial distribution of this contamination. The second objective of this study was to obtain the lead isotope compositions of atmospheric deposition, ores and coal cinder from the LZSP, and coal cinder from the power plant, to clarify the source of Pb contamination in this area. The third objective of this study was to analyze the Pb isotope ratios found in plant samples to identify the dominant route of Pb contamination and accumulation in the plants of this region.

**Table 1** Decay processes of $^{238}$U, $^{235}$U, and $^{232}$Th and their half-lives

| Reaction | Decay constant (year$^{-1}$) | Half-life (years) |
|---|---|---|
| $^{238}_{92}U \rightarrow {}^{206}_{82}Pb + 8^4_2He + 6\beta^-$ | $1.55125 \times 10^{-10}$ | $4.468 \times 10^9$ |
| $^{235}_{92}U \rightarrow {}^{207}_{82}Pb + 7^4_2He + 4\beta^-$ | $9.8485 \times 10^{-10}$ | $7.038 \times 10^8$ |
| $^{232}_{90}Th \rightarrow {}^{208}_{82}Pb + 6^4_2He + 4\beta^-$ | $4.9475 \times 10^{-11}$ | $1.4008 \times 10^{10}$ |

## Methods and materials

### Sampling

The study region is an industrial area in western China with long and narrow topography and an altitude difference of 200 m between the two sides (Fig. 1). The total area of this region is approximately 40 km$^2$. Two reservoirs, which are connected by a river, are located in the northwest and southeast directions. The prevailing wind directions in winter and spring are NW and N, respectively, with SE winds prevailing during the summer. According to meteorological data taken from a 3-year continuous record, the annual average wind speed is 2.19 ms$^{-1}$ and the maximum wind speed is 19.0 ms$^{-1}$. This region belongs to the continental monsoon climate zone. The average annual temperature of the region is 11.4 °C and precipitation is 616.3 mm. A power plant and a smelting plant (LZSP) are located in this area.

A total of ten passive bulk deposition samplers were collected from A1–A10 using cylinder-shaped samplers over a 3-month period from November 2012 to February 2013. The inner diameter and the height of the cylinder-shaped samplers are 15 and 30 cm, respectively. The samplers were fixed at height in range of 3–

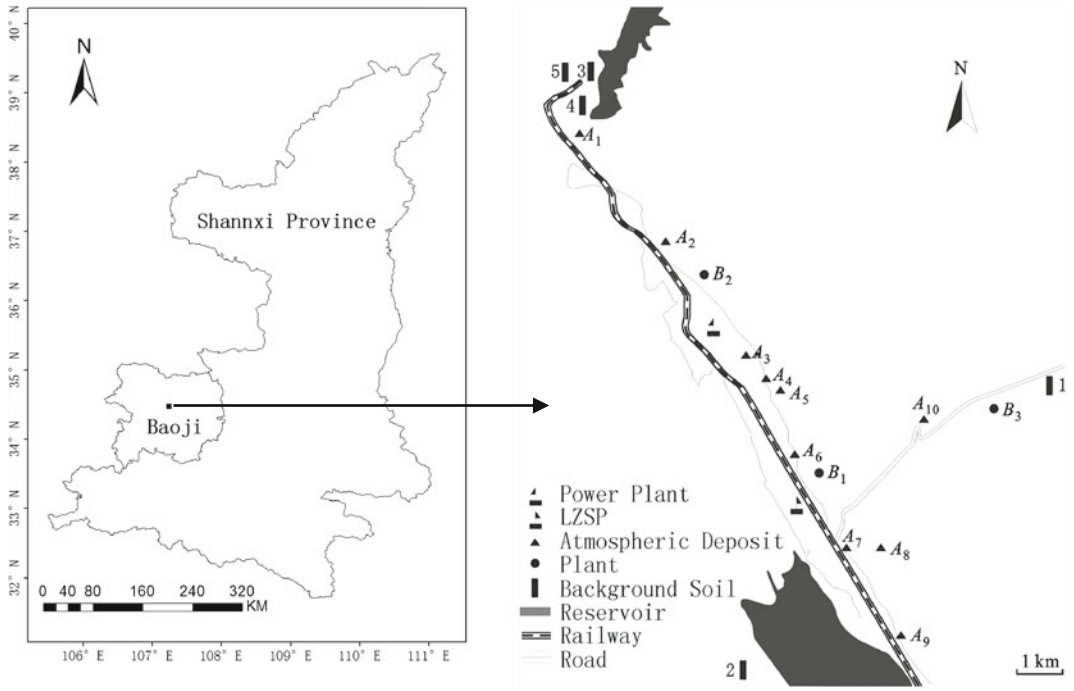


**Fig. 1** Study area and sampling locations

8 m from the ground. Among the sampling sites, the most northerly, marked as A1, is 5 km from the power plant and 10 km from LZSP. For comparison, sampling site A9, at the southerly end of the research area, has the reverse distances at 10 km from the power plant, 5 km from LZSP. Samples of ore, coal cinder, and wheat (B1–B3) were all collected through field sampling from June to August 2013. Five sites (1–5), approximately 5 km away from the study area, were selected as background sites. Soil samples comprising of five individual cores collected at 0–20 cm were collected from these background sites and bagged for transport to the laboratory.

## Sample preparation and analysis

Different pre-processing methods were employed for the different samples. The air-dried soil and deposition samples were sieved through a 2-mm polyethylene sieve to remove stones and other debris. After homogenization, the sub-samples were then crushed in a mortar and passed through a 1-mm sieve to obtain fine particles. Due to the hardness of the ore and coal cinder samples, an ore crushing mill was used for these samples. After crushing, the ore and coal cinder samples underwent a similar process to the soil samples. Plant samples, which are easily dissolved by strong acid, were washed with Milli-Q water and separated into roots (underground) and shoots (above ground). The resulting sub-samples were dried at 75 °C until the sample mass remained constant. It is important that fresh plant samples are treated as soon as possible after collection. All samples were properly stored for subsequent analysis.

To measure the concentration of heavy metals, the prepared samples were processed using the strong acid method (Wong et al. 2002). Briefly, a mass of 0.1 g of sample was placed in a Teflon crucible and submerged in a mixed solution of concentrated $HNO_3$ and $HCLO_4$ (4:1). The sample was then heated progressively to 190 °C in a heating block until it was nearly dry. A total of 20 ml of 5 % (*v*/*v*) $HNO_3$ was added to the crucible and subsequently heated at 70 °C for 1 h. After cooling, samples were decanted into polyethylene tubes and analyzed using an Inductively Coupled Plasma–Atomic Emission Spectrometer (ICP-AES, Perkins Elmer Optima DV3300). The standard reference materials (GSS1-8), which were utilized to control accuracy, were randomly inserted in the analytical process resulting in a precision and bias of <10 %.

Analyses of Pb isotopic ratios of the solutions were achieved by ICP-MS (Perkin Elmer Sciex Elan 6100 $DRC^{PLUS}$) after dilution using 2 % $HNO_3$. The standard error of the Pb isotopic ratios was typically <0.3 %. The

certified standard reference material NIST981 was analyzed after every ten sample solutions, to calibrate and verify the precision. The measured ratios of $^{208}Pb/^{204}Pb$, $^{206}Pb/^{204}Pb$, and $^{207}Pb/^{204}Pb$ were 36.7151, 15.4894, and 16.9409, respectively, consistent with the certified value of 36.7219, 15.4963, and 16.9405, respectively (Todt et al. 1993).

### Statistical analysis

Figures were generated with Excel. The statistical result of correlation coefficients between trace metals was achieved using SPSS 17.0 (SPSS Inc., Chicago, IL).

## Results and discussion

### Heavy metal concentrations

As shown in Table 2, concentrations of Pb, Cd, Cu, Cr, and Zn, resulting from atmospheric deposition show a large range of values across the sampling sites. Values of background soils and the soil environmental quality standards (China) are also provided in Table 2 for comparison. All of the trace metals investigated, with the exception of Cr, exceeded the background soil values and the national soil environmental quality standards (China). This suggests considerable anthropogenic sources of heavy metals as a consequence of rapid industrial development in the study area. Lead, cadmium, and zinc are the principal contaminants in this region with copper having a lesser impact. The dominant anthropogenic sources of heavy metals in this area are from coal combustion in a power plant and smelting activities in LZSP. To identify which of these sources of heavy metals has the most impact, the spatial distribution of the sampling sites was investigated.

Concentrations of Pb, Zn, and Cd in atmospheric deposition in the region are depicted in Fig. 2. The most polluted sampling sites were A7, A8, and A9, which are located in the downwind direction of the LZSP. The contamination of these three sites is more significant than the other sites. As distance from the LZSP is increased, the concentration of Pb, Zn, and Cd gradually decreases. Although A6 is much closer to the LZSP than A9, the level of contamination is much greater at A9. This demonstrates the importance of the influence of prevailing winds transporting greater concentrations of pollutants in this direction. These findings point to the sources of heavy metals in this area being principally derived from LZSP. The reason for the low degree of contamination for A10 is that the site is located at a higher elevation than the remaining sampling sites and was less affected by the anthropogenic sources.

**Table 2** Concentrations ($mg \cdot kg^{-1}$) of heavy metals in atmospheric deposition

| | Pb | Cd | Cu | Cr | Zn |
|---|---|---|---|---|---|
| A1 | 35.78 | 1.12 | 78.96 | 32.75 | 174.4 |
| A2 | 223.36 | 2.93 | 127.62 | 30.71 | 56.61 |
| A3 | 1113.38 | 3.23 | 75.22 | 65.4 | 146.63 |
| A4 | 1603.29 | 37.02 | 133.82 | 165.18 | 68.59 |
| A5 | 1423.7 | 33.5 | 86.16 | 69.4 | 701.86 |
| A6 | 2706.5 | 68.13 | 88.29 | 44.29 | 1703.46 |
| A7 | 5429.24 | 186.74 | 270.86 | 77.39 | 6725.35 |
| A8 | 5416.68 | 148.09 | 173.18 | 43.21 | 6677.84 |
| A9 | 5014.52 | 150.73 | 223.63 | 62.13 | 11125.53 |
| A10 | 475.68 | 1.59 | 25.05 | 26.23 | 623.4 |
| Range | 35.78–5429.24 | 1.12–186.74 | 25.05–270.86 | 26.23–165.18 | 56.61–11125.53 |
| Mean | 2344.213 | 63.308 | 128.279 | 61.669 | 2800.367 |
| Background value[a] | 23.12 | 0.19 | 29.07 | 68.20 | 76.58 |
| Soil environmental quality standard (China)[b] | 80 | 0.8 | 100 | 350 | 300 |

[a] The mean value of five background soils

[b] The value was published by CNEMC (1990)

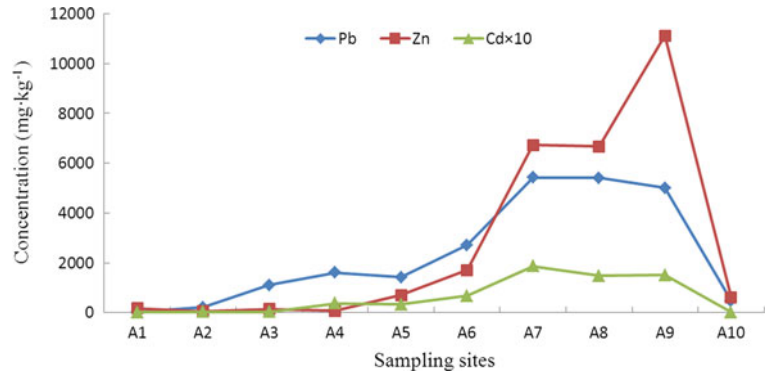


**Fig. 2** Concentrations ($mg \cdot kg^{-1}$) of Pb, Zn, and Cd in atmospheric deposition

To examine the distribution relationships of heavy metals in the atmospheric deposition in the study area, the correlation coefficients between various metals were calculated. Table 3 shows that the relationships between Pb, Cd, and Zn are significantly correlated. This is in agreement with (Bacon and Dinev 2005) who found these metals to be the major contaminants around non-ferrous metal smelter. The correlation between Cu and these three metals is significant, which indicates that Cu may also be a derivative of the smelting operation. As the correlation between Cr and other metals is not significant, it is posited that the Cr in this area comes from the power plant rather than the smelter.

## Lead isotope ratios of the samples

In order to further examine the potential sources of atmospheric Pb in this area, the Pb isotope ratios of all samples were determined. Results from this analysis are provided in Table 4. The range of the $^{208}Pb/^{204}Pb$, $^{207}Pb/^{204}Pb$, and $^{206}Pb/^{207}Pb$ ratios of the deposits include 37.4848–37.6499, 15.5618–15.5740, and 1.1187–1.1276, respectively. Owing to the lack of data concerning Pb isotope ratios in environmental media in Shannxi Province, the Pb isotopic signatures of background soils collected in this study and those found in other cities in China are employed to evaluate the analytical results (see Table 5). By comparison, the $^{206}Pb/^{207}Pb$ ratios of the deposits in this area were generally smaller than the $^{206}Pb/^{207}Pb$ ratios that are derived from the background soils and aerosols collected in other Chinese cities. These results reflect that anthropogenic Pb is an important source of atmospheric deposits in this region.

The relationship between $^{208}Pb/^{207}Pb$ and $^{206}Pb/^{207}Pb$ ratios of the deposition samples are clearly shown in the Fig. 3. This figure also provides the ratios of $^{208}Pb/^{207}Pb$ and $^{206}Pb/^{207}Pb$ relating to anthropogenic Pb sources in this area, i.e., coal combustion and ore of lead-zinc smelting. In general, the contribution of natural Pb is very small, and the anthropogenic sources are the predominant factor influencing lead in atmospheric deposits. Decreasing $^{206}Pb/^{207}Pb$ ratios are associated with an increase in Pb concentration, which is highlighted at the sample sites of A7–A9 which had the highest Pb concentrations and the lowest $^{206}Pb/^{207}Pb$ ratios. This is in agreement with previous studies (Sutherland et al. 2003) and shows that Pb and Zn smelting plays a crucial role in Pb contamination for the most polluted sites in this area. For the remaining sample sites, the Pb in the deposits is derived from both the power plant and the LZSP source. As the Pb concentration of A7–A9 exceeds the Pb concentration from the remaining sample sites, the LZSP is considered to be the major source of atmospheric lead deposition in this region. The $^{206}Pb/^{207}Pb$ ratios of the air deposits collected from sites A7–A9 are lower than the $^{206}Pb/^{207}Pb$ ratios of known sources, which reflect the possible existence of other sources of Pb. As these three sites are the most polluted sites, it is hypothesized that the ore of LZSP has more than one source; however, further investigations are required to prove this conjecture.

**Table 3** Correlation coefficients between the trace metals

| | Pb | Zn | Cr | Cu | Cd |
|---|---|---|---|---|---|
| Pb | 1 | | | | |
| Zn | 0.89** | 1 | | | |
| Cr | 0.12** | −0.04** | 1 | | |
| Cu | 0.83** | 0.80** | 0.27** | 1 | |
| Cd | 0.92** | 0.90** | 0.12** | 0.89** | 1 |

** Significance to 0.01

**Table 4** Pb isotopic ratios of all samples

| Sample type | Sample ID | $n$ ($^{208}Pb/^{204}Pb$) | $n$ ($^{207}Pb/^{204}Pb$) | $n$ ($^{206}Pb/^{207}Pb$) |
|---|---|---|---|---|
| Atmospheric Deposits | A1 | 37.6499 ± 0.0008 | 15.5740 ± 0.0003 | 1.1276 ± 0.0003 |
| | A2 | 37.6201 ± 0.0007 | 15.5690 ± 0.0003 | 1.1262 ± 0.0002 |
| | A3 | 37.5934 ± 0.0009 | 15.5685 ± 0.0003 | 1.1242 ± 0.0003 |
| | A4 | 37.5306 ± 0.007 | 15.5635 ± 0.0002 | 1.1217 ± 0.0002 |
| | A5 | 37.5202 ± 0.009 | 15.5636 ± 0.0003 | 1.1209 ± 0.0003 |
| | A6 | 37.5257 ± 0.009 | 15.5648 ± 0.0003 | 1.1207 ± 0.0003 |
| | A7 | 37.4848 ± 0.008 | 15.5630 ± 0.0003 | 1.1187 ± 0.0003 |
| | A8 | 37.4885 ± 0.007 | 15.5618 ± 0.0002 | 1.1194 ± 0.0002 |
| | A9 | 37.5069 ± 0.009 | 15.5654 ± 0.0004 | 1.1197 ± 0.0004 |
| | A10 | 37.5936 ± 0.008 | 15.5693 ± 0.0003 | 1.1245 ± 0.0003 |
| Plant | Root1 | 37.8088 ± 0.009 | 15.5801 ± 0.0003 | 1.1343 ± 0.0003 |
| | Root2 | 37.8167 ± 0.0015 | 15.5828 ± 0.0004 | 1.1338 ± 0.0006 |
| | Root3 | 37.5653 ± 0.0010 | 15.5682 ± 0.0003 | 1.1213 ± 0.0004 |
| | Shoot1 | 37.7222 ± 0.009 | 15.5767 ± 0.0004 | 1.1304 ± 0.0004 |
| | Shoot2 | 37.7458 ± 0.007 | 15.5799 ± 0.0002 | 1.1315 ± 0.0003 |
| | Shoot3 | 37.5286 ± 0.008 | 15.5663 ± 0.0003 | 1.1203 ± 0.0003 |
| Ore | / | 37.5351 ± 0.0010 | 15.5784 ± 0.0004 | 1.1246 ± 0.0004 |
| Coal (power plant) | / | 37.6495 ± 0.008 | 15.5914 ± 0.0003 | 1.1312 ± 0.0003 |
| Coal (LZSP) | / | 37.3623 ± 0.0006 | 15.5648 ± 0.0002 | 1.1203 ± 0.0002 |
| Soil | Background1 | 38.8774 ± 0.0013 | 15.6633 ± 0.0005 | 1.1960 ± 0.0006 |
| | Background2 | 38.8657 ± 0.0011 | 15.6654 ± 0.0004 | 1.1950 ± 0.0004 |
| | Background 3 | 38.8136 ± 0.0009 | 15.6551 ± 0.0004 | 1.1913 ± 0.0003 |
| | Background 4 | 38.9716 ± 0.0007 | 15.6719 ± 0.0003 | 1.2000 ± 0.0003 |
| | Background 5 | 38.9927 ± 0.0006 | 15.6728 ± 0.0002 | 1.2012 ± 0.0002 |

**Table 5** Pb isotopic ratios of aerosols collected in various Chinese cities

| Sample sites | Sample season | $n$ ($^{206}Pb/^{207}Pb$) | $n$ ($^{208}Pb/^{207}Pb$) |
|---|---|---|---|
| Harbin[a] | Summer | 1.148 | 2.436 |
| | Winter | 1.172 | 2.460 |
| Changchun[a] | Summer | 1.145 | 2.441 |
| | Winter | 1.166 | 2.463 |
| Dalian[a] | Summer | 1.138 | 2.441 |
| | Winter | 1.135 | 2.441 |
| Najing[a] | Summer | 1.166 | 2.460 |
| | Winter | 1.161 | 2.460 |
| Guiyang[a] | Summer | 1.189 | 2.465 |
| | Winter | 1.185 | 2.461 |
| Shanghai[a] | Summer | 1.159 | 2.450 |
| | Winter | 1.156 | 2.449 |

[a] The values were reported by Mukai et al. (2001)

To trace the Pb contamination pathways for the accumulation of trace metals in plants, Pb isotope ratios of wheat in this area were analyzed. Figure 4 shows the isotopic composition of Pb in shoots and roots of wheat at three sampling sites (B1–B3 in Fig. 1) together with the $^{208}Pb/^{207}Pb$, and $^{206}Pb/^{207}Pb$ ratios of atmospheric deposits and background soils. The Pb isotopic composition of wheat was distinctly different to those derived from background soils. If no another sources of Pb were present, the isotopic composition of plants should be similar to those found in background soils. This leads us to the conclusion that other sources of Pb are present. The regression line drawn between atmospheric deposits and background soils (Fig. 4, $R^2 = 0.998$) passes directly through the value for plants meaning that wheat in this area could be defined as a binary relationship between the atmospheric deposits and the background soils. The following equation was employed to calculate

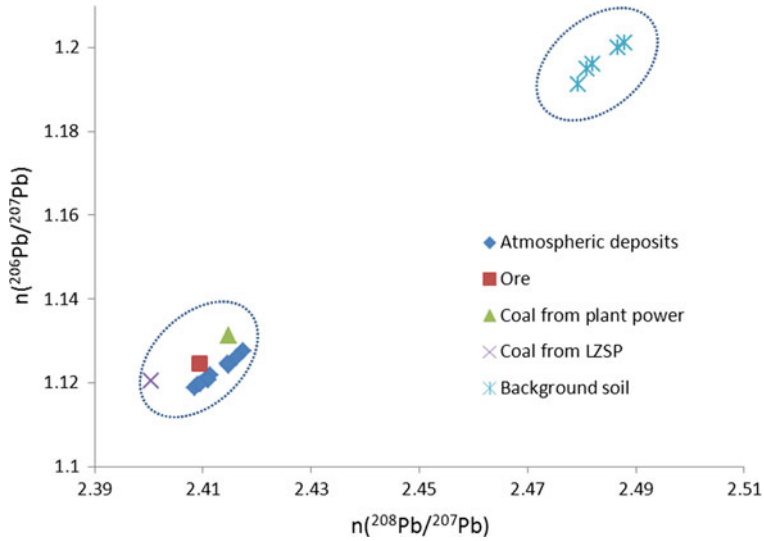


**Fig. 3** The relationship between $^{208}Pb/^{207}Pb$, and $^{206}Pb/^{207}Pb$ ratios of the samples

the contribution of Pb from atmospheric deposits (Monna et al. 1997).

$$X1 = \frac{\left(^{206}Pb/^{204}Pb\right)_{SAMP} - \left(^{206}Pb/^{204}Pb\right)_{soils}}{\left(^{206}Pb/^{204}Pb\right)_{AD} - \left(^{206}Pb/^{204}Pb\right)_{soils}} \quad (1)$$

, where X1 is the percentage contribution of atmospheric deposits and $(^{206}Pb/^{204}Pb)_{SAMP}$, $(^{206}Pb/^{204}Pb)_{AD}$, and $(^{206}Pb/^{204}Pb)_{soils}$ are the isotope ratios of the samples in plants, atmospheric deposits and background soils, respectively. The results range from 85 to 99 %, indicating that atmospheric deposits are the major source of Pb contamination in plants. The difference in the Pb isotope ratios between the shoot and root of one plant confirmed that Pb was entering the plants through both pathways of soil to root and air to leaf (Hu and Ding 2009).

## Conclusion

This study shows that atmospheric deposition concentrations of Pb, Zn, and Cd in the survey region were significantly elevated compared with the soil background and national soil standard values (China). The most polluted sites were located in the downwind direction of the LZSP. These results provide strong evidence that heavy metals are principally derived from anthropogenic sources, such as smelting and coal combustion. The lead isotope ratios of atmospheric deposition in this

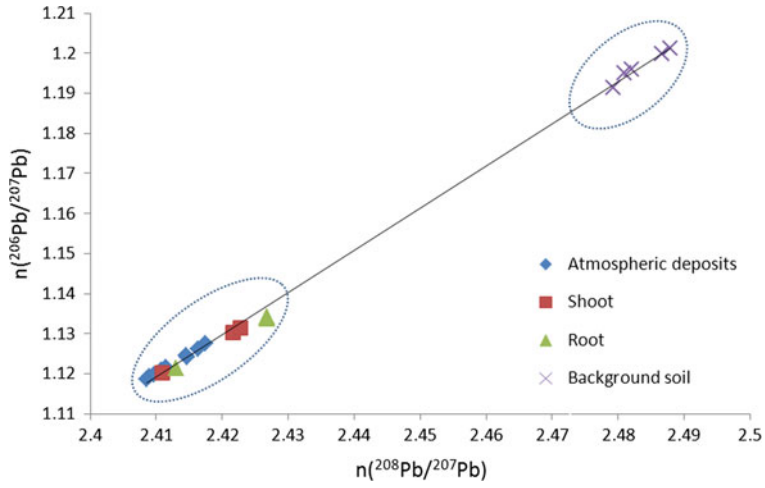


**Fig. 4** Pb isotope ratios of plants and other samples

region also confirmed that the LZSP is the major source of contamination in this area. It is worth mentioning that the most polluted sites had a low value of $^{206}Pb/^{207}Pb$ ratio, reflecting that there may be additional sources of Pb. The isotopic composition of Pb in plants demonstrates that atmospheric deposition is the principal source of metal accumulation in plants in this area.

**Acknowledgments** The research was supported by the Shaanxi Provincial Academy of Environmental Science, Institute of Earth Environment, Chinese Academy of Sciences, and The Research Special Fund for Public Welfare Projects of Ministry of Environmental Protection of China (MEPC) (Grant NO. 201109053).

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