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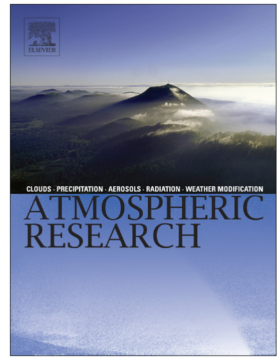

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Biomonitoring trace element contamination impacted by atmospheric deposition in China's remote mountains

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Abstract

The purpose of this study is to better understand the key driving factors affecting the distribution of trace elements in remote mountainous areas at a national scale. In this regard, eight trace elements (As, Cd, Cr, Cu, Ni, Pb, Sb, Zn) in mosses were investigated at thirty remote mountains across China. The sources and possible factors controlling the distribution of trace elements in the mosses were identified through multivariate statistical analysis and Pb isotopic ratios. The results showed that the concentrations of trace elements in the mosses were higher than the levels in most European countries but comparable to those in Chinese cities. Mosses had the highest concentration levels of Cd and Sb and slightly lower levels of other trace elements. Spatially, moss contamination with the trace elements was significantly higher in the eastern, southern and southwestern China than in other regions. Mining-related activities, fuel combustion and other industrial emissions were identified as the main anthropogenic sources of As, Cd, Pb, Sb and Zn, whereas Cr, Cu and Ni were mainly from natural sources. The precipitation from monsoons clearly altered the distribution of anthropogenic elements in the mosses. The results of this study indicate that the trace elements in the mosses of China's remote mountains were affected by regional anthropogenic emissions, and the monsoon-dominated trace element transport and deposition should be highlighted at a large regional scale. These findings provide baseline data for future targeting policies to protect remote ecosystems from long-term inputs of trace elements via

long-range atmospheric transportation.

Keywords: Trace elements; moss monitoring; source identification; Pb isotopes; atmospheric deposition; mountain regions

1. Introduction

Trace elements emitted into the atmosphere by various anthropogenic sources are significant due to their threat to ecological safety and human health (Duan et al., 2018; Komarek et al., 2008). Extensive research has been conducted to determine the distribution, sources and toxicity of atmospheric trace elements over the past few decades in population centers, such as megacities, and industrial and economic areas around the world (Fernández-Camacho et al., 2012; Hao et al., 2018; Luo et al., 2016; Moreno et al., 2011; Pan et al., 2015). However, very little attention has been given to the trace element contamination in remote mountainous areas, which feature relatively pristine ecological environments with high levels of biodiversity, rare species, and few population centers or tourists. Remote mountains, especially high mountains, are sensitive to global changes and can trap airborne trace elements because of the effects of cold condensation based on the temperature dependence of precipitation scavenging (Bing et al., 2016a, 2018). Monitoring the contamination of trace elements in multiple remote mountain ecosystems can facilitate the identification of regional emissions within these ecosystems.

It is a challenge to identify trace element contamination in remote mountains due to the harsh environmental conditions. Mosses can accumulate trace elements from the atmosphere, and thus, they are suitable for monitoring the spatial patterns of the elements over large regions (Harmens et al., 2010; Kłos et al., 2018; Kolon et al., 2015; Lequy et al., 2017). The suitability and possibility of using mosses for cross-regional and extensive surveys are based on their moderate costs compared with *in situ* measurements of bulk deposition. In Europe, using mosses to monitor atmospheric contamination of trace elements has been conducted since 1990, in up to thirty-five countries every five years (Harmans et al., 2008; Nickle and Schröder, 2017). This survey has demonstrated a decreasing trend of atmospheric contamination of trace elements (Harmens et al., 2015; Schröder and Pesch, 2010). In China, there are no networks monitoring atmospheric contamination of trace elements in mosses at the cross-regional or national scale. In this study, we selected thirty remote mountains in China to investigate the atmospheric contamination of trace elements through moss monitoring. The mosses were sampled mostly in forest areas, aiming to assess forest exposure to the elements in terms of bioaccumulation.

Monitoring biological indicators with fingerprint techniques has made it possible to distinguish the source of anthropogenic elements (Bing et al., 2016b; Shotyk et al., 2015). Among these isotopic fingerprints, the Pb isotopic tracing technique has been more extensively applied due to its comprehensive dataset around the world (Bi et al., 2017; Cheng and Hu, 2010; Cong et al., 2011; Dong et al., 2017; Ferrat et al., 2012; Kaste et al., 2003; Klaminder et al., 2008; Komárek et al., 2008; Steinnes et al., 2005). Meanwhile, multivariate statistical methods (e.g., factor analysis, cluster analysis) have been broadly applied to determine the source of trace elements in the environment and thus support the interpretation of the sources or influencing factors of an element. The GeoDetector is a statistical method that analyzes the spatial heterogeneity and identifies the driving factors of variables (Wang et al., 2010; Wang and Xu, 2017). It not only detects the effect of a single independent variable on dependent variable but also analyzes the interactive relationship between independent variables. The q statistic value varied from 0 to 1 and was proposed to indicate the effect strength. Moreover, the GeoDetector uses both qualitative and quantitative data and allows the sample numbers of each type of variable to be as few as two. This method does not ask the dataset to meet a normal distribution, reducing the influence from collinearity between variables and thus improving the reliability and validity of the estimates. These methods are believed to improve our insight into the key factors affecting the distribution of potential toxic elements in remote mountains at a large spatial scale.

In last few decades, China has undergone rapid development through industrialization and urbanization with substantial exploitation and utilization of resources and energy. With the imbalance of regional industrial structures, the distribution of resource and energy consumption varies significantly in China. In general, coal is the dominant energy source for power generation in China, and most of the thermal power stations are in the central and northern regions. Thus, coal combustion is the primary contributor to pollution emissions (Chen et al., 2013; Li et al., 2017; Tian et al., 2014). In addition to coal, fuel consumption is higher in eastern China than in other regions due to that region’s diverse economy (Liang and Zhang, 2009). Another important resource is polymetallic ores, which are mainly distributed in southern, southwestern and northwestern China. Mining and metal smelting are potential sources of trace elements in the surrounding environment (Bi et al., 2007). Trace elements released into the atmosphere are transported at short or long distances based on atmospheric circulation (Bing et al., 2014, 2018; Shotyk et al., 2016). We, therefore, hypothesized that the accumulation of trace elements in the mosses of remote mountains would respond to the spatial distributions of resource exploitation and energy consumption at the national scale in China.

This study aimed to address the question of whether the anthropogenic emissions of trace elements have a legacy effect on the China's remote mountains. By designing a national-scale program, we targeted mosses from thirty remote mountains across China to a) investigate the spatial distribution of trace elements including As, Cd, Cr, Cu, Ni, Pb, Sb and Zn and assess the contamination states of the mosses; b) distinguish natural versus anthropogenic sources of the trace elements through multivariate statistical analysis and Pb isotopic ratios; and c) decipher the driving factors of the distribution of these elements in the mosses. This is the first time using mosses as the bioindicators of trace element contamination to understand anthropogenic effects on the remote mountain areas across China.

2. Materials and methods

2.1. Study area

Thirty remote mountainous areas across China were selected (Figure S1), and these mountains are located in the national and provincial nature reserves. The spatial ranges of the study area are 21-53° latitude, 100-128° longitude and 285-4225 m above sea level (a.s.l.) altitude. The mean annual temperature (MAT) and precipitation (MAP) in the study area are -9.2-32.4 °C and 265-2816 mm, respectively. Specific characteristics of each sampling location are presented in Table S1.

2.2. Sample collection and preparation

At each mountain, the sampling sites were chosen based on the local altitudinal gradient, dominant vegetation types and the existence of mosses. In non-growing season of 2012 and 2013, the epigeic mosses were carefully collected in at least five spots at each site (Figure S2a). The nearest distance of the adjacent spots was approximately 5 m in order to obtain representative samples at a site. In total, 166 moss samples, including 22 moss families, were collected (Table S1). The samples were placed in polyethylene zip-bags and transported to the laboratory. After removing the fake roots (Figure S2b), the moss samples were oven-dried at 60 ^{o}C to reach a constant weight. Then, the samples were pulverized by a crusher with stainless steel tri-rotors so that they could pass through a 100-mesh Nylon screen for the chemical analysis.

2.3. Chemical analysis

The moss samples were digested with a mixture of HNO_3 and H_2O_2. Briefly, approximately 0.2 g of the sample digested in a digestion tank (50 mL TFM-PTFE, Berghof DAB-2, Germany) with 2.5 mL HNO_3 and 0.5 mL H_2O_2 at 195-210 ^{o}C. After cooling, 0.5 mL 1:1 HCl was used to dilute the solution to 25 mL for the analysis. The standard solution SPEX^{TM} was used as the standard. Quality control was implemented by the analysis of blanks, duplicate samples and reference materials (GBW07603 and GBW07604). The concentrations of As, Cd, Cr, Cu, Ni, Pb, Sb and Zn in the diluted digestion solution were determined by inductively coupled plasma mass spectrometry (ICP-MS). The concentrations of major elements (e.g., Al, Ti) were also measured by inductively coupled plasma atomic emission spectrometer (ICP-AES, Leeman Labs, Profile DV). The precision and accuracy were routinely below 5% through the relative standard deviation of the repeated samples and standard reference materials, respectively.

The moss Pb isotope ratios ($^{206}Pb/^{207}Pb$ and $^{208}Pb/^{206}Pb$) in the digestion solution described above were determined by the ICP-MS (Agilent 7700x). A standard reference material from the United State National Institute of Standards and Technology-SRM 981 was selected for the instrument calibration ($^{208}Pb/^{206}Pb$ = 2.1681 ± 0.0008, $^{207}Pb/^{206}Pb$ = 0.9146 ± 0.00033), and the standard material of GBW04426 from China was used for the quality control ($^{208}Pb/^{206}Pb$ = 2.1280 ± 0.00016, $^{207}Pb/^{206}Pb$ = 0.8677 ± 0.00007). According to multiple measurements of GBW04426 with the calibration of SRM 981, the ratios of $^{208}Pb/^{206}Pb$ and $^{207}Pb/^{206}Pb$ were 2.135 ± 0.0016 and 0.869 ± 0.0009, respectively. The precision of the $^{208}Pb/^{206}Pb$ and $^{207}Pb/^{206}Pb$ ratios through the replicate analysis of the SRM 981 was <0.07% and <0.12%, respectively.

2.4. Calculations

The enrichment factor (EF) of trace elements in the mosses was calculated as follows:

$$EF = (Me/Al)_{sample}/(Me/Al)_{background}$$

where $(Me/Al)_{sample}$ is the concentration ratio of a trace element and Al in the mosses, whereas

$(Me/Al)_{background}$ is the ratio in the upper crust (Taylor and McLennan, 1995). The EF values < 5 are considered as having no atmospheric contribution, 5-10 as having slight contamination, 10-100 as having moderate contamination, 100-1000 as having higher contamination, and > 1000 as having high or extreme contamination (Steinnes and Friedland, 2006).

2.5. Statistical analysis

The model of the GeoDetector was applied to determine the driving factors determining the distribution of the trace elements in the mosses, which has been introduced in details elsewhere (Wang et al., 2010, 2016). One-way ANOVA (Fisher test, $p < 0.05$) was used to identify the significant difference between the element concentrations, EFs and Pb isotopic ratios; factor analysis, cluster analysis and regression analysis were applied to classify the elements in the samples based on their concentrations. All statistical analyses in this study were performed by the software packages of Office Excel 2010, SPSS 16.0, Origin 8.0 and ArcGIS 9.3 in Windows.

3. Results and discussion

3.1. Trace element concentrations in mosses

The concentrations of trace elements in the mosses are shown in Table 1. Compared with the compiled data in other cities and countries, the concentrations of the eight trace elements in the mosses of China's remote mountains are comparable to those in the main Chinese cities, whereas they are one order of magnitude higher than those in European countries. Moreover, the concentrations of these elements are significantly higher in our study area than in the Canadian high arctic regions, but they show similar values to those in the Qiyi Glacier on Tibetan Plateau, except for Cd. The comparison reveals that the trace elements studied in China's remote mountains are generally at a high level, suggesting the possibility of atmospheric contamination.

Accumulation of the elements in moss species is determined by their species-dependent characteristics such as growth rates, surface roughness, and dry mass (Aboal et al., 2017; Čeburnis and Valiulis, 1999; Kempter et al., 2010). In our study, twenty-two families of the mosses were observed in the thirty remote mountain areas (Table S1). It is a big challenge to compare element concentrations based on the species level in such national scale investigation with abundant moss species in China. As a result, the element concentrations for moss families with more than three individuals were selected and analyzed for differences in accumulations of the elements in the mosses. We observed little difference in the concentrations of the trace elements among the moss families (Table S2). In general, *Dicranaceae* serves as a better monitor for As, Cd, Pb, Sb and Zn, whereas the *Hylocomiaceae*'s ability to indicate trace element contamination is minimal except for Cd and Cr. Similar accumulation concentrations of trace elements in multiple moss families were observed. This may be related to two possible interpretations. On the one hand, if the loadings of the trace elements are low in the atmosphere, the various mosses would carry out preferential and/or sensitive uptake of the nonessential elements based on their annual production. On the other hand, with high loadings of the elements in the environment, mosses would become more enriched with them during their growth periods (normally several years) as they take up essential nutrients. Apparently, the higher concentrations of the eight trace elements (especially nonessential elements) in our mosses compared with those in other regions worldwide (Table 1) supports the high loadings of these trace elements in the remote mountains.

3.2. Spatial hotspots of trace element contamination by the moss biomonitoring

The concentrations of all the trace elements in the mosses showed a large spatial heterogeneity by the CVs, especially for Cr, Cu and Ni (Table 1). This was also observed based on the concentrations of trace elements at each mountain (Fig. 1). The concentrations of the trace elements in the mosses showed significant correlation with latitude and longitude ($p < 0.05$, Fig. 2). The relatively higher concentration values were observed in the ranges of approximately 24-34^{o}N and 107-120^{o}E, corresponding to the southern areas of China (Fig. 1).

According to the division of Chinese administrative regions, the concentrations of As, Cd, Cr, Cu, Ni, Pb and Sb were significantly higher in eastern, southern and southwestern China than in other regions ($p < 0.05$, Table S3). Although the concentrations of Zn did not present spatial differences, its highest value was observed in eastern China. The distribution patterns of the trace elements in the mosses of China's remote mountains are generally in accordance with those in Chinese soils (Chen et al., 2015; Zhao et al., 2015) and precipitation in China's natural terrestrial ecosystems (Zhu et al., 2016). The relatively high geochemical background and intense human activity in the southern, eastern and southwestern China may be related to the distribution of trace elements in the mosses (Luo et al., 2016).

The EFs of the trace elements in the mosses further indicate the contamination level of the mosses in the remote mountains. The contamination levels of the eight trace elements were clearly different in these mountains through monitoring the moss (Table 2). According to the contamination classification, mosses showed the higher and/or high contamination with Cd and Sb; mosses were moderately contaminated with As, Pb and Zn; and mosses had little to no contamination with Cr, Cu and Ni (Table 2). The spatial distribution of the EFs of the trace elements was correlated with the concentrations of the elements, which were higher in the southern, eastern and southwestern China compared with other regions ($p < 0.05$), despite of a slight difference in the highest EFs of each element (Table S4). This outcome supports the fact that the local and/or regional emissions of trace elements in these areas contribute to the contamination of the mosses in the remote mountains.

Moss is an effective bioindicator of atmospheric metal contamination in the environment (Harmens et al., 2010; Shotyk et al., 2015; Meyer et al., 2015). The markedly higher EFs of As, Cd, Pb, Sb and Zn in the mosses from the remote mountains explicitly demonstrate their potential to originate from non-crustal sources. We also calculated the EFs of other elements such as Ba, Fe, Ca, Mg, Mn, Sr and Ti in the mosses (Table S5). Except for several elements that may be affected by dry (e.g., air dust) and wet deposition and/or biological processes (Brahney et al., 2015; Tipping et al., 2014), the EFs of these elements were generally lower than 5.0, indicating no contamination. This suggests that the remote mountains are less influenced by trace elements from natural origins and that the trace elements in the mosses are mainly from the atmosphere.

3.3. A regional perspective on the sources of trace elements

Multivariate statistical analysis is conducive for screening the classification of elements in a complex environment. The results of factor analysis (Table S6) revealed the distinctive sources of the trace elements in the mosses. As, Cd, Pb, Sb and Zn were substantially different from crustal elements (e.g., Al, Ti), indicating their non-crustal and anthropogenic sources. However, Cr, Cu and Ni presented weak relationships with both crustal and anthropogenic elements. This suggests that they are sourced from both natural and anthropogenic origins.

Lithogenic elements, such as Al and Ti, in the atmosphere are almost always derived from weathering processes of soils or rocks (Di Palma et al., 2017; Kempter et al., 2017). In our study, we also observed significant relationships between Al and Ti in the mosses ($R^2 = 0.629$, $p < 0.001$).

A regression analysis was applied to establish the relationship of Ti with the trace elements (Fig. 3). The correlation of Ti with Cd and Sb was not significant, which further evidences their anthropogenic source from atmospheric deposition. Although Cu did not correlate significantly with Ti, it did correlate with Ti after removing the strikingly high concentrations of Cu at the Wuzhi and Nanling Mountains ($p < 0.001$). This indicates that Cu in the mosses is derived from natural sources. Meanwhile, a similar case was also observed for Cr and Ni, although they showed a significant correlation with Ti.

The results of the regression analysis for As, Pb and Zn (Fig. 3) showed distinct differences from the results of factor analysis and cluster analysis. This difference is attributed to the heterogeneity and complexity of the sources of the elements in the mosses at a large spatial scale. To ascertain the different sources of the trace elements in the mosses, we re-established the relationships between the trace elements and Ti at the mountains within each Chinese administrative division (Table S7). All trace elements in the mountain mosses from northeastern China showed significantly positive correlations with Ti, and same was also observed for elements such as As, Cr, Ni in northern and northwestern China, whereas other trace elements, except Cr, did not correlate with Ti in eastern, southern and southwestern China. This observation further supports the result that the spatial differences in trace element sources cause the contradictory results of the different statistical analysis.

Shotyk et al. (2016) concluded that airborne dust was the dominant source of trace elements in peat moss from the bogs of northern Alberta, which was supported by the significant relationships between the trace elements and lithogenic elements. This significant correlation of the trace elements with Ti reflects the naturally occurring air dust contribution to them. However, the transport of trace elements in dust depends on the size of the particulate matter particles. Particles larger than 10 μm have a relatively short life-time in the atmosphere, whereas particles smaller than 1 μm can be permanently suspended (Bargagli, 1998; Shotyk et al., 2015). Considering the remote locations of the mountains studied, it is not likely that the coarse dust beyond our study area or from other regions or countries deposited in these remote mountains. Instead, based on the results of the EFs (Table S4), we conclude that As, Cr, Cu and Ni in the mountain mosses from northeastern, northern and northwestern China are probably from the local, naturally occurring dust, and Cd, Pb, Sb and Zn with relatively high EFs in northern and northwestern China are likely from anthropogenic emissions. For the mountains in eastern, southern and southwestern China, the other trace elements, except Cr that is from naturally occurring dust, in the mosses are also mainly from anthropogenic emissions.

3.4. Anthropogenic contribution evidenced by Pb isotopic ratios

Based on the Pb isotopic ratios in various natural and anthropogenic materials, the Pb sources in mosses can be identified, which helps identify the possible sources of other trace elements that correlated well with Pb. The Pb isotopic ratios in the mosses across China's remote mountains showed broad ranges: 1.121-1.205 (mean ± SE: 1.166 ± 0.001) for $^{206}Pb/^{207}Pb$ and 2.053-2.198 (2.106 ± 0.001) for $^{208}Pb/^{206}Pb$ (Table S8). In addition, these ratios exhibited significant difference among the administrative regions of China, which indicated the spatial anthropogenic Pb sources for the mosses. We compiled large quantities of Pb isotopic ratios in possible source materials in China at a large spatial scale, including polymetallic ores (n = 3364), local C-horizon soil (parent materials, also see Table S8, n = 451), air particulate matters (PMs, n = 155), vehicle exhaust (n = 56), Chinese loess (n = 58), and coal combustion emissions (n = 18) (Fig. 4).

In comparing $^{206}Pb/^{207}Pb$ versus $^{208}Pb/^{206}Pb$ (Fig. 4A), we observed three marked characteristics of the Pb isotopic ratios in the mosses: a) they were relatively constrained among the dataset, indicating similar sources of Pb in general; b) they were significantly different from the ratios in Chinese loess, local C horizon soil, vehicle exhaust and air dust in industrial areas; and c) they overlapped with ratios in polymetallic ores, coal combustion emissions and air particulate matters (PMs) in cities, despite some specific differences (Fig. 4B). Although the remote mountains are far from human activity and industrial centers and located in nature reserves, the Pb in the mosses was not mainly from local sources due to the marked difference in its isotopic ratios from those in the local soil. However, remote geographic isolation is a possible reason for the difference in the moss Pb isotopic ratios for vehicle exhaust and PMs of industrial areas that can be deposited at short distances. Moreover, the moss Pb isotopic ratios were striking different from those in vehicles exhaust, which confirms that the phase-out of Pb-contained gasoline reduced Pb emissions from vehicles, and it is not a major source of Pb in the atmosphere of the remote areas.

Mining and metal smelting are considered as the main sources of Pb in the mosses. The polymetallic ores showed identical Pb isotopic ratios to mosses, and spatially, the ratios in the mosses corresponded to the ratios in the ores, except the case in the northwestern China. This indicates that the local mining-related emissions are the dominant source of Pb in the mosses. As one of the largest global producers and consumers of metals/metalloids, China possesses diversified and large-scale resources of polymetallic ores (Li et al., 2014). The exploitation and utilization of these resources, especially illegal operations or inadequate enforcement of environmental protection laws for several small and medium mining enterprises, has resulted in large quantities of trace elements being released to the environment (Zhao et al., 2015), which induced atmospheric metal contamination. For example, Zhang et al. (2009) observed high concentrations of trace elements in PM_{10} (As: 15239 mg/kg, Cd: 210 mg/kg, Cu: 673 mg/kg, Sb: 445 mg/kg, Pb: 8053 mg/kg, and Zn: 13151 mg/kg) in a mountain mining area of South China, which were several orders of magnitude higher than those in the air dust of non-mining areas of northwestern China (Jilili et al., 2015).

The Pb in the atmosphere is commonly a mixture of multiple sources. The Pb isotopic ratios in the mountain mosses from the northwestern China did not overlap well with those in local ores, but did well with coal combustion emissions from other regions. This suggests that the emissions from coal combustion contribute to Pb accumulation in the mosses. Energy consumption, especially coal combustion that accounts for more than 70% of the total energy consumption, has been regarded as a major emission source of trace elements in China for decades (Cheng and Hu, 2010; Chen et al., 2013). In 2007, the atmospheric emissions from coal utilization in China were estimated as As 2205 t, Cd 245 t, Cr 8218 t, Ni 2308 t, Pb 12547 t and Sb 547 t (Tian et al., 2013). Moreover, the Pb isotopic ratios in the mosses were also similar to those in the air PMs of major cities from other regions, which means that the long-range transport of fine particulates with Pb is another source of Pb.

3.5. Significance of mutual influences from climate and anthropogenic emissions

Environmental conditions, including forest type, precipitation, predominant wind and its speed, terrain, in moss habitats regulate the distribution of trace elements in mosses (Bing et al., 2014; Kempter et al., 2010). We compiled both qualitative and quantitative variables that included natural and anthropogenic sources to determine the driving factors of trace element accumulation in the mosses using the model of GeoDetector (Table S9). The results showed that monsoon and MAP were the dominant natural factors influencing the distribution of most trace elements in the mosses, and industrial production and smog dust emissions were the dominant anthropogenic sources (Table 3).

The types of monsoons reflect climate conditions, including temperature, precipitation, and dominant wind direction and intensity. Many studies have reported that the trace elements in terrestrial ecosystems correlated well with wet precipitation (Cong et al., 2015; Pan and Wang, 2015; Zhu et al., 2016). In our study, we also observed the significant relationship of anthropogenic elements (As, Cd, Pb, Sb) with MAP and monsoon type (Table 3), which suggests that wet precipitation plays a key role in the accumulation of these elements in the mosses. In summer, the eastern and southwestern monsoons bring much more rainfall to the southern regions of China. Mountains have substantial effects on the dynamical atmospheric structure of the monsoons (Wu et al., 2014), which can alter dry and wet deposition in the forms of precipitation, fog and clouds, depending on altitude (Fomba et al., 2015). As a result, the wet deposition may increase the amount of trace elements in remote mountains with atmospheric circulation. In winter, the westerly wind passes through the northern China where the main pollution emitting sectors exist (Wang et al., 2017). This may be the reason for the difference in the amounts of trace elements in the mosses between the northern and southern regions of China. As a whole, the significant correlation between anthropogenic elements in the mosses and monsoon type and MAP reveals the effects of short- or long-range atmospheric transport on the distribution of the elements in the mountains.

The accumulation of trace elements in the mosses was related to industrial production and smog dust emissions (Table 3), which is in accordance with the results of the Pb isotopic ratios (Fig. 4). Current industrial development has been a major contributor of trace elements into the atmosphere, and fine particles (especially nanoscale), which can be transported long distances through atmospheric circulation, are important carriers of the trace elements. Moreover, As, Cd and Sb in the mosses showed a significant correlation with the emissions from fuel consumption, and a weaker correlation was observed for Pb and Zn, indicating the important contribution of fuel combustion to these atmospheric trace elements. Coal consumption showed a significant relationship with Cu and Ni but not with the anthropogenic elements, which is not in agreement with the results of the Pb isotopic tracing. The limited dataset on coal consumption may be a reason for this result. Meanwhile, coal consumption did not represent the emissions from coal combustion due to the multiple roles of coal in industrial production (Chen et al., 2013).

The altitude of mountains is a variable commonly related to local temperature and precipitation, geographical distribution and vegetation composition and is believed to alter the deposition of trace elements in moss tissues (Aboal et al., 2017; Lequy et al., 2017). However, we only observed a significant correlation between altitude and Cd (Table 3) and the weak relationship of altitude with Cu, Pb and Zn (Figure S3). This result, on the one hand, is attributed to the different altitude ranges at each mountain, where most of the sampling sites are located at the altitudes below 2000 m a.s.l. (Table S1). On the other hand, the role of altitude in intercepting trace elements in the atmosphere cannot be highlighted at the national scale due to the different patterns of local emissions and long-range atmospheric transport of anthropogenic elements. In contrast, at the local scale, the distribution patterns of trace elements in mosses in conjunction with altitude have been observed (Gerdol and Bragazza, 2006; Lee et al., 2005).

In mountain forest ecosystems, forest type is an important factor controlling trace element deposition on the forest floor through canopy types, leaf interception and/or washing, etc. (Bing et al., 2014; Lequy et al., 2017; Uehara and Kume, 2012). Unexpectedly, we did not observe that forest type affected the distribution of trace elements in the mosses, except for Zn (Table 3). This result suggests that the simple classification of plant types (Table S9) is probably not adequate to interpret the complicated effects of vegetation on the accumulation of trace elements in the mosses. Instead, more specific indices such as tree species, canopy density, leaf surface area and throughfall need to be further explored.

3.6. Responses of atmospheric trace elements to regional human activity

According to the discussion above, the type of trace elements (especially As, Cd, Pb, Sb and Zn) in the mosses of China's remote mountains was related to the anthropogenic release of emissions, including mining-related emissions (e.g., mining, metal smelting) and long-range atmospheric transport from coal combustion or other industrial emissions (Table 3, Fig. 4). This partly supports our hypothesis that the types of trace elements present in the mosses of remote mountains were related to the resource exploitation and energy consumption in China. However, the spatial distribution patterns of anthropogenic elements in the mosses were not completely consistent with the patterns of regional resource exploitation and energy consumption. Based on the Pb isotopic ratios, the Pb in the mosses was mainly from regional mining of polymetallic ores, except in the northwestern China. This is not in agreement with the patterns of resource and energy consumption in the northern and northeastern China, where coal consumption is dominant (Tian et al., 2014). In addition to the dominant effect of mining-related emissions, we found that monsoon climate was a key driving factor for the distribution of anthropogenic elements in the mosses. Recently, in China, cities with a high population density are a major source of fine particulates (Han et al., 2015), and the spatial spillover and regional transmission of these particulates, under the effects of meteorological conditions, have been evidently observed (Cheng et al., 2017). This mechanism of atmospheric deposition of trace elements from anthropogenic emissions in the remote mountains may be notable. The results from the Pb isotopic ratios in the PMs and coal combustion also provided evidence that the Pb from the southeastern and southwestern regions was a potential contributor to Pb in the northern and northwestern regions because of the effects of the East Asian monsoon and Indian monsoon (Bing et al., 2014; Zheng et al., 2015). In contrast, the westerly wind did not bring anthropogenic elements to the central and eastern regions due to the relatively slow development of western China, which was demonstrated by the inconsistent Pb isotopic ratios between the mosses and loess. In the future, numerical modeling based on the moss biomonitoring needs to be conducted to understand the transport processes and deposition of trace elements in the remote mountains, which will improve our knowledge of the regional and/or global patterns of atmospheric transport of the trace elements.

4. Conclusions

The investigation of trace elements in the mosses at thirty remote mountains across China observed relatively higher contamination of As, Cd, Pb, Sb and Zn compared with other elements. Moss contamination with the trace elements was significantly higher in the eastern, southern and southwestern China than in other regions. Mining-related activities, fuel combustion and industrial emissions were the main anthropogenic sources of As, Cd, Pb, Sb and Zn in the mosses. Monsoon-dominated trace element transport and deposition regulated the distribution of the anthropogenic elements in the mosses. The trace elements in the mosses of China's remote mountains to a large extent responded to the regional anthropogenic emissions.

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Table 1 Statistical characteristic of trace element concentrations (mg/kg) in the mosses of this study and other reports

Regions		As	Cd	Cr	Cu	Ni	Pb	Sb	Zn	Refs
	Min.	0.5	0.10	1.7	5.8	2.9	3.0	0.07	28.1	This study
	Max.	26.8	4.65	466	2064	594	390	6.31	1266	
	Mean	5.0	1.31	30.4	48.9	18.2	54.6	1.09	132	
Mts. in China	Median	4.2	1.08	17.4	29.2	11.3	46.4	0.80	112	
(n=166)	SE[a]	0.3	0.07	3.6	12.8	3.7	3.4	0.08	8.6	
	Skewness	2.3	1.3	5.9	11.4	11.3	3.4	2.5	6.9	
	Kurtosis	8.6	1.6	49.9	138.2	136.7	20.8	7.3	66.8	
	CV[b]	72.7	70.4	150.8	337.1	259.1	80.2	96.5	84.2	
Cities in China										
Wuxi (n=49)	Mean (ND)	ND	0.93	15.1	36.5	10.1	31.2	ND	206	Yan et al. (2016)
Taizhou (n=60)	Mean (2012)	ND	1.05	30.9	48.0	21.0	44.2	ND	233	Zhou et al. (2017)
Xuzhou (n=55)	Mean (2013)	ND	0.82	26.3	28.0	20.1	33.0	ND	155	Liu et al. (2016)
Chengdu (n=15)	Mean (ND)	3.61	0.99	89.1	69.4	306	23.8	ND	172	Ge et al. (2013)
28 countries in Europe	Mean (1990)	ND	0.36	1.83	7.52	2.17	14.7	ND	46.7	Harmens et al. (2010)
	Mean (1995)	0.26	0.29	1.47	7.04	1.81	8.75	ND	39.0	
	Mean (2000)	0.22	0.20	1.82	6.19	2.00	6.29	ND	36.3	
	Mean (2005)	0.21	0.18	1.81	6.25	1.74	4.19	0.17[c]	33.0	
Canadian high	Mean	1.17	0.092	6.14	4.71	2.76	0.992	ND	19.2	Wilkie and

arctic (n = 59)	(2007)									Farge (2011)
Qiyi Glacier in Tibetan Plateau (n = 17)	Mean (2009)	9.15	0.234	34.5	140	ND	26.2	ND	170	Ma and Li (2014)

[a] Standard error; [b] Coefficient of variance (%); [c] Mean of the medians; ND: No data; The date in the bracket of the second column represent the sampling time.

Table 2 Classification of trace element contamination according to their enrichment factors at each mountain within six Chinese administrative divisions

Mts.	Divisions	0<EF≤5	5<EF≤10	10<EF≤50	50<EF≤100	EF>100
DX	NE China	Cr	As, Cu, Ni, Pb	Sb, Zn	Cd	--
XX	NE China	Cr, Cu, Ni	As, Pb	Sb, Zn	Cd	--
CB	NE China	Cr, Ni	As, Cu	Pb, Sb, Zn	--	Cd
BCW	North China	Cr, Cu, Ni	As, Pb	Sb, Zn	Cd	--
GD	North China	Cr, Ni	As, Cu	Pb, Sb, Zn	--	Cd
HS	North China	Cr, Cu, Ni, Pb, Zn	As, Sb	Cd	--	--
SHB	North China	Cr, Cu, Ni, Pb	As, Sb, Zn	Cd	--	--
WYZ	North China	Cr	Cu, Ni	As, Pb, Sb, Zn	--	Cd
AS	NW China	--	Cr, Cu	As, Ni, Pb, Sb, Zn	--	Cd
JF	NW China	Cr	Cu, Ni	As, Pb, Sb, Zn	--	Cd
QF	NW China	Cr	Cu, Ni	As, Pb, Sb, Zn	--	Cd
SYK	NW China	Cr, Cu, Ni, Pb, Zn	As, Sb	Cd	--	--
TB	NW China	Cr	As, Cu, Ni	Pb, Sb, Zn	--	Cd
AL	SW China	Cr	--	As, Cu, Ni, Pb, Zn	Sb	Cd
FJ	SW China	--	Cr, Cu, Ni	As, Pb, Zn	--	Cd, Sb
GG	SW China	Cu	As, Ni	Cr, Pb, Sb, Zn	--	Cd
LG	SW China	Cr	--	As, Cu, Ni, Pb, Zn	--	Cd, Sb
LJ	SW China	Cr	Cu, Ni	As, Pb, Zn	Sb	Cd
DH	South China	Cr, Ni, Zn	Pb	Cd	Sb	--
JG	South China	Cr, Cu, Ni	Zn	As, Pb, Sb, Zn	--	Cd
NL	South China	Cr	--	As, Zn	Ni, Pb	Cd, Cu, Sb
SN	South China	Cr	Cu, Ni	As, Pb, Zn	Sb	Cd
SWDS	South China	--	Cr, Ni	As, Cu, Pb, Zn	--	Cd, Sb
WZ	South China	--	Cr	As, Pb, Zn	Sb	Cu, Ni, Cd
DB	East China	As, Cu, Cr, Ni, Pb, Zn	Sb	Cd	--	--
DY	East China	Cu	As, Ni	Cr, Pb, Sb, Zn	--	Cd
JGS	East China	--	--	As, Cr, Cu, Ni, Pb, Zn	--	Cd, Sb
LQ	East China	Cr, Cu, Ni	--	As, Pb, Sb, Zn	--	Cd
TM	East China	--	Cr, Cu, Ni	As, Pb, Sb, Zn	--	Cd
WG	East China	Cu, Ni	--	As, Cr, Pb, Zn	Sb	Cd

AL: Mt. Ailao, AS: Mt. Ao, BCW: Mt. Baicaowa, CB: Mt. Changbai, DB: Mt. Dabie, DH: Mt. Dinghu, DX: Daxinganling, DY: Mt. Daiyun, FJ: Mt. Fajing, GD: Mt. Guandi, GG: Mt. Gongga, HS: Mt. Han, JF: Mt. Jifeng, JG: Mt. Jiugong, JGS: Mt. Jinggang, LG: Mt. Leigong, LJ: Mt. Luoji, LQ: Mt. Longquan, NL: Mt. Nanling, QF: Mt. Qingfengxia, SHB: Mt. Saihanba, SN: Mt. Shennongjia, SWDS:

Mt. Shiwandashan, SYK: Suyukou, TB: Mt. Taibai, TM: Mt. Tianmu, WG: Mt. Wugong, WYZ: Wuyuezhai, WZ: Mt. Wuzhi, XX: Xiaoxinganling

Table 3 The detection of the factors influencing trace element distribution in the mosses

		Forest type	Monsoon type	Altitude	MAP	Industrial production	Coal consumption	Fuel consumption	Smog dust emission
As	q	0.090	**0.154**	0.029	**0.117**	**0.229**	0.022	**0.148**	**0.163**
	p	0.447	**0.000**	0.847	**0.041**	**0.000**	0.837	**0.038**	**0.000**
Cd	q	0.064	**0.237**	**0.188**	**0.196**	**0.364**	0.066	**0.112**	**0.213**
	p	0.441	**0.000**	**0.009**	**0.000**	**0.000**	0.471	**0.043**	**0.000**
Cr	q	0.054	0.080	0.043	**0.150**	**0.117**	0.040	0.046	0.065
	p	0.382	0.012	0.741	**0.000**	**0.014**	0.347	0.299	0.137
Cu	q	0.013	**0.154**	0.035	0.088	**0.204**	**0.448**	0.090	**0.449**
	p	0.836	**0.004**	0.685	0.048	**0.004**	**0.000**	0.064	**0.000**
Ni	q	0.012	**0.132**	0.033	0.096	**0.196**	**0.434**	0.067	**0.439**
	p	0.883	**0.011**	0.733	0.036	**0.005**	**0.000**	0.125	**0.000**
Pb	q	0.029	**0.176**	0.051	**0.174**	**0.230**	0.016	0.107	**0.192**
	p	0.849	**0.000**	0.511	**0.000**	**0.000**	0.897	0.130	**0.000**
Sb	q	0.107	**0.219**	0.021	**0.263**	**0.267**	0.014	**0.158**	**0.210**
	p	0.192	**0.000**	0.926	**0.000**	**0.000**	0.864	**0.030**	**0.000**
Zn	q	**0.259**	0.011	0.033	0.069	0.118	0.026	0.104	0.071
	p	**0.011**	0.816	0.758	0.082	0.070	0.881	0.066	0.312

Fig. 1 The average concentrations of the trace elements in the mosses of the remote mountains

Fig. 2 The relationships of the trace element concentrations with latitude and longitude

Fig. 3 Linear regressions of the trace elements versus Ti in the mosses of all mountains

Fig. 4 The diagrams of $^{206}Pb/^{207}Pb$ versus $^{208}Pb/^{206}Pb$ in the mosses (squares) and other materials. The isotopic ratios of Pb in the mosses and Chinese ores were compiled based on the administrative region to clearly observe the local contribution to Pb; meanwhile, the ratios in the coal emissions and PMs were also presented considering the spatial difference. Fig.4A shows natural and anthropogenic isotopic ratios of Pb in the mosses and other materials, and Fig. 4B highlights the anthropogenic endmember of Pb. The references for the Pb isotopic data in other materials can be found in the *Supplementary Information*

Fig. 1

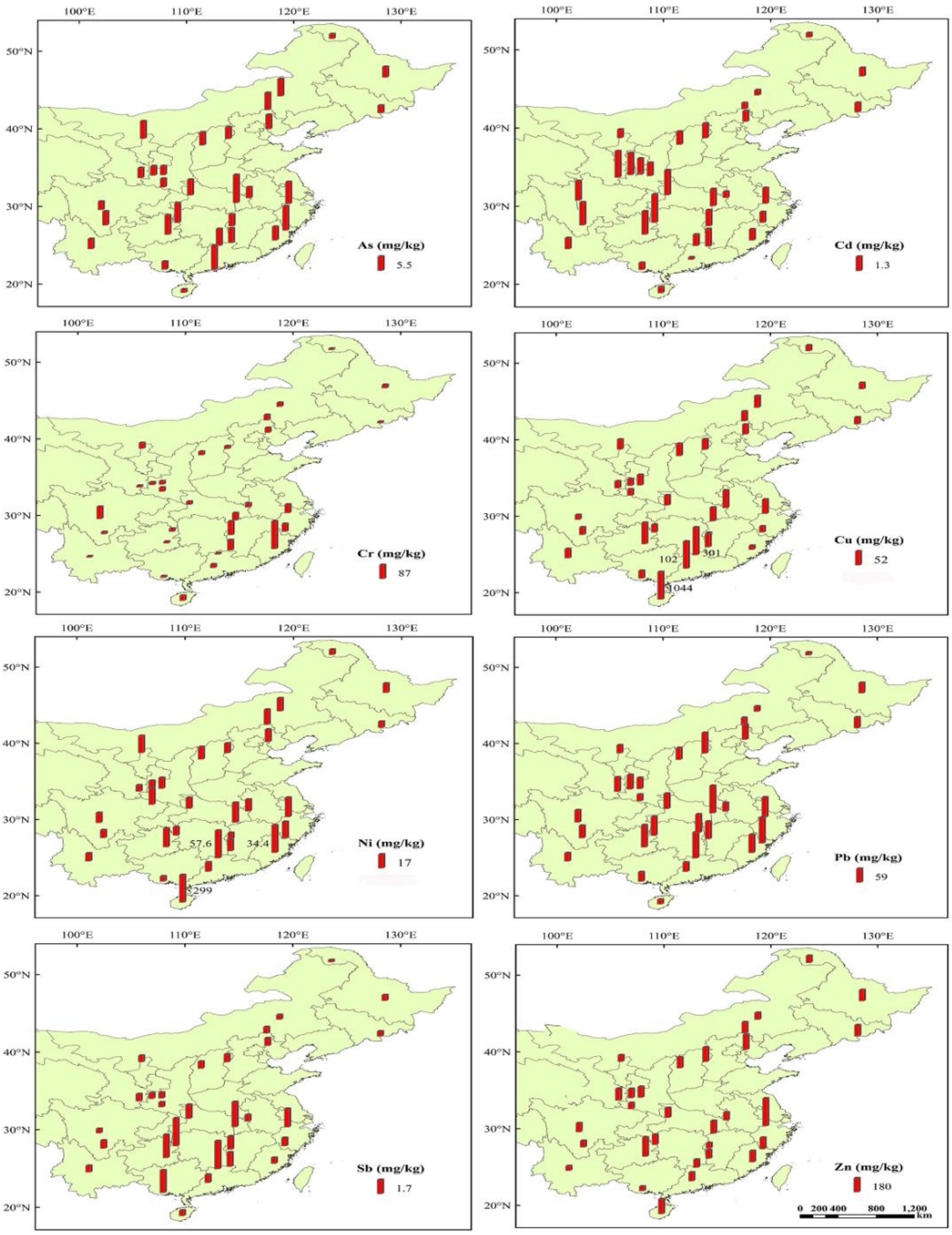

As (mg/kg)
5.5
Cd (mg/kg)
1.3
Cr (mg/kg)
87
Cu (mg/kg)
52
102
301
1044
Ni (mg/kg)
17
57.6
34.4
299
Pb (mg/kg)
59
Sb (mg/kg)
1.7
Zn (mg/kg)
180
0 200 400 800 1,200 km

Fig. 2

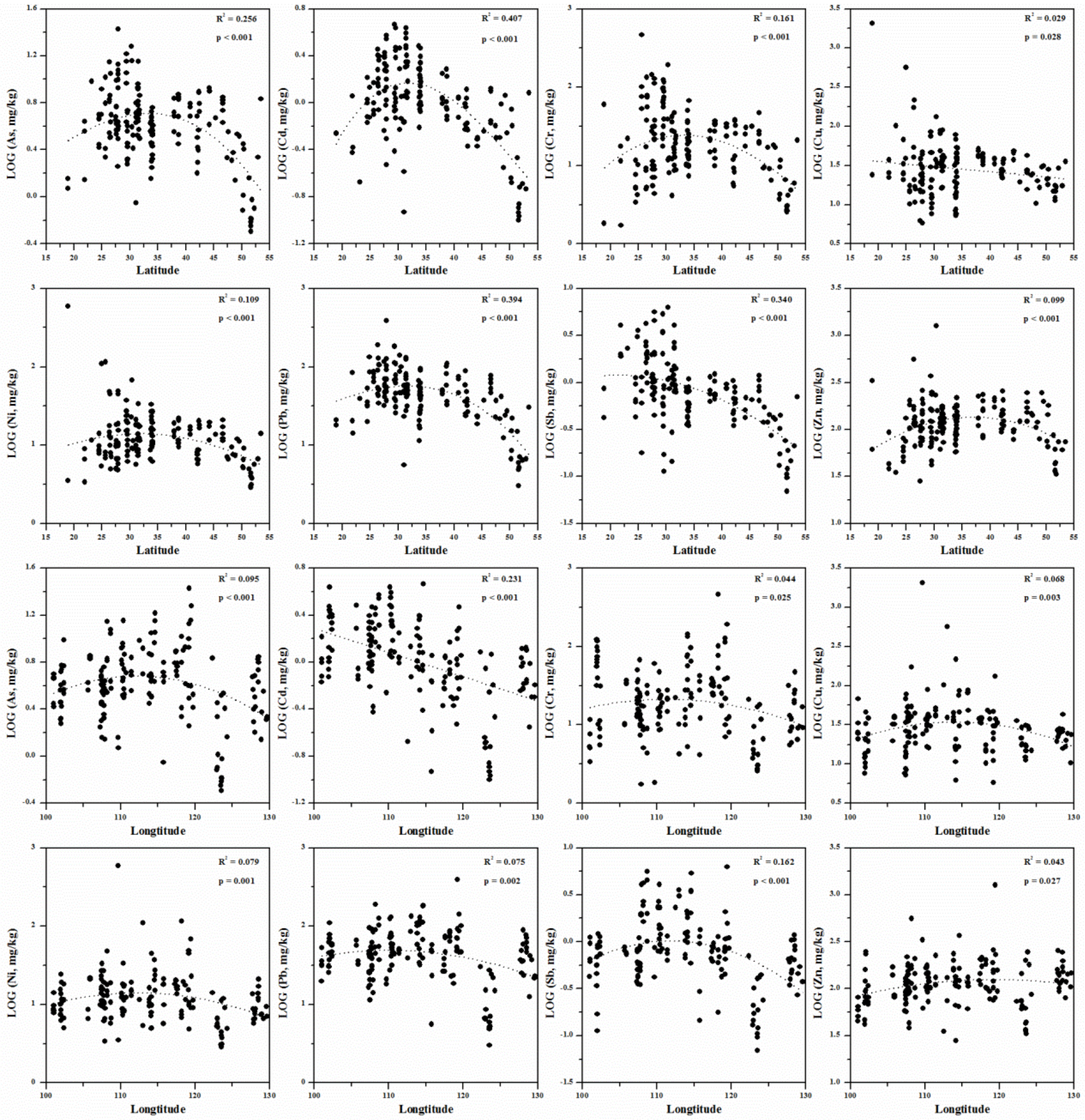

LOG (As, mg/kg)
LOG (Cd, mg/kg)
LOG (Cr, mg/kg)
LOG (Cu, mg/kg)
LOG (Ni, mg/kg)
LOG (Pb, mg/kg)
LOG (Sb, mg/kg)
LOG (Zn, mg/kg)
Latitude
Longtitude
R² = 0.256
p < 0.001
R² = 0.407
p < 0.001
R² = 0.161
p < 0.001
R² = 0.029
p = 0.028
R² = 0.109
p < 0.001
R² = 0.394
p < 0.001
R² = 0.340
p < 0.001
R² = 0.099
p < 0.001
R² = 0.095
p < 0.001
R² = 0.231
p < 0.001
R² = 0.044
p = 0.025
R² = 0.068
p = 0.003
R² = 0.079
p = 0.001
R² = 0.075
p = 0.002
R² = 0.162
p < 0.001
R² = 0.043
p = 0.027

Fig. 3

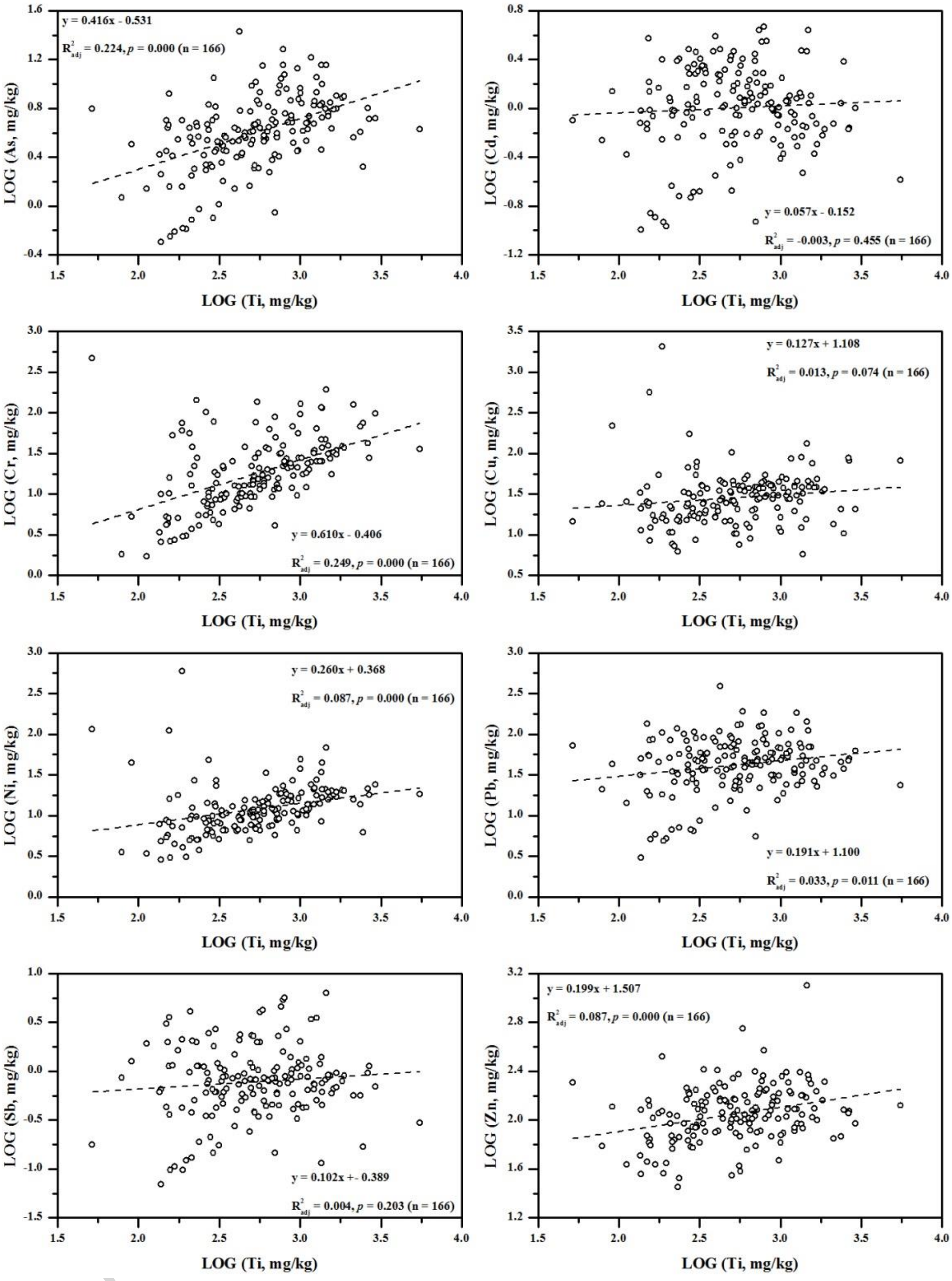
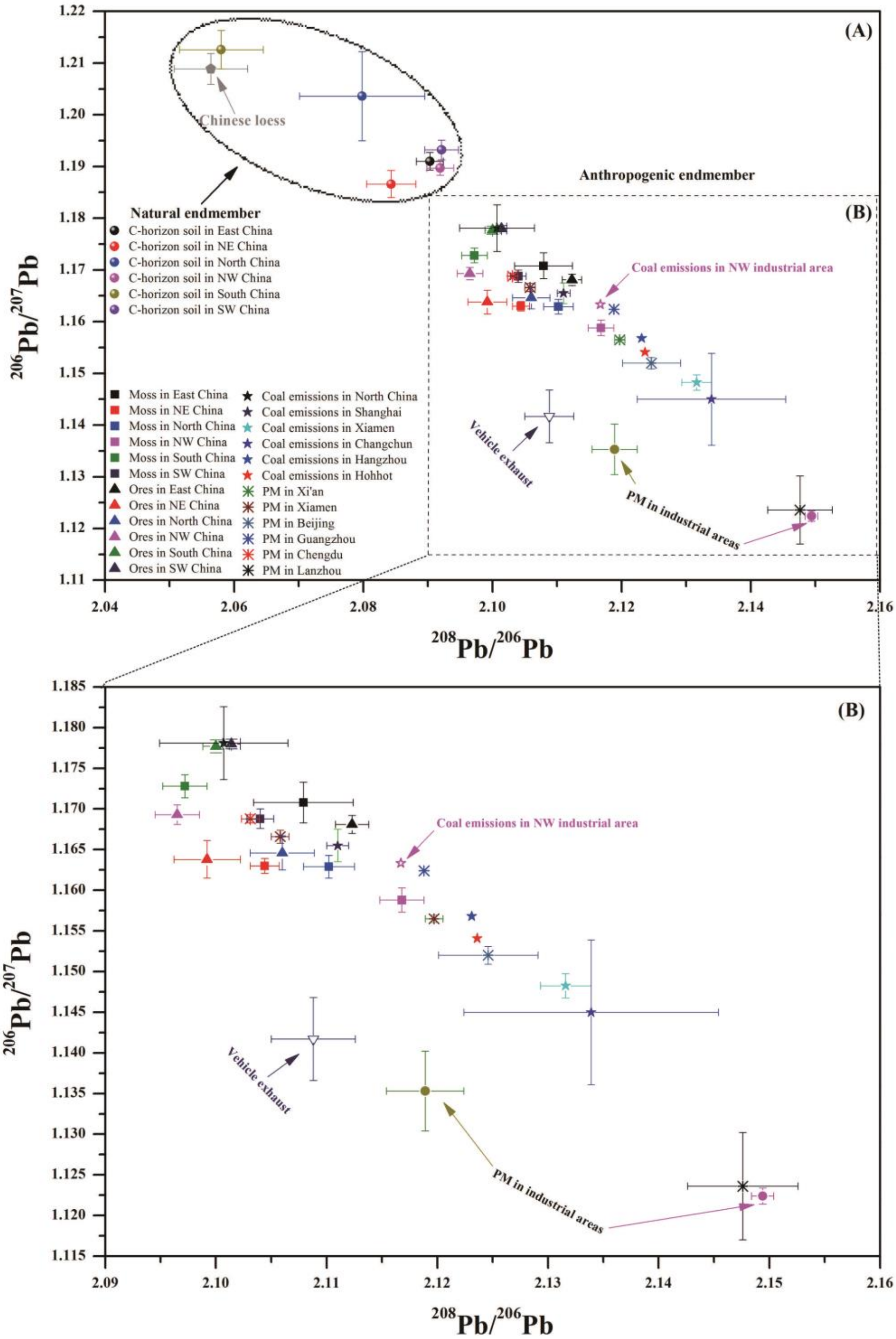

y = 0.416x - 0.531
R²adj = 0.224, p = 0.000 (n = 166)
LOG (As, mg/kg)
LOG (Ti, mg/kg)
y = 0.057x - 0.152
R²adj = -0.003, p = 0.455 (n = 166)
LOG (Cd, mg/kg)
LOG (Ti, mg/kg)
y = 0.610x - 0.406
R²adj = 0.249, p = 0.000 (n = 166)
LOG (Cr, mg/kg)
LOG (Ti, mg/kg)
y = 0.127x + 1.108
R²adj = 0.013, p = 0.074 (n = 166)
LOG (Cu, mg/kg)
LOG (Ti, mg/kg)
y = 0.260x + 0.368
R²adj = 0.087, p = 0.000 (n = 166)
LOG (Ni, mg/kg)
LOG (Ti, mg/kg)
y = 0.191x + 1.100
R²adj = 0.033, p = 0.011 (n = 166)
LOG (Pb, mg/kg)
LOG (Ti, mg/kg)
y = 0.102x + - 0.389
R²adj = 0.004, p = 0.203 (n = 166)
LOG (Sb, mg/kg)
LOG (Ti, mg/kg)
y = 0.199x + 1.507
R²adj = 0.087, p = 0.000 (n = 166)
LOG (Zn, mg/kg)
LOG (Ti, mg/kg)

Fig. 4


(A)
(B)
Chinese loess
Natural endmember
Anthropogenic endmember
C-horizon soil in East China
C-horizon soil in NE China
C-horizon soil in North China
C-horizon soil in NW China
C-horizon soil in South China
C-horizon soil in SW China
Moss in East China
Moss in NE China
Moss in North China
Moss in NW China
Moss in South China
Moss in SW China
Ores in East China
Ores in NE China
Ores in North China
Ores in NW China
Ores in South China
Ores in SW China
Coal emissions in North China
Coal emissions in Shanghai
Coal emissions in Xiamen
Coal emissions in Changchun
Coal emissions in Hangzhou
Coal emissions in Hohhot
PM in Xi'an
PM in Xiamen
PM in Beijing
PM in Guangzhou
PM in Chengdu
PM in Lanzhou
Coal emissions in NW industrial area
Vehicle exhaust
PM in industrial areas
$^{206}Pb/^{207}Pb$
$^{208}Pb/^{206}Pb$

Highlights

- Investigation of trace elements in mosses from thirty China's remote mountains
- Relatively high contamination of mosses by As, Cd, Pb, Sb and Zn
- Hotspots of trace element contamination in eastern, southern and southwestern China
- Mining, fuel combustion and industrial emissions as main anthropogenic sources
- Monsoon regulating the transport and deposition of anthropogenic elements in mosses