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# Continental-scale geochemical survey of lead (Pb) in mainland China's pedosphere: Concentration, spatial distribution and influences



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#### ABSTRACT

The China Geochemical Baseline (CGB) project (2008–2012) obtained comparable, continental-scale, geochemical data including lead (Pb) concentrations in the pedosphere. Some 3382 topsoil samples were collected at depths of 0–25 cm. Lead concentrations in soil were determined by inductively coupled plasma mass spectrometry (ICP-MS) using 4-acid extraction under strict quality control. The median Pb concentration throughout China was 22.1 mg/kg and varied between 15.6 and 30 mg/kg according to the morphological landscape. There were predominantly low Pb concentrations in northwestern China and much higher concentrations in southeastern China. This is mainly due to differences in climate and landscape, geology (parent rocks) and the presence of mineralization. The arid and semi-arid landforms in northern and northwestern China had low Pb concentrations. High Pb concentrations were most often linked to geology (occurrence of granite) and climate. The most important Pb anomalies occurred in the southern and southwestern parts of China in areas with known polymetallic mineralization. Such anomalies may be further enhanced by mining or smelting activities.

#### 1. Introduction

Lead (Pb) is a potentially toxic element that is part of a suite of elements often incorrectly called 'heavy metals' (Duffus, 2002; Chapman, 2007) and has received particular attention in recent decades (Reimann et al., 2012a; Zhu et al., 2013a, b; Bi et al., 2015a, b; Marx et al., 2016; Fabian et al., 2017; Kushwaha et al., 2018). Exposure to Pb can cause serious toxic reactions (Chiodo et al., 2007). According to Needleman (2004), 0.6% of the world's disease burden is caused by Pb pollution, and the World Health Organization (WHO, 2016) reported that, in 2013, there were 853,000 Pb-related deaths worldwide. The situation is more serious in developing countries than developed ones (WHO, 2016). As the largest developing country, and one that generates large Pb emissions, China should pay more attention to the increased Pb levels in blood (Han et al., 2018), which are of particular concern for children (Li et al., 2012). Many studies have demonstrated that there is a significant correlation between Pb concentrations in blood and those in soil or sediments (Ren et al., 2006; Mielke et al., 2007; Laidlaw and Taylor, 2011). To manage and prevent Pb poisoning, it is vital to know the concentration and spatial distribution of Pb in the pedosphere

(Mielke et al., 2011). However, very little research has addressed Pb pollution in China at the national level.

The pedosphere is the outermost layer of the Earth that is composed of soil and subject to soil formation processes. Humans have investigated the concentration and spatial distribution of elements in the pedosphere for the past 100 years. To date, average concentrations of Pb in the upper crust have been extensively studied (Clarke, 1889; Clarke and Washington, 1924; Goldschmidt, 1954; Taylor, 1964; Shaw et al., 1986; Wedepohl, 1995; Rudnick and Gao, 2003). However, little is known of its spatial distribution in the Earth's surface or near-surface environments (Wang et al., 2016). Therefore, Darnley et al. (1995) proposed the Global Geochemical Baselines project to obtain comparable continental-scale geochemical data across national borders (Reimann et al., 2012b). China developed the leadership and funding to take this project forward (Xie and Cheng, 2001; Smith et al., 2012; Yao et al., 2014). The China Geochemical Baseline (CGB) project was initiated from 2008 to 2012 and covers all of China (Wang, 2012; Wang et al., 2015a). In the CGB project, catchment sediments or alluvial soils were selected as the best sampling media for representing the distribution of elements in continental-scale geochemical surveys (Xie and

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Cheng, 1997; Salminen, 2005; Caritat et al., 2009, 2018; Wang et al., 2015a).

Landscape, geological background (parent rocks), quaternary materials, vegetation cover, climate (especially precipitation) and, last but not least, human activities, such as land use, population and traffic density, and the distribution of major industries, all need to be considered when interpreting geochemical maps at the continental scale (Reimann et al., 2012b). To provide all this information is beyond the scope of this paper. This study uses data from the CGB project to 1) present the concentration and spatial distribution of Pb in the Chinese pedosphere, and 2) investigate influences such as geological background, landscape, climate, and mineralization.

#### 2. Materials and methods

# 2.1. Sample collection

Selecting the optimal sampling media is a critical problem in establishing geochemical baselines over very large areas, in this case, 9.6 million km<sup>2</sup>. In the CGB project, catchment sediments and alluvial soils were selected because they can provide high-resolution and comparable baseline data nationwide (Xie and Cheng, 1997; Salminen, 2005; Caritat et al., 2009, 2018; Wang et al., 2015a). The sampling methodology was developed for China's diverse terrain, which includes mountains, hills, plains, deserts, grassland, loess and karst terrain. Specifically, the sampling media were: 1) floodplain sediment or alluvial soil from plains and hilly terrain near exorheic rivers in eastern China, 2) overbank sediment from exorheic river systems in mountainous terrain in southwestern China, and 3) catchment basin and lake sediments from desert and semi-desert terrains, respectively. Sampling was conducted from 2008 to 2012. Two sampling sites were allocated to each CGB grid cell of 1° (longitude) × 40' (latitude), approximately equal to  $80 \times 80 \, \mathrm{km}^2$  in size. Top samples are generally collected from the A-horizon at depths of 0-25 cm. Each sample is composited from three samples collected at the vertex of an equilateral triangle with 50 m on a side. The weight of each sample was about 5 kg. The number of field duplicate samples exceeded 3% of the total number of samples. In total, 3382 samples were collected from 1500 CGB grid cells at an average density of 1 per 3000 km<sup>2</sup> across mainland China (See Fig. 1).

Details of the sampling media, site distribution, and quality control are provided in our previous work (Wang et al., 2015a).

# 2.2. Laboratory analysis and data quality control

All samples were air-dried and sieved through a 2 mm nylon mesh screen in the laboratory. Samples were subsequently ground to  $<74\,\mu m$  (200 mesh) in an agate mill for analysis. Sample analysis was conducted in the IGGE laboratory (Institute of Geophysical and Geochemical Exploration).

A 0.250 g aliquot was weighed and placed into a 25 ml test tube to which 10 ml HF, 5 ml HNO $_3$ , 2 ml HClO $_4$  and 8 ml of aqua regia were added. The test tube was heated in a boiling water bath for 1 h and shaken once during the course of decomposition. After cooling, 1 ml supernatant was diluted with HNO $_3$  solution (2%) to 10 ml. Then, the solution was used to determine the total Pb concentration by inductively coupled plasma mass spectrometry (ICP-MS). The detection limit was 2 mg/kg and the reportable rate was as high as 100%.

The standardized quality control (QC) procedures consisted of 1) field training for all sampling participants; 2) field sampling checking by random selection of more than 5% of the sampling sites; 3) collection of 3% field duplicate samples; 4) blank insertion of 10% laboratory replicate samples; and 5) insertion of four standard reference materials into each batch of 50 routine samples (Caritat et al., 2018). The passing percentage rates of national standard reference materials, laboratory replicate samples and field duplicates were 100%, 99.9%, and 99.5%, respectively. The Pb analytical scheme and quality monitoring system for the CGB project are described in detail elsewhere ( Zhang et al., 2012; Wang et al., 2015a). In summary, the CGB project obtained high-quality Pb data.

# 2.3. Data analysis and mapping

Geochemical maps of Pb were drawn up using Geoexpl2012 software, which was developed by the Development and Research Center of the China Geological Survey. Raw analytical data were interpolated to generate a regular output grid of  $80 \times 80 \, \mathrm{km}$  using an exponentially-weighted moving average model (Wang et al., 2015). Eighteen-shade color map classes were created according to the following percentile Pb



Fig. 1. Map of CGB project sampling sites.

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Table 1
Percentiles of Pb concentrations (mg/kg) found in top sediment/alluvial soil samples from the CGB project.

N	Min	P2.5	P25	P50	P75	P85	P97.5	Max	AM	GM	STD
3382	< 2.0	11.8	17.6	22.1	28.2	34.4	72.1	1386	29.3	23.6	48.7

Notation: N = number of samples; P = percentile (P50 = median); Min = minimum; Max = maximum; AM = arithmetical mean; GM = geometric mean; STD = standard deviation.

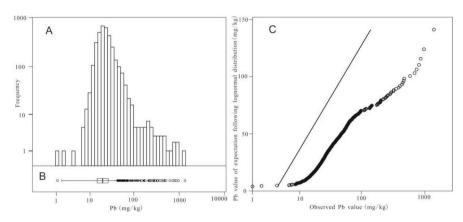


Fig. 2. Histograms (A), boxplot(B), and Q-Q plot (C) displaying the distribution of Pb concentrations (mg/kg) in top sediment/alluvial soil samples of the CGB project.

concentrations: P2.5, P5, P10, P15, P20, P25, P30, P40, P50, P60, P70, P75, P80, P85, P90, P95, and P97.5, which are equivalent to 11.8, 13.0, 14.6, 15.6, 16.7, 17.6, 18.5, 20.2, 22.1, 24.1, 26.6, 28.2, 30.9, 34.4, 40.6, 54.6, and 72.1 mg/kg.

The data was statistically analyzed using the statistical package SPSS v17.0 (SPSS Inc.). Correlation analysis between Pb and  $Al_2O_3$ , CaO, MgO,  $Na_2O$ ,  $K_2O$ ,  $Fe_2O_3$ , MnO concentration was carried out without the outliers of Pb concentration (lower than P2.5 and higher than P97.5). The database of the major elements ( $Al_2O_3$ , CaO, MgO,  $Na_2O$ ,  $K_2O$ ,  $Fe_2O_3$ , and MnO) is available at http://www.globalgeochemistry.com/en/index.php.

# 3. Results

# 3.1. General abundance of Pb in China

Summary statistics of Pb concentrations determined by the CGB project are shown in Table 1. Pb concentrations ranged from < 2 to 1386 mg/kg, with a median of 22.1 mg/kg. The average concentration was 29.3 mg/kg (95% confidence interval, CI: [27.7, 30.9] mg/kg). The 95% range (P2.5–P97.5) of Pb concentrations was 11.8–72.1 mg/kg. The median Pb concentration is comparable to that found by the Environmental Geochemical Monitoring Networks (EGMON) project (23 mg/kg, Xie and Cheng, 2001), which was a continental-scale geochemical baseline study conducted in 1994–1996 (Cheng et al., 1997).

The histograms show approximately lognormal distributions (Fig. 2), which suggests that Pb concentrations may be mainly related to natural factors and only slightly influenced by human activities. The

geometric mean value of Pb in this study was 23.6 mg/kg, which is just a little lower than the background value reported for Chinese topsoil (26 mg/kg; CNEMC, 1990) and that reported for Chinese stream sediments (27 mg/kg; Chi and Yan, 2007). There were 15.21% samples with Pb concentrations higher than the Grade I Chinese Soil Guideline (35 mg/kg), while 2.16% exceeded the Grade II Chinese Soil Guideline for agricultural soils (80 mg/kg).

The Pb concentrations reported in various loose sediments in China are summarized in Table 2. They are ranked as follows: agricultural soil (25.6 mg/kg, data from the First National Soil Pollution Survey, Chen et al., 2015) > topsoil (23.5 mg/kg, CNEMC, 1990) > vurban soils of 31 metropolises (23.4 mg/kg, Cheng et al., 2014) > floodplain sediments (23mg/kg, data from EGMON project, Xie and Cheng, 2001).

Pb concentrations in the continental crust and rocks are summarized in Table 3. The Pb concentration in the Chinese pedosphere (29.3 mg/kg, this study) is higher than those in the continental crust, which vary from 8 to 16 mg/kg. This indicates that Pb could be enriched in the soil formation process. In terms of rock types, Pb concentrations in granite are the highest, followed by shale, schist, sandstone, slate, phyllite, gneiss, intermediate rocks, siliceous rocks, basic rocks, carbonate rocks, marble, and ultramafic rocks (Table 3). It could be concluded that the more acidic the rock is, the higher the Pb concentration.

# 3.2. General spatial distribution of Pb and its influences in China

The most noticeable pattern in the Pb distribution shown on the geochemical map of topsoil samples (Fig. 3) is an increasing trend of Pb concentration from the northwest to southeast. Latitude 32° N can be

**Table 2**Pb concentration (mg/kg) in various loose sediments throughout China.

Soil type	Sample size	Parameter	Pb concentration	Reference
Topsoil Floodplain sediments Agricultural Soil Stream sediments Urban soil of 31 metropolises	4095 529 38393 869 1011	M M M AM M	23.5 23 25.6 27 23.4	CNEMC (1990) Xie and Cheng (2001) Chen et al. (2015) Chi and Yan (2007) Cheng et al. (2014)

Notation: AM = arithmetical mean; M = median.

**Table 3**Pb concentration (mg/kg) in continental crust and rocks.

Type		Pb	Reference
Crust	Continental crust	16	Goldschmidt, 1933
	Continental crust	16	Vinogradov (1962)
	Continental crust in eastern China	15	Chi and Yan (2007)
	Continental crust	14.8	Wedepohl (1995)
	Continental crust	12.5	Taylor (1964)
	Continental crust	11	Rudnick and Gao (2003)
	Continental crust	8	Taylor and McLenan, 1985
Rocks	Granites in China	26	Yan and Chi (2005)
	Granites in China	26	Shi et al. (2005)
	Acidic rocks in China	24	Yan and Chi (2005)
	Shales in eastern China	23	Yan and Chi (2005)
	Acidic rocks	20	Vinogradov (1962)
	Schists in eastern China	19	Chi and Yan (2007)
	Sandstones in eastern China	18	Yan and Chi (2005)
	Slates in eastern China	18	Chi and Yan (2007)
	Phyllites in eastern China	18	Chi and Yan (2007)
	Gneisses in eastern China	16	Chi and Yan (2007)
	Intermediate rocks in China	15.5	Chi and Yan (2007)
	Siliceous rocks in eastern China	14	Chi and Yan (2007)
	Basic rocks in China	13	Yan and Chi (2005)
	Carbonate rocks in eastern China	8.7	Yan and Chi (2005)
	Marbles in eastern China	8.6	Chi and Yan (2007)
	Ultramafic rocks in China	8	Yan and Chi (2005)

used to divide mainland China into southern and northern China. The median values of Pb are 30.97 and 19.73 mg/kg in the south and north, respectively, with the Pb-south:Pb-north ratio being 1.57.

Low Pb concentrations (here defined as Pb content < P25; blue regions on map, Fig. 3) are distributed in arid and semi-arid areas (arid desert basins, Gobi Desert, semi-desert grassland and loess) in the northern and northwestern parts of China. Here, the soil is mostly composed of windblown sand and is unpolluted or only slightly polluted by human activities (Wei et al., 1991; Zhou et al., 2015; Wang et al., 2015a).

High Pb concentrations (Pb content > P75; yellow regions on map) can be interpreted as a lithological effect. It will be discussed later in this paper.

Pb anomalies (Pb content > P85; orange and red regions of the map), both in upper and deeper samples, were mainly distributed in the southern and southwestern parts of China. These anomalies were caused by a combination of geological background, climate, and land-scape. They are especially associated with mineralization, such as that of the Gangdise metallogenic belt, the Nujiang-Lancangjiang-Jinshajiang metallogenic belt, the metallogenic province at the southwestern border of the Yangtze Plate, and the southeastern coastal metallogenic belt, which all have many lead-zinc deposits (Hu et al., 2007; Huang et al., 2011; Mao et al., 2002; Zhang et al., 2013).

Pb anomalous centers (Pb content > P97.5; dark red regions of the map) were exactly correlated with polymetallic mineralization or polymetallic mining activities. These occur in areas such as A-H in Fig. 3. Pb concentration in topsoil sample was as high as 809 mg/kg near the Baiyinnuoer Pb—Zn deposit (A in Fig. 3), which is more than 40 times the background concentration of Pb in Inner Mongolia (CNEMC, 1990). This higher concentration is probably due to mining activities. However, it cannot be ignored that some deposits, which mainly located in North China, are only marked by weak anomalies. This is mainly due to the lower geochemical background of Pb in North China.

#### 4. Discussion

# 4.1. Comparison of Pb concentrations in different countries

Pb concentrations vary greatly in different countries and regions. China and Australia, and American and European countries, have all conducted continental-scale geochemical surveys (Caritat et al., 2018), including the EGMON (Cheng et al., 1997) and CGB projects in China (Wang et al., 2015a), the National Geochemical Survey of Australia (NGSA) (Caritat and Cooper, 2011; Reimann and Caritat, 2017), the Soil Geochemical Landscapes (SGL) project in the conterminous United States (Smith et al., 2013), and the Forum of European Geological Surveys (FOREGS) (Salminen, 2005) and Geochemical Mapping of Agricultural Soils (GEMAS) projects in Europe (Reimann et al., 2012a, 2014a,b). Table 4 summarizes comparable data for Pb in loose sediments or soils from these projects (Liu et al., 2015; Caritat et al., 2018). Chi and Yan (2007) reported that the arithmetic mean Pb concentration

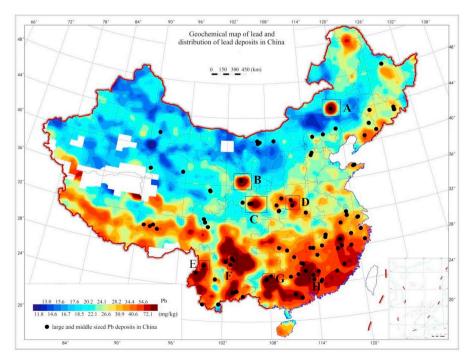


Fig. 3. Distribution of moderate and large Pb deposits in geochemical map of top sediment/alluvial soil samples in China. Baiyinnuoer Pb—Zn deposit in Inner Mongolia (A), Lanzhou-Baiyin Ag—Pb—Zn ore-field in Gansu (B), Fengxian Pb—Zn ore-field in Shannxi (C), Lingbao-Lushi Pb—Zn ore-field in Henan (D), Huize Pb—Zn ore-field (F) and the largest Pb—Zn deposit in Asia- Jinding Pb—Zn ore-field(E) in Yunnan, Hechi Pb—Zn ore-field in Guangxi (G), Fankou Pb—Zn ore-field in Guangdong (H).

Table 4
Total Pb concentrations (mg/kg) in loose sediments of China, Australia, USA, and Europe.

Region (Survey)	Sample type	N	Analytical method	Parameter	Pb (mg/kg)	References
China	Catchment sediments and alluvial soil	3382	ICP-MS (HF + HNO <sub>3</sub> + HClO <sub>4</sub> + aqua regia)	M	22.1	This study
	Floodplain sediments	529	ICP-MS (HF + HNO <sub>3</sub> +HClO <sub>4</sub> +aqua regia)	M Range	23 2.9–201.4	Xie and Cheng, 2001
USA	Soil	4841	ICP-MS (HF + HCl+ HNO <sub>3</sub> +HClO <sub>4</sub> )	AM M Range	25.8 18.1 < 0.5–12400	Smith et al. (2013)
Australia	Top catchment outlet sediments	1315	ICP-MS (HF + HNO <sub>3</sub> )	M range	12.9 0.5–1530	Caritat and Cooper (2011)
Europe (GEMAS)	Agricultural soil	2211	XRF (Fusion)	M range	21 < 3–1309	Reimann et al. (2012b)
Europe (FOREGS)	Topsoil	843	XRF (Fusion)	M AM Range	22.6 32.6 5.3–970	Salminen (2005)
	Floodplain sediments	747		M AM Range	22 54.1 4.0–7080	

Notation: N = number of the samples; AM = arithmetic mean; GM = geometric mean; M = median.

in stream sediments in China was 27 mg/kg, which is lower than that in Europe (38.6 mg/kg, Salminen, 2005). The median concentration of Pb in China (23 mg/kg in the EGMON project and 22.1 mg/kg in the CGB project) is similar with that in Europe (22 mg/kg in the FOREGS project) (Table 4). In Europe, the FORGES project reported floodplain sediments to have similar topsoil Pb concentrations to those in agricultural soils (as reported by the GEMAS project; Salminen, 2005; Reimann et al., 2012a). The median concentrations of Pb in loose sediments or soils in the USA (18.1 mg/kg) and Australia (12.9 mg/kg) are relatively low (see Table 4).

# 4.2. Factors influencing Pb concentration and distribution in China

# 4.2.1. Geomorphologic landscapes

The geomorphologic landscape plays an important role in the supergene geochemical behavior of Pb (Fortescue, 1992; Yin, 1999). The landscape varies significantly across China, as shown in Fig. 4. Hilly or low mountainous areas, alluvial plains, and forests are distributed in eastern and northeastern China. Arid desert terrain, including desert basins, the Gobi Desert, semi-desert grassland and loess plateaus, are concentrated in northern and northwestern China. Southwestern China

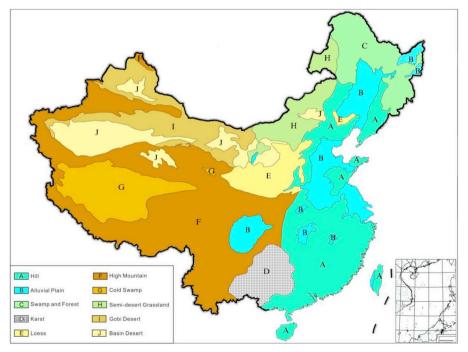
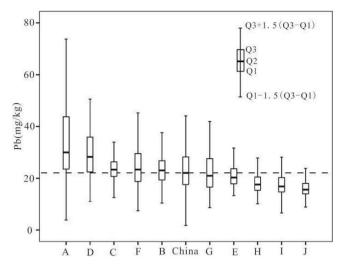


Fig. 4. Geomorphologic landscape terrains in China (after Wang et al., 2015b).



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Fig. 5. Boxplots displaying statistical variation of Pb concentration (mg/kg) in top sediment/alluvial soil samples in different geomorphological terrains of China.

is dominated by high mountains. The Pb statistics for different land-scapes are listed in Fig. 5 and Table 5. The median Pb concentrations of ten surveyed landscapes are very broad, ranging from 15.6 to 30.0 mg/kg, which indicates the influence of the geomorphological landscape on Pb distribution. Karst and hilly terrain have the highest median Pb concentrations among these landscapes in both upper and deeper samples, while desert basins have the lowest. The high Pb values in the karst terrain are due to Pb becoming enriched in red earth and laterite during the process of the chemical decomposition of carbonates (Yin, 1999), and Pb—Zn mineralization in southwestern China where karst terrain occurs. The low Pb values in desert basin terrain are due to the dominance of quartz-rich windblown sand in the samples (Reimann et al., 2012a).

# 4.2.2. Parent materials or geological background

Elemental distribution patterns in continental-scale geochemical maps are mainly determined by natural variation (Reimann et al., 2009). Parent material controls at least 50% of the variability in soil Pb (Yin, 1999; Reimann et al., 2012a; Fabian et al., 2017). Pb is mainly hosted as an isomorphism in the lattice of potash feldspar and biotite, which are rock-forming minerals. In the soil formation process, some potash feldspar and biotite are preserved and some are converted to clay minerals like illite. These minerals have one thing in common—they all contain Al and K. Correlation analysis shows that Pb concentrations were significantly correlated to Al<sub>2</sub>O<sub>3</sub> and K<sub>2</sub>O

**Table 6**Results of correlation analysis ( $R^2$ ) between Pb and Al<sub>2</sub>O<sub>3</sub>, K<sub>2</sub>O, Fe<sub>2</sub>O<sub>3</sub>, MnO concentrations

	$Al_2O_3$	$K_2O$	$Fe_2O_3$	MnO
Pb	0.432**	0.263**	0.306**	0.170**

<sup>\*\*</sup>p < 0.01.

(Table 6), indicating that Pb concentration is determined by the parent materials. The significant positive correlation between the concentration of Pb and  $Fe_2O_3$  and MnO may be because Pb can be adsorbed on the surface of Fe–Mn oxides (Han et al., 2012).

Granites are more enriched in Pb than other rock types (see Table 3). To further study the influence of geological background on Pb concentration, the spatial distribution of the correlation between Pb concentration and granites is shown in Fig. 6. High Pb concentrations match well with the granite distribution outline, especially in the Mesozoic giant granites province in South China, on the concentration map of Pb (see Fig. 6).

#### 4.2.3. Climate or weathering

The CIA is widely used as an indicator of the degree of weathering of source areas (e.g. Kasanzu et al., 2008; Négrel et al., 2015). The weathering history of CGB topsoil samples was evaluated using the chemical index of alteration (CIA), defined by Nesbitt and Young (1982) as CIA =  $[mAl_2O_3/(mAl_2O_3 + mCaO^* + mNa_2O + mK_2O)] \times 100$ . Where m represents molar proportions, and CaO\* silicate-borne CaO. Due to lack of data, this fraction of silicate-borne CaO could not be calculated for both datasets discussed here. Therefore, we observe a certain bias due to Ca bound in carbonates and gypsum at continentalscale. Areas underlain by, or containing, carbonates and gypsum have very low CIA values (Reimann et al., 2012b). The calculated CIA values show that most of the CGB topsoil samples fall within the intermediate and low weathering range (45% of the samples had CIA values of 60-80, 27% had 40-60), and 20% of soils had low CIA values (< 50), while less than 8% of the samples were in the intense weathering range (80–100) (Servaraj and Arthur, 2006). Chemical weathering processes play an important role in elemental transportation and soil formation. Because Ca, Na, and Mg are removed from the soil profile during soil development (Négrel et al., 2015), Pb concentration (varying from P2.5 to P97.5) has a significantly negative correlation with Na2O, CaO, and MgO concentration (Table 7). According to the scatter plot in Fig. 7, Pb concentration (varying from P2.5 to P97.5) is found to have an exponential relationship with CIA. Weathering (soil formation) leads to an increase in Pb concentration in soils. The results suggest that the Pb distribution is related to climate or weathering.

Table 5
Characteristics of Pb concentration (mg/kg) in top sediment/alluvial soil samples in the different landscapes of China.

Landscape	N	Min	P25	P50	P75	P85	Max	AM	GM	STD
Whole China	3382	1	17.6	22.1	28.2	34.4	1385.6	29.3	23.6	48.7
Hill (A)	633	3.8	23.6	30	43.7	54.4	808.7	39.6	32.7	46.2
Karst (D)	126	11.1	22.5	28.3	35.8	41.9	1385.6	53.6	23.1	7.8
Swamp and forest (C)	218	10.7	20.8	23.4	26.3	28.3	147.5	24.6	23.6	10.2
High mountain (F)	923	1	18.8	23.4	29.6	36.1	859.5	33.4	32.2	141
Alluvial plain (B)	335	10.4	19.4	23.1	26.8	29.4	70.3	24.1	21.5	72.8
Cold swamp (G)	140	8.6	16.6	21	27.6	32	290	25.9	25.4	55.7
Loess (E)	170	13.3	17.9	20.3	23.6	26.3	968	27.2	22.3	25.5
Semi-desert grassland (H)	215	10.1	15.4	17.6	20.5	22.3	39.6	18.3	17.8	4.4
Gobi Desert (I)	424	6.6	14.8	16.9	20.3	22.1	35.8	17.6	17.1	4.4
Basin desert (J)	198	1.8	14	15.6	18	19.9	26.2	16.1	15.6	3.8

Notation: N = number of samples; P = percentile (P50 = median); Min = minimum; Max = maximum; AM = arithmetic mean; GM = geometric mean; STD = standard deviation.

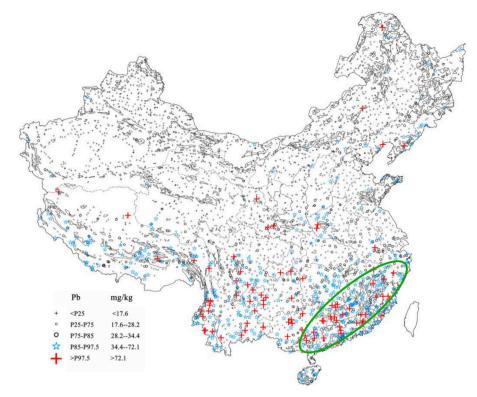


Fig. 6. Map of Pb concentration in top sediment/alluvial soil samples of the CGB project, and granite distribution in China (Green ellipse-Mesozoic giant granite province in South China). (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

Table 7 Results of correlation analysis ( $R^2$ ) between Pb and MgO, Na<sub>2</sub>O, CaO concentrations

	MgO	Na <sub>2</sub> O	CaO
Pb	-0.183**	-0.304**	-0.305**

<sup>\*\*</sup>p < 0.01.

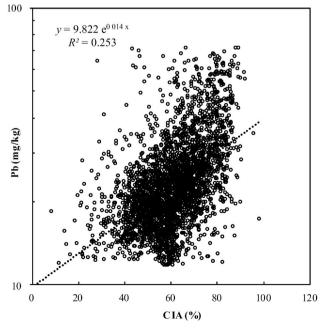


Fig. 7. Scatterplot of Pb concentration vs. CIA.

#### 4.2.4. Pb mineralization

Ore-forming processes commonly affect large areas of the earth's crust, resulting in large-scale geochemical anomalies (Wang et al., 2007). In the European GEMAS project, it was observed that Pb anomalies coincide with known Pb mineral belts or deposits (Reimann et al., 2012a). A similar conclusion was reached in this study: that nearly 80% of the extremely high Pb concentrations (red cross in Fig. 8) fall in the Pb metallogenic belt. This further indicates that most Pb anomalies (dark red color in Fig. 3) are exactly correlated with polymetallic mineralization. Moreover, smelting plants are always located near the deposits, and smelting activities could enhance the pollution level of Pb (Li et al., 2014; Bi et al., 2017).

# 5. Conclusions

This paper provides the first continental-scale geochemical survey of the general abundance and distribution of Pb in the Chinese pedosphere. The 95% range (P2.5-P97.5) of Pb concentrations was 11.8-72.1 mg/kg with a median value of 22.1 mg/kg. The spatial distribution pattern of Pb shows an increasing trend from the northwest to southeast. Low Pb concentrations are distributed in the arid and semiarid landforms of the northern and northwestern parts of China. Weathering (soil formation) leads to an increase in Pb concentration in soils. Landscapes play an important role in the supergene geochemical behavior of Pb. High Pb concentrations match well with the distribution of granite, and Pb anomalies are mainly distributed in southern and southwestern parts of China. These are mainly related to climate and polymetallic mineralization, which is probably enhanced by mining and smelting activities. A significant correlation between Pb and Al<sub>2</sub>O<sub>3</sub>, K2O, Fe2O3, and MnO concentrations indicates that the spatial distribution of Pb is associated with parent rocks. Taken together, these results suggest that the concentration and distribution of Pb are predominantly related to parent materials, landscape, climate, and mineralization.

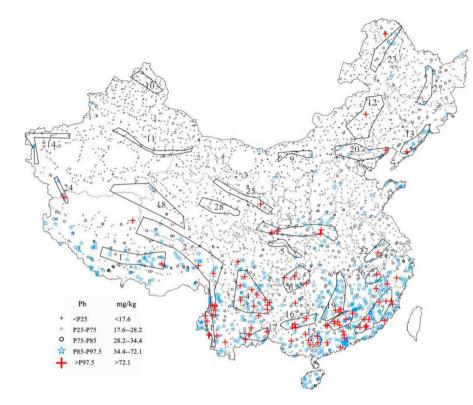


Fig. 8. Map of Pb concentrations in the top sediment/ alluvial soil samples of the CGB project, and the Pb metallogenic belt (Zhang et al., 2013). 1-Gangdise in Tibet, 2-Sanijang area in southwestern China, 3-Oilian-Qinling orogenic belt, 4-junction area of Sichuan, Yunnan and Guizhou, 5-North verge of Yangtze platform, 6-western Hunan and Hubei, 7-southern Fujian and eastern Guangdong, 8-southern Jiangxi and northern Guangdong, 9-Langshan mountain and Zhaertai area, 10-Altay, Xinjiang, 11-East Tianshan, Xinjiang, 12-Tuquan-Ongniud Banner in Daxinganling, 13-southern Jilin and northern Liaoning, 14-Kashi-Hetian in Xinjiang, 15-eastern Guangxi and western Guangdong, 16-Hechi-Rongxian in northern Guangxi, 17-Honghe-Wenshan in southeastern Yunnan, 18eastern Kunlun orogenic belt, 19-eastern Hunan and western Jiangxi, 20-Yanshan-Liaoning area, 21-Changning-Lancang area in Yunnan, 22-middle and lower Yangtze River, 23-Derbugan-Sanhe area in Inner Mongolia, 24-southern Tarim basin, 25-northern Oilian orogenic belt, 26-northern Wuyi mountain, 27-Xiaoxing'an mountains and Zhangguangcai mountains, 28-Qilian orogenic belt.

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#### Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.apgeochem.2018.11.003.

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