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Status of lead accumulation in agricultural soils across China (1979–2016)

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ABSTRACT

The first national-scale assessment of lead (Pb) contamination in agricultural soils across China was conducted based on > 1900 articles published between 1979 and 2016. Pb concentrations, temporal and spatial variations, and influencing factors were analyzed. Children's blood lead levels (BLLs) were also estimated using the integrated exposure uptake biokinetic (IEUBK) model. Pb concentrations in different areas of China varied greatly, which was closely associated with the distribution of Pb-related industries, especially Pb-zinc mine smelting, non-ferrous polymetallic mine smelting, e-waste recycling, and leaded gasoline consumption. The year 2000 was a significant transition year for Pb concentrations, with a rapid increase pre-2000 and a subsequent slow upward trend. Pb concentrations were found to be strongly associated with indicators of economic and social development including gross domestic product (GDP), population size, and vehicle ownership. Leaded gasoline, coal combustion, and non-ferrous smelting were the main sources of atmospheric Pb during the different periods. Predicted BLLs were higher in South China than those in the north. This study details the overall Pb contamination status of agricultural soils in China, and thus provides insights for policymakers with respect to pollution prevention measures.

1. Introduction

Lead (Pb) is considered one of the most hazardous heavy metals due to its toxic effects on the environment and human health (Lee and Tallis, 1973). As a result, Pb pollution has received growing attention in recent decades (K. Chen et al., 2014). Epidemiologic studies have shown that even at low concentrations, chronic exposure to Pb can result in adverse effects on the blood and central nervous system, especially in young children (Lansdown et al., 1974). In 2013, it was estimated that Pb pollution resulted in 0.6% of the world's disease burden (Needleman, 2004) and 853,000 deaths (WHO, 2016).

In the past five decades, 800,000 t of Pb have been released into the environment globally, much of which has accumulated in soil resulting in serious levels of pollution (Chen et al., 2016). As the world's largest developing country, with large emissions of Pb, the Chinese government has given much attention to Pb poisoning. Over the last two decades, with nationwide industrialization and rapid urbanization, environmental pollution and associated health problems in China have become increasingly severe in urban and suburban areas, especially with regard to heavy metal enrichment in different environmental media (Yang et al., 2014). Approximately 82% of contaminated agricultural soils in China contain toxic inorganic pollutants, such as Pb, cadmium (Cd), chromium (Cr), and arsenic (As) (R. Chen et al., 2014). After Cd, Pb is the second-most prevalent heavy metal in the environment based on two recent Chinese government ministry surveys (Zhao et al., 2015). According to the first National Soil Pollution Investigation of China from 2005 to 2013, 1.50% of soil samples were polluted with Pb (Zhao et al., 2015). Direct ingestion of dust and soil in hand-to-mouth activity is an important pathway of Pb exposure to humans, especially for children (Mielke and Reagan, 1998). The average Pb blood levels are higher in Chinese children than in other countries (Z.X. Han et al., 2018). From 2009 to 2011, 10 major Pb poisoning episodes caused by heavily polluted enterprises occurred in China, affecting > 4000 children (Ji et al., 2011). As the largest developing country in the world, understanding of the evolution of Pb contamination in agricultural soils and the critical influencing factors associated with regional economic development is still lacking in China. This is particularly serious when considering the contamination characteristics of Pb in soils and the difficulty of developing effective policies for soil pollution prevention and control. In recent years, review and research articles have provided assessments of Pb pollution in Chinese soils (Chen et al., 2015; Z.X. Han et al., 2018; Yang et al.,

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2018); however, given the breadth of these previous studies, the accumulation of Pb in agricultural soils in China had not yet been examined in depth.

In this study, we conducted systematic statistical analysis on levels of Pb contamination in agricultural soils in China from 1979 to 2016 based on > 1900 articles according to the latest standard (GB15618-2018). Specifically, we studied the response of Pb soil contamination to the rapid economic and social development in China, by considering the GDP, population size, and vehicle ownership. To the best of our knowledge, this review is the first to systematically investigate Pb contamination in agricultural soils at a national scale in China.

2. Data collection and sources

Data were collected from the Web of Science, Elsevier Science Direct, Science Online, Chinese periodical full texts, and China national knowledge infrastructure (CNKI) databases. In total, 1901 papers published between 1979 and 2016 were collected using the keywords 'Pb', 'agricultural soil', and 'China'. In addition, the following eligibility criteria were applied to these papers. Studies focusing on specific areas that have severe Pb pollution caused by accidental events were excluded. For example, literature irrelevant to the scope of this study (Wang et al., 2011), that did not identify the agricultural soil (Xu et al., 2016), which described research conducted in special geographical location (Liu et al., 2013), and with sampling sites that were not clearly defined (Song et al., 2009) were excluded. Most of the resulting studies that were retained for our analysis described data for soils collected from depths of 0–20 cm. The sampling and processing methods used in these studies are all widely accepted by the scientific community. Preservation and transportation of the soil samples were executed according to the Technical Specification for SEPAC (State Environmental Protection Agency of China) (2004). Across the studies, various methods were used to measure heavy metal concentrations in their respective soil samples, but atomic absorption spectroscopy (AAS) and inductively coupled plasma-mass spectrometry (ICP-MS) were most commonly used. Importantly, strict quality control had to have been used in the analysis procedure for a study to include in our sample pool.

Data on soil Pb concentrations from 1300 sampling sites were obtained for agricultural regions distributed across 30 provinces throughout China (Fig. 1) including 22 provinces, four autonomous regions, and four municipalities. Each of the reporting studies investigated only one or a few sites that were suspected of being contaminated. By collating this existing research, our study provides a relatively comprehensive and national-scale assessment of Pb contamination of agricultural soils. As shown in Fig. 1, more sites in the eastern and central regions of China were identified than in the western region, which implies that economically developed regions have been the focus of more intensive research on Pb soil pollution.

3. Results and discussion

3.1. Concentrations and distribution of Pb

Relatively less Pb data were identified pre-2000, implying that concerns have been rising in China over the issue of Pb contamination in soils over the past few decades. Based on this, we divided the available Pb data into four periods: 1979–2000, 2001–2005, 2006–2010, and 2011–2016 (Fig. 2). A similar method of temporal division has been well established in previous research (Z.X. Han et al., 2018).

As shown in Fig. 2a, the mean Pb concentrations in agricultural soil showed a slow but increasing trend after 2000. Pb concentrations peaked (90.58 mg/kg) during the period 2006–2010. Based on mass balance theory, differences in the input and output of heavy metals in agricultural soils will influence the dynamic balance of heavy metals over time (Yi et al., 2018). In recent years the Chinese government has made significant efforts to address soil contamination by implementing a series of measures to cut-off sources of pollution and avoid further pollution. This includes: the launch of the first national soil quality and pollution survey between April 2006 to December 2013 backed by a budget of approximately $125 million; enforcing a new "Environment Protection Law" on January 1, 2015; and issuing an Action Plan on Soil Pollution Prevention and Control (APSPPC) on May 31, 2016. This "war" on soil pollution in China has proven effective; currently, the Chinese government plans to close approximately 500 industrial sites involved in soil pollution and aims to invest 3% of the total economic output of Shanghai (approximately 2220 billion RMB) to address the problem. This suggests there is strong potential for increasing the rate of remediation and establishing strong technical capabilities and delivery to address soil pollution in China (Ken Research, 2015). Generally, heavy metals persist in soils for a long time after their introduction. Therefore, the output of heavy metals from soils is relatively stable over time. Thus, a decrease in inputs and relatively constant outputs may be responsible for the observed reduction in Pb concentrations during the period 2011–2016 (Belon et al., 2012).

Considering regional differences, we classified the agricultural soils in China into five regions (the South China Sea and Macao were not evaluated due to limited data) (Fig. S1). A clear geographical distribution in Pb concentrations was apparent, with soil Pb concentrations in South China higher than other regions (Fig. 2b). Mean Pb concentrations in all the five regions were higher than the national soil background value for China (25.56 mg/kg) (Yang et al., 2018), indicating that Pb has been introduced into agricultural soils from exterior sources associated with anthropogenic activities. The mean soil pH value in major Chinese croplands (Guo et al., 2010) is corresponded to the Environmental quality standard (GB15618-2018) for soils with pH range between 5.5 and 6.5. However, except for South China, the mean Pb concentrations were lower than the risk control value (500 mg/kg) and the risk screening value (90 mg/kg) of the soil environmental quality standard for China (GB15618-2018). It is noteworthy that Pb concentrations in some sites did exceed the risk control value (500 mg/kg) in East, South, and West China, which indicates priority areas for control. For example, Pb concentrations ranged from 14.3 mg/kg to 2000 mg/kg in soil samples from five battery plants in East China (Jin et al., 2015). In Hunan Province, South China, soil located 4 km from a smelter was severely contaminated, with maximum concentrations of Pb as high as 1200 mg/kg in vegetable plots and paddy fields (Li et al., 2011). Soil samples from a cornfield near smelting sites in Hezhang County, Guizhou, West China, contained extremely high levels of Pb (60–14,000 mg/kg) (Bi et al., 2006). Yang et al. (2018) found that high Pb concentrations across industrial and agricultural regions in South China, such as in Guangdong and Guangxi Province. South China has become one of the most rapidly developing regions in China over the past few decades with industrial and agricultural activities, municipal development, and widespread use of chemicals being associated with serious pollution problems in this region (Wang et al., 2013).

The mean Pb concentrations reported in agricultural soils in the USA (Holmgren, 1993), Europe (Reimann et al., 2012), Poland (Piotrowska et al., 1994), and Iran (Ahmadi et al., 2017) are 12.3 mg/kg, 16 mg/kg (median value), 16.4 mg/kg, and 6.124 mg/kg, respectively. The United States Environmental Protection Agency (EPA) standard for Pb in bare soil is 400 mg/kg in playground areas and 1200 mg/kg for non-playground areas (USEPA, 2000). According to these standards, the levels of Pb contamination in China are comparatively lower; approximately 1.04% and 0.5% of the sites identified in South China and in West China, respectively, had Pb concentrations above the EPA standards. The reported Pb concentrations in Northeast, East, and North China do not exceed these limits.

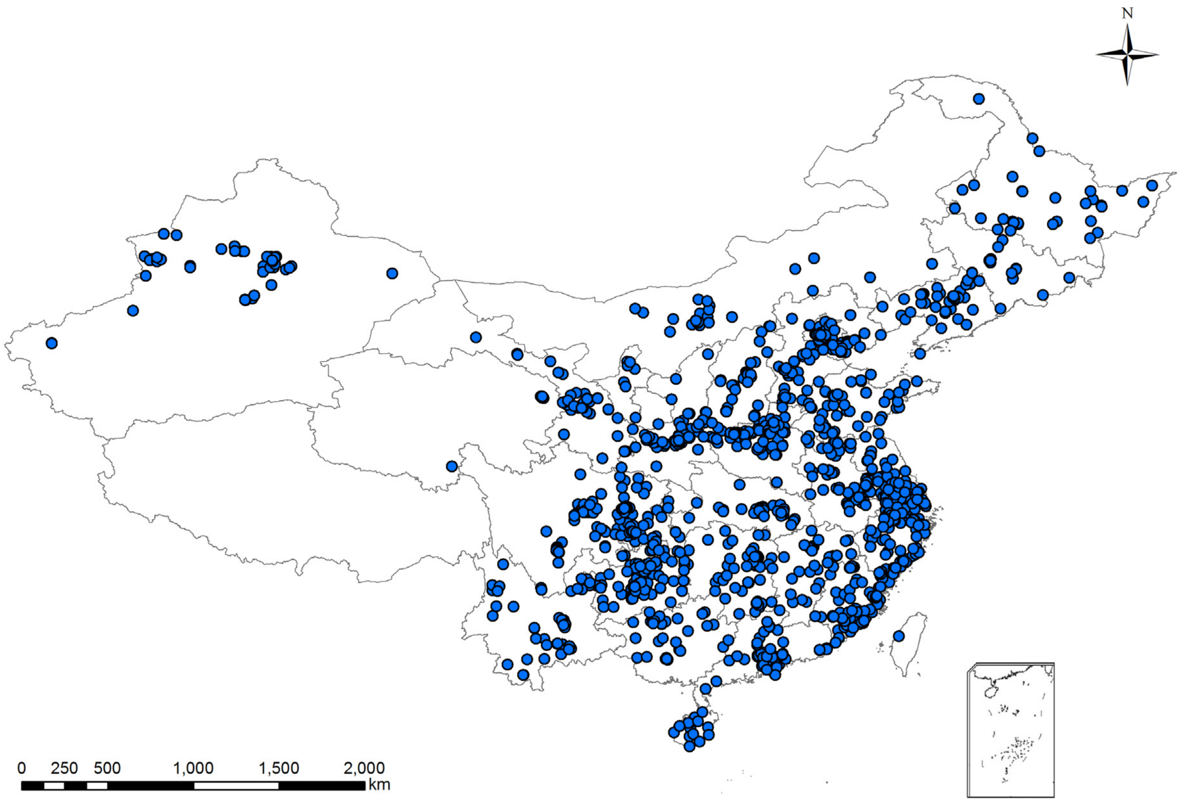

Fig. 1. Areas with studies of Pb occurrence in agricultural soils in China selected for this analysis.

3.2. Geographical variation from 1979 to 2016

The mean Pb concentrations in soils across China during the four studied time periods (1979–2000, 2001–2005, 2006–2010, and 2011–2016) are presented in Figs. 3 and S3.

Pb concentrations in agricultural soils were generally low during the period 1979–2000, ranging from 1.01 mg/kg to 670.5 mg/kg. Hunan Province has experienced the most contamination since 1979, which may partially explain the high blood levels of Pb in this region (Qiu et al., 2015). In 2001–2005, high Pb concentrations were mainly distributed in the regions of Liaoning, Gansu, Yunnan, Zhejiang, and Hunan Province. High soil Pb concentrations found in these provinces correspond with the locations of major Pb-zinc smelting plants in Huludao in Liaoning Province, Hui County in Gansu Province, and Wenzhou in Zhejiang Province, as well as extensive non-ferrous metal smelting in Yunnan (Li et al., 2012). Mining and related activities (e.g., metal processing and smelting) discharge large amounts of wastewater, flue gas, and solid waste to the environment, which often contain Pb (Wen et al., 2015). Thus, Pb enters into soils through atmospheric diffusion, surface runoff flushing, and weathering (Ettler et al., 2008). During the two most recent periods (2006–2010 and 2011–2016), high Pb concentrations have generally occurred in the southeastern hills, such as the Yunnan-Guizhou Plateau and the southern part of the Yangtze River. This is mainly due to the rapid development of Pb-

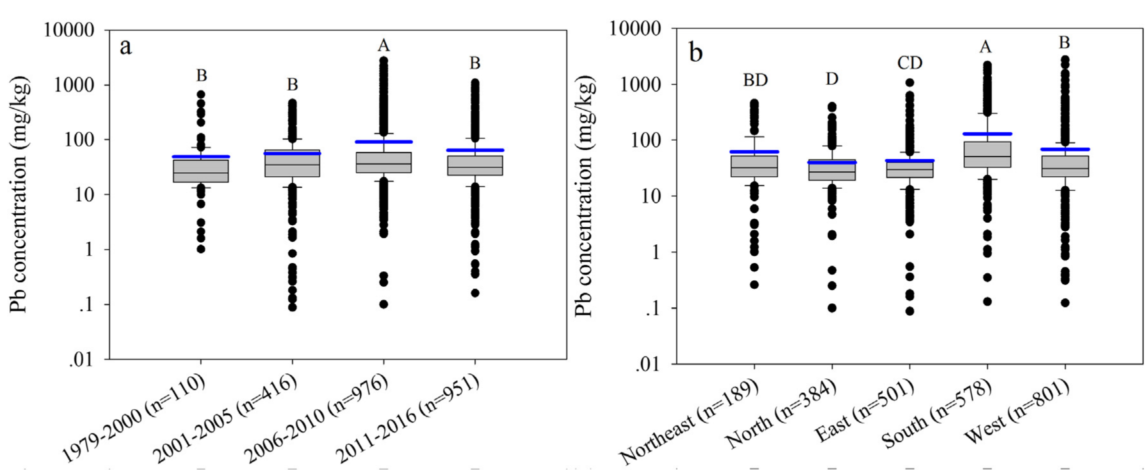

Fig. 2. Mean Pb concentrations in agricultural soils measured during different periods (a) and in different regions (b) in China. The black horizontal line represents the median and the blue horizontal line represents the mean. The boxes represent the interquartile range and the whiskers represent the 10th–90th percentiles. The number of samples in each case is shown in parentheses. Distinct capital letters indicate differences among time periods or regions ($P < 0.05$). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

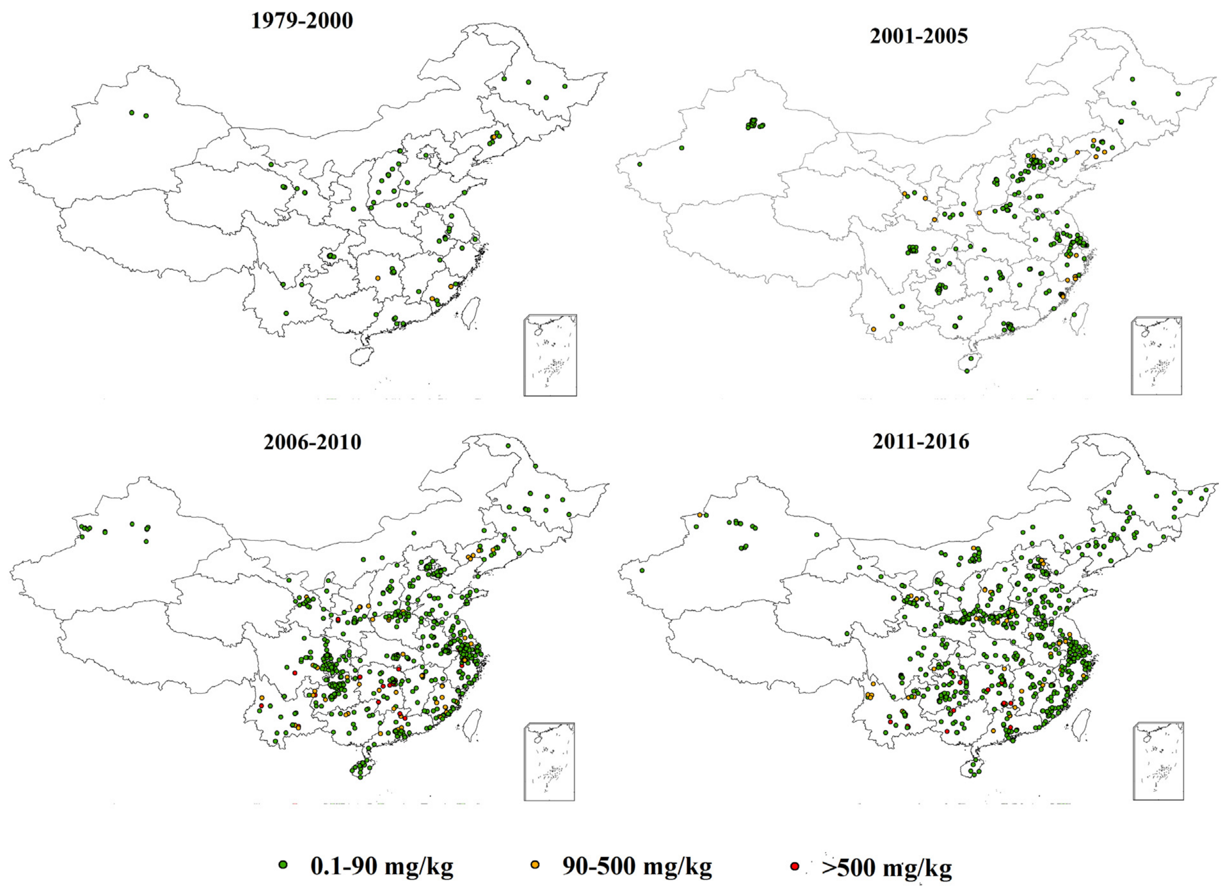

Fig. 3. Spatial distribution of mean Pb concentrations in China from 1979 to 2016.

related industries including Pb smelting, Pb-acid battery production and recycling, and wire rope production. Potential industrial sources of Pb include the Youxi Pb-zinc mine in Fujian, the Qixiashan Pb-zinc-silver polymetallic mine in Jiangsu, the Tongling non-ferrous polymetallic mine in Anhui, the Changqing Pb-zinc smelting plant in Baoji, and the Huanjiang Pb-zinc mine in Guangxi.

Fig. S2 shows the distribution of Pb-zinc ore resources across China (Zhang et al., 2016). It can be seen that most mining areas are located in the southern regions of China, which is consistent with the patterns of mineral exploitation. The linkage between mining activities and Pb concentrations in soils across China is, therefore, evident. This concurs with a previous study by Li et al. (2014), which stated that the I_{geo} values for Pb and Zn are higher than the I_{geo} values of other heavy metals in Pb−Zn mining areas (Li et al., 2014). Moreover, the coastal regions of Fujian and Guangdong Province are both economically developed areas, with high population densities and many factories. Electronic waste recycling activities have also been found to contribute to Pb accumulation in soils in China. Taizhou in Zhejiang is currently dominated by e-waste recycling businesses, on a scale similar to that of Guiyu in Guangdong (Shen et al., 2008). In the Pearl River Delta and the Yangtze River Delta, industrial wastewater discharge is a major source of Pb contamination (Z.X. Han et al., 2018). Concentrations of Pb in northern China were generally low, which is consistent with the findings of the first national soil pollution survey (Chen et al., 2016). It is well known that land use patterns, the intensity of human activity, pollution history, and the distance from emission sources lead to varying degrees of soil pollution. Provinces such as Heilongjiang, Jilin, Xinjiang, and Gansu in northeast and northwestern regions emit far lower amounts of heavy metals because of a lower coal-fired power plant capacity, a lower coal consumption rate by industrial sector, and fewer inhabitants (Tian et al., 2012).

As shown in Fig. 3, Pb concentrations in most regions have been increasing since 1979. Since the beginning of the Reform and Opening-Up program in the 1970s, the economic situation in China has changed dramatically. Along with the industrialization and urbanization of cities undergoing rapid economic development, soil pollution by heavy metals has been both serious and widespread in China. It is clear that numerous studies have been conducted in China on the issue of Pb contamination in soils after the reform in the 1970s. This reflects that over the past few decades, concern has been growing with respect to the issue of Pb pollution in China.

3.3. Temporal variation from 1979 to 2016

Fig. 4 shows Pb concentrations in agricultural soils during the period 1979–2016. The observed temporal trend can be divided into two stages: rapid increase from 1979 to 2000 that followed a trend for rapid industrial development and economic growth, and a subsequent slow upward trend from 2001 to 2016 (Fig. 4). The rate at which Pb concentrations have increased gradually decline after 2000, which may reflect the phasing-out of leaded gasoline in China in the 1990s—and particularly from 2000 (Xu et al., 2012; Bi et al., 2017). For example, previous studies have indicated that restricting the use of leaded gasoline can substantially reduce the emission of Pb into the atmosphere (Miller and Friedland, 1994). In China, total Pb emissions decreased from 13,700 t in 1990 to 9600 t in 2009 (Li et al., 2012). Economic development and good hygiene practices have also contributed to this observed reduction. Because of its substitution by cleaner fuels, such as natural gas and liquefied petroleum gas, especially in urban areas, the use of coal in the residential sector has decreased during the past few decades, resulting in direct fall in the emission of Pb (Tian et al., 2012). In 2008, emissions of Pb from residential coal use were estimated to be

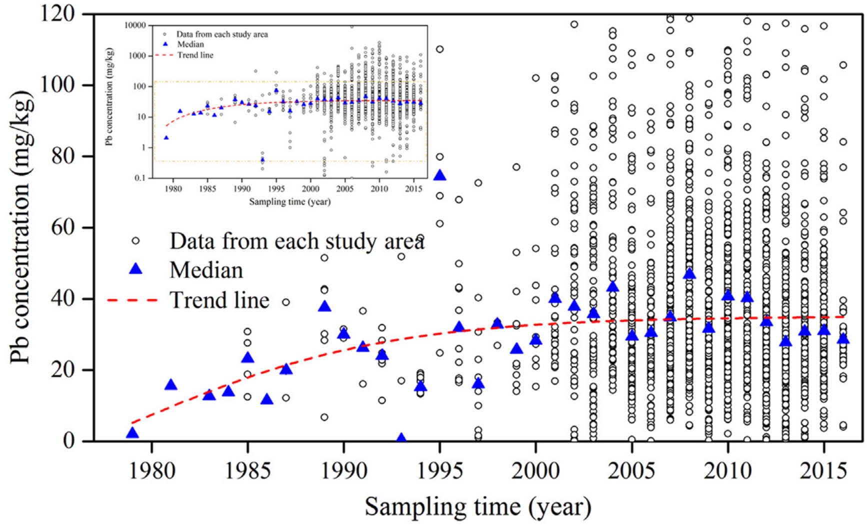

Fig. 4. Temporal trend of Pb concentrations in agricultural soils from 1979 to 2016. Pb concentrations in the range 0–120 mg/kg have been enlarged for clarity.

663.86 t, which was below the level for 1980 (Tian et al., 2012).

In response to the serious pollution problems associated with Pb and other heavy metals, the State Council of China approved the 12th Five-year Plan for the Comprehensive Prevention and Control of Heavy Metal Pollution in 2011. Since then, national and local efforts to decrease Pb pollution from heavily polluting enterprises have been implemented. Management measures have included the phasing-out of leaded gasoline and the closing or merging of heavily polluting enterprises. In addition, many Pb-polluting industries have moved from large cities to medium-sized and small cities, and have moved further into rural areas, often from eastern China to western China (Yan et al., 2013).

3.4. Factors influencing soil Pb contamination in China

China is the world's largest developing country with the world's second-largest economy. China has become the largest producer of raw and refined Pb and its largest consumer. Pb contamination has been recognized in China since the early 1920s (L.F. Han et al., 2018); however, the trends in Pb contamination in agricultural soils in response to China's rapid economic and social development have remained uncertain. In this study, we determined the main anthropogenic sources of Pb in agricultural soils. As shown in Fig. 5, Pb concentrations in agricultural soils were significantly correlated with the GDP (P = 0.0035), total national population (P < 0.0001), and car ownership (P = 0.004). High heavy metal concentrations are usually related to industrial activities, traffic, and a high population density (Ahmed and Ishiga, 2006), which also show interactive effects (Tang et al., 2013; Hao et al., 2008). Furthermore, rapid and substantial economic growth has been accompanied by the expansion of certain industries, such as the automotive industry. China's large population generates significant amounts of waste, of which some is able to enter surrounding agricultural soils (Taghipour et al., 2013). The increasing population is boosting the demand for vehicles, which is causing a rapid increase in car use. Expanding car ownership, heavy traffic flows, and the use of low-grade gasoline have resulted in automobiles becoming one of the leading sources of air pollution (Walraven et al., 2014).

Atmospheric deposition is an important pathway through which Pb accumulates in soils (Schröder et al., 2010). To estimate the sources of Pb contamination further, Pb emissions from anthropogenic activities and their temporal variations in China were assessed (Table S1). The methods we used for these estimations included material balances, emission factors, and source measurements, all of which have been applied in a previous study (Li et al., 2012). Fig. 6 shows that Pb emissions have displayed a large fluctuation over the last 30 years. Between 1990 and 2000, the combustion of motor vehicle gasoline was the largest source of Pb emissions in China, with 115,000 t of Pb being emitted into the atmosphere, representing 80% of total Pb emissions (Li et al., 2012). However, Pb emissions from this source have reduced substantially from 2001 (the unleaded gasoline period) and currently account for just 5% of the total Pb emissions (Li et al., 2012). As a result, coal combustion became the main source of Pb pollution after

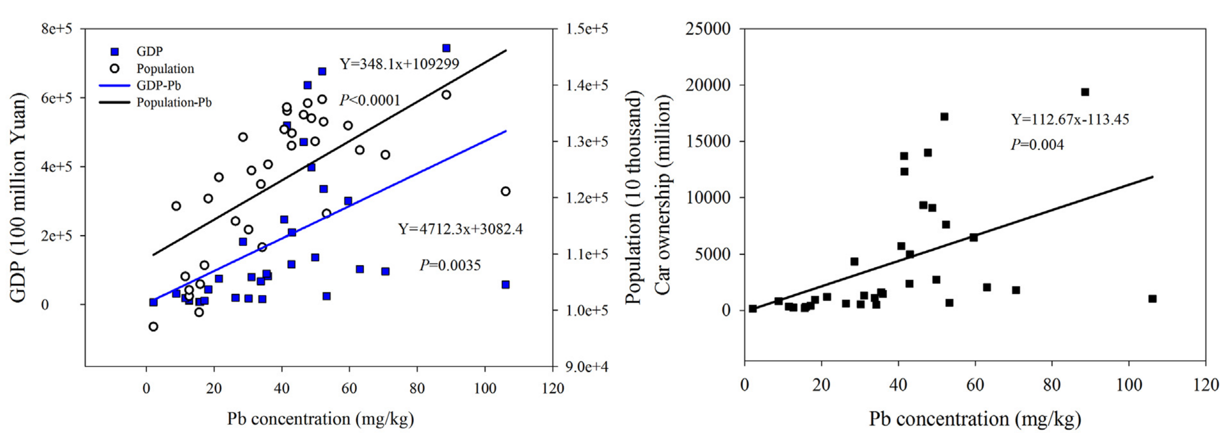

Fig. 5. Relationships between Pb concentrations in agricultural soils vs. GDP, vs. population size, and vs. car ownership.

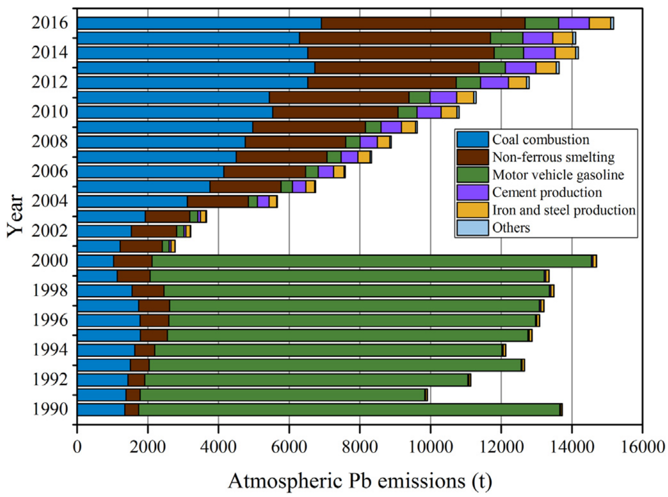

Fig. 6. Atmospheric Pb emissions from several sources in China during the period 1990–2016.

2000. This, while leaded gasoline has been phased out, other Pb emission sources—mainly coal combustion and non-ferrous smelting—have grown rapidly due to the growing demand for energy and the increase in industrial production (L.F. Han et al., 2018). Following a rapid increase in the urban population, energy consumption in urban buildings has driven a rise in Pb emissions (Li et al., 2012). Therefore, emissions have increased rapidly again from 2002 onward. The efficient control of atmospheric Pb emissions should be given much more attention by the government and public; regulatory policies need to be established to reduce Pb emissions from numerous Pb-related industries. Additionally, there is an urgent need to develop new and renewable energy sources including wind, solar, and hydro power if coal consumption is to be reduced.

3.5. Estimation of blood lead levels (BLLs)

Blood Pb poisoning is widely viewed as the biggest health problem resulting from soil contamination with Pb (Laidlaw et al., 2017). Blood Pb poisoning is a worldwide health problem, especially in developing countries (Wang and Zhang, 2006). The World Health Organization (WHO) estimates that the sources of Pb exposure in children include dust and soil (45%), food (47%), water (6%), and air (1%) (Z.X. Han et al., 2018). Previous studies have also shown that soil is an important pathway of Pb exposure to children in China (Z.X. Han et al., 2018).

The integrated exposure uptake biokinetic (IEUBK) model is an empirically validated model (Goodrum et al., 1996) used to estimate Pb exposure from different sources in children aged 0–7 years old. The model exposure parameters associated with Pb concentrations in multiple environmental media, daily consumption rates (e.g., inhalation rates, and ingestion rates of soil/dust, grain, vegetables, and water), and absorption fractions of Pb from different environmental media can be found in the supplementary material. One of the prerequisites for developing policies aimed at reducing the soil exposure to Pb and for eliminating elevated BLLs is an accurate assessment of the contribution from soil exposure sources. In our analysis, we assumed that other exposure pathways including dust, water, and diet would be the same in different regions of China, with only the effect of soil being taken into account. Parameter values of the IEUBK model were listed in Tables S2 and S3.

The predicted BLLs are higher in children aged 2–3 years than those in other age groups, with many studies showing peak BLLs within this age range (Laidlaw et al., 2014). This is probably attributable to the common hand-to-mouth behavior among toddlers (Zhong et al., 2017). For Chinese children, BLLs have declined since 2000 (Z.X. Han et al., 2018) and the simulated BLLs for the period 2011–2016 are considered to reflect the current situation most accurately. In the present study, the mean BLLs of all provinces were lower than the threshold value (100 μg/L) for China. It has been reported that even if blood Pb levels are lower than 100 μg/L, or even 50 μg/L, serious cognitive impairment and neurobehavioral deficits could occur (Gilbert and Weiss, 2006). There is still no internationally recognized blood Pb poisoning threshold (Lanphear et al., 2005). It should be noted that BLLs were higher in Hunan Province than that in other regions, which corresponds to its rich nonferrous metal and nonmetallic mineral deposits and the severe soil contamination that has occurred in this region (Li et al., 2018). Chinese children were found to have higher BLLs than those in developed countries; the average BLL of children in American was 8.4 μg/L in 2013–2014 (Tsoi et al., 2016) and was < 30 μg/L in Europe in 2009 (Bierkens et al., 2011). According to the US National Health Nutrition and Examination Survey (NHANES), blood Pb levels have decreased for the entire population of 1- to 5-year-olds between 1976 and 2014 from 149 μg/L to 8.4 μg/L (Benson et al., 2016; Tsoi et al., 2016). This remarkable decline was due to two major legislative initiatives. Specifically, Pb was phased-out of gasoline and lead-based paints (Benson et al., 2016). The critical concentration value for blood Pb poisoning has been lowered from 100 to 35 μg/L in Germany (Wilhelm et al., 2010) and from 100 to 50 μg/L in the USA (CDC, 2012). However, a higher level of 100 μg/L is still applied in China. Such a high threshold limit for blood Pb level may bring about the underestimation of adverse health risks, and it is not suitable for prevention. Indeed, it is important to prevent Pb exposure rather than responding after exposure has taken place.

Analysis of the BLLs of children aged 0–7 years have been performed in urban China, whereas children in rural China have not been assessed (Zhong et al., 2017). To obtain more precise BLL information, it would be beneficial for the Chinese government to implement free screening of BLLs in children, both in urban and rural areas.

4. Conclusion and perspectives

There was clear spatial variation in Pb concentrations at a national scale in China. The distribution of Pb hotspots indicated that mining and smelting activities, wastewater irrigation, and e-waste recycling have made significant contributions to Pb accumulation in agricultural soils. South regions in China are identified as priority regions for control implementation. The year 2000 was a significant transition year for Pb concentrations, with rapid increase before 2000 and a subsequent slow upward trend. Economic growth, population expansion, and the growing number of automobiles have made substantial contributions to the accumulation of Pb in agricultural soils. Predicted BLLs of all provinces were lower than the threshold value (100 μg/L) for China. There have been many investigations of Pb levels in agricultural soils in China. However, most have focused on regions undergoing rapid economic and industrial development. Therefore, to determine the status of Pb contamination more fully, more research should be conducted in Northwest China. Diet is the predominant contributor to Pb exposure among the preschool urban population in China (Zhong et al., 2017). Therefore, further nationwide investigations are needed to understand the soil-to-crop transfer of Pb and the assessment of human health risks from Pb soil contamination.

5. Limitations

There were a number of limitations in this study. First, the distribution of sample sites was not spatially uniform and, therefore, cannot completely represent the situation with respect to Pb concentrations in agricultural soils across China. Furthermore, the limited number of data records in some provinces may not accurately reflect the extent of Pb accumulation across entire provincial regions. In addition, most previous studies have been conducted in more polluted areas. Thus, assessments based on the dataset compiled in this study may overestimate the extent of Pb contamination in agricultural soils in China. Further verification of these results is, therefore, necessary.

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Declaration of competing interest

All authors of this article declare that they have no conflicts of interest regarding the publication of this paper.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.envint.2019.05.025.

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