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Soil contamination in China: studies on the status, priorities, policies, management and risk assessment

Yiming Sun

Submitted for the degree of Doctor of Philosophy

December 2019

Abstract

As China is trying to balance economic development, environmental safety and human health, the Government has released strategic plans and legislation for soil contamination management. Aspects of the quality of China's soils and management of soil contamination in China are addressed in this thesis. Soil environmental quality standards and science-based risk assessment of contaminants in soils are evaluated.

China and the UK use different risk-based approaches to derive soil screening or guideline values (SSVs; SGVs) for contaminants. The approaches are compared and values derived for 6 illustrative contaminants. China's SSVs are derived using an approach developed in the US as follows: for carcinogens, acceptable level of risk (ACR) is set at 10^{-6} and the SSVs calculated as 10^{-6} divided by the soil exposure and toxicity data; for non-carcinogens, the hazard quotient is 1 and the SSV is calculated as 1 divided by the soil exposure and toxicity data. The UK's SGVs are calculated by the CLEA model, for which the Average Daily Exposure (ADE) from soil sources by a specific exposure route equals the health criteria values (HCVs) for that route, whether for carcinogens or a non-carcinogens. The UK's CLEA model is also used here to derive SSVs with Chinese input parameters. China's SSVs, the UK's SGVs and values for Chinese conditions derived using the UK approach were as follows (mg/kg): As, <1, 35, 20; Cd, 20, 18, 11; Cr (VI), <1, 14, 29; benzene, 1, 1, 2; toluene, 1200, 3005, 3800; ethyl-benzene, 7, 930, 1200. The difference in toxicity assessment and risk characterization for carcinogens results in the biggest difference in SSVs between the 2 countries. However, for non-carcinogenic substances, the difference of SSVs calculation method and SSVs is small. In the

future, China can use the UK method to strengthen its toxicity assessment and risk characterization for carcinogenic substances.

Data was made available for this thesis from an extensive field and analytical campaign of human exposure to heavy metals in China. This was used to calculate the relative contributions of exposure to As, Cd, Cr, Hg and Pb from environmental media (air, water, soils) via the inhalation, drinking and the diet for different regions of China. Dietary exposure dominated, contributing ~90-97% of the total exposure for these elements. Exposure differences were observed with gender, age and region. This survey information can be used to derive exposures from soil-borne sources.

Soil organic matter (SOM) and pH are critical soil properties strongly linked to carbon storage, nutrient cycling and crop productivity, but there is a lack of information on changes in these soil properties over time for China. This study used data from Chinese soil surveys to examine changes in soil pH and SOM across different land uses (dry farmland, paddy fields, grassland, woodland, unused land), with surface soil (0-20 cm) collected in the periods 1985-90 (Survey 1; 890 samples) and 2006-10 (Survey 2; 5005 samples) from two contrasting areas. In the southern part of China, the mean pH of paddy soils fell over the two decades between surveys - from pH 5.81 to 5.19 ($p<0.001$), while dry farmlands in the northern sampling area fell slightly (from pH 8.15 to 7.82; $p<0.001$). The mean SOM content of dry farmland soil rose in both areas and the mean SOM of paddy fields in the southern area also rose (all $p<0.001$). Woodland soil pH in the south increased from 4.71 to 5.29 ($p<0.001$) but no significant difference was measured in the woodlands of the northern area, although the trend increased. The SOM content

of woodland top soils rose in the northern ($p=0.003$) and southern ($p<0.001$) study areas. The implications and potential causes of these changes are discussed and suggestions made as to how large-scale soil sampling campaigns can be designed to monitor for changes and potential controlling factors.

Because of rapid urbanization in China, the demand for land for urban development is increasing. To upgrade and modernize, China has also moved many major industries and factories from urban centres to less populated areas. With the high economic value of urban land, the transformation and utilization of the brownfield areas left behind has become important economically and socially. Strong scientific, regulatory and decision-making frameworks are needed, to ensure practical, careful and wise use of central and local Government resources, to manage the re-use and regeneration of these brownfield sites. The final chapter provides a thorough review of the background, context, regulations, policies and management procedures to develop and utilize brownfields in developed countries such as the US and UK, and identifies some of the priorities for brownfield governance and redevelopment in China. It is proposed: to establish a monitoring body, to identify shared responsibilities and inputs of various stakeholders, to establish brownfield databases, and to set up a remediation advisory system with technology support as future priorities of brownfield management. Recommendations are made for future research, to support China's strategic management of soil resources.

Acknowledgement

I appreciate the opportunity of I working with four supervisors, Prof. Kevin Jones, Prof. Kirk Semple, Dr. Hong Li and Prof. Guanlin Guo, who have different backgrounds and are all willing to provide expertise, data and technical support during the four years. Prof. Kevin Jones is the first supervisor leading me to the scientific research when I studied master degree and also the person who recommended me to here, the wonderful journey. He is not only my supervisor, but also life mentor. I learned from Kevin how to tell story, positioning contents in an article and how to make important things simple, concise, clear and clean. He always prepares to provide suggestions on science and life, and lets me know every option is possible if reasonable and he encourages me a lot, which made me more confident especially at my whole stage of PhD study. Prof. Kirk Semple is the supervisor always providing me valuable suggestions. During the four years PhD study, he always responded promptly to the question or request from me on research progress and paper comments. Hong Li is active with full of ideas in his mind. It is always rewarding to discuss with him on ideas, data and technical problems. Prof. Guanlin Guo the supervisor leading me to the practical research, when I was in CRAES, China as an exchanging PhD student. I respect them, as they are not only supervisors but also friends, mentors and sometimes parents. By working with them as a group, I become a better person.

Thanks also to colleagues in Centre for Ecology & Hydrology-Lancaster, Dr. Aidan Keith who helped me for data analysis and paper comments, Dr. Lisa Norton who contributed the knowledge of the Countryside Survey. They all kindly provided support during the process that

I was developing my projects. Also, thanks to friends and colleagues in LEC, CEH, CRAES and my office, who provided the warmest companionship during the four years.

I would like to thank the Friend's programme Research Travel Conference Fund to support me for attending the academic conference in China during my doctoral research.

Last but not least, I would like to express the deepest thanks to my parents, Xiaoyu Sun and Yanping Wang, for their selfless love, support and encouragement throughout the past three years, for which I will always be grateful. I wish to dedicate this dissertation to my family.

Yiming Sun

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List of papers

I. Yiming Sun, Hong Li, Guanlin Guo, Kirk T. Semple, Kevin C. Jones. (2019). Soil contamination in China: Current priorities, defining background levels and standards for heavy metals. Journal of Environmental Management, 251, 109512.

*Yiming Sun wrote the paper, with supervision from Kevin C. Jones, Hong Li, Guanlin Guo and Kirk T. Semple. Kevin C. Jones joined in designing the idea***.**

II. Yiming Sun, Jicai Wang, Guanlin Guo, Hong Li, Kevin C. Jones. (2019). A comprehensive comparison and analysis of soil screening values in China and UK. Environmental Pollution, 113404.

Yiming Sun and Jicai Wang developed the model, conducted the research and wrote the paper with supervision from Kevin C. Jones, Guanlin Guo, and Hong Li.

III. Yiming Sun, Guanlin Guo, Huading Shi, Mengjiao Liu, Aidan Keith, Hong Li and Kevin C Jones. (2019). Decadal shifts in soil pH and organic matter differ between land uses in contrasting regions in China. Submitted to Science of the Total Environment.

Yiming Sun conducted the research and wrote the paper with data support from Huading Shi and Mengjiao Liu, with supervision from Kevin C. Jones who joined in designing the ideas, Aidan Keith joined in data analysis and interpretation, Guanlin Guo, and Hong Li reviewed the manuscript.

IV. Yiming Sun, Hong Li, Guanlin Guo, Kirk T. Semple, Kevin C. Jones. Re-development of urban brownfield sites in China: motivation, history, policies and improved management. Submitted to Science of the Total Environment.

Yiming Sun conducted the research and wrote the paper with data support from Hong Li, and Kirk Semple, with supervision from Kevin C. Jones who joined in designing the ideas.

Appendix 3. Zhang Xiuge, Danlu Wang, Bin Zou, Jian Xu, Limin Lei, Zhenglei Li, Yan Tao, Yiming Sun, Hao Zhang, Kevin C Jones, Feiyang Qiao, Ju Huang. Total human environmental exposure and risk assessment of residents for metals in typical areas of China. Submitted to Environmental Health Perspectives.

Zhang Xiuge conducted the research and wrote the paper with the data support from Danlu Wang, Bin Zou, Limin Lei, Zhenglei Li, Yan Tao, Ju Huang, with the supervision from Jian Xu, Yiming Sun helped the manuscript revision, Kevin C. Jones and Hao Zhang joined in designing the ideas and comment on the manuscript.

1. The soils and land use of China

1.1 The soil resources of China

Soils are essential for the functioning of our planet and to support human kind. They underpin ecosystems, enable agricultural production, support the supply of clean water and the development of human civilisations (Singer and Munns, 1991, Brady et al., 2008). Economic development, quality of life and advanced civilisations are directly dependent on soil resources. It is vital that soils are well managed and protected and that their quality is safeguarded for the future. This is critical for the future of China, the most populated country on Earth (Pierzynski et al., 2005, Donahue et al., 1983).

China has a land area of 9.6 million square kilometres, which accounts for ~6.5% of the Earth's surface, nearly 20% of the Earth's land surface (Zhao, 1989). The management of this land and soils needs to support a population of ~1.4 billion people, a range of diverse ecosystems and natural habitats. The importance of managing our planet's soils is enshrined in international sustainable development goals (Keesstra et al., 2016, Liu et al., 2010, Bouma, 2014) and is increasingly recognised and incorporated into the intentions of the Chinese government (Huang et al., 2010, State Council, 2016).

China is a vast territory, covering many climatic zones and habitats (Ding and Gong, 1987). A complex range of natural conditions combine to give a rich array of soil types and diverse land resources, reflecting differences in climate, geology, land use and management (Figure 1-4). Broadly, China is divided into a cold temperate zone, a temperate zone, a warm temperate zone, a subtropical zone and a tropical zone from north to south under the various temperature conditions (Figure 1). There is also an increasing trend of precipitation from west to east, so that China is divided into humid, semi-humid, semi-arid and arid areas. China’s natural vegetation from the southeast to the northwest can be roughly classified into forest, grassland and desert (Gong, 1990) (Figure 2). Seven main terrestrial ecosystem categories (farmland, forest, grassland, water body and wetland, desert, settlement and other ecosystems) cover the whole landmass of China (Figure 3). Land use types include cultivated land, forest land, grassland, waters, residential land and unused land. According to the Ministry of Natural Resources of the People's Republic of China, in 2016 there was 645 million hectares of agricultural land in the country, including 135 million hectares of arable land, 14.3 million hectares of ‘gardens’ (parcels of land where people cultivate fruits, tea etc), 253 million hectares of forest land, 219 million hectares of pasture and 38.6 million hectares of construction land (Ministry of Natural Resources of the People's Republic of China, 2017). Under the influence of geographic and climatic factors, China has 41 different soil types (Figure 4). The geographical distribution of soil pH is related to ocean-continent precipitation and – in many areas – the underlying geology. It can broadly be summarized into an acid south region, alkali north region, acid coastal region and alkali inland region (Dai et al., 2009). Soil organic matter is also affected by these climatic and geographical factors, which lead to significant differences in soil organic matter content between different regions of China (central southwest China > northeast China > south China > east China > north China > northwest China) (Wang et al., 2000).

China’s arable land, woodland and grassland areas rank as the 4th, 8th and 3rd respectively in the world (Gong et al., 2005). Although China’s arable area ranks fourth in the world, the per capita area of land in China is significantly lower than the world’s per capita (1.5 mu per capita in

China compared to 5.5 mu per capita as the world average) (1 mu =666 m^2, mu is a unit of rural land division in China). It is a similar story for grassland (5.3 mu versus 11.4 mu) and forest land (1.8 mu versus 9.8 mu) (Zhao, 1989). This obviously shows a greater pressure on China's land resources than the global average.

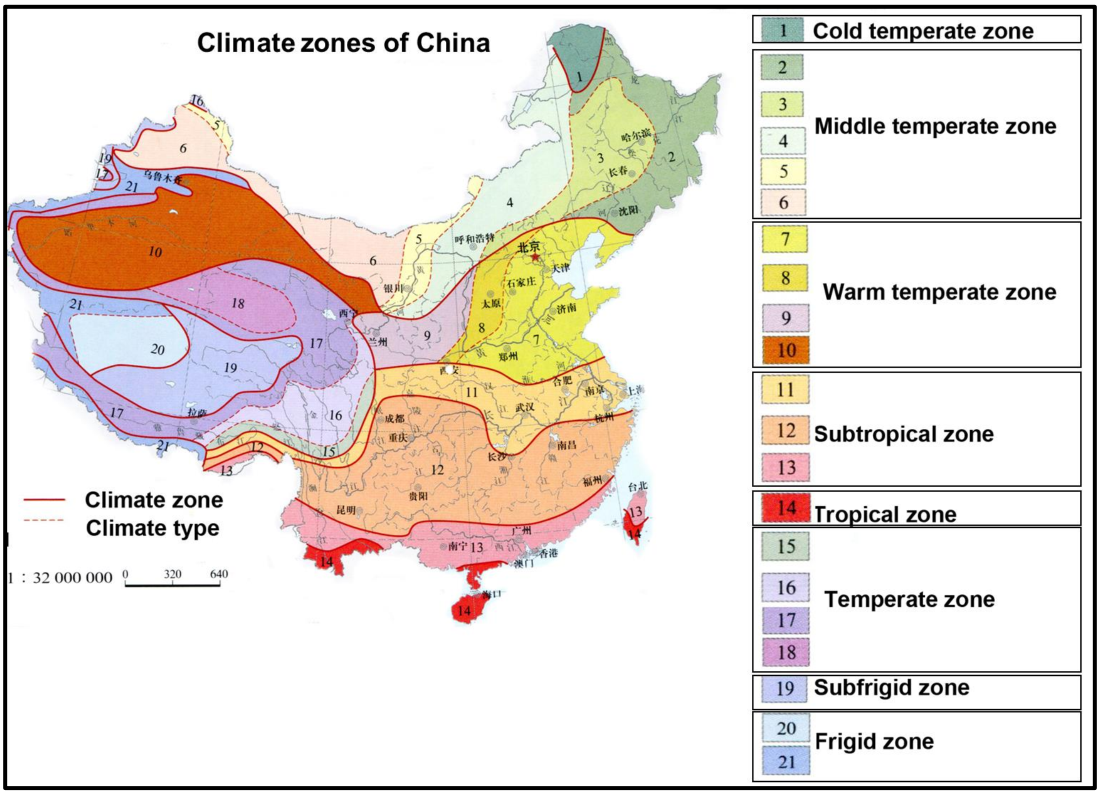
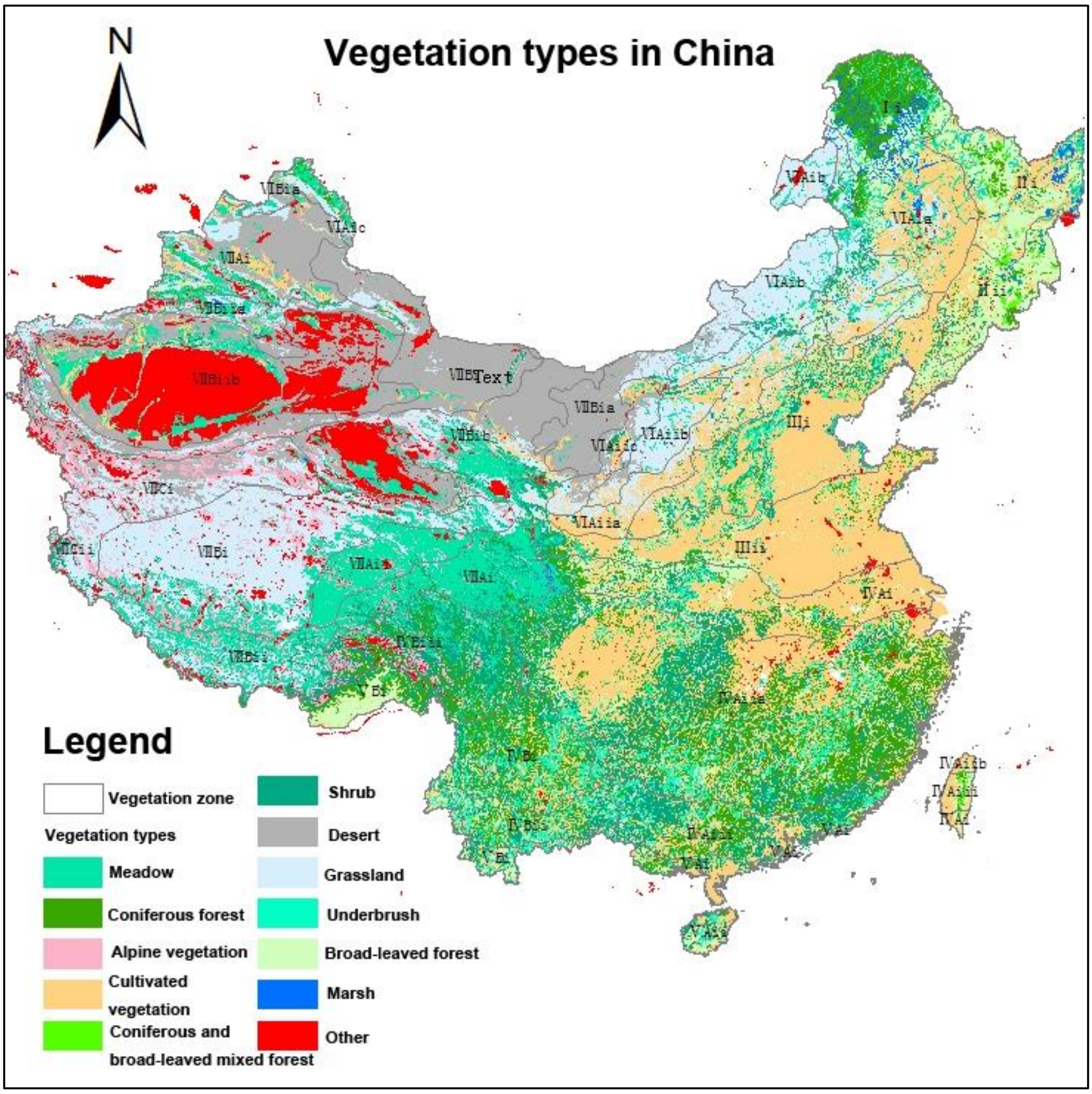


Figure 1 The map of climate zones of China based on the China geographic atlas in 2012 (Geographic Data Sharing Infrastructure, 2019).



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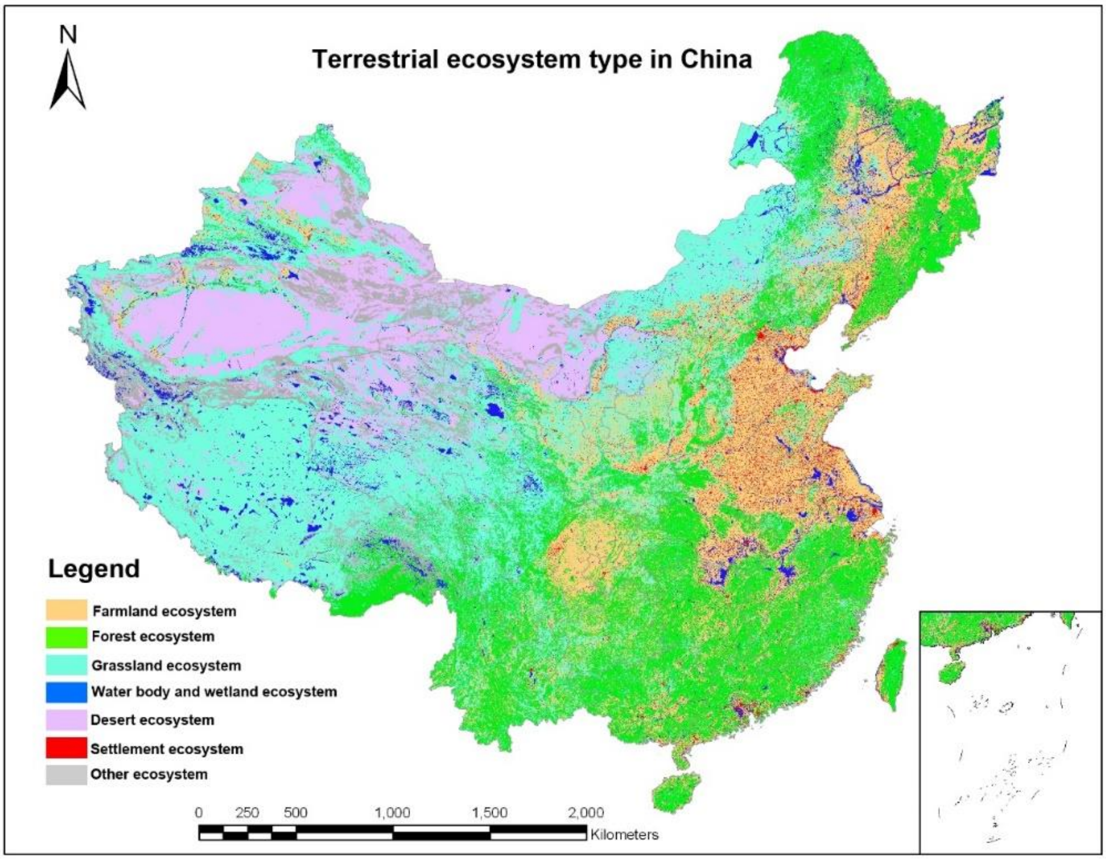

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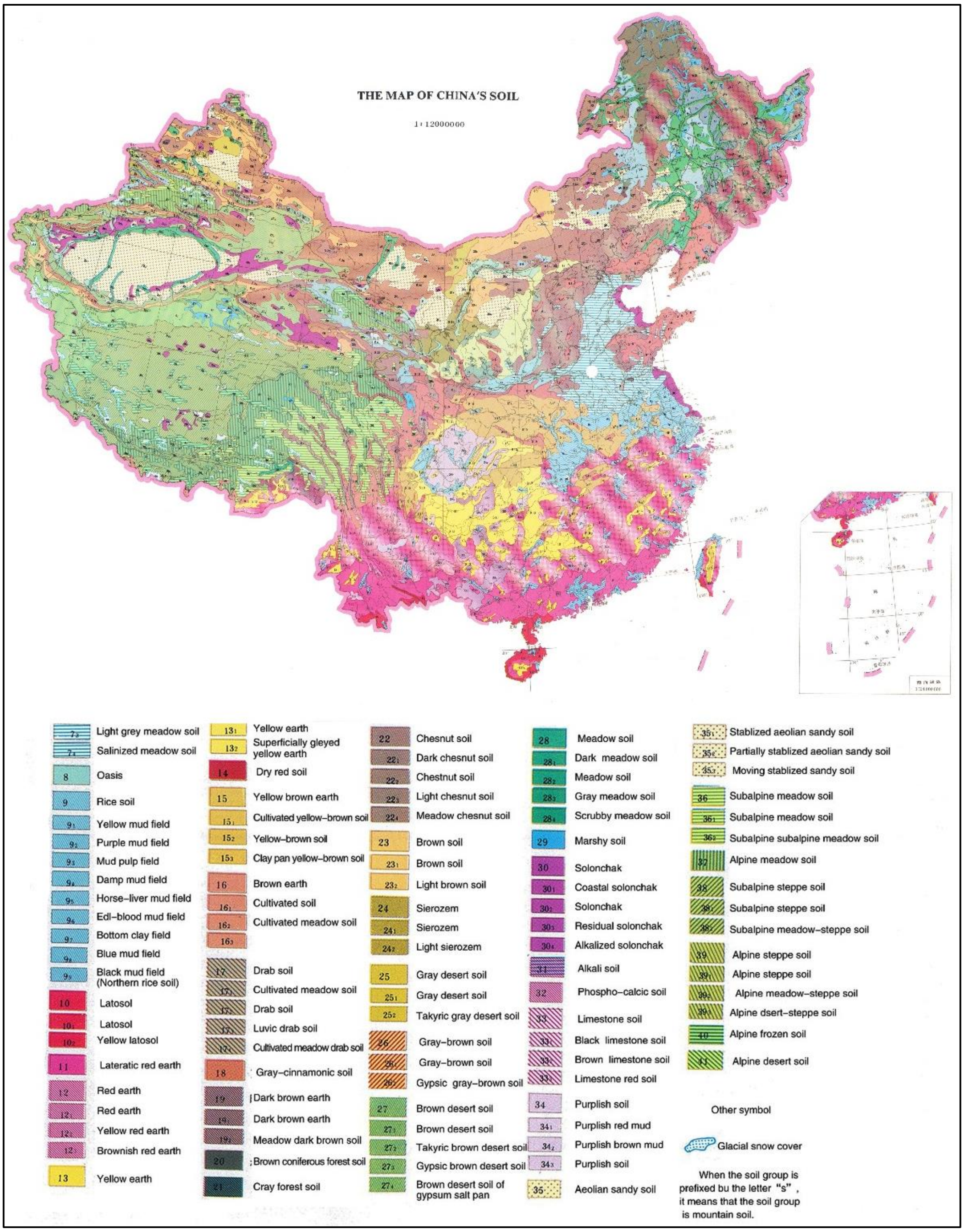


Figure 4. The map of China's soil types (National Environmental Protection Agency and China National Environmental Monitoring Centre, 1994).

1.2 Soil resource distribution in China

As China has pushed for greater food security, the quantity and quality of soil resources strongly influences food production. The soil resource varies greatly between the regions in China (Figures 1-4). As a result, the distribution of ~1.4 billion people in China is very uneven (Figure 5). There is also an uneven distribution of gross domestic product (GDP) across China (Figure 6). With the rapid economic development of China and the active urbanisation policy, the urban area, urban construction land area and urban population density have all been increasing sharply (Figure 7 A-C). Between 2009-2018 China's urban population increased by 0.186 billion and the rural population decreased by 0.125 billion (Figure 7 D). These dramatic social and economic changes will have a complex effect on land use and resources (Zhao, 1989, Sheng et al., 2019). In short, more people are consuming more food and natural resources, with growing pressure on the higher quality lower altitude lands to the east of China.

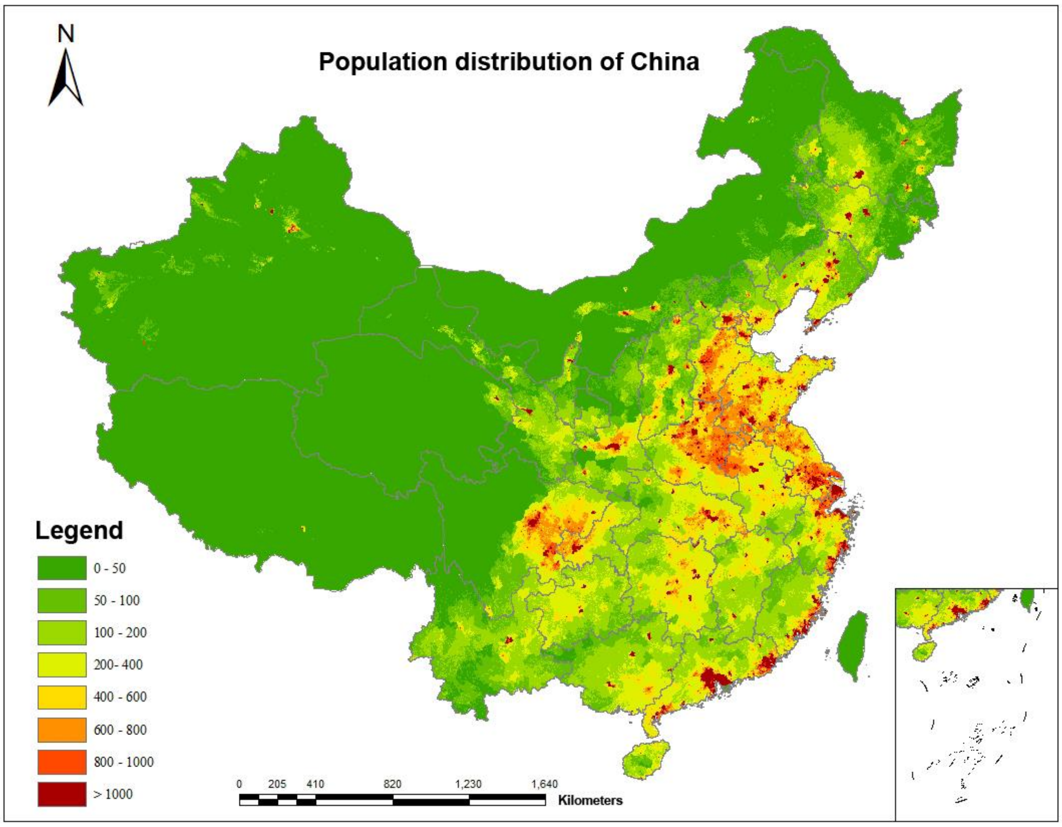
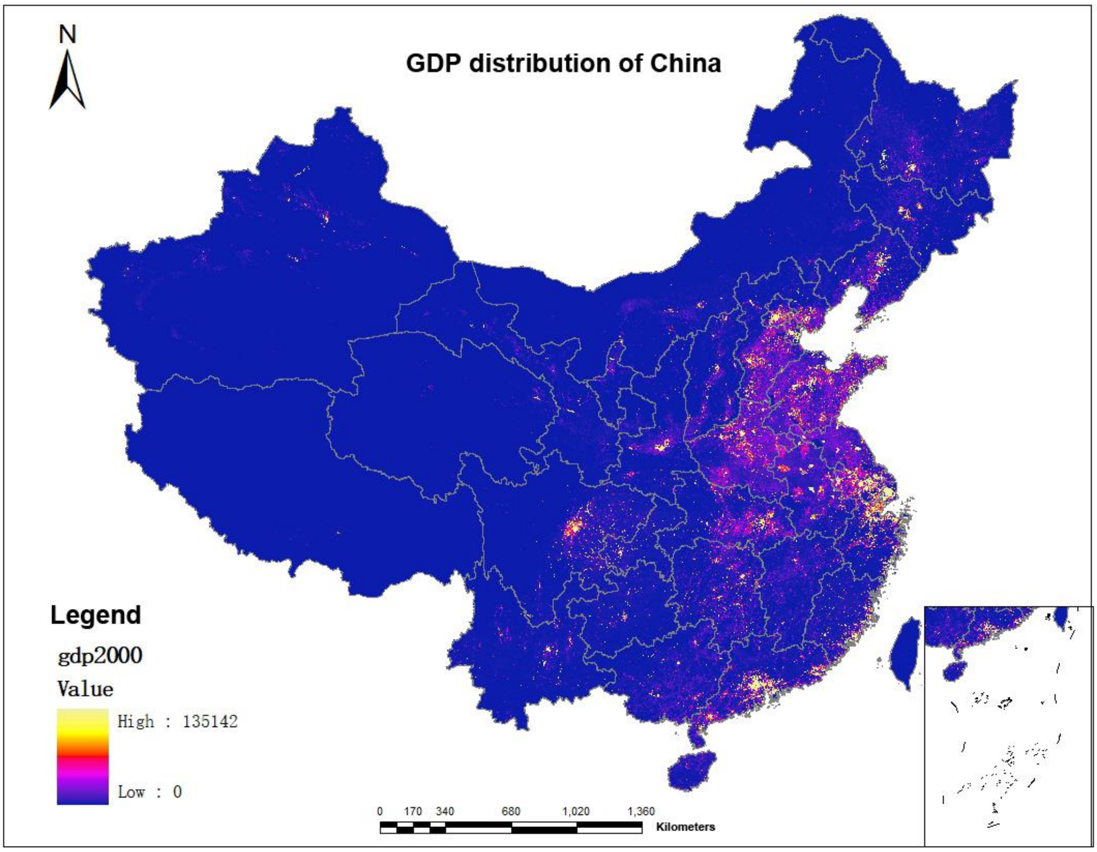


Figure 5. The map of population distribution of China in 2000 (Resource and Environmental Data Cloud Platform, 2019).



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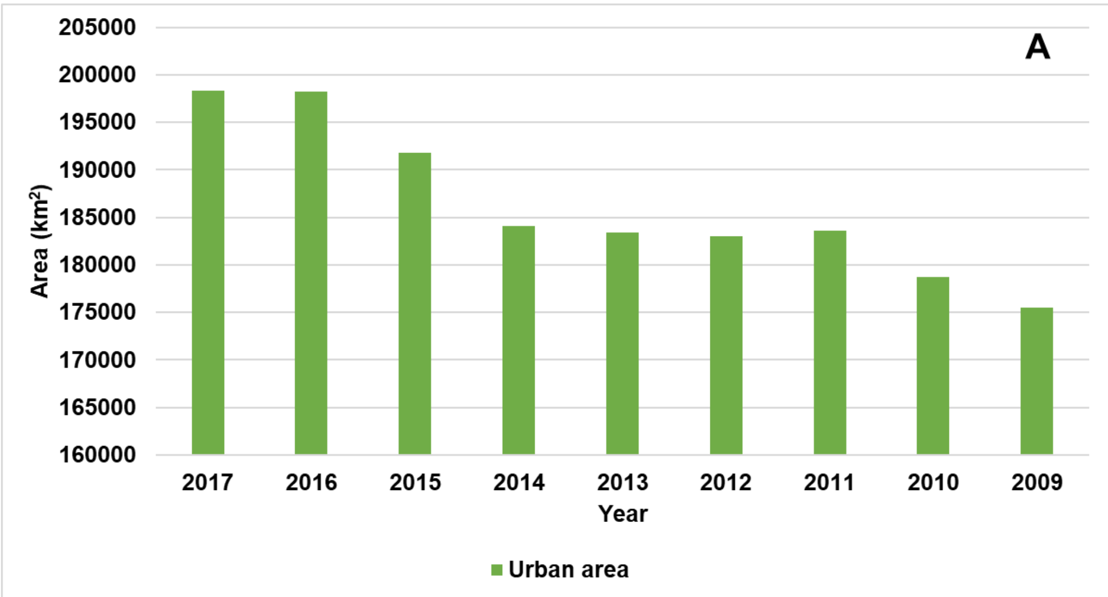

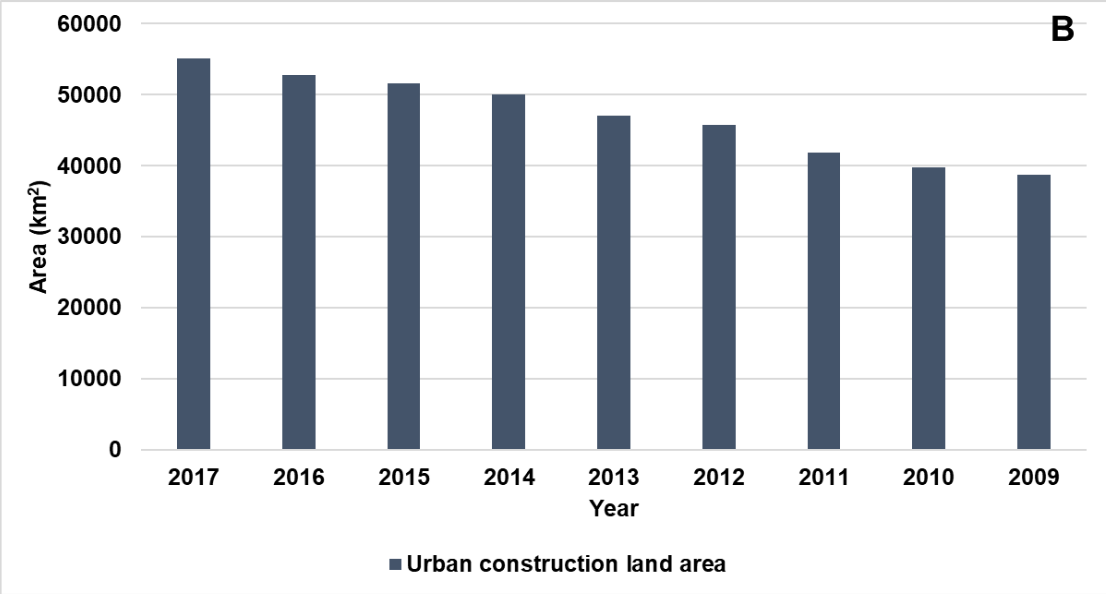
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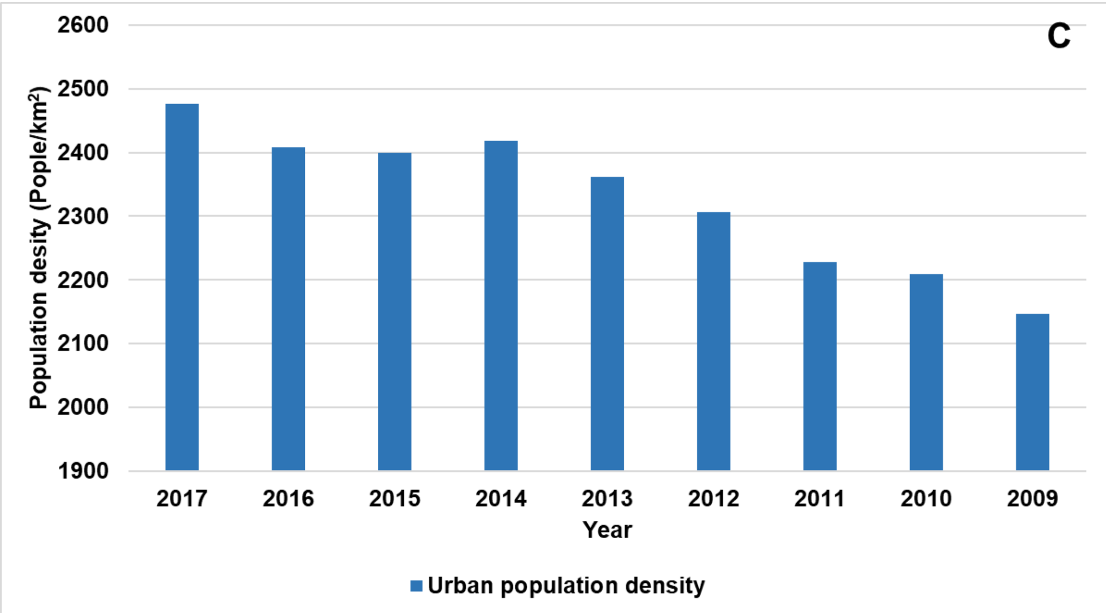
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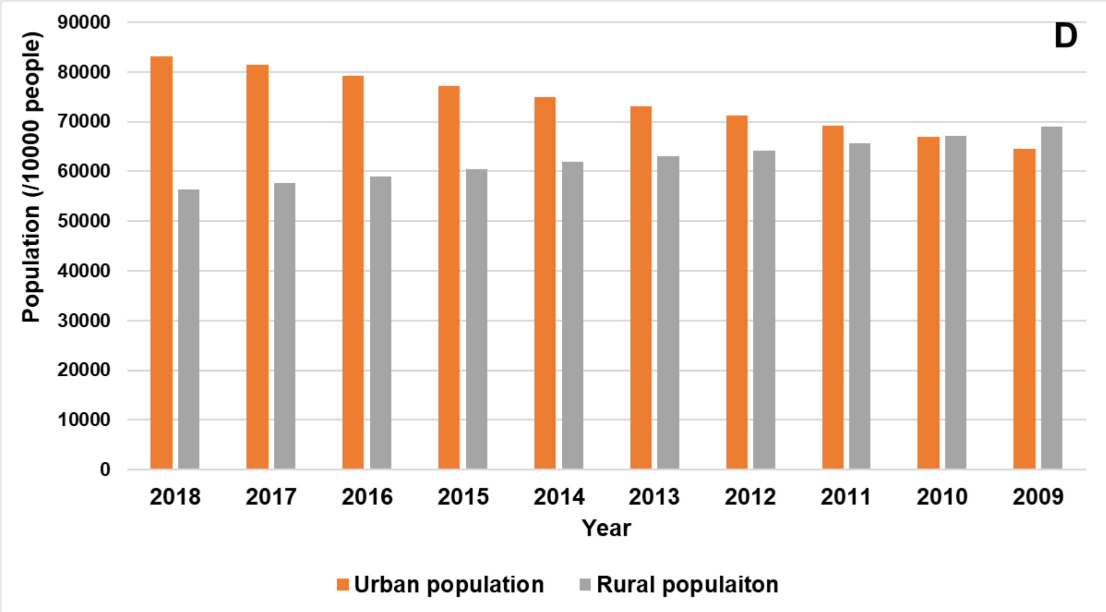
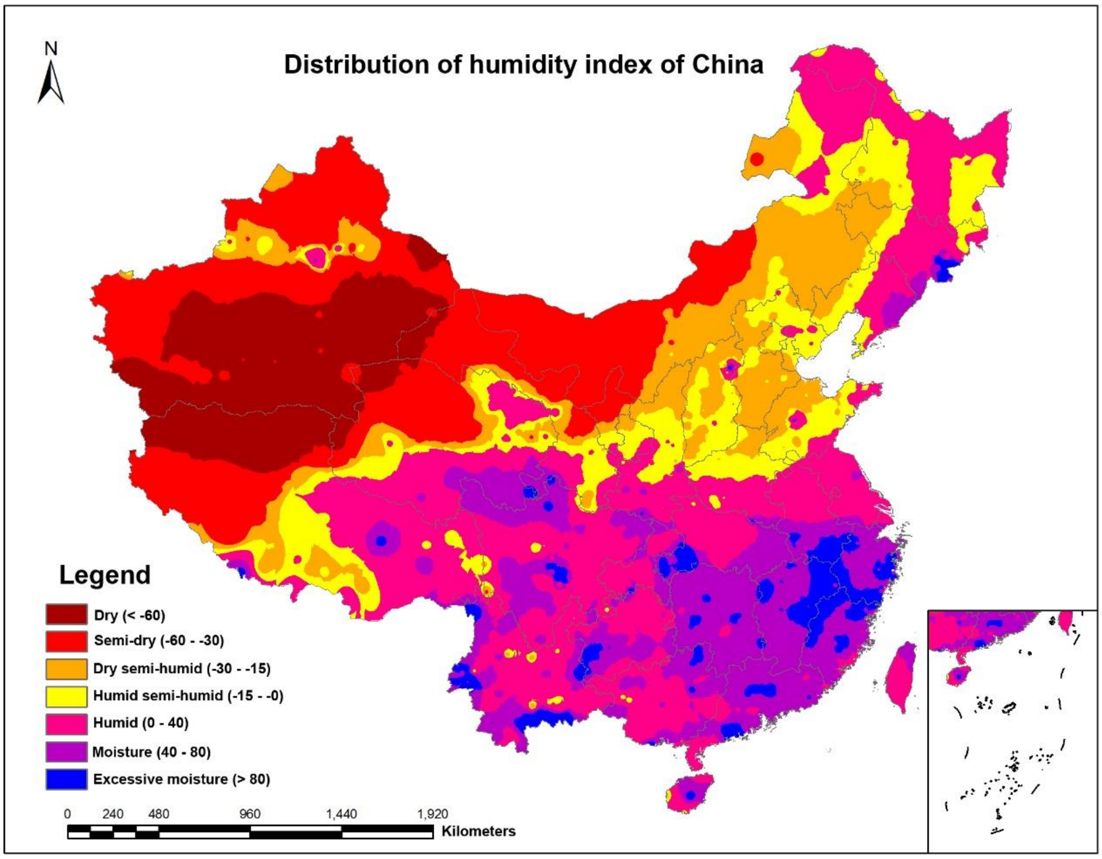


Figure 7. A: Distribution of urban area from 2009 to 2017; B: Distribution of urban construction land area from 2009 to 2017; C: Distribution of urban population density from 2009 to 2017; D: Distribution of urban and rural population from 2009 to 2018. Population unit is 10,000 people. Data from National Bureau of Statistics (National Bureau of Statistics of China, 2019).

China's 'Soil system classification: theory, method, practice' broadly divides China's soil resources into three major soil areas (Gong, 1999) - the humid, semi-humid and dry soil areas. China's humidity index increases from west to east China (Figure 8). The humid soil area in the southeast region accounts for 41.6% of the country, with a dense population and developed economy. It contains 81% of the country's population and 72.2% of the arable land. The arid northwest soil region accounts for 35.7% of the national area. Due to the constraints of drought and alpine terrain in this region, the range of water and soil quality and quantity are sharp, the ecological environment is fragile, the economy is undeveloped, and the population and proportion of arable land are small, accounting for only 4% and 8.2%, respectively. The semi-dry soil area in the central part of China has obvious transitional characteristics, accounting for 22.7% of the total land area, 15% of the population and 19.6% of the arable land in the country

(Figure 9). Industrial development is lagging behind in this area and the proportion of agricultural land is high (Gong et al., 2005).



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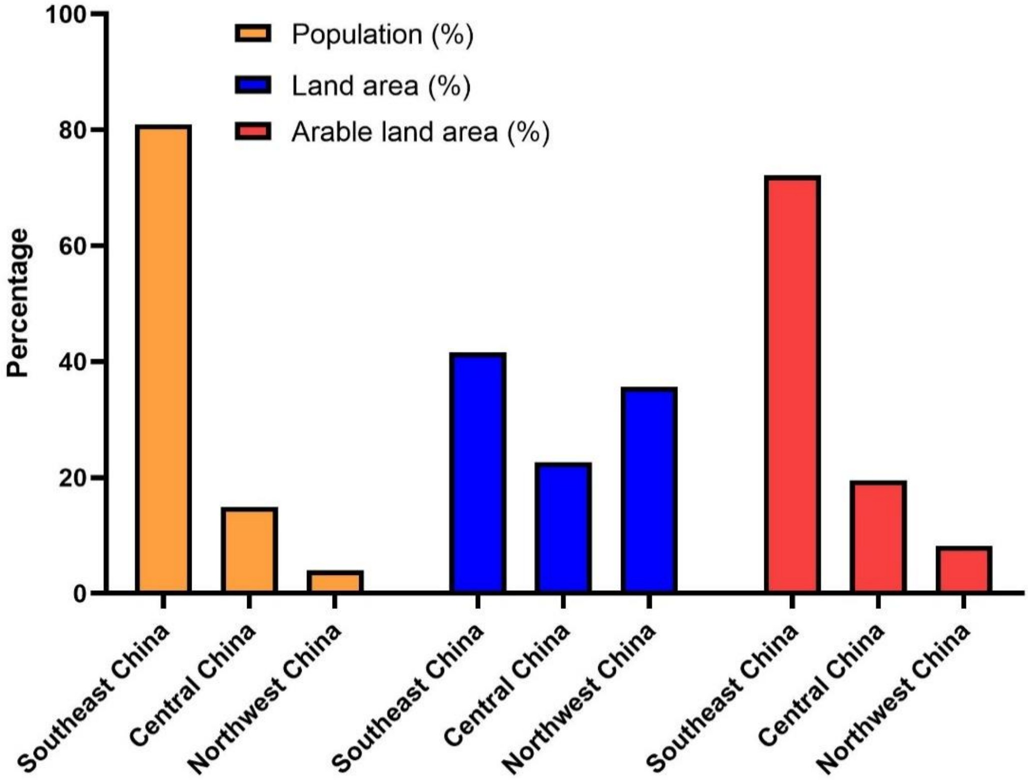

Figure 9. The distribution of population, land area and arable land area in three Chinese soil regions.

2. Management of land and soils in China – dramatic changes and pressures

After the founding of the People's Republic of China in 1949, China has mainly implemented the public land ownership system. China's Constitution stipulates that 'land in cities belongs to the state. Land in rural and suburban areas is owned by the state, in addition to those stipulated by law and is owned collectively; house sites, self-reserved land, and self-retained mountains are also collectively owned'. Since 1949, the reform of China's rural land system can be roughly divided into three stages: the land reform period (1949-1953), the cooperative and the people's commune period (1953-1978), and the household contract operation (1978-present) (Figure 10) (Jiang and Tan, 2019, Liu and Cheng, 2007, Ding, 2003).

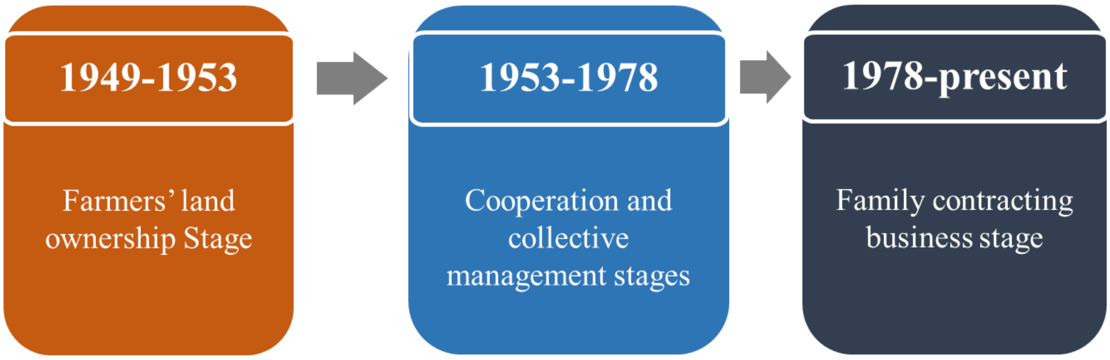

Figure 10. The stages of land policy in China (Ding, 2003, Liu and Cheng, 2007, Jiang and Tan, 2019).

The land reform period (1949-1953) guided the reform of the rural land system in China. The land system of feudal and semi-feudal exploitation was abolished and a land system where farmers have their own land was implemented. The "Land Reform Law of the People's Republic of China" promulgated and implemented in 1950 stipulates that China implements a land system owned by farmers, and farmers have the right to freely operate, buy, sell, and lease their land. Since then, the Central People's Government has promulgated the General Rules for the Organization of Farmers' Associations, the Resolution on Division of Rural Classes, and the Regulations on Land Reform in Urban and Suburban Areas, to further promote the reform of the land property rights system. At this stage, according to the status of individual farmer production and operation and the needs of the country's economic development, the state believed that it was necessary to promote "organization" of farmers, develop mutual assistance and cooperation among farmers, guide farmers on the road to prosperity, and change outdated agriculture. In particular, the "Land Reform Law of the People's Republic of China" promulgated in June 1950 clearly stipulated and adjusts the line, principles and policies of land reform, and guided the country in formulating land reform. This is the landmark law of this stage, and its main content is the abolition of feudal exploitation. At the beginning of 1953, the country basically completed the land reform, the direct combination of the means of production and labour, the abolition of feudal land ownership, and promoted the recovery and development of the rural economy (Ding, 2003 , Liu and Cheng, 2007, Jiang and Tan, 2019).

In the cooperative and the people's commune period (1953-1978) China carried out agricultural socialist transformation. It experienced the transformation from ‘agricultural production mutual aid groups’ to ‘advanced agricultural cooperatives’ – basically a transition from peasant ownership to collective ownership. During this stage, in order to carry out agricultural socialist transformation, China went from agricultural production mutual aid groups to primary agricultural cooperatives to advanced agricultural cooperatives. Land and other means of production are owned by farmers and handed over to cooperatives for unified use and receive a certain amount of compensation. During the process of collective ownership and cancellation of the means of production, the land system achieved a transition from peasant ownership to collective ownership (Ding, 2003 , Liu and Cheng, 2007, Jiang and Tan, 2019).

During the household contract operation (1978-present) period, historic changes have taken place in China's rural land policy, and the changes have mainly revolved around household responsibility systems for joint production. Since the reform and opening up of China, the focus of the country's work has shifted to economic construction. Economic reform began in rural areas, and the core was land policy. Support was given to policies for the upsurge of the household contract responsibility system in rural areas. During this period, the state continuously strengthened the protection of land laws and policies, and consolidated and improved the basic rural management system with collective land ownership and household contract management. From the late 1990s, the reform and innovation of the rural land system attracted attention as an important way to solve the problems of agriculture, rural areas and farmers. In the field of research, it has mainly focused on a series of issues, such as the construction of rural land system and rural democratic system, land system and industrialization, urbanization construction, sustainable land use and land system reform, research on land system and agricultural ecological environment issues, rural land system and farmers' rights safeguards (Ding, 2003 , Liu and Cheng, 2007, Jiang and Tan, 2019).

In order to understand and better utilise the soils and land, the Chinese government has conducted several soil environmental surveys (Figure 11) (Zhu and Liu, 1990). This began with descriptions of soil types (taxonomy) in the 1930s and the preparation of regional and national soil maps. After the founding of the People's Republic of China, large-scale regional soil surveys and specialized comprehensive scientific investigations were carried out. In 1958-1959 and 1979, two national soil surveys and large-scale soil mapping were carried out. In recent years, with the intensification of soil pollution, the country has gradually strengthened its leadership and guidance on soil environmental protection, and various relevant departments have also carried out soil environmental monitoring in their respective fields. During the 'Sixth Five-Year Plan' (1981-1985), 'Seventh Five-Year Plan' (1986-1990) and 'Eighth Five-Year Plan' (1991-1995), the Ministry of Environmental Protection carried out a survey on the background value of selected elements in soils; during 2001-2003, a special survey on soil environmental quality in sewage irrigation areas and organic food production bases was carried out. In recent years, the Ministry of Agriculture has also begun to pay attention to the prevention and control of agricultural soil pollution. From 2002-2004, special investigations were carried out on the soils of agricultural production areas in some cities (Lu et al., 2014, Liu and Cheng, 2007).

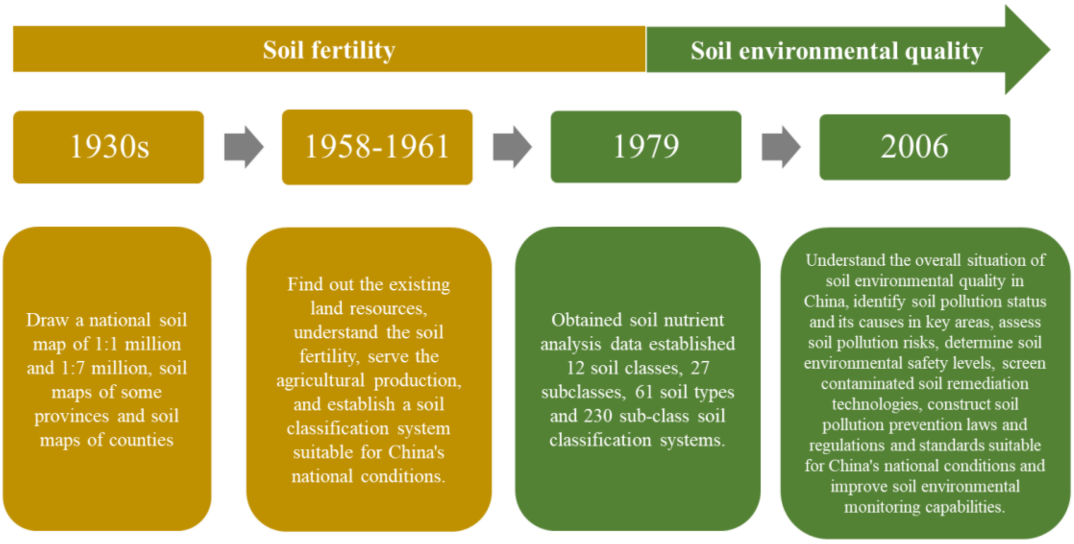

Figure 11. Process of national soil surveys in China (Zhu and Liu, 1990 , Lu et al., 2014, Liu and Cheng, 2007).

The previous section and this one has given some background to the social and political changes affecting China over the last ~70 years. These have had important effects on land use, resource management and soil quality. These are difficult to quantify and understand on a large scale (see Figures 12). China's "Classification Standard for Land Use Status" (GB/T 2010-2017) combines land use data into six categories: grassland, forest land, cultivated land, construction land, water area and other land. In summary, this time period has seen the area of forest land, waters and construction land increase and the area of grassland, other land and arable land decrease. The increases in forest land and water areas mainly occurred in the western region; the increase in the area of construction land mainly occurred in the eastern and central regions; the decrease in the area of grassland and other land occurred mainly in the western region; the decrease in cultivated land mainly occurred in the eastern region (Figure 12) (Zhu et al., 2019).

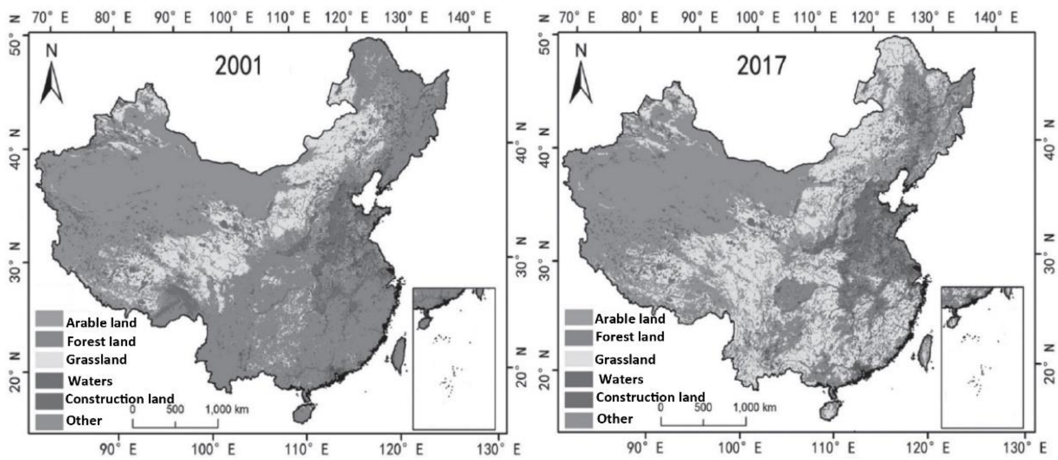

Figure 12. Changes in land use types in China between 2001 and 2017 (from Zhu et al., (2019)).

During the ~70 years summarised in section 2 and here, many factors have changed to affect land use and soil management. A key factor is more intensive crop production, a stated aim of the Government. This has been achieved, through the change in land ownership and management, greater use of fertilisers and pesticides, more demand for water etc. But there have been unwanted side-effects, including more soil erosion, more/larger scale desertification, flooding, soil and water pollution (Liu and Diamond, 2005). For example, according to available statistics from the International Institute for Applied Systems Analysis (IIASA) for assessing artificially induced soil degradation (ASSOD) in South and Southeast Asia, it is estimated to be 465 million km^2, most of which are slightly degraded, which accounting for 66% of total area (~307 million hm^2).

China is currently in an important period of industrialization, urbanization and ecological civilization (Liu and Diamond, 2005, Hesselberg, 2005). Cultivated land resources are increasingly subject to competition from industrial and urban land use. The trend of 'non-agriculturalization' of some arable land risks seriously affecting the country's food safety, and has attracted widespread attention at home and abroad. In response to the rapid economic development, the contradiction between non-agricultural and agricultural land has become increasingly prominent. During the 15 years from 1978 to 1993, the apparent area of arable land in China decreased by 4.288 million hm^2 (Zhu, 1997).

The National Bureau of Statistics released a report showing that China's GDP in 2018 had increased by 175 times compared to 1952. China's contribution to world economic growth averages 30% annually, lifting more than 700 million people out of poverty and contributing more than 70% to global poverty reduction. As mentioned above, these huge achievements have also created a series of environmental problems.

China's soil resources are facing many prominent problems such as soil erosion, reduced fertility of cultivated land, land desertification, soil salinization and soil acidification. All are very important pressures on China's soil resources. However, this thesis focusses on a number of aspects of soil contamination and soil quality, and the impacts this can have.

3. Environmental pollution pressures in China

As just introduced, China is facing major environmental problems (see Figure 13). Specifically, contamination of air, water and soil have become high priorities for the Government and the public. Indeed, it has been estimated that environmental problems have become one of the main obstacles restricting further economic development in China (Zeng, 2012). According to a World Bank Survey, China’s “environmental crisis” will consume 8% to 12% of GDP per year. Of the 20 most polluted cities in the world today 13 are in China. The global shift towards industrialisation in the developing world has focussed in China, as China has become a ‘factory for the world’. The relative shortage of resources, fragile ecological environment and insufficient environmental capacity have become major issues in China’s development (Li et al., 2015).

Figure 13. The major environmental problems in China

China's development through the 1970s and later has resulted on obvious impacts on air and water quality (Huang et al., 2010, Liu and Diamond, 2005). The Chinese Government has taken active steps to improve this situation. However, effects on soils and ground-waters are generally less visible and the understanding and soil protection issues has been slower to materialise. Whereas air and water pollution are closely linked to emissions and discharges, the soil is the key 'storage compartment' for many environmental pollutants. It can act as a reservoir or sink for pollutants, potentially storing them for many decades or even centuries (Zhou et al., 2016). The focus on air and water has begun to give improvements, for example the air quality of Beijing has improved in recent years. Generally the technologies and measures needed to improve air and water quality are well known and the Government's investments in these areas will give obvious benefits.

A series of laws and regulations have been introduced, such as the "Water Pollution Control Law" (first released in 2008), "Air Pollution Control Law" (first released in 1987), "Administrative Measures for Construction Projects", etc. (Zheng et al., 1997, Miao et al., 2006). The situation with soils is more complex. Soils receive many inputs of contaminants – from agriculture, industry, mining, atmospheric deposition, wastes etc, and the storage capacity, fate and behaviour of contaminants in different soils can vary tremendously. In addition, techniques to treat soils are less well understood and the long-term effects of soil contamination on soil processes and ecosystem/human health are much more poorly understood. The public awareness of soil pollution issues is weaker than that of air and water pollution. Government policy makers may lack awareness of soil resources, soil quality, soil functions, and social values of the soil, and measures to actively protect soil have been slower to develop and implement. In recent years, however, the Government has realised the need to safeguard and protect China's soil resources, and regulations are being developed. It is therefore very important and timely to provide data, studies and advice to help China better manage it's soils.

4. Soil pollution in China

4.1 The general situation

Soil pollution has become a widespread and serious problem in many regions of the world (China Environment and Development International Cooperation Committee, 2015 , Mirsal, 2008, Yaron et al., 1996). China has been eager to learn from these experiences. Because the rapid urbanization and huge industrialization expansion in China started later (the past ~30 years), China has been slower to focus on soil pollution (Chen, 1991). Nonetheless, following a survey in 2014, conducted by the Ministry of Environmental Protection and the Ministry of Land and Resources, a National Polluted Soil Investigation Bulletin was published and highlighted the scale of the problem for China. The national standard for soil pollutants (a range of heavy metals and a few selected organic pollutants) was exceeded in >16% of all the soils analysed (Ministry of Environmental Protection of the People's Republic of China and Ministry of Land and Resources of the People's Republic of China, 2014). This survey was comprehensive and included all land use types. It has motivated the Chinese Government to conduct and fund a number of initiatives, including more soil survey work, focussed attention on brownfield site management, investigations into soil treatment and remediation technologies and a national ‘10-point management plan’ (see below for more details).

4.2 Types of soil pollution

In general, soil pollutants are divided into two categories: organic and inorganic. Organics include pesticides, aromatic compounds, solvents, phenols, cyanide, oil, synthetic detergents and so on. Inorganics cover fertiliser nutrients (e.g. N, P, K), heavy metals (e.g. Pb, Cd, Zn, Cu, As, Hg), radioactive elements (Cs, Sr), metalloids (As, Se), and so on (Ministry of Environmental Protection of the People's Republic of China and Ministry of Land and Resources of the People's Republic of China, 2014, Fu et al., 2016) (Figure 14).

Many studies have reported the reasons, sources and pathways of soil pollution in China (Fu et al., 2016, Zhou et al., 2014, Zhu et al., 2008). Figure 14 summarises some of the main sources of soil contamination. This highlights industrial, agricultural, transport related and 'daily life' sources. A useful distinction is between localised high concentrations (e.g. around a point source industry or factory or landfill) and widespread diffusive sources (e.g. fertiliser applications and atmospheric deposition). This thesis considers both – widespread changes in soil pH across regions of China are shown in **Paper III** of this thesis, and discussion of 'hotspots' or brownfield urban sites is the topic of **Paper IV**.

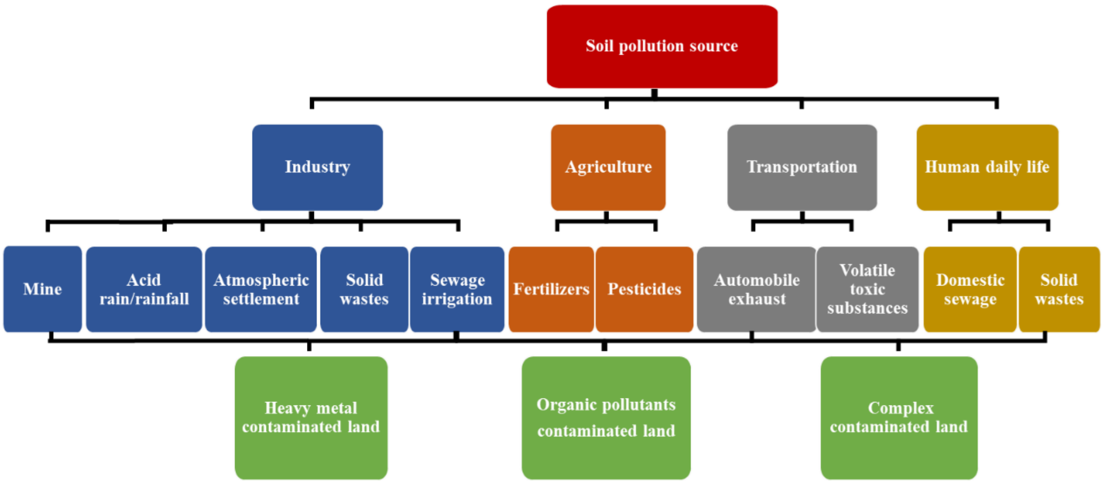

Figure 14. Four typical types of soil contaminated land in China.

Important industry sources include waste water, mining dust, waste residues and electronic wastes, which contaminate soil by various ways (Zhou et al., 2014). Fertilizers and pesticides are two main ways of agricultural pollution source, for example, phosphate fertilizer often contains other chemical elements (Cd, Sr, Ra, Th etc.). In China, the Cd amount in phosphate fertilizer is 0.2-2.5 mg/kg; if this is applied to soil over the long term, this results in Cd contamination (Fu et al., 2016). Agricultural land can also receive inputs from animal wastes and wastewaters. Besides industry and agriculture, transportation and human daily life also result in release of contaminants. Vehicle exhaust and volatile toxic substances are produced in the process of transportation. For example, Pb, Cd, Cu, Zn, etc. are produced by burning of gasoline, fuel and lubricating oil, and from the wear of tires (Zhou et al., 2014). Sources associated with human daily life are mainly from domestic sewage and solid wastes, for example, detergents, pharmaceuticals and personal care products are discharged from domestic wastewater and concentrated into sludges, while solid wastes can contain metals, plastics, packaging materials, etc. Some toxic substances may be produced during the process of waste degradation in landfills etc (Zhou et al., 2014).

This way of classifying soil contaminants is common and well known. However, the relative importance of these sources for China it is not clear yet and studies are needed to identify and accurately quantify these sources: a) as a source to land; b. as a source to human exposure. (i.e. source inventories and exposure inventories are needed). This approach, towards a more quantitative risk assessment is developed in **Paper II** of the thesis. Another key issue is that many contaminants occur naturally in soils, or may be present at low levels without causing any harm. This has resulted in international schemes to define 'background' or 'uncontaminated' levels in soils and guidelines or standards above which concerns are raised. China has been actively developing this understanding and schemes, as discussed in more detail in **Paper I**.

The National Soil Pollution Bulletin released in 2014 – mentioned earlier – is the first step to providing better quantitative information for the country. The exceedance of standards applied to 16.1% of all the many thousands of samples which were analysed. This broke down with different land use types as follows: arable land: 19.4%, woodland: 10%, grassland: 10.4%, unused land: 11.4%. A further breakdown of information is by pollutant type (see Table 1). Inorganic exceedances were greater than organics or 'complex mixtures' of pollutants (Ministry of Environmental Protection of the People's Republic of China and Ministry of Land and Resources of the People's Republic of China, 2014). These results and others are discussed in greater detail in **Paper I** of the thesis. In general, research and management in China has so far focussed on heavy metals in soils (see **Paper I**), while organic chemicals are less well studied and understood (**Paper II**). The behaviour of contaminants in soils is controlled by soil pH and organic matter content, so changes in these properties in soils is critical to understand (**Paper IV**).

Table 1 The exceedance of China's soil quality standards in the samples taken for the 2014 national survey (Ministry of Environmental Protection of the People's Republic of China and Ministry of Land and Resources of the People's Republic of China, 2014).
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Type	Pollutants	Percentage of soils over the guideline values (%)	Proportion of different degrees of pollution (%)			
			Slight	Mild	Moderate	Severe
Inorganic	Cd	7.0	5.2	0.8	0.5	0.5
	Hg	1.6	1.2	0.2	0.1	0.1
	As	2.7	2.0	0.4	0.2	0.1
	Cu	2.1	1.6	0.3	0.15	0.05
	Pb	1.5	1.1	0.2	0.1	0.1
	Cr	1.1	0.9	0.15	0.04	0.01
	Zn	0.9	0.75	0.08	0.05	0.02
	Ni	4.8	3.9	0.5	0.3	0.1
Organic	HCH	0.5	0.3	0.1	0.06	0.04
	DDT	1.9	1.1	0.3	0.25	0.25
	PAHs	1.4	0.8	0.2	0.2	0.2

Notes: HCH: Hexachlorocyclohexane; DDT: Dichlorodiphenyltrichloroethane; PAHs: Polycyclic Aromatic Hydrocarbons.

Values in each column: the percentage of exceedance sites of soil standard value (GB15618-1995) accounting for total surveyed sites (these standard values are discussed in **Paper I**).

The survey generated data according to land use/type. The following summarises some of interesting trends (Ministry of Environmental Protection of the People's Republic of China and Ministry of Land and Resources of the People's Republic of China, 2014):

- Of the 5,846 soil samples taken from 690 sites occupied by enterprises, 36.3% exceeded guideline value. These sites mainly involved ferrous metals, non-ferrous metals, leather products, paper, petroleum and coal, chemicals and medicines, fibres rubber, plastics, and minerals production.
- Of the 775 soil points in 81 industrial wastelands surveyed, 34.9% exceeded a guideline value for at least one pollutant. The main pollutants were Zn, Hg, Pb, Cr, As and PAHs, which mainly involved the chemical industry, mining and metallurgy.
- Of the 1672 soil sites surveyed in 70 mining areas, 33.4% exceeded a standard. The main pollutants were Cd, Pb, As and PAHs. Cd, As and Pb pollution in the soil around non-ferrous metal mining areas was noted as most serious.
- Of the 2523 soil points in the 146 industrial parks surveyed, over-standard points accounted for 29.4%. Among them, the main pollutants in metal smelting industrial parks and the surrounding soil were Cd, Pb, Cu, As and Zn; PAHs were high in and around chemical works.

These reports heightened interest and concern in China about soil contamination problems. However, it should be noted that the survey had a disproportionate focus on urban/industrial areas and was not a survey made in proportion to land use cover. This has the effect of 'biasing' the results towards land more likely to be contaminated. The high exceedances should not be taken as meaning that agricultural, grassland or woodland would have such frequent exceedances of standards, for example.

4.2.1 Soil heavy metal contamination

Hu et al (2014) suggested that heavy metal pollution in China is mainly caused by mining, the manufacture of products that contain metals, sewage irrigation and fertilizer application. They estimated there are >1.5 million sites in China where exposure to heavy metals could be an issue. It is not known if this figure is accurate, but it gives a sense of the potential scale of the issues for assessment in China. Some other key points concerning heavy metals in soils are:

- In urban areas, heavy metals can be present in industrial sites which are subject to redevelopment/re-use (**Paper IV**) (Hu et al., 2014, Luo et al., 2011). It is important to assess when the levels exceed safety values, and when/how to remediate the land;
- In rural areas agricultural, and mining/industrial activities can contaminate crop lands, potentially resulting in transfers of elevated levels into food crops, grazing animals and the human diet (**Appendix 3**);
- Long-term atmospheric deposition, applications of fertilizers and crop residues can cause changes in soil pH, organic matter, reduce soil fertility and reduce crop yield

(**Paper III**). When the soil is contaminated by heavy metals, these underlying changes could affect crop growth and dietary exposures (Zhuang, 2015);

- In summary, reliable source apportionment and risk assessment procedures are needed to support China's management of heavy metals in soils (see **Paper II**).

4.2.2 Organic contaminants in soils

'Organic contaminants' is a general term covering many types and classes of chemicals. For example, at present, there are about 50 kinds of chemical pesticides in widespread use, including organic phosphorus pesticides, organic chlorine pesticides, carbamates, phenoxycarboxylic acids, phenols, and amines. In addition, petroleum hydrocarbons, Polycyclic Aromatic Hydrocarbons (PAHs), polychlorinated biphenyls (PCBs), flame retardants, pharmaceuticals, and solvents are also common organic contaminants that can reach the soil. According to statistics, during 1996 to 2000, the amount of pesticides used in agriculture in China was about 230,000 tons per year. At present, China's pesticide production ranks second in the world, but manufacture can be inefficient and result in local contamination problems (Han and Chi, 2007, Zhuang, 2015). At the moment there is still little regulation, assessment and control of trace organic chemicals in China's soils and ground-waters. There is great interest in remediation techniques, which could remove organics from soils (e.g. bioremediation, soil venting) (Cai et al., 2011), but still little systematic risk assessment to assess whether such techniques are needed or are reliable.

4.2.3 Complex contamination

Long-term industrial production activities and the diversification of production processes can give rise to complex mixtures of contaminants in soils (Ramakrishnan et al., 2011, Dong et al., 2015). For example, a range of heavy metals, tars, oils and PAHs could often occur together in brownfield sites (Wilcke, 2000, Khillare et al., 2007, Yang et al., 2014). The risk assessment typically addresses contaminants individually, but they could exert effects as mixtures and be difficult to remediate (Yang et al., 2017, Gauthier et al., 2014).

4.3 Effects of soil pollution

Soil pollution refers to the occurrence of a chemical substances in soil caused by human activities, which changes soil quality and function, leads to soil degradation, and/or damages the structures of buildings with their bases in soil (Luo et al., 2015, Zhuang, 2015, Mirsal, 2008). The main hazards are reflected in four aspects:

(1) Soil pollution causes severe direct economic losses: According to preliminary statistics (Liu, 2015), there are about 10 million hm^2 of polluted arable land in the country, and 13-16 million hm^2 of farmland is being polluted by organic pollutants (Zhang, 2016). The excess rate of pesticide residues on major agricultural products is as high as 16-20%. The land area occupied by material storage and land destruction is 133,000 hm^2. It has been estimated that various economic losses of about 20 billion yuan are caused every year due to soil pollution that reduces the production of grain by more than 10 million tons (Zhang, 2016).

(2) Soil pollution can cause a decline in quality of crop products: The content of heavy metals such as Cd, As, and Pb in vegetables and fruits can exceed standards or guidelines for food products, because of uptake from soils.

(3) Soil pollution can result in elevated human exposure and health concerns: Soil contaminants in agricultural soils can reach the human diet. Around contaminated sites, contaminants can also transfer via vapours and ground-waters to people living nearby (**Appendix 3**).

(4) Soil pollution can cause other ecological problems: Soil pollution affects the survival and reproduction of plants, soil animals and microorganisms, and endangers normal soil ecological processes and ecosystem service functions. Pollutants in the soil may undergo transformation and migration, and then enter surface water, groundwater, and the atmosphere, affecting the quality of surrounding environmental media.

5. Soil management in China: observations on the current situation and priorities

5.1 Policies and regulations of soil management

To enable soil contamination to be evaluated and managed reliably and to be based on sound science, China needs to develop policies, risk assessment methodologies, soil standards/guideline values, and the necessary management structures and expertise. From an international perspective, the US, Environmental Protection Agency (USEPA) and central/state government have released legislation and technical guidelines to manage soil contaminated land. Examples include the Comprehensive Environmental Response Compensation and Liability Act (CERCLA) (United States Environmental Protection Agency, 1980), the Superfund Amendments and Reauthorization ACT (SARA) (United States Environmental Protection

Agency, 1986), the Small Business Liability Relief and Brownfields Revitalization Act, Superfund guidelines of health risk assessment in contaminated land etc. In the UK, legislation and guidelines were issued to govern contaminated land (e.g. the Inter-departmental Commission for Redevelopment of Polluted Sites (ICRCL) (1979). The UK's procedures are quite mature. For example, between 1988-93, 19% of brownfield sites in the UK were converted into green field sites. Such legislation, guidelines, technical reports and management schemes show the importance attached to soil contamination management internationally (Hu, 2019, Liang and Yang, 2013).

Compared with some developed countries, the regulations on soil pollution in China are not yet so mature, but China has the advantage to learn efficiently from other countries and to potentially reach a leading position internationally (see Chapter IV – the brownfield one - for detailed discussion). Indeed, in May 2016, the State Council publicly released its landmark document - the Action Plan for the Prevention and Control of Soil Pollution (China's '10-Point Soil Plan'). This was included in China's thirteenth 5-Year Plan, the key strategic statement and ambition for the country. It was developed in accordance with the decisions of the Party Central Committee and the State Council and prepared by the Ministry of Environment, Ministry of Agriculture, Ministry of Land Resources, the Forestry Bureau, and the Legislative Affairs Office of the State Council. The 10-Point Soil Plan is ambitious and an internationally leading statement of intent. It targets systematic and comprehensive planning and actions for soil pollution control in China. It will formulate specific "timetables" for soil pollution control in China (Table 2) (State Council, 2016). In August 2018, the Fifth Session of the Standing Committee of the Thirteenth National People's Congress passed the Soil Pollution Control Law, and it was implemented from January 1, 2019. It clearly lists public participation as a basic principle for the prevention and control of soil pollution. It stipulates the provisions of information disclosure, social supervision and environmental damage compensation, and provides a solid legal guarantee for the public's right to environmental information and participation. Table 2 details the ten points.

Table 2. The 10-Point Soil Plan in China released in 2016.

Plan	Content	Remarks relating to urban Brownfield
1	Conduct soil pollution survey and master soil environmental quality	Deeply developing soil environmental quality survey; construction of monitoring network for soil environmental quality; Improving the information management level of soil environment
2	Promoting legislation on prevention and control of soil pollution and establishing and perfecting the system of laws and regulations and standards	Speed up the legislative process; Systematic construction of standard system; Enhancing supervision and law enforcement in an all-round way
3	Implementing classified management of agricultural land to guarantee the environmental safety of agricultural production	Classification of soil environmental quality of agricultural land; Enhancing protection effectively; Efforts to promote safe utilization; Fully implementing strict control; Strengthening soil environmental management of forest and grassland gardens
4	Implementing access management of construction land to prevent habitat environmental risks	Clear management requirements; Implementing regulatory responsibility; Strict access to land
5	Strengthen the protection of non-polluted soil and strictly control new soil pollution	Strengthening environmental management of unused land; Preventing new pollution of

		construction land; Strengthen the control of spatial layout
6	Strengthen the supervision of pollution sources and do a good job in preventing soil pollution	Strict control of industrial and mining pollution; Controlling agricultural pollution; Reducing domestic pollution
7	Conduct pollution control and remediation to improve regional soil environmental quality	Define the subject of governance and restoration; Establishment of governance and rehabilitation planning; To carry out management and restoration in an orderly manner; Supervise the implementation of objectives and tasks
8	Strengthen research and development of science and technology to promote the development of environmental protection industry	Strengthening the study of soil pollution prevention and control; Strengthen the popularization of applied technology; Promoting the development of governance and restoration industries
9	Bringing the Government's Leading Role into Full Play and Constructing a Soil Environmental Management System	Strengthening government leadership; Play a market role; strengthen social supervision; carry out propaganda and education
10	Strengthen objective assessment and strictly investigate responsibility	Define the main responsibility of local government; strengthening department coordination and linkage; implementing corporate responsibility; Strict assessment and assessment;

This national plan helped to set the framework and motivation for this PhD research in 2016. Indeed, Plan points 1, 2, 6, 8, 9 and 10 are addressed by aspects of the research presented here.

Together, this thesis address some ‘big picture’ issues for China which address China’s national needs for:

i. learning current best practice and policies from developed countries which have led on soil pollution research, policies, regulations and management (Papers I, II, V);

ii. defining background and contaminated values, setting soil EQSs and guidelines (Papers I, II);

iii. using and developing risk assessment tools and techniques to adapt/apply to the Chinese situation (Papers II, III);

iv. improving the knowledge and understanding of the role of soil contamination in controlling human exposure (Paper II and III);

v. utilising the large databases on China's soils and their properties, to better understand underlying changes over time and under different land uses (Paper III);

vi. detailed plans for better management of contaminated land (brownfield) sites and the science-based risk assessment methods to evaluate them (Paper II and V).

5.2 Soil environmental quality standards and guidelines

To define and resolve soil pollution problems, it is necessary to be able to define what constitutes 'clean', 'background', 'contaminated' and 'polluted' soils. Many countries, have developed such values. This is a 'cornerstone' to being able to assess and manage soil contamination. Definition of the background levels requires good knowledge of the national soils resource, from survey data (see **Paper I**). Definition of standards requires a sound soil risk assessment framework and knowledge of exposure pathways (see **Paper II**). Depending on the national environmental management and regulatory processes, different countries have different approaches. Examples include the Soil Guideline Values (2009) in the UK, the US Soil Screening Levels (2002), the Intervention Values (2009) in the Netherlands, and Environmental Quality Standards (EQS) (1991 and 1994) in Japan.

In China, EQSs for soils (GB15618-1995) were first officially released in 1995. Since then, relevant soil EQSs have been developed further, by referring to GB15618-1995 as a basic standard, but deriving values for different land uses, soil and crop types etc (Li et al., 2016). For example, the MEP issued a series of standards for 'Green foods' (NY/T391-2000), milk and dairy products (GB/T 1807.5-2003), and edible agricultural products (HJ/T 332-2006). These have been developed to safeguard the food chain and to recognise that risks are different from different soil types, land uses and products. At the time of writing, there are 63 standards related to soil environmental protection in China and the number of standards released by the MEP-PRC has increased, especially in the last 5 years (Li et al., 2016). For example, a series of technical guidelines (e.g. Technical guidelines for risk assessment of contaminated sites (HJ 25.3-2014)) were released to guide soil contamination surveys, risk assessment and monitoring etc. by referring to the US EPA's soil risk assessment system. In 2018 new soil quality guidance (Risk control standard for soil contamination of agricultural land GB15618-2018) was issued, to replace GB15618-1995 and took effect on 1st August 2018. A risk control standard for soil contamination of development land (GB36600-2018) was also issued and came into force at the same time, to protect human health. These will be discussed further in the later chapters.

5.3 Soil risk assessment

The idea of risk-based management for contaminants in soils has developed rapidly and is widely recognized in developed countries (Ferguson and Denner, 1993, Petts et al., 1997). This is a management strategy to protect human health and ecological safety under different land uses. Due to its practicality and operability, developed countries such as the US, the UK, Canada, the Netherlands, and Australia have established soil contaminant risk assessment guidelines and risk-based soil EQSs.

The risk-based soil EQS value is generally derived from the generic assessment criteria of the potential risk of sensitive receptors on contaminated soil, and is used to provide a basis for the preliminary determination of contaminated sites as the next step in the management of contaminated sites. The toxicity of pollutants and the exposure pathway(s) of sensitive receptors are the key factors in the derivation process (Environment Agency, 2004). Figure 15 summarises the general approach.

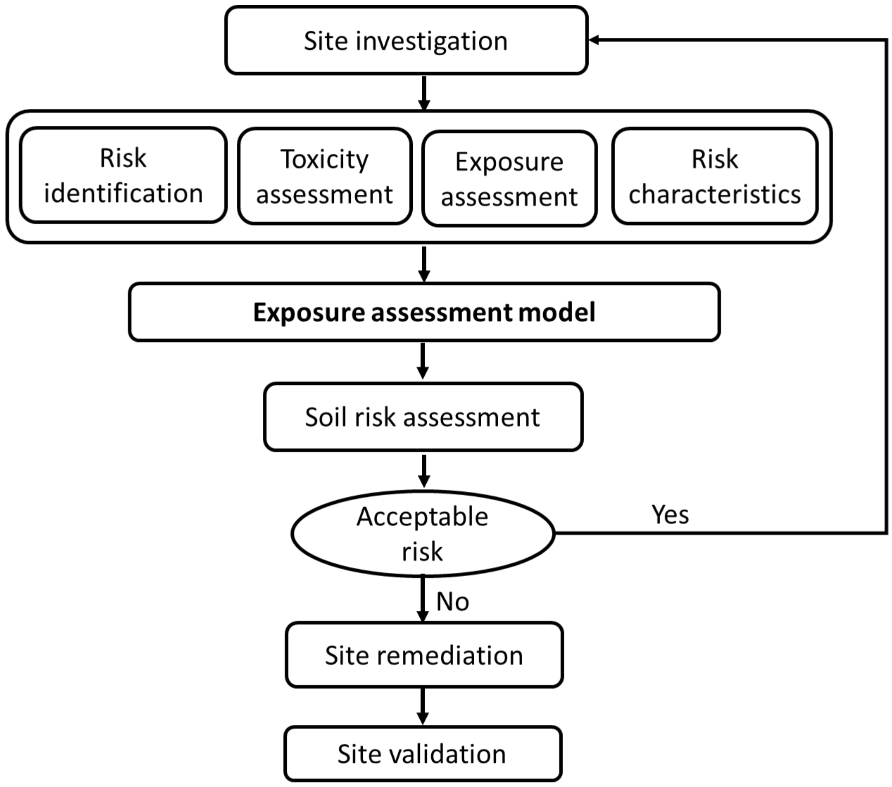


Figure 15. Example work flow of soil contamination management and the role of exposure assessment model (Environment Agency, 2004).

Soil risk assessment procedures are used and evaluated in the thesis. **Paper II** makes some comparisons of risk-based models used in China and the UK to derive Soil Screening Values (SSVs) for different contaminants. **Appendix 3** considers the assessment of total human environmental exposure to metals (from food, water, air, soil).

5.4 The need for reliable surveys, data and interpretation

The scale of China's economic growth, the size of the country and its population, and the diversity of its climate and ecosystems mean there is great demand to understand the spatial and temporal variability in the Chinese environment (Liu and Diamond, 2005). Following scientific and regulatory focus on China's air and water quality, the Government is now prioritising soil quality (State Council, 2016). Point 1 of the 10-Point Soil Plan specifically focusses on the need for good survey data for China. Knowledge and effective management of China's basic soil resources is essential, requiring careful and systematic surveying of soil elements, soil properties and related environment factors. For example, soil pH and soil organic matter (SOM) are critically important properties of soils (Brady et al., 2008, Sposito, 2008). Understanding their variability, range and any underlying changes is fundamentally important for agriculture/food security, land use management and the environmental sciences. This is investigated in detail in **Paper III**.

Understanding the cause and condition of soil pollution in China (**Paper I**) is the prerequisite and basis for all control work. However China lacks comprehensive and detailed soil pollution data. It is an indispensable first step to focus on detailed investigation and continuous monitoring of the pollution situation of agricultural land and enterprises in key industries in China, and to conduct detailed and reliable surveys. Surveys can form a large national soil database, which provide the solid platform to observe and predict soil dynamic changes, and provide important guidance for soil management (**Paper III**).

6. Summary and specific objectives

This Chapter has made it clear that:

- China is reliant on its soils resources to support the health and wellbeing of its people, its plans for economic development and its natural environment;
- The Chinese Government has made clear its intentions to tackle soil contamination problems, improving and safeguarding the nations soils resource;

- It is a critical time for China to develop the tools, data, expertise and approaches to manage the issues of soil contamination.

This background and context provided the motivation for the research described in this thesis. The thesis broadly addresses the following objectives:

(1) to understand the current priorities and define background levels and standards for heavy metals in China's soils;

(2) to compare methodologies to derive SSVs in China and the UK and make recommendations on the approaches;

(3) to assess total human environmental exposure and risk assessment of residents for metals, including assessing the pathways for soil-borne pollutants;

(4) to investigate changes in soil pH and soil organic matter over recent decades for different land uses and regions;

(5) to address the history and management of Chinese brownfield sites by learning from developed countries, highlighting priorities and suggesting improvements for future management and government planning.

To achieve the research objectives, more than 300 publications were reviewed to obtain information about the situation, policy, management and soil environmental standards in China and developed countries.

The specific studies presented here:

(1) Review the current priorities and define background levels and standards of soil heavy metal in China, which has been discussed in **paper I**;

(2) Compare SSVs derived in China and the UK in **paper II** and assess total human environmental exposure and risk assessment of residents for metals in **Appendix 3**;

(3) Investigate the change of soil pH and soil organic matter in different land uses and regions in **Paper III**;

(4) Illustrate the Chinese brownfield history and management by learning from developed countries in **Paper IV** for future brownfield management and government planning.

Paper III In the final Chapter of the thesis some conclusions and recommendations are made, which are aimed at advising on the implications for better soils management in China and future research studies.

I

Soil contamination in China: current priorities, defining background levels and standards for heavy metals

Yiming Sun (1), Hong Li (1), Guanlin Guo (2), Kirk T. Semple (1), Kevin C. Jones (1)

(1) Lancaster Environment Centre, Lancaster University, Lancaster, LA1 4YQ, UK

(2) State Key Laboratory of Environmental Criteria and Risk Assessment, Chinese Research Academy of Environmental Sciences, Beijing 100012.P.R.China

Abstract

The Chinese Government is working to establish an effective framework in managing soil contamination. Heavy metal contamination is key to the discussion about soil quality, health and remediation in China. Soil heavy metal contamination in China is briefly reviewed and the concepts of background values and standards discussed. The importance of contaminated land and its management for China food security and urbanization are discussed. Priorities for China's next steps in developing an effective research and management regime are presented. We propose that critically important to the science-based risk assessment of contaminants in soils is the incorporation of speciation and bioavailability into the measurement and evaluation criteria. Consideration of soil biology/ecological endpoints will be necessary to protect ecosystem health. National and regional/local scenarios of land use type/usage will address residential/urban re-use of industrial land as well as varying agricultural scenarios.

Keywords: Soil contamination, soil sampling, risk assessment, land use, China, United Kingdom

1. Introduction

Soil pollution refers to the occurrence of some substances in soil caused by human activities, which can change soil quality and function, lead to soil degradation, damage basic soil structures and has the potential to harm human and environmental health. Soil pollution has been identified as a key national priority in China, with an increase of reports on agricultural land and human health affected by soil pollution (Luo et al., 2015). With economic growth and industrial restructuring in China, soil pollution from abandoned sites in urban areas has also drawn attention and concern regarding the safety of human settlements and human health in industrial and brownfield sites (Cao and Guan, 2007; Luo et al., 2015). According to the National Investigation Bulletin of Soil Pollution Status (NIBSPS) issued by the Ministry of Environmental Protection of the People's Republic of China (MEP-PRC), investment in soil remediation will reach up to RMB 4,633,000 million (£ 526,000 million). This is a huge financial commitment, so it is critical that sound science and knowledge are applied to the decisions that determine how this money will be spent. There is still work to be done in China to improve information to define soil background conditions and pollution status, the relevant science and policies needed to set soil quality standards, the assessment system for site evaluation and soil remediation strategies and technologies (State Council, 2016). The importance of soil pollution and degradation in China has now been recognized at the highest level, with specific requirements included in China 13th Five-Year National Development Plan and the Fifth Plenum of the 19th Central Committee of the Communist Party of China (MEP-PRC, 2016).

This paper focusses on an assessment of some of the priorities relating to heavy metals in

Chinese soils. Soil heavy metal pollution has become a widespread and serious problem globally. Heavy metals are present naturally in soils, but elevated levels may be derived from agricultural activities, urbanization, industrialization and other human activities. To define and resolve pollution problems, it is therefore necessary to be able to define what constitutes ‘clean’, ‘background’ and ‘contaminated’ and ‘polluted’ soils. Following surveys and analysis of heavy metals in soils, many countries such as the United Kingdom, the United States and the Netherlands have developed such values. Depending on the national environmental management and regulatory processes, different countries have different approaches. Examples include the *Soil Guideline Values (2009)* in the UK, the US *Soil Screening Levels (2002)*, the *Intervention Values (2009)* in the Netherlands, and *Environmental Quality Standards (EQS) (1991 and 1994)* in Japan.

China first developed its own Soil Environmental Quality Standards (SEQS) in 1995 (GB15618-1995) (Xia, 1996). So far, there are 63 current standards related to soil environmental protection in China and the number of standards released by the MEP-PRC has increased, especially in the last 5 years (Li et al., 2016). Following China previous focus on air and water quality, the Government has now turned a focus onto soils and groundwater, publishing a landmark ‘10-Measures for Soil Pollution Action Plan’ in 2016 (State Council, 2016). Its purpose is to manage, control and prevent soil pollution, to gradually improve soil quality in China. The Plan’s first action recommends conducting surveys on soil pollution to better define the status of China’s soil resources. GB15618-1995 first defined SEQS values for 8 heavy metals in China to apply to the whole country. Later, relevant soil quality standards in China were developed by referring to GB15618-1995 as a basic standard (Li et al., 2016). For example, the MEP-PRC issued a series of standards, such as ones for ‘Green food-technical conditions for environmental areas’ (NY/T391-2000), but these are rarely applied in practice. In contrast, European countries conduct soil pollution control mainly through a series of systematic assessment methods. These are based on different land use type, soil specificity or local environmental factors for understanding the risks either to the environment or human health. In China over the past 20 years, a general soil standard value (GB15618-1995) was applied to the whole country, without considering soil specificity and integrated environmental factors. During this period, in order to meet development needs, some regional standards for soil risk assessment were also set; for example, Beijing issued screening levels for soil environmental risk assessment of sites (DB11/T 811) in 2011.

A particular challenge for China is the country size and hence the range of soil types and conditions. Heavy metal concentrations vary naturally in soils, as a function of the geology, climate, land use etc. Hence, the Soil Environment Background Value (SEBV) will vary across the country. SEQS values were originally set nationally, so now there is an important discussion about whether different SEQS are needed regionally/locally. When managing contaminated sites, SEQS and SEBV will affect the selection, formulation and cost of remediation strategies. Internationally, different countries have prioritized soil pollution and management in different ways. China has been eager to learn from this (Luo et al., 2015; Wang et al., 2005; Wen et al., 2010; Xia and Luo, 2007) and – given the Government’s stated aim to manage its soil pollution problems effectively - has the opportunity to put in place sound, strong policies and management structures. An interesting comparison is with the UK, which has a long history and legacy of contaminated land problems and a mature environmental regulatory system (Luo et al., 2009; Wu, 2007). The 10-Measures Soil Pollution Action Plan strongly recommended that: surveys of the soil pollution situation be conducted; regulations and laws of soil pollution prevention be amended; soil pollution control and remediation be promoted; and a control system to prevent soil pollution be introduced (State Council, 2016).

Given the highly topical nature of soil contamination issues in China, in this paper we focus on the following questions: 1. What is the situation of soil heavy metal contamination in China? 2. What factors affect the background value of heavy metals in soils? 3. What are the background values of selected heavy metals and how do they compare between China and the UK? 4. What can we learn from the UK about soil survey methodologies and soil environmental standard assessment? 5. What are future priorities and next steps for China in its management of soil pollution?

2. Heavy metal soil pollution in China

As in other countries, key sources of heavy metals to Chinese soils include: metal mining and smelting; industrial activities, power generation; agricultural activities, including fertilizer and animal manure amendments; waste disposal activities; urbanization, transportation. In some regions, high contamination of the soil occurs around point sources, for example, mines and smelters – giving high but generally localized problems. In other situations, for example, agricultural soils, contamination may be lower, but important as a direct route of food contamination (Fu et al., 2016; Zhou et al., 2014; Zhu et al., 2008). In some countries, inventories have been published which estimate the relative importance of these different sources to the national soil resource. This approach would be very helpful in China, because it provides a scientific basis to prioritize source reductions; we are not aware that this exercise has been performed in China yet. The distinction between 'high level hotspots' of contaminated land (e.g. brownfield sites; mines) and diffusive agricultural sources may also be important in management. For example, should there be different standards for agricultural land? Can brownfield sites be cleaned by simply excavating and removing dirty soils from important areas of re-development? (Cao and Guan, 2007; Li et al., 2015; Wang et al., 2014; Wei and Yang, 2010).

Over the past decades, heavy metal contamination has increased worldwide, following large-scale mining and industrial releases (Li et al., 2014). Ultimately much of the metal from such activities reaches the soil, via wastes, disposal and atmospheric deposition. China has undergone huge and rapid urbanization and industrial expansion over the past thirty years, which will have resulted in increased release of heavy metals to the environment, and the burden of metals held in surface soils (Chen, 1991). It has been estimated that nearly 20 million hectares of arable land has been polluted by heavy metals, such as Cd, Pb, Cu, and Zn in China, accounting for approximately 20% of the total arable land area (Lin, 2004; Zhao et al., 2007). Although information is not available on a site-specific basis, national soil surveys conducted by the MEP-PRC and the Ministry of Land and Resources of the People's Republic of China (MLR-PRC) (2005 to 2013) concluded that 16.1% of national land (based on sampling points, including arable land, some woodland, grassland, unused land, construction land) was contaminated (i.e. exceeded background values), of which >80% were exceeded by inorganic contaminants. Contamination may have been with heavy metals and other inorganic contaminants, and/or with organic chemicals. Cd was responsible for most exceedances, accounting for 7% of national land. It should be noted that there was a sampling bias, because soils were not sampled in proportion to the national land coverage. Nonetheless, the Ministries concluded that “The overall situation looked not optimistic, of which, the situation of arable land and industrial abandoned land are the most severe”. In addition, in recent years, many reports in China have highlighted contamination and poisoning of animal and human health through the food chain. Prominent cases include Cd in rice, Pb and As poisoning incidents (Song et al., 2013). Hu et al. (2014) concluded that Pb, Cr, As, Cd and Hg constituted the five most important heavy metal contaminants in Chinese soils.

3. Soil background values and factors that affect them

‘Soil environment background values’ (SEBV) are the concentration of elements or components in soil with little influence from human activities (Connor and Shacklette, 1975; Wang and Yang, 1990). They reflect the underlying geology and soil formation processes; hence they vary between locations and are commonly expressed as a range of values for a particular country or region. The background value could change over time, if environmental processes (including background human activities) affect the burden in the soil. So, absolute uncontaminated or pristine soils may be difficult to identify, since industrial activities have emitted heavy metals and other contaminants into the atmosphere. In general, the SEBV is a relative concept (Xia and Luo, 2006). The geochemical background refers to the normal abundance of an element in barren earth material or the normal range of an element in certain areas. The concept of geochemical background aims to distinguish the normal and abnormal concentration of elements. In the exploration geochemistry field, it may be an indicator of an ore occurrence, while for environmental geochemistry, it may be an indicator of a contaminated or insufficient element (Cheng et al., 2014; Hawkes and Webb, 1963). It is assumed that the range of SEBVs give ‘clean’ soils where there are no adverse ‘pollution’ effects. Hence, contaminated soils are defined with levels of heavy metals (and other constituents) above the SEBV (Xia and Luo, 2006). This is why it is so important to conduct carefully designed surveys of contaminants in soils, because precise definition of the SEBV will determine whether the soil is contaminated and to what degree.

As just noted, the SEBV is affected by various abiotic and biotic factors that change in space and time. For example, parent materials, soil chemical properties, topographic factors, hydrological factors, human activities, geological factors, weathering and leaching conditions of parent material driven by climatic factors etc. (Chen and Wang, 1987; China National Environmental Monitoring Centre, 1990; Fu and He, 1992, Liang and Zhang, 1988). Parent materials are a direct and main factor influencing the SEBV in many studies (Chen and Wang, 1987; Nair and Cottenie, 1971; Oertel, 1961). Climatic factors indirectly affect the SEBV by controlling weathering and leaching processes. Chen and Wang (1987) found soil types and parent materials to be the main drivers that led to a decline from south to north in Shanxi province and from southeast to northwest, based on a survey of distribution trends and factors affecting the SEBV. Deng et al. (1986) showed that the main factors affecting background values are different from region to region in Beijing; topographical characteristics are the main driver in plain regions, while in mountain regions it is parent materials. Hydrological and topographical factors directly influence soil formation factors, soil surface runoff, soil surface temperature, the degree of surface erosion etc., and then influence soil parent materials and soil elemental composition. For example, fine clays which are richer in heavy metals than sands and silts, will accumulate in floodplains and result in higher concentrations than in hillslope soils. Thus, the determination of SEBVs needs to be based on statistical analyses, with careful consideration given to sampling design and soil sample collecting, statistical tests of sample frequency distribution, data distribution patterns and eigenvalues, to show the range of background values with a given confidence interval (China National Environmental Monitoring Centre, 1990). Further details on these issues are given later in the paper.

3.1 Deriving soil background values in China and the UK

The UK has a widely varying geology for a comparatively small country, areas of heavy metal mineralization, a long history of mining and industrial activity, a legacy of soil contamination, and a lot of experience in the management and regulation of soil contamination. The development planning process has been used to deal with contaminated sites since the creation of the current UK land use planning system in 1947 (Luo et al., 2009). Soil remediation of contaminated sites has been carried out since the 1960s, often with low cost and pragmatic solutions (Ferguson, 1999). In 1976, the Inter-departmental Committee on the Redevelopment of Contaminated Land (ICRCL) was established as the first central institutional mechanism to clearly address this issue. In the Environmental Protection Act of 1990, the provision for registers of potentially contaminated sites was included. The current system of regulation was created in the 1995 Environment Act. In 2005, the Contaminated Land Exposure Assessment (CLEA) model was published with a series of soil guideline values and toxicological reports on key soil contaminants. Hence, the UK has developed a relatively effective management system from longstanding practice. China is experiencing a situation like the UK had several decades ago, although on different scales. As noted earlier, mining and industrial activities have become extensive in parts of China, whilst urban areas have expanded and re-developed on sites with a legacy of contamination. In rural areas, there are several examples where agricultural land and community areas have become contaminated too (Liu et al., 2013; Shi et al., 2008; Zhuang, 2015).

The UK approach to contaminated land management is underpinned by a series of comprehensive surveys of soil contaminants, which allow a clear definition of the typical levels, ranges and distributions of elements. It is therefore useful to compare the situation with China, which has undertaken national surveys too and is planning further work of this kind.

4. Experience from surveying UK soils

In the UK, there have been different national soil surveys in past decades. In the late 1970s, soil information in England and Wales was incomplete, knowledge of regional soil geochemistry was limited, available soil maps only covered ~25% of the area, and the existing information was not based on a representative and unbiased sampling strategy. Thus, between 1978 and 1983, the National Soil Inventory (NSI) carried out a survey of soil background metal concentrations in England and Wales (McGrath and Loveland, 1992).

In 1996, the Royal Commission on Environmental Pollution (RCEP) published the nineteenth report on the Sustainable Use of Soil, which stressed the need for the assessment and monitoring of soil quality, including certain chemical and physical attributes, and some biological parameters. A second survey – the so-called Countryside Survey – therefore began in 1998. The purpose has been to assess and monitor soil quality over time, by returning to the same locations over several years (Barr et al, 2003).

A third survey was conducted in 2011/12, to give guidance on normal levels of contaminants - to support revision of the Part 2A Contaminated Land Statutory Guidance. This was conducted by the British Geological Survey (BGS) in England and Wales (Johnson et al., 2012). A summary comparison of the survey designs and methodologies is presented in Table 1.

Table 1. Comparison of sampling methods across three national soil surveys in the UK.

	NSI	CS 2000	BGS
Study area	England, Wales	England, Wales, Scotland	England, Wales
Time	1978-1983	1998-2000	2011-2012
Sampling density	5 km × 5 km	1 km × 1 km	G-based: Urban: 1 km × 1 km; rural: 2 km^2; NSI (XRFS): 5 km × 5 km
Quadrat size	20 m × 20 m	14 m × 14 m	20 m × 20 m
Sample number (per quadrat)	1	3	5
Sampling depth	0-15 cm deep topsoil	15 cm deep x 8 cm dia.	topsoil: 0-15 cm; surface soil: 0-2 cm; deeper soil: >30 cm
Investigated elements	Al, Ba, Cd, Cr, Co, Cu, Fe, Pb, Mg, Mn, Ni, P, K, Na, Sr, Zn	Cd, Cr, Cu, Pb, Ni, V and Zn	As, Cd, Cu, Hg, Ni and Pb
Number of soil samples	5691	1081	42133
Analytical value	Range, mean, median, maximum, minimum	Mean, standard deviation, median, maximum value and minimum value	Mean, range, minimum, maximum

Notes: NSI: National Soil Inventory; CS2000: Countryside Survey 2000; BGS: British Geographical Survey.

As Table 1 shows, ~50,000 UK surface (0-15 cm) soil samples have been taken and analyzed, albeit with slightly different purposes and with a focus on different land use types. A comparison of a selection of heavy metals from these surveys is given in Table 2.

Table 2. Comparison of range, mean and median of four comparative heavy metals (Cd, Cu, Ni, Pb) from the results of NSI, CS2000 and BGS surveys (all concentrations in mg/kg).

Cd	**Range**	**Mean**	**Median**
NSI	<0.2-41	0.8	0.7
CS2000	0-11	0.49	0.3
BGS	0.3-20	0.5	0.3
Cu	**Range**	**Mean**	**Median**
NSI	1.2-1508	23	18
CS2000	0.3-448	18	14
BGS	<1-5326	27	20
Ni	**Range**	**Mean**	**Median**
NSI	0.8-440	25	22.6
CS2000	0-1890	24	16.3
BGS	1-506	25	23
Pb	**Range**	**Mean**	**Median**
NSI	3-16338	74	40
CS2000	1.3-20600	88	37
BGS	3-10000	72	41
pH	**Range**	**Mean**	**Median**
NSI			
CS2000	3.4-8.71	5.72	5.58
BGS			
Soil organic matter	**Range**	**Mean**	**Median**
NSI			
CS2000	2-98.02	29.11	12.57
BGS			

Notes: Median values are the most commonly reported values for background soils. They are more meaningful than mean values, which can be biased by a few extreme polluted values. See, for example Davies (1983) paper on soil Pb values.

Median values (Davies, 1983) provide a good way to compare the data: the 3 surveys gave the following values for the elements presented in Table 2: Cd – 0.7, 0.3 and 0.3; Cu – 14, 18 and 20; Ni, 23, 16 and 23; Pb – 40, 38 and 41. The main conclusions from Tables 1 and 2 are: i. despite different sampling and analytical methods, the general soil quality determined by the 3 surveys is very similar. ii. The sampling, preparation procedures and – crucially - the large number of samples taken provides a robust way to determine the typical range of heavy metal concentrations in soils.

5. Surveying Chinese soils

The earliest research on SEBVs in some selected city areas of China (Beijing, Nanjing, Guangzhou etc.) was in the mid-1970s. Subsequently, in 1978, SEBVs for 9 elements in agricultural soils and crops were surveyed in 13 provinces. In 1982, a background value survey was listed in the national key scientific and technological projects, which was carried out in a few of the main climate zones in northeast China, Yangzi River basin, Pearl River Basin etc. In 1990, a large-scope and systematic survey for SEBVs was carried out across the whole of China, covering all 29 provinces, cities and autonomous regions. These survey data were summarized in a book entitled China Soil Element Background Value (China National Environmental Monitoring Centre, 1990). From 2005, the MEP-PRC and the Ministry of Land Resources launched a national soil pollution survey to capture the distribution data and to look for changes in the 20 years since the 1990 survey. It covered all arable land and parts of the woodland, grassland, unused and construction land. In 2014, a national bulletin on site-specific soil pollution status was published, to summarize the pollution situation without detailed site-specific or soil survey data.

A comparison of Tables 2 and 4 shows close agreement for Chinese and UK Cu and Ni background values. The median value for Cd in UK soil is ~0.3 mg/kg, about 3 times higher than that of the Chinese 1990 survey. UK Pb median values were ~40 mg/kg, against a Chinese median of 24 mg/kg. This might be explained by the UK's long history and density of Pb mining and inefficient smelting operations (Davies, 1983).

Table 3. Sampling details for the 1990 national soil survey in China

Study area	Covered 29 provinces, cities and autonomous regions
Time	1990
Sampling density	East areas: 30×30 km^2 per study point; Central areas: 50×50 km^2 per study point; west areas: study point from 80×80 km^2 per study point
Soil profile	1.5m × 0.8m × 1.2m (Length × width × depth)
Sample depth	A layer: 0-20cm, B layer: 50cm, C layer: 100cm
Investigated elements	As, Cd, Co, Cr, Cu, F, Hg, Mn, Ni, Pb, Se, V, Zn
Number of soil samples	4095
Analytical value	Maximum value, minimum value, arithmetic mean and geometric mean

Table 4. The range, mean and median of four comparative heavy metals from the 1990 China soil survey (all concentrations in mg/kg).

China soil survey 1990	Range	Mean	Median
Cd	0.001-13.4	0.097	0.079
Cu	0.33-272	23	21
Ni	0.06-628	27	25
Pb	0.68-1143	26	24

6. Methodologies for determining background concentrations in soil

6.1 Statistical methods used for UK soils

6.1.1 Countryside Survey 2000

All elements were analyzed in different environments, classified according to Land Class and eighteen broad habitats and major soil groups and Countryside Vegetation System Aggregate Vegetation Class. Data are typically presented as figures (box-plots, scatterplots, frequency histograms etc.) to summarize the variation in different environmental factors. Mean values, standard deviations, median, maximum value and minimum values are commonly calculated to represent the primary analysis (Black et al., 2002).

6.1.2 National Soil Inventory

For all variables, the range, mean, median, maximum, minimum, skewness and kurtosis were calculated for both transformed and log10-transformed data (except for pH). Box plot analysis was performed for the data of Cd, Co, Cr, Cu, Ni, Pb and Zn. Correlation analysis was performed on soil element concentrations (log10-tranformed data). Principal component analysis (PCA) was performed on all datasets, including all elemental concentrations, organic carbon and pH to provide an overall view of the relations among variables. Simple or multiple linear regression analysis was used to exclude the outlier data (McGrath and Loveland 1992).

6.1.3 British Geological Survey

Values for contaminant domain normal background concentrations were calculated by a study of a contaminant’s population distribution. Skewness coefficient and octile skew were used as statistical measures. Percentiles for the domain data sets for each contaminant were generated along with calculations of percentile confidence intervals. The upper limit for a normal background concentration has been as the upper confidence limit of the 95th percentile (Johnson et al. 2012).

6.2 Statistical methods used in Chinese soils

Relevant information (soil types, parent materials, topography, latitude, longitude, vegetation, land use types, administrative regions etc.) of 4,095 typical soil profiles, together with the chemical analytical data were stored in a database of Chinese soil background values. In summary, soil types were divided into 41 statistical units, parent materials were divided into 21 units, and administrative regions were divided into 34 units, so in total, every element has 97 statistical units. Frequency distribution graphs are available for different elements, with the maximum, minimum, arithmetic mean and geometric mean values were presented. For elements with a log-normal distribution, the geometric mean (M) was used to represent the data distribution, the geometric standard deviation (D) to represent the level of dispersion, and M/D^2-MD^2 for the range of 95% confidence interval. For the elements with a normal distribution, the arithmetic mean ($\bar{x}$) was used to represent the data distribution, the arithmetic standard deviation (s) for the level of dispersion and $\bar{x}\pm2s$ for the range of 95% confidence interval (China National Environmental Monitoring Centre, 1990).

6. Soil standards

Environmental Quality Standards (EQSs) for soils (GB15618-1995) in China were officially released in 1995. They were derived based on several factors: data on the soil background in China; data from soil ecological tests; data from geographically anomalous areas in China and information on soil standards or guidelines from abroad (MEP-PRC, 1995; Wu and Zhou, 1991).

These EQSs set the maximum acceptable concentration of pollutants and relevant monitoring methods in the soil based on different soil functions/uses, protection targets and soil properties. Three types of standard were set: Type I is protective of soils in national nature reserves, centralized drinking water resources, tea plantations, pasture and other protected areas, and the goal is to basically maintain the natural background level. Type II is applicable to the soil in general farmland, land for growing vegetables, tea plantations, orchards, pasture etc., where the goal is to not cause harm and pollution to plants and the environment. Type III is applicable to woodland soil, and farmland soils near to high background soils of more pollutant capacity and mineral fields, where the goal is basically to not cause harm and pollution to plants and the environment. Chinese EQSs take account of the soil pH value, cropping pattern and soil cation exchange capacity (see Table 5 for details) (MEP-PRC, 1995).

Table 5. Soil environment quality standards adopted in China for general farmland (mg/kg). See text for definition of Type I, II and III.

Standard		Type I soil	Type II soil			Type III Soil
Soil pH		Natural background	<6.5	6.5~7.5	>7.5	>6.5
Cd		0.20	0.30	0.30	0.30	1.0
Hg		0.15	0.30	0.50	1.0	1.5
As	Paddy field	15	30	25	20	30
As	Non-irrigated farmland	15	40	30	25	40
Cu	Farmland	35	50	100	100	400
Cu	Orchard	--	150	200	200	400
Pb		35	250	300	350	500
Cr	Paddy field	90	250	300	350	400
Cr	Non-irrigated farmland	90	150	200	250	300
Zn		10	200	250	300	500
Ni		40	40	50	60	200

In order to protect agricultural soil, control agricultural soil contamination risk, safeguard agricultural product security, the normal growth of crops and soil ecological environment, China has been working on the development of new soil environmental quality standards. Twenty years after the release of GB15618-1995, the new soil quality guidance (Risk control standard for soil contamination of agricultural land GB15618-2018) was issued in 22nd June 2018, to replace GB15618-1995 and to take effect on 1st August 2018 (Table 6 & 7). This standard regulates the soil risk screening value and risk intervention value in agricultural land, and the requirements of monitoring, implementation and supervision. These values were derived from human health risk assessment procedures. In addition, a risk control standard for soil contamination of development land (GB36600-2018) was also issued, to come into force at the same time to protect human health and living environmental security.

Table 6 Screening values of soil pollution risk of agricultural land (basic items) (mg/kg).

Pollutant		Risk screening value			
		pH≤5.5	5.5<pH≤6.5	6.5<pH≤7.5	pH>7.5
Cd	Paddy	0.3	0.4	0.6	0.8
Cd	other	0.3	0.3	0.3	0.6
Hg	Paddy	0.5	0.5	0.6	1
Hg	other	1.3	1.8	2.4	3.4
As	Paddy	30	30	25	20
As	other	40	40	30	25
Pb	Paddy	80	100	140	240
Pb	other	70	90	120	170
Cr	Paddy	250	250	300	350
Cr	other	150	150	200	200
Cu	Paddy	150	150	200	200
Cu	other	50	50	100	190
Ni		60	70	100	190
Zn		200	200	250	300

To quote the new guidance: ‘The value of the main pollutant content in the soil when the quality and safety of edible agricultural products, crop growth or the soil ecological environment are or may have adverse effects. If the content of pollutants in soil is lower than this value, the risk of soil pollution such as non-conformity of quality and safety standards in edible agricultural products, may generally be ignored. If there may be a risk of soil pollution, soil environmental monitoring and coordinated monitoring of agricultural products should be strengthened, and in principle, safe use measures should be taken. Agricultural land is classified into three types – arable land (paddy, irrigated land, dry land), garden (orchard, tea garden) and pasture (natural pasture and artificial pasture). In this standard, 'others' include all kinds of land except for paddy.'

Table 7 Intervention values of soil pollution risk of agricultural land (mg/kg).

Pollutant	Risk intervention value			
	pH≤5.5	5.5<pH≤6.5	6.5<pH≤7.5	pH>7.5
Cd	1.5	2.0	3.0	4.0
Hg	2.0	2.5	4.0	6.0
As	200	150	120	100
Pb	400	500	700	1000
Cr	800	850	1000	1300

Notes: Risk intervention value in this standard refers to the value of the main pollutant content in the soil when it causes or may cause serious effects on the quality and safety of edible agricultural products. If the content of pollutants in the soil exceeds this value, the risk of soil pollution, such as non-compliance with quality and safety standards, is high, and strict control measures shall be taken in principle.

A particular challenge for China at the present time is that specific areas are considering variants to the national standards, to reflect their particular challenges. For example, areas (jurisdictions at province or city level) with high background values, or particular soil/crop systems may wish to adopt more pre-cautionary limits. In other situations, standards may be considered as targets for remediation of contaminated sites. Although not published from the national survey of China, there are many studies which have reported geographical variations, with some provinces having high background values, for example (Cheng et al., 2014; Cheng and Tian, 1993; Dong et al., 2007; He et al., 2006; Pan and Yang, 1988). Hunan Province is an interesting example. It has a long history (~2,700 years) of non-ferrous metal mining and metal resources, which began to be extensively exploited in the 1980s. With industrial development, many incidents and impacts from heavy metal pollution have been widely reported in this area. For example, in June 2014, 315 children living around Dapu industrial area in Hengdong county were reported with excessive Pb concentrations in their blood, 10 of which had been sub-chronically poisoned. Another heavy metal survey published in November 2014 from an environmental protection organization showed that the As content of river sediments exceeded national standards by 700 times and the Cd content in some paddy soils exceeded the standards by 200 times in the Sanshiliuwan mining area from Chenzhou City (Cao and Li, 2014). Regulations have been issued by the province – for example - an 'Implementation Plan (2012-2015)' of heavy metal pollution control in the Xiangjiang river basin, which has been set to close illegal factories, control industrial pollution sources and decrease heavy metal emissions and remediate the legacy contaminated sites. In 2016, standards for soil remediation of heavy metal contaminated sites (DB43/T1165-2016) were issued by the Environmental Protection Department of Hunan and Hunan Provincial Bureau of quality and technical supervision. These provided the remediation standard for 11 heavy metals in residential land, commercial land and industrial land (Table 8). These remediation targets are higher than the national standard in

GB15618-1995. For example, DB43/T1165-2016 values are: Cd-7, 20, 20 in residential land, commercial land and industrial land, respectively; GB15618-1995 values are: Cd-0.3, 0.3, 0.6, 1 in Standard II pH <6.5, 6.5-7.5, >7.5, Standard III, respectively; GB15618-2018 screening values are: Cd-0.3, 0.4, 0.6, 0.8 for paddy pH≤5.5, 5.5<pH≤6.5, 6.5<pH≤7.5, pH>7.5, respectively. Details of how Hunan's standards were derived are not clear, but they may be pragmatic and risk-based.

Table 8. The remediation standard of heavy metal in contaminated sites for Hunan Province (first 3 columns), compared to the national standards (all concentrations in mg/kg).

Elements	Residential land	Commercial land	Industrial land	National Standard I soil	National Standard II soil			National Standard III soil
pH					<6.5	6.5-7.5	>7.5	>6.5
Pb	280	600	600	35	250	200	200	400
As	50	70	70	15	30	25	20	30
Cd	7	20	20	0.2	0.3	0.3	0.6	1
Hg	4	20	20	0.15	0.3	0.5	1	1.5
Cr	400	610	800	90	250	300	350	500
Cr^{+6}	5	30	30	--	--	--	--	--
V	200	250	250	--	--	--	--	--
Mn	2000	5000	10000	--	--	--	--	--
Cu	300	500	500	35	50	100	100	400
Zn	500	700	700	100	200	250	300	500
Sb	30	60	60	--	--	--	--	--

Note: see Table 5 for more information on the national standards.

EQSs are considered impractical as remediation targets, because of the high costs/time required to achieve such a level of clean-up (Cai et al., 2006; Qiu et al., 2007; Wen et al., 2010; Wu and Zhou, 1991; Xia and Luo, 2007; Yu et al., 2010). A crucial aspect of any remediation targets is the after-use of the land. Land for residential or agricultural use would require more stringent limits than amenity land, for example.

The new soil EQSs have added some further details. For example, they add one other organic contaminant – benzo-a-pyrene, and GB15618-2018 gives a newly added soil screening value and soil control value at the pH level of 5.5. Under GB36600-2018, different land uses are considered when developing soil EQSs. These two standards provided one method for identifying soil heavy metal contamination; If the content of pollutants exceeds the screening value, but is not higher than the background value of the soil environment, it is not included in the management of contaminated land. However, some considerations are still not resolved; for example, more contaminants still need to be considered and soil ecological protection still needs to be addressed.

7. Current situation and future priorities

China is highly reliant on its 'best quality' soils for food security and agricultural production. It has been estimated that 20% of China total arable land is contaminated (Lin, 2004; Zhao et al., 2007). This may be different from the situation in most developed countries, where a higher proportion of agricultural land is not contaminated. For example, a higher (~93%) proportion of European agricultural land is considered safe for food production (Tóth et al., 2016). The reality is that China will need to produce food for human consumption on soils which are already deemed 'contaminated'. Scientifically based risk assessments are necessary to inform practical decisions about the most practical land use options. For example, important research is currently being conducted in China and elsewhere to understand where and how 'contaminated' land can be used to support food production. This requires knowledge of soil chemistry and soil-crop plant transfers of contaminants (Tangahu et al., 2011; Xu et al., 2005).

If China makes these changes/meets these priorities, it can be leading the world in approaches to contaminated land management. It is in this context that China is committed to conducting the most detailed and comprehensive soil survey to date. MEP-PRC carried out a nation survey covered 6,300,000 km^2 soil area from April 2005 to December 2013, and several geochemistry surveys by MLR-PRC have been completed from 1999 to 2014, which covered 68% of total arable land (MEP-PRC, 2016). There are several areas where revisions are being considered, to bring China to a leading position internationally. It is hoped that the new regulatory approaches can be further developed to:

1. Increase the range of analytes for which standards are set. Most focus so far has been on inorganics, but there is a wide array of organic contaminants for which standards can be set.
2. Critically important to the science-based risk assessment of contaminants in soils is the incorporation of speciation and bioavailability into the measurement and evaluation criteria. On initial screening, soils and sites may be deemed 'contaminated', but after a second tier analysis they may be shown to be suitable for crop production and use. Selection of appropriate and validated measurement and evaluation tools is a priority. If this is done in a scientifically transparent and defensible way, China will have a robust and internationally leading system for soil management in place.
3. Derivation of standards has focused on human receptor endpoints. However, consideration of soil biology/ecological endpoints will be necessary to protect ecosystem health.
4. National and regional/local scenarios of land use type/usage. This addresses

residential/urban re-use of industrial land, as well as varying agricultural scenarios, such as different agricultural systems and cropping regimes.

Acknowledgements

We are grateful to Dr Lisa Norton and Dr Aidan Keith in Centre for Ecology & Hydrology-Lancaster for their contribution to the knowledge of the Countryside Survey.

Summary

In this Chapter, the situation and priority of soil contamination in China were reviewed by comparing soil background surveys and soil environmental standards between China and the UK. Soil survey and monitoring, and soil environmental standards based on the derivation of risk-based assessment method need to be as the priority of soil contamination management and policy development in China. For a long time, China has used the Environmental Quality Standard for Soils (GB15618-1995) published in 1995. In 2018, new EQS (soil screening values) for agricultural and construction lands were released. In order to further assess whether the new soil screening values are suitable for China and what need to be improved in China's soil environmental standards, the comparisons of soil screening values between China and UK were explored in the next Chapter.

II

A comprehensive comparison and analysis of soil screening values derived and used in China and the UK

Yiming Sun (1,2), Jicai Wang (1), Guanlin Guo (1)*, Hong Li (2,3), Kevin Jones (2)

1 State Key Laboratory of Environmental Criteria and Risk Assessment, Chinese Research Academy for Environmental Sciences, Beijing 100012.P.R.China

2 Lancaster Environment Centre (LEC), Lancaster University, Lancaster, LA1 4YQ, UK

3 Centre for Ecology and Hydrology, Maclean Building, Wallingford OX10 8BB, UK

#Yiming Sun and Jicai Wang are co-first authors.

Abstract

China and the UK use different risk-based approaches to derive soil screening or guideline values (SSVs; SGVs) for contaminants. Here we compare the approaches and the derived values for 6 illustrative contaminants. China's SSVs are derived using an approach developed in the US as follows: for carcinogens, acceptable level of risk (ACR) is set at 10^{-6} and the SSVs calculated as 10^{-6} divided by the soil exposure and toxicity data; for non-carcinogens, the hazard quotient is 1 and the SSV is calculated as 1 divided by the soil exposure and toxicity data. The UK's SGVs are calculated by the CLEA model, for which the Average Daily Exposure (ADE) from soil sources by a specific exposure route equals the health criteria values (HCVs) for that route, whether for carcinogens or a non-carcinogens. The UK's CLEA model is also used here to derive SSVs with Chinese input parameters. China's SSVs, the UK's SGVs and values for Chinese conditions derived using the UK approach were as follows (mg/kg): As, <1, 35, 20; Cd, 20, 18, 11; Cr (VI), <1, 14, 29; benzene, 1, 1, 2; toluene, 1200, 3005, 3800; ethyl-benzene, 7, 930, 1200. By comparing the differences in toxicity assessment and risk characterization, exposure assessment and parameter types in the methodologies to obtain SSVs in China and the UK, and by combining the CLEA model with Chinese parameterisation, these comparisons highlight that the difference in toxicity assessment and risk characterization methods of carcinogens results in the biggest difference in SSVs between the 2 countries. However, for non-carcinogenic substances, the difference of SSVs calculation method and SSVs is small. The difference in SSVs for carcinogenic substances is also related to the route of exposure. For volatile organic compounds, the presence of indoor respiratory exposure pathways greatly reduces the differences caused by toxicity assessment and risk characterization methods. For non-volatile substances such as heavy metals, the effects of toxicity assessment and risk characterization methods are significant. The SSV of As obtained by the CLEA model with Chinese parameters is closer to the background value of soil in China. In the management of non-volatile contaminated sites such as heavy metals in China, the CLEA model can be used for risk assessment and calculation of site specific SSVs. In the future, China can use the UK method to strengthen its toxicity assessment and risk characterization methods for carcinogenic substances, to reduce the uncertainty in the risk assessment of contaminated sites and improve the scientific management of contaminated sites.

Keywords: soil pollution, soil screening values, soil guideline values, China, UK

Capsule: Improvements are suggested to derive China’s national soil screening values.

1. Introduction

Soil pollution has become a widespread and serious problem in many regions of the world (Cachada et al. 2018; Chen et al. 2014; Barsova et al. 2019; Ramón and Lull 2019; Kumar et al. 2018). In the past thirty years, environmental risk assessment has been widely adopted in many countries to manage soil pollution in contaminated land, and some countries (e.g. United States, United Kingdom, Netherlands, Canada and Australia) have developed risk based approaches to derive contaminant-specific values, to help with management of contaminant scenarios. These are designed to protect human health and manage soil pollution in accordance with national regulations. Soil screening values (SSVs) are derived by risk based approaches, and provide an important support tool for contaminated land management (United States Environmental Protection Agency 1996; Swartjes et al. 2012; Environmental Agency 2009a; CCME 2006; National Environmental Protection Council 2011). SSVs are used to categorise the risk of soil contamination. For a specific land use, if the concentration of a given contaminant is less than the SSV, it is defined as being of no risk to human health. If it exceeds the SSV, this may trigger further surveys, risk assessments, potential changes of land use or remediation measures, depending on the national processes (United States Environmental Protection Agency 1996; CCME 2006; CCME 2006; National Environmental Protection Council 2011; Ministry of Environmental Protection of the People's Republic of China 2018a). However, SSVs in different countries are different in terms of definition, numerical values and inference methods. For example, Zhou et al (2016) found standard values for arsenic varied by country, land use and definition. Generally, standard values of industrial land are higher than those of commercial, residential and agricultural land. Standard values of some countries and regions place more emphasis on soil properties, soil types and extractants, not land types. Carlon et al. (2007) conducted a comprehensive analysis of SSVs in different European countries and found it was subject to geographical, biological, socio-cultural, regulatory, political and scientific factors. The value of SSVs in different countries in Europe are different in value and usage, and the influence of different factors is different. Many factors combine to result in differences, namely: i. the approaches used to derive the SSVs (e.g. hazard identification, toxicity assessment, exposure assessment and risk characterization); ii. the descriptors/parameters selected (e.g. the population/soil/site characteristics and building structure; the environmental conditions and parameters values); iii. The proposed land use or level of 'acceptable risk' (Claudio et al. 2007; Song 2011; Wang and Lin 2016; Xu et al. 2013). Due to these reasons, SSVs derived and used in different countries can be different, resulting in different management options being selected for the same soil concentration in different places. Thus, using scientific derivation methods, matching the parameter values of regional characteristics, and calculating the soil screening value to meet the risk level of policy requirements is the basis for scientific management of contaminated soils in a country and region, which is necessary to derive methods, parameter values, etc. In this paper, a comparative analysis of SSVs is carried out, to provide scientific reference for method selection and parameter determination. Our focus is China, as explained below.

The UK's approach is one of the most established. Over the last 20 years, the Environment Agency (EA) has systematically released a series of regulations, standards and science reports to introduce how to deal with soil contamination in the UK (Environmental Agency 2009a; Environmental Agency 2009b; Environmental Agency 2009c; Environmental Agency 2009d).

Soil Guideline Values (SGVs) have been derived and widely applied to the investigation and management of contaminated land (Environmental Agency 2009a). SGVs are defined as a starting point for evaluating long-term and *on-site* exposure risks to human health from chemicals in soil, below which the long-term human health risks are tolerable or minimal, above which further investigation should be undertaken. It uses the CLEA (Contaminated Land Exposure Assessment) model to derive SGVs.

In recent years, the Chinese central government and local government has started to pay attention to soil pollution by taking a series of actions (Hou et al. 2017; Li et al. 2017). For example, in 2016, the 10-Chapter Soil Pollution Action Plan was issued. It's purpose is to manage, control and prevent soil pollution and improve soil quality in China (People's Daily, 2016). An early priority is to conduct relevant surveys of soil pollution, to define baselines of soil environmental quality (State Council 2016). In August 2018, the Chinese government released national standards for contaminants in agricultural soils and contaminated land, Soil Screening Values (SSVs) (Ministry of Environmental Protection of the People's Republic of China 2018a; Ministry of Environmental Protection of the People's Republic of China 2018b). China's SSVs are also derived using a risk-based approach. Figure 1 shows the procedures used to derive SSVs in China and the UK.

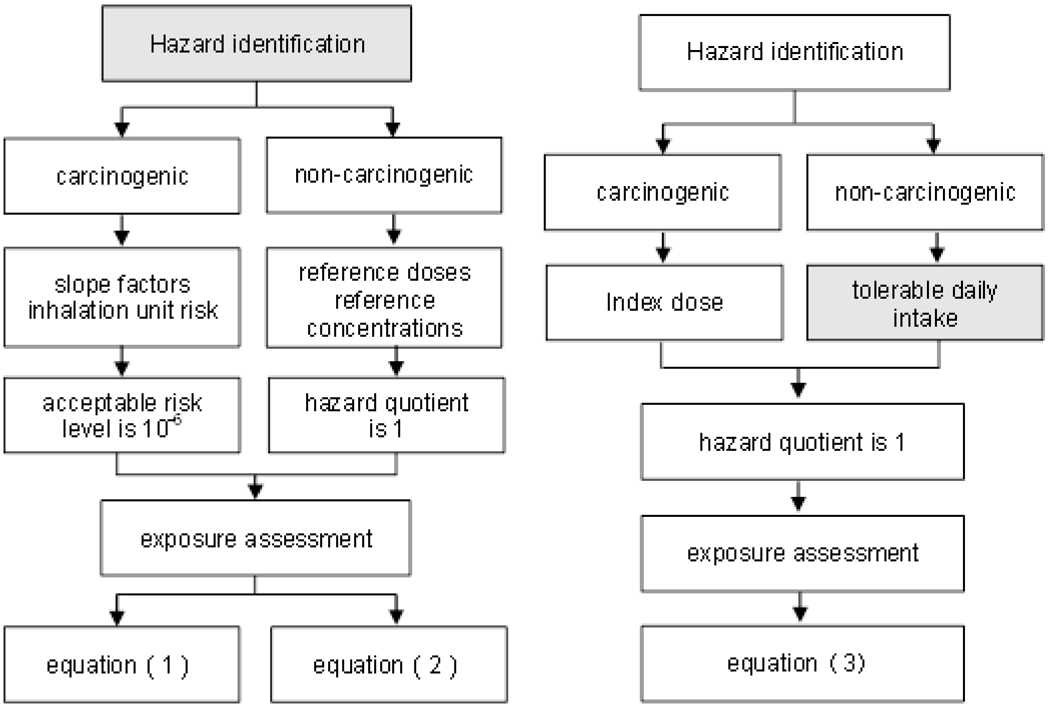


Figure 1. The derivation procedure for SSVs in China (Left) and the UK (Right).

The purpose and objectives of the study were therefore to:

(1) compare the derivation method of SSVs in China and the UK in terms of toxicity assessment, risk characterization and exposure assessment;

(2) identify the key differences in SSVs between China and UK and their main factors;

(3) provide some suggestions for the improvement of China's national and local SSVs standard setting.

To achieve these goals, six chemicals were selected as examples, 3 inorganic and 3 organic, for which SSVs have been published. These are As, Cd, hexavalent Cr, benzene, toluene and ethyl-benzene.

2. The approaches to derive SSVs in China and the UK

The basic principle in each case is to derive a soil concentration which gives an acceptable level of risk (ACR), using knowledge of the contaminant behavior in soils, an assessment of exposure and toxicological information.

2.1 The Chinese approach

China's approach is based on that used in the USA and developed by the US Environmental Protection Agency (US EPA). Carcinogenic and non-carcinogenic substances are treated differently in the SSV calculation process. For carcinogenic substances without an effect threshold, it's ACR is set at 10^{-6}, the SSVs is calculated as follows (Ministry of Environmental Protection of the People's Republic of China 2014):

For carcinogens:

$$RCVS = \frac{ACR}{R_{oral} \times SF_o + R_{dermal} \times SF_d + R_{inhalation} \times SF_i} \quad \text{Equation (1)}$$

For a threshold non-carcinogenic substance, its ACR is named as acceptable hazard quotient (AHQ) and set at 1, the SSVs is calculated as follows:

For non-carcinogens:

$$HCVS = \frac{AHQ \times SAF}{\frac{R_{oral}}{RfD_{oral}} + \frac{R_{dermal}}{RfD_{dermal}} + \frac{R_{inhalation}}{RfD_{inhalatio}}} \quad \text{Equation (2)}$$

Where: *RCVS* is the SSVs for carcinogens, mg/kg;

HCVS is the SSVs for non-carcinogens, mg/kg;

ACR is the acceptable risk level, 10^{-6};

AHQ is the hazard quotient, 1;

R_i is route of exposure (oral, inhalation and dermal), mg/kg-d;

SF_i is the slope factor (oral, inhalation and dermal), 1/(mg/kg-d);

RfD_i is the reference dose (oral, inhalation and dermal), mg/kg-d;

SAF is soil allocation factor, dimensionless.

The AHQ for a threshold non-carcinogenic substance is 1, and the SSV (Equation 2) is calculated as 1 divided by the soil exposure and toxicity data (reference dose, etc.). The process can be done with Microsoft Excel or other calculation programs. Three exposure routes are considered: oral intake, dermal contact and inhalation intake. Residential and commercial land uses are considered separately.

2.2 The UK approach

The UK's SGVs are based on the four steps of hazard identification, toxicity assessment, exposure assessment and risk characterization of contaminated soil, using the CLEA model - a defined framework and methodology (Environmental Agency 2009d). The basic principle used to establish SGVs is to set the soil concentration, of which the Average Daily Exposure (ADE) from soil sources by a particular exposure route equals the health criteria values (HCVs) for that route. The CLEA software estimates the ADE to soil contaminants by adults and children living or working on contaminated land over long periods of time, and compares this estimate to HCVs (Equation (3)) (Environmental Agency 2009b; Environmental Agency 2009d). Again exposure via ingestion through the mouth (oral), absorption through the skin (dermal), and inhalation through the mouth and nose are considered. SGVs are derived for three generic land use scenarios: residential, allotment (gardening to grow food) and commercial.

$$\frac{CR_{oral}}{HCV_{oral}} + \frac{CR_{dermal}}{HCV_{dermal}} + \frac{CR_{inhalation}}{HCV_{inhalatio}} = 1$$ Equation（3）

Where: C is the representative concentration of the chemical in soil, mg/kg;

Rx is the ratio of ADE from the soil source over the soil concentration for exposure route x (oral, inhalation and dermal), mg/kg·d over mg/kg;

HCVx is health criteria value for exposure route x (oral, inhalation and dermal) mg/kg·d.

2.3 Highlighted differences between the Chinese and UK approaches

2.3.1 Toxicity assessment and risk characterization

China's chemical substances hazard identification and toxicity assessment use the US EPA's Integrated Risk Information System (IRIS). According to the characteristics of the dose-response relationship, carcinogens are considered with a non-threshold limit, while non-carcinogens have a threshold. For carcinogens, the oral slope factor (SF) gives a plausible upper-bound estimate of the probability of a response per unit intake of a chemical over a lifetime, and the inhalation unit risk (IUR) is defined as the upper-bound excess lifetime cancer risk estimated to result from continuous inhalation exposure to a chemical at a concentration of 1 $\mu g/m^3$ in air (https://www.epa.gov/risk/regional-screening-levels-rsls-users-guide). The SFs and IURs in the IRIS database are used for toxicity assessment. Non-carcinogens use the reference dose (RfD) as an estimate of a daily oral exposure or the reference concentrations (RfC) to estimate inhalation exposure (https://www.epa.gov/risk/regional-screening-levels-rsls-users-guide). The UK's EA Human Health Toxicological Assessment of Contaminants in Soil Science Report also classifies the toxic effects of substances into threshold-toxic and non-threshold toxicities based on dose-response characterisation, but the names, values and derived method of HCVs is different from IRIS. For non-carcinogens, threshold-based substances use tolerable daily intakes (TDIs) as HCVs for human health toxicity assessment, and non-threshold substances with an Index Dose (ID) as HCVs (Environmental Agency 2009b). The TDI is defined as an estimate of the amount of a contaminant that can be ingested daily over a lifetime without appreciable health risk. ID expressed as a daily dose which is likely to be associated with a negligible risk of carcinogenic effect over a specified duration of exposure.

According to the IRIS, for carcinogens, the Benchmark Dose (BMD) or No Observed Adverse Effect Level (NOAEL) or Lowest Observed Adverse Effect Level (LOAEL) were used in the dose response curve to derive the SF or IUR for humans through a linear mathematical model with an acceptable cancer risk level of 10^{-6}; for non-carcinogens the BMD, NOAEL, LOAEL or categorical regression were used with uncertainty factors (UF) to derive the RfD or RfC to reflect the daily intake limitation. In the UK both the TDI and the ID are derived as the same method as RfD in the US using the BMD, NOAEL or LOAEL with UF. So the main difference of toxicity assessment between China and UK is the hazard characterisation of carcinogens. China adopted the quantitative dose-response modelling from US, while the UK use the non-quantitative extrapolation.

Due to the differences in toxicity assessment methods, the risk characterization approach and the structure of SSVs calculation equation is inevitably different between two countries. In China, for carcinogens, the soil screening value is calculated by dividing the ACR 10^{-6} by the product of soil exposure and carcinogenic slope factor (Equation (1)); for non-carcinogens the soil screening value is calculated by dividing the product of the reference dose and the AHQ 1 by the soil exposure (Equation 2). The UK CLEA model calculates the SGVs, whether carcinogenic or non-carcinogenic, by dividing the product of the HCV by 1 by the soil exposure (Equation 3). For carcinogens, there is no clearly acceptable risk level of cancer (although Theoretically, the health risk corresponding to the dose should be 10^{-5}) (Environmental Agency

2009b), and the ID is significantly different from the carcinogenic slope factor derived from the linear mathematical model. It is more similar to the reference dose of Chinese non-carcinogens in application and value. If equation 3 is rewritten, its form is exactly the same as equation 2. This shows that the difference in the toxicity assessment and risk characterization of carcinogens is the fundamental reason for the different formulae for soil screening values of carcinogens in the two countries.

2.3.2 Exposure assessment

At present, countries including the UK, the US, the Netherlands, and China have established exposure assessment models for the risk assessment of soil pollution (CCME 2011; Environmental Agency 2009c; United States Environmental Protection Agency 1992; Brand et al. 2007). Exposure assessment is an important aspect of human health risk assessment for soil contaminants and a model basis for soil screening value calculations to assess the amount of soil pollutants that may be exposed to human. The exposure pathways considered by both Chinese and English in soil exposure assessment include ingestion, dermal contact and inhalation, and the exposures of each pathways are calculated using the corresponding models. The differences in exposure assessment are mainly reflected in three aspects: a) the sensitive receptor will vary with the type of land use; b) the source of soil contaminants will vary depending on the type of land use; c) the exposure assessment model equation is different for the same exposure pathway. For example, the sensitive receptors of residential land use in China are children (0-6 years old) and adults (female), depending on the carcinogenic and non-carcinogenic effects of the agent, while the UK CLEA model only considers female children

(1-6 age class) for both carcinogenic and non-carcinogenic effects. For soil contaminant sources, the Chinese model only considers direct soil contaminant exposure, and there is no source of self-produced crops. The CLEA model can consider not only direct soil contaminant exposure, but also indirect exposure such as ingestion of self-produced crops. The soil ingestion rate, the soil particulate emission model and the vapor intrusion model for exposure assessment are the largest differences between the two countries in the exposure models. In this study, in order to limit the comparison in a unified framework, the scenarios of residential land use, without the self-produced crops ingestion exposure pathway, is selected - mainly comparing the models and model parameters of each exposure pathway in China and the UK.

2.3.2.1 Exposure assessment models

In the selected residential land use, the exposure pathways in China and the UK CLEA model are soil ingestion, dermal contact, inhalation of dust and volatile organic compounds vapors indoors or outdoors. The exposure assessment equation consists of the daily soil intake rate (R_x) of different exposure routes, the exposure duration (ED), exposure frequency (EF), body weight (BW) and average time (AT), where the exposure duration (ED) and the average time (AT) vary with toxic effects and sensitive receptors. The exposure frequency (EF) and the daily soil intake rate (R_x) are related to the exposure route and different in the equations, parameters and values between the two countries (see Table 1).

It can be seen from Table 1 that in addition to considering the oral soil ingestion rate of adults, the oral soil ingestion rate of children in China is 100 mg more than that of children in the UK. The dermal contact daily soil intake rate in the Chinese and UK calculation equations are completely consistent with each other. The inhalation route can be divided into inhalation of soil dust and inhalation of VOC vapors in the two countries. For the inhalation of soil dust, the PM_{10} is used in the Chinese equation to calculate the daily soil intake rate of soil particles in the indoor and outdoor ambient air multiplied by the ratio of particles from the soil in the indoor and outdoor air, p_i and p_o, respectively. The exposure frequency (EF) of children and adults are divided into the indoor exposure frequency (EFI_{inh}) and the outdoor exposure frequency (EFO_{inh}). The UK CLEA model adopts the particulate emission factor (PEF) to calculate the rate of daily soil intake exposed to ambient air in children. The PEF is calculated by the air diffusion factor Q/C_{wind} and the inverse of soil PM_{10} emission flux $J_{w,}$ where Q/C_{wind} can take the monitoring data of different cities, J_w is calculated with wind speed values (Environmental Agency 2009b; Environmental Agency 2009d). For inhalation of vapors, China assumes the source of vapors from surface soils, subsurface soils and indoor air. The CLEA model assumes vapors from surface soils and indoor air. The rate of daily vapor intake exposed to ambient air from the source of surface soils is characterized by the volatilization factor (VF_{sur}), and since the VF_{sur} equations adopted in two countries were recommended by the American Society for Testing and Materials (ASTM) or the US EPA, the formula is very similar. The difference in detail is that the air diffusion factor is calculated by DF_{oa} in China and by Q/C_{wind} in the CLEA model. For calculation of the vapor intrusion concentration in indoor air from the contaminated subsurface soil located below the bottom of the building floor or foundation, the equations used in China and in the CLEA model are completely different. This is because China adopts the VF_{ind} equation of the ASTM to calculate the vapor intrusion concentration for indoor air, but the CLEA model uses the soil vapor concentration to calculate the indoor air concentration by multiplying the attenuation factor α; the pressure difference between the soil air and indoor air is 0 Pa in China, and in the CLEA model is 3.1 Pa.

Table 1. Comparison of exposure model and parameters in China and UK.

Item	China	UK
Sensitive receptor	Carcinogen: children (0-6 years old) and adult (Female)	Children（Female, 1-6 age class）
	Non-carcinogen: children (0-6 years old)	
Assessment model	Carcinogen: $ER_x = \frac{R_{x-c} \times ED_c \times EF_{x-c}}{BW_c \times AT} + \frac{R_{x-a} \times ED_a \times EF_{x-a}}{BW_a \times AT}$	$ER_x = \frac{R_{x-c} \times ED_c \times EF_{x-c}}{BW_c \times AT}$
	Non-carcinogen: $ER_x = \frac{R_{x-c} \times ED_c \times EF_{x-c}}{BW_c \times AT}$	
	Where：ER_x is the daily human exposure for a exposure pathway x， mg/kg-d; R_x is the daily soil intake rate for exposure route x for children (c) or adults (a),mg/d; ED is exposure duration for children (c) or adults (a), a; EF_x is exposure frequency for children (c) or adults (a), which can be subdivided into indoor (EFI) and outdoor (EFO) exposure frequencies, d/a; BW is body weight to children (c) or adults (a), kg; AT is average time, d.	
Exposure duration (ED)	Carcinogen: children: 6 years; adult: 24 years	Children: 6 years
	Non-carcinogen: children: 6 years	
Exposure frequency (EF)	Soil ingestion and dermal contact: children: 350d/y, adult: 250d/y	Soil ingestion and dermal contact: Age class 1: 180 d/y; Age class 2-6: 365 d/y
	Indoor inhalation: children :262.5d/y, adult: 187.5d/y	Indoor and outdoor inhalation: Age class 1-6: 365 d/y
	Outdoor inhalation: children: 87.5d/y, adult: 62.5d/y	
Average time (AT)	Carcinogen: 27740d	2190d
	Non-carcinogen: 2190d	
Oral	Children soil ingestion rate: R_{oral-c}= 200 mg/d	Children soil ingestion rate: R_{oral-c}=100 mg/d
	Adult soil ingestion rate:	

	$R_{oral-a} = 100$ mg/d	
Dermal	Children: $R_{der-c} = A_{skin-c} \times AF_c \times n \times ABS_d$ Adult: $R_{der-a} = A_{skin-a} \times AF_a \times n \times ABS_d$	$R_{der-c} = A_{skin-c} \times AF_c \times n \times ABS_d$
	Where: A_{skin} is the exposure skin area for children (c) or adult (a), cm^2; AF is soil-to-skin adherence factor for children (c) or adult (a), mg/cm^2; n is the number of daily soil contact events, d^{-1}; ABS_d is the dermal absorption fraction, unitless.	
Inhalation of dust	Children: $R_{inh-c} = PM_{10} \times RF \times V_{inh-c} \times (po \times EFO_c + pi \times EFI_c)$ Adult: $R_{inh-a} = PM_{10} \times RF \times V_{inh-a} \times (po \times EFO_a + pi \times EFI_a)$	Outdoor: $R_{inh-c} = \frac{1}{PEF} \times V_{inh-c} \times \frac{T_{site-o}}{24}$ Indoor: $R_{inh-c} = \left(\frac{1}{PEF} + TF \times DL\right) \times V_{inh-c} \times \frac{T_{site-i}}{24}$
	Where: RF is Retention fraction of inhaled particulates in body; V_{inh} is daily inhalation rate for children (c) or adults (a),m^3/d; p_o is the fraction of soil-borne particulates in outdoor air, unitless; p_i is the fraction of soil-borne particulates in indoor air, unitless; PEF is particulate emission factor, m^3/kg; T_{site} is exposure time of indoor (i) and outdoor (o), h/day; TF is the soil-to-dust transport factor according to soil type, g/g dw; DL is the indoor dust loading factor, g/m^3.	
Inhalation of vapors from outdoor surface soil	$R_{inh} = C_s \times VF_{sur} \times (V_{inh-c} + V_{inh-a})$ $VF = \frac{\rho_b}{DF_{oa}} \times \sqrt{\frac{4 \times D_{eff} \times H'}{\pi \times \tau \times 31536000 \times K_{sw} \times \rho_b}}$ $DF_{oa} = \frac{U_{air} \times W \times \delta_{air}}{A}$	$R_{inh} = C_S \times VF_{sur} \times V_{inh-c} \times \left(\frac{T_{site}}{24}\right)$ $VF = \frac{\rho_s}{\frac{1}{10} \times Q/C_{wind}} \times \sqrt{\frac{4 \times D_{eff} \times H'}{\pi \times \tau \times 31536000 \times K_{sw} \times \rho_s}}$
	Where: VF_{sur} is the volatilisation factor from soil to outdoor ambient air, kg/m^3; C_s is the total contaminants concentration in soil, mg/kg; D_{eff} is the effective chemical diffusion coefficient in soil, cm^2/s; H' is the air-water partition coefficient at ambient temperature, cm^3/cm^3; ρ_b is the bulk soil density, g/cm^3; ρ_s is the dry bulk soil density, g/cm^3; τ is the averaging time for surface emission vapor flux, y; K_{sw} is the total soil-water partition coefficient, cm^3/g; Q/C_{wind} is the air dispersion factor, g/cm^2 per kg/m^3; U_{air} is the ambient air velocity in mixing zone, cm/s; W is the width of source-zone area, cm; A is the source-zone area, cm^2;	

	δ_{air} is the mixing zone height, cm; T_{site} is the outdoor site occupancy period, h/d.	
Inhalation of vapors indoor air	$R_{inh} = C_s \times VF_{ind} \times \left(V_{inh-c} + V_{inh-a}\right)$	$R_{inh} = C_{vap} \times \alpha \times V_{inh-c}$
	Where: VF_{ind} is the volatilization factor from subsurface soil to indoor, kg/m^3; C_{vap} is the soil vapor concentration, mg/m^3; α is the attenuation factor, unitless.	
Inhalation of vapors from outdoor subsurface soil	$R_{inh} = C_s \times VF_{sub} \times \left(V_{inh-c} + V_{inh-a}\right)$ $VF_{suboa1} = \dfrac{1}{\left(1 + \dfrac{DF_{oa} \times L_s}{D_s^{eff}}\right) \times \dfrac{K_{sw}}{H'}}$	-

3. Results

3.1 Effect of methods on SSVs

In order to confirm the influences of derivation methods on the calculation result of SSVs between the two countries, As, Cd, Cr (VI), benzene, toluene and ethylbenzene were selected as representative heavy metals and volatile organic compounds (VOCs) under the exposure scenario of residential land use (without self-produced crops ingestion). The SSVs of these 6 substances in China and the UK were calculated by the methods mentioned above and the results are shown in Table 2. It can be seen that for the non-carcinogenic substances (Cd, toluene), the difference of SSVs in China and the UK is relatively small: Cd (China: 20 mg/kg, UK: 18 mg/kg; toluene (China: 1200 mg/kg, UK: 3000 mg/kg). For carcinogens (As, Cr (VI), benzene, ethylbenzene)), the differences are large: As (China: <1 mg/kg, UK: 35 mg/kg), Cr (VI) (China: <1 mg/kg, UK: 14 mg/kg), ethylbenzene (China: 7 mg/kg, UK: 930 mg/kg). Due to the difference of hazard identification, the SSVs derived for ethylbenzene highlight major differences. This indicates that volatiles the contribution of inhalation via vapor pathway significantly reduces the differences caused by other elements, especially the inhalation of vapors indoor air, which can be inferred from the exposure pathway contribution showed in Figure 2. The different calculation procedures, especially for carcinogens, make the SSVs under the current conditions of China and UK differ substantially.

Table 2. Soil screening values of selected chemicals derived from different models (mg/kg)

Chemical	Toxicity	China	UK	China in CLEA Model
Arsenic	Carcinogenic	<1*	35	20
Cadmium	Non-Carcinogenic	20	18	11
Chromium(VI)	Carcinogenic	<1*	14(C4SL child)	29(C4SL child)
Benzene	Carcinogenic	1	1.1	1.6
Toluene	Non-Carcinogenic	1200	3000	3800
Ethylbenzene	Carcinogenic(China) Non-Carcinogenic(UK)	7.2	930	1200

Note: * Since the soil screening value of As calculated by the Chinese model is < 1, the soil screening value of 20 mg/kg of As released in China is the result of replacing the soil environmental background value, which is not the calculated value of the model. In order to make this study comparable, the screening value of As in China is still calculated by the model.

3.2 Effect of parameters on SSVs

To further understand the impact of parameter values on the SSVs between the two countries, the CLEA model was run with Chinese input parameters to calculate screening values of these six chemicals. These values can be compared with the SSVs calculated in section 3.1, to analyse the differences caused by parameters in China and UK. Details of the Chinese parameter inputs to replace default values are given in the Supplementary Information (SI) and summarized in Tables 3 and 4. The calculated results are shown in Table 2 (as ‘China in the CLEA model’). It can be seen that SSVs calculated in this way are very close to the UK SSVs. The influence of the parameters of SSVs in UK and China is far less than the impact of the formulation method. However, it is meaningful to find the parameters that cause the difference between the two countries to further reduce the difference of SSVs in China and UK.

Table 3. Land use and receptor parameters of China input into the CLEA model

Land use	Unit	Age class					
		1	2	3	4	5	6
EF (soil and dust ingestion) [a]	day yr^{-1}	350	350	350	350	350	350
EF (consumption of homegrown produce) [a]	day yr^{-1}	0	0	0	0	0	0
EF (dermal contact, indoor) [a]	day yr^{-1}	350	350	350	350	350	350
EF (dermal contact, outdoor) [a]	day yr^{-1}	350	350	350	350	350	350
EF (inhalation of dust and vapor, indoor) [a]	day yr^{-1}	262.5	262.5	262.5	262.5	262.5	262.5
EF (inhalation of dust and vapor, outdoor) [a]	day yr^{-1}	87.5	87.5	87.5	87.5	87.5	87.5
Occupancy Period (indoor) [b]	hr day^{-1}	22.3	21.5	21.3	21.3	21.5	21.5
Occupancy Period (outdoor) [b]	hr day^{-1}	1.7	2.5	2.6	2.5	2.2	2.2
Soil to dermal adherence factor (indoor)*	mg cm^{-2} day^{-1}	2.0E-01	2.0E-01	2.0E-01	2.0E-01	2.0E-01	2.0E-01
Soil to dermal adherence factor (outdoor)*	mg cm^{-2} day^{-1}	2.0E-01	2.0E-01	2.0E-01	2.0E-01	2.0E-01	2.0E-01
Soil and dust ingestion rate [a]	g day^{-1}	2.0E-01	2.0E-01	2.0E-01	2.0E-01	2.0E-01	2.0E-01
Receptor		**Age class**					
		1	**2**	**3**	**4**	**5**	**6**
Body weight [b]	kg	7.9	10.8	13.2	15.3	17.4	19.2
Body height [c]	m			1	1.06	1.12	1.18
inhalation rate	m^3 day^{-1}	4.6	5.4	6	7.6	8.1	8.4
Max exposed dermal fraction (indoor)*	m^2 m^{-2}	0.14	0.18	0.22	0.24	0.27	0.28
Max exposed dermal fraction (outdoor)*	m^2 m^{-2}	0.14	0.18	0.22	0.24	0.27	0.28

Note: a: refer to the values in the "Technical Guidelines for Risk Assessment of Contaminated Sites (HJ25.3-2014) (Ministry of Environmental Protection of the People's Republic of China

2004)". b: "Exposure factors handbook of Chinese Population (0-5 years old), (6-17 years old) (Ministry of Environmental Protection of the People's Republic of China 2013)". c: "National Physical Fitness Monitoring 2014" (http://www.sport.gov.cn/n16/n1077/n1422/7331093.html). *: refer to the default values in the CLEA model.

Table 4. Soil and building properties of China input into the CLEA model

Soil properties for	value	Unit
Porosity, total *	0.43	$cm^3\ cm^{-3}$
Porosity, air-filled [d]	0.30	$cm^3\ cm^{-3}$
Porosity, water-filled [d]	0.13	$cm^3\ cm^{-3}$
Residual soil water Content [d]	0.20	$cm^3\ cm^{-3}$
Saturated hydraulic conductivity [d]	7.90E-05	$cm\ s^{-1}$
van Genuchten shape parameter (*m*) *	3.20E-01	dimensionless
Bulk density [d]	1.50	$g\ cm^{-3}$
Threshold value of wind speed at 10m [d]	2.00	$m\ s^{-1}$
Empirical function (Fx) for dust model *	1.22	dimensionless
Ambient soil temperature [d]	298	K
Building properties for		
Building footprint [d]	9.00E+00	m^2
Living space air exchange rate [d]	0.5	hr^{-1}
Living space height (above ground) [d]	2.2	m
Living space height (below ground) [d]	0.0	m
Pressure difference (soil to enclosed space) *	3.1	Pa
Foundation thickness [d]	3.50E-01	m
Floor crack area [d]	2.40E+01	cm^2
Dust loading factor *	5.00E+01	$\mu g\ m^{-3}$
Air dispersion model		
Mean annual windspeed (10m) [d]	2.00	$m\ s^{-1}$
Air dispersion factor at height of 0.8m *	2400	$g\ m^{-2}$ per $kg\ m^{-3}$
Air dispersion factor at height of 1.6m *	0.00	$g\ m^{-2}$ per $kg\ m^{-3}$
Fraction of site with hard or vegetative cover	0.00	$m^2\ m^{-2}$
Vapor model		
Default soil gas ingress rate	-	$cm^3\ s^{-1}$
Depth to top of source (beneath building) [d]	100	cm
Depth to top of source (no building)	0	cm
Time average period for surface emissions [d]	6	years

User defined effective air permeability [d]	1.00E-08	cm^2

Note: d: refer to the values in the "Preparation instructions for "soil environment quality risk control standard for soil contamination of development land (Trial)" (Environmental Agency 2009c). * refer to the default values in the CLEA model.

4. Discussion

4.1 The reasons for differences of SSVs in China and UK

The SSVs calculation for China referred to the toxicity assessment of the US EPA's Integrated Risk Information System (IRIS), with SSVs calculated for carcinogens according to equation (1) in 2.1 and an acceptable cancer risk level of 10^{-6}, while non-carcinogens are calculated according to equation (2) in 2.1 with a hazard quotient of 1. However, in the UK CLEA model, SSVs are calculated for carcinogens and non-carcinogens using equation (3) in 2.1. The effect of these differences on SSV toxicity assessment and risk characterization can be seen from the calculation results of the selected chemicals in Section 3.1. Although both China and UK identified As as a carcinogen (in the UK, the oral and inhalation ID of As associated with a minimum excess risk of cancer were determined, in which the oral intake ID is based on British drinking water standards. The risk is equivalent to the cancer risk of around 40 to 400 in 100,000[8]. If the equations, toxicity values and other parameters (such as daily soil intake rate, exposure frequency, exposure duration, etc.) are not the same, the calculated result in China was < 1 mg/kg and in the UK was 35 mg/kg. If the CLEA model was used with Chinese input parameters, the toxicity values and equations were unified; the As SSV for China was of the same order of magnitude as the UK and very close to the Chinese As soil background value. The result was also the same for Cr (VI). However, for the non-carcinogens, (Cd and toluene), the SSVs were very similar. In contrast, the SSVs derived for ethylbenzene (which is identified as a carcinogen in China and a non-carcinogen in the UK) were very different, because of the differences in toxicity assessment and risk characterization (see Table 5).

So, in conclusion, the toxicity values and risk characterization methods were the factors most affecting the SSV differences between China and the UK, especially for carcinogens, while choices in model parameters has only a minor effect.

Table 5. Toxicity values of selected chemicals in China and UK

Chemical	China				UK CLEA Model			
	SF_o mg/kg-d	IUR $(ug/m^3)^{-1}$	RfD_o mg/kg-d	RfC_i mg/m^3	TDI_o ug/kg-d	ID_o ug/kg-d	TDI_i ug/kg-d	ID_i ug/kg-d
Arsenic	1.5E+00	4.3E-03				3.0E-01		2.0E-03
Cadmium			1.0E-03	1.0E-05	3.6E-01		1.4E-03	
Chromium(VI)	5.0E-01	8.4E-02				4.4E-01		3.4E-04
Benzene	5.5E-02	7.8E-06				2.9E-01		1.4E+00
Toluene			8.0E-02	5.0E+00	2.23E+02		1.4E+03	
Ethylbenzene	1.1E-02	2.5E-06			1.0E+02		2.2E+02	

Note: SF_o is oral slope factor; IUR is inhalation unit risk; RfD_o is oral reference dose; RfC_i is inhalation reference concentrations; TDI_o and TDI_i is tolerable daily intakes of oral (o) and inhalation (i); ID_o and ID_i is index dose of oral (o) and inhalation (i).

4.2 The identification of key parameters affecting on SSVs

From Table 2, it can be seen the SSVs calculated by substituting Chinese parameters into the CLEA model are still different from the SSVs in UK, indicating that the parameters also contribute to the differences of SSVs in China and UK. Therefore, analysis and identification of key parameters can help us to understand the differences of SSVs in China and UK. Here, we determine the important parameters of the difference of SSVs in China and UK by calculating the contribution rate of each exposure route of different substances to SSVs. Firstly, the exposure route with the highest contribution rate to SSVs is determined, and then the parameters that have a greater influence on the exposure route are analyzed to determine the important parameters affecting the difference of SSVs in China and UK. Figure 2 summarises the calculated contributions of the different exposure pathways for each of the selected chemicals, under the 3 scenarios. For the heavy metals, the contributions of the oral ingestion, dermal contact and inhalation of dust pathways is >80%, for each method of SSV derivation. However, for the VOCs, inhalation of vapors from the indoor air pathway dominated (see Figure 2). On the other hand, for SSVs of heavy metals (except Cr), the contribution of imported intake (not distinguishing from background intake) is greater than 50%, which is the most influential exposure pathway, so the effect of parameters of oral exposure pathways are important for heavy metal screening. The value for oral soil ingestion is 200 mg/kg in China, 100 mg/kg in the UK. The SSVs of heavy metal (except Cr) calculated by the CLEA model in China is reduced by half. So, for heavy metals, we consider that the key parameter affecting the screening value of China and the UK is the oral daily soil intake. However, for volatile organic compounds, it is not very difference from SSVs in UK and China calculated by using Chinese parameter into the CLEA model. China is between 1.2 and 1.4 times higher than the UK value, indicating there is less impact of the parameters on the VOC SSVs is less than for heavy metals. In addition, the contribution rate of indoor respiratory exposure to the SSVs of volatile organic compounds is greater than 50%, which is the most important route. From the comparative analysis in Section 2.3.2.1, the attenuation factor α is the main parameter of the SSVs for indoor air volatiles in the CLEA model, so the parameters involved in α also have a certain influence.

It should be stressed that these assessments are generic conditions; in real environmental circumstances, site-specific and person/individual specific differences can vary considerably and need to be captured in site- and population/community-specific scenarios (Claudio et al. 2007).

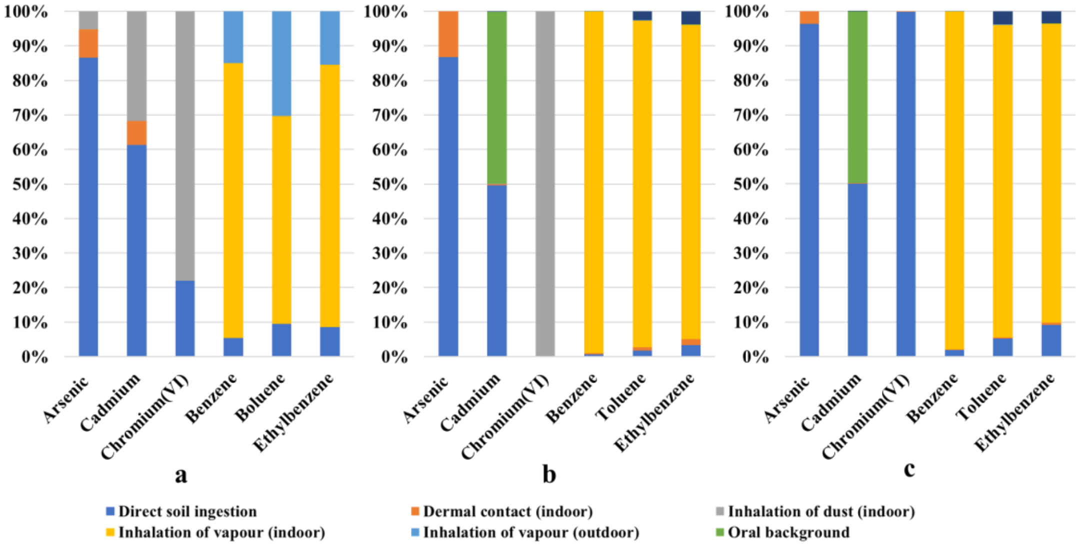


Figure 2. Exposure pathway contribution in different derivation methods of SSVs (a: China, b: UK in CLEA model, c: China in CLEA model)

C

5. Conclusion

Based on the above discussion, we can conclude that the differences in SSVs between China and the UK are mainly reflected in the aspects of toxicity assessment and risk characterization methods, exposure assessment and differences in parameters. Among them, toxicity assessment and risk characterization are important factors that cause differences in soil screening values. They not only determine the type of hazardous effect and the toxicity values, but also determine the characterization method of risks. The compared results indicate that differences in toxicity assessment and risk characterization can cause large differences in SSVs without considering the effects of other factors. Although there are some differences in exposure assessment, the model structure and parameter type used in the exposure assessment of the two countries are very similar. The biggest difference is mainly in the model of inhalation pathway. For VOCs, the contribution rate of inhalation pathways is much higher than others, and the contribution of this pathway significantly reduces the differences caused by other factors. The difference of the parameters also has some impact on the screening values, but the degree of influence is relatively weak. For example, although the SSVs of As and Cd in China and UK calculated by the CLEA model using Chinese parameters are of the same order, the SSVs in China are smaller than those for the UK. In the future, the key to reducing the difference of SSVs between China and UK is to increase consistency in toxicity assessment, risk characterization and exposure assessment models. In particular, China should implement toxicity assessment and risk characterization studies based on its own conditions due to the large variation of derived SSVs compared with soil background value, such as arsenic, and an in-depth study of the exposure assessment model for the indoor inhalation of volatiles should also be conducted. Under the current conditions, human health risk assessment has been applied to the risk assessment of pollutants in different environmental media (air, water and soil). Due to differences of environmental media, there may be some differences in behavioural patterns and exposure assessment to different environmental media. However, China and UK did not have an in-depth analysis of this issue, nor did it conduct an in-depth study of the uncertainty of the soil exposure assessment model. Therefore, the two countries should further study the human exposure behavior to soil and have certain revise in exposure assessment models, in order to reduce the uncertainty caused by the inadequate understanding of the exposure.

Acknowledgement

We are grateful to Dr Lisa Norton and Dr Aidan Keith in Centre for Ecology & Hydrology-Lancaster for their contribution to the knowledge of the soil risk assessment in UK. The authors are grateful for funding from the National Natural Science Foundation of China (Grant no. 41571311) and the National High Technology Research and Development Program of China (863 Program) (No. 2013AA06A206).

Summary

In this Chapter, soil screening values between China and UK differ in toxicity assessment, risk characterization, exposure assessment and differences in parameters. The biggest differences are toxicity assessment and risk characterization, of which, difference in toxicity assessment between China and UK are the classification of carcinogenic and non-carcinogenic equations. For the derivation of soil environmental standards, the soil environmental factors should also be considered (e.g. soil pH and soil organic matter). It is well known that soil pH and soil organic matter are important soil properties, which effect soil fertility, agricultural productivity, plant growth and human health. In order to better improve soil environmental standards and soil contamination management in China, it is necessary to analyze the change of soil pH and soil organic matter in a country with such big environmental differences.

III

TITLE: Decadal shifts in soil pH and organic matter differ between land uses in contrasting regions in China

RUNNING TITLE: pH and organic matter changes in Chinese soils

Yiming Sun (1,2), Guanlin Guo (1)*, Huading Shi (1), Mengjiao Liu (1), Aidan Keith (3), Hong Li (2,3) and Kevin C Jones (2)*

1. State Key Laboratory of Environmental Criteria and Risk Assessment, Chinese Research Academy for Environmental Sciences, Beijing 100012.P.R.China
2. Lancaster Environment Centre (LEC), Lancaster University, Lancaster, LA1 4YQ, UK
3. Centre for Ecology and Hydrology, Library Avenue, Bailrigg, Lancaster LA1 4AP, UK

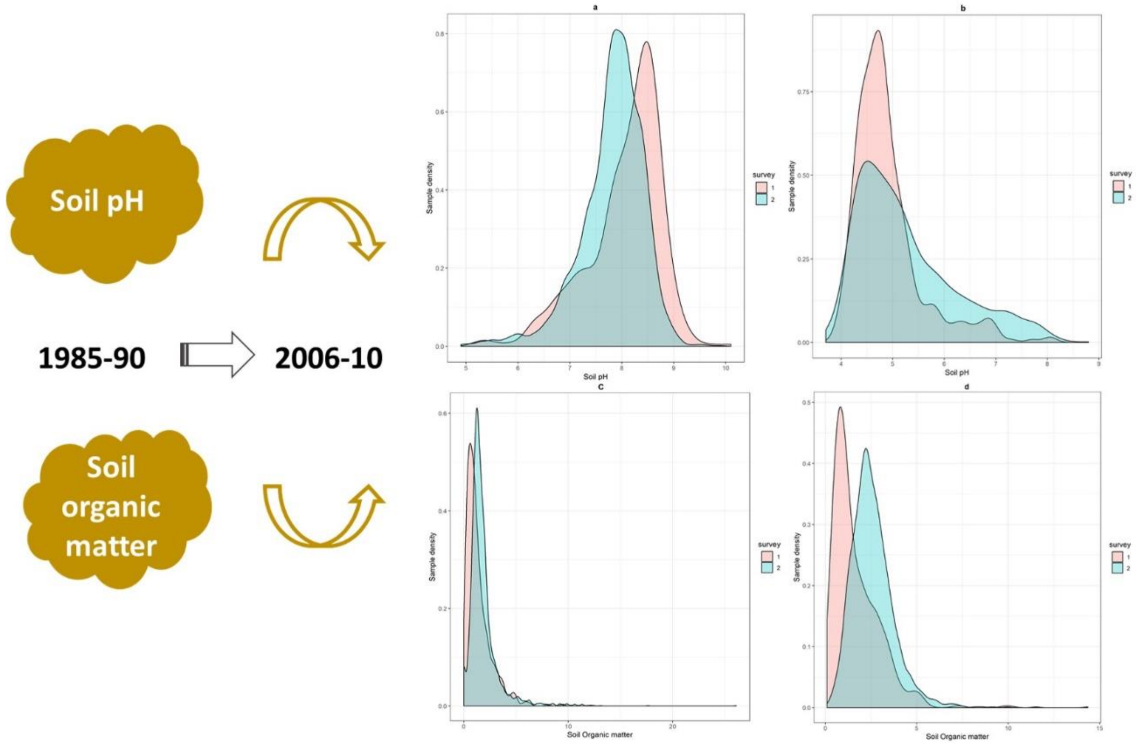
*Author for correspondence: k.c.jones@lancaster.ac.uk and guogl@craes.org.cn

Abstract

Soil organic matter (SOM) and pH are critical soil properties strongly linked to carbon storage, nutrient cycling and crop productivity. Land use is known to have a dominant impact on these key soil properties, but we often lack the ability to examine temporal trajectories across extensive spatial scales. Large-scale monitoring programmes provide the data to evaluate these longer-term changes, and under different climatic conditions. This study used data from Chinese soil surveys to examine changes in soil pH and SOM across different land uses (dry farmland, paddy fields, grassland, woodland, unused land), with surface soil (0-20 cm) collected in the periods 1985-90 (Survey 1; 890 samples) and 2006-10 (Survey 2; 5005 samples) from two contrasting areas. In the southern part of China the mean pH of paddy soils fell sharply over the two decades between surveys - from pH 5.81 to 5.19 ($p<0.001$), while dry farmlands in the northern sampling area fell slightly (from pH 8.15 to 7.82; $p<0.001$). The mean SOM content of dry farmland soil rose in both areas and the mean SOM of paddy fields in the southern area also rose (all $p<0.001$). Woodland soil pH in the south showed an increase from 4.71 to 5.29 ($p<0.001$) but no significant difference was measured in the woodlands of the northern area, although the trend increased. The SOM content of woodland top soils rose in the northern ($p=0.003$) and southern ($p<0.001$) study areas. The implications and potential causes of these changes over the two decade timespan between surveys are discussed and suggestions made as to how large scale soil sampling campaigns can be designed to monitor for changes and potential controlling factors.

Key words: Soil change; land use; soil surveys; woodland; paddy fields; agriculture

Graphical Abstract



1. Introduction

The scale of China's economic growth, the size of the country and its population, and the diversity of its climate and ecosystems mean there is great demand to understand the spatial and temporal variability in the Chinese environment. Following scientific and regulatory focus on China's air and water quality, the Government is now prioritising soil quality (State Council, 2016). Knowledge and effective management of China's basic soil resources is essential, requiring careful and systematic surveying of the terrestrial environment. Soil pH and soil organic matter (SOM) are critically important properties of soils. Understanding their variability, range and any underlying changes is fundamentally important for agriculture/food security, land use management and the environmental sciences. Soil pH is important for crop production, nutrient chemistry, soil organisms and in shaping plant community composition in natural ecosystems. SOM is critical for soil structure and workability, the ability of soils to store nutrients and water, and for the global C cycle. China's agricultural land is critical for food production and its diverse landscape is critical for the balance of natural ecosystems.

China covers 7.7% of the world's total farmland (Cai and Barry, 1994) and therefore any systematic changes have global implications. Some recent and high profile studies have reported underlying rapid changes in Chinese soils. For example, Guo et al. (2011) reported significant acidification of major Chinese croplands between the 1980s and the early 2000s, while Fang et al. (2007) and Tang et al. (2018) presented evidence of the impacts of human activities on carbon sequestration in China's soils and ecosystems. However, there is still a shortage of systematic information from which to evaluate the spatio-temporal ranges and variations in the pH and SOM of Chinese soils across different land uses. Large-scale surveys have been undertaken in China at different times and co-ordinated by different Ministries but the datasets are not widely available or evaluated yet. Here we report on pH and SOM data obtained for two time periods (1985-90 and 2006-10) across two important and climatically different parts of China. These data sets provide the opportunity to evaluate temporal trajectories in key soil properties across land use types at an extensive spatial scale, thus critically advancing the knowledge base needed to manage China's vast soils and land resources. In this paper we therefore explore the distribution of pH and SOM values for the two surveys, and test whether changes over two decades are significant; importantly, we look at differences within the main broad land-use types to determine whether temporal changes are land-use specific and consistent across the two contrasting regions. The findings are discussed in relation to other studies for China and internationally and consider the wider implications for China's land use management. Furthermore, we consider how future regional/national surveys of China's soil resources can be designed and co-ordinated in the light of international experiences, to ensure the most reliable information, capable of detecting underlying changes is obtained.

2. Material and methods

2.1. Study areas

Two major surveys of Chinese soils have been conducted by Government Ministries. The first was between 1985-90, the second was more comprehensive, with more samples taken over the period 2006-2010 (see **Table S1**). For this study, two regions were selected from those national surveys, one in the north and one in the south (see **Figure 1**).

Area 1 (north) covers 218,000 km^2. Land use types include dry farmland, paddy fields, woodland (including coniferous forest, broadleaf forest, coniferous-broadleaf forest, and shrub), grassland and unused land. Dry farmland dominates in Area 1, with wheat, maize, rice, beans and other crops being common. However, the land use in Area 1 has also undergone big changes (see **Table S2**); arable land, grassland and unused land have decreased, but woodland, garden and construction land have increased (Wu et al., 2015). Area 1 has a temperate semi-humid and semi-arid continental climate. Summers are hot and humid with high rainfall; winter is cold and dry. The most widely distributed soil types are brown earths. The main zonal soils also showed succession from the southeast to northwest, from brown earths to chestnut soils (chestnut brown soil) (Hao et al., 2017).

Area 2 (south) covers 178,000 km^2 of varying terrain, with high land in the north and lower land in the south, near the coast. It has a tropical and subtropical monsoon maritime climate. Igneous rocks dominate around a third of the province. Elsewhere it has the full range from ultrabasic to acid rocks, with acidic granite a major component (Lin et al., 2006). Three main soil types occur - latosols (pH 4.5-5.5), lateritic red soils (pH 4.5-5.6) and red soils (pH 4.5-6) (Lian, 2002). Their formation is influenced by strong soil leaching, because of the sub-tropical high rainfall conditions (Lian, 2002). Major land use types include paddy fields, a range of fruit and vegetable crops (or collectively defined ‘dry agricultural land’), woodlands (including coniferous forest, broadleaf forest, coniferous-broadleaf forest, and shrub), grasslands and unused land. Paddy fields make up the largest type, accounting for 27% of the whole area (Guo et al., 2011). A huge urbanization programme and rapid development of the economy has had a significant effect in changing the composition of land use types. The composition of land use in Area 2 has changed significantly from the 1990s, with a decrease of arable land and the increase of urbanisation, industrial and mining land (Tang, 2008) (see **Table S2 and S3**).

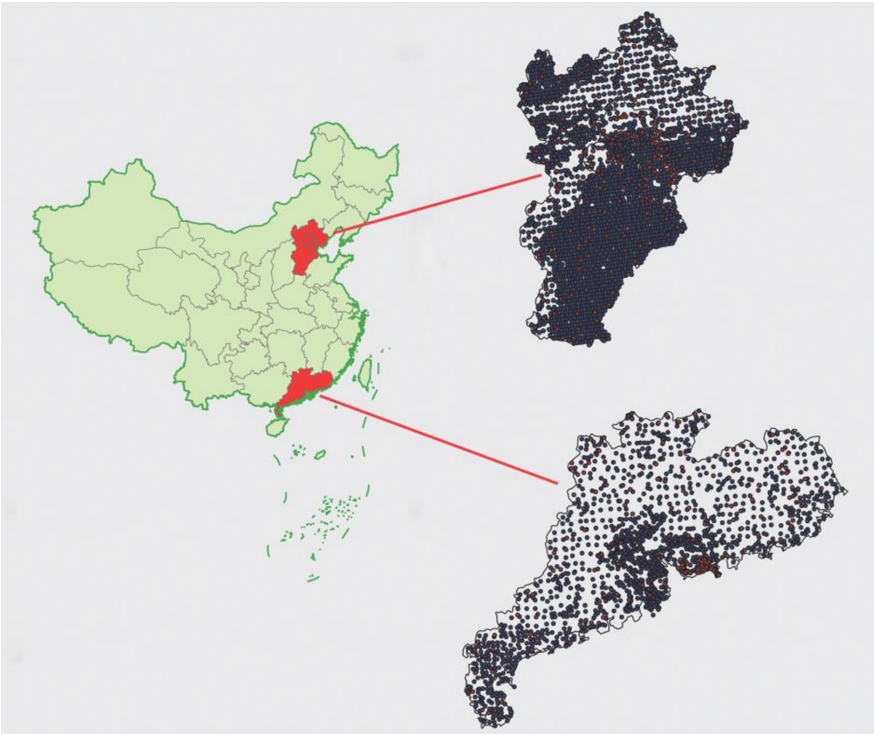


Figure 1: Soil sampling sites in north (Area 1) and south (Area 2) of China.

2.2. Soil surveys

The Chinese National Environmental Monitoring Centre (CNEMC), the Chinese Academy of Sciences (CAS), the MEP Chinese Research Academy of Environmental Sciences (CRAES) and a number of universities in China were also involved in these activities. Sampling sites were randomly selected using a grid method for the two surveys, with consideration of different environmental factors including soil types, vegetation types, land uses, soil texture etc (see **Supplementary Information** for further information). Topsoil (0-20 cm) was collected and stones, litter and large roots removed. Soil samples were dried at room temperature and then gently ground to pass through a 2 mm sieve. 100 g dry samples were used for chemical analysis. Soil pH was determined, depending on the salinity and OM status of the soils, as follows: a 2.5:1 ratio of water or saline solution for acid soils with 1 mol KCl/L, neutral and alkaline soils with 0.01 mol $CaCl_2$/L); a ratio of 5:1 for saline soil; a ratio of 10:1 for litter-rich and peat soil. SOM (%) was determined by heated oxidation with $K_2Cr_2O_7$-H_2SO_4 (185 °C), followed by back titration by $FeSO_4$ (see **Table S1**). The number of samples taken in the two surveys differed, with a more comprehensive survey conducted in 2006-2010. In summary, data was available as follows: Area 1: 1985-1990 – 500 samples, 2006-2010 – 3132 samples; Area 2: 1985-1990 – 390 samples, 2006-2010 – 1873 samples (**Table 1**).

2.3. Data analysis

Unpaired t-tests were used to examine differences in soil pH and SOM between surveys for whole areas and for separate land use types in these areas. The formula for the unpaired t-test is:

$t = \frac{\bar{X}1-\bar{X}2}{\sqrt{\frac{S1^2}{N1}+\frac{S2^2}{N2}}}$, where $\bar{x}1$, s_1^2 and N1 are the first sample mean, sample variance and sample size; x2, s_2^2 and N2 are the second sample mean, sample variance and sample size. R software was used for statistical analyses (R Core Team, 2016). The distribution of soil pH and SOM data for all samples and samples from individual land use types were visualised in the ggplot2 package (Wickham, 2016) using geom_density to produce smoothed sample densities for comparison of the surveys, and geom_hex was used to plot relationships between soil pH and SOM within land use types.

3. Results

Table 1 presents the summary of soil pH and SOM data from the surveys. **Table 2** and **3** give details of soil pH and SOM, respectively, according to land use type.

Table 1: Soil pH and organic matter in Area 1 (north) and Area 2 (south) from 1985-90 to 2006-10

Site	Year	Sample number	Soil pH		Organic matter	
			Mean	Median	Mean	Median
Area 1	1985-90	500	8.05 (6.7-8.9)	8.25	1.37 (0.23-3.7)	1.00
	2006-10	3132	7.81 (6.7-8.6)	7.9	1.83 (0.48-4.31)	1.49
	1985-90	390	4.90	4.8	1.65	1.23

Area 2			**(4.2-6.4)**		**(0.38-3.92)**	
	2006-10	**1873**	**5.26**	**5**	**2.58**	**2.41**
			(4.2-7.3)		**(1.06-4.62)**	

3.1. Characterization of pH and SOM distribution and variation

Mean (and median) pH values for all the soils sampled in Area 1 were 8.05 (8.25) in 1985-90 (n=500) and 7.81 (7.9) in 2006-10 (n = 3132) (i.e. an apparent decline). In Area 2 mean (and median) values for all the soil samples were 4.90 (4.8) in 1990 (n = 390) and 5.26 (5.0) in 2006 (n = 1873) (i.e. an apparent increase). However, it is important to note that the sites sampled and the distribution of samples across land uses differed between the surveys. The apparent overall differences in soil pH values between the two surveys are significant for soil pH (see **Table 2** for statistics**; Figure 2a, b**) and SOM (see **Table 2** for statistics; **Figure 2c, d**) but need to be seen as indicative only, with consideration given the shifts in land use composition.

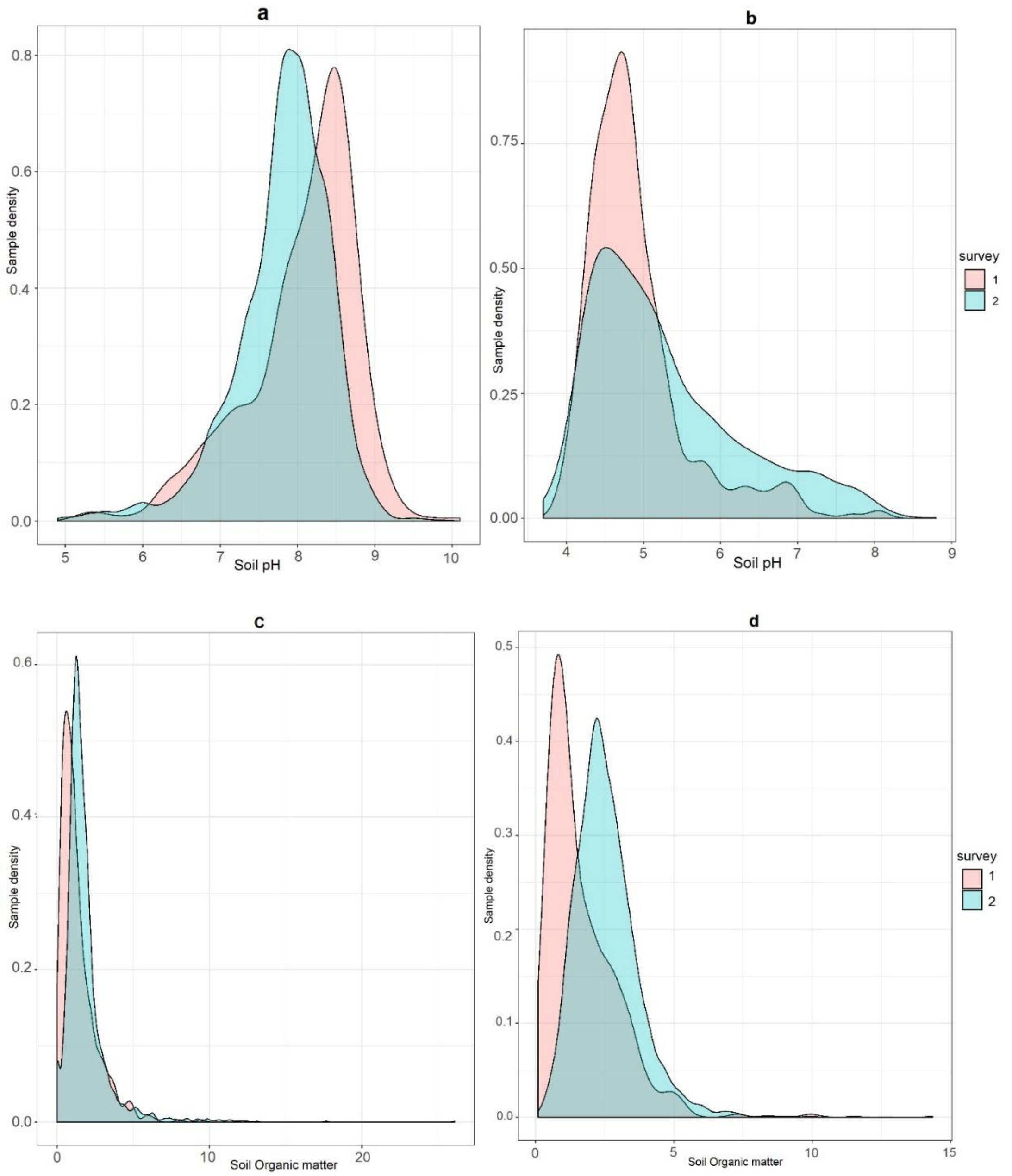


Figure 2: Sample density of pH and SOM values from both surveys for the two study regions. a. soil pH in Area 1; b. soil pH in Area 2; c: SOM in Area 1; d: SOM in Area 2. Survey 1 (pink) carried out from 1985 to 1990; Survey 2 (blue) carried out from 2006 to 2010.

More confidence can be placed on direct comparisons with those land use types that were most comprehensively sampled in both surveys. In this regard, in Area 1 the woodland (n = 101/515 in 1985-90/2006-10) and dry farmland soils (n = 334/2283) can be most confidently compared. At the level of land use type, the pH trends were different compared to each area overall, with dry farmland being significantly lower ($p<0.001$) in 2006-10 (mean = 7.82) than 1985-90 (mean = 8.15). Woodland soils were not significantly different between surveys. Repeating the test of differences between surveys, using only the subset of samples which were taken in the same locations (n = 73/27) also showed a significant reduction in soil pH from 1985-90 to 2006-10 for dry farmland ($t_{1,47} = 2.31$, $p = 0.025$). There were not sufficient samples in the same locations to do this for the other land use types. The grassland soils data summarised in **Table 2** also show an apparently significant decrease with time, but the number of samples available from 1990 was limited, so these grassland trends should be treated with some caution.

Table 2: Topsoil pH across different land use types in Area 1 and 2 in the 1985-90 and 2006-10 surveys. df = degrees of freedom.

	Land use type	N		Estimate (mean)		T-value	95 percent confidence interval		DF	P-value
		1985-90	2006-10	1985-90	2006-10					
	Dry farmland	334	2283	8.15	7.82	9.05	0.26	0.40	447.37	< 0.001

Area 1	**Grassland**	**17**	**196**	**8.52**	**7.88**	**4.04**	**0.31**	**0.98**	**20.10**	**<0.001**
	Paddy field	**6**	**45**	**8.03**	**7.91**	**0.84**	**-0.19**	**0.44**	**10.63**	**0.42**
	Unused land	**42**	**93**	**7.95**	**7.74**	**1.52**	**-0.07**	**0.47**	**49.03**	**0.14**
	Woodland	**101**	**515**	**7.70**	**7.82**	**-1.34**	**-0.29**	**0.06**	**115.34**	**0.18**
Area 2	**Dry farmland**	**23**	**163**	**4.71**	**5.11**	**-2.89**	**-0.67**	**-0.12**	**53.81**	**0.005**
	Grassland	**0**	**3**	**--**	**--**	**--**	**--**	**--**	**--**	**--**
	Paddy field	**66**	**1061**	**5.81**	**5.19**	**4.72**	**0.36**	**0.88**	**91.451**	**<0.001**
	Unused land	**0**	**4**	**--**	**--**	**--**	**--**	**--**	**--**	**--**
	Woodland	**301**	**642**	**4.71**	**5.29**	**-17.22**	**-0.65**	**-0.51**	**1251.2**	**< 0.001**

In Area 2, the woodland soils in 2006-10 (n = 642, mean = 5.29) were also higher ($p<0.001$) than in 1985-90 (n = 301, mean = 4.71), while paddy field soils were markedly lower in 2006-10 (n = 1061, mean = 5.19) than in 1985-90 (5.81) ($p<0.001$). It is noted that these mean values are derived from a wide range of soil pH values in each survey/land use, as highlighted by **Figure 3**.

Other statistically significant differences over time are summarised in **Table 2**, but it should be noted that sample numbers were more limited in these cases.

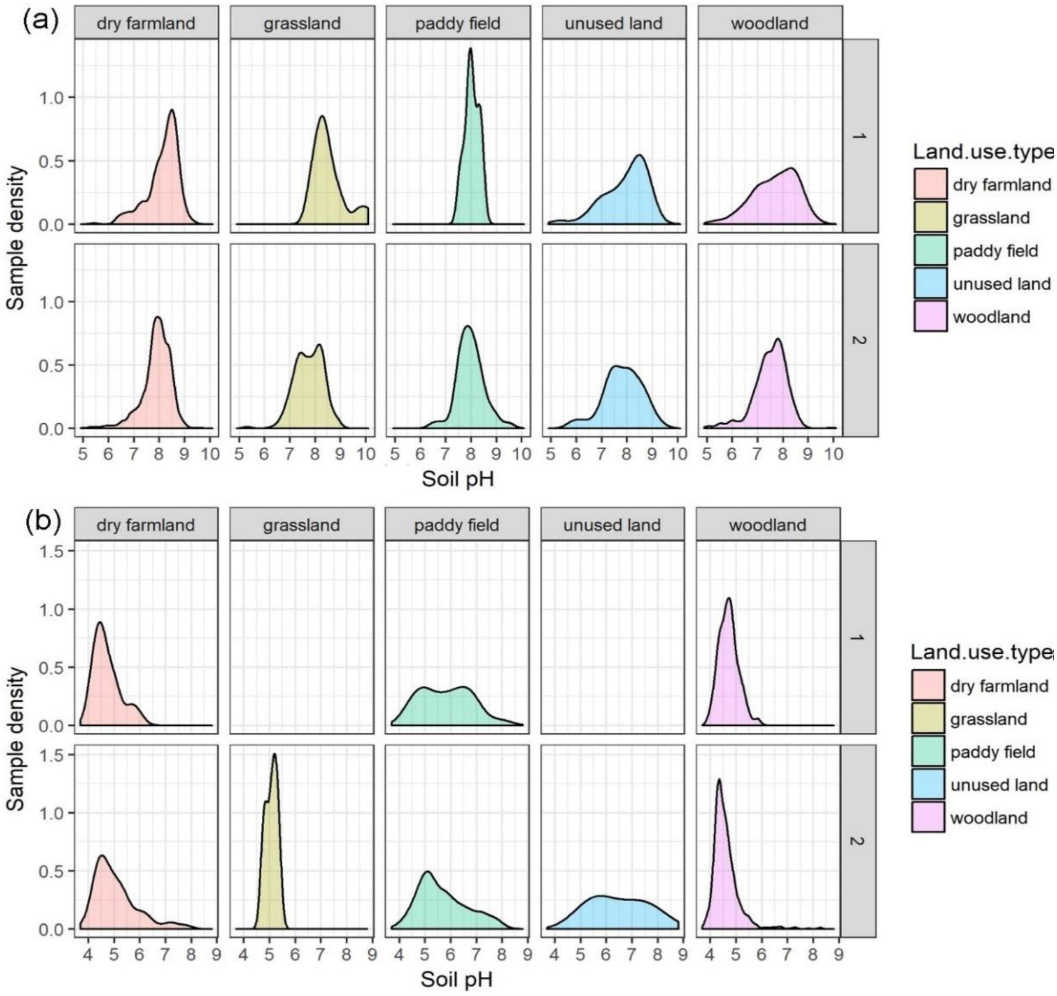


Figure 3: Sample density of soil pH values for each land use type in (a) Area 1 and (b) Area 2. Surveys carried out from 1985-90 and 2006-10.

In general, soil pH in Area 1 is higher (range 6.7-8.9) than that in Area 2 (range 4-7). Area 1 has more saline soils with higher soil pH. The distribution of soil pH values in different land use types is shown in **Figure 3**. The most complete information (i.e. greatest number of samples) is available for paddy field soils, dry farmland and woodland soils. In Area 1 the soil pH range is similar across all land use types – for example the mean for both dry farmland and woodland was 7.82 in the 2006-2010 survey. In Area 2, although mean values in 2006-10 were similar (paddy field 5.19; woodland 5.29; dry farmland 5.11), the range of values were rather different (see **Figure 3**).

3.2. Land use and SOM

In Area 1 decreasing SOM followed the sequence woodland > dry farmland > paddy field (see **Table 3** and **Figure 4**). In Area 2, the sequence was less clear and showed some differences between the two surveys: in 1985-90, woodland > paddy field > dry farmland; in 2006-10, paddy field > dry farmland > woodland (see **Table 3** and **Figure 4**).

Table 3: Soil organic matter (0-20 cm) across different land use types in Areas 1 and 2 in the 1985-90 and 2006-10 surveys.

Site	Land use type	N		Estimate (mean)		T-value	95 percent confidence interval		DF	P-value
		1985-90	2006-10	1985-90	2006-10					
	Dry farmland	334	2283	1.35	1.81	-6.69	-0.59	-0.32	561.43	<0.001

Area 1	**Paddy field**	**6**	**45**	**1.22**	**1.74**	**-1.38**	**-1.41**	**0.37**	**7.00**	**0.21**
	Woodland	**101**	**515**	**1.39**	**1.89**	**-3.00**	**-0.81**	**-0.17**	**133.71**	**0.003**
Area 2	**Dry farmland**	**23**	**163**	**1.23**	**2.59**	**-6.71**	**-1.77**	**-0.95**	**42.46**	**<0.001**
	Paddy field	**66**	**1061**	**1.63**	**2.67**	**-6.56**	**-1.35**	**-0.72**	**89.22**	**<0.001**
	Woodland	**301**	**642**	**1.68**	**2.55**	**-10.88**	**-1.03**	**-0.71**	**419.17**	**<0.001**

The overall in mean SOM content increased from 1985-1990 to 2000-2006 in both Area 1 soils (mean of 1.37% (median = 1.00%) to 1.83% (1.49%), and Area 2 soils (1.65% (1.23%) to 2.58% (2.41%)). These represent large relative differences in the two decade time interval. However, as noted previously for overall differences in soil pH, the apparent overall change in SOM summarised in **Table 1** and **Figure 2** need to be interpreted along with additional information, because the sites sampled and the distribution of samples across land uses differed between the surveys. It is therefore important to look at the land use types separately.

In Area 1, the statistically significant results were for dry farmland, woodland and grassland, with the caveat noted above about the limited number of grassland samples analysed from 1985-90. Dry farmland SOM increased from 1.35% to 1.81% ($p<0.001$), woodland from 1.39% to 1.89% ($p=0.003$) and grassland from 0.93 to 1.89% ($p<0.001$). In Area 2, dry farmland, paddy field and woodland SOM all showed statistically significant ($p<0.001$) increases, from 1.23 to 2.59%, from 1.63 to 2.67% and from 1.68 to 2.55%, respectively (see **Table 3** and **Figure 4**).

Repeating the test of differences between surveys using only the subset of samples which were taken in the same locations (n = 73/27) also showed a significant increase in SOM from 1985-90 to 2006-10 for dry farmland ($t_{1,45} = 2.02$, $p = 0.049$). As for soil pH, there were insufficient samples taken in the same locations to do this for the other land use types.

Previous studies have explored the relationship between SOM and pH for soils across China and different regions (e.g. see Dai et al. (2009)). The relation between these important two variables is complex and highly variable, because it depends on many factors – notably geology, climate, vegetation types, soil microbiology, and land use management. There were no clear relationships between SOM and pH within each land use types, neither by region or survey (see **Figure S1)**.

In summary, the key results from this study are as follows:

Agricultural soils - the mean pH of paddy soils in Area 2 fell sharply ($p<0.001$) between 1985-90 and 2006-10 - from pH 5.81 to 5.19, while dry farmlands in the north fell slightly (8.15-7.82) but significantly ($p<0.001$) too. The mean SOM content of dry agricultural land rose sharply ($p<0.001$) in both Area 1 and Area 2. The mean SOM of the Area 2 paddy fields also rose significantly ($p<0.001$).

Woodland soils – woodland soil pH in Area 2 showed a net increase ($p<0.001$) from 4.71 to 5.29; no statistically significant difference was measured in the woodlands of Area 1. The SOM content of woodland top soils, rose sharply, in the northern ($p=0.003$) and southern ($p<0.001$) study areas, respectively.

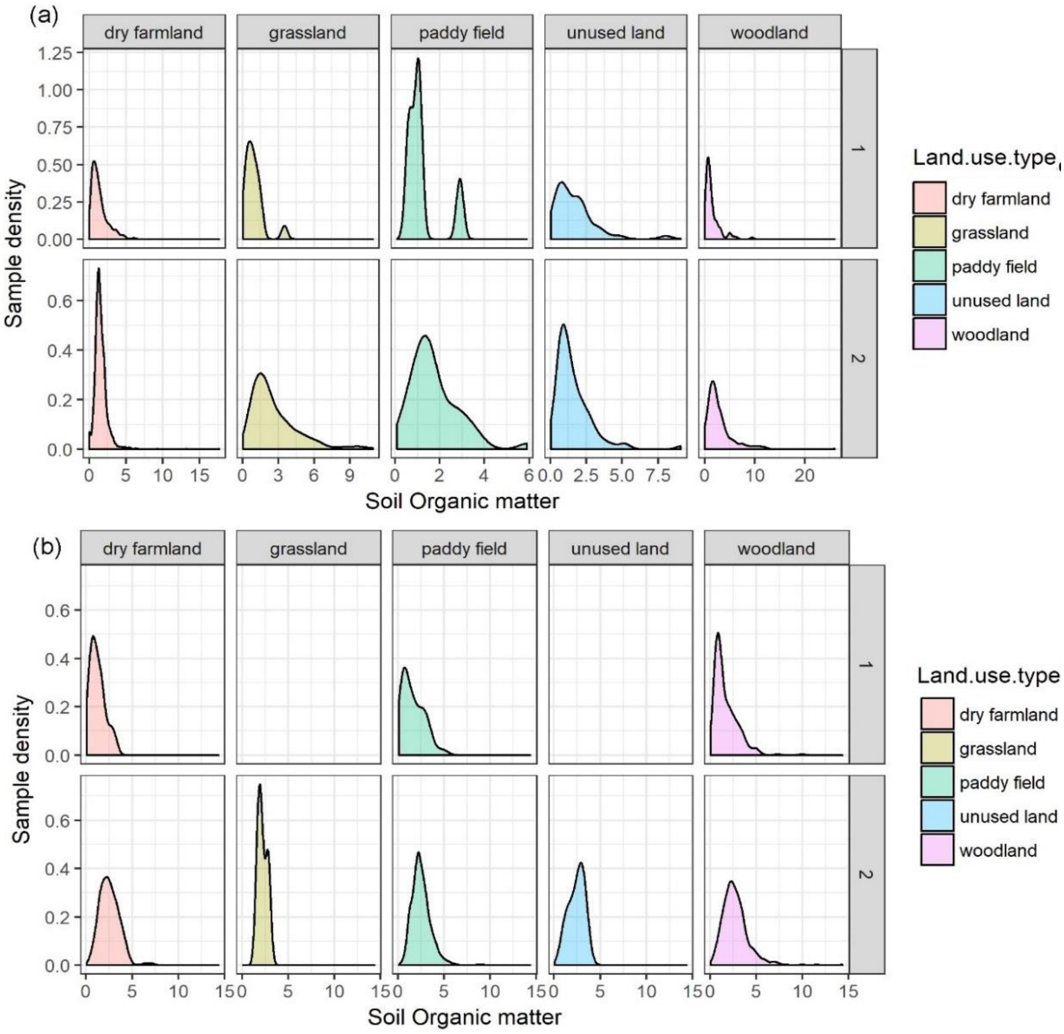


Figure 4: Sample density of soil organic matter values for each land use type in (a) Area 1 and (b) Area 2. 1: survey carried out from 1985 to 1990; 2: survey carried out from 2006 to 2010.

4. Discussions

The changes in soil pH and SOM across two contrasting regions of China represent major differences in the two decade time window of this study. They have significant implications for carbon storage, nutrient cycling and crop productivity, and need to be understood to optimise land management in different environmental contexts and avoid degradation of China's soil resources. Agricultural soils of the different regions demonstrated variable change depending on specific land use type; soil pH in dry farmlands decreased in the north and increased in the south, whereas paddy field soils decreased in both regions but to different extents. In woodland soils, there were increases in soil pH in both regions, though this was only significant in the south. Soil organic matter tended to increase in all land use types but to a greater extent in the south where soil types generally had lower pH and climate is sub-tropical. Interactions between the composition of land use and environmental conditions play a key role in determining the trajectory of soil quality at large spatial scales. Below we discuss these findings in more detail in terms of other large-scale studies of soil change, potential causes of change and the implications for future management and monitoring.

4.1. Have such rapid changes in soil pH and SOM been reported before?

Previous studies have reported underlying recent and rapid changes in soil pH in Chinese soils. For example, Guo et al. (2010) found soil pH in major Chinese crop-production areas significantly decreased from the 1980s to the 2000s. They compared cropland soil pH in the 1980s and 2000s using results from two nationwide surveys, 154 paired sites and long-term agricultural sites. They reported declines in pH under cash crop systems and under cereals, with the size of reduction influenced by soil type and soil pH range (i.e. some function of buffering capacity). For example, leached red soils (typically pH~5) in southern China declined by 0.23-

0.30 pH units, while fluvo-aquic soils in the north declined by 0.27-0.58 units. They were able to show the relative contributions of different processes to increased acidity followed the sequence: processes related to N-cycling > base cation uptake by crops > acid deposition. The widespread use of N fertilisers, they argued, accounted for most of the decline in soil pH. Guo et al. (2018) observed paddy soil pH decreased by an overall 0.6-unit from 1980 to 2010 in Jiangxi Province. Guo et al. (2011) also reported soil pH in Guangdong Province decreased from 5.7 to 5.44 based on ca. 30-year data. The dataset reported here adds important information with a systematic assessment of soil pH and SOM in all the main land use types, highlighting temporal changes in agricultural and woodland soils. Yang et al. (2015) reported a significant decreasing trend in soil pH occurred in broadleaved forests and minor changes occurred in coniferous or mixed coniferous and broadleaved forests by using historical soil inventory data from the 1980s and a data set synthesized from literature published after 2000 in the forest ecosystem. Soil pH of tea plantation decreased from 1980s to 2010 based on 2058 soil samples from 19 provinces (Yan et al., 2020). A significant decreasing in different soil depths trend from 1980s to 2010s in Chengdu Plain of China (Li et al., 2020).

Probably the world's most systematic assessments of long-term soil changes have been conducted in the UK, with a combination of long-term (>100 years) controlled arable and pasture grassland agricultural plot trials at Rothamsted Research station (Blake et al., 1999; Johnston et al., 1986) and the Great Britain *Countryside Survey* across a wide range of habitats, with several thousand samples taken in 1978-2007 (Keith et al., 2015; Reynolds et al., 2013). These provide support to our study with comparable findings across a similar time period, namely: the generally significant increase in pH across most UK habitats from 1978 to 2007, by up to 0.6-0.8 pH units for some; there are some differences comparing England and Scotland, highlighting broad regional differences. Soil C concentrations decreased in arable and horticulture habitats (considered most equivalent in terms of land use intensity to ‘dry farmland’ in this study), but increased under broadleaved/mixed woodlands (Reynolds et al., 2013). The controlled Rothamsted experiments provide the clearest controlled and quantifiable evidence of changes in pH linked to atmospheric deposition and N inputs (Hütsch et al., 1994), together with increasing soil C in response to organic matter amendments of farmland (e.g. addition of straw stubble and livestock manures) (Powlson et al., 2011a; Powlson et al., 2011b). Increases in soil pH in recent decades in some UK soils have been linked to reduced sulphur acid deposition inputs (Blake and Goulding, 2002; Emmett et al., 2010), as the UK’s emissions from coal combustion, industry and domestic heating sources have declined (Emmett et al., 2010).

4.2. What factors could cause such changes?

Changes in topsoil pH and SOM over time are caused by a shift in the balance between inputs and losses. For pH, this is the balance between H ion inputs from soil weathering, acidifying atmospheric deposition and additions in fertilisers and plant residues. For SOM, it is the balance between the rate of accumulation of the C stock (from photosynthesis, C additions in leaf litter, stubble and residue incorporation) and the rate of decomposition/leaching/other losses. The systems studied here differ in their inputs/losses and their ability to buffer changes. Paddy field soils have very different inputs/losses to woodland systems, for example. To understand the changes seen in the systems studied here, it is therefore necessary to consider inputs/losses, and other large-scale environmental and management factors, that have changed over recent decades to shift the balance of hydrogen ions and soil C stocks in the different Chinese ecosystems studied here.

The loss of soil C can be relatively rapid (e.g. after moving from grassland to arable, or following ploughing/disturbance), compared to the length of time and inputs required to build up soil C stocks. Active management of the C inputs added to agricultural soils can have major impacts on C stocks. A long-term study from Thomsen and Christensen (2004) reported SOM clearly and persistently increased with the annual application of straw and ryegrass. For example, when the amount of straw returned was 4 t/hm^2, 8 t/hm^2 and 12 t/hm^2, after 18 years, soil C increased by 12%, 21% and 30%, respectively.

China's 'dry' agricultural lands have seen great changes in land management practices over recent decades, through the Land Reform, the drive towards agricultural self-sufficiency, greater use of fertilisers and pesticides, and often with changes in agricultural practices (Fei et al., 2010; Han et al., 2017; He et al., 2018; Zhao et al., 2018). Some of these changes have been imposed/adopted regionally. Such factors include: greater incorporation of crop residues; greater addition of livestock manures; high fertiliser loadings and use of pest control agents; mechanisation and changes in the crops grown and cropping patterns. Similarly, China's 'wet' agricultural lands (paddy fields) have also seen shifts in practice, which have resulted in dramatic gains in rice yields in China since the 1950s. These include: improved varieties of rice; changes to the incorporation of crop residues; much greater fertiliser use and changing inputs via atmospheric deposition; and changes in irrigation practice or cropping patterns. These changes also differ between regions and land use types, which makes it difficult to predict how the SOM inputs and C cycling have been impacted; China's agricultural extension service farm plots can potentially provide an important resource to conduct systematic studies of the factors influencing SOM (and pH) trends. Woodland systems and soils have also witnessed changes in several factors, which can influence the SOM dynamics of topsoils. These include: shifts in the proportions of primary and secondary woodland; the degree of active woodland management (e.g. clearance/felling/species mix/planting programmes); changing atmospheric loadings of CO_2 and nutrients, which can affect woodland productivity and C storage. Future work is needed, to systematically monitor soil changes and to assess the contribution of these drivers in controlling the pH and SOM content of China's soils resource, to help explain the trends seen here and in other studies.

Guo et al. (2010) published a comprehensive survey of soil pH in Area 2, where they were able to compare soil types from the 1980s with data from 2002-07. They focussed on trend differences between soil types. Alluvial soils from river valleys and the Pearl River Delta increased in soil pH, while red soils and paddy soils decreased. They also noted how major land use changes and agricultural practices, including urbanisation, acid mine drainage and excessive fertiliser use, had influenced the province. These important factors cannot easily be studied with our survey results, because precise information on soil types, locations and agricultural inputs are not known. However, the survey data presented here adds to the body of evidence showing rapid changes in critical soil properties in Chinese soil systems.

4.3. What are the main implications of the changes reported here?

This study shows that the basic properties of Chinese soils are changing quickly - they are dynamic, not static, systems. Rates of change in soil pH are fast and in line with some other recent published work from China and the UK that demonstrate significant change on decadal timescales. Perhaps the greatest concern is that agricultural soil pH is declining, notably that of paddy field soils, which supply rice – the key staple foodstuff – to much of China's population. Greater acidity, particularly in the pH 4–6 range, can induce Al and Fe toxicity in crop plants, affect nutrient availability, soil fertility and crop yields. Reversing agricultural soil acidification is costly and labour/resource intensive.

4.4. How can future surveys be conducted to verify underlying trends and shed light on causes?

China is committed to soil surveys – with large resources and man-power at its disposal. This is clear from the scale and intensity of the national surveys already conducted. For example, the most recent national survey of soil pollutant quality (for selected heavy metals and organic contaminants) in the 2000s took many thousands of samples across China. Indeed, another national survey is being conducted now. However, what this study shows is that it is critical to be able to improve the quality of information obtained from such surveys, to give definitive information on the extent and scale of underlying changes in soil pH and SOM, and to yield information to explain the causes, in a way that is not possible from this study. This needs very careful design, handling and analysis, to ensure thorough statistical interpretation can be assured, capable of detecting underlying changes and their causes. This is not simply a matter of analysing large numbers of samples. Knowledge of other national soil monitoring programmes and experience operating the long-running GB Countryside Survey in the UK are valuable in guiding future soil monitoring programmes in China, and the following aspects of monitoring are considered important:

Sampling strategy: Survey designs for national sampling strategies across Europe include, amongst others, systematic or gridded sampling and stratified random sampling (Van Leeuwen, 2017). These designs allow selection of sampling locations to be representative of the prevailing composition of land uses and soil types, and provide unbiased estimates to enable upscaling. Since land use can change over time, a survey sampling design which is not based on land use types is more flexible and temporal estimates can be reported with and without land use change. The Countryside Survey uses the ITE Land Classification (Bunce et al., 1996) which stratifies Great Britain according to major environmental gradients (e.g. climate, geology, topography). In a stratified random survey, it is important to consider sample replication within strata and power analyses may be needed for different reporting classifications and metrics, particularly if devolved or regional reporting is required.

Co-location of data: Measurements taken from the same sampling locations provide the basis for robust integrated modelling of different data. The most effective soil monitoring programmes would combine collections of biological, chemical and physical properties, along with functional measures of the soil, and the assessment of the plant community. The unit of replication for strata is a 1 km square in the survey design of the Countryside Survey but, for soil monitoring, there are five sampling plots within each 1 km square; soil, vegetation and habitat data are linked in these plots and this co-location has been exploited in a variety of integrated modelling activities (Caruso et al., 2019; Maskell et al., 2013; Norton et al., 2018; Reynolds et al., 2013). It is important to capture detailed data on the plant community in conservation areas or national parks, where indirect drivers may be causing changes in vegetation composition that are not picked up in intensively managed habitat or with a coarse land use type. Other data such as climate and landscape-level metrics are linked at the 1 km resolution.

Sample archives: The Countryside Survey has air-dried and frozen soil samples, which are catalogued and stored in dedicated archives. This means that new analyses can be undertaken on stored samples and, importantly, comparisons of methods can be made when they are updated or change.

Repeated sampling: Large-scale monitoring often evaluates data as a population of samples, for example those from different land uses as done in this study. Sampling the same set of locations over time (e.g. every 5–10 years) provides the strongest statistical basis to analyse changes over time. In order to do this, it is important that precise sampling locations can be re-located in subsequent surveys; this is done using GPS coordinates, detailed written descriptions and plot and landscape photographs for CS. Statistical analyses, however, should be flexible enough to accommodate a mixture of old, repeat and new sampling locations (Scott, 2008); it is therefore very important to have a systematic schema for uniquely identifying sampling locations, so that data can be efficiently handled and combined for analyses. Recent Chinese papers discuss some of these issues in detail (Peng et al., 2016; Song et al., 2017).

Acknowledgements

We would like to thank the National Natural Science Foundation (Grant no. 41571311) of China for their support.

Summary

Overall, China's soil is undergoing change, which differs between different land uses and surveys. Therefore, the impact of these changes needs to be considered, when determining soil standards. Currently, China is taking active actions to tackle soil management. In addition, China is meeting the shortage of land resources in urban areas. However, how to better utilize and redevelop brownfield sites in urban areas is still an unsolved problem in China. Therefore, brownfield management methods in China, UK and US are discussed in the next Chapter.

Supplemental information for

TITLE: Decadal shifts in soil pH and OM differ between land uses in contrasting regions in China

RUNNING TITLE: pH and organic matter changes in Chinese soils

	Survey	
	1985-90	2006-10
Organization	China National Environmental Monitoring Centre; Department of geography, Peaking University; Institute of soil ecology, Chinese Academy of Sciences	China National Environmental Monitoring Centre; Chinese Academy of Sciences; Universities
Sampling method	Systematic random	Systematic random
Sampling depth	0-20 cm	0-20 cm
Number of soil samples	890 (Area 1: 500, Area 2: 390)	5005 (Area 1: 3132, Area 2: 1873)
Soil pH analytical method	The ratio of water or saline solution (acid soil is 1 mol/L KCl, neutral and alkaline are 0.01 mol/L $CaCl_2$) to soil is 2.5:1, saline soil using 5:1, litter layer and peat soil using 10:1.	The ratio of water or saline solution (acid soil is 1 mol/L KCl, neutral and alkaline are 0.01 mol/L $CaCl_2$) to soil is 2.5:1, saline soil using 5:1, litter layer and peat soil using 10:1.
SOM method (%)	$K_2Cr_2O_7$-H_2SO_4, back titration by $FeSO_4$	$K_2Cr_2O_7$-H_2SO_4, back titration by $FeSO_4$

Table S1: Sampling and analytical methods employed in the two surveys.

	Land use type						
	Arable land	Garden	Woodland	Grassland	Other agricultural land	Construction land	Unused land
Number of samples in 2000	766	69	458	82	55	275	464
Ratio of soil sample in each land use type to total samples in all land use type in 2000	0.35	0.03	0.21	0.04	0.03	0.13	0.21
Number of soil samples in 2013	721	101	540	49	92	296	386
Ratio of soil sample in each land use type to total samples in all land use type in 2013	0.33	0.05	0.25	0.02	0.04	0.14	0.18
Annual rate of change	-0.45%	3.6%	1.4%	-3.1%	5.3%	0.59%	-1.3%

Table S2: Illustrative land use types and their coverage in Area 1 (2000-2013).

Year	Urbanization level (%)	Non-agricultural population (ten thousand people)	Agricultural land (ten thousand hm²)	Construction land (ten thousand hm²)	Unused land (ten thousand hm²)
1996	30.6	2170	1518	142	137
1997	31	2173	1513	147	136
1998	31.2	2219	1511	150	136
1999	31.2	2276	1509	153	135
2000	31.2	2338	1508	155	134
2001	31.6	2391	1506	156	135
2002	36.2	2767	1500	161	135
2003	47.7	3682	1497	165	135
2004	48.7	3798	1493	168	135
2005	51.7	4082	1494	171	132

Table S3: Urbanization level in China from 1996 to 2005.

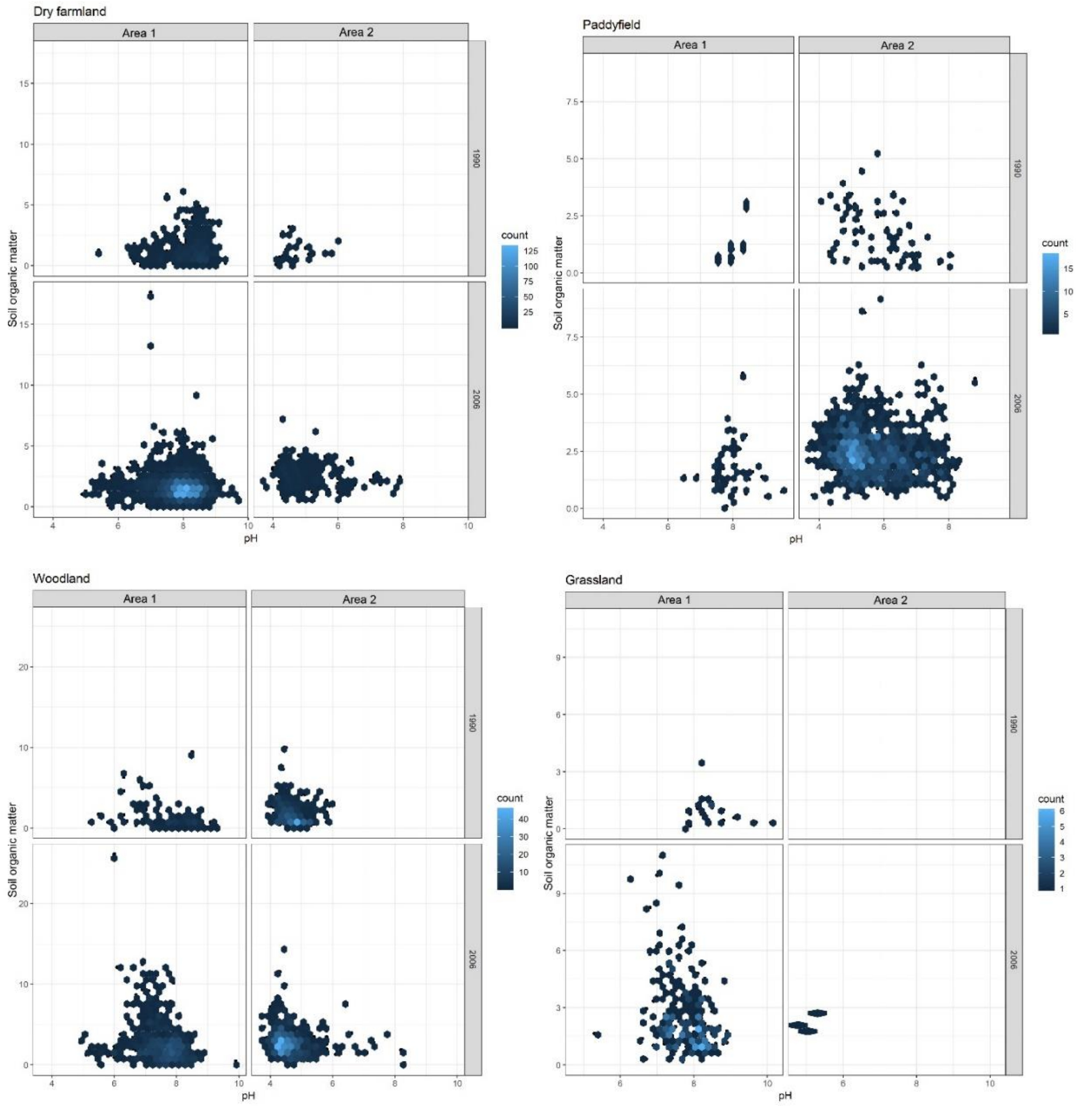


Figure S1: Scatterplots of Soil organic matter (SOM) and pH for the major land use types covered in both surveys.

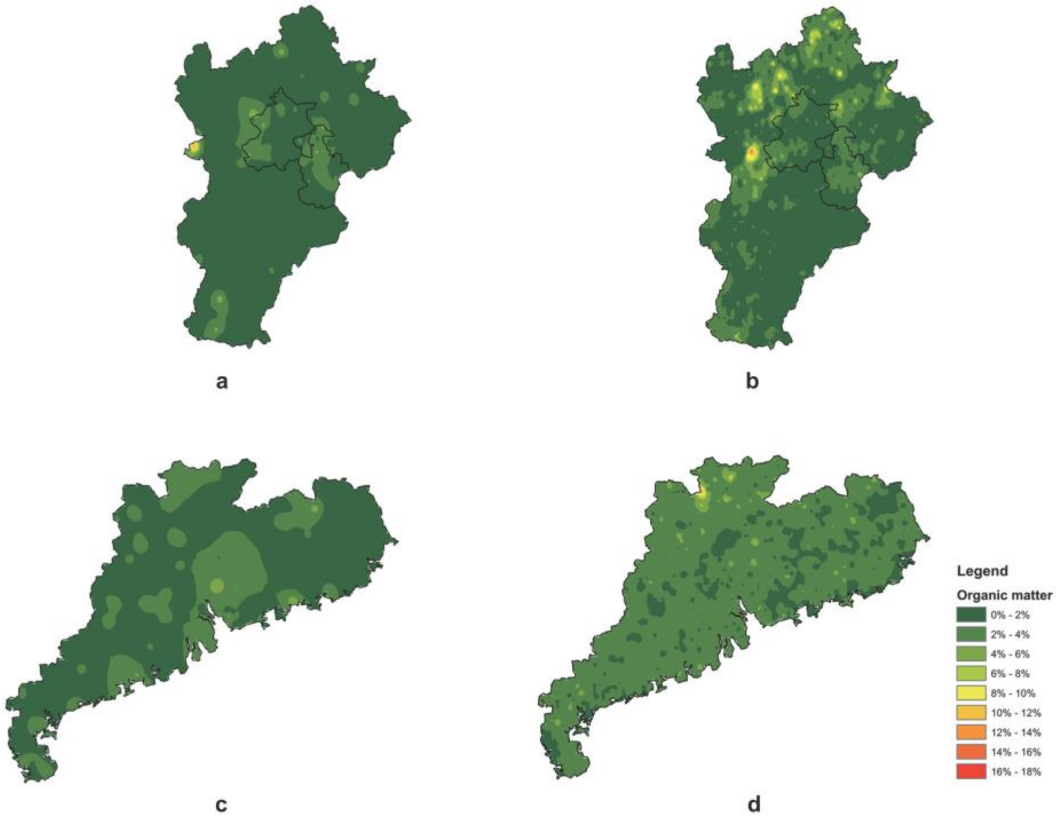
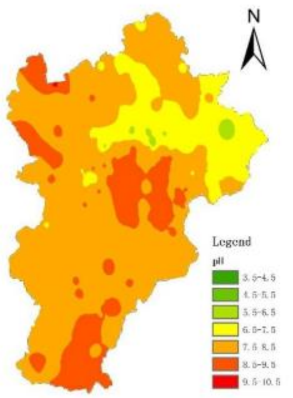


Figure S2: Map of modelled soil organic matter plotted in Areas 1 and 2 from 1990 to 2013 by using the spatial interpolation method of inverse distance weight. a. Area 1 in 1990, b. Area 1 in 2013, c. Area 2 in 1990, d. Area 2 in 2016.



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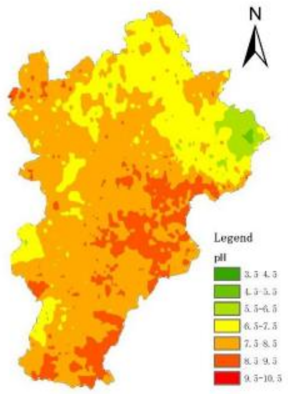

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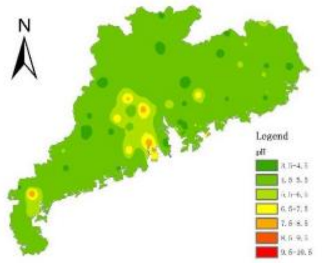

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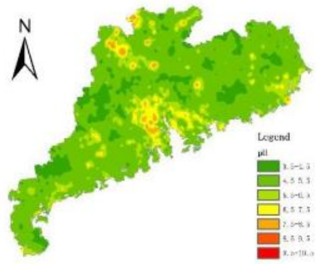

d

Figure S3: Maps of soil pH plotted in Area 1 and 2 from 1990 to 2013 by using the spatial interpolation method of inverse distance weight. a. Area 1 in 1990, b. Area 1 in 2013, c. Area 2 in 1990, d. Area 2 in 2016.

IV

Re-development of urban brownfield sites in China: motivation, history, policies and improved management

Yiming Sun[1], Hong Li[1,2], Kirk Semple[1], Frederic Coulon[3], Qing Hu[4], Jingyang Gao[6], Guanlin Guo[5] and Kevin C Jones[1]*

[1]Lancaster Environment Centre (LEC), Lancaster University, Lancaster, LA1 4YQ, UK

[2]Centre for Ecology and Hydrology, Library Avenue, Bailrigg, Lancaster LA1 4AP, UK

[3]School of Water, Energy and Environment, Cranfield University, MK43 0AL, UK

[4]Southern University of Science and Technology, 518055, Shenzhen, Guangdong, China

[5]Technical Centre for Agricultural and Rural Ecology and Environment, Ministry of Ecology and Environment, Beijing 100012, PR China

[6]Beijing Huanding Environmental Big Data Institute, 100083, Beijing, China

Abstract

Rapid urbanization in China has resulted in increased demand for land in towns and cities. To upgrade and modernize, China has also moved many major industries from urban centres to less populated areas. With the high economic value of urban land, the transformation and utilization of brownfield areas have become important economically and socially. The Chinese government has recognized the need for strong frameworks to safeguard soil and groundwater quality, with brownfield sites a key category for management. Strong scientific, regulatory and decision-making frameworks are needed and being adopted, to ensure practical, careful and wise use of central and localized Government resources, to manage the re-use and regeneration of these brownfield sites. This paper reviews the context, policies and management procedures to develop brownfield sites in countries with a history of brownfield management, and discusses China's current situation and priorities for brownfield governance and redevelopment. These include: clarification of brownfield site soil quality standards and risk assessment procedures; and the responsibilities of different national and local level agencies; establishment of a national expert committee to advise on best practice, policy and process; the use of registered brownfield databases at national, provincial, municipal and county levels; set up soil pollution prevention fund at provincial level.

Keywords: brownfield, urbanization, urban soils, management, China, USA, UK

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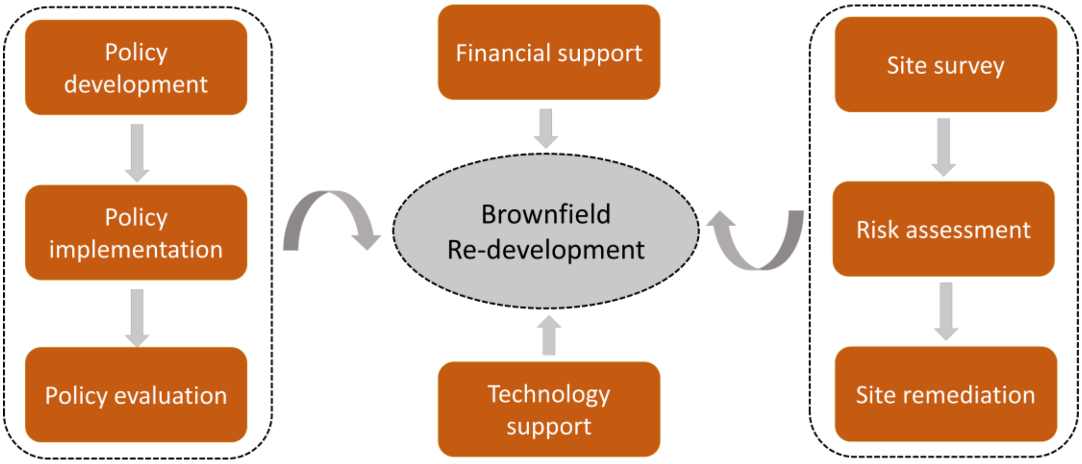
Policy development
Policy implementation
Policy evaluation
Financial support
Brownfield Re-development
Technology support
Site survey
Risk assessment
Site remediation

1. Introduction

1.1 Urbanization in China

Over the past 40 years of reform and development, China has undergone remarkable economic growth. The scale of China's urbanisation and the number of growing large metropolitan regions where this urbanisation is concentrated are globally unprecedented. Many older industrial facilities along the edge of, or within, the city boundaries are being relocated or closed, leaving behind derelict, underused and abandoned land contaminated by the former industrial activities. At the same time, the continuous outward shift of urban boundaries and the expansion of territorial jurisdictions of cities, primarily through the expropriation of surrounding rural land and its integration into urban areas, means that these new urban and peri-urban expansions increase fragmentation of the landscape (Coulon et al., 2016b). In effect, fragmentation decreases connectivity, causes green space loss and impacts upon the ecology and function of green space. The restoration of the functionality of green space often requires restoring the ecological connectivity of this green space within the city matrix and enhancing the biodiversity therein.

With the rapid development of urbanization, land resource is indeed becoming increasingly valuable. Despite the differences in urban structures in China and other countries, all countries face an ongoing trend towards urbanization, re-densification and an increased stock of marginal land. While many municipalities and initiatives worldwide are pursuing green and sustainable urban development, transparent indicator-based evaluation systems are necessary to ensure that planning and action do indeed lead to increased sustainability and to a higher quality of life for the population in cities. A shared endeavour is therefore needed to promote the development and implementation of the vacated and abandoned land in China which is often called ‘brownfield’ in western countries. This has been identified as a priority for environmental regulation and management in China (Liang and Yang, 2013, Xue et al., 2012, State Council, 2016). China is engaged in serious efforts to implement brownfield redevelopment on a large scale. However, in contrast to the UK, the US and other countries, the Chinese government for various reasons like retaining competitiveness, intends to initially introduce the brownfield redevelopment framework on a smaller scale through a number of pilot studies so that it has a better basis for assessing its large scale and full coverage in the longer run.

1.2 The Brownfield concept

The term ‘brownfield’ was first used in the Comprehensive Environmental Response, Compensation and Liability Act (CERCLA) in the United States in 1980 (Foley, 2014). The main purpose of the Act was to solve the problem of legacy soil pollution in industrial sites. Around 1990, the term brownfield also occurred in British planning regulations, which refers to ‘previously developed land’ (PDL) as unused or exploitable land, including vacant, abandoned land and currently used land with the potential for redevelopment (Hu, 2019, Dunstone, 2013, Adams and Watkins, 2002). In the UK, ‘brownfield’ is widely understood to be abandoned or vacant land that can be redeveloped in accordance with planning policies or urban revitalization goals (Sam et al., 2017). In the United States, ‘brownfield’ is generally interpreted as occupied or contaminated land (Xue et al., 2012, Zhao et al., 2014, Marker, 2018).

Alker et al. (2000) proposed a comprehensive definition – ‘a brownfield site is any land or premises which has previously been or developed and is not currently fully in use, although it may be partially occupied or utilised. It may also be vacant, derelict or contaminated. Therefore, a brownfield site is not available for immediate use without intervention’. Other useful terms include: derelict land – ‘Land damaged by industrial or other development that is incapable of beneficial use without treatment’ (DoE, 1995) and contaminated land – ‘an indication of the presence of some biological, chemical or physical hazard on or within a site that would require some treatment before the site could be reused.’

1.3 Brownfield in China

In China, the term brownfield was first mentioned by Niu (2001) when he introduced the US brownfield definition and regulations and they can be applied to real case studies in China (Niu, 2001). According to the World Bank's ‘Waste Management in China: Problems and Suggestions’ issued in 2010, there were ‘at least 5,000 brownfield sites’ in China. In reality, this estimate is likely to be 1-2 orders of magnitude greater (Li and Li, 2010).

The national soil survey published in April 2014 by the Environmental Protection and Land and Resources Ministries of China revealed the significant challenges China (Zhao et al., 2015, Changsheng et al., 2016) is facing with soil pollution. Extrapolation of the soil survey indicated there were substantial areas (36% of sampling points) within the vicinity of industrially contaminated sites being potentially contaminated (Ministry of Environmental Protection of the People's Republic of China and Ministry of Land and Resources of the People's Republic of China, 2014).

In 2008 the Ministry of Environmental Protection issued the ‘Opinions on Strengthening the Prevention and Control of Soil Pollution’, and put forward corresponding action measures namely: (1) completing the investigation of soil pollution situation in a ‘comprehensive’ way; (2) establishing a soil environmental monitoring network; (3) compiling and completing national and local soil pollution prevention and control plans; (4) establishing policies and laws for soil pollution prevention and control; and (5) establishing a management system framework such as laws and regulations (Ministry of Environmental Protection of the People's Republic of China, 2008).

China’s State Council – the central government’s main governing body - released the ‘Action plan for soil pollution prevention and control (10-point soil Action plan)’ in 2016. This has been heralded as a key development in identifying and prioritizing wise use and management of China’s soils resources. The 10-point plan presents the requirements, work plan and main goals of China’s national soil contamination prevention priorities (State Council, 2016) (Table SI).

The Chinese authorities also committed over 30 billion RMB within the countries twelfth Five-Year Plan to address soil pollution, along with a specific plan of action for the prevention and control of soil pollution coming into force during the period of the 13th Five-Year Plan (2016-20). This along with the development of the nation's first specific national law on the control and prevention of soil pollution being drafted by China's Environment Ministry demonstrates the commitment for long term soil management and regeneration of industrialized sites. China has set very ambitious targets for a high percentage of contaminated sites to be used by 2020 and beyond and established a soil quality standards system (State Council, 2016). It further promotes on-site remediation; as well as opening up of the monitoring services market. This paper is timed to showcase highlight the needs and opportunities arising from rapid urbanisation and the changes in land use resulting from industrial change. This has left a legacy of vast polluted industrial and commercial areas (also called brownfields) and marginal land areas in China. The paper also highlights the remaining challenges and opportunities for the brownfield market in China.

2. UK and US experiences of brownfield redevelopment

2.1 The development of brownfield management in US

In the late 1970s, some contaminated land incidents raised government and public attention in the US. In 1980, the CERCLA was released to deal with these. This required the owners, users and polluters of real estate to bear the consequences of land pollution, and to clarify the cost of land governance through the form of law.

In 1986, the Superfund Amendments and Re-authorization Act (SARA) updated some provisions: 1. To emphasize the importance of technological innovation in permanent remediation and remediation of hazardous waste sites; 2. Ensure environmental laws and standards of the Federal and States Governments should be taken into account when implementing Superfund operations. 3. A new executive body and dispute settlement mechanism was proposed. 4. Increase the involvement of state governments in each phase of the Superfund Plan; 5. Pay more attention to the human health problems caused by hazardous waste sites; 6. Encourage more citizens to participate in the decision-making of site restoration process; 7. Increase the investment of trust funds.

As mentioned in section 2.1, the framework for the rehabilitation of contaminated sites in the US mainly includes the CERCLA passed in 1980. This bill, often referred to as the 'Superfund Law', establishes the 'polluter pays' principle, which stipulates that different parties (legally defined as "potentially responsible parties") are responsible for remediating historically contaminated sites. In addition, the 'Superfund Law' authorizes the US Environmental Protection Agency to force any potential responsible party to pay for the remediation of the site. The sharing of site remediation costs and the sharing of responsibilities will be resolved between potential responsible parties. However, the CERCLA also has been criticized for shortcomings, including causing many legal proceedings, causing burdens on small businesses, and insufficient participation of State governments and local communities (the main actions are the responsibility of the federal government). Further to this, several investors and developers are discouraged from becoming involved due to uncertainties with regards to responsibilities and lability, leaving sites empty or undeveloped, and eventually becoming brownfield. These shortcomings of the law have gradually been corrected through multiple rounds of amendments and reforms to the Superfund program over the years, including the 2002 Small-Scale Corporate Responsibility Mitigation and Brownfield Revitalization Act and other brownfield-related Projects and plans (Figure SI). The revised Superfund Law is now welcomed by various stakeholders. These amendments and reforms are practical lessons for developing countries like China. In addition, the lessons learned from the US Superfund Act include the high cost of remediation of contaminated sites, and scientific management such as controlling the spread of existing pollution is more effective than site remediation in many cases.

2.2 The development of brownfield management of UK

In the UK, the Interdepartmental Commission for Redevelopment of Polluted Sites (ICRCL) was the first to address the problem of contaminated sites. It is responsible for providing advice and guidance on health hazards caused by the reuse of contaminated sites and coordinating recommendations on remediation measures. The Committee issued Guidelines 59/83 in 1987 (second edition, July 1987) to guide practitioners to deal with different types of hazards and pollution. In 1990, the UK enacted the Environmental Protection Law, which first legislated to regulate contaminated land. In 1998, the National Land Use Database (NLUD) was established and began to identify and address the management of brownfield sites. In the database land use is divided into 51 categories and begins to evaluate the suitability of redevelopment of brownfield sites and other sites. In 2000, the Environmental Protection Agency asked local governments to confirm the treatment of contaminated land. The guidelines define ‘trigger values’ (thresholds and action values) of land for different planning purposes. These triggers were officially cancelled by the Department of Environment, Food and Rural Affairs (DEFRA) in 2002 and in 2005, the sustainable development strategy has been highly valued in the planning and development of land in the UK. The UK government believes that brownfield governance and redevelopment is key to promoting economic growth and maintaining social development, while minimizing environmental impact (Figure SII).

In UK brownfield governance, the government has played a leading role, and the brownfield risk management and restoration policy promoted by it has achieved good results. From 1988 to 1993, 19% of brownfield sites in the UK were converted into green-field sites. Brownfield treatment has improved the quality of urban environments and reduced the pressure on rural land development. The NLUD database shows that about 28,810 hectares (45%) of brownfield land may be suitable for residential use, so the UK's brownfield management has reuse as a starting point, to use market drivers to realize its economic benefits. In 1998, the government policy was that 60% of new homes to be built in 2008 or the renovation of existing residences needed to be carried out on brown fields. This goal was achieved ahead of schedule in 2002, and by 2008 this indicator reached 80%.

Combining the experience of the US and the UK, successful brownfield governance has had the following factors: 1. Established legal and regulatory guarantee systems; 2. Pay attention to public participation in the whole process of brownfield governance and remediation redevelopment and linking contaminated land re-use and remediation to the planning process; 3. Established a brownfield register, regularly publish brownfield information, and mobilize the enthusiasm of all stakeholders; 4. Built a funds guarantee system, including financial allocation (national government provides special fund for brownfield redevelopment), tax relief (making full use of market mechanisms, reducing the cost of redevelopment of brownfield sites by private enterprises and encouraging private investment to enter the field of brownfield redevelopment); 5. "polluter pays" system (Units and individuals that cause damage to land and environment are required to assume corresponding responsibilities for pollution control).

3. The process of contaminated urban soil management in China

3.1 Legal system for brownfield governance and redevelopment

Since the mention of soil control in the Environmental Protection Law of 1989, China has issued about 36 national-level documents related to soil pollution control (Li et al., 2015), such as laws and regulations and technical guidelines (see Table SII), of which 17 are related to urban brownfield reuse.

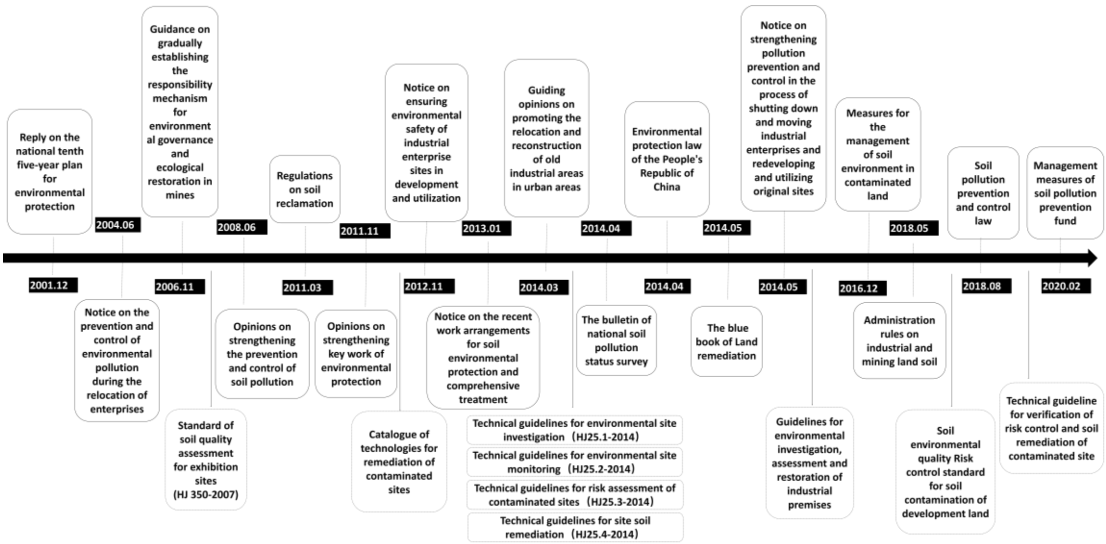

Figure 1. Timeline of contaminated land regulations in China

In June 2004, the State Environmental Protection Administration issued the “Notice on Effectively Preventing and Controlling Environmental Pollution in the Process of Enterprise Relocation”, which first raised the issue of soil pollution for soil redevelopment. Figure 1 details some key steps that followed. By December 2016, the then Ministry of Environmental

Protection issued the “Measures for the Management of the Soil Environment in Contaminated Land”, which stipulates the soil environmental investigation and risk assessment system, the risk management and control system of contaminated land, and the contaminated land governance and restoration system. In general, China's brownfield governance policy development can be divided into three stages: Problem outbreak period (2004-2008); Policy exploration period (2009-2014); Policy establishment development period (2015-present). This period is coming to an end; the next steps will see enactment of the policies to try and solve urban contamination problems. This needs strong policies and laws, together with good knowledge and practical actions at the national, regional and local scale.

In China, the policy and regulatory framework for brownfield management needs to consider China's national conditions, such as:

1. large differences in economic and social development levels between different regions;
2. availability of supporting infrastructure, such as landfills and transport, storage and disposal facilities;
3. the level of competence, knowledge and technical skills;
4. length of pollution history, extent and nature of site pollution;
5. the consequences of exposure risks.

Therefore, it seems more prudent to choose regional and phased approaches based on the national guidelines to establish a framework for contaminated site management.

In the most recent five years, China entered the ‘Policy establishment development period’ of soil contamination management. Many key regulations and laws were issued in 2018-20, with establishment of laws and regulations on “Industrial and Mining Soil Management Methods”, “Law on the Prevention and Control of Soil Pollution”, “Land Use Survey Manuals for Key Industries”, “Certification Methods for Construction Land Responsible Persons”, “Performance Evaluation Methods of the Central Finance Ecological Environmental Protection Special Fund” and “Management methods for soil pollution control funds”.

Land ownership is a major difference when comparing brownfield management in China with that in the UK and the USA. China’s land ownership is completely state controlled; individuals or businesses only have the right of land use and typically properties are bought or leased for 40~70 years from the government (fState Council, 1990). So, governance has an absolute control right in brownfield management, financing and supervision, which means state ownership makes the responsibility and management pathways potentially easier for China. Nonetheless, China has many of the usual challenges in brownfield management. These include multiple levels of government control (see Figure 2); multiple stakeholders i.e. different Ministries, planning and development offices, expert groups, residents’ groups etc. China is currently undergoing institutional reforms, optimizing and integrating the multiple sectors related to soil, water and marine fields, centralizing them into the Ministry of Ecology and Environment, and establishing corresponding professional departments and technical support units. For example, in 2018, to prioritize environmental management and harmonize the decision-making processes, he former Ministry of Environmental Protection formed a new Ministry of Ecology and Environment by integrating the former Ministry of Land and Resources, the former Water Conservancy Department, the former Marine Bureau, the former Agriculture Department and the former Development and Reform Commission related to environmental management. It exercises the responsibilities of the supervisor of the ecological environment in a unified manner, focusing on strengthening the four major functions of the formulation of the ecological environment system, namely – formulating policies and regulations; monitoring and evaluation, supervision and enforcement, and supervision and accountability.

Since the founding of the People's Republic of China in 1949 and the simultaneous abolition of privatization, there has been no substitute for public ownership of land and other natural resources. Article 74 of the General Principles of Civil Law of China states: ‘The collectively owned land belongs to the village peasant collectively in accordance with the law’. According to the newly revised Land Management Law, China adheres to the socialist public ownership of land, i.e. the collective ownership of land by the people. According to Chinese law, the State Council formulates land use policies on behalf of collective ownership and implements ownership. The State Council authorizes relevant ministries and subordinate (city and provisional) governments to exercise the property rights of natural resources in protected areas. In fact, the central government plays a leading role in implementing, protecting and supervising the property rights arrangement of natural landscape resources. However, this leadership role has not yet been assessed in terms of the effectiveness of land use management. The government has allocated part of its budget for the maintenance, planning and management of brownfield sites. However, according to the current land use management system, all levels of government (i.e. central, provincial, municipal, local) need to explain the types and scale of land use to planners, owners and operators. Therefore, the interpretation of ownership and the implementation of land use policies related to ownership may be a major issue for future governance of brownfield sites.

With legislation, there is a solid basis for the construction of an urban soil environmental management system. That legislation needs to be enforced, and to be workable, fair and just. Both the United States and the United Kingdom have specific laws on soil protection. They provide a legal basis for soil environmental protection and stipulate a management system. They clarify the rights and obligations of the main body of governance and urge local governments and their departments to follow the law. The prescribed steps, methods or procedures are managed. In terms of legislation, countries tend to establish precise procedures, evaluate according to local conditions, implement regulations on urban contaminated sites, and at the same time achieve the goal of improving the effectiveness of urban contaminated sites, by gradually improving scientific and technological standards. Therefore, central government should act as a monitoring body to release the standards of enforcement and supervise the results of enforcement, while the local governments and agencies need to enforce their power by following national policies. For this aspect, China has issued Law on the Prevention and Control of Soil Pollution in August 2018 and carried out in January 2019, which aims to protect and improve the ecological environment, prevent and control soil pollution, protect public health, promote the sustainable use of soil resources, promote the construction of ecological civilization, and promote sustainable economic and social development (National People's Congress, 2018).

Under the framework of national laws and regulations and related standards, local governments need to carry out the research method of natural backgrounds, by combining the spatial heterogeneity of soil types and the actual characteristics of contaminated plots considerations for risk management and control in high background value areas, key points for technical reviews, public participation and information disclosure etc., which will improve the environmental management system of local contaminated land, so that the local government's soil environmental management can be implemented.

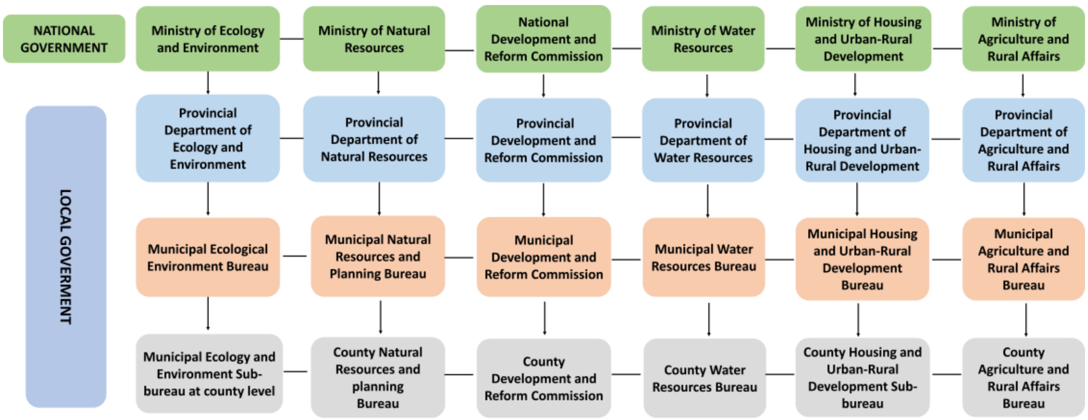

Figure 2. The framework involved in the Chinese soil environmental governance system

3.2 The registration information system for brownfield sites in China

Experience from Western countries indicates that suspected contaminated sites should be investigated and screened, and a professional database of contaminated sites need to be established. The database of brownfield sites needs to hold detailed records of the location, size and nature of potential contaminated sites. Such information needs to be held locally, to inform city planning and development, while information on large, hazardous or priority sites will be needed nationally. It will also help later users of land to understand its basic conditions and avoid an imbalance of information between developers and owners. If an accident occurs during the subsequent use, the relevant data provided by the brownfield database can be used to trace the responsibilities of the parties and provide land governance information and governance process data for future brownfield pollution control. Based on the brownfield database, brownfield sites can be managed hierarchically according to the pollution situation of brownfield sites, and classification of site management and development can be implemented. China has now begun to instigate such a scheme. In 2016, China's Ministry of Ecology and Environment released the Measure for the Management of Soil Environment in Contaminated Land. It provided a procedure for suspected contaminated land, from the definition of suspected contaminated land to supervision. Suspected contaminated land was considered as land that has been engaged in production and operation activities in non-ferrous metal smelting, petroleum processing, chemical, coking, electroplating, tanning and other industries, and has been engaged in hazardous waste storage, utilization, and disposal activities. The Ministry recommended the establishment of a national soil environmental management information system for contaminated sites. It requires local environmental protection authorities at or above the county level to organize the construction and application of information systems on contaminated land within their respective administrative areas. The owner of the suspected contaminated land and the land user is required to fill in and submit online information about the suspected contaminated land and related activities through the contaminated land information system. The MEE then implements information sharing with the urban and rural planning, land and resources departments, through the information system. The list of suspected contaminated sites should be regularly updated. The land use right holder is required to carry out site investigation, risk assessment and remediation evaluation procedures, in accordance with relevant national environmental standards and technical specifications, compile a preliminary survey report, a detailed survey report, a risk assessment report, a risk control plan, a contaminated land remediation plan, a governance and remediation evaluation report of contaminated land. This is uploaded and administered through the contaminated site information system and the main contents of the reports are open to the public. According to the recent released ‘China’s Soil Pollution Prevention and Control Law’ in 2018, the State also implemented a system of risk management and remediation of soil pollution on construction land, different stakeholders have different responsibilities (National People's Congress, 2018).

3.3 Responsibility system for brownfield governance

In the process of constructing an urban soil environmental management system, developed countries have continuously strengthened the unified supervision of central environmental authorities to enhance their authority. At the same time, they emphasize the appropriate decentralization of local environmental management institutions, such as through the rational expansion of state (provincial) local governments, the environmental administrative authority of the environmental protection agencies, or the establishment of branches directly under the central government, and fully mobilize the local expertise in the governance of the city. Countries have stipulated in their legislation the responsibilities and authorities of relevant departments in detail, avoiding the emergence of conflicts of power and mutual promotion. In terms of the competent authorities, the UK and the US authorize the environmental management departments to have strong enforcement powers, to ensure that the polluters fulfil their obligations. They pay attention to the division of responsibilities between the central and local governments and give full play to the initiative of local governments. The US has given the EPA powerful law enforcement powers, imposing heavy penalties on polluters, greatly improving the environmental protection awareness of enterprises; the UK has given local environmental protection and health departments more comprehensive powers, including planning, investigation, administrative enforcement, and will include all sectors related to the environment into the EPA, ensuring a high degree of unity. The strength of law enforcement and the efficiency of execution have been improved. However, in the process of environmental law enforcement, the decentralized functions of environmental protection departments in China lead to the decentralization of law enforcement power. China can draw lessons from UK and US.

A national expert advisory committee was recently established, which can give technical and specialist advice to the various stakeholders involved in brownfield assessment and redevelopment. The system requires a national set of soil quality standards and an accepted risk assessment scheme to be followed for urban brownfield re-development. Some cities may choose to modify these values for local purposes, but the national standards will have a strong theoretical and scientific basis, with the goal of protecting human and ecological health. The National Expert Committee can carry advice and instruct on governance issues, site investigations to relevant management departments, provide program support for risk assessment, reconstruction and post-reconstruction management, and propose key research areas and tasks. The expert committee should be composed of experts from various research fields and stakeholder groups. At the same time, an ‘Environmental Pollution Reconstruction’ or brownfield management Supervision Committee would work with the local environmental protection department, to supervise and evaluate the risk assessment and remediation work at specific sites. The committee would exercise the rights conferred by the state, directly manage each member and supervise the relevant subordinate units and form an effective program cycle chain (program establishment-program evaluation-implementation supervision-effect feedback).

On December 18, 2019, the MEE set up an Expert Advisory Committee on Soil Ecology and Environmental Protection covering more than 60 people with different specialties in soil, groundwater, and agriculture and rural affairs. This group of experts will serve as a think tank role in advancing ecological environmental protection in the fields of soil, agriculture and rural areas, and groundwater.

3.4 Brownfield governance fund and responsibility system

Although the issue of division of responsibility in the case of site contamination is not an easy task, most international regulations and policy frameworks adhere to the “polluter pays principle”. In the US, the Superfund Act seeks to recover remediation costs from potential responsible parties. Although its implementation has caused controversy and proved to be time-consuming and costly, most stakeholders have recognized that the ‘polluter pays principle’ has effectively changed the environmental behaviour of enterprises, making them pay more attention to corporate environmental responsibility. Experience in managing the US Superfund process has shown:

1). It is necessary to seek methods for determining the responsible party for pollution of sites with multiple discharges, such as landfills and responsible persons for dumping sites;

2). Effective methods must be sought to reduce the legal and administrative costs incurred by governments and small businesses for the risk of site responsibility;

3). Management and law enforcement agencies need to consider the limited effectiveness of tracking those responsible for inability to cover remediation costs;

4). Site remediation is extremely expensive and must ensure a sustainable funding mechanism.

In the UK, the opposite is true, with the private sector promoting and funding most land development and rehabilitation projects. In some countries, the responsibility for contaminated sites is determined on a clear scale. The level begins with the polluter; if the polluter does not pay the remediation cost, the responsibility is transferred to the owner of the land; the responsibility is transferred to the government only if the landowner does not pay the remediation fee. In addition, there are special mechanisms for dealing with uninformed landowners’ responsibilities.

Under the China’s policy framework, the basic principles of ‘who pollutes and who is responsible’ also have been clarified (Measures for Environmental Management in Contaminated land, 2016) (Ministry of Ecology and Environment of People's Republic of China, 2016) as follows:

1. In accordance with the principle of "who pollutes, who governs", the unit or individual that causes soil pollution shall bear the main responsibility for the control and restoration.

2. If the responsible subject changes, the unit or individual who inherits its creditor's rights or debts after the change shall bear relevant responsibilities.

3. If the responsible subject is lost or the responsible subject is not clear, the local people's government at the county level shall bear relevant responsibilities according to law.

4. Where the land use right is transferred in accordance with the law, the land use right transferee or the responsible person agreed upon by both parties shall bear the relevant responsibilities.

5. If the land use right is terminated, the original land use right holder shall bear relevant responsibilities for the soil pollution caused during the use of the land.

6. The lifelong responsibility system shall be implemented in the treatment and remediation of soil pollution.

However it is still necessary to further clarify the responsibilities of the various departments and comprehensively regulate the brownfield governance process. Measures for the Administration of Special Funds for the Prevention and Control of Soil Pollution (Ministry of Finance of the People's Republic of China and Ministry of Ecology and Environment of People's Republic of China, 2019) released by Ministry of Finance support the scopes including: (1) Detailed investigation, monitoring and evaluation of soil pollution; (2) Investigation and risk assessment of construction land and agricultural land; (3) prevention and control of soil pollution sources; (4) Management and control of soil pollution risks; (5) Remediation and treatment of soil pollution; (6) Support the establishment of provincial soil pollution prevention funds; (7) Enhancement of soil environmental supervision capabilities and other content closely related to improvement of soil environmental quality. Measures for the Evaluation of the Performance of the Central Government's Ecological and Environmental Protection Special Funds (Consultation Draft). And the Ministry of Finance reviews and determines the amount of funding arrangements of the relevant provinces, autonomous regions, and municipalities (hereinafter referred to as the provinces) in accordance with the allocation proposals made by the Ministry of Ecology and Environment (Ministry of Finance of the People's Republic of China and Ministry of Ecology and Environment of People's Republic of China, 2019). According to ‘Management measures of soil pollution prevention fund’ newly released by Ministry of Ecology and Environment in 2020 (Ministry of Finance, 2020). China has set up soil pollution prevention fund in provincial level. It refers to the establishment by the provincial, autonomous region, municipality directly under the Central Government, and single-planned cities (hereinafter referred to as the province) of the budget through separate budget or co-funding with social capital, and adopting marketization methods such as equity investment to exert guidance and leverage effects to guide society various types of capital investment in the prevention and control of soil pollution, government investment funds to support the development of the soil remediation industry.

3.5 Management of contaminated site remediation and the context of strategic city planning

Early national policies emphasized multi-functional restoration (permanent contaminant removal) (such as in the US and the Netherlands). In most developed countries today, the overall trend of remediation tends to use ‘applicability’ as a target for remediation (i.e. the re-used land needs to be ‘fit for purpose’ rather than returned to a pristine condition. In other words, the required level of remediation/soil quality targets depend on the intended land use. They are generally classified into agricultural, residential, and industrial/commercial uses. Site risk assessment and remediation objectives therefore usually need to consider the current or future land use.

For China, most of the brownfield industrial sites attracting attention are in cities, and some are even in major real estate development areas. After redevelopment and to gain the greatest land price, these sites can be used for residential or commercial purposes. Therefore, returning contaminated sites to their original uncontaminated state appears to be a conservative and attractive option. However, many sites can have a pollution history of half a century or more, and given the time constraints of redevelopment, the time available for remediation is very limited. Expensive remediation costs and development time constraints can make it unrealistic to remediate contaminated sites to a standard that can be used for any purpose. In addition, technologies that can effectively achieve rigorous remediation goals may not be available. Considering other potential land uses, such as industrial park sites, park green belts, or golf courses, may be a pragmatic and more economical options. The re-uses of the brownfield sites were called as soft re-use of brownfields that are not based on built constructions or infrastructure in the study of National People's Congress (2018), and they also suggested a

“Brownfield Opportunity Matrix” (Coulon et al., 2016a) to understand the sustainability of the services and provide a structure for the overall valuation of restoration work.

The Chinese government have released a series of regional regulations about soil remediation (Li et al., 2015). The Ministry of Ecology and Environment has officially issued the Technical guidelines for risk assessment of soil contamination of land for construction (HJ 25.3-2019), but in fact the soil environmental quality standards (GB15618-1995) and the soil environmental quality evaluation standards for exhibition land (HJ350-2007) have not been revised or abolished, resulting in two problems. The two evaluation methods are parallel, and the target values determined by the two different methods are obviously different, which brings difficulties to the remediation work. In 2018, China released its latest soil standards: soil screening values and intervention values (Ministry of Environmental Protection of the People's Republic of China, 2018), however, there is no a clear remediation value released in China so far. The suitability, costs and time for various remediation technologies also needs a system for independent testing, advice and verification. In recent years, a number of physical, chemical and biological treatment methods have been applied at brownfield sites and there are many claims for patents and commercially valued technologies. It is critical that these are scientifically evaluated, so that credible and defensible decisions can be made over remediation targets and costs. Without this, the whole environmental engineering and remediation sector may ultimately be undermined.

On October 29, 2019, the National Environmental Benchmark Expert Committee was established in Beijing and governed by Ministry of Ecology and Environment of People’s

Republic of China (Former Ministry of Environmental Protection) (Ministry of Ecology and Environment of People's Republic of China, 2019). The benchmark expert committee needs to establish a national expert team to focus on the restoration work, and provide professional consultation, advice and technical support for the restoration work of different brownfield types.

4. Suggestions for future Chinese urban brownfield management and development

Although China is tackling actively environmental pollution and especially soil pollution, there is still considerable scope for strengthening the implementation of environmental policies and redeveloping brownfield sites (Li et al., 2018). There is a need for enhanced coordination of governance, implementation of a risk-based approach, and funding mechanisms. Developing a coherent and integrated framework for brownfield management and redevelopment (Figure 3) is an urgent and long-term strategic task for China that will contribute to a resource-saving and environment-friendly society. The timing is seen as optimal as China is in an unprecedented stage of urbanization and industrialization. China has already invested and/or committed significant resources and efforts in implementing a brownfield redevelopment with the objective of promoting eco-industrial and eco-friendly development. By promoting the coexistence of a healthy economy and environmental health, China attempts to integrate environmental management so as to meet environmental, economic and community development goals of modern cities.

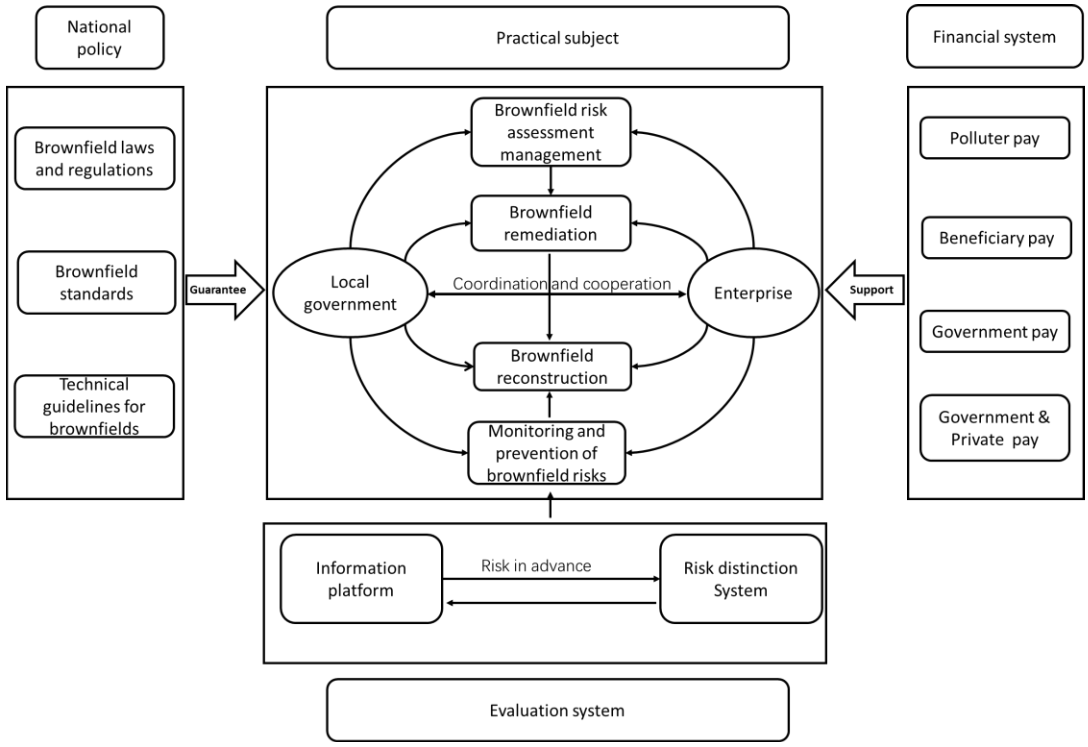

Figure 3. Proposed Chinese urban brownfield management framework (expanded from Chen and Xu (2017).

While the UK and US have legislated for the re-development of brownfield land and have the redevelopment of brownfield land as a systematic regional or national project, the specific situation of each brownfield site is different (e.g. the soil type, the contaminant mixture and level, the site planned use etc). The general model therefore is that the central government should guide the management and redevelopment of brownfield sites from a macro perspective, while the specific practical work should be promoted and managed by local governments and relevant stakeholders, according to local conditions and priorities (Figure SIII-SIV). These experiences have been learned as references by China. China has a stronger platform for control, informed decision-making and management than western models. China can use its strong central and provincial planning capabilities and the single land public ownership system, which provides a huge opportunity for the overall planning of land use and site rehabilitation in the future. For example, the government can ensure the preferential supply of brownfield to the land market, by means of a land reserve system and preferential auction. It could also instigate schemes to use the land for defined purposes and benefits, such as providing renewable energy, or development of soil-free three-dimensional agriculture. The government is responsible for providing land use rights and plays a guiding role. It can also enforce strong monitoring and policy implementation, encourage sound technology, monitoring and site management and regulate/authorize experts and professional bodies.

Acknowledgements

We would like to thank the Dr. Aidan Keith from Centre for Ecology and Hydrology for UK brownfield technical information support.

Supplemental information for

TITLE: Re-development of urban brownfield sites in China: motivation, history, policies and improved management

Table SI. The 10-point plan in China released in 2016

Plan	Content	Remarks relating to urban Brownfield
1	Conduct soil pollution survey and master soil environmental quality	Deeply developing soil environmental quality survey; construction of monitoring network for soil environmental quality; Improving the information management level of soil environment.
2	Promoting legislation on prevention and control of soil pollution and establishing and perfecting the system of laws and regulations and standards	Speed up the legislative process; Systematic construction of standard system; Enhancing supervision and law enforcement in an all-round way.
3	Implementing classified management of agricultural land to guarantee the environmental safety of agricultural production	Classification of soil environmental quality of agricultural land; Enhancing protection effectively; Efforts to promote safe utilization; Fully implementing strict control; Strengthening soil environmental management of forest and grassland gardens.
4	Implementing access management of construction land to prevent habitat environmental risks	Clear management requirements; Implementing regulatory responsibility; Strict access to land.
5	Strengthen the protection of non-polluted soil and strictly control new soil pollution	Strengthening environmental management of unused land; Preventing new pollution of construction land; Strengthen the control of spatial layout.

6	Strengthen the supervision of pollution sources and preventing soil pollution	Strict control of industrial and mining pollution; Controlling agricultural pollution; Reducing domestic pollution.
7	Conduct pollution control and remediation to improve regional soil environmental quality	Define the subject of governance and restoration; Establishment of governance and rehabilitation planning; To carry out management and restoration in an orderly manner; Supervise the implementation of objectives and tasks.
8	Strengthen research and development of science and technology to promote the development of environmental protection industry	Strengthening the study of soil pollution prevention and control; Strengthen the popularization of applied technology; Promoting the development of governance and restoration industries.
9	Bringing the government's leading role into full play and constructing a soil environmental management system	Strengthening government leadership; Play a market role; strengthen social supervision; carry out propaganda and education.
10	Strengthen objective assessment and strictly investigate responsibility	Define the main responsibility of local government; strengthening department coordination and linkage; implementing corporate responsibility; Strict assessment and assessment.

Table SII. Management documents related to contaminated sites issued by the state

Date	Release agency	Laws/Regulations
2001.12	State Council	Reply on the National Environmental Protection Tenth Five-Year Plan
2004.6	State Environmental Protection Administration	Notice on Effectively Preventing and Controlling Environmental Pollution in the Process of Enterprise Relocation

2004.8	The eleventh meeting of the Standing Committee of the Tenth National People's Congress	Land Management Law of the People's Republic of China
2006.2	State Environmental Protection Administration in conjunction with the Ministry of Finance and the Ministry of Land and Resources	Guiding Opinions on the Progressive Establishment of Responsibility Mechanisms for Mine Environmental Governance and Ecological Restoration
2006.11	State Environmental Protection Administration	Notice on Issuing the “Eleventh Five-Year Plan” National Science and Technology Support Plan
2008.6	Ministry of Environmental Protection	Opinions on Strengthening Soil Pollution Control
2009.8	Office of the State Council	Guidance on strengthening prevention and control of heavy metal pollution
2011	Ministry of Land and Resources	Rules for the Preparation of Land Reclamation Plans
2011.2	Office of the State Council	Twelfth Five-Year Plan" for Comprehensive Prevention and Control of Heavy Metal Pollution
2011.03	State Council	Soil reclamation regulations
2011.10	State Council	Opinions of the State Council on Strengthening Key Tasks of Environmental Protection
2012.11	Ministry of Environmental Protection and other four ministries	Notice on Ensuring the Environmental Safety of Industrial Enterprise Sites in Development and Utilization
2013.01	State Council	Notice on Printing and Distributing the Work Arrangements for Soil Environmental Protection and Comprehensive Management in the Near Future

2014	Ministry of Land and Resources	Regulations on Acceptance of Land Improvement Projects
2014.3	Office of the State Council	Guiding Opinions on Promoting the Relocation and Transformation of Old Industrial Zones in Urban Areas
2014.4	Ministry of Environmental Protection, Ministry of Land and Resources	National Survey Bulletin on Soil Pollution
2014.4	the eighth meeting of the Standing Committee of the Twelfth National People's Congress	Environmental Protection Law of the People's Republic of China
2014.5	Land Remediation Center of the Ministry of Land and Resources	Blue Book on Land Remediation
2014.5	Ministry of Environmental Protection	Notice on Strengthening the Prevention and Control of Pollution in the Process of Shutdown, Relocation and Redevelopment of Industrial Sites
2014.12	Ministry of Agriculture	National Announcement of Quality of Cultivated Land
2014.12	Ministry of Land and Resources	Announcement on the Results of Major Data Surveys and Assessments of National Cultivated Land Quality
2016.5	State Council	Action Plan for Soil Pollution Prevention
2016.12	Ministry of Environmental Protection	Environmental Management Measures for Contaminated Soil (Trial)
2017.9	Ministry of Environmental Protection, Ministry of Agriculture	Administrative Measures for Agricultural Land Soil Environment (Trial)
2018.5	Ministry of Environmental Protection	Measures for the Management of Soil Environment of Industrial and Mining Lands (Trial)
2018.8	Standing Committee of the National People's Congress	Soil Pollution Control Law

2019	Ministry of Ecology and Environment of People's Republic of China	National Environmental Benchmark Expert Committee established
2019	Ministry of Finance of the People's Republic of China and Ministry of Ecology and Environment of People's Republic of China	Measures for the Administration of Special Funds for the Prevention and Control of Soil Pollution
2020.	Ministry of Finance, Ministry of Ecology and Environment, Ministry of Agriculture and Rural Affairs, Ministry of Natural Resources, Ministry of Housing and Urban-Rural Development, State Forestry and Grassland Bureau	Management measures of soil pollution prevention fund
Date	**Release agency**	**Standards of Soil Quality**
1995.7	Ministry of Environmental Protection	Soil environmental quality standard (GB15618-1995)
2006.11	Ministry of Environmental Protection	Environmental Quality Evaluation Standards for the Origin of Edible Agricultural Products (HJ332-2006)
2007.6	Ministry of Environmental Protection	Environmental Quality Evaluation Standards for Greenhouse Vegetable Producing Areas (HJ333-2006)
2007.6	State Environmental Protection Administration, General Administration of Quality Inspection and Quarantine	Evaluation Standards of Soil Environmental Quality for Exhibition Land (Interim) (HJ 350—2007)
2013	Ministry of Land and Resources	Land reclamation quality control standards (TD/T1036-2013)
2015.8	Ministry of Environmental Protection	Technical specifications for soil environmental quality assessment (draft for comments)
2018.6	Ministry of Environmental Protection	Soil Environmental Quality Agricultural Land Soil Pollution Risk Control Standard (GB15618-2018）

2018.6	Ministry of Environmental Protection	Soil Environmental Quality, Soil Pollution Risk Control Standards for Construction Land (GB36600-2018)
2019	Ministry of Ecology and Environment	Technical guidelines for risk assessment of soil contamination of land for construction (HJ 25.3-2019)
Date	**Release agency**	**Soil Remediation Technology Management**
2014.2	Ministry of Environmental Protection	Technical Guidelines for Site Environmental Investigation (HJ25.1-2014)
2014.2	Ministry of Environmental Protection	Technical Guidelines for Site Environmental Monitoring (HJ25.2-2014)
2014.2	Ministry of Environmental Protection	Technical Guidelines for Risk Assessment of Contaminated Sites (HJ25.3-2014)
2014.2	Ministry of Environmental Protection	Guidelines for Soil Remediation Technology of Contaminated Sites (HJ25.4-2014)
2014.2	Ministry of Environmental Protection	Terms of Contaminated Sites (HJ682-2014)
2014.10	Ministry of Environmental Protection	Contaminated Site Remediation Technology Catalog (First Batch)
2014.10	Ministry of Environmental Protection	Guidelines for Project Management of Contaminated Soil Remediation in Agricultural Land (Trial)
2014.10	Ministry of Environmental Protection	Guide to Extraction of Plants from Contaminated Soil of Agricultural Land (Trial)
2014.11	Ministry of Environmental Protection	Guidelines for Environmental Investigation, Assessment and Restoration of Industrial Enterprise Sites (Trial)
2015.08	Ministry of Environmental Protection	Guidance on Screening Guidance of Soil Pollution Risk for Construction Land

2018.12	Ministry of Environmental Protection	Technical Guidelines for Risk Control of Contaminated Land and Evaluation of Soil Remediation Effect

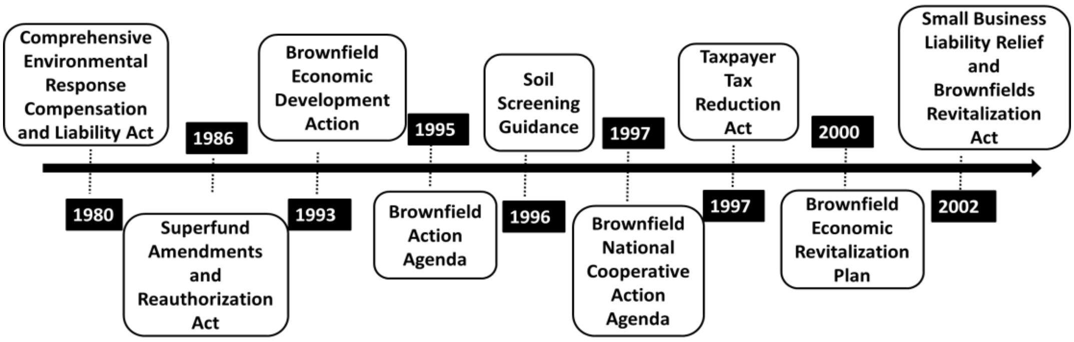

Figure SI. Timetable of brownfield regulations in US

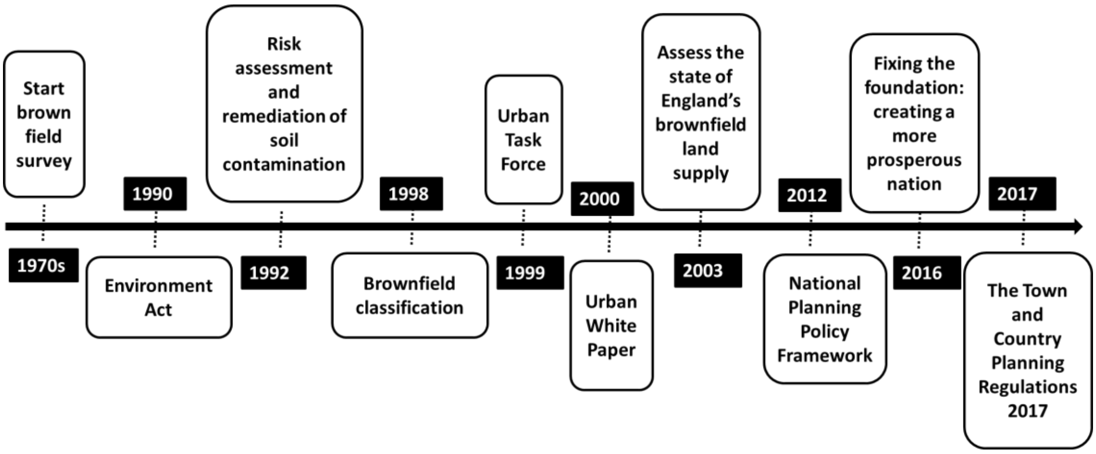

Figure SII. Timetable of brownfield regulations in the UK

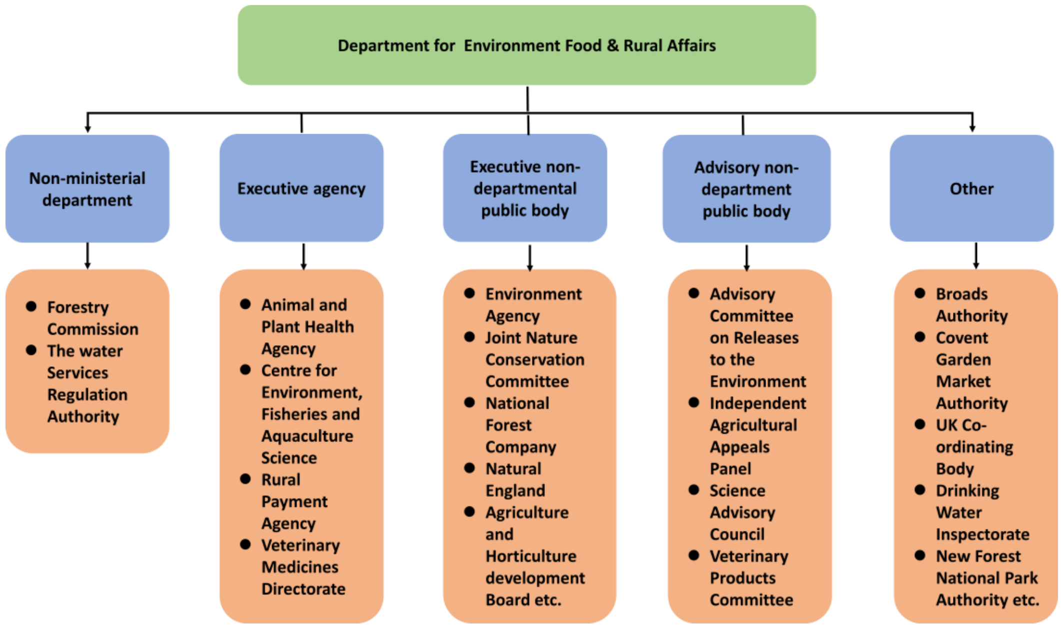

Figure SIII. The UK soil environmental governance framework

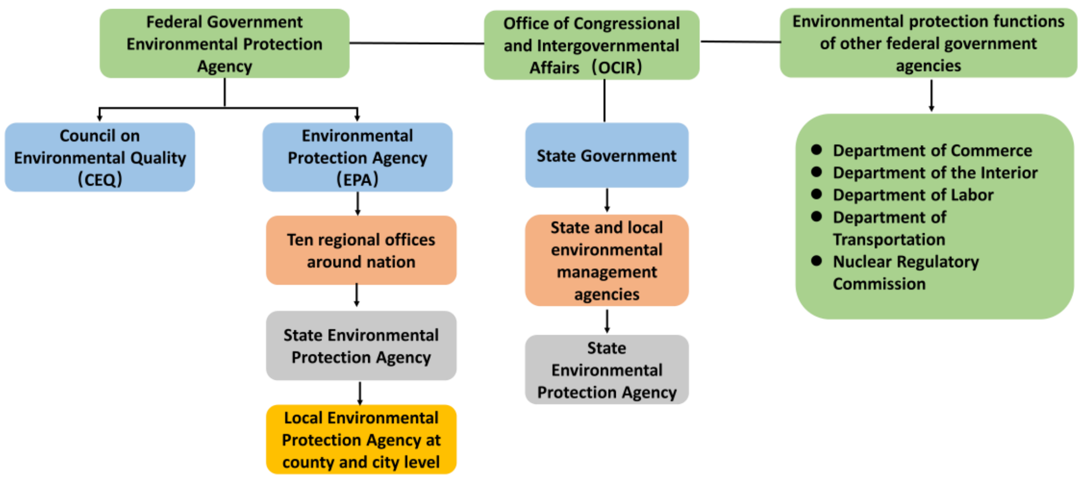

Figure SIV. The US soil environmental governance framework

7. Concluding remarks and recommendations

7.1 General comments

With the rapid development of China's economy and urbanization and the intensification of agriculture in the past decades, the amount of contaminated land has been increasing. Soil pollution has therefore become an urgent problem for sustainable soil management in China. The National Polluted Soil Investigation Bulletin released in 2014 drew attention to the proportion of China’s soils that may exceed soil quality standards. China’s ‘10-Point Soil Plan’ released by the State Council in 2016 sets out strategic priorities for China’s research and management of the soils resource. This research thesis can make a timely contribution by helping to summarize the situation of soil pollution in China, providing new data on soil change and developing tools, data, expertise and methods.

7.2 Specific research findings and conclusions

Paper I generally reviewed the situation, survey methodology, soil environmental standard assessment of soil contamination in China. Priorities for China’s next steps is to develop an effective research and management regime. **(1)** Critically important to the science-based risk assessment of contaminants in soils is the incorporation of speciation and bioavailability into the measurement and evaluation criteria. On initial screening soils and sites may be deemed ‘contaminated’, but after a second tier analysis they may be shown to be suitable for crop production and use. **(2)** Selection of appropriate and validated measurement and evaluation tools is a priority. If this is done in a scientifically transparent and defensible way, China will have a robust and internationally leading system for soil management in place. **(3)** It is necessary to consider the soil biology/ecological endpoints for protecting ecosystem health. **(4)** National and regional/local scenarios of land use type/usage will address residential/urban re-use of industrial land as well as varying agricultural scenarios.

Paper II compared the differences in toxicity assessment and risk characterization, exposure assessment and parameter types in the methodologies to obtain SSVs in China and the UK, and combined the CLEA model with Chinese parameterisation. These comparisons highlight that **(1)** the difference in toxicity assessment and risk characterization methods of carcinogens results in the biggest difference in SSVs between the two countries. The differences in SSVs also varied in carcinogenic and volatile properties of substances. **(2)** The difference in SSVs for carcinogenic substances is related to the route of exposure, but for non-carcinogenic substances, the difference of SSVs calculation method and SSVs is small. For volatile organic compounds, the presence of indoor respiratory exposure pathways greatly reduces the differences caused by toxicity assessment and risk characterization methods. For non-volatile substances such as heavy metals, the effects of toxicity assessment and risk characterization methods are significant. **(3)** The SSV of As obtained by the CLEA model with Chinese parameters is closer to the background value of soil in China. In the management of non-volatile contaminated sites such as heavy metals in China, the CLEA model can be used for risk assessment and calculation of site specific SSVs.

Appendix 3 found **(1)** the total human environmental exposure levels varied between five metals (Cr > As > Pb > Cd > Hg), between gender (females > males) and between ages (less than 60 > equal or more than 60). **(2)** In all exposure media, the diet contributed the most to the total environmental exposure levels of Hg, Cd, As, Pb and Cr. In terms of exposure routes, the exposure contributions of Hg, Cd, As, Pb and Cr via ingestion were highest. **(3)** The contributions of exposure media and routes were different among age groups. The exposure contributions of residents aged 60 years old and above through non-dietary medium and ingestion route were higher than others. **(4)** Non-dietary and dietary exposure had different contributions to non-carcinogenic risks and carcinogenic risks.

Paper III found **(1)** in the southern part of China the mean pH of paddy soils fell sharply (from pH 5.81 to 5.19) over the two decades between surveys (1985-90 and 2006-10), while dry farmlands in the northern sampling area fell slightly (from pH 8.15 to 7.82). The mean SOM content of dry farmland soil rose in both areas (from 1.35 to 1.81 in northern part of China; from 1.23 to 2.59 in southern part of China; $p<0.001$) and the mean SOM of paddy fields in the southern area also rose (from 1.63 to 2.67; $p<0.001$). **(2)** Woodland soil pH in the south showed an increase from 4.71 to 5.29 but no significant difference was measured in the woodlands of the northern area, although the trend increased. The SOM content of woodland top soils rose in the northern and southern study areas. **(3)** The implications and potential causes of these changes over the two-decade timespan between surveys need to be considered for soil nutrients/potential toxins intake from soil to food production and how large scale soil sampling campaigns can be designed to monitor for changes and potential controlling factors.

Paper IV generally reviewed the history, innovation and management of brownfield sites in China, the UK and US. Guidance is provided on how to improve the legal system and establish a monitoring body, recognize the responsibilities of key stakeholders, improve the brownfield database or brownfield survey system for China and establish a remediation system with technology support to aid management of China’s brownfield sites.

7.3 General conclusions

The review of soil heavy metal pollution highlighted, the situation of arable land and industrial abandoned land as severe issues for China to address (**research objective 1**). China will need to produce food for human consumption on soils which are already deemed “contaminated”. Therefore, scientifically based risk assessment is necessary to inform practical decisions about land use options and management decisions.

The comparison analysis of SSVs derived using methodologies used in China and the UK highlights the need and opportunity for China to develop scientifically sound tools for risk assessment. The differences in SSVs between China and the UK are mainly reflected in the aspects of toxicity assessment and risk characterization methods, exposure assessment and differences in parameters (**research objective 2**). The key to reducing the difference of SSVs between China and the UK is to increase consistency in toxicity assessment, risk characterization and exposure assessment models. China should implement toxicity assessment and risk characterization studies based on its own conditions.

Human exposure and health risk assessment data were obtained in a major study co-ordinated by the Chinese Research Academy of Environmental Sciences (Ministry of Ecology and Environment of the People’s Republic of China) and made available for inclusion in this thesis. This study reinforces that the diet is the main source of human exposure to heavy metals for the general population. Soil-borne metals can supply the diet via plant-based foodstuffs and this can be estimated from the study’s database (**research objective 3**).

Results from the analysis of soil pH and organic matter showed they varied in different land uses and two time periods (**research objective 4**). Interactions between the composition of land use and environmental conditions play a key role in determining the trajectory of soil quality at large spatial scales. They have significant implications for carbon storage, nutrient cycling and crop productivity, and need to be understood to optimise land management in different environmental contexts and avoid degradation of China's soil resources.

Overall, the findings in this study suggested that the current situation of soil pollution management is at the stage of "rapid development" with a lack of an effective management scheme. By comparing the background, context, regulations, policies and management procedures of brownfield sites among China and UK and US (**research objective 5**), areas for improvements were suggested. These included establishing a brownfield database, improving the legal system, recognizing the responsibilities of each government department, clarifying the shared responsibilities of stakeholders and making full use of advantage of state ownership.

7.4 Recommendations for future work

Future work is suggested to further understand the soil impacts and management of soil pollution, with considerations of food safety, human health and soil sustainability development in China.

Soil quality standards: Soil EQSs are now defined for 9 pollutants in agricultural land in China (GB15618-2018). However, they are still needed for other substances. Only 3 organics are currently listed: hexachlorocyclohexane, dichlorodiphenyltrichloroethane (DDT) and benzo(a)pyrene. As the production and use of hexachlorocyclohexane and DDT ceased in 1983, the effects of these two pesticides in the soil will gradually decrease over time. In China's agricultural production, due to the heavy use of chemical fertilizers, pesticides and sewage irrigation, organic pollutants such as polychlorinated biphenyls (PCBs), polycyclic aromatic hydrocarbons (PAHs), and pesticides in the soil may be added in future.

Exposure and risk assessment: Based on the experiences of risk assessment in other developed countries, the methods of exposure and risk assessment are well established and easily adapted to the Chinese situation. China can use the UK method (CLEA model) to strengthen its toxicity assessment and risk characterization methods for carcinogenic substances, to reduce the uncertainty in the risk assessment of contaminated sites and improve the specific management of contaminated sites. Although China can learn from other countries, the approach needs to better formulate the suitable toxicity assessment, exposure assessment and risk characterization for China’s situation.

Surveillance and monitoring of soils: Once standards are established, soils need to be surveyed and monitored for contaminants. Soil surveys have been carried out in China previously, but they can be improved by collecting from the same points, to improve the consistency and comparability of samples between sampling years. With careful design, it will be possible to better monitor underlying changes in soil properties and contaminant levels. China has large databases on soil properties and contaminants already, but with little thorough interpretation of the data so far. In the future, more information and interpretation can be ‘mined’ from existing samples and databases, as shown in **Paper III**.

Assessment and use of bioavailability: The focus in China so far has been on soil standards and risk assessment, based on 'total concentrations'. The current soil EQSs use the total amount as a standard. Therefore, the current soil EQSs are applicable to soils with high availability of heavy metals, while soils with low availability of heavy metals cannot reflect their true situation. It is likely that the availability of metals in soils will be very different in the two soils regions discussed in Paper III, for example. This will over-estimate the problems heavy metals pose in China's soils. Other countries have begun to use measures of 'bioavailable' contaminants and China can move in this direction.

Impacts of changing soil properties: Paper III reported underlying changes in the basic properties of soils – their pH and SOM. Changes in these properties can have very important effects on the availability of soil nutrients and potential toxins (e.g. Al, Mn, Fe), as shown in Figure 16 and Figure 17. The changes in soil properties reported here are highlighted in the Figure. However, whether this will affect the soil nutrients and contaminants and whether they will transfer more into the crops remains unknown. This is an important topic for further research.

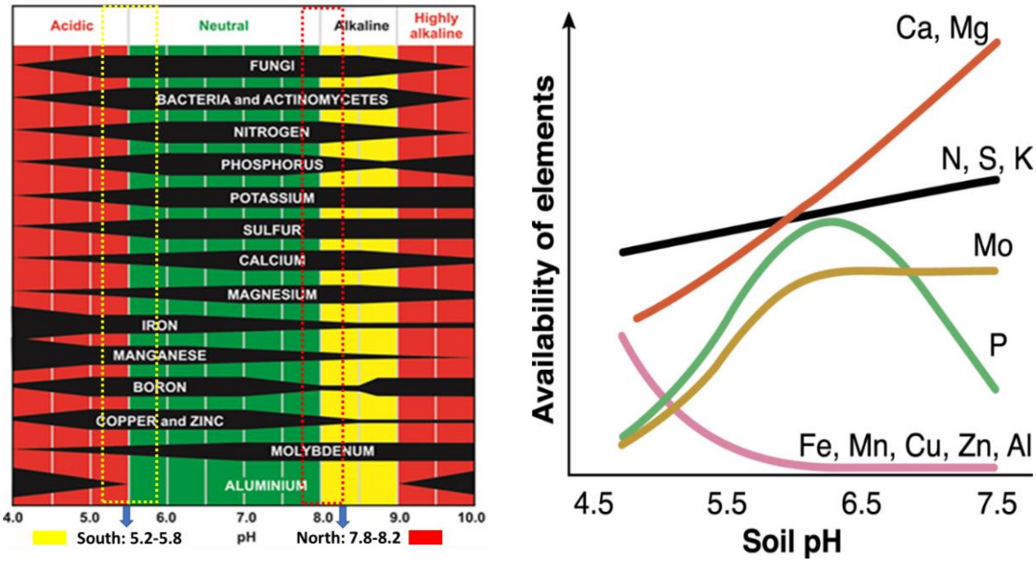

Figure 16. **LEFT**: Summarised representation of the effects of pH on nutrient availability. The width of the bar represents the relative availability of that particular nutrient at that pH level (Source: http://fertsmart.dairyingfortomorrow.com.au/dairy-soils-and-fertiliser-manual/chapter-7-managing-limiting-soil-factors/7-6-soil-ph/#target-7-6-6). Yellow and red dotted box represents the soil pH ranges of agricultural land in South of China and North of China reported in this research (Paper III), respectively. **RIGHT**: The relationship between soil pH and nutrient availability (source: https://www.agric.wa.gov.au/soil-acidity/effects-soil-acidity?page=0%2C1).

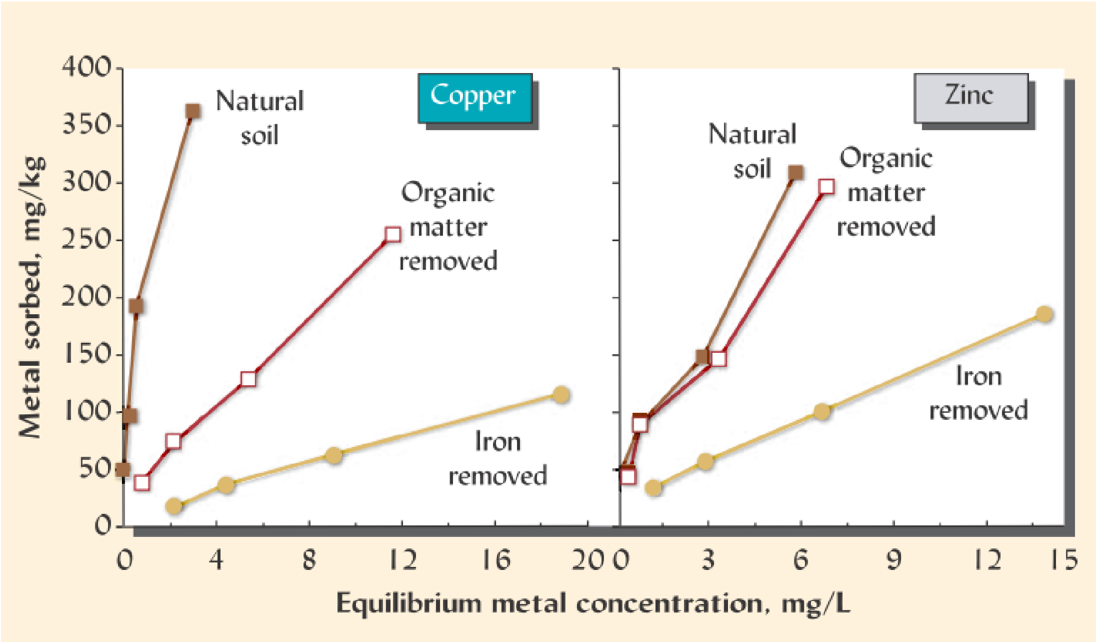

Figure 17. Sorption of Cu and Zn affected by removal of the organic and iron components of the soil. The large role that sorption on soil organic matter (and iron oxides) plays in the solubility of Cu and the much smaller role of organic matter with regard to Zn (Source: The Nature and Properties of Soils, 15^{th} edition by Ray R. Weil and Nyle C. Brady in 2017).

Impacts of contaminant levels on soil processes: most work in China so far has considered the implications of soil contaminants on potential human exposure. However, the impacts on soil processes, soil fertility and soil health are also fundamental to understand. For example, does soil pollution affect the cycling of major nutrients? Is carbon turnover and storage affected by soil pollution?

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Appendix 1

Journal of Environmental Management 251 (2019) 109512

Contents lists available at ScienceDirect

Journal of Environmental Management

journal homepage: www.elsevier.com/locate/jenvman

Research article

Soil contamination in China: Current priorities, defining background levels and standards for heavy metals

Yiming Sun[a], Hong Li[a], Guanlin Guo[b], Kirk T. Semple[a], Kevin C. Jones[a,*]

[a] Lancaster Environment Centre, Lancaster University, Lancaster, LA1 4YQ, UK
[b] State Key Laboratory of Environmental Criteria and Risk Assessment, Chinese Research Academy of Environmental Sciences, Beijing, 100012, PR China

ARTICLE INFO

Keywords:
Soil contamination
Soil sampling
Risk assessment
Land use
China
United Kingdom

ABSTRACT

The Chinese Government is working to establish an effective framework in managing soil contamination. Heavy metal contamination is key to the discussion about soil quality, health and remediation in China. Soil heavy metal contamination in China is briefly reviewed and the concepts of background values and standards discussed. The importance of contaminated land and its management for China food security and urbanization are discussed. Priorities for China's next steps in developing an effective research and management regime are presented. We propose that critically important to the science-based risk assessment of contaminants in soils is the incorporation of speciation and bioavailability into the measurement and evaluation criteria. Consideration of soil biology/ecological endpoints will be necessary to protect ecosystem health. National and regional/local scenarios of land use type/usage will address residential/urban re-use of industrial land as well as varying agricultural scenarios.

1. Introduction

Soil pollution refers to the occurrence of some substances in soil caused by human activities, which can change soil quality and function, lead to soil degradation, damage basic soil structures and has the potential to harm human and environmental health. Soil pollution has been identified as a key national priority in China, with an increase of reports on agricultural land and human health affected by soil pollution (Luo et al., 2015). With economic growth and industrial restructuring in China, soil pollution from abandoned sites in urban areas has also drawn attention and concern regarding the safety of human settlements and human health in industrial and brownfield sites (Cao and Guan, 2007; Luo et al., 2015). According to the National Investigation Bulletin of Soil Pollution Status (NIBSPS) issued by the Ministry of Environmental Protection of the People's Republic of China (MEP-PRC), investment in soil remediation will reach up to RMB 4,633,000 million (£526,000 million). This is a huge financial commitment, so it is critical that sound science and knowledge are applied to the decisions that determine how this money will be spent. There is still work to be done in China to improve information to define soil background conditions and pollution status, the relevant science and policies needed to set soil quality standards, the assessment system for site evaluation and soil remediation strategies and technologies (State Council, 2016). The importance of soil pollution and degradation in China has now been recognized at the highest level, with specific requirements included in China 13th Five-Year National Development Plan and the Fifth Plenum of the 19th Central Committee of the Communist Party of China (MEP-PRC, 2016).

This paper focusses on an assessment of some of the priorities relating to heavy metals in Chinese soils. Soil heavy metal pollution has become a widespread and serious problem globally. Heavy metals are present naturally in soils, but elevated levels may be derived from agricultural activities, urbanization, industrialization and other human activities. To define and resolve pollution problems, it is therefore necessary to be able to define what constitutes 'clean', 'background' and 'contaminated' and 'polluted' soils. Following surveys and analysis of heavy metals in soils, many countries such as the United Kingdom, the United States and the Netherlands have developed such values. Depending on the national environmental management and regulatory processes, different countries have different approaches. Examples include the *Soil Guideline Values (2009)* in the UK, the US *Soil Screening Levels (2002)*, the *Intervention Values (2009)* in the Netherlands, and *Environmental Quality Standards (EQS) (1991 and 1994)* in Japan.

China first developed its own Soil Environmental Quality Standards (SEQS) in 1995 (GB15618-1995) (Xia, 1996). So far, there are 63 current standards related to soil environmental protection in China and the number of standards released by the MEP-PRC has increased, especially in the last 5 years (Li et al., 2016). Following China previous focus on

* Corresponding author.
E-mail address: k.c.jones@lancaster.ac.uk (K.C. Jones).

https://doi.org/10.1016/j.jenvman.2019.109512
Received 14 March 2019; Received in revised form 1 September 2019; Accepted 1 September 2019
Available online 25 September 2019

air and water quality, the Government has now turned a focus onto soils and groundwater, publishing a landmark '10-Measures for Soil Pollution Action Plan' in 2016 (State Council, 2016). Its purpose is to manage, control and prevent soil pollution, to gradually improve soil quality in China. The Plan's first action recommends conducting surveys on soil pollution to better define the status of China's soil resources. GB15618-1995 first defined SEQS values for 8 heavy metals in China to apply to the whole country. Later, relevant soil quality standards in China were developed by referring to GB15618-1995 as a basic standard (Li et al., 2016). For example, the MEP-PRC issued a series of standards, such as ones for 'Green food-technical conditions for environmental areas' (NY/T391-2000), but these are rarely applied in practice. In contrast, European countries conduct soil pollution control mainly through a series of systematic assessment methods. These are based on different land use type, soil specificity or local environmental factors for understanding the risks either to the environment or human health. In China over the past 20 years, a general soil standard value (GB15618-1995) was applied to the whole country, without considering soil specificity and integrated environmental factors. During this period, in order to meet development needs, some regional standards for soil risk assessment were also set; for example, Beijing issued screening levels for soil environmental risk assessment of sites (DB11/T 811) in 2011.

A particular challenge for China is the country size and hence the range of soil types and conditions. Heavy metal concentrations vary naturally in soils, as a function of the geology, climate, land use etc. Hence, the Soil Environment Background Value (SEBV) will vary across the country. SEQS values were originally set nationally, so now there is an important discussion about whether different SEQS are needed regionally/locally. When managing contaminated sites, SEQS and SEBV will affect the selection, formulation and cost of remediation strategies.

Internationally, different countries have prioritized soil pollution and management in different ways. China has been eager to learn from this (Luo et al., 2015; Wang et al., 2005; Wen et al., 2010; Xia and Luo, 2007) and – given the Government's stated aim to manage its soil pollution problems effectively - has the opportunity to put in place sound, strong policies and management structures. An interesting comparison is with the UK, which has a long history and legacy of contaminated land problems and a mature environmental regulatory system (Luo et al., 2009; Wu, 2007). The 10-Measures Soil Pollution Action Plan strongly recommended that: surveys of the soil pollution situation be conducted; regulations and laws of soil pollution prevention be amended; soil pollution control and remediation be promoted; and a control system to prevent soil pollution be introduced (State Council, 2016).

Given the highly topical nature of soil contamination issues in China, in this paper we focus on the following questions: 1. What is the situation of soil heavy metal contamination in China? 2. What factors affect the background value of heavy metals in soils? 3. What are the background values of selected heavy metals and how do they compare between China and the UK? 4. What can we learn from the UK about soil survey methodologies and soil environmental standard assessment? 5. What are future priorities and next steps for China in its management of soil pollution?

2. Heavy metal soil pollution in China

As in other countries, key sources of heavy metals to Chinese soils include: metal mining and smelting; industrial activities, power generation; agricultural activities, including fertilizer and animal manure amendments; waste disposal activities; urbanization, transportation. In some regions, high contamination of the soil occurs around point sources, for example, mines and smelters – giving high but generally localized problems. In other situations, for example, agricultural soils, contamination may be lower, but important as a direct route of food contamination (Fu et al., 2016; Zhou et al., 2014; Zhu et al., 2008). In some countries, inventories have been published which estimate the relative importance of these different sources to the national soil resource. This approach would be very helpful in China, because it provides a scientific basis to prioritize source reductions; we are not aware that this exercise has been performed in China yet. The distinction between 'high level hotspots' of contaminated land (e.g. brownfield sites; mines) and diffusive agricultural sources may also be important in management. For example, should there be different standards for agricultural land? Can brownfield sites be cleaned by simply excavating and removing dirty soils from important areas of re-development? (Cao and Guan, 2007; Li et al., 2015; Wang et al., 2014; Wei and Yang, 2010).

Over the past decades, heavy metal contamination has increased worldwide, following large-scale mining and industrial releases (Li et al., 2014). Ultimately much of the metal from such activities reaches the soil, via wastes, disposal and atmospheric deposition. China has undergone huge and rapid urbanization and industrial expansion over the past thirty years, which will have resulted in increased release of heavy metals to the environment, and the burden of metals held in surface soils (Chen, 1991). It has been estimated that nearly 20 million hectares of arable land has been polluted by heavy metals, such as Cd, Pb, Cu, and Zn in China, accounting for approximately 20% of the total arable land area (Lin, 2004; Zhao et al., 2007). Although information is not available on a site-specific basis, national soil surveys conducted by the MEP-PRC and the Ministry of Land and Resources of the People's Republic of China (MLR-PRC) (2005–2013) concluded that 16.1% of national land (based on sampling points, including arable land, some woodland, grassland, unused land, construction land) was contaminated (i.e. exceeded background values), of which > 80% were exceeded by inorganic contaminants. Contamination may have been with heavy metals and other inorganic contaminants, and/or with organic chemicals. Cd was responsible for most exceedances, accounting for 7% of national land. It should be noted that there was a sampling bias, because soils were not sampled in proportion to the national land coverage. Nonetheless, the Ministries concluded that "The overall situation looked not optimistic, of which, the situation of arable land and industrial abandoned land are the most severe". In addition, in recent years, many reports in China have highlighted contamination and poisoning of animal and human health through the food chain. Prominent cases include Cd in rice, Pb and As poisoning incidents (Song et al., 2013). Hu et al. (2014) concluded that Pb, Cr, As, Cd and Hg constituted the five most important heavy metal contaminants in Chinese soils.

3. Soil background values and factors that affect them

'Soil environment background values' (SEBV) are the concentration of elements or components in soil with little influence from human activities (Connor and Shacklette, 1975; Wang and Yang, 1990). They reflect the underlying geology and soil formation processes; hence they vary between locations and are commonly expressed as a range of values for a particular country or region. The background value could change over time, if environmental processes (including background human activities) affect the burden in the soil. So, absolute uncontaminated or pristine soils may be difficult to identify, since industrial activities have emitted heavy metals and other contaminants into the atmosphere. In general, the SEBV is a relative concept (Xia and Luo, 2006). The geochemical background refers to the normal abundance of an element in barren earth material or the normal range of an element in certain areas. The concept of geochemical background aims to distinguish the normal and abnormal concentration of elements. In the exploration geochemistry field, it may be an indicator of an ore occurrence, while for environmental geochemistry, it may be an indicator of a contaminated or insufficient element (Cheng et al., 2014; Hawkes and Webb, 1963). It is assumed that the range of SEBVs give 'clean' soils where there are no adverse 'pollution' effects. Hence, contaminated soils are defined with levels of heavy metals (and other constituents) above the SEBV (Xia and Luo, 2006). This is why it is so important to conduct carefully designed surveys of contaminants in soils, because precise definition of the SEBV will determine whether the soil is contaminated and to what degree.

As just noted, the SEBV is affected by various abiotic and biotic factors that change in space and time. For example, parent materials, soil chemical properties, topographic factors, hydrological factors, human activities, geological factors, weathering and leaching conditions of parent material driven by climatic factors etc. (Chen and Wang, 1987; China National Environmental Monitoring Centre, 1990; Fu and He, 1992; Liang and Zhang, 1988). Parent materials are a direct and main factor influencing the SEBV in many studies (Chen and Wang, 1987; Nair and Cottenie, 1971; Oertel, 1961). Climatic factors indirectly affect the SEBV by controlling weathering and leaching processes. Chen and Wang (1987) found soil types and parent materials to be the main drivers that led to a decline from south to north in Shanxi province and from southeast to northwest, based on a survey of distribution trends and factors affecting the SEBV. Deng et al. (1986) showed that the main factors affecting background values are different from region to region in Beijing; topographical characteristics are the main driver in plain regions, while in mountain regions it is parent materials. Hydrological and topographical factors directly influence soil formation factors, soil surface runoff, soil surface temperature, the degree of surface erosion etc., and then influence soil parent materials and soil elemental composition. For example, fine clays which are richer in heavy metals than sands and silts, will accumulate in floodplains and result in higher concentrations than in hillslope soils. Thus, the determination of SEBVs needs to be based on statistical analyses, with careful consideration given to sampling design and soil sample collecting, statistical tests of sample frequency distribution, data distribution patterns and eigenvalues, to show the range of background values with a given confidence interval (China National Environmental Monitoring Centre, 1990). Further details on these issues are given later in the paper.

3.1. Deriving soil background values in China and the UK

The UK has a widely varying geology for a comparatively small country, areas of heavy metal mineralization, a long history of mining and industrial activity, a legacy of soil contamination, and a lot of experience in the management and regulation of soil contamination. The development planning process has been used to deal with contaminated sites since the creation of the current UK land use planning system in 1947 (Luo et al., 2009). Soil remediation of contaminated sites has been carried out since the 1960s, often with low cost and pragmatic solutions (Ferguson, 1999). In 1976, the Inter-departmental Committee on the Redevelopment of Contaminated Land (ICRCL) was established as the first central institutional mechanism to clearly address this issue. In the Environmental Protection Act of 1990, the provision for registers of potentially contaminated sites was included. The current system of regulation was created in the 1995 Environment Act. In 2005, the Contaminated Land Exposure Assessment (CLEA) model was published with a series of soil guideline values and toxicological reports on key soil contaminants. Hence, the UK has developed a relatively effective management system from longstanding practice. China is experiencing a situation like the UK had several decades ago, although on different scales. As noted earlier, mining and industrial activities have become extensive in parts of China, whilst urban areas have expanded and redeveloped on sites with a legacy of contamination. In rural areas, there are several examples where agricultural land and community areas have become contaminated too (Liu et al., 2013; Shi et al., 2008; Zhuang, 2015).

The UK approach to contaminated land management is underpinned by a series of comprehensive surveys of soil contaminants, which allow a clear definition of the typical levels, ranges and distributions of elements. It is therefore useful to compare the situation with China, which has undertaken national surveys too and is planning further work of this kind.

4. Experience from surveying UK soils

In the UK, there have been different national soil surveys in past decades. In the late 1970s, soil information in England and Wales was incomplete, knowledge of regional soil geochemistry was limited, available soil maps only covered ~25% of the area, and the existing information was not based on a representative and unbiased sampling strategy. Thus, between 1978 and 1983, the National Soil Inventory (NSI) carried out a survey of soil background metal concentrations in England and Wales (McGrath and Loveland, 1992).

In 1996, the Royal Commission on Environmental Pollution (RCEP) published the nineteenth report on the Sustainable Use of Soil, which stressed the need for the assessment and monitoring of soil quality, including certain chemical and physical attributes, and some biological parameters. A second survey – the so-called Countryside Survey – therefore began in 1998. The purpose has been to assess and monitor soil quality over time, by returning to the same locations over several years (Barr et al., 2003).

A third survey was conducted in 2011/12, to give guidance on normal levels of contaminants - to support revision of the Part 2A Contaminated Land Statutory Guidance. This was conducted by the British Geological Survey (BGS) in England and Wales (Johnson et al., 2012). A summary comparison of the survey designs and methodologies is presented in Table 1.

Table 1
Comparison of sampling methods across three national soil surveys in the UK.

Parameters	NSI	CS 2000	BGS
Study area	England, Wales	England, Wales, Scotland	England, Wales
Time	1978–1983	1998–2000	2011–2012
Sampling density	5 km × 5 km	1 km × 1 km	G-based: Urban: 1 km × 1 km; rural: 2 km^2; NSI (XRFS): 5 km × 5 km
Quadrat size	20 m × 20 m	14 m × 14 m	20 m × 20 m
Sample number (per quadrat)	1	3	5
Sampling depth	0–15 cm deep topsoil	15 cm deep × 8 cm dia.	topsoil: 0–15 cm; surface soil: 0–2 cm; deeper soil: > 30 cm
Investigated elements	Al, Ba, Cd, Cr, Co, Cu, Fe, Pb, Mg, Mn, Ni, P, K, Na, Sr, Zn	Cd, Cr, Cu, Pb, Ni, V and Zn	As, Cd, Cu, Hg, Ni and Pb
Number of soil samples	5691	1081	42133
Analytical value	Range, mean, median, maximum, minimum	Mean, standard deviation, median, maximum value and minimum value	Mean, range, minimum, maximum

Notes: NSI: National Soil Inventory; CS2000: Countryside Survey 2000; BGS: British Geographical Survey.

Table 2
Comparison of range, mean and median of four comparative heavy metals (Cd, Cu, Ni, Pb) from the results of NSI, CS2000 and BGS surveys (all concentrations in mg/kg).

	Range	Mean	Median
Cd			
NSI	< 0.2–41	0.8	0.7
CS2000	0–11	0.49	0.3
BGS	0.3–20	0.5	0.3
Cu			
NSI	1.2–1508	23	18
CS2000	0.3–448	18	14
BGS	< 1-5326	27	20
Ni			
NSI	0.8–440	25	22.6
CS2000	0–1890	24	16.3
BGS	1–506	25	23
Pb			
NSI	3–16338	74	40
CS2000	1.3–20600	88	37
BGS	3–10000	72	41

Notes: Median values are the most commonly reported values for background soils. They are more meaningful than mean values, which can be biased by a few extreme polluted values. See, for example Davies (1983) paper on soil Pb values.

As Table 1 shows, ~50,000 UK surface (0–15 cm) soil samples have been taken and analyzed, albeit with slightly different purposes and with a focus on different land use types. A comparison of a selection of heavy metals from these surveys is given in Table 2.

Median values (Davies, 1983) provide a good way to compare the data: the 3 surveys gave the following values for the elements presented in Table 2: Cd – 0.7, 0.3 and 0.3; Cu – 14, 18 and 20; Ni, 23, 16 and 23; Pb – 40, 38 and 41. The main conclusions from Tables 1 and 2 are: i. despite different sampling and analytical methods, the general soil quality determined by the 3 surveys is very similar. ii. The sampling, preparation procedures and – crucially - the large number of samples taken provides a robust way to determine the typical range of heavy metal concentrations in soils.

5. Surveying Chinese soils

The earliest research on SEBVs in some selected city areas of China (Beijing, Nanjing, Guangzhou etc.) was in the mid-1970s. Subsequently, in 1978, SEBVs for 9 elements in agricultural soils and crops were surveyed in 13 provinces. In 1982, a background value survey was listed in the national key scientific and technological projects, which was carried out in a few of the main climate zones in northeast China, Yangzi River basin, Pearl River Basin etc. In 1990, a large-scope and systematic survey for SEBVs was carried out across the whole of China, covering all 29 provinces, cities and autonomous regions. These survey data were summarized in a book entitled China Soil Element Background Value (China National Environmental Monitoring Centre, 1990). From 2005, the MEP-PRC and the Ministry of Land Resources launched a national soil pollution survey to capture the distribution data and to look for changes in the 20 years since the 1990 survey. It covered all arable land and parts of the woodland, grassland, unused and construction land. In 2014, a national bulletin on site-specific soil pollution status was published, to summarize the pollution situation without detailed site-specific or soil survey data.

Table 4
The range, mean and median of four comparative heavy metals from the 1990 China soil survey (all concentrations in mg/kg).

China soil survey 1990	Range	Mean	Median
Cd	0.001–13.4	0.097	0.079
Cu	0.33–272	23	21
Ni	0.06–628	27	25
Pb	0.68–1143	26	24

A comparison of Tables 2 and 4 shows close agreement for Chinese and UK Cu and Ni background values. The median value for Cd in UK soil is ~0.3 mg/kg, about 3 times higher than that of the Chinese 1990survey. UK Pb median values were ~40 mg/kg, against a Chinese median of 24 mg/kg. This might be explained by the UK's long history and density of Pb mining and inefficient smelting operations (Davies, 1983).

6. Methodologies for determining background concentrations in soil

6.1. Statistical methods used for UK soils

6.1.1. Countryside Survey 2000

All elements were analyzed in different environments, classified according to Land Class and eighteen broad habitats and major soil groups and Countryside Vegetation System Aggregate Vegetation Class. Data are typically presented as figures (box-plots, scatterplots, frequency histograms etc.) to summarize the variation in different environmental factors. Mean values, standard deviations, median, maximum value and minimum values are commonly calculated to represent the primary analysis (Black et al., 2002).

6.1.2. National Soil Inventory

For all variables, the range, mean, median, maximum, minimum, skewness and kurtosis were calculated for both transformed and log10-transformed data (except for pH). Box plot analysis was performed for the data of Cd, Co, Cr, Cu, Ni, Pb and Zn. Correlation analysis was performed on soil element concentrations (log10-tranformed data). Principal component analysis (PCA) was performed on all datasets, including all elemental concentrations, organic carbon and pH to provide an overall view of the relations among variables. Simple or multiple linear regression analysis was used to exclude the outlier data (McGrath and Loveland, 1992).

6.1.3. British Geological Survey

Values for contaminant domain normal background concentrations were calculated by a study of a contaminant's population distribution. Skewness coefficient and octile skew were used as statistical measures. Percentiles for the domain data sets for each contaminant were generated along with calculations of percentile confidence intervals. The

Table 3
Sampling details for the 1990 national soil survey in China.

Study area	Covered 29 provinces, cities and autonomous regions
Time	1990
Sampling density	East areas: 30 × 30 km^2 per study point; Central areas: 50 × 50 km^2 per study point; west areas: study point from 80 × 80 km^2 per study point
Soil profile	1.5 m × 0.8 m × 1.2 m (Length × width × depth)
Sample depth	A layer: 0–20 cm, B layer: 50 cm, C layer: 100 cm
Investigated elements	As, Cd, Co, Cr, Cu, F, Hg, Mn, Ni, Pb, Se, V, Zn
Number of soil samples	4095
Analytical value	Maximum value, minimum value, arithmetic mean and geometric mean

upper limit for a normal background concentration has been as the upper confidence limit of the 95th percentile (Johnson et al., 2012).

6.2. Statistical methods used in Chinese soils

Relevant information (soil types, parent materials, topography, latitude, longitude, vegetation, land use types, administrative regions etc.) of 4095 typical soil profiles, together with the chemical analytical data were stored in a database of Chinese soil background values. In summary, soil types were divided into 41 statistical units, parent materials were divided into 21 units, and administrative regions were divided into 34 units, so in total, every element has 97 statistical units. Frequency distribution graphs are available for different elements, with the maximum, minimum, arithmetic mean and geometric mean values were presented. For elements with a log-normal distribution, the geometric mean (M) was used to represent the data distribution, the geometric standard deviation (D) to represent the level of dispersion, and M/D^2-MD^2 for the range of 95% confidence interval. For the elements with a normal distribution, the arithmetic mean (symbol of mean value) was used to represent the data distribution, the arithmetic standard deviation (s) for the level of dispersion and symbol of mean value ± 2s for the range of 95% confidence interval (China National Environmental Monitoring Centre, 1990).

7. Soil standards

Environmental Quality Standards (EQSs) for soils (GB15618-1995) in China were officially released in 1995. They were derived based on several factors: data on the soil background in China; data from soil ecological tests; data from geographically anomalous areas in China and information on soil standards or guidelines from abroad (MEP-PRC, 1995; Wu and Zhou, 1991). These EQSs set the maximum acceptable concentration of pollutants and relevant monitoring methods in the soil based on different soil functions/uses, protection targets and soil properties.

Three types of standard were set: Type I is protective of soils in national nature reserves, centralized drinking water resources, tea plantations, pasture and other protected areas, and the goal is to basically maintain the natural background level. Type II is applicable to the soil in general farmland, land for growing vegetables, tea plantations, orchards, pasture etc., where the goal is to not cause harm and pollution to plants and the environment. Type III is applicable to woodland soil, and farmland soils near to high background soils of more pollutant capacity and mineral fields, where the goal is basically to not cause harm and pollution to plants and the environment. Chinese EQSs take account of the soil pH value, cropping pattern and soil cation exchange capacity (see Table 5 for details) (MEP-PRC, 1995).

In order to protect agricultural soil, control agricultural soil contamination risk, safeguard agricultural product security, the normal growth of crops and soil ecological environment, China has been working on the development of new soil environmental quality standards. Twenty years after the release of GB15618-1995, the new soil quality guidance (Risk control standard for soil contamination of agricultural land GB15618-2018) was issued in 22nd June 2018, to replace GB15618-1995 and to take effect on 1st August 2018 (Tables 6 and 7). This standard regulates the soil risk screening value and risk intervention value in agricultural land, and the requirements of monitoring, implementation and supervision. These values were derived from human health risk assessment procedures. In addition, a risk control standard for soil contamination of development land (GB36600-2018) was also issued, to come into force at the same time to protect human health and living environmental security.

To quote the new guidance: 'The value of the main pollutant content in the soil when the quality and safety of edible agricultural products, crop growth or the soil ecological environment are or may have adverse effects. If the content of pollutants in soil is lower than this value, the risk of soil pollution such as non-conformity of quality and safety standards in edible agricultural products, may generally be ignored. If there may be a risk of soil pollution, soil environmental monitoring and coordinated monitoring of agricultural products should be strengthened, and in principle, safe use measures should be taken. Agricultural land is classified into three types – arable land (paddy, irrigated land, dry land), garden (orchard, tea garden) and pasture (natural pasture and artificial pasture). In this standard, 'others' include all kinds of land except for paddy'.

Table 5
Soil environment quality standards adopted in China for general farmland (mg/kg). See text for definition of Type I, II and III.

Soil pH		Natural background	< 6.5	6.5–7.5	> 7.5	> 6.5
Standard		Type I soil	Type II soil			Type III Soil
Cd		0.20	0.30	0.30	0.30	1.0
Hg		0.15	0.30	0.50	1.0	1.5
As	Paddy field	15	30	25	20	30
As	Non-irrigated farmland	15	40	30	25	40
Cu	Farmland	35	50	100	100	400
Cu	Orchard	–	150	200	200	400
Pb		35	250	300	350	500
Cr	Paddy field	90	250	300	350	400
Cr	Non-irrigated farmland	90	150	200	250	300
Zn		10	200	250	300	500
Ni		40	40	50	60	200

Table 6
Screening values of soil pollution risk of agricultural land (basic items) (mg/kg).

Pollutant	Risk screening value				
		pH ≤ 5.5	5.5 < pH ≤ 6.5	6.5 < pH≤7.5	pH > 7.5
Cd	Paddy	0.3	0.4	0.6	0.8
Cd	other	0.3	0.3	0.3	0.6
Hg	Paddy	0.5	0.5	0.6	1
Hg	other	1.3	1.8	2.4	3.4
As	Paddy	30	30	25	20
As	other	40	40	30	25
Pb	Paddy	80	100	140	240
Pb	other	70	90	120	170
Cr	Paddy	250	250	300	350
Cr	other	150	150	200	200
Cu	Paddy	150	150	200	200
Cu	other	50	50	100	190
Ni		60	70	100	190
Zn		200	200	250	300

Table 7
Intervention values of soil pollution risk of agricultural land (mg/kg).

Pollutant	Risk intervention value			
	pH ≤ 5.5	5.5 < pH ≤ 6.5	6.5 < pH≤7.5	pH > 7.5
Cd	1.5	2.0	3.0	4.0
Hg	2.0	2.5	4.0	6.0
As	200	150	120	100
Pb	400	500	700	1000
Cr	800	850	1000	1300

Notes: Risk intervention value in this standard refers to the value of the main pollutant content in the soil when it causes or may cause serious effects on the quality and safety of edible agricultural products. If the content of pollutants in the soil exceeds this value, the risk of soil pollution, such as non-compliance with quality and safety standards, is high, and strict control measures shall be taken in principle.

A particular challenge for China at the present time is that specific areas are considering variants to the national standards, to reflect their particular challenges. For example, areas (jurisdictions at province or city level) with high background values, or particular soil/crop systems may wish to adopt more pre-cautionary limits. In other situations, standards may be considered as targets for remediation of contaminated sites. Although not published from the national survey of China, there are many studies which have reported geographical variations, with some provinces having high background values, for example (Cheng et al., 2014; Cheng and Tian, 1993; Dong et al., 2007; He et al., 2006; Pan and Yang, 1988). Hunan Province is an interesting example. It has a long history (~2700 years) of non-ferrous metal mining and metal resources, which began to be extensively exploited in the 1980s. With industrial development, many incidents and impacts from heavy metal pollution have been widely reported in this area. For example, in June 2014, 315 children living around Dapu industrial area in Hengdong county were reported with excessive Pb concentrations in their blood, 10 of which had been sub-chronically poisoned. Another heavy metal survey published in November 2014 from an environmental protection organization showed that the As content of river sediments exceeded national standards by 700 times and the Cd content in some paddy soils exceeded the standards by 200 times in the Sanshiliuwan mining area from Chenzhou City (Cao and Li, 2014). Regulations have been issued by the province – for example - an 'Implementation Plan (2012–2015)' of heavy metal pollution control in the Xiangjiang river basin, which has been set to close illegal factories, control industrial pollution sources and decrease heavy metal emissions and remediate the legacy contaminated sites. In 2016, standards for soil remediation of heavy metal contaminated sites (DB43/T1165-2016) were issued by the Environmental Protection Department of Hunan and Hunan Provincial Bureau of quality and technical supervision. These provided the remediation standard for 11 heavy metals in residential land, commercial land and industrial land (Table 8). These remediation targets are higher than the national standard in GB15618-1995. For example, DB43/T1165-2016 values are: Cd-7, 20, 20 in residential land, commercial land and industrial land, respectively; GB15618-1995 values are: Cd-0.3, 0.3, 0.6, 1 in Standard II pH < 6.5, 6.5–7.5, > 7.5, Standard III, respectively; GB15618-2018 screening values are: Cd-0.3, 0.4, 0.6, 0.8 for paddy pH ≤ 5.5, 5.5 < pH ≤ 6.5, 6.5 < pH≤7.5, pH > 7.5, respectively. Details of how Hunan's standards were derived are not clear, but they may be pragmatic and risk-based.

EQSs are considered impractical as remediation targets, because of the high costs/time required to achieve such a level of clean-up (Cai et al., 2005; Qiu et al., 2007; Wen et al., 2010; Wu and Zhou, 1991; Xia and Luo, 2007; Yu et al., 2010). A crucial aspect of any remediation targets is the after-use of the land. Land for residential or agricultural use would require more stringent limits than amenity land, for example.

The new soil EQSs have added some further details. For example, they add one other organic contaminant – benzo-a-pyrene, and GB15618-2018 gives a newly added soil screening value and soil control value at the pH level of 5.5. Under GB36600-2018, different land uses are considered when developing soil EQSs. These two standards provided one method for identifying soil heavy metal contamination; If the content of pollutants exceeds the screening value, but is not higher than the background value of the soil environment, it is not included in the management of contaminated land. However, some considerations are still not resolved; for example, more contaminants still need to be considered and soil ecological protection still needs to be addressed.

8. Current situation and future priorities

China is highly reliant on its 'best quality' soils for food security and agricultural production. It has been estimated that 20% of China total arable land is contaminated (Lin, 2004; Zhao et al., 2007). This may be different from the situation in most developed countries, where a higher proportion of agricultural land is not contaminated. For example, a higher (~93%) proportion of European agricultural land is considered safe for food production (Tóth et al., 2016). The reality is that China will need to produce food for human consumption on soils which are already deemed 'contaminated'. Scientifically based risk assessments are necessary to inform practical decisions about the most practical land use options. For example, important research is currently being conducted in China and elsewhere to understand where and how 'contaminated' land can be used to support food production. This requires knowledge of soil chemistry and soil-crop plant transfers of contaminants (Tangahu et al., 2011; Xu et al., 2005). If China makes these changes/meets these priorities, it can be leading the world in approaches to contaminated land management. It is in this context that China is committed to conducting the most detailed and comprehensive soil survey to date. MEP-PRC carried out a nation survey covered 6,300,000 km^2 soil area from April 2005 to December 2013, and several geochemistry surveys by MLR-PRC have been completed from 1999 to 2014, which covered 68% of total arable land (MEP-PRC, 2016). There are several areas where revisions are being considered, to bring China to a leading position internationally. It is hoped that the new regulatory approaches can be further developed to:

1. Increase the range of analytes for which standards are set. Most focus so far has been on inorganics, but there is a wide array of organic contaminants for which standards can be set.
2. Critically important to the science-based risk assessment of contaminants in soils is the incorporation of speciation and bioavailability into the measurement and evaluation criteria. On initial screening, soils and sites may be deemed 'contaminated', but after a second tier analysis they may be shown to be suitable for crop production and use. Selection of appropriate and validated

Table 8
The remediation standard of heavy metal in contaminated sites for Hunan Province (first 3 columns), compared to the national standards (all concentrations in mg/kg).

Elements	Residential land	Commercial land	Industrial land	National Standard I soil	National Standard II soil			National Standard III soil
pH					< 6.5	6.5–7.5	> 7.5	> 6.5
Pb	280	600	600	35	250	200	200	400
As	50	70	70	15	30	25	20	30
Cd	7	20	20	0.2	0.3	0.3	0.6	1
Hg	4	20	20	0.15	0.3	0.5	1	1.5
Cr	400	610	800	90	250	300	350	500
Cr^{+6}	5	30	30	–	–	–	–	–
V	200	250	250	–	–	–	–	–
Mn	2000	5000	10000	–	–	–	–	–
Cu	300	500	500	35	50	100	100	400
Zn	500	700	700	100	200	250	300	500
Sb	30	60	60	–	–	–	–	–

Notes: see Table 5 for more information on the national standards.

measurement and evaluation tools is a priority. If this is done in a scientifically transparent and defensible way, China will have a robust and internationally leading system for soil management in place.

3. Derivation of standards has focused on human receptor endpoints. However, consideration of soil biology/ecological endpoints will be necessary to protect ecosystem health.
4. National and regional/local scenarios of land use type/usage. This addresses residential/urban re-use of industrial land, as well as varying agricultural scenarios, such as different agricultural systems and cropping regimes.

Acknowledgements

We are grateful to Dr Lisa Norton and Dr Aidan Keith in Centre for Ecology & Hydrology-Lancaster for their contribution to the knowledge of the Countryside Survey.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.jenvman.2019.109512.

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Appendix 2

Environmental Pollution 256 (2020) 113404

Contents lists available at ScienceDirect

Environmental Pollution

journal homepage: www.elsevier.com/locate/envpol

A comprehensive comparison and analysis of soil screening values derived and used in China and the UK☆

Yiming Sun [a, b, c, 1], Jicai Wang [b, 1], Guanlin Guo [a, b, *], Hong Li [c, d], Kevin Jones [c]

[a] Technical Centre for Agricultural and Rural Ecology and Environment, Ministry of Ecology and Environment, Beijing 100012, PR China
[b] State Key Laboratory of Environmental Criteria and Risk Assessment, Chinese Research Academy for Environmental Sciences, Beijing 100012, PR China
[c] Lancaster Environment Centre (LEC), Lancaster University, Lancaster, LA1 4YQ, UK
[d] Centre for Ecology and Hydrology, Maclean Building, Wallingford OX10 8BB, UK

A R T I C L E I N F O

Article history:
Received 1 June 2019
Received in revised form
8 October 2019
Accepted 14 October 2019
Available online 19 October 2019

Keywords:
Soil pollution
Soil screening values
Soil guideline values
China
UK

A B S T R A C T

China and the UK use different risk-based approaches to derive soil screening or guideline values (SSVs; SGVs) for contaminants. Here we compare the approaches and the derived values for 6 illustrative contaminants. China's SSVs are derived using an approach developed in the US as follows: for carcinogens, acceptable level of risk (ACR) is set at 10^{-6} and the SSVs calculated as 10^{-6} divided by the soil exposure and toxicity data; for non-carcinogens, the hazard quotient is 1 and the SSV is calculated as 1 divided by the soil exposure and toxicity data. The UK's SGVs are calculated by the CLEA model, for which the Average Daily Exposure (ADE) from soil sources by a specific exposure route equals the health criteria values (HCVs) for that route, whether for carcinogens or a non-carcinogens. The UK's CLEA model is also used here to derive SSVs with Chinese input parameters. China's SSVs, the UK's SGVs and values for Chinese conditions derived using the UK approach were as follows (mg/kg): As, <1, 35, 20; Cd, 20, 18, 11; Cr (VI), <1, 14, 29; benzene, 1, 1, 2; toluene, 1200, 3005, 3800; ethyl-benzene, 7, 930, 1200. By comparing the differences in toxicity assessment and risk characterization, exposure assessment and parameter types in the methodologies to obtain SSVs in China and the UK, and by combining the CLEA model with Chinese parameterisation, these comparisons highlight that the difference in toxicity assessment and risk characterization methods of carcinogens results in the biggest difference in SSVs between the 2 countries. However, for non-carcinogenic substances, the difference of SSVs calculation method and SSVs is small. The difference in SSVs for carcinogenic substances is also related to the route of exposure. For volatile organic compounds, the presence of indoor respiratory exposure pathways greatly reduces the differences caused by toxicity assessment and risk characterization methods. For non-volatile substances such as heavy metals, the effects of toxicity assessment and risk characterization methods are significant. The SSV of As obtained by the CLEA model with Chinese parameters is closer to the background value of soil in China. In the management of non-volatile contaminated sites such as heavy metals in China, the CLEA model can be used for risk assessment and calculation of site specific SSVs. In the future, China can use the UK method to strengthen its toxicity assessment and risk characterization methods for carcinogenic substances, to reduce the uncertainty in the risk assessment of contaminated sites and improve the scientific management of contaminated sites.

1. Introduction

Soil pollution has become a widespread and serious problem in many regions of the world (Cachada et al., 2018; Chen et al., 2014; Barsova et al., 2019; Ramón and Lull, 2019; Kumar et al., 2018). In the past thirty years, environmental risk assessment has been widely adopted in many countries to manage soil pollution in contaminated land, and some countries (e.g. United States, United Kingdom, Netherlands, Canada and Australia) have developed risk

☆ This paper has been recommended for acceptance by Baoshan Xing.

* Corresponding author. Technical Centre for Agricultural and Rural Ecology and Environment, Ministry of Ecology and Environment, Beijing 100012, PR China.
E-mail addresses: guogl@craes.org.cn (G. Guo), k.c.jones@lancaster.ac.uk (K. Jones).
[1] Yiming Sun and Jicai Wang are co-first authors.

https://doi.org/10.1016/j.envpol.2019.113404

based approaches to derive contaminant-specific values, to help with management of contaminant scenarios. These are designed to protect human health and manage soil pollution in accordance with national regulations. Soil screening values (SSVs) are derived by risk based approaches, and provide an important support tool for contaminated land management (United States Environmental Protection Agency, 1996; Swartjes et al., 2012; Environmental Agency, 2009a; CCME, 2006; National Environment Protection Council, 2011). SSVs are used to categorise the risk of soil contamination. For a specific land use, if the concentration of a given contaminant is less than the SSV, it is defined as being of no risk to human health. If it exceeds the SSV, this may trigger further surveys, risk assessments, potential changes of land use or remediation measures, depending on the national processes (United States Environmental Protection Agency, 1996; Environmental Agency, 2009a; CCME, 2006; National Environment Protection Council, 2011; Ministry of Environmental Protection of the People's Republic of China, 2018a). However, SSVs in different countries are different in terms of definition, numerical values and inference methods. For example, Zhou et al. (2016) found standard values for arsenic varied by country, land use and definition (Zhou et al., 2016). Generally, standard values of industrial land are higher than those of commercial, residential and agricultural land. Standard values of some countries and regions place more emphasis on soil properties, soil types and extractants, not land types. Carlon et al. (Carlon, 2007) conducted a comprehensive analysis of SSVs in different European countries and found it was subject to geographical, biological, socio-cultural, regulatory, political and scientific factors. The value of SSVs in different countries in Europe are different in value and usage, and the influence of different factors is different. Many factors combine to result in differences, namely: i. the approaches used to derive the SSVs (e.g. hazard identification, toxicity assessment, exposure assessment and risk characterization); ii. the descriptors/parameters selected (e.g. the population/soil/site characteristics and building structure; the environmental conditions and parameters values); iii. The proposed land use or level of 'acceptable risk' (Claudio et al., 2007; Song et al., 2011; Wang and Lin, 2016; Xu et al., 2013). Due to these reasons, SSVs derived and used in different countries can be different, resulting in different management options being selected for the same soil concentration in different places. Thus, using scientific derivation methods, matching the parameter values of regional characteristics, and calculating the soil screening value to meet the risk level of policy requirements is the basis for scientific management of contaminated soils in a country and region, which is necessary to derive methods, parameter values, etc. In this paper, a comparative analysis of SSVs is carried out, to provide scientific reference for method selection and parameter determination. Our focus is China, as explained below.

The UK's approach is one of the most established. Over the last 20 years, the Environment Agency (EA) has systematically released a series of regulations, standards and science reports to introduce how to deal with soil contamination in the UK (Environmental Agency, 2009a; Environmental Agency, 2009b; Environmental Agency, 2009c; Environmental Agency. Sci, 2009). Soil Guideline Values (SGVs) have been derived and widely applied to the investigation and management of contaminated land (Environmental Agency, 2009a). SGVs are defined as a starting point for evaluating long-term and *on-site* exposure risks to human health from chemicals in soil, below which the long-term human health risks are tolerable or minimal, above which further investigation should be undertaken. It uses the CLEA (Contaminated Land Exposure Assessment) model to derive SGVs.

In recent years, the Chinese central government and local government has started to pay attention to soil pollution by taking a series of actions (Hou and Li, 2017; Li et al., 2017). For example, in 2016, the 10-Chapter Soil Pollution Action Plan was issued. It's purpose is to manage, control and prevent soil pollution and improve soil quality in China (People's Daily, 2016). An early priority is to conduct relevant surveys of soil pollution, to define baselines of soil environmental quality (Council, 2016). In August 2018, the Chinese government released national standards for contaminants in agricultural soils and contaminated land, Soil Screening Values (SSVs) (Ministry of Environmental Protection of the People's Republic of China, 2018a, 2018b). China's SSVs are also derived using a risk-based approach. Fig. 1 shows the procedures used to derive SSVs in China and the UK.

The purpose and objectives of the study were therefore to:

(1) compare the derivation method of SSVs in China and the UK in terms of toxicity assessment, risk characterization and exposure assessment;
(2) identify the key differences in SSVs between China and UK and their main factors;
(3) provide some suggestions for the improvement of China's national and local SSVs standard setting.

To achieve these goals, six chemicals were selected as examples, 3 inorganic and 3 organic, for which SSVs have been published. These are As, Cd, hexavalent Cr, benzene, toluene and ethylbenzene.

2. The approaches to derive SSVs in China and the UK

The basic principle in each case is to derive a soil concentration which gives an acceptable level of risk (ACR), using knowledge of the contaminant behavior in soils, an assessment of exposure and toxicological information.

2.1. The Chinese approach

China's approach is based on that used in the USA and developed by the US Environmental Protection Agency (US EPA). Carcinogenic and non-carcinogenic substances are treated differently in the SSV calculation process. For carcinogenic substances without an effect threshold, it's ACR is set at 10^{-6}, the SSVs is calculated as follows (Ministry of Environmental Protection of the People's Republic of China, 2014):

For carcinogens:

$$RCVS = \frac{ACR}{R_{oral} \times SF_o + R_{dermal} \times SF_d + R_{inhalation} \times SF_i} \quad (1)$$

For a threshold non-carcinogenic substance, its ACR is named as acceptable hazard quotient (AHQ) and set at 1, the SSVs is calculated as follows:

For non-carcinogens:

$$HCVS = \frac{AHQ \times SAF}{\frac{R_{oral}}{RfD_{oral}} + \frac{R_{dermal}}{RfD_{dermal}} + \frac{R_{inhalation}}{RfD_{inhalatio}}} \quad (2)$$

where: *RCVS* is the SSVs for carcinogens, mg/kg;
HCVS is the SSVs for non-carcinogens, mg/kg;
ACR is the acceptable risk level, 10^{-6};
AHQ is the hazard quotient, 1;
R_i is route of exposure (oral, inhalation and dermal), mg/kg-d;
SF_i is the slope factor (oral, inhalation and dermal), 1/(mgkg-d);
RfD_i is the reference dose (oral, inhalation and dermal), mg/kg-d;
SAF is soil allocation factor, dimensionless.

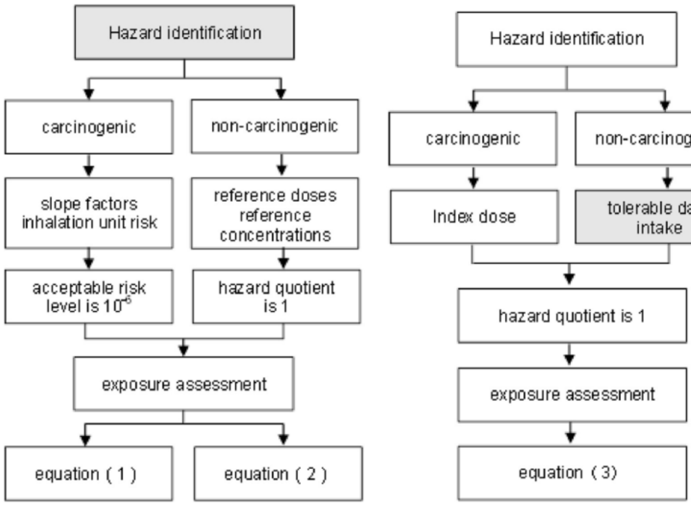

Fig. 1. The derivation procedure for SSVs in China (Left) and the UK (Right).

The AHQ for a threshold non-carcinogenic substance is 1, and the SSV (Equation (2)) is calculated as 1 divided by the soil exposure and toxicity data (reference dose, etc.). The process can be done with Microsoft Excel or other calculation programs. Three exposure routes are considered: oral intake, dermal contact and inhalation intake. Residential and commercial land uses are considered separately.

2.2. The UK approach

The UK's SGVs are based on the four steps of hazard identification, toxicity assessment, exposure assessment and risk characterization of contaminated soil, using the CLEA model - a defined framework and methodology (Environmental Agency. Sci, 2009). The basic principle used to establish SGVs is to set the soil concentration, of which the Average Daily Exposure (ADE) from soil sources by a particular exposure route equals the health criteria values (HCVs) for that route. The CLEA software estimates the ADE to soil contaminants by adults and children living or working on contaminated land over long periods of time, and compares this estimate to HCVs (Equation (3)) (Environmental Agency, 2009b; Environmental Agency. Sci, 2009). Again exposure via ingestion through the mouth (oral), absorption through the skin (dermal), and inhalation through the mouth and nose are considered. SGVs are derived for three generic land use scenarios: residential, allotment (gardening to grow food) and commercial.

$$\frac{CR_{oral}}{HCV_{oral}}+\frac{CR_{dermal}}{HCV_{dermal}}+\frac{CR_{inhalation}}{HCV_{inhalatio}}=1 \quad (3)$$

where: C is the representative concentration of the chemical in soil, mg/kg;

Rx is the ratio of ADE from the soil source over the soil concentration for exposure route x (oral, inhalation and dermal), mg/kg·d over mg/kg;

HCVx is health criteria value for exposure route x (oral, inhalation and dermal) mg/kg·d.

2.3. Highlighted differences between the Chinese and UK approaches

2.3.1. Toxicity assessment and risk characterization

China's chemical substances hazard identification and toxicity assessment use the US EPA's Integrated Risk Information System (IRIS). According to the characteristics of the dose-response relationship, carcinogens are considered with a non-threshold limit, while non-carcinogens have a threshold. For carcinogens, the oral slope factor (SF) gives a plausible upper-bound estimate of the probability of a response per unit intake of a chemical over a lifetime, and the inhalation unit risk (IUR) is defined as the upper-bound excess lifetime cancer risk estimated to result from continuous inhalation exposure to a chemical at a concentration of 1 μg/m^3 in air (United States Environmental Protection Agency). The SFs and IURs in the IRIS database are used for toxicity assessment. Non-carcinogens use the reference dose (RfD) as an estimate of a daily oral exposure or the reference concentrations (RfC) to estimate inhalation exposure (United States Environmental Protection Agency). The UK's EA Human Health Toxicological Assessment of Contaminants in Soil Science Report also classifies the toxic effects of substances into threshold-toxic and non-threshold toxicities based on dose-response characterization, but the names, values and derived method of HCVs is different from IRIS. For non-carcinogens, threshold-based substances use tolerable daily intakes (TDIs) as HCVs for human health toxicity assessment, and non-threshold substances with an Index Dose (ID) as HCVs (Environmental Agency, 2009b). The TDI is defined as an estimate of the amount of a contaminant that can be ingested daily over a lifetime without appreciable health risk. ID expressed as a daily dose which is likely to be associated with a negligible risk of carcinogenic effect over a specified duration of exposure.

According to the IRIS, for carcinogens, the Benchmark Dose

(BMD) or No Observed Adverse Effect Level (NOAEL) or Lowest Observed Adverse Effect Level (LOAEL) were used in the dose response curve to derive the SF or IUR for humans through a linear mathematical model with an acceptable cancer risk level of 10^{-6}; for non-carcinogens the BMD, NOAEL, LOAEL or categorical regression were used with uncertainty factors (UF) to derive the RfD or RfC to reflect the daily intake limitation. In the UK both the TDI and the ID are derived as the same method as RfD in the US using the BMD, NOAEL or LOAEL with UF. So the main difference of toxicity assessment between China and UK is the hazard characterization of carcinogens. China adopted the quantitative dose-response modelling from US, while the UK use the non-quantitative extrapolation.

Due to the differences in toxicity assessment methods, the risk characterization approach and the structure of SSVs calculation equation is inevitably different between two countries. In China, for carcinogens, the soil screening value is calculated by dividing the ACR 10^{-6} by the product of soil exposure and carcinogenic slope factor (Equation (1)); for non-carcinogens the soil screening value is calculated by dividing the product of the reference dose and the AHQ 1 by the soil exposure (Equation (2)). The UK CLEA model calculates the SGVs, whether carcinogenic or non-carcinogenic, by dividing the product of the HCV by 1 by the soil exposure (Equation (3)). For carcinogens, there is no clearly acceptable risk level of cancer (although Theoretically, the health risk corresponding to the dose should be 10^{-5}) (Environmental Agency, 2009b), and the ID is significantly different from the carcinogenic slope factor derived from the linear mathematical model. It is more similar to the reference dose of Chinese non-carcinogens in application and value. If equation (3) is rewritten, its form is exactly the same as equation (2). This shows that the difference in the toxicity assessment and risk characterization of carcinogens is the fundamental reason for the different formulae for soil screening values of carcinogens in the two countries.

2.3.2. Exposure assessment

At present, countries including the UK, the US, the Netherlands, and China have established exposure assessment models for the risk assessment of soil pollution (CCME, 2006; Environmental Agency, 2009c; United States Environmental Protection Agency, 1992; Brand et al., 2007). Exposure assessment is an important aspect of human health risk assessment for soil contaminants and a model basis for soil screening value calculations to assess the amount of soil pollutants that may be exposed to human. The exposure pathways considered by both Chinese and English in soil exposure assessment include ingestion, dermal contact and inhalation, and the exposures of each pathways are calculated using the corresponding models. The differences in exposure assessment are mainly reflected in three aspects: a) the sensitive receptor will vary with the type of land use; b) the source of soil contaminants will vary depending on the type of land use; c) the exposure assessment model equation is different for the same exposure pathway. For example, the sensitive receptors of residential land use in China are children (0–6 years old) and adults (female), depending on the carcinogenic and non-carcinogenic effects of the agent, while the UK CLEA model only considers female children (1–6 age class) for both carcinogenic and non-carcinogenic effects. For soil contaminant sources, the Chinese model only considers direct soil contaminant exposure, and there is no source of self-produced crops. The CLEA model can consider not only direct soil contaminant exposure, but also indirect exposure such as ingestion of self-produced crops. The soil ingestion rate, the soil particulate emission model and the vapor intrusion model for exposure assessment are the largest differences between the two countries in the exposure models. In this study, in order to limit the comparison in a unified framework, the scenarios of residential land use, without the self-produced crops ingestion exposure pathway, is selected - mainly comparing the models and model parameters of each exposure pathway in China and the UK.

2.3.2.1. Exposure assessment models. In the selected residential land use, the exposure pathways in China and the UK CLEA model are soil ingestion, dermal contact, inhalation of dust and volatile organic compounds vapors indoors or outdoors. The exposure assessment equation consists of the daily soil intake rate (R_x) of different exposure routes, the exposure duration (ED), exposure frequency (EF), body weight (BW) and average time (AT), where the exposure duration (ED) and the average time (AT) vary with toxic effects and sensitive receptors. The exposure frequency (EF) and the daily soil intake rate (R_x) are related to the exposure route and different in the equations, parameters and values between the two countries (see Table 1).

It can be seen from Table 1 that in addition to considering the oral soil ingestion rate of adults, the oral soil ingestion rate of children in China is 100 mg more than that of children in the UK. The dermal contact daily soil intake rate in the Chinese and UK calculation equations are completely consistent with each other. The inhalation route can be divided into inhalation of soil dust and inhalation of VOC vapors in the two countries. For the inhalation of soil dust, the PM_{10} is used in the Chinese equation to calculate the daily soil intake rate of soil particles in the indoor and outdoor ambient air multiplied by the ratio of particles from the soil in the indoor and outdoor air, p_i and p_o, respectively. The exposure frequency (EF) of children and adults are divided into the indoor exposure frequency (EFI_{inh}) and the outdoor exposure frequency (EFO_{inh}). The UK CLEA model adopts the particulate emission factor (PEF) to calculate the rate of daily soil intake exposed to ambient air in children. The PEF is calculated by the air diffusion factor Q/C_{wind} and the inverse of soil PM_{10} emission flux $J_{w,}$ where Q/C_{wind} can take the monitoring data of different cities, J_w is calculated with wind speed values (Environmental Agency, 2009b; Environmental Agency. Sci, 2009). For inhalation of vapors, China assumes the source of vapors from surface soils, subsurface soils and indoor air. The CLEA model assumes vapors from surface soils and indoor air. The rate of daily vapor intake exposed to ambient air from the source of surface soils is characterized by the volatilization factor (VF_{sur}), and since the VF_{sur} equations adopted in two countries were recommended by the American Society for Testing and Materials (ASTM) or the US EPA, the formula is very similar. The difference in detail is that the air diffusion factor is calculated by DF_{oa} in China and by Q/C_{wind} in the CLEA model. For calculation of the vapor intrusion concentration in indoor air from the contaminated subsurface soil located below the bottom of the building floor or foundation, the equations used in China and in the CLEA model are completely different. This is because China adopts the VF_{ind} equation of the ASTM to calculate the vapor intrusion concentration for indoor air, but the CLEA model uses the soil vapor concentration to calculate the indoor air concentration by multiplying the attenuation factor α; the pressure difference between the soil air and indoor air is 0 Pa in China, and in the CLEA model is 3.1 Pa.

3. Results

3.1. Effect of methods on SSVs

In order to confirm the influences of derivation methods on the calculation result of SSVs between the two countries, As, Cd, Cr (VI), benzene, toluene and ethylbenzene were selected as representative heavy metals and volatile organic compounds (VOCs) under the exposure scenario of residential land use (without self-produced

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Table 1
Comparison of exposure model and parameters in China and UK.

Item	China	UK
Sensitive receptor	Carcinogen: children (0–6 years old) and adult (Female) Non-carcinogen: children (0–6 years old)	Children (Female, 1–6 age class)
Assessment model	Carcinogen: $ER_x = \frac{R_{x-c} \times ED_c \times EF_{x-c}}{BW_c \times AT} + \frac{R_{x-a} \times ED_a \times EF_{x-a}}{BW_a \times AT}$ Non-carcinogen: $ER_x = \frac{R_{x-c} \times ED_c \times EF_{x-c}}{BW_c \times AT}$ Where:ER_x is the daily human exposure for a exposure pathway x,mg/kg-d; R_x is the daily soil intake rate for exposure route x for children (c) or adults (a),mg/d; ED is exposure duration for children (c) or adults (a), a; EF_x is exposure frequency for children (c) or adults (a), which can be subdivided into indoor (EFI) and outdoor (EFO) exposure frequencies, d/a; BW is body weight to children (c) or adults (a), kg; AT is average time, d.	$ER_x = \frac{R_{x-c} \times ED_c \times EF_{x-c}}{BW_c \times AT}$
Exposure duration (ED)	Carcinogen: children: 6 years; adult: 24 years Non-carcinogen: children: 6 years	Children: 6 years
Exposure frequency (EF)	Soil ingestion and dermal contact: children: 350 d/y, adult: 250 d/y Indoor inhalation: children:262.5 d/y, adult: 187.5 d/y Outdoor inhalation: children: 87.5 d/y, adult: 62.5 d/y	Soil ingestion and dermal contact: Age class 1: 180 d/y; Age class 2–6: 365 d/y Indoor and outdoor inhalation: Age class 1–6: 365 d/y
Average time (AT)	Carcinogen: 27740d Non-carcinogen: 2190d	2190d
Oral	Children soil ingestion rate: $R_{oral\text{-}c} = 200$ mg/d Adult soil ingestion rate: $R_{oral\text{-}a} = 100$ mg/d	Children soil ingestion rate: $R_{oral\text{-}c} = 100$ mg/d
Dermal	Children: $R_{der-c} = A_{skin-c} \times AF_c \times n \times ABS_d$ Adult: $R_{der-a} = A_{skin-a} \times AF_a \times n \times ABS_d$ Where:A_{skin} is the exposure skin area for children (c) or adult (a), cm^2; AF is soil-to-skin adherence factor for children (c) or adult (a), mg/cm^2; n is the number of daily soil contact events, d^{-1}; ABS_d is the dermal absorption fraction, unitless.	$R_{der-c} = A_{skin-c} \times AF_c \times n \times ABS_d$
Inhalation of dust	Children: $R_{inh-c} = PM_{10} \times RF \times V_{inh-c} \times (po \times EFO_c + pi \times EFI_c)$ Adult: $R_{inh-a} = PM_{10} \times RF \times V_{inh-a} \times (po \times EFO_a + pi \times EFI_a)$ Where:RF is Retention fraction of inhaled particulates in body; V_{inh} is daily inhalation rate for children (c) or adults (a),m^3/d; p_o is the fraction of soil-borne particulates in outdoor air, unitless; p_i is the fraction of soil-borne particulates in indoor air, unitless; PEF is particulate emission factor, m^3/kg; T_{site} is exposure time of indoor (i) and outdoor (o), h/day; TF is the soil-to-dust transport factor according to soil type, g/g dw; DL is the indoor dust loading factor, g/m^3.	Outdoor: $R_{inh-c} = \frac{1}{PEF} \times V_{inh-c} \times \frac{T_{site-o}}{24}$ Indoor: $R_{inh-c} = \left(\frac{1}{PEF} + TF \times DL\right) \times V_{inh-c} \times \frac{T_{site-i}}{24}$
Inhalation of vapors from outdoor surface soil	$R_{inh} = C_s \times VF_{sur} \times (V_{inh-c} + V_{inh-a})$ $VF = \frac{\rho_b}{DF_{oa}} \times \sqrt{\frac{4 \times D_{eff} \times H'}{\pi \times \tau \times 31536000 \times K_{sw} \times \rho_b}}$ $DF_{oa} = \frac{U_{air} \times W \times \delta_{air}}{A}$ Where:VF_{sur} is the volatilization factor from soil to outdoor ambient air, kg/m^3; C_s is the total contaminants concentration in soil, mg/kg; D_{eff} is the effective chemical diffusion coefficient in soil, cm^2/s; H′ is the air-water partition coefficient at ambient temperature, cm^3/cm^3; ρ_b is the bulk soil density, g/cm^3; ρ_s is the dry bulk soil density, g/cm^3; τ is the averaging time for surface emission vapor flux, y; K_{sw} is the total soil-water partition coefficient, cm^3/g; Q/C_{wind} is the air dispersion factor, g/cm^2 per kg/m^3; U_{air} is the ambient air velocity in mixing zone, cm/s; W is the width of source-zone area, cm; A is the source-zone area, cm^2; δ_{air} is the mixing zone height, cm; T_{site} is the outdoor site occupancy period, h/d.	$R_{inh} = C_S \times VF_{sur} \times V_{inh-c} \times \left(\frac{T_{site}}{24}\right)$ $VF = \frac{\rho_s}{\frac{1}{10} \times Q/C_{wind}} \times \sqrt{\frac{4 \times D_{eff} \times H'}{\pi \times \tau \times 31536000 \times K_{sw} \times \rho_s}}$
Inhalation of vapors indoor air	$R_{inh} = C_s \times VF_{ind} \times (V_{inh-c} + V_{inh-a})$ Where:VF_{ind} is the volatilization factor from subsurface soil to indoor, kg/m^3; C_{vap} is the soil vapor concentration, mg/m^3; α is the attenuation factor, unitless.	$R_{inh} = C_{vap} \times \alpha \times V_{inh-c}$

(*continued on next page*)

Table 1 (*continued*)

Item	China	UK
Inhalation of vapors from outdoor subsurface soil	$R_{inh} = C_s \times VF_{ind} \times (V_{inh-c} + V_{inh-a})VF_{suboal} = \frac{1}{\left(1 + \frac{DF_{oa} \times L_s}{D_s^{eff}}\right) \times \frac{K_{sw}}{H'}}$	–

crops ingestion). The SSVs of these 6 substances in China and the UK were calculated by the methods mentioned above and the results are shown in Table 2. It can be seen that for the non-carcinogenic substances (Cd, toluene), the difference of SSVs in China and the UK is relatively small: Cd (China: 20 mg/kg, UK: 18 mg/kg; toluene (China: 1200 mg/kg, UK: 3000 mg/kg). For carcinogens (As, Cr (VI), benzene, ethylbenzene)), the differences are large: As (China: <1 mg/kg, UK: 35 mg/kg), Cr (VI) (China: <1 mg/kg, UK: 14 mg/kg), ethylbenzene (China: 7 mg/kg, UK: 930 mg/kg). Due to the difference of hazard identification, the SSVs derived for ethylbenzene highlight major differences. This indicates that volatiles the contribution of inhalation via vapor pathway significantly reduces the differences caused by other elements, especially the inhalation of vapors indoor air, which can be inferred from the exposure pathway contribution showed in Fig. 2. The different calculation procedures, especially for carcinogens, make the SSVs under the current conditions of China and UK differ substantially.

3.2. Effect of parameters on SSVs

To further understand the impact of parameter values on the SSVs between the two countries, the CLEA model was run with Chinese input parameters to calculate screening values of these six chemicals. These values can be compared with the SSVs calculated in section 3.1, to analyse the differences caused by parameters in China and UK. Details of the Chinese parameter inputs to replace default values are summarized in Tables 3 and 4. The calculated results are shown in Table 2 (as 'China in the CLEA model'). It can be seen that SSVs calculated in this way are very close to the UK SSVs. The influence of the parameters of SSVs in UK and China is far less than the impact of the formulation method. However, it is meaningful to find the parameters that cause the difference between the

Table 2
Soil screening values of selected chemicals derived from different models (mg/kg).

Chemical	Toxicity	China	UK	China in CLEA Model
Arsenic	Carcinogenic	<1[a]	35	20
Cadmium	Non-Carcinogenic	20	18	11
Chromium (VI)	Carcinogenic	<1[a]	14(C4SL child)	29(C4SL child)
Benzene	Carcinogenic	1	1.1	1.6
Toluene	Non-Carcinogenic	1200	3000	3800
Ethylbenzene	Carcinogenic (China) Non-Carcinogenic (UK)	7.2	930	1200

Note:
[a] Since the soil screening value of As calculated by the Chinese model is < 1, the soil screening value of 20 mg/kg of As released in China is the result of replacing the soil environmental background value, which is not the calculated value of the model. In order to make this study comparable, the screening value of As in China is still calculated by the model.

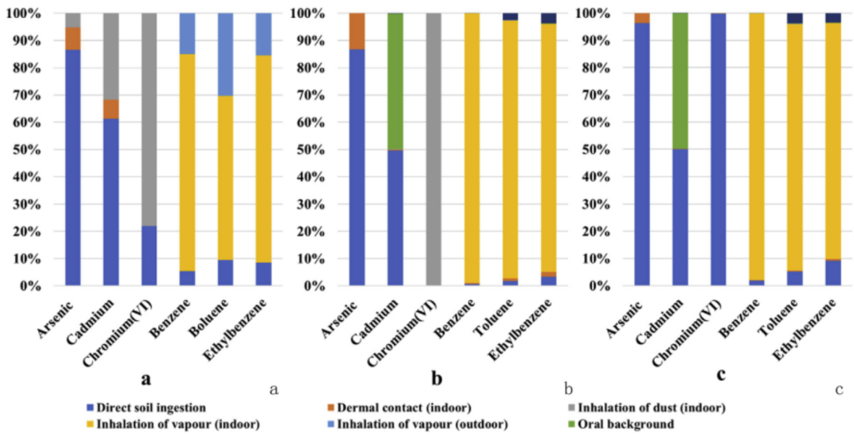

Fig. 2. Exposure pathway contribution in different derivation methods of SSVs. (a: China, b: UK in CLEA model, c: China in CLEA model).

Table 3
Land use and receptor parameters of China input into the CLEA model.

Land use	Unit	Age class					
		1	2	3	4	5	6
EF (soil and dust ingestion) [a]	day yr^{-1}	350	350	350	350	350	350
EF (consumption of homegrown produce)[a]	day yr^{-1}	0	0	0	0	0	0
EF (dermal contact, indoor) [a]	day yr^{-1}	350	350	350	350	350	350
EF (dermal contact, outdoor)[a]	day yr^{-1}	350	350	350	350	350	350
EF (inhalation of dust and vapor, indoor)[a]	day yr^{-1}	262.5	262.5	262.5	262.5	262.5	262.5
EF (inhalation of dust and vapor, outdoor)[a]	day yr^{-1}	87.5	87.5	87.5	87.5	87.5	87.5
Occupancy Period (indoor) [b]	hr day^{-1}	22.3	21.5	21.3	21.3	21.5	21.5
Occupancy Period (outdoor)[b]	hr day^{-1}	1.7	2.5	2.6	2.5	2.2	2.2
Soil to dermal adherence factor (indoor)[d]	mg cm^{-2} day^{-1}	2.0E-01	2.0E-01	2.0E-01	2.0E-01	2.0E-01	2.0E-01
Soil to dermal adherence factor (outdoor)[d]	mg cm^{-2} day^{-1}	2.0E-01	2.0E-01	2.0E-01	2.0E-01	2.0E-01	2.0E-01
Soil and dust ingestion rate[a]	g day^{-1}	2.0E-01	2.0E-01	2.0E-01	2.0E-01	2.0E-01	2.0E-01
Receptor		**Age class**					
		1	**2**	**3**	**4**	**5**	**6**
Body weight[b]	kg	7.9	10.8	13.2	15.3	17.4	19.2
Body height[c]	m			1	1.06	1.12	1.18
inhalation rate	m^3 day^{-1}	4.6	5.4	6	7.6	8.1	8.4
Max exposed dermal fraction (indoor)[d]	m^2 m^{-2}	0.14	0.18	0.22	0.24	0.27	0.28
Max exposed dermal fraction (outdoor)[d]	m^2 m^{-2}	0.14	0.18	0.22	0.24	0.27	0.28

Note:
[a] Refer to the values in the "Technical Guidelines for Risk Assessment of Contaminated Sites (HJ25.3–2014) (Ministry of Environmental Protection of the People's Republic of China, 2014)".
[b] "Exposure factors handbook of Chinese Population (0–5 years old), (6–17 years old) Ministry of Environmental Protection of the People's Republic of China, 2013.".
[c] "National Physical Fitness Monitoring 2014" (http://www.sport.gov.cn/n16/n1077/n1422/7331093.html).
[d] Refer to the default values in the CLEA model.

Table 4
Soil and building properties of China input into the CLEA model.

Soil properties for	value	Unit
Porosity, total[b]	0.43	cm^3 cm^{-3}
Porosity, air-filled[a]	0.30	cm^3 cm^{-3}
Porosity, water-filled[a]	0.13	cm^3 cm^{-3}
Residual soil water Content[a]	0.20	cm^3 cm^{-3}
Saturated hydraulic conductivity[a]	7.90E-05	cm s^{-1}
van Genuchten shape parameter (*m*)[b]	3.20E-01	dimensionless
Bulk density[a]	1.50	g cm^{-3}
Threshold value of wind speed at 10m[a]	2.00	m s^{-1}
Empirical function (Fx) for dust model[b]	1.22	dimensionless
Ambient soil temperature[a]	298	K
Building properties for		
Building footprint[a]	9.00E+00	m^2
Living space air exchange rate[a]	0.5	hr^{-1}
Living space height (above ground)[a]	2.2	m
Living space height (below ground)[a]	0.0	m
Pressure difference (soil to enclosed space)[b]	3.1	Pa
Foundation thickness[a]	3.50E-01	m
Floor crack area[a]	2.40E+01	cm^2
Dust loading factor[b]	5.00E+01	μg m^{-3}
Air dispersion model		
Mean annual windspeed (10m)[a]	2.00	m s^{-1}
Air dispersion factor at height of 0.8m[b]	2400	g m^{-2} per kg m^{-3}
Air dispersion factor at height of 1.6m[b]	0.00	g m^{-2} per kg m^{-3}
Fraction of site with hard or vegetative cover	0.00	m^2 m^{-2}
Vapor model		
Default soil gas ingress rate	–	cm^3 s^{-1}
Depth to top of source (beneath building)[a]	100	cm
Depth to top of source (no building)	0	cm
Time average period for surface emissions[a]	6	years
User defined effective air permeability[a]	1.00E-08	cm^2

Note:
[a] Refer to the values in the "Preparation instructions for "soil environment quality risk control standard for soil contamination of development land (Trial)" (Environmental Agency, 2009c).
[b] Refer to the default values in the CLEA model.

two countries to further reduce the difference of SSVs in China and UK.

4. Discussion

4.1. The reasons for differences of SSVs in China and UK

The SSVs calculation for China referred to the toxicity assessment of the US EPA's Integrated Risk Information System (IRIS), with SSVs calculated for carcinogens according to equation (1) in 2.1 and an acceptable cancer risk level of 10^{-6}, while non-carcinogens are calculated according to equation (2) in 2.1 with a hazard quotient of 1. However, in the UK CLEA model, SSVs are calculated for carcinogens and non-carcinogens using equation (3) in 2.1. The effect of these differences on SSV toxicity assessment and risk characterization can be seen from the calculation results of the selected chemicals in Section 3.1. Although both China and UK identified As as a carcinogen (in the UK, the oral and inhalation ID of As associated with a minimum excess risk of cancer were determined, in which the oral intake ID is based on British drinking water standards. The risk is equivalent to the cancer risk of around 40 to 400 in 100,000 (Environmental Agency, 2009a). If the equations, toxicity values and other parameters (such as daily soil intake rate, exposure frequency, exposure duration, etc.) are not the same, the calculated result in China was <1 mg/kg and in the UK was 35 mg/kg. If the CLEA model was used with Chinese input parameters, the toxicity values and equations were unified; the As SSV for China was of the same order of magnitude as the UK and very close to the Chinese As soil background value. The result was also the same for Cr (VI). However, for the non-carcinogens, (Cd and toluene), the SSVs were very similar. In contrast, the SSVs derived for ethylbenzene (which is identified as a carcinogen in China and a non-carcinogen in the UK) were very different, because of the differences in toxicity assessment and risk characterization (see Table 5).

So, in conclusion, the toxicity values and risk characterization methods were the factors most affecting the SSV differences between China and the UK, especially for carcinogens, while choices in model parameters has only a minor effect.

Table 5
Toxicity values of selected chemicals in China and UK.

Chemical	China				UK CLEA Model			
	SF_o mg/kg-d	IUR $(ug/m^3)^{-1}$	RfD_o mg/kg-d	RfC_i mg/m^3	TDI_o ug/kg-d	ID_o ug/kg-d	TDI_i ug/kg-d	ID_i ug/kg-d
Arsenic	1.5E+00	4.3E-03				3.0E-01		2.0E-03
Cadmium			1.0E-03	1.0E-05	3.6E-01		1.4E-03	
Chromium (VI)	5.0E-01	8.4E-02				4.4E-01		3.4E-04
Benzene	5.5E-02	7.8E-06				2.9E-01		1.4E+00
Toluene			8.0E-02	5.0E+00	2.23E+02		1.4E+03	
Ethylbenzene	1.1E-02	2.5E-06			1.0E+02		2.2E+02	

Note: SF_o is oral slope factor; IUR is inhalation unit risk; RfD_o is oral reference dose; RfC_i is inhalation reference concentrations; TDI_o and TDI_i is tolerable daily intakes of oral (o) and inhalation (i); ID_o and ID_i is index dose of oral (o) and inhalation (i).

4.2. The identification of key parameters affecting on SSVs

From Table 2, it can be seen the SSVs calculated by substituting Chinese parameters into the CLEA model are still different from the SSVs in UK, indicating that the parameters also contribute to the differences of SSVs in China and UK. Therefore, analysis and identification of key parameters can help us to understand the differences of SSVs in China and UK. Here, we determine the important parameters of the difference of SSVs in China and UK by calculating the contribution rate of each exposure route of different substances to SSVs. Firstly, the exposure route with the highest contribution rate to SSVs is determined, and then the parameters that have a greater influence on the exposure route are analyzed to determine the important parameters affecting the difference of SSVs in China and UK. Fig. 2 summarises the calculated contributions of the different exposure pathways for each of the selected chemicals, under the 3 scenarios. For the heavy metals, the contributions of the oral ingestion, dermal contact and inhalation of dust pathways is >80%, for each method of SSV derivation. However, for the VOCs, inhalation of vapors from the indoor air pathway dominated (see Fig. 2). On the other hand, for SSVs of heavy metals (except Cr), the contribution of imported intake (not distinguishing from background intake) is greater than 50%, which is the most influential exposure pathway, so the effect of parameters of oral exposure pathways are important for heavy metal screening. The value for oral soil ingestion is 200 mg/kg in China, 100 mg/kg in the UK. The SSVs of heavy metal (except Cr) calculated by the CLEA model in China is reduced by half. So, for heavy metals, we consider that the key parameter affecting the screening value of China and the UK is the oral daily soil intake. However, for volatile organic compounds, it is not very different from SSVs in UK and China calculated by using Chinese parameter into the CLEA model. China is between 1.2 and 1.4 times higher than the UK value, indicating there is less impact of the parameters on the VOC SSVs is less than for heavy metals. In addition, the contribution rate of indoor respiratory exposure to the SSVs of volatile organic compounds is greater than 50%, which is the most important route. From the comparative analysis in Section 2.3.2.1, the attenuation factor α is the main parameter of the SSVs for indoor air volatiles in the CLEA model, so the parameters involved in α also have a certain influence.

It should be stressed that these assessments are generic conditions; in real environmental circumstances, site-specific and person/individual specific differences can vary considerably and need to be captured in site- and population/community-specific scenarios (Claudio et al., 2007).

5. Conclusion

Based on the above discussion, we can conclude that the differences in SSVs between China and the UK are mainly reflected in the aspects of toxicity assessment and risk characterization methods, exposure assessment and differences in parameters. Among them, toxicity assessment and risk characterization are important factors that cause differences in soil screening values. They not only determine the type of hazardous effect and the toxicity values, but also determine the characterization method of risks. The compared results indicate that differences in toxicity assessment and risk characterization can cause large differences in SSVs without considering the effects of other factors. Although there are some differences in exposure assessment, the model structure and parameter type used in the exposure assessment of the two countries are very similar. The biggest difference is mainly in the model of inhalation pathway. For VOCs, the contribution rate of inhalation pathways is much higher than others, and the contribution of this pathway significantly reduces the differences caused by other factors. The difference of the parameters also has some impact on the screening values, but the degree of influence is relatively weak. For example, although the SSVs of As and Cd in China and UK calculated by the CLEA model using Chinese parameters are of the same order, the SSVs in China are smaller than those for the UK. In the future, the key to reducing the difference of SSVs between China and UK is to increase consistency in toxicity assessment, risk characterization and exposure assessment models. In particular, China should implement toxicity assessment and risk characterization studies based on its own conditions due to the large variation of derived SSVs compared with soil background value, such as arsenic, and an in-depth study of the exposure assessment model for the indoor inhalation of volatiles should also be conducted. Under the current conditions, human health risk assessment has been applied to the risk assessment of pollutants in different environmental media (air, water and soil). Due to differences of environmental media, there may be some differences in behavioural patterns and exposure assessment to different environmental media. . However, China and UK did not have an in-depth analysis of this issue, nor did it conduct an in-depth study of the uncertainty of the soil exposure assessment model. Therefore, the two countries should further study the human exposure behavior to soil and have certain revise in exposure assessment models, in order to reduce the uncertainty caused by the inadequate understanding of the exposure.

Acknowledgement

We are grateful to Dr Lisa Norton and Dr Aidan Keith in Centre for Ecology & Hydrology-Lancaster for their contribution to the knowledge of the soil risk assessment in UK. The authors are grateful for funding from the National Natural Science Foundation of China (Grant no. 41571311) and the National High-tech R&D Program (863 Program) (No. 2013AA06A206).

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Appendix 3

Total human environmental exposure to selected heavy metals: A national study for China

Xiuge Zhao[a]*, Danlu Wang[a], Bin Zou[c], Jian Xu[a], Limin Lei[b], Zhenglei Li[b], Yan Tao[b], Yiming Sun[d], Hao Zhang[d,] Kevin C Jones[d]*, Feiyang Qiao[b], Ju Huang[b]

a. State Key Laboratory of Environmental Criteria and Risk Assessment, Chinese Research Academy of Environmental Sciences, Beijing 100012, China

b. College of Earth and Environmental Sciences, Lanzhou University; Key Laboratory of Environmental Pollution Prediction and Control, Gansu Province， Lanzhou 730000, China

c. School of Geosciences and Info-Physics, Central South University, Changsha 410086, China

d. Lancaster Environment Centre, Lancaster University, Lancaster, LA1 4YQ, UK

Corresponding authors:

Name: Xiuge Zhao, E-mail: hkyzhaoxg@163.com

Address: State Key Laboratory of Environmental Criteria and Risk Assessment, Chinese Research Academy of Environmental Sciences, Beijing 100012, China

Name: Kevin Jones, E-mail: k.c.jones@lancaster.ac.uk

Address: Lancaster Environment Centre, Lancaster University, Lancaster, LA1 4YQ, UK

Acknowledgments: The research leading to these results has received funding from the Ministry of Ecology and Environment of the People's Republic of China (EH [2017]-15-14). We thank all the volunteers and participants contributing to this study in Dalian Ocean University, East China University of Science and Technology, Gansu Academy of Eco-environmental Sciences, Huazhong University of Science and Technology, Shanxi University and Sichuan University.

Conflict of interest: The authors declare no conflict of interest.

Abstract:

BACKGROUND: Heavy metals are a public health concern that have sub-chronic and chronic effects on human health. The Global Burden of Disease study concluded that 9% of all deaths are due to environmental pollution, with heavy metals accounting for 11% of those.

OBJECTIVES: This study was designed to quantify current human exposure to 5 key heavy metals – arsenic, cadmium, chromium, lead and mercury – for the Chinese population, examining differences with age, gender, urban/rural locations in representative regions of China and dietary composition. The main routes of exposure, via diet and drinking water ingestion, inhalation and skin contact were quantified. We derive these estimates for different regions of China and assess the health risks.

METHODS: We conducted an extensive field and analytical campaign (>4100 samples) to make the most reliable estimates to date of human exposure to As, Cd, Cr, Hg and Pb from environmental and exposure media (air, water, soils, diet) in China. Volunteers participated to provide survey data and duplicate diet samples.

RESULTS: The average total human environmental exposure mg/(kg·d) in China to the 5 metals decreased in the order of Cr (3.8×10-3) > As (1.4×10-3) > Pb (8.4×10-4) > Cd (1.2×10-4) > Hg (4.7×10-5). Dietary exposure contributed most to the total environmental exposure of Hg, Cd, As, Pb and Cr (up to 94%, 97%, 88%, 95% and 94%, respectively). Total human environmental exposure (per kg body weight) was higher for females than males. Residents aged 60 and above had the lowest exposures.

CONCLUSIONS: The non-carcinogenic risk levels for the general population of China are all within the maximum acceptable level and the carcinogenic risk to Cr, As and Pb are all below the acceptable levels of 10-4.

Keywords: dietary; exposure route; health risk; metal; non-dietary; total exposure

1. Introduction

Heavy metals are a public health concern that have sub-chronic and chronic effects on human health (Wang et al. 2001; Glorennec et al. 2016; Soomro et al. 2019). The Global Burden of Disease study concluded that 9% of all deaths are due to environmental pollution, with heavy metals accounting for 11% of those (Landrigan et al. 2018). Most concern for the general population centres on mercury (Hg), cadmium (Cd), arsenic (As), lead (Pb) and chromium (Cr). Mercury can damage the central nervous system and increases the prevalence of cardiovascular disease, especially in adults (Mozaffarian 2009; Rice 2004; Yaginuma-Sakurai et al. 2010). Cadmium can affect the bone and skeletal system (Akesson et al. 2006), accumulates in the cardiovascular system (Schutte et al. 2008), and can damage liver and kidney function (Wallin et al. 2013). Arsenic can affect development, and the nervous, respiratory, and cardiovascular systems; it can also cause skin lesions and skin cancer (ATSDR 2016; Naujokas et al. 2013; Yoshida et al. 2004). Lead causes harmful effects on the nervous, renal and endocrine systems (Navas-Acien et al. 2007; Weinhold 2004). Chromium is an essential element for maintaining healthy insulin activity (Wu and Li 2015), but may cause lung cancer, skin ailments and teratogenic effects (Jena and Singh 2017) when exposure is too high.

Humans are continuously exposed to heavy metals, via multi-media transfers from the air, water, soil and diet. The different exposure routes are via inhalation, ingestion and dermal contact (Huang et al. 2018; Li et al. 2011; Ruby and Lowney 2012; Yu et al. 2017; Dong and Hu 2011; Glorennec et al. 2016; James and Meliker 2013; Lucas et al. 2012; Zhuang et al. 2014). A full understanding of human exposure and health risks from metals requires the measurement of environmental concentrations, dietary intakes and assessment of multiple exposure routes (Lioy et al. 1988). Full human health risk assessment should estimate the nature and probability of adverse health effects for people who may be exposed now or in the future (NRC 1983; Ott, 1990). To inform controls and policy, it is important to be able to quantify the contribution of different media and routes of exposure (Kennedy et al. 2015; Dong and Hu 2011)). The US EPA conducted the first national total exposure study in 1979 (Lioy et al. 1988). Other countries, including Japan (Aung et al. 2004; Kawabe et al. 2003; Ohno et al. 2010) and Korea (Lee et al. 2003) have also conducted total exposure studies. Dietary exposure is generally shown to contribute >50% of the total exposure to metals; differences in dietary sources, patterns, food preparations and behavior will result in different exposures within and between countries (Cubadda et al. 2017; Lee et al. 2003; Wu and Li 2015; Glorennec et al. 2016; Kurzius-Spencer et al. 2014; Yeganeh et al. 2012).

There are some differences between exposure levels and health risk levels of the various metals. A study showed that the ingestion route of As, Cr contributed more than 85% of total exposure levels, but the ingestion routes of the non-cancer risk of Cr and the carcinogenic risk of As contributed 41% and 11% of total non-cancer risk and carcinogenic risk, respectively (Cao et al. 2014). Even though the concept of total human exposure was proposed many years ago, most studies on exposure and health risk assessment of heavy metals have been focused on a single exposure medium or route, and on residents living near industrial enterprises with metal emissions (Cai et al. 2015; Cao et al. 2014; Cubadda et al. 2017). These studies seldom target the total exposure of the population through multiple media and routes or for many metals. In China and many other rapidly developed/ing countries, the environment has suffered from widespread and diffusive emissions of heavy metals during rapid industrialization and urbanization, resulting in enrichment of metals in environmental media. Responsible national management makes it necessary to carry out representative surveys, monitoring, assessment and research, to provide key points of reference for health risk prevention and control. This study, funded by the Chinese Ministry of Ecology and Environment, is part of a major commitment to environmental and public health management in China.

China is a huge country, spanning very different natural environments, degrees of industrialization and urbanization, and people with widely different habits, diets, behaviour and cooking habits. We therefore:

Used questionnaire survey and individual track monitoring methods, to study total human exposure of Hg, Cd, As, Pb and Cd for residents living in 6 typical areas of China;

Quantified non-dietary exposure (via indoor air, outdoor air, traffic air, water and soil);

Quantified dietary exposure;

Estimated the aggregate non-cancer and carcinogenic health risk levels for non-dietary and dietary exposure through all the exposure routes.

The project was a major undertaking, involving analysis of 4156 samples and participation of ~3855 volunteers and 90 collaborative scientists.

The results of the total human exposure assessments in this study give key national reference data for China. It will support development and revision of environmental health standards, and the formulation of pollution/exposure control policies for China and other countries.

2 Materials and methods

2.1 Ethics Statement

This research was approved by Institutional Review Boards (IRB) of Public Health in Tongji Medical College of Huazhong University of Science and Technology. All participants were informed about the objectives and methods of the study before the investigation. Written consents were obtained from all participants.

2.2 Study areas and basic design

The study locations were selected to be representative of general conditions, across different geographical, social, economic and residential areas; different dietary patterns are represented by the regions selected, which were away from any known specific hot-spots of heavy metal contamination. Taiyuan, Shanghai, Wuhan, Dalian, Lanzhou and Chengdu were chosen as the study areas. They are, respectively: an inland coal-powered city (Taiyuan; population 4.34 M); a coastal industrial city (Dalian; 6.99 M); a Tier-1 city and business-oriented metropolis (Shanghai; 24.20 M); a port city on a major river artery (Wuhan; 10.77 M); an industrial city located in an inland basin (Chengdu; 3.71 M) and a dusty city located in an inland arid regions (Lanzhou; population 3.71 M). According to the mainland provinces classification, Taiyuan, Dalian and Lanzhou are located in the North, Northeast and Northwest, respectively. They represent the Northern area resident dietary patterns, with generally higher intake of staple foods and a slightly lower the intake of vegetables than in southern area; wheat is the main staple food. Shanghai, Wuhan and Chengdu are located in the East, South and Southwest, respectively. They represent the southern area resident dietary pattern, with a higher intake of meat, aquatic products and vegetables than in the northern areas; rice is the main staple food of the southern area.

Healthy adults aged 18 and above were selected randomly, but with the requirement that they should be from different households and resident at the same place for >6 months. A total of 3855 validated surveys were collected from volunteers, distributed as follows; 1982 urban and 1873 rural residents; 1862 males and 1993 females; 1587 young adults (18-44 years old), 1119 middle-aged adults (45-59) years old, 1149 older adults (60 years old and above). About 10% (382) of the subjects were selected to participate in monitoring of typical exposure scenario. This covered information on location, drinking water types, fuel/heating types etc. Exposure samples of indoor air, outdoor air, traffic air, drinking water, soil, and diet were collected for metals analysis.

2.3 Sample collection and analysis

2.3.1 Questionnaire survey

Information was obtained about the volunteers by questionnaire and face-to-face surveys. Data included: body weight, height, skin surface area, intake rates (dietary, water and inhalation), time-activity factors and other residential factors.

2.3.2 Field sampling

Air (i.e. indoor, outdoor, traffic), drinking water, soil and dietary samples related to personal exposure were collected. The indoor air samples were collected with mini air samplers (BUCK PL-5, AP Buck Inc., USA) with quartz filters (37mm, Whatman In C, USA, all filters treated at 400 °C, 6 h) at a flow rate of 2 L/min. Sampling was for >60 hours over 3 days, including a weekend. The outdoor and traffic air samples were collected with a mid-volume air sampler (TH-150 series, Wuhan Tianhong Instruments Co., Ltd., China) with a quartz filter (90mm, Whatman Inc., USA) at 100L/min. In total, 583 indoor, 874 outdoor and 271 traffic air samples were collected. They were then stored at <4 °C before preparation for analysis. Sampling followed the national protocols detailed in Technical Specifications for Monitoring of Indoor Air Quality (HJ/T 167-2004), Technical regulation for selection of ambient air quality monitoring stations (on trial) (HJ 664-2013) and Manual methods for ambient air quality monitoring (HJ/T 194-2005).

Drinking water samples were collected from the household tap or vat used most frequently by the volunteer. Samples were collected into pre-cleaned polyethylene bottles (HNO3 added as preservative) and stored at <4°C before analysis. In total 1065 samples were collected, according the national Standard examination methods for drinking water-Collection and preservation of water samples (GB/T 5750.2-2006).

Soil samples (0-20 cm) were collected close to the main resident areas or nearby farmland. In total 146 samples were collected, following the national standard of Soil Testing Part1: Soil sampling, processing and reposition (NY/T 1121.1-2006).

Diet samples were collected, using the duplicate diet method for 3 daily meals. Each sample was uniformly mixed from the daily meal samples, freeze-dried and placed in cold storage before analysis. A total of 1217 diet samples were collected following the Procedural regulations regarding monitoring of pollutants in the produce of agriculture, animal husbandry and fishery (NY/T398-2000) and Dietary survey method-Part2: Weighting method (WS/T 426.2-2013).

2.3.3 Sample treatment and analysis

Air samples were digested with HNO3-HCl, using a microwave digestion system. Hg and As were determined by an atomic fluorescence method (national standard Ambient air-Determination of mercury and its compounds-Cold atomic fluorescent (HJ 542-2009)); Cd, Pb and Cr were determined by ICP-MS (national standard of Ambient air and stationary source emission-Determination of metals in ambient particulate matter-Inductively coupled plasma/mass spectrometry (ICP-MS) (HJ 657-2013).

Drinking water samples were passed through a 0.45μm filter before analysis. Hg and As were detected by atomic fluorescence (national standard Water Quality-Determination of Mercury, Arsenic, Selenium, Bismuth and Antimony-Atomic Fluorescence Spectrometry (HJ694-2014); Cd, Pb and Cr were determined by ICP-MS (Standard examination methods for drinking water-Metal parameters (GB/T5750.6-2006)).

Soil samples were air-dried, disaggregated with an agate mortar, then sieved (100 mesh, pore size 0.149 mm). A uniform powder was obtained, then 0.1g weighed and treated with HNO3-HCl in a microwave digestion system. Hg and As were determined by atomic fluorescence according to U.S. EPA Method 200.8 Revision 5.4, and Cd, Pb and Cr determined by ICP-MS according to the national standard Soil and sediment-determination of mercury, arsenic, selenium, bismuth, antimony -Microwave dissolution/Atomic Fluorescence Spectrometry (HJ 680-2013).

The diet samples were freeze-dried, disaggregated with an agate pestle and mortar (100 mesh, pore size 0.149 mm), weighed (0.5g) and digested with HNO3 in a microwave digestion system. Hg and As were determined according to the National food safety standards-Determination of total mercury and organic mercury in food(GB5009.17-2014) and National food safety standards-Determination of total arsenic and inorganic arsenic in food(GB5009.11-2004), and Cd, Pb and Cr were determined following the National food safety standards-Determination of multiple elements in food (GB5009.268-2016).

Certified reference material and reagent blanks were included in each digestion batch and used for quality control of the analysis process.

2.4 Exposure assessment

2.4.1 Air

Air exposure assessment considered inhalation of indoor, outdoor and traffic air using the following equation:

$$ADD_{air}=\frac{C_a \times InhR \times ET \times EF \times ED}{BW \times AT} \quad (1)$$

Where: ADD_{air} is the average daily dose (mg/(kg·d)) of an element via the inhalation of air; C_a is the air concentration (mg/m^3), InhR is the inhalation rate (m^3/h), ET is the exposure time (h/d), EF is the exposure frequency (d/year), ED is the exposure duration (year), BW is the body weight (kg) and AT is the average time (d). The InhR was calculated from the BW and height using an inhalation rate model (USEPA 2011). The ET and BW were collected from the human behavior pattern questionnaire. The correlation exposure factors are given in Tables S1 and S2.

2.4.2 Water

This considered exposure through ingestion and dermal contact routes using the following equations:

$$ADD_{water}=ADD_{w\text{-}oral}+ADD_{w\text{-}dermal} \quad (2)$$

$$ADD_{w\text{-}oral}=\frac{C_w\times IngR_w\times EF\times ED}{BW\times AT} \quad (3)$$

$$ADD_{w\text{-}dermal}=\frac{C_w\times SA_w\times PC\times CF_w\times ET\times EF\times ED}{BW\times AT} \quad (4)$$

Where $ADD_{w\text{-}oral}$ is the average daily dose (mg/(kg·d)) of an element via the ingestion of water and ($ADD_{w\text{-}dermal}$) via dermal contact of water. C_w is the water concentration (mg/L), $IngR_w$ is the ingestion rate of water (L/d), SA_w is the surface area of the skin exposed to pollutants of water (cm^2), PC is the dermal absorption factor (cm/h) and CF_w is the volume conversion factor ($1L/1000cm^3$). The SA_w was calculated from the body weight and height, using the body surface model (USEPA 2011). The $IngR_w$, ET and BW were collected from the human behavior pattern survey in this study. Correlation exposure factors are given in Tables S1 and S2.

2.4.3 Soil

Soil exposure assessment considered inhalation, ingestion and dermal contact. The daily dose via total soil exposure daily dose was derived from the following equations:

$$ADD_{soil}=ADD_{s\text{-}dermal}+ADD_{s\text{-}oral}+ADD_{s\text{-}inh} \quad (5)$$

$$ADD_{s-inh}=\frac{C_s\times InhR\times EF\times ED}{PEF\times BW\times AT} \quad (6)$$

$$ADD_{s\text{-}oral}=\frac{C_s\times IngR_s\times CF_s\times FI\times EF\times ED}{BW\times AT} \quad (7)$$

$$ADD_{s\text{-}dermal}=\frac{C_s\times CF\times SA_s\times AF\times ABS_d\times EF\times ED}{BW\times AT} \quad (8)$$

Where ADD_{soil} is the total average daily dose (mg/(kg·d)) of an element from soil, the sum of $ADD_{s\text{-}dermal}$, $ADD_{s\text{-}oral}$ and $ADD_{s\text{-}inh}$. C_s is the soil concentration (mg/kg), PEF is the particulate emission factor (m^3/kg), $IngR_s$ is the ingestion rate of soil (mg/d), CF_s is the conversion factor (10^{-6}), FI is the digestive tract absorption factor (dimensionless), SA_s is the surface area of the skin exposed to pollutants of soil (in cm^2/event), AF, the skin adherence factor (mg/cm^2) and ABS_d is the dermal absorption factor (dimensionless). The SA_s was calculated the same way as SA_w. Values for $IngR_s$ (MEE 2013), FI, AF (MEE 2014) and ABS_d (USEPA 2004) were collected from the literature and technical data. Further details are given in Tables S1 and S2.

2.4.4 Dietary exposure assessment

This was calculated as:

$$\mathrm{ADD_{food}}=\frac{\mathrm{C_f \times IngR_f \times FI \times EF \times ED}}{\mathrm{BW \times AT}} \qquad (9)$$

Where ADD_{food} is the average daily dose (mg/(kg·d)) of an element via dietary ingestion, C_s is the element concentration in the diet (mg/kg), $IngR_f$ is the dietary ingestion rate (mg/d), FI is the digestive tract absorption factor (dimensionless) (see also Tables S1 and S2).

The total daily dose (ADD_{total}) is then:

$$\mathrm{ADD_{total}=ADD_{air}+ADD_{water}+ADD_{soil}+ ADD_{food}} \qquad (10)$$

2.5 Risk calculation

2.5.1 Non-cancer risk

The hazard quotient (HQ) for lifetime non-cancer risk can be calculated by dividing the ADD from each exposure route by a reference dose (RfD) and the hazard index (HI), which is the sum of HQ for each route. The HQ and HI are defined as follows (USEPA 1989):

$$\mathrm{HQ}=\frac{\mathrm{ADD}}{\mathrm{RfD}} \qquad (11)$$

$$\mathrm{HI}=\sum \mathrm{HQ_i} \qquad (12)$$

There are three thresholds (mg/(kg·d)): Rfd_i for inhalation, Rfd_o for ingestion, Rfd_d for dermal contact. In the case of HI>1, there may be concern for potential non-carcinogenic effects. If HI≤1, the assumption is that adverse health effects are unlikely to occur. The values of RfD_i, RfD_O and RfD_d are given in Table S2.

2.5.2 Cancer risk

The life-time cancer risk (LICR) is estimated as the incremental probability of an individual developing cancer over their lifetime due to exposure to a potential carcinogen. The LiCR is defined as (USEPA 1989):

$$\mathrm{LICR} = \mathrm{ADD} \times \mathrm{SF} \tag{13}$$

$$\mathrm{TLICR} = \sum \mathrm{LICR_i} \tag{14}$$

The slope factor (SF) has three thresholds ((kg·d)/mg): SF_i for inhalation, SF_o for ingestion, SF_d for dermal contact. They can estimate the increased cancer risk from a lifetime of inhalation, ingestion or dermal contact exposure to a substance, respectively. Risks in the range of 10^{-6}-10^{-4} are deemed 'acceptable' (USEPA 1991). SF_i, SF_O and SF_d values are given in Table S2.

2.6 Exposure contribution

The relative source contribution (RSC) (Howd et al. 2004) is the contribution of the total daily exposure to a chemical that is attributed to or allocated to different media or routes (accounting for multi-route exposures) in calculating acceptable levels of chemicals in total environmental exposure. It is defined as follows:

$$\mathrm{RSC_i} = \frac{\mathrm{ADD_i}}{\mathrm{ADD_{total}}} \times 100\% \tag{15}$$

2.7 Statistical analysis

Descriptive statistics, exposure and health risks for each metal are presented as means, medians and inter-quartile ranges. An alpha level of 0.05 was chosen to determine the significance.

3 Results and discussions

3.1 Metal concentration

Measured concentrations of Hg, Cr, As, Pb and Cd in air, water, soil and diet samples are presented in Figure S1 and Table S3. The metals concentrations in each medium were compared with the published national standards. It was found that the mean values of Hg, Cd and Pb in indoor air, outdoor air and traffic air were below annual mean thresholds and As was above the annual mean threshold of the Ambient air quality standards (GB2762-2012). Because of a lack of total Cr standard values in air, it was not possible to assess the relationship with standards. Our concentrations of Hg, Cr, As, Pb and Cd in water were all below the limits value of Standard for drinking water quality (GB5749-2006). Moreover, when compared to the risk intervention values for soil contamination of agricultural land in the Soil environmental quality control standards for soil contamination of agricultural land (GB15618-2018), concentrations of heavy metals did not exceed their target values. The metals concentrations in dietary samples were also all below the thresholds for metals in major food categories regulated by China Food Safety National Standard for Maximum Levels of Contaminants in Foods (GB2762-2012).

3.2 Daily exposures

3.2.1 Total daily exposure

In general, the total human environmental exposure levels of five metals decreased in the order Cr (3.76×10^{-3} mg/(kg·d)) >As (1.41×10^{-3} mg/(kg·d)) >Pb (8.45×10^{-4} mg/(kg·d)) >Cd (1.22×10^{-4} mg/(kg·d)) >Hg (4.72×10^{-5} mg/(kg·d)) (Table S4). They were all below the best estimate levels of the Joint FAO/WHO Expert Committee on Food Additives. The total human environmental exposure levels in this study are similar to the limited data reported for other countries, but there were some differences e.g. the exposure level of As was higher than Japan (4.10×10^{-4} mg/(kg·d)) (Kawabe et al. 2003) while Pb was lower than Korea (9.97×10^{-4} mg/(kg·d)) (Lee et al. 2003). In terms of different areas, the total environmental exposure of Hg and As was higher in Dalian than other areas ($P<0.05$), Cd and Pb exposures were higher in Chengdu than other areas ($P<0.05$), and Cr exposure was highest in Lanzhou ($P<0.05$).

Rural versus urban environmental exposure levels showed some interesting statistically significant ($P<0.05$) differences (Figure 1). The environmental exposure levels of Hg, Cd and Pb in rural areas were higher than urban areas, by 1.16, 1.06 and 1.17 times, respectively; As and Cr were lower in rural areas than urban areas (0.97 and 0.99 times), respectively. There were gender differences in environmental exposure levels for all the elements. Values for women (when expressed per unit body weight) were higher than for men ($P<0.05$) (PUT EACH ELEMENT NAME IN THE RIGHT ORDER PLEASE 1.16,1.16,1.19,1.13, and 1.11 times, respectively). Similar results have been seen for As in the US (Meacher et al. 2002).

Differences between age groups were also apparent for all the elements studied; young adults were the highest category (Table S4, Figure 1). Different age groups in different areas differed from each other, e.g. the As exposure level in Dalian of middle-aged adults was higher than others ($P<0.05$). The non-dietary (Figure S2, Table S5) and dietary (Figure S3, Table S6) exposure level sequence (i.e. Hg lowest, Cd, As, Pb and Cr highest) was the same as for total human exposure. Dietary exposure was much greater than non-dietary exposure (see below).

The dietary exposure levels of Hg, Cd and Cr found in this study were higher than those reported in the Fourth China total diet study; As and Pb values were similar in both surveys (Wu and Li 2015). Dietary exposure is highly related to the mix of food groups, food preparation methods and cooking styles which are typical for an area. In this study, the dietary exposure levels of Hg and As in Dalian were higher than in other areas ($P<0.05$), probably associated with the higher aquatic/seafood intakes in this area (Figure 2) (Wang and Wang 2014; Zhang et al. 2013).

Hg, Cd and Pb exposure decreased in order ingestion > inhalation > dermal contact; As and Cr exposure decreased in order ingestion > dermal contact > inhalation (Table S7). Ingestion was clearly the dominant exposure route for all 5 elements studied, as found in studies in the US (Kurzius-Spencer et al. 2014), the UK (Rowbotham 2000), France (Glorennec et al. 2016), Japan (Aung et al. 2004; Kawabe et al. 2003), Korea (Lee et al. 2003) and other studies in China (Cao et al. 2014; Cao et al. 2016).

3.2.2 Dominance of diet to the total exposure

As the previous section showed, the diet dominates total human exposure to the 5 elements studied. The dietary contribution averaged the following contributions to the total exposure (Hg - 94%, Cd - 97.5%, As - 87.8%, Pb - 95.4% and Cr - 94.4%, respectively (Figure 3, Table S8)). Combining diet and drinking water, the contribution of the ingestion route to the total environmental exposure levels followed the sequence Hg, Cd, As, Pb and Cr, at 99.99%, 99.96%, 97.83%, 99.764% and 99.91%, respectively (Figure 3, Table S9). The exposure contribution of the dietary medium or ingestion route was >95% in several other studies (Rowbotham 2000; Glorennec et al. 2016; Kawabe et al. 2003; Kurzius-Spencer et al. 2014; Lee et al. 2003; Ohno et al. 2010; Yeganeh et al. 2012). The metal exposure contribution of the diet was higher in rural areas than in urban areas. Females had a higher Hg, Cd, As, Pb and Cr exposure contribution from the diet than males, and a larger contribution of dietary exposure to the total exposure. The dietary exposure contribution of different age groups was 18-44 years old>45-59 years old>60 years old and above. According to relevant research, cereals, meat, aquatic products and vegetables generally make the biggest contribution to dietary exposure for Cr. For the other elements, major contributors – on average - tend to be as follows: Pb - cereals and vegetables; Cd – cereals; As - aquatic products, meat and vegetables; Hg - aquatic products, cereals and vegetables (Wu and Li 2015). In terms of the dietary structure, staple foods, meat, vegetables and aquatic products account for the main part of the diet in the survey areas, contributing >50% of the intake (Figure 2). In general, the high content of each metal in cereals may make the staple food a major component of the dietary exposure contribution; it can also generate some regional differences in China, with some areas predominantly rice-based and others with more wheat.

3.2.3 Non-dietary exposure contributions

In terms of non-dietary exposure contributions (Table S8), this was proportionally highest for As (12.2%), followed by Hg, Cr, Pb and Cd, for which it was >10%. The non-dietary exposure contribution of Hg, Cd and As were all higher in Lanzhou than the other areas (e.g. As, up to 35.8%. For Pb and Cr the non-dietary contribution in Taiyuan was higher than the other areas. On the whole, the non-dietary contribution of metals in Lanzhou, Wuhan, and Taiyuan were higher than the other areas, except for Cr. This may be related to urban industries. The rural area non-dietary exposure contribution was usually higher than urban areas (P>0.05) – except for Hg. However, the proportion of the exposure from non-dietary media such as drinking water, soil, air, and so on in rural areas were higher than in urban areas. Hg, Cd, As, Pb, and Cr non-dietary exposure contributions were generally higher for males than females (P>0.05). There are some differences between countries in the respect. In the US, the inorganic As non-dietary exposure contribution was higher for females than males (Meacher et al. 2002), while in Iran the Cd, Pb and Cr non-dietary exposure contribution for males was higher than for females (Yeganeh et al. 2012). The non-dietary exposure contribution for different age groups followed the sequence: 60 years old and above>45-59 years old>18-44 years old. The exposure contribution of ingestion route to the total environmental exposure levels was the highest for Hg, Cd, As, Pb and Cr, which were 99.99%, 99.96%, 97.83%, 99.76% and 99.91%, respectively (Figure 3, Table S9).

3.3 Risk characteristic

3.3.1 life-time non-cancer risks

In general, the average life-time non-carcinogenic risk level of Hg, Cd, As, Pb and Cr in 95th percentiles were all < 1, and followed the sequence As>Cr>Pb>Cd>Hg (Figure 4, Table S10). Therefore, the non-carcinogenic risk levels of metals were within an acceptable range. In terms of exposure media, the non-carcinogenic risk level of As in non-dietary and dietary exposure were both higher than the other metals, Hg was lowest for non-dietary exposure and Pb was lowest for dietary exposure (Table S11). The non-carcinogenic risk levels of non-dietary routes were higher than for the dietary route, except for Hg. In terms of exposure routes, the non-carcinogenic risk levels of As from inhalation, ingestion and dermal contact routes were all higher than for the other metals; Hg was lowest for inhalation and dermal contact, and Pb was lowest for ingestion (Table S12). The non-carcinogenic risk levels of Hg and As via ingestion were higher than for other routes, while for Cd, Pb and Cr inhalation was higher than other routes; the non-carcinogenic risk levels of dermal contact were lower than other routes except for Hg. The non-carcinogenic risk levels of Hg, Cd, As and Cr in Dalian were higher than other areas, while for Pb it was higher in Chengdu than other areas (Table S10). The non-carcinogenic risk for rural residents was higher than for urban residents, except for Cd. In terms of gender, the non-carcinogenic risk of males was higher than females, except for Hg. The non-carcinogenic risk levels of different age groups were ranked as 18-44 years old>45-59 years old>60 years old and above.

In terms of the non-dietary exposure contribution of Cd, As, Pb and Cr, non-carcinogenic risks were 67.70%，67.52%，88.75% and 57.99%, respectively, which were higher than via dietary exposure (Table 1). The non-carcinogenic risk contribution of Hg from non-dietary exposure was lower than from dietary exposure. Comparing the non-carcinogenic risk contributions with gender and age groups in different media, it was found that the contribution of non-dietary exposure in males was higher than females, while the dietary exposure in females was higher than males, and the contribution of non-dietary and dietary exposure for older adults were higher than the other age classes. As for the non-carcinogenic exposure risk contribution of each exposure route, Hg and As were higher through the ingestion route than other routes, and the contribution through the ingestion route were 98.87% and 65.64%, respectively (Table 2). The non-carcinogenic exposure risk contribution of Cd, Pb and Cr were ordered as inhalation> ingestion> dermal contact, and the inhalation route contribution were 64.50%, 83.16% and 46.31%, respectively. The contribution of non-carcinogenic risk to inhalation and ingestion routes of urban residents were higher than rural, by an average of 1.16 and 1.13 times. The non-carcinogenic risk contribution of rural residents through dermal contact exposure was 36.6 times higher than for urban dwellers. The contribution of non-carcinogenic risk through inhalation and dermal contact exposure routes were 1.15 and 1.35 times higher for males than for females, while the contribution through ingestion was 1.15 times higher for females than males. The non-carcinogenic risk contribution through inhalation exposure was higher for young adults than other age groups.

3.3.2 Life-time cancer risks

Based on the current EPA cancer slope factor for Cr, As, Pb, the life-time cancer risks associated with dietary and non-dietary intake were estimated. In general, the carcinogenic risk level of Cr, As and Pb was acceptable (10-4) in 95th percentiles, in the order Cr>As> Pb (Figure 5, Table S13). Cr could pose potential carcinogenic risk, in accordance with the result reported by Wang et al. (2019). The carcinogenic risk levels of rural residents were higher than urban; males were higher than females for Cr. In terms of exposure media, the carcinogenic risk levels of non-dietary exposure followed the order Cr>As>Pb, while for dietary exposure was As>Cr >Pb, respectively (Table S14). The carcinogenic risk levels of non-dietary exposure were higher than for dietary exposure, except for Pb. In terms of exposure routes, the carcinogenic risk levels of ingestion showed As>Cr>Pb, and for inhalation and dermal contact Cr>As>Pb. (Table S15). The carcinogenic risk levels of ingestion were highest (except for Cr), while dermal contact carcinogenic risk levels were lowest (except for As). The characteristic of carcinogenic risk of As, Pb and Cr in regions, urban-rural areas, genders and age groups were consistent with the non-carcinogenic risk. The carcinogenic risk of As and Cr were mainly from non-dietary exposure contributions, at 66.29% and 84.19%, respectively. However, the carcinogenic risk of Pb was mainly from dietary exposure contributions, at 68.64% (Table 3). The carcinogenic risk contribution of non-dietary exposure for males was higher than for females.

3.4 Limitations and uncertainties

Due to the toxicity database being unavailable, the health risk of Cr was calculated by the relevant parameters of $Cr6+$ in the evaluation of total Cr, which may lead to an overestimate of the risk of total chromium. The uncertainty of health risk was minimized through reducing the uncertainty of parameters; for example the exposure factors were all from one-by-one human behavior pattern surveys, except for the soil intake rate parameters.

4 Conclusions

This study shows that the total exposure level of residents were ordered as Cr>As>Pb>Cd>Hg, consistent with the concentration in environmental media. Differences between regions, urban-rural locations, gender and age were statistically significant. Dietary intake was the dominant route of total human exposure (with the contribution at least 87% for the different metals). Overall, the dietary exposure influence declined with increased age and the non-dietary exposure influence rose as age increased. It highlighted that non-dietary exposure and ingestion exposure have a greater influence on older age groups, while dietary exposure, inhalation and dermal contact exposure have a greater influence on the middle age group. According to health risk, the non-carcinogenic risk levels of the 5 elements followed the sequence As>Cr>Pb>Cd>Hg, while the carcinogenic risk showed Cr>As>Pb. The non-carcinogenic risk levels are all within the maximum acceptable level of 1 and the carcinogenic risk levels are all below the acceptable levels in 10-4; the health risk levels are all acceptable. The non-carcinogenic risks of all metals decreased with age. The carcinogenic risk of As and Pb is mainly through the ingestion route and Cr is mainly through the inhalation route. In general, it is very important to have a detailed total human exposure study before conducting environmental exposure to health risk control and carrying out targeted risk prevention and control work.

We have done a detailed and comprehensive research to heavy metal total human exposure and health risk through multiple media and routes. It provides a reference for health risk prevention and control.

Acknowledgments

The research leading to these results received funding from the Ministry of Ecology and Environment of the People's Republic of China (EH [2017]-15-14). We thank all volunteers and the participants contributing to this study in Dalian Ocean University, East China University of Science and Technology, Gansu Academy of Eco-environmental Sciences, Huazhong University of Science and Technology, Shanxi University and Sichuan University.

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Tables

Table 1. The contribution (%) of HI in each exposure medium

Metal		Hg		Cd		As		Pb		Cr	
		Non-dietary	Dietary	Non-dietary	Dietary	Non-dietary	Dietary	Non-dietary	Dietary	Non-dietary	Dietary
All		7.19	92.81	67.70	32.30	67.52	32.48	88.75	11.25	57.99	42.01
Region	Taiyuan	6.32	93.68	63.35	36.65	79.60	20.40	98.65	1.35	73.12	26.88
	Dalian	0.72	99.28	84.51	15.49	25.85	74.15	87.42	12.58	60.50	39.50
	Shanghai	2.66	97.34	31.46	68.54	67.06	32.94	79.60	20.40	79.54	20.46
	Wuhan	12.60	87.40	68.87	31.13	83.60	16.40	94.88	5.12	69.02	30.98
	Chengdu	2.17	97.83	60.32	39.68	46.38	53.62	74.32	25.68	47.52	52.48
	Lanzhou	17.58	82.42	94.19	5.81	98.21	1.79	95.63	4.37	21.32	78.68
Area	Urban	7.63	92.37	68.28	31.72	65.16	34.84	87.68	12.32	51.33	48.67
	Rural	6.73	93.27	67.09	32.91	70.02	29.98	89.89	10.11	65.04	34.96
Gender	Male	7.94	92.06	70.75	29.25	69.61	30.39	90.27	9.73	60.77	39.23
	Female	6.48	93.52	64.84	35.16	65.56	34.44	87.34	12.66	55.39	44.61
Age	18-44	6.21	93.79	67.88	32.12	61.02	38.98	86.72	13.28	58.11	41.89
	45-59	7.91	92.09	65.91	34.09	71.77	28.23	89.66	10.34	61.14	38.86
	≥60	7.84	92.16	69.18	30.82	72.35	27.65	90.68	9.32	54.76	45.24

Table 2. The contribution (%) of HI in each exposure route

Metal		Hg			Cd			As			Pb			Cr		
		Inhalation	Ingestion	Dermal	Inhalation	Ingestion	Dermal	Inhalation	Ingestion	Dermal	Inhalation	Ingestion	Dermal	Inhalation	Ingestion	Dermal
All		0.29	98.87	0.84	64.50	33.95	1.55	29.55	65.64	4.81	83.16	11.49	5.35	46.31	43.62	10.07
Region	Taiyuan	0.16	98.95	0.88	60.54	38.05	1.40	6.79	87.98	5.23	95.33	1.52	3.14	55.55	31.65	12.80
	Dalian	0.22	99.70	0.08	83.60	15.73	0.67	17.15	81.39	1.46	80.77	12.80	6.43	53.63	39.69	6.68
	Shanghai	0.42	99.53	0.05	29.06	70.82	0.12	48.75	50.24	1.01	78.06	20.85	1.09	77.66	20.68	1.65
	Wuhan	0.31	99.21	0.48	66.58	32.91	0.51	17.13	76.64	6.23	92.01	5.35	2.63	55.44	34.21	10.36

Metal		Hg			Cd			As			Pb			Cr		
		Inhalation	Ingestion	Dermal	Inhalation	Ingestion	Dermal	Inhalation	Ingestion	Dermal	Inhalation	Ingestion	Dermal	Inhalation	Ingestion	Dermal
	Chengdu	0.51	99.09	0.40	54.82	41.86	3.31	15.43	75.37	9.20	64.95	25.86	9.18	35.17	53.08	11.75
	Lanzhou	0.15	96.97	2.88	89.10	7.82	3.08	71.17	23.40	5.43	86.19	4.57	9.24	5.06	79.04	15.91
Area	Urban	0.34	99.50	0.16	66.54	33.36	0.09	30.45	69.41	0.14	87.26	12.50	0.24	49.00	50.18	0.81
	Rural	0.24	98.21	1.55	62.33	34.57	3.10	28.59	61.65	9.76	78.83	10.41	10.76	43.46	36.67	19.87
Gender	Male	0.34	98.63	1.04	67.43	30.81	1.76	31.87	62.56	5.57	84.26	9.95	5.79	47.76	40.70	11.54
	Female	0.24	99.11	0.65	61.76	36.88	1.36	27.37	68.52	4.11	82.14	12.92	4.94	44.95	46.34	8.70
Age	18-44	0.36	98.99	0.65	65.06	33.67	1.27	29.87	66.66	3.47	82.25	13.49	4.26	49.69	43.09	7.21
	45-59	0.26	98.64	1.10	62.22	35.74	2.04	29.92	63.75	6.33	82.51	10.60	6.89	46.41	40.70	12.90
	≥60	0.21	98.95	0.84	65.94	32.59	1.47	28.74	66.07	5.19	85.07	9.59	5.34	41.55	47.19	11.27

Table 3. The contribution (%) of TLICR in each exposure medium

Metal		As		Pb		Cr	
		Non-dietary	Dietary	Non-dietary	Dietary	Non-dietary	Dietary
All		66.29	33.71	31.36	68.64	84.19	15.81
Region	Taiyuan	83.27	16.73	64.06	35.94	94.19	5.81
	Dalian	20.81	79.19	19.71	80.29	92.20	7.80
	Shanghai	56.28	43.72	13.32	86.68	97.21	2.79
	Wuhan	85.74	14.26	35.31	64.69	93.14	6.86
	Chengdu	49.96	50.04	11.83	88.17	86.73	13.27
	Lanzhou	96.30	3.70	38.88	61.12	45.09	54.91
Area	Urban	64.58	35.42	29.16	70.84	82.71	17.29
	Rural	68.10	31.90	33.70	66.30	85.75	14.25
Gender	Male	68.10	31.90	34.21	65.79	85.34	14.66
	Female	64.59	35.41	28.70	71.30	83.11	16.89
Age	18-44	63.61	36.39	33.46	66.54	89.29	10.71
	45-59	68.96	31.04	32.46	67.54	84.64	15.36
	≥60	67.39	32.61	27.39	72.61	76.69	23.31

Table 4. The contribution (%) of TLICR in each exposure route

Metal		As			Pb			Cr		
		Inhalation	Ingestion	Dermal Contact	Inhalation	Ingestion	Dermal Contact	Inhalation	Ingestion	Dermal Contact
All		10.72	82.67	6.62	25.10	72.46	2.44	77.08	16.36	6.56
Region	Taiyuan	1.17	93.20	5.63	55.47	41.65	2.88	84.98	7.23	7.79
	Dalian	4.91	92.62	2.48	14.69	83.01	2.29	89.86	7.86	2.28
	Shanghai	17.59	80.81	1.60	9.51	90.27	0.21	96.76	2.84	0.40
	Wuhan	3.55	89.28	7.17	29.05	69.56	1.38	87.76	7.88	4.37

Metal		As			Pb			Cr		
		Inhalation	Ingestion	Dermal Contact	Inhalation	Ingestion	Dermal Contact	Inhalation	Ingestion	Dermal Contact
	Chengdu	5.17	83.83	10.99	8.83	89.44	1.73	79.94	13.56	6.50
	Lanzhou	31.17	57.62	11.21	28.71	65.63	5.66	28.18	55.27	16.55
Area	Urban	12.10	87.66	0.24	26.38	73.51	0.11	81.81	17.77	0.41
	Rural	9.25	77.39	13.36	23.75	71.35	4.90	72.07	14.87	13.06
Gender	Male	11.95	80.33	7.71	27.41	69.68	2.90	77.29	15.16	7.55
	Female	9.56	84.85	5.59	22.94	75.05	2.00	76.88	17.49	5.63
Age	18-44	11.97	82.89	5.13	27.52	70.36	2.12	83.87	11.13	5.00
	45-59	10.11	81.21	8.69	25.50	71.46	3.03	75.52	15.98	8.49
	≥60	9.57	83.78	6.65	21.37	76.33	2.29	69.22	23.97	6.81

Figures

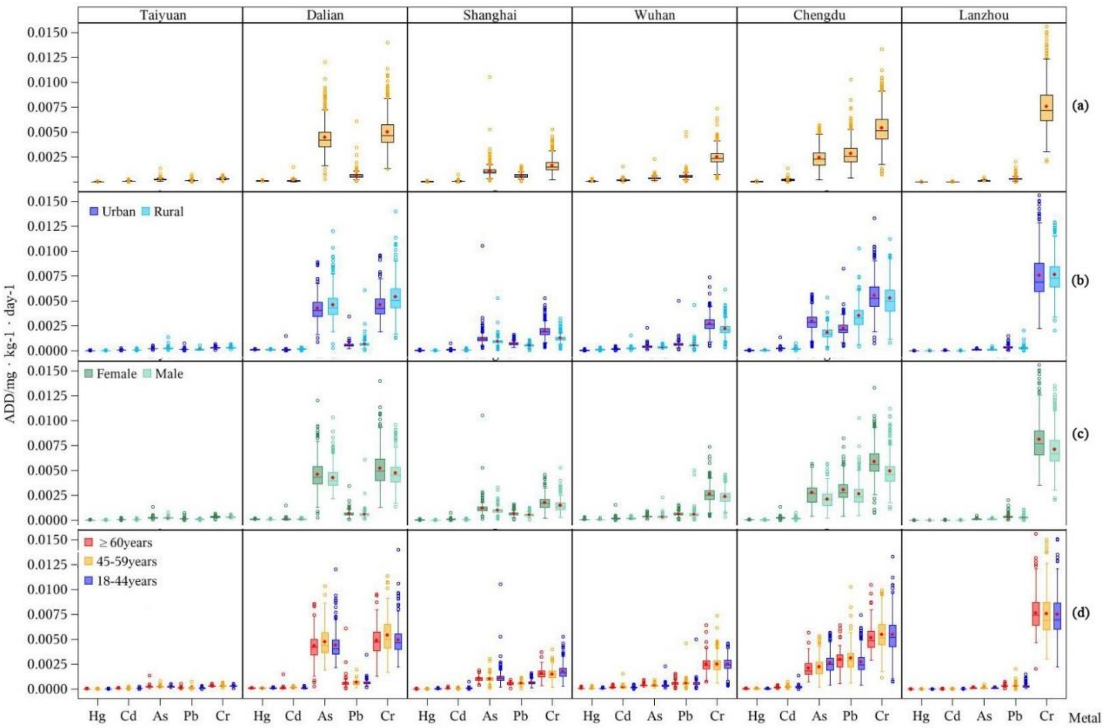

Figure 1. The total daily exposure doses of metals in different areas, sex, and age groups. Boxes represent 25th, 50th (shown by the horizontal line) and 75th percentiles. Whiskers indicate 5th and 95th values and the circles show outliers, the red filled dots in boxes show mean values. The first row in the figure marked (a) is the total exposure level of areas (in orange); the second row in the figure marked (b) is the total exposure level of urban(in blue) and rural(in light blue) areas; the third row in the figure marked (c) is the total exposure level of male(in green) and female(in light green); the fourth row in the figure marked (d) is the total exposure level of age groups(>60 years old in red, 45-59 years old in yellow and 18-44 years old in blue). For the detailed data was shown in Table S4.

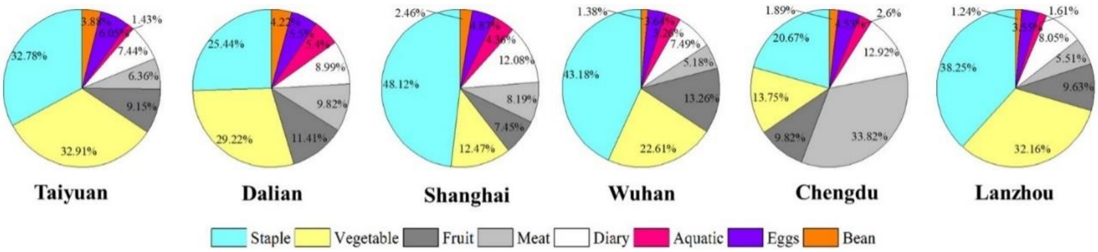

Figure 2. The ratio of dietary intake of different types of food in different seasons in typical areas.

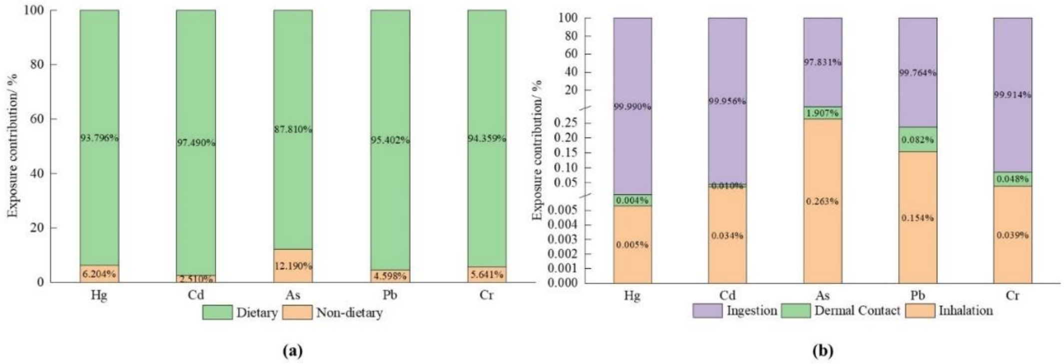

Figure 3. The exposure contribution of Hg, Cd, As, Pb and Cr. The bars in the figure marked (a) is the exposure contribution of each media; (b) is the total exposure contribution of each routes. For the detailed data was shown in Table S8 and Table S9.

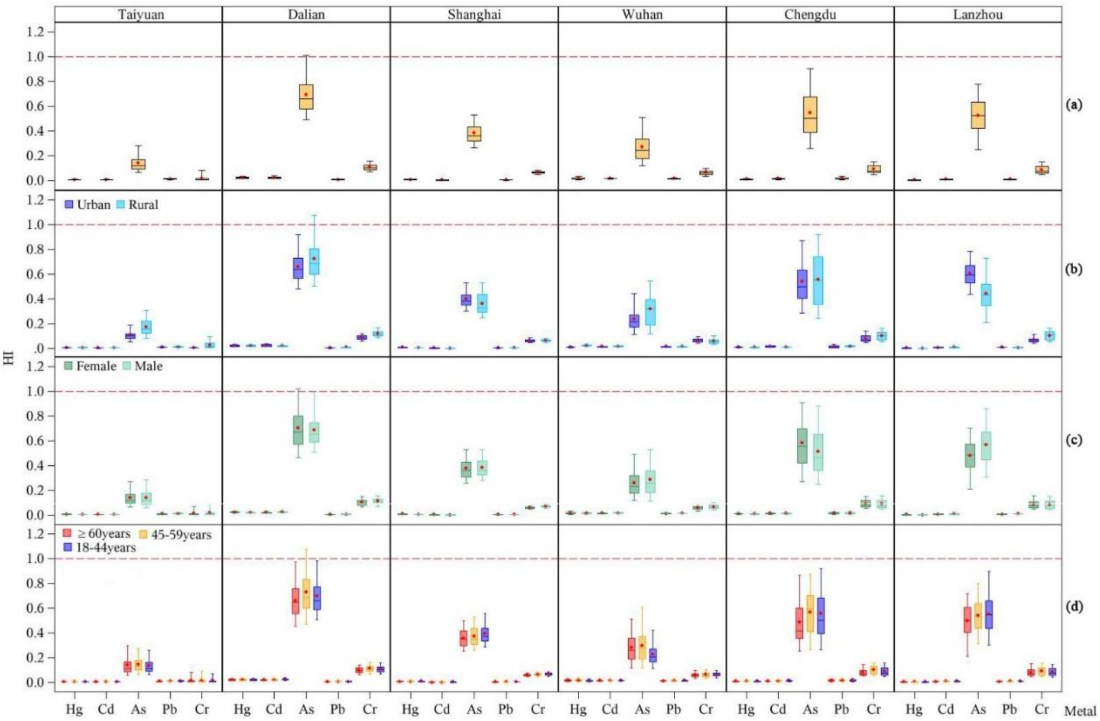

Figure 4. The hazard index (HI) of Hg, Cd, As, Pb and Cr. Boxes represent 25^{th}, 50^{th} (shown by the horizontal line) and 75^{th} percentiles. Whiskers indicate 5^{th} and 95^{th} values and the circles

show outliers, the red filled dots in boxes show mean values. The first row in the figure marked (a) is the total HI of areas (in orange); the second row in the figure marked (b) is the total HI of urban (in blue) and rural (in light blue) areas; the third row in the figure marked (c) is the total HI of male (in green) and female (in light green); the fourth row in the figure marked (d) is the total HI of age groups (>60 years old in red, 45-59 years old in yellow and 18-44 years old in blue). For the detailed data was shown in Table S10.

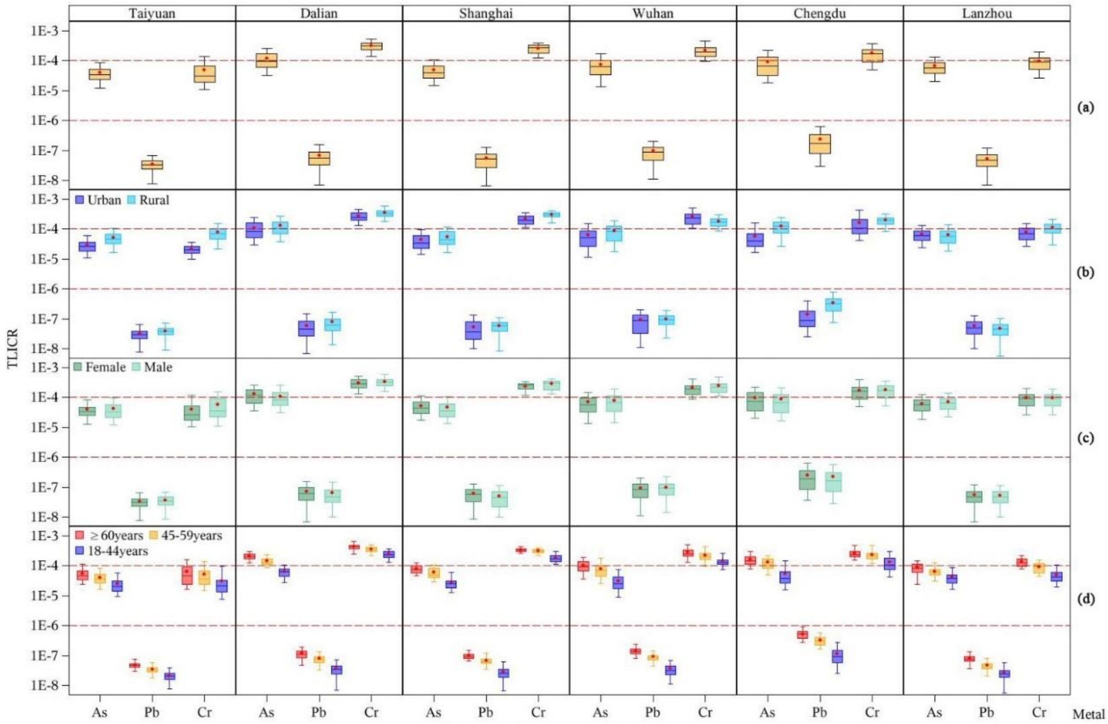

Figure 5. The total life time cancer risk (TLICR) of As, Pb and Cr. Boxes represent 25th, 50th (shown by the horizontal line) and 75th percentiles. Whiskers indicate 5th and 95th values and the circles show outliers, the red filled dots in boxes show mean values. The first row in the figure marked (a) is the total TLICR of areas (in orange); the second row in the figure marked (b) is the total TLICR of urban (in blue) and rural (in light blue) areas; the third row in the figure marked (c) is the total TLICR of male (in green) and female (in light green); the fourth row in the figure marked (d) is the total TLICR of age groups(>60 years old in red, 45-59 years old in yellow and 18-44 years old in blue). For the detailed data was shown in Table S13.

Supplemental information for

Total human environmental exposure and risk assessment of residents for metals in typical areas of China

Xiuge Zhao[a,*], Danlu Wang[a], Bin Zou[c], Jian Xu [a], Limin Lei[b], Zhenglei Li[b], Yan Tao[b], Yiming Sun[d], Kevin Jones[d,*], Feiyang Qiao[b], Ju Huang[b]

Table S1. Summary of Exposure Factor from the behavior pattern survey in this study

Area		BW (kg)	InhR* (m^3/d)	SA (m^2)	Time-activity factors related to air exposure (h/d)			Time in contact with soil (h/d)	Time spent showering/bathing (min/d)	Time spent swimming (min/month)	$IngR_f$ (g/d)	$IngR_w$ (L/d)
					indoor	outdoor	transit					
Taiyuan	Mean	63.91	13.32	1.67	19.58	3.52	1.00	0.81	8.53	119.15	999.49	1.29
	P5	50.00	9.89	1.44	16.50	1.00	0.25	0.00	1.93	6.00	820.97	0.53
	P95	80.00	17.74	1.89	22.58	6.50	2.08	6.96	16.00	900.00	1322.00	2.30
Dalian	Mean	65.94	13.73	1.71	18.83	4.56	1.00	1.81	8.17	243.22	1097.84	1.11
	P5	50.00	10.20	1.47	16.11	1.65	0.37	0.00	0.50	90.00	816.78	0.48
	P95	80.00	18.52	1.94	21.68	7.33	1.60	8.00	16.25	564.00	1605.93	2.20
Shanghai	Mean	63.14	13.60	1.66	20.49	2.35	1.28	0.10	5.40	191.41	979.10	1.22
	P5	50.00	10.05	1.45	19.43	1.28	0.50	0.00	0.00	75.00	817.57	0.61
	P95	79.00	18.52	1.89	21.35	3.41	2.40	0.50	14.88	297.00	1340.32	2.14
Wuhan	Mean	61.01	12.93	1.63	19.76	3.43	0.99	0.56	10.60	165.39	1065.91	1.55
	P5	50.00	9.61	1.44	15.65	1.00	0.33	0.00	2.78	30.00	820.52	0.70
	P95	75.00	17.62	1.84	22.50	8.00	2.00	3.00	22.92	315.00	1518.21	2.92
Chengdu	Mean	58.78	13.45	1.60	19.59	3.65	0.99	1.56	6.50	202.05	1087.70	1.92
	P5	45.00	9.66	1.37	14.88	1.03	0.50	0.00	1.20	45.00	846.87	0.83
	P95	77.00	18.42	1.88	22.24	8.36	1.48	7.33	16.75	560.00	1504.28	2.87
Lanzhou	Mean	61.76	13.29	1.64	18.65	4.30	1.21	2.00	4.30	169.81	1091.43	1.49
	P5	49.00	9.58	1.42	14.71	1.27	0.36	0.00	0.25	18.00	822.67	0.39
	P95	80.00	18.21	1.88	21.80	7.68	2.86	8.00	11.00	600.00	1673.18	3.01

Note: BW is body weight, InhR is inhalation rate, SA is the surface area of the skin, $IngR_f$ is the ingestion rate of dietary, $IngR_w$ is the ingestion rate of water.

*Inhalation rate was calculated by the method from *Exposure factors handbook: 2011 edition* of USA.

Table S2. Some parameters in formula of this study

Parameter		Reference value	Source
Ingestion rate of soil (IngRs mg/d)		50	Ministry of Ecology and Environment of the People's Republic of China, 2013a. Exposure Factors Handbook of Chinese Population (Adults). China Environmental Science Press, Beijing, pp262
Digestive tract absorption factor (FI)		When calculate the risk we need to use 0.07 for Hg, 0.025 for Cd, Pb and Cr	Ministry of Ecology and Environment of the People's Republic of China, 2014. Technical guidelines for risk assessment of contaminated sites HJ 25.3-2014, China Environmental Science Press, Beijing.
Skin adherence factor (AF $mg\cdot cm^{-2}$)		0.2	
Dermal absorption factor (PC $cm\cdot hr^{-1}$)		0.0018 for As, 0.000004 for Pb, 0.002 for Cr, 0.001 for Cd, 0.001 for Hg	U.S.EPA.Dermal Exposure Assessment:Principles and Applications .EPA /600/8-91/011B,http://www.epa.gov,1992.P5-9,pp5-11
Particulate emission factor (PEF $m^3\cdot kg^{-1}$)		1.36×10^9 $m^3\cdot kg^{-1}$	U.S.EPA.Supplemental Guidance for Developing Soil Screening Levels for Superfund Sites. Washington, DC: Office of Emergency and Remedial Response 2001. pp4-17
Dermal absorption factor (ABS_d)		0.001 for Hg, Cd, Pb and Cr, 0.03 for As	U.S. EPA, RAGS, Part E, Supplemental Guidance for Dermal Risk Assessment, Interim Guidance, 2001.pp3-16
Averaging time (AT days)		For non-cancer, AT = ED×365; for cancer, AT=74.8×365	US EPA, Risk Assessment Guidance for Superfund (RAGS), Volume I: Human Health Evaluation Manual (Part E, Supplemental Guidance for Dermal Risk Assessment) Final. Washington, DC: Office of Superfund Remediation and Technology Innovation. 2009.pp3-9
Reference dose (Rfd mg/(kg·d))	Inhalation (Rfd_i)	0.0000039 for As, 0.0000026 for Cd, 0.000026 for Cr, 0.000052 for Pb, 0.000078 for Hg	Oak Ridge National Laboratory. 2014. Risk Assessment Information System. http://rais.ornl.gov/
	Oral (Rfd_o)	0.0003 for As, 0.001 for Cd(diet), 0.0005for Cd(water), 0.003 for Cr, 0.02 for Pb, 0.0003 for Hg	
	Dermal (Rfd_d)	0.0003 for As, 0.000025 for Cd, 0.000075 for Cr, 0.0005 for Pb, 0.000021 for Hg	
slope factor (SF (kg·d)/mg)	Inhalation (SF_i)	17 for As, 6.9 for Cd, 320 for Cr, 0.046 for Pb	
	Oral (SF_o)	1.5 for AS, 0.5 for CR, 0.0085 for Pb	
	Dermal (SF_d)	1.5 for As, 20 for Cr, 0.0085 for Pb	

Table S3. The concentration of Hg, Cd, As, Pb and Cr in each exposure media

Metal/Area		Air (ng/m^3)						Water (ug/L)		Soil (mg/kg)		Dietary (mg/kg)	
		Indoor		Outdoor		Traffic							
		Mean	SD	Mean	SD	Mean	SD	Mean	SD	Mean	SD	Mean	SD
Hg	Taiyuan	0.0884	0.0922	0.1177	0.1547	0.0736	0.0581	0.0786	0.0505	0.3437	0.2039	0.0019	0.0010
	Dalian	0.5344	0.3610	0.0776	0.1545	0.0409	0.0447	0.0219	0.0071	0.0174	0.0081	0.0057	0.0009
	Shanghai	0.2808	0.2726	0.0728	0.0639	0.1316	0.1853	0.0360	0.0623	0.0475	0.0182	0.0022	0.0027
	Wuhan	0.2317	0.2596	0.1556	0.2060	0.1228	0.1148	0.2159	0.0802	0.1411	0.1404	0.0032	0.0058
	Chengdu	0.3900	0.2225	0.4638	0.2184	0.3859	0.2446	0.0156	0.0119	0.0273	0.0169	0.0023	0.0013
	Lanzhou	0.0335	0.0000	0.0335	0.0000	0.0335	0.0000	0.0871	0.1992	0.1132	0.1845	0.0006	0.0006
Cd	Taiyuan	0.2531	1.0224	5.2880	8.9224	3.5782	7.6937	0.0365	0.0906	0.2331	0.0168	0.0048	0.0049
	Dalian	7.4929	7.5006	0.9735	0.8392	0.9308	0.9273	0.0339	0.0543	0.1818	0.0550	0.0073	0.0287
	Shanghai	0.0877	0.1250	1.0355	0.6863	1.2300	0.6187	0.0262	0.0317	0.0627	0.0478	0.0056	0.0159
	Wuhan	3.1340	1.9754	2.7130	2.5143	4.0425	4.9511	0.0906	0.2034	0.3609	0.1331	0.0111	0.0128
	Chengdu	1.8112	2.9920	2.5688	5.8980	4.6738	2.6815	0.0739	0.1300	0.6214	0.1965	0.0121	0.0322
	Lanzhou	2.2613	1.8178	2.2696	2.0923	2.8019	2.7900	0.0686	0.1192	0.3255	0.1031	0.0011	0.0007
As	Taiyuan	2.0284	4.0595	12.1287	14.6305	9.9746	13.7202	1.2931	1.0721	14.7465	0.3157	0.0133	0.0163
	Dalian	61.2209	37.7657	14.0963	12.7509	16.7960	16.7632	0.7598	0.2381	6.2317	1.6265	0.2584	0.1547
	Shanghai	80.4316	15.3088	53.6993	23.0226	49.9476	22.0741	0.7432	0.4278	47.1527	2.6068	0.0695	0.1461
	Wuhan	17.8879	31.1827	15.6420	23.8006	17.7805	17.7644	1.6312	0.9603	23.2766	27.2539	0.0182	0.0382
	Chengdu	21.8004	44.5631	25.3882	38.2094	12.2283	11.4844	0.8257	1.5678	41.7146	11.3461	0.1566	0.9331
	Lanzhou	198.1877	146.9202	15.9545	12.7369	17.0536	4.7740	1.1309	0.8087	10.8302	4.6536	0.0041	0.0029
Pb	Taiyuan	52.1868	71.7565	88.7678	120.0660	134.6531	179.3419	0.5475	3.3550	26.0649	3.0684	0.0075	0.0114
	Dalian	32.8558	45.7860	22.6809	16.8970	21.6822	15.6361	0.4823	0.8040	19.0614	6.3569	0.0363	0.0670
	Shanghai	9.3882	6.3887	52.4963	21.8471	67.0774	23.6062	0.5725	0.7375	24.3955	12.7201	0.0403	0.0252
	Wuhan	75.7678	53.8845	82.6832	77.7236	91.1360	57.6071	0.7880	1.1078	29.7873	19.6817	0.0340	0.1029
	Chengdu	45.4704	87.1023	38.5795	38.4146	77.4802	62.0002	0.2718	0.7208	47.5838	12.4194	0.1466	0.2110
	Lanzhou	40.5518	19.4927	55.5460	39.2515	60.7419	44.3270	0.3224	0.6452	24.0903	16.4072	0.0186	0.0369
Cr	Taiyuan	1.3156	1.7600	91.5891	183.8794	39.7904	49.0158	2.5523	1.4761	72.9743	1.6990	0.0159	0.0124
	Dalian	216.9877	224.4217	9.9009	6.6944	7.8011	3.3127	1.0654	0.9365	33.2896	7.7342	0.2863	0.1935
	Shanghai	154.3363	147.4898	107.2905	42.0496	102.3120	50.5978	0.3420	0.0826	87.0713	9.7624	0.1073	0.1412
	Wuhan	105.1244	92.5590	29.5483	23.8958	32.1461	20.4263	8.3331	2.6606	64.7511	43.1377	0.1305	0.1318
	Chengdu	60.9754	98.7472	83.2174	54.6349	51.5664	32.5525	0.8694	4.7250	69.4974	15.8267	0.2785	0.3239
	Lanzhou	7.8016	6.3944	14.6547	8.0869	17.8816	8.9446	0.7672	0.5249	67.2916	5.0184	0.4140	0.2006

Table S4. The total exposure average daily dose (expressed as mg/(kg·d)) of Hg, Cd, As, Pb and Cr

Metal		Hg			Cd			As			Pb			Cr		
		Mean	95%UCI	95%LCI	Mean	95%UCI	95%LCI	Mean	95%UCI	95%LCI	Mean	95%UCI	95%LCI	Mean	95%UCI	95%LCI
All		4.72E-05	4.83E-05	4.61E-05	1.22E-04	1.24E-04	1.19E-04	1.41E-03	1.47E-03	1.36E-03	8.45E-04	8.78E-04	8.13E-04	3.76E-03	3.85E-03	3.67E-03
Region	Taiyun	3.14E-05	3.19E-05	3.09E-05	7.95E-05	8.10E-05	7.79E-05	2.41E-04	2.47E-04	2.35E-04	1.28E-04	1.31E-04	1.25E-04	3.28E-04	3.33E-04	3.23E-04
	Dalian	9.74E-05	9.94E-05	9.54E-05	1.35E-04	1.41E-04	1.29E-04	4.45E-03	4.56E-03	4.35E-03	6.45E-04	6.71E-04	6.20E-04	5.01E-03	5.13E-03	4.88E-03
	Shanghai	3.48E-05	3.56E-05	3.39E-05	8.68E-05	8.99E-05	8.37E-05	1.10E-03	1.15E-03	1.06E-03	6.30E-04	6.45E-04	6.15E-04	1.62E-03	1.68E-03	1.57E-03
	Wuhan	6.86E-05	7.17E-05	6.55E-05	2.06E-04	2.13E-04	2.00E-04	3.78E-04	3.88E-04	3.68E-04	6.17E-04	6.41E-04	5.93E-04	2.50E-03	2.56E-03	2.44E-03
	Chengdu	4.36E-05	4.45E-05	4.26E-05	2.14E-04	2.21E-04	2.08E-04	2.44E-03	2.59E-03	2.30E-03	2.83E-03	2.92E-03	2.74E-03	5.41E-03	5.53E-03	5.28E-03
	Lanzhou	1.29E-05	1.32E-05	1.25E-05	2.08E-05	2.13E-05	2.03E-05	1.18E-04	1.21E-04	1.15E-04	3.20E-04	3.31E-04	3.08E-04	7.59E-03	7.75E-03	7.43E-03
Area: Taiyun	Urban	3.02E-05	3.10E-05	2.95E-05	7.66E-05	7.87E-05	7.46E-05	2.06E-04	2.11E-04	2.01E-04	1.35E-04	1.39E-04	1.30E-04	3.17E-04	3.24E-04	3.10E-04
	Rural	3.26E-05	3.34E-05	3.19E-05	8.26E-05	8.48E-05	8.03E-05	2.78E-04	2.88E-04	2.69E-04	1.21E-04	1.25E-04	1.17E-04	3.41E-04	3.48E-04	3.33E-04
Dalian	Urban	9.81E-05	1.01E-04	9.53E-05	1.02E-04	1.12E-04	9.15E-05	4.26E-03	4.39E-03	4.12E-03	5.95E-04	6.22E-04	5.67E-04	4.57E-03	4.72E-03	4.42E-03
	Rural	9.68E-05	9.96E-05	9.40E-05	1.63E-04	1.69E-04	1.58E-04	4.62E-03	4.77E-03	4.46E-03	6.89E-04	7.30E-04	6.48E-04	5.38E-03	5.56E-03	5.20E-03
Shanghai	Urban	3.74E-05	3.88E-05	3.61E-05	9.97E-05	1.05E-04	9.44E-05	1.24E-03	1.32E-03	1.17E-03	7.22E-04	7.43E-04	7.00E-04	1.99E-03	2.07E-03	1.92E-03
	Rural	3.21E-05	3.30E-05	3.13E-05	7.41E-05	7.68E-05	7.14E-05	9.63E-04	1.00E-03	9.21E-04	5.40E-04	5.54E-04	5.25E-04	1.26E-03	1.30E-03	1.21E-03
Wuhan	Urban	4.54E-05	4.72E-05	4.35E-05	1.85E-04	1.90E-04	1.80E-04	4.00E-04	4.15E-04	3.84E-04	6.56E-04	6.87E-04	6.25E-04	2.72E-03	2.80E-03	2.63E-03
	Rural	9.96E-05	1.04E-04	9.49E-05	2.35E-04	2.47E-04	2.22E-04	3.49E-04	3.60E-04	3.38E-04	5.66E-04	6.03E-04	5.29E-04	2.21E-03	2.29E-03	2.12E-03
Chengdu	Urban	4.52E-05	4.65E-05	4.39E-05	2.22E-04	2.31E-04	2.12E-04	2.99E-03	3.24E-03	2.75E-03	2.25E-03	2.33E-03	2.17E-03	5.53E-03	5.71E-03	5.36E-03
	Rural	4.17E-05	4.31E-05	4.03E-05	2.06E-04	2.15E-04	1.98E-04	1.82E-03	1.89E-03	1.75E-03	3.50E-03	3.63E-03	3.36E-03	5.26E-03	5.45E-03	5.07E-03
Lanzhou	Urban	1.55E-05	1.59E-05	1.50E-05	2.26E-05	2.33E-05	2.19E-05	1.19E-04	1.22E-04	1.15E-04	3.60E-04	3.76E-04	3.45E-04	7.58E-03	7.83E-03	7.34E-03
	Rural	1.02E-05	1.05E-05	9.90E-06	1.89E-05	1.95E-05	1.84E-05	1.18E-04	1.22E-04	1.13E-04	2.78E-04	2.92E-04	2.63E-04	7.60E-03	7.80E-03	7.40E-03

Metal		Hg			Cd			As			Pb			Cr		
		Mean	95%UCI	95%LCI	Mean	95%UCI	95%LCI	Mean	95%UCI	95%LCI	Mean	95%UCI	95%LCI	Mean	95%UCI	95%LCI
Gender	Male	4.35E-05	4.50E-05	4.21E-05	1.12E-04	1.16E-04	1.09E-04	1.29E-03	1.36E-03	1.22E-03	7.92E-04	8.36E-04	7.47E-04	3.56E-03	3.68E-03	3.44E-03
	Female	5.06E-05	5.22E-05	4.91E-05	1.30E-04	1.35E-04	1.26E-04	1.53E-03	1.62E-03	1.44E-03	8.95E-04	9.43E-04	8.48E-04	3.95E-03	4.08E-03	3.81E-03
Age	18-44years	4.91E-05	5.07E-05	4.75E-05	1.30E-04	1.35E-04	1.26E-04	1.79E-03	1.89E-03	1.69E-03	1.01E-03	1.06E-03	9.56E-04	3.92E-03	4.05E-03	3.79E-03
	45-59years	4.56E-05	4.78E-05	4.35E-05	1.18E-04	1.23E-04	1.12E-04	1.20E-03	1.30E-03	1.11E-03	8.01E-04	8.64E-04	7.37E-04	3.57E-03	3.74E-03	3.39E-03
	≥60years	4.62E-05	4.81E-05	4.42E-05	1.13E-04	1.18E-04	1.08E-04	1.09E-03	1.18E-03	1.00E-03	6.61E-04	7.10E-04	6.12E-04	3.73E-03	3.90E-03	3.56E-03

Table S5. The non-dietary exposure average daily dose (expressed as $10^{-6}\times g\cdot kg^{-1}\cdot day^{-1}$)

Metal		Hg			Cd			As			Pb			Cr		
		Mean	95%UCI	95%LCI	Mean	95%UCI	95%LCI	Mean	95%UCI	95%LCI	Mean	95%UCI	95%LCI	Mean	95%UCI	95%LCI
All		1.82	1.89	1.75	1.58	1.63	1.53	38.70	39.80	37.60	17.90	18.50	17.30	74.70	77.70	71.80
Region	Taiyuan	1.64	1.71	1.58	0.79	0.84	0.73	31.90	33.30	30.50	14.30	16.10	12.50	63.00	65.90	60.10
	Dalian	0.39	0.41	0.38	0.61	0.63	0.59	18.20	18.90	17.40	12.30	12.80	11.70	27.60	29.20	26.10
	Shanghai	0.76	0.79	0.72	0.73	0.77	0.68	23.30	24.80	21.80	13.90	14.60	13.20	19.70	21.90	17.50
	Wuhan	5.55	5.76	5.35	2.86	3.03	2.68	57.90	60.70	55.10	28.70	30.10	27.30	230.00	239.00	222.00
	Chengdu	0.55	0.57	0.53	2.66	2.79	2.53	55.70	60.10	51.30	24.30	26.30	22.30	69.90	77.10	62.60
	Lanzhou	2.01	2.14	1.87	1.86	1.99	1.74	44.50	46.40	42.50	14.70	15.70	13.70	41.00	43.30	38.70
Area	Urban	2.09	2.20	1.99	1.46	1.53	1.39	29.40	30.40	28.40	11.40	11.90	11.00	55.70	59.90	51.60
	Rural	1.53	1.61	1.44	1.71	1.78	1.63	48.50	50.40	46.60	24.80	25.90	23.80	94.80	98.80	90.70
Gender	Metal	1.83	1.94	1.72	1.58	1.66	1.50	39.70	41.40	38.10	18.10	18.80	17.30	75.80	80.30	71.30
	Female	1.81	1.90	1.72	1.58	1.65	1.51	37.70	39.20	36.30	17.80	18.70	16.90	73.70	77.60	69.80
Age	18-44	1.53	1.63	1.44	1.52	1.59	1.45	33.40	35.00	31.70	15.70	16.40	14.90	56.90	60.60	53.20
	45-59	1.90	2.04	1.76	1.57	1.67	1.47	43.80	46.00	41.60	19.70	20.70	18.60	83.90	90.00	77.90
	≥60	2.14	2.27	2.00	1.68	1.78	1.57	41.10	42.90	39.20	19.40	20.70	18.10	90.30	96.20	84.40

Table S6. The dietary exposure average daily dose (expressed as $10^{-4}\times mg/(kg\cdot d)$)

Metal		Hg			Cd			As			Pb			Cr		
		Mean	95%UCI	95%LCI	Mean	95%UCI	95%LCI	Mean	95%UCI	95%LCI	Mean	95%UCI	95%LCI	Mean	95%UCI	95%LCI
All		0.45	0.47	0.44	1.20	1.23	1.17	13.70	14.30	13.20	8.27	8.60	7.95	36.80	37.80	35.90
Region	Taiyun	0.30	0.30	0.29	0.79	0.80	0.77	2.09	2.14	2.04	1.14	1.17	1.11	2.65	2.70	2.61
	Dalian	0.97	0.99	0.95	1.34	1.40	1.28	44.30	45.40	43.30	6.33	6.59	6.07	49.80	51.00	48.60
	Shanghai	0.34	0.35	0.33	0.86	0.89	0.83	10.80	11.20	10.30	6.16	6.31	6.02	16.00	16.60	15.50
	Wuhan	0.63	0.66	0.60	2.03	2.10	1.97	3.20	3.30	3.10	5.89	6.13	5.65	22.70	23.30	22.10
	Chengdu	0.43	0.44	0.42	2.12	2.18	2.05	23.90	25.30	22.40	28.10	29.00	27.20	53.40	54.60	52.10
	Lanzhou	0.11	0.11	0.11	0.19	0.19	0.19	0.74	0.76	0.72	3.05	3.16	2.94	75.50	77.10	73.90

Metal		Hg			Cd			As			Pb			Cr		
		Mean	95%UCI	95%LCI	Mean	95%UCI	95%LCI	Mean	95%UCI	95%LCI	Mean	95%UCI	95%LCI	Mean	95%UCI	95%LCI
Area	Urban	0.42	0.43	0.40	1.17	1.21	1.13	14.10	14.90	13.30	7.68	8.02	7.34	37.10	38.40	35.90
	Rural	0.49	0.51	0.48	1.23	1.28	1.19	13.40	14.20	12.60	8.90	9.46	8.33	36.60	37.90	35.20
Gender	Male	0.42	0.43	0.40	1.11	1.14	1.07	12.50	13.20	11.80	7.74	8.18	7.29	34.80	36.10	33.60
	Female	0.49	0.50	0.47	1.29	1.33	1.24	14.90	15.80	14.00	8.78	9.25	8.31	38.70	40.10	37.40
Age	18-44	0.48	0.49	0.46	1.29	1.33	1.25	17.60	18.60	16.60	9.94	10.50	9.40	38.60	40.00	37.30
	45-59	0.44	0.46	0.42	1.16	1.22	1.11	11.60	12.50	10.60	7.81	8.44	7.18	34.80	36.60	33.00
	≥60	0.44	0.46	0.42	1.11	1.16	1.06	10.50	11.40	9.63	6.42	6.90	5.93	36.40	38.10	34.70

Table S7. The total environmental exposure average daily dose (expressed as mg/(kg·d)) in different routes

Metal		Hg			Cd			As			Pb			Cr		
		Ingestion	Inhalation	Dermal contact	Ingestion	Inhalation	Dermal contact	Ingestion	Inhalation	Dermal contact	Ingestion	Inhalation	Dermal contact	Ingestion	Inhalation	Dermal contact
All		4.72×10^{-5}	2.20×10^{-9}	9.77×10^{-10}	1.21×10^{-4}	2.18×10^{-8}	4.24×10^{-9}	1.41×10^{-3}	5.26×10^{-7}	7.26×10^{-6}	8.44×10^{-4}	4.51×10^{-7}	3.58×10^{-7}	3.76×10^{-3}	7.72×10^{-7}	7.82×10^{-7}
Region	Taiyuan	3.14×10^{-5}	8.83×10^{-10}	1.42×10^{-9}	7.95×10^{-5}	9.65×10^{-9}	2.28×10^{-9}	2.37×10^{-4}	3.41×10^{-8}	4.02×10^{-6}	1.27×10^{-4}	5.73×10^{-7}	2.05×10^{-7}	3.27×10^{-4}	1.63×10^{-7}	6.42×10^{-7}
	Dalian	9.74×10^{-5}	3.64×10^{-9}	3.64×10^{-10}	1.35×10^{-4}	4.99×10^{-8}	3.46×10^{-9}	4.45×10^{-3}	4.44×10^{-7}	3.07×10^{-6}	6.45×10^{-4}	2.84×10^{-7}	2.97×10^{-7}	5.00×10^{-3}	1.46×10^{-6}	6.57×10^{-7}
	Shanghai	3.48×10^{-5}	2.54×10^{-9}	9.02×10^{-11}	8.68×10^{-5}	2.23×10^{-9}	8.99×10^{-11}	1.10×10^{-3}	6.99×10^{-7}	1.52×10^{-6}	6.30×10^{-4}	1.55×10^{-7}	2.29×10^{-8}	1.62×10^{-3}	1.32×10^{-6}	9.35×10^{-8}
	Wuhan	6.86×10^{-5}	2.34×10^{-9}	1.51×10^{-9}	2.06×10^{-4}	2.81×10^{-8}	2.26×10^{-9}	3.70×10^{-4}	1.58×10^{-7}	7.85×10^{-6}	6.16×10^{-4}	7.07×10^{-7}	2.20×10^{-7}	2.50×10^{-3}	9.21×10^{-7}	5.86×10^{-7}
	Chengdu	4.35×10^{-5}	3.82×10^{-9}	8.07×10^{-10}	2.14×10^{-4}	2.10×10^{-8}	9.30×10^{-9}	2.42×10^{-3}	3.27×10^{-7}	1.96×10^{-5}	2.83×10^{-3}	5.61×10^{-7}	9.58×10^{-7}	5.40×10^{-3}	8.30×10^{-7}	1.12×10^{-6}
	Lanzhou	1.29×10^{-5}	3.22×10^{-10}	1.53×10^{-9}	2.08×10^{-5}	2.08×10^{-8}	7.69×10^{-9}	1.09×10^{-4}	1.46×10^{-6}	7.48×10^{-6}	3.19×10^{-4}	4.04×10^{-7}	4.32×10^{-7}	7.59×10^{-3}	9.63×10^{-8}	1.50×10^{-6}
Area	Urban	4.38×10^{-5}	2.46×10^{-9}	2.02×10^{-10}	1.18×10^{-4}	2.31×10^{-8}	2.40×10^{-10}	1.44×10^{-3}	5.99×10^{-7}	2.27×10^{-7}	7.79×10^{-4}	4.63×10^{-7}	1.36×10^{-8}	3.77×10^{-3}	7.88×10^{-7}	3.60×10^{-8}
	Rural	5.08×10^{-5}	1.92×10^{-9}	1.80×10^{-9}	1.25×10^{-4}	2.05×10^{-8}	8.48×10^{-9}	1.37×10^{-3}	4.49×10^{-7}	1.47×10^{-5}	9.13×10^{-4}	4.39×10^{-7}	7.22×10^{-7}	3.75×10^{-3}	7.55×10^{-7}	1.57×10^{-6}
Gender	Male	4.35×10^{-5}	2.40×10^{-9}	1.15×10^{-9}	1.12×10^{-4}	2.41×10^{-8}	4.85×10^{-9}	1.28×10^{-3}	5.90×10^{-7}	8.31×10^{-6}	7.91×10^{-4}	4.93×10^{-7}	4.10×10^{-7}	3.56×10^{-3}	8.35×10^{-7}	9.08×10^{-7}
	Female	5.06×10^{-5}	2.01×10^{-9}	8.18×10^{-10}	1.30×10^{-4}	1.97×10^{-8}	3.67×10^{-9}	1.52×10^{-3}	4.66×10^{-7}	6.28×10^{-6}	8.95×10^{-4}	4.11×10^{-7}	3.09×10^{-7}	3.95×10^{-3}	7.13×10^{-7}	6.65×10^{-7}
Age	18-44	4.91×10^{-5}	2.82×10^{-9}	7.14×10^{-10}	1.30×10^{-4}	2.53×10^{-8}	3.68×10^{-9}	1.79×10^{-3}	5.56×10^{-7}	6.06×10^{-6}	1.01×10^{-3}	4.59×10^{-7}	3.20×10^{-7}	3.92×10^{-3}	9.21×10^{-7}	6.22×10^{-7}
	45-59	4.56×10^{-5}	1.88×10^{-9}	1.20×10^{-9}	1.18×10^{-4}	1.91×10^{-8}	5.44×10^{-9}	1.19×10^{-3}	5.26×10^{-7}	9.49×10^{-6}	8.00×10^{-4}	4.59×10^{-7}	4.59×10^{-7}	3.56×10^{-3}	7.31×10^{-7}	1.00×10^{-6}
	≥60	4.62×10^{-5}	1.65×10^{-9}	1.12×10^{-9}	1.13×10^{-4}	1.96×10^{-8}	3.85×10^{-9}	1.09×10^{-3}	4.84×10^{-7}	6.75×10^{-6}	6.60×10^{-4}	4.31×10^{-7}	3.12×10^{-7}	3.73×10^{-3}	6.07×10^{-7}	7.92×10^{-7}

Table S8. The exposure contribution of each media (expressed as %)

Metal		Hg		Cd		As		Pb		Cr	
		Non-dietary	Dietary	Non-dietary	Dietary	Non-dietary	Dietary	Non-dietary	Dietary	Non-dietary	Dietary
All		6.2040	93.7960	2.5101	97.4899	12.1898	87.8102	4.5977	95.4023	5.6408	94.3592
Region	Taiyun	5.3351	94.6649	1.0444	98.9556	13.2300	86.7700	10.4507	89.5493	18.8859	81.1141
	Dalian	0.4268	99.5732	0.5436	99.4564	0.4507	99.5493	2.1556	97.8444	0.5828	99.4172
	Shanghai	2.2090	97.7910	0.8710	99.1290	2.3130	97.6870	2.3373	97.6627	1.4350	98.5650
	Wuhan	12.0329	87.9671	1.5035	98.4965	15.5127	84.4873	5.2515	94.7485	9.9148	90.0852
	Chengdu	1.2821	98.7179	1.3828	98.6172	2.9577	97.0423	0.8302	99.1698	1.3372	98.6628
	Lanzhou	15.0372	84.9628	9.1356	90.8644	35.8055	64.1945	5.7149	94.2851	0.5642	99.4358
Area	Urban	7.1985	92.8015	1.9178	98.0822	10.6961	89.3039	2.7266	97.2734	3.8489	96.1511
	Rural	5.1517	94.8483	3.1368	96.8632	13.7705	86.2295	6.5776	93.4224	7.5369	92.4631

Metal		Hg		Cd		As		Pb		Cr	
		Non-dietary	Dietary	Non-dietary	Dietary	Non-dietary	Dietary	Non-dietary	Dietary	Non-dietary	Dietary
Gender	Male	6.7518	93.2482	2.8389	97.1611	13.3677	86.6323	4.9967	95.0033	5.8979	94.1021
	Female	5.6922	94.3078	2.2028	97.7972	11.0894	88.9106	4.2248	95.7752	5.4005	94.5995
Age	18-44	5.3111	94.6889	2.1150	97.8850	9.0113	90.9887	3.5755	96.4245	4.0834	95.9166
	45-59	6.7438	93.2562	2.6754	97.3246	13.8917	86.1083	5.1202	94.8798	6.6483	93.3517
	≥60	6.9116	93.0884	2.8947	97.1053	14.9226	85.0774	5.5005	94.4995	6.8106	93.1894

Table S9. The exposure contribution of each route (expressed as %)

Metal		Hg			Cd			As			Pb			Cr		
		Ingestion	Inhalation	Dermal contact	Ingestion	Inhalation	Dermal contact	Ingestion	Inhalation	Dermal contact	Ingestion	Inhalation	Dermal contact	Ingestion	Inhalation	Dermal contact
All		99.9903	0.0053	0.0044	99.9561	0.0343	0.0096	97.8307	0.2627	1.9066	99.7638	0.1541	0.0821	99.9137	0.0387	0.0476
Region	Taiyuan	99.9924	0.0030	0.0046	99.9839	0.0132	0.0029	98.5691	0.0143	1.4166	99.3473	0.4970	0.1557	99.7702	0.0503	0.1795
	Dalian	99.9957	0.0040	0.0004	99.9487	0.0489	0.0024	99.9104	0.0110	0.0786	99.9014	0.0490	0.0496	99.9550	0.0315	0.0134
	Shanghai	99.9921	0.0076	0.0003	99.9970	0.0029	0.0001	99.7787	0.0727	0.1486	99.9692	0.0268	0.0040	99.8964	0.0967	0.0070
	Wuhan	99.9918	0.0057	0.0025	99.9841	0.0149	0.0011	97.8649	0.0461	2.0890	99.8207	0.1347	0.0446	99.9309	0.0404	0.0288
	Chengdu	99.9886	0.0094	0.0020	99.9845	0.0107	0.0048	98.8892	0.0146	1.0963	99.9482	0.0230	0.0287	99.9610	0.0170	0.0220
	Lanzhou	99.9816	0.0028	0.0156	99.8477	0.1093	0.0430	92.5089	1.3293	6.1619	99.6617	0.1507	0.1876	99.9777	0.0014	0.0209
Area	Urban	99.9930	0.0061	0.0009	99.9653	0.0341	0.0006	99.6146	0.3154	0.0700	99.8533	0.1433	0.0035	99.9660	0.0326	0.0014
	Rural	99.9873	0.0044	0.0082	99.9464	0.0346	0.0190	95.9429	0.2070	3.8501	99.6691	0.1656	0.1653	99.8584	0.0451	0.0965
Gender	Male	99.9882	0.0062	0.0055	99.9481	0.0402	0.0117	97.3808	0.3116	2.3076	99.7193	0.1783	0.1023	99.8937	0.0445	0.0619
	Female	99.9921	0.0045	0.0034	99.9636	0.0289	0.0075	98.2510	0.2170	1.5320	99.8054	0.1315	0.0632	99.9324	0.0333	0.0343
Age	18-44	99.9898	0.0067	0.0035	99.9583	0.0344	0.0073	98.4646	0.2313	1.3042	99.8063	0.1360	0.0577	99.9281	0.0404	0.0315
	45-59	99.9893	0.0048	0.0059	99.9533	0.0337	0.0130	97.1750	0.2738	2.5512	99.7179	0.1755	0.1066	99.8995	0.0429	0.0576
	≥60	99.9917	0.0039	0.0044	99.9558	0.0349	0.0093	97.5937	0.2953	2.1110	99.7497	0.1583	0.0920	99.9076	0.0322	0.0602

Table S10. The hazard index (HI) of Hg, Cd, As, Pb and Cr

Metal		Hg			Cd			As			Pb			Cr		
		Mean	P5	P95	Mean	P5	P95	Mean	P5	P95	Mean	P5	P95	Mean	P5	P95
All		1.11E-02	2.33E-03	2.66E-02	1.17E-02	2.63E-03	2.48E-02	4.23E-01	9.51E-02	8.24E-01	1.04E-02	3.35E-03	2.15E-02	7.14E-02	6.20E-03	1.43E-01
Region	Taiyun	7.40E-03	5.30E-03	1.05E-02	5.85E-03	3.10E-03	9.40E-03	1.39E-01	6.29E-02	2.79E-01	1.16E-02	7.12E-03	1.84E-02	1.75E-02	4.20E-03	8.16E-02
	Dalian	2.28E-02	1.55E-02	3.38E-02	2.28E-02	1.52E-02	3.56E-02	6.93E-01	4.93E-01	1.01E+00	6.86E-03	3.17E-03	1.09E-02	1.07E-01	6.91E-02	1.58E-01
	Shanghai	8.15E-03	5.79E-03	1.20E-02	3.08E-03	2.13E-03	4.32E-03	3.81E-01	2.66E-01	5.32E-01	3.82E-03	2.84E-03	4.98E-03	6.57E-02	5.03E-02	8.26E-02
	Wuhan	1.61E-02	6.55E-03	3.12E-02	1.63E-02	1.17E-02	2.23E-02	2.71E-01	1.17E-01	5.11E-01	1.48E-02	1.04E-02	1.96E-02	6.40E-02	3.44E-02	9.95E-02
	Chengdu	1.02E-02	6.60E-03	1.60E-02	1.40E-02	7.22E-03	2.16E-02	5.48E-01	2.58E-01	9.05E-01	1.62E-02	6.03E-03	3.10E-02	9.19E-02	5.04E-02	1.54E-01

Metal		Hg			Cd			As			Pb			Cr		
		Mean	P5	P95	Mean	P5	P95	Mean	P5	P95	Mean	P5	P95	Mean	P5	P95
	Lanzhou	3.08E-03	1.78E-03	5.10E-03	8.97E-03	5.23E-03	1.38E-02	5.27E-01	2.46E-01	7.78E-01	9.04E-03	5.37E-03	1.25E-02	8.70E-02	4.79E-02	1.51E-01
Area	Urban	1.03E-02	2.89E-03	2.47E-02	1.19E-02	3.00E-03	2.85E-02	4.15E-01	8.45E-02	7.78E-01	9.90E-03	3.33E-03	1.94E-02	6.22E-02	5.06E-03	1.17E-01
	Rural	1.20E-02	2.07E-03	2.91E-02	1.15E-02	2.43E-03	2.30E-02	4.32E-01	1.19E-01	8.76E-01	1.10E-02	3.39E-03	2.19E-02	8.12E-02	1.09E-02	1.53E-01
Gender	Male	1.02E-02	2.22E-03	2.47E-02	1.24E-02	2.58E-03	2.75E-02	4.29E-01	9.19E-02	7.99E-01	1.13E-02	3.69E-03	2.25E-02	7.39E-02	6.49E-03	1.44E-01
	Female	1.19E-02	2.46E-03	2.85E-02	1.11E-02	2.71E-03	2.28E-02	4.18E-01	9.86E-02	8.40E-01	9.65E-03	3.13E-03	2.04E-02	6.92E-02	6.07E-03	1.42E-01
Age	18-44	1.15E-02	2.40E-03	2.65E-02	1.33E-02	2.76E-03	2.91E-02	4.58E-01	9.91E-02	8.78E-01	1.07E-02	3.57E-03	2.51E-02	7.64E-02	6.80E-03	1.45E-01
	45-59	1.07E-02	2.32E-03	2.73E-02	1.06E-02	2.48E-03	2.29E-02	4.15E-01	9.77E-02	8.31E-01	1.07E-02	3.31E-03	2.15E-02	7.11E-02	6.58E-03	1.46E-01
	≥60	1.08E-02	2.28E-03	2.63E-02	1.06E-02	2.68E-03	2.10E-02	3.83E-01	8.49E-02	7.62E-01	9.74E-03	3.10E-03	1.80E-02	6.49E-02	4.84E-03	1.33E-01

Table S11. The hazard index (HI) of Hg, Cd, As, Pb and Cr in each exposure medium

Metal		Hg		Cd		As		Pb		Cr	
		Non-dietary	Dietary	Non-dietary	Dietary	Non-dietary	Dietary	Non-dietary	Dietary	Non-dietary	Dietary
All		4.98E-04	1.06E-02	8.71E-03	3.00E-03	2.62E-01	1.61E-01	9.41E-03	1.03E-03	4.07E-02	3.07E-02
Region	Taiyuan	4.62E-04	6.94E-03	3.88E-03	1.97E-03	1.15E-01	2.45E-02	1.15E-02	1.42E-04	1.53E-02	2.21E-03
	Dalian	1.55E-04	2.26E-02	1.94E-02	3.35E-03	1.73E-01	5.20E-01	6.06E-03	7.91E-04	6.52E-02	4.15E-02
	Shanghai	2.12E-04	7.94E-03	9.31E-04	2.15E-03	2.55E-01	1.27E-01	3.05E-03	7.70E-04	5.24E-02	1.34E-02
	Wuhan	1.40E-03	1.47E-02	1.12E-02	5.08E-03	2.33E-01	3.76E-02	1.41E-02	7.36E-04	4.51E-02	1.89E-02
	Chengdu	2.14E-04	1.00E-02	8.71E-03	5.29E-03	2.68E-01	2.80E-01	1.27E-02	3.51E-03	4.75E-02	4.45E-02
	Lanzhou	5.45E-04	2.53E-03	8.49E-03	4.73E-04	5.18E-01	8.65E-03	8.66E-03	3.81E-04	2.40E-02	6.29E-02
Area	Urban	5.29E-04	9.73E-03	9.02E-03	2.92E-03	2.50E-01	1.65E-01	8.94E-03	9.60E-04	3.12E-02	3.09E-02
	Rural	4.65E-04	1.15E-02	8.38E-03	3.08E-03	2.75E-01	1.57E-01	9.91E-03	1.11E-03	5.08E-02	3.05E-02
Gender	Male	5.11E-04	9.73E-03	9.61E-03	2.76E-03	2.82E-01	1.47E-01	1.03E-02	9.67E-04	4.48E-02	2.90E-02
	Female	4.86E-04	1.14E-02	7.87E-03	3.22E-03	2.44E-01	1.75E-01	8.55E-03	1.10E-03	3.69E-02	3.23E-02
Age	18-44	4.27E-04	1.11E-02	1.00E-02	3.22E-03	2.52E-01	2.06E-01	9.49E-03	1.24E-03	4.42E-02	3.22E-02
	45-59	5.23E-04	1.02E-02	7.70E-03	2.91E-03	2.79E-01	1.36E-01	9.77E-03	9.76E-04	4.21E-02	2.90E-02
	≥60	5.72E-04	1.03E-02	7.86E-03	2.78E-03	2.59E-01	1.24E-01	8.94E-03	8.02E-04	3.46E-02	3.03E-02

Table S12. The hazard index (HI) of Hg, Cd, As, Pb and Cr in each exposure route

Metal	Hg			Cd			As			Pb			Cr		
	Inhalation	Ingestion	Dermal contact	Inhalation	Ingestion	Dermal contact	Inhalation	Ingestion	Dermal contact	Inhalation	Ingestion	Dermal contact	Inhalation	Ingestion	Dermal contact

All		2.82E-05	1.10E-02	4.65E-05	8.39E-03	3.15E-03	1.70E-04	1.35E-01	2.64E-01	2.42E-02	8.67E-03	1.06E-03	7.16E-04	2.97E-02	3.13E-02	1.04E-02
Region	Taiyuan	1.13E-05	7.32E-03	6.74E-05	3.71E-03	2.04E-03	9.10E-05	8.75E-03	1.17E-01	1.34E-02	1.10E-02	1.59E-04	4.11E-04	6.26E-03	2.73E-03	8.56E-03
	Dalian	4.67E-05	2.27E-02	1.73E-05	1.92E-02	3.41E-03	1.39E-04	1.14E-01	5.69E-01	1.02E-02	5.46E-03	8.06E-04	5.93E-04	5.63E-02	4.17E-02	8.75E-03
	Shanghai	3.26E-05	8.11E-03	4.29E-06	8.56E-04	2.22E-03	3.59E-06	1.79E-01	1.97E-01	5.06E-03	2.98E-03	7.88E-04	4.57E-05	5.10E-02	1.35E-02	1.25E-03
	Wuhan	3.00E-05	1.60E-02	7.19E-05	1.08E-02	5.36E-03	9.06E-05	4.06E-02	2.04E-01	2.62E-02	1.36E-02	7.70E-04	4.40E-04	3.54E-02	2.08E-02	7.81E-03
	Chengdu	4.90E-05	1.02E-02	3.84E-05	8.09E-03	5.54E-03	3.72E-04	8.38E-02	3.99E-01	6.53E-02	1.08E-02	3.54E-03	1.92E-03	3.19E-02	4.50E-02	1.50E-02
	Lanzhou	4.13E-06	3.00E-03	7.30E-05	8.01E-03	6.49E-04	3.08E-04	3.74E-01	1.27E-01	2.49E-02	7.78E-03	3.98E-04	8.65E-04	3.71E-03	6.33E-02	2.00E-02
Area	Urban	3.16E-05	1.02E-02	9.61E-06	8.87E-03	3.06E-03	9.58E-06	1.54E-01	2.61E-01	7.57E-04	8.89E-03	9.74E-04	2.72E-05	3.03E-02	3.14E-02	4.80E-04
	Rural	2.46E-05	1.19E-02	8.56E-05	7.88E-03	3.24E-03	3.39E-04	1.15E-01	2.68E-01	4.90E-02	8.43E-03	1.14E-03	1.44E-03	2.90E-02	3.12E-02	2.10E-02
Gender	Male	3.08E-05	1.02E-02	5.46E-05	9.27E-03	2.91E-03	1.94E-04	1.51E-01	2.50E-01	2.77E-02	9.48E-03	9.88E-04	8.19E-04	3.21E-02	2.96E-02	1.21E-02
	Female	2.58E-05	1.18E-02	3.89E-05	7.57E-03	3.37E-03	1.47E-04	1.19E-01	2.78E-01	2.09E-02	7.91E-03	1.12E-03	6.19E-04	2.74E-02	3.29E-02	8.86E-03
Age	18-44	3.62E-05	1.15E-02	3.40E-05	9.75E-03	3.37E-03	1.47E-04	1.43E-01	2.96E-01	2.02E-02	8.83E-03	1.26E-03	6.39E-04	3.54E-02	3.26E-02	8.29E-03
	45-59	2.42E-05	1.06E-02	5.70E-05	7.33E-03	3.05E-03	2.17E-04	1.35E-01	2.49E-01	3.16E-02	8.83E-03	1.00E-03	9.19E-04	2.81E-02	2.97E-02	1.33E-02
	≥60	2.11E-05	1.08E-02	5.35E-05	7.54E-03	2.94E-03	1.54E-04	1.24E-01	2.36E-01	2.25E-02	8.29E-03	8.25E-04	6.23E-04	2.33E-02	3.11E-02	1.06E-02

Table S13. The total life time cancer risk (TLICR) of As, Pb and Cr

Metal		As			Pb			Cr		
		Mean	P5	P95	Mean	P5	P95	Mean	P5	P95
All		7.32E-05	1.59E-05	1.93E-04	9.00E-08	1.68E-08	3.11E-07	1.83E-04	1.98E-05	4.21E-04
Region	Taiyuan	4.08E-05	1.25E-05	8.96E-05	3.52E-08	1.44E-08	5.96E-08	4.91E-05	1.09E-05	1.39E-04
	Dalian	1.21E-04	3.34E-05	2.61E-04	6.94E-08	1.62E-08	1.50E-07	3.20E-04	1.41E-04	5.24E-04
	Shanghai	4.96E-05	1.53E-05	1.08E-04	5.58E-08	1.52E-08	1.15E-07	2.59E-04	1.24E-04	3.90E-04
	Wuhan	7.46E-05	1.37E-05	1.71E-04	9.63E-08	2.10E-08	1.99E-07	2.24E-04	9.71E-05	4.64E-04
	Chengdu	9.17E-05	1.84E-05	2.24E-04	2.36E-07	3.93E-08	6.22E-07	1.77E-04	4.92E-05	3.71E-04
	Lanzhou	6.58E-05	2.09E-05	1.32E-04	5.39E-08	1.64E-08	1.09E-07	9.56E-05	2.66E-05	1.96E-04
Area	Urban	6.09E-05	1.43E-05	1.69E-04	7.26E-08	1.59E-08	1.95E-07	1.64E-04	1.62E-05	4.20E-04
	Rural	8.63E-05	1.88E-05	2.17E-04	1.08E-07	1.93E-08	4.17E-07	2.03E-04	3.99E-05	4.22E-04
Gender	Male	7.16E-05	1.54E-05	1.84E-04	8.64E-08	1.62E-08	2.83E-07	1.92E-04	2.24E-05	4.34E-04

Metal		As			Pb			Cr		
		Mean	P5	P95	Mean	P5	P95	Mean	P5	P95
	Female	7.48E-05	1.70E-05	2.03E-04	9.33E-08	1.80E-08	3.26E-07	1.74E-04	1.84E-05	4.12E-04
Age	18-44	4.36E-05	1.25E-05	1.05E-04	5.08E-08	1.36E-08	1.70E-07	1.39E-04	1.61E-05	3.15E-04
	45-59	7.98E-05	2.46E-05	1.88E-04	9.74E-08	2.71E-08	3.49E-07	1.96E-04	2.33E-05	4.18E-04
	≥60	1.08E-04	2.76E-05	2.45E-04	1.37E-07	3.93E-08	4.80E-07	2.29E-04	2.33E-05	5.02E-04

Table S14. The total life time cancer risk (TLICR) of As, Pb and Cr in each exposure medium

Metal		As		Pb		Cr	
		Non-dietary	Dietary	Non-dietary	Dietary	Non-dietary	Dietary
All		4.43E-05	2.89E-05	1.79E-08	7.21E-08	1.61E-04	2.14E-05
Region	Taiyuan	3.46E-05	6.19E-06	2.19E-08	1.33E-08	4.73E-05	1.83E-06
	Dalian	2.12E-05	9.94E-05	1.13E-08	5.81E-08	2.94E-04	2.64E-05
	Shanghai	2.78E-05	2.18E-05	6.05E-09	4.98E-08	2.51E-04	7.54E-06
	Wuhan	6.54E-05	9.18E-06	2.80E-08	6.83E-08	2.08E-04	1.55E-05
	Chengdu	5.18E-05	3.99E-05	2.18E-08	2.14E-07	1.55E-04	2.25E-05
	Lanzhou	6.36E-05	2.20E-06	1.75E-08	3.64E-08	4.25E-05	5.31E-05
Area	Urban	3.44E-05	2.64E-05	1.45E-08	5.81E-08	1.44E-04	2.03E-05
	Rural	5.48E-05	3.15E-05	2.15E-08	8.69E-08	1.80E-04	2.26E-05
Gender	Male	4.62E-05	2.54E-05	1.94E-08	6.70E-08	1.72E-04	2.00E-05
	Female	4.26E-05	3.22E-05	1.65E-08	7.68E-08	1.51E-04	2.27E-05
Age	18-44	2.58E-05	1.78E-05	1.18E-08	3.90E-08	1.30E-04	9.33E-06
	45-59	5.08E-05	2.90E-05	2.00E-08	7.74E-08	1.76E-04	2.03E-05
	≥60	6.36E-05	4.42E-05	2.44E-08	1.13E-07	1.90E-04	3.93E-05

Table S15. The total life time cancer risk (TLICR) of As, Pb and Cr in each exposure route

Metal		As			Pb			Cr		
		Inhalation	Ingestion	Dermal Contact	Inhalation	Ingestion	Dermal Contact	Inhalation	Ingestion	Dermal Contact
All		5.73E-06	6.02E-05	7.34E-06	1.34E-08	7.45E-08	2.02E-09	1.50E-04	2.21E-05	1.07E-05
Region	Taiyuan	4.14E-07	3.59E-05	4.46E-06	1.85E-08	1.54E-08	1.29E-09	3.72E-05	2.39E-06	9.49E-06
	Dalian	4.54E-06	1.13E-04	3.04E-06	8.04E-09	5.97E-08	1.66E-09	2.85E-04	2.66E-05	8.66E-06
	Shanghai	6.87E-06	4.12E-05	1.56E-06	4.14E-09	5.15E-08	1.32E-10	2.50E-04	7.69E-06	1.28E-06
	Wuhan	1.84E-06	6.35E-05	9.28E-06	2.22E-08	7.26E-08	1.47E-09	1.97E-04	1.75E-05	9.18E-06
	Chengdu	2.78E-06	7.10E-05	1.79E-05	1.39E-08	2.17E-07	4.95E-09	1.41E-04	2.31E-05	1.37E-05
	Lanzhou	1.74E-05	4.06E-05	7.77E-06	1.29E-08	3.85E-08	2.53E-09	2.14E-05	5.35E-05	2.08E-05
Area	Urban	6.27E-06	5.44E-05	1.97E-07	1.30E-08	5.96E-08	6.81E-11	1.43E-04	2.08E-05	4.33E-07
	Rural	5.15E-06	6.63E-05	1.49E-05	1.39E-08	9.04E-08	4.09E-09	1.58E-04	2.34E-05	2.16E-05
Gender	Male	6.33E-06	5.68E-05	8.48E-06	1.47E-08	6.95E-08	2.33E-09	1.59E-04	2.07E-05	1.25E-05
	Female	5.16E-06	6.34E-05	6.28E-06	1.23E-08	7.93E-08	1.74E-09	1.42E-04	2.34E-05	9.05E-06
Age	18-44	4.08E-06	3.52E-05	4.32E-06	9.06E-09	4.04E-08	1.29E-09	1.24E-04	9.63E-06	5.96E-06
	45-59	6.07E-06	6.39E-05	9.77E-06	1.45E-08	8.02E-08	2.66E-09	1.62E-04	2.10E-05	1.37E-05

Metal	As			Pb			Cr		
	Inhalation	Ingestion	Dermal Contact	Inhalation	Ingestion	Dermal Contact	Inhalation	Ingestion	Dermal Contact
≥60	7.67E-06	9.10E-05	9.15E-06	1.84E-08	1.16E-07	2.40E-09	1.74E-04	4.03E-05	1.44E-05

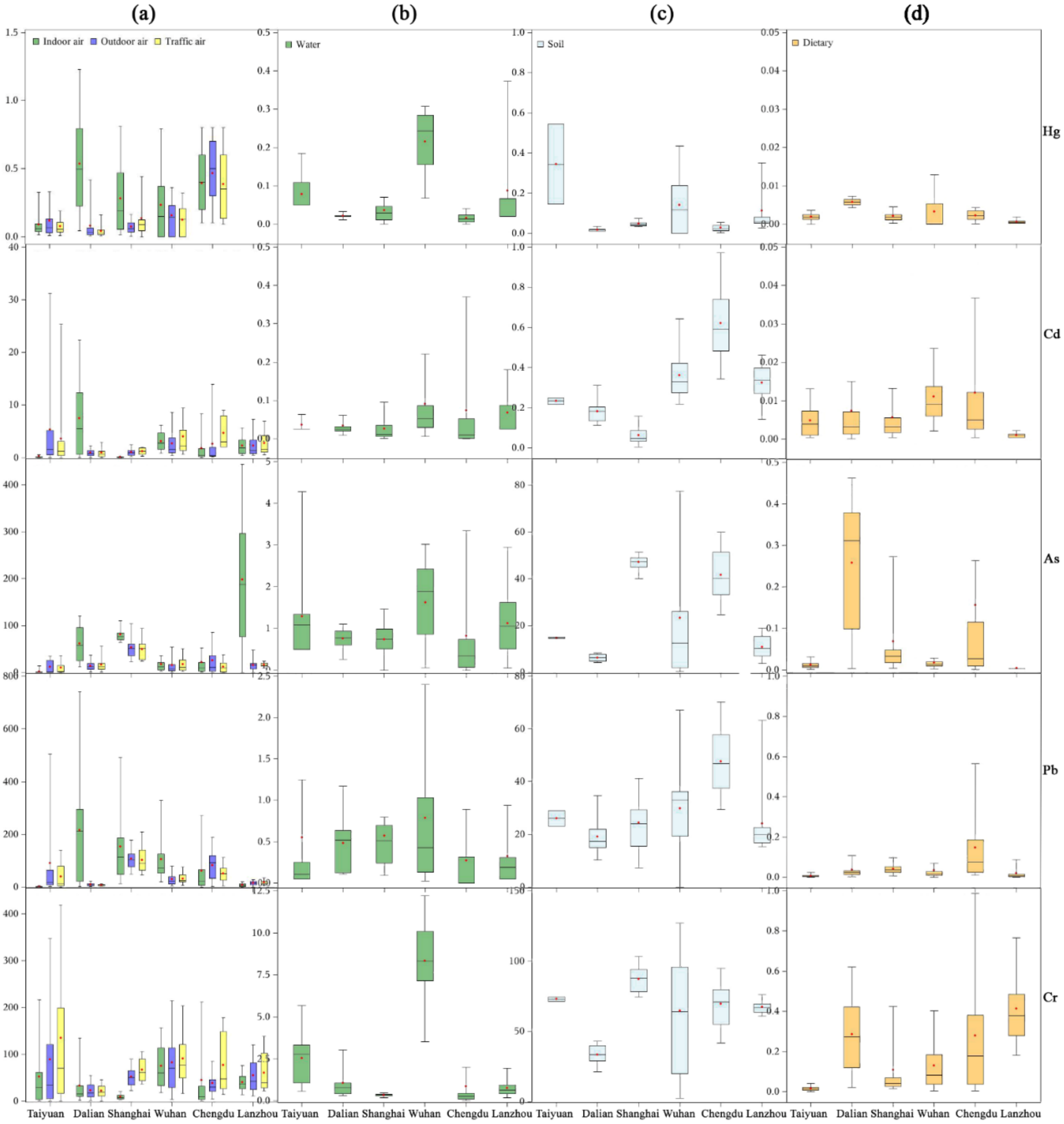

Figure S1. The concentration of Hg, Cd, As, Pb and Cr in each exposure media. Boxes represent 25th, 50th (shown by the horizontal line) and 75th percentiles. Whiskers indicate 5th and 95th values and the red filled dots show mean values. The first column in the figure marked (a) is the concentration of indoor air (box in green), outdoor air (box in blue) and the traffic air (box in yellow) in ng/m^3; the second column in the figure marked (b) is the concentration of water(box in green) in μg/L; the third column in the figure marked (c) is the concentration of soil(box in light blue) in mg/kg; the fourth column in the figure marked (d) is the concentration of dietary(box in orange) in mg/kg. For the detailed data was shown in Table S3.

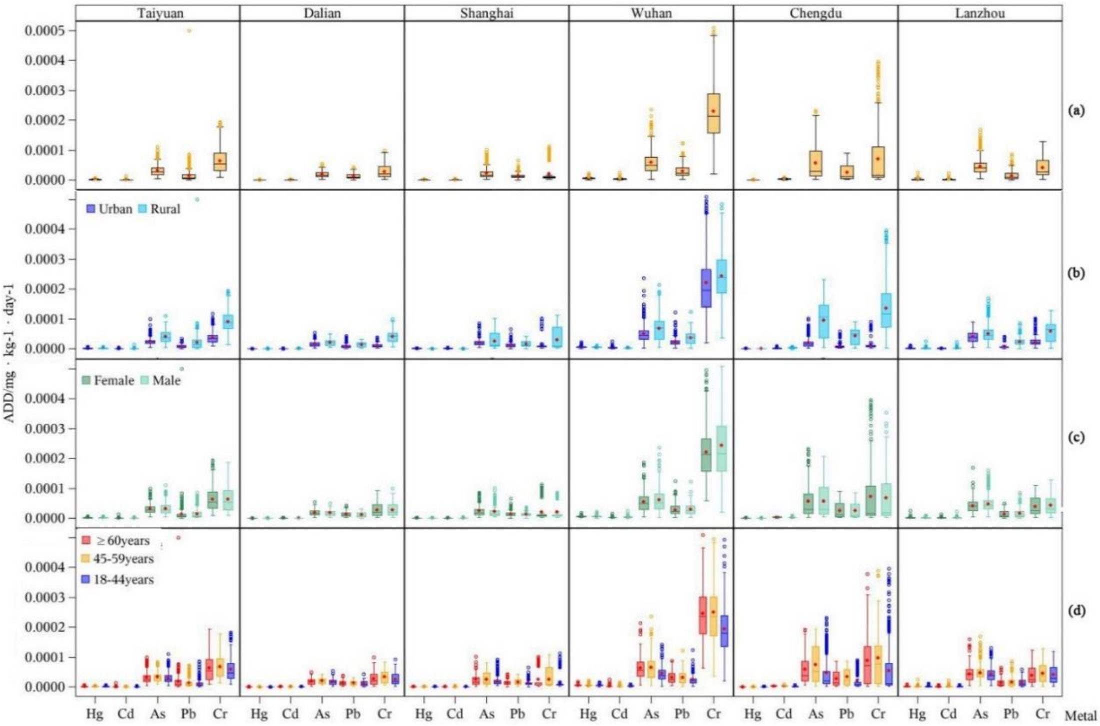

Figure S2. The non-dietary daily exposure doses of metals in different areas, sex, and age groups. Boxes represent 25th, 50th (shown by the horizontal line) and 75th percentiles. Whiskers indicate 5th and 95th values and the circles show outliers, the red filled dots in boxes show mean values. The first row in the figure marked (a) is the non-dietary exposure level of areas (in orange); the second row in the figure marked (b) is the non-dietary exposure level of urban(in blue) and rural(in light blue) areas; the third row in the figure marked (c) is the non-dietary exposure level of male(in green) and female(in light green); the fourth row in the figure marked (d) is the non-dietary exposure level of age groups(>60 years old in red, 45-59 years old in yellow and 18-44 years old in blue). For the detailed data was shown in Table S5.

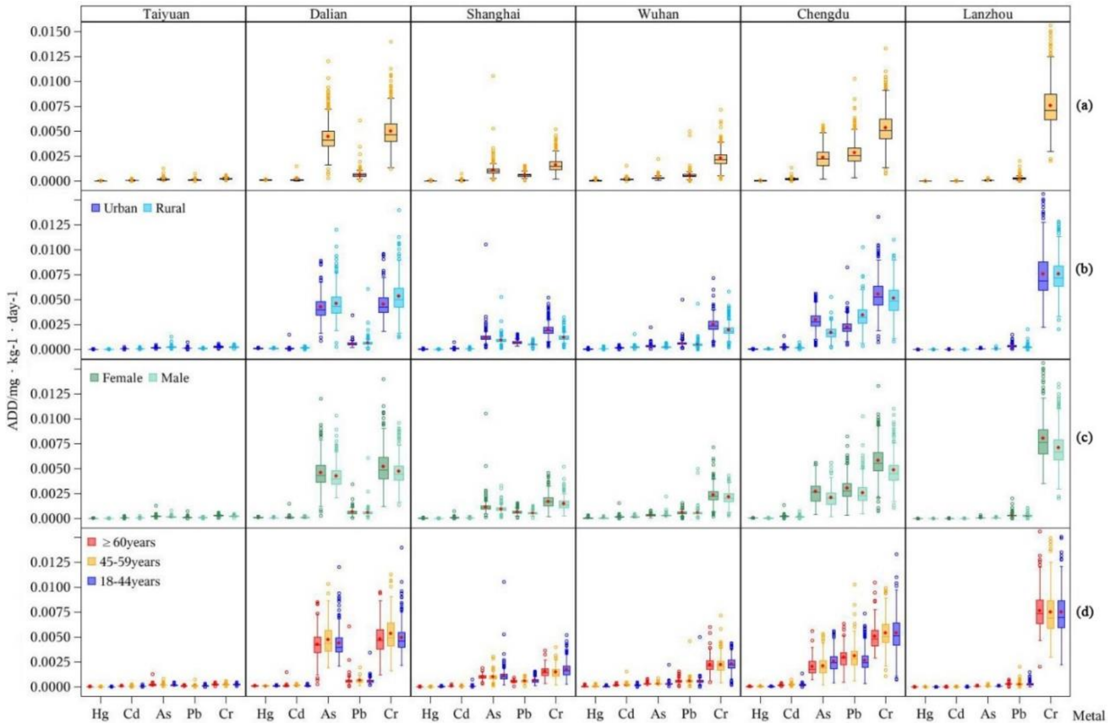

Figure S3. The dietary daily exposure doses of metals in different areas, sex, and age groups. Boxes represent 25th, 50th (shown by the horizontal line) and 75th percentiles. Whiskers indicate 5th and 95th values and the circles show outliers, the red filled dots in boxes show mean values. The first row in the figure marked (a) is the dietary exposure level of areas (in orange); the second row in the figure marked (b) is the dietary exposure level of urban(in blue) and rural(in light blue) areas; the third row in the figure marked (c) is the dietary exposure level of male(in green) and female(in light green); the fourth row in the figure marked (d) is the dietary exposure level of age groups(>60 years old in red, 45-59 years old in yellow and 18-44 years old in blue). For the detailed data was shown in Table S6.