Spatial-Temporal Analysis of Heavy Metal Water Pollution and the Impact on Public Health in China

by

Huixuan Li

A thesis submitted to the Graduate Faculty of
Auburn University
in partial fulfillment of the
requirements for the Degree of
Master of Science in Geography

Auburn, Alabama
May 9, 2015

Keywords: Heavy metal water pollution, public health, cancers, GIS, spatial statistics, China

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Approved by

Yingru Li, Chair, Assistant Professor of Geography, Department of Geosciences
Luke Marzen, Professor of Geography, Department of Geosciences
Ming-Kuo Lee, Professor of Geology, Department of Geosciences

Abstract

Water is the source of life. China’s water pollution has become a burning research field because of the severely polluted conditions. 60 percent of the country's rivers were not potable according to the State Environmental Protection Administration of China. This study will advance the knowledge of heavy metal water pollution in China from a spatial-temporal perspective. Specifically, this study addressed the following: (1) spatial patterns of heavy metal water pollution levels were analyzed using data collected within prefecture-level cities from 2004 to 2011 using GIS techniques. (2) Spatial statistical methods were used to examine the underlying socioeconomic and physical factors behind water pollution including human activities (transportation, urbanization, industrialization, globalization, agriculture, mining), and environmental characteristic (hydrology and vegetation coverage). (3) Impacts of heavy metal water pollution on public health were analyzed using kernel estimation and multiple regression analysis. The results show that highest pollution levels are most concentrated in central and east areas of China. Transportation, urbanization, economic development, agriculture, mining activities, as well as greenery coverage are all significantly related to heavy metal water pollution. As expected, the analytical results of seven cancer diseases were consistent with the distribution of heavy metals. Five clusters of cancer incidences were detected in

Beijing, Shanghai, Guangzhou, Shenyang, and Wuhan. In addition, the analytical findings will provide valuable information for policy-makers to initiate and adjust protocols and strategies for protecting water sources and controlling water pollution; thus improving the quality of living environments.

Key words: heavy metal water pollution, public health, cancers, China, GIS, Spatial Statistics

Acknowledgement

Studying abroad in the United States at Auburn University is the most important turning point in my life. I am honored to have Dr. Luke Marzen’s support to obtain this opportunity. I feel extremely fortunate to have Dr. Yingru Li as my advisor. She more than just teaches me professional knowledge; she also serves as an excellent mentor. I cannot imagine finishing this Master’s degree successfully without her. Her influence is so profound and positive, and I am very proud of what I have become-- professional and confident. I am also very grateful to have Dr. Ming-Kuo Lee and Dr. Luke Marzen as my committee members. They provided so much professional advice which I benefited quite well from.

I would like to send my best regard to my friends and family. My dearest mom, who loves me unconditionally and took great care of me when she visited the United States. She helped me through the most difficult time. I want to thank my father, who provided so much help on data collection and professional opinions on my research. I also thank Andy Hug, who is the best friend and English teacher ever and helped me in revising my thesis. Finally, I am very grateful to have the support from the Geography and Geology Department of Auburn University and all the people who gave me their valuable comments and suggestions.

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1. Introduction

People’s standards of living continue to increase, along with their desires to live long and healthy lives (Zhang et al., 2010). Water is the source of life; however, only 0.26% of the Earth’s total water is usable fresh water. According to a United Nations’ report, water quality problems have caused about 3.5 million deaths annually (UNDESA, 2014). Water pollution studies have become a research frontier due to the importance of academic, public and political concerns (Ebenstein, 2012; Schwarzenbach et al., 2010; Zhang et al., 2010; Li et al., 2014). Although heavy metals widely exist in natural environments, heavy metal pollution is undoubtedly accelerated by anthropogenic activities. In addition, the food chain, which serves as the main intake of pollutants by humans, can cause severe long-term health problems at both local scales, such as enclosed water systems and global scales (Lake, et. al., 2003; Meng, et. al. 2008; Li et al., 2014; Wang et al., 2014).

China is facing serious shortages of water resources and severe water pollution issues have caused great concern, especially from heavy metals. The total amount of water resources in China ranks fourth in the world; however, the water supply per capita is only one-fourth of the world average (World Resource Institute, 2007; Bao et al., 2012). According to China's State Environmental Protection

Administration (SEPA), 60 percent of the country's rivers suffered from pollution so severely that they were not potable. For example, the lead levels of Chinese rivers are approximately 44 times higher than the water quality standard based on a National Academy of Sciences report. Heavy metal contamination in water bodies can cause long-term human health risks creating significant consequences in Chinese economic development, and limiting environmental sustainability (Liu and Diamond, 2005; Carr et al., 2008). Given the academic and political concerns, investigating heavy metal water pollution and its consequences have become hot topics. Many studies have focused on one specific region such as a river or a province of China (Zhai, et al., 2008; Meng et al., 2008; Huang et al., 2010; Bao et al., 2012; Jiang et al., 2012; Sheng et al., 2012; Wang et al., 2013). However, studies concerning spatial-temporal patterns of water pollution across mainland China are limited and merit further investigation. The goal of this study is to investigate the spatial-temporal variation of heavy metal water pollution, to explore the underlying social-economic and environmental factors, and to examine their impacts on public health in China. Three research questions are proposed: (1) Is the heavy metal water pollution in China sensitive to spatial autocorrelation? (2) Is the heavy metal water pollution positively related to human activities and physical environment? (3) How does heavy metal water pollution impact public health indicated by cancer patient incidences?

This thesis mainly investigates five types of heavy metal water pollution levels and seven types of cancers and their correlated factors. To fulfill the research objectives, the thesis is organized as five chapters, namely, introduction, literature review, methodology, results and interpretation, and conclusions and significance. Following the Introduction section, the Literature Review summarizes previous water pollution research and applied methods. The interactions between heavy metal water pollution and human health studies are also included. The thesis objectives are proposed based on the limitations of previous research at the end of the literature review section. The Methodology section discusses study area, data and data sources, followed by, applied methods, including kriging models, spatial statistics etc. The Results and Interpretation section summarizes the spatial and temporal variations of heavy metal water pollution and public health conditions, in addition to the interactions between China's socioeconomic transitions, heavy metal water contamination, and cancer incidences. The conclusions and significance section highlights results and findings, in addition to limitations and future directions.

2. Literature Review

This section briefly summarizes achievements of previous studies, basic theories and methods used in investigating heavy metal contamination associated with water pollution, as well as the interaction between heavy metals and human health.

2.1 Water pollution research

The rapid development of industrialization in England in the 1850s resulted in the extreme pollution of Thames River. People directly discharged untreated sewage and industrial wastewater into the river producing serious water pollution. This event received global attention and made people realize the importance of environmental protection. Many scholars have investigated water pollution, and previous studies have mainly contributed to identifying water pollution sources and controlling contamination (Coskun et al., 2006; Asadi et al., 2007; Satapathy et al., 2009; Singare et al., 2011; Dede et al., 2013). Scholars use collected water samples from a local scale water system to analyze parameters including temperature, pH, turbidity, etc. by employing physical-chemical experiments. Then they compare the concentration of heavy metals in samples with the national water quality standards to evaluate potential human health risks (Meng et al., 2008; Awomeso et al., 2010, Liu et al., 2011; Wang et al., 2013). Previous studies have interpreted both point source (Wang et al., 2008) and non-point source water pollutions (Zhang et al., 1996). GIS-based methods have been widely used to monitor water pollution levels (Coskun et al., 2006) and to analyze the spatial distribution of pollution patterns (Kwon et al., 2012).

2.2 Water pollution studies of China

Water pollution issues in China have attracted increasing attention since the 1990s (Zhang et al., 1996). Rising water pollution problems captured global attention, along with China's rapid economic growth and industrialization process (Ebenstein, 2012). Increasing public concerns about this issue have led to extensive studies regarding China's water pollution, especially concerning heavy metal contamination and its impact on public health (Lee et al., 2010; Schwarzenbach et al., 2010; Bao et al., 2012; Ebenstein, 2012; Jiang et al., 2012; Sheng et al., 2012; Wang et al., 2013). According to World Health Organization (WHO) records, over 26 million Chinese people suffer from effects of dental fluorosis due to elevated fluoride levels in their drinking water, and over one million suffer from skeletal fluorosis, another condition thought to be attributable to drinking water.

Zhong et al. (2012) investigated the relationship of heavy metal contamination among water, paddy soil and rice in Xiangyin County, in central south China. They provided scientific evidence informing residents and local governments to take action to control water pollution and preserve drinking water quality to increase human health. Meng et al. analyzed pollution levels of seven heavy metals (arsenic, cadmium, chromium, copper, mercury, lead, zinc) in Bohai Bay (Meng et al., 2008) and identified polluted areas. Yang and Liu (2012) divided heavy metal pollutants into carcinogenic and non-carcinogenic categories to investigate pollution levels from tributaries of the Haihe River, China. Many case studies followed this methodology to investigate heavy metal pollution and evaluate health risk through direct and indirect food chain intake from a local enclosed ecosystem (Liu et al., 2011; Feng and Zhao, 2012; Zhang and Wang, 2013). Statistical methods including linear regression (Zhang et al., 2010; Liu et al., 2013), spatial regression (Ebenstein, 2012) and statistical hypothesis testing (Lee et al., 2010; Liu et al., 2011) are also widely used for measuring heavy metal contamination levels and analyzing their relationship with relevant factors (Huang, et al., 2010; Wang et al., 2013).

Scholars have examined the relevant factors of China's water pollution from the socioeconomic perspective (Schwarzenbach et al., 2010). Industrial wastewater is one of the most serious and common sources of water pollution with its high toxicity and untraceable characteristics (Wang et al., 2008; Ebenstein, 2012). Municipal sewage contributes significantly to ecotoxicological effects on water bodies due to the process of urbanization (Schwarzenbach et al., 2010). Given that China is a large agricultural country, the extensive use of pesticides and fertilizers is of great concern for ground and surface water contamination with biological chemicals (Schwarzenbach et al., 2010). Human activities, such as transportation, is another influencing factor, which can cause accidental spills leading to air and soil pollution (DeCatanzaro and Cvetkovic, 2009; Feng and Zhao, 2012). Surface or ground water could also be indirectly contaminated by air pollution settlement (Zhang et al., 2010) or directly contaminated by soil (Liu et al., 2011; Wang et al., 2013). Similarly, nuclear energy has caused fatal radiation accidents and severe pollution problems (Sovacool, 2010; Hippel, 2011). Mining waste, hospital waste, hazardous waste, etc. are also water pollution sources (Schwarzenbach et al., 2010). Globalization, industrialization, economic growth, transportation, and urbanization are all inseparable anthropogenic activities, which can lead to emission of pollutants (Lee et al., 2010; Li and Wei, 2010a; Wen and Yang, 2010). In addition, the hydrologic characteristics of water bodies, land use types, and greenery coverage profoundly influence water quality (Alexander et al., 2007; Liu et al., 2009).

2.3 GIS-based methods applied in water pollution investigation

With the popularity and development of GIS technology, more scholars have used GIS techniques to study water quality and pollution from a spatial perspective (Yang and Liu, 2012; Wang et al., 2013). British scholars first used GIS to study water pollution problems in the late 1980s (Foster, 2000). They applied GIS methods in mapping water distribution, modeling sewer networks, detecting leakage, analyzing customer records and complaints, identifying and quantifying point and non-point water pollution sources, and managing drinking water quality (Foster, 2000). GIS-based methods have also been widely used to monitor water pollution levels (Coskun, et al, 2006) and analyze spatial distributions of pollution patterns (Kwon et al., 2012). A GIS-based screen model (Ag-PIE) was used to assess the agricultural land use pressure and the consequences on surface and groundwater. This model was more appropriate for a synoptic view at a continental scale rather than a local scale analysis (Giupponi and Vladimirova, 2006). Scholars also identified the source of diffused water pollution (perfluorinated compound, PFC) in Japan and collected its component data and then extracted those data from the GIS database. They used GIS to visualize the spatial distribution of the pollution elements and explored those factors through multiple linear regression. This study showed the effectiveness of using a GIS-based approach to identify and implement spatial distributions on nonpoint pollution sources (Zushi and Masunaga, 2011). A similar approach was taken using GIS as a decision support system to draw thematic maps and to classify the pollution level of agricultural drainage water, which can be potentially reused for irrigation purposes depending on water degradation (Shaban et al., 2010). This study could assist the government in controlling and managing water pollution, in addition to monitoring national water quality.

2.4 Statistical analysis and Kriging model

Statistical methods have been commonly used to examine the spatial pattern of water pollution (Yang and Liu, 2012; Wang et al., 2013). Statistical methods including linear regression (Zhang et al., 2010; Liu et al., 2013), spatial regression (Ebenstein, 2012) and statistical hypothesis testing (Lee et al., 2010; Liu et al. 2011) are widely used for measuring contamination levels and analyzing the relationship between heavy metal contamination levels and relevant factors (Wang et al., 2013). For instance, Lee et al. (2010) used the Environmental Kuznets Curve (EKC) hypothesis with a generalized method of moments (GMM) approach and dynamic panel regression to investigate the water pollution levels. The authors analyzed the underlying socioeconomic variables such as population, trade, GDP per capita etc. in four regional groups including, Africa, Asia and Oceania, America, and Europe. They confirmed that regional economic development through industrialization and urbanization can cause water pollution.

Another statistical method known as kriging has become an important geo-statistical tool in analyzing water pollution. Previous studies have proven that kriging has irreplaceable advantages because of its high accuracy and low bias (Yang et al., 2004; Li et al., 2005; Simasuwannarong et al., 2012; Wu and Li, 2013). There are four types of kriging models: simple kriging, ordinary kriging (Satapathy et al., 2009), universal kriging, and residual kriging (Sluiter, 2009; Wu and Li, 2013). Kriging operates on the assumption that the predicted value of a function at an unsampled point is calculated by using a weighted average of the known values of the function at sampled points in the neighborhood of the unsampled point. The method is mathematically closely related to linear regression analysis. Both theories derive a best linear unbiased estimator based on covariance assumptions. Kriging models have become commonly used in many fields, including Meteorology and Climatology (Wu and Li, 2013), Hydrogeology (Tonkin and Larson, 2002; Wycisk et al., 2013), Natural resources, etc. (Richmond, 2003; Emery, 2005). Wu and Li (2013) used residual kriging to estimate air temperature with variables of latitude, longitude and elevation in the U.S. Their research has demonstrated this methodological approach with the reliability of predicting temperature and the flexibility of adding suitable variables.

2.5 Health impacts of heavy metal water pollution

Heavy metal water pollution has caused global concern in almost every aspect of human lives, including agricultural irrigation, surface and ground water supplies, marine ecosystems, etc. Heavy metal water pollution has many consequences including economic loss, irreversible environmental damage and most importantly, human health problems (Schwarzenbach et al., 2010; Wang, et al., 2013). Generally, excessive lead and mercury can permanently damage the nervous system and brain. Cadmium and arsenic accumulation have toxic effects on important human organs, such as the liver, lung, kidney, skin, etc. (Duruibe et al., 2007; Morais et al., 2012; Liu et al, 2014). Drinking water contaminated with arsenic has been known to cause of skin, bladder, and lung cancers. Therefore, it is classified as a Group 1 carcinogen by the International Agency for Research on Cancer (IARC, 2002; Dauphiné et al. 2013; Oberoi et al. 2014). The well-known Itai-itai disease was triggered by intake of cadmium polluted rice in Japan. Another heavy metal water pollution disaster also occurred in Japan in the 1960s. Local residents ingested mercury poisoned seafood and suffered from Minamata disease, causing severe body deformation (Zhai, et al., 2008; Wang et al., 2013).

Severe pollution issues and increasing awareness human health have attracted more attentions from scholars to investigate the impacts of heavy metal pollution on human living conditions in China (Zhou, et al. 2008; Zhou, et al. 2008; Kerger et al., 2009; Liu, 2010; Bale, 2014). Based on Beaumont et al. (2008) study of Liaoning Province, the mortality rates of stomach and lung cancers during 1970-1978 were unusually high compared to cancers caused by polluted hexavalent chromium found in drinking water. Liu (2010) emphasized the phenomenon of cancer villages in China. Based on this research, Hebei, Henan, Guangdong, Jiangsu, and Hunan Provinces ranked as the top 5 in terms of the numbers of cancer villages. A crisis caused by Cadmium poisoned rice emerged in Hunan Province and Guangdong Province in 2013, causing tremendous political concerns and negative impacts on both public health and socioeconomic environment (Bale, 2014; Tan, 2014).

2.6 Limitations and research objectives

Based on the reviews of previous studies, three aspects concerning China's water pollution need to be further investigated. Firstly, due to the lack of available water pollution data, few spatial analyses have been done on heavy metal contamination in major Chinese cities at a national scale. Most of the previous research focused on one specific region such as a river or a province of China (Zhai, et al., 2008; Meng et al., 2008; Huang et al., 2010; Bao et al., 2012; Jiang et al., 2012; Sheng et al., 2012; Wang et al., 2013). Secondly, multiple mechanisms need to be taken into consideration to examine the spatial pattern of heavy metal water pollution. Previous studies mainly deliberated on a single factor in a local area (Yang and Liu, 2012). For example, one such study was located near an electroplating facility, and industrialization was considered to be the only factor that caused water pollution in that area (Liu et al., 2011). In fact, the cumulative risk factors have greater impacts than individual factors. Thirdly, the research on the consequences of heavy metal water pollution in China remains limited and its impacts on public health merit further investigation. Use of spatial and statistical tools to analyze China's cancer incidence patterns and correlations among heavy metal water pollution, socioeconomic impacts and human health, are rather rare (Zhai et al., 2008; Beaumount et al., 2008; Chen et al., 2013; Wang et al., 2013; Jiang et al., 2014; Li et al., 2014; Liu et al., 2014).

To compensate for deficiencies in previous research, this study addresses the following three research objectives. First, GIS and spatial statistical techniques are used to analyze the spatial patterns of heavy metal water pollution levels of China's major cities based on the water pollution data collected from 159 cities between 2004 and 2011. Second, after considering the interwoven factors behind water pollution, heavy metal water pollution levels are examined using socioeconomic attributes including transportation, urbanization, industrialization, globalization, economic development, and agricultural and mining activities. Physical factors including the green area percentage of each city, the distance between a city and the nearest major water body, and hydrologic characteristics are also examined. Third, spatial patterns of cancer patients' distributions are analyzed, in addition to interactions between China's socioeconomic transitions, heavy metal water contamination, and cancer incidences.

3. Methodology

3.1 Study area

Mainland China has 31 provincial-level units, including 4 municipalities (Beijing, Tianjin, Shanghai and Chongqing) and 27 provinces (Fig. 1). They are traditionally grouped into three regions: eastern, central and western (Li and Wei, 2010b). The eastern region plays a leading role in the economic development considering its strong advantages in attracting foreign trading partners and investment. This region has also benefited from the government's preferential policies as well as favorable natural environment and geographic location. The central region is highly populated and serves as the center of China's politics and agricultural economy. The western region has complex and vast terrain with sparse population. Although less developed, the large land area and rich mineral resources provide the western region with great opportunities for potential development. China's administrative system is conveyed through multiple levels of governments, consisting of provinces, prefecture-level cities, county, townships, and villages' or citizens' committees (Li and Wei, 2010a). Prefecture-level cities were chosen as analytical units since they are formal cities which have a well-defined metropolitan area (Dhakal Fotheringham 2009). Generally, prefecture-level cities are large administrative units containing one or more urban districts serving as the central urban area and is often surrounded by a number of rural counties. The main fresh water resources in China include rivers and lake systems (Fig. 1). China's major rivers include Yangtze River (Changjiang), Yellow River (Huang He), Pearl River (Zhujiang), Huaihe River, Liaohe River, and the Songhua River. The major lakes include Qinghai Lake (saltwater lake), Poyang Lake, Dongting Lake, Taihu Lake, and Hongze Lake (freshwater lakes) (China Statistical Yearbook, 2005).

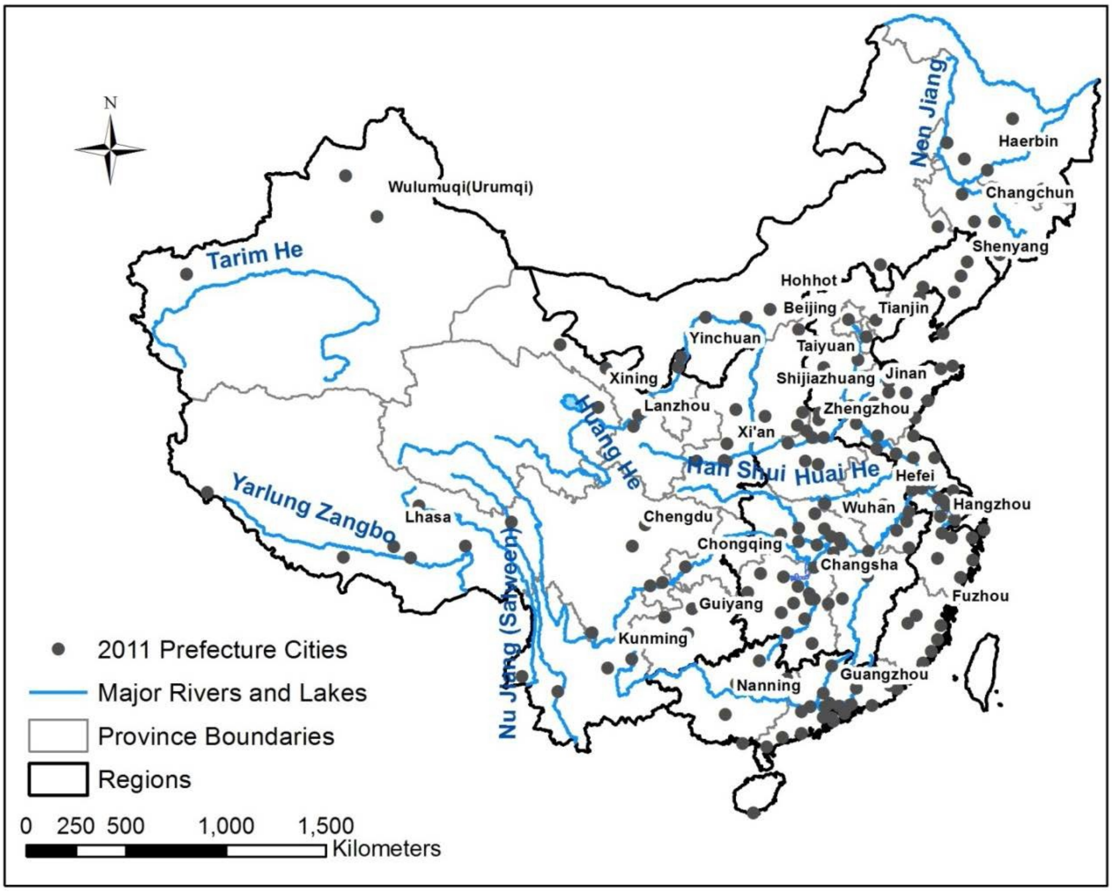

Figure 1: Major water bodies, and prefecture-level cities of China

3.2 Data and data sources

This study uses four types of data (Table 1). First, GIS shapefiles were downloaded from the China Data Center (http://chinadatacenter.org/). Second, water pollution data and parameters were obtained from the Institution of Public & Environmental Affairs (http://www.ipe.org.cn/pollution/status.aspx). Third, social-economic data used to explain the water pollution patterns was obtained from China City Statistical Yearbooks. Fourth, health data, including types of cancers and patient information were collected and summarized based on Chinese Cancer Registry Annual Reports. The time period of total analytical data covers from 2004 to 2011. For investigating heavy metal water pollution, five models were constructed with five different dependent variables (Y), namely the levels of heavy metal pollutants (unit as tons) such as Arsenic (As), Chromium (Cr), Mercury (Hg), Cadmium (Cd) and Lead (Pb). According to recent literature, the independent variables include transportation, urbanization, industrialization, globalization, agricultural development, constant GDP per capita (GDPPC), distances between cities and major water bodies (based on the official published China' map, 1cm/76 kilogram as radius scale), hydrological properties of water bodies, the percentage of green area per city, and the percentage of persons employed in mining and quarrying.

Table 1. Water pollution data: 2004 and 2011.

Classes	Variables	Indicators
Dependent Variable	Heavy metal pollutants	As, Cr, Hg, Cd and Pb
Independent Variables	Transportation (TRA)	Highway/Railway density
	Urbanization (URB)	Population Density
	Industrialization (IND)	Industrial output / total GDP
	Globalization (GLO)	Foreign direct investment per capita
	Agricultural (AGR)	Agricultural output / total GDP
	Economic Development (ECO)	GDP per capita
	Employment (EMP)	% of population employed in mining and quarrying
	Distance (DIS)	Distance to a major water body
	Green land (GRE)	Percentage of green area
	Hydrology (HYD)	Upper, middle and lower reaches

Note: Chemical element symbols: Arsenic - As, Chromium - Cr, Mercury - Hg, Cadmium - Cd, Lead – Pb

To analyze the impacts of heavy metal water pollution on human health, esophagus, stomach, colon, rectum, liver, trachea/bronchus/lung (TBL), and brain/nervous system (BN) all were chosen as dependent variables to build seven multiple linear regression models; one for each cancer type. Independent variables consistently included all five heavy metals and all independent variables from previous pollution models. Heavy metal pollution data in 2004 and sewage treatment rates were chosen as control variables for pollution investigation and health impact evaluation respectively.

Table 2. Human health data: 2004 and 2009

Classes	Variables	Indicators
Dependent Variable	Cancer diseases	Number of registered cancer patients
Independent Variables	Transportation (TRA)	Highway/Railway density
	Urbanization (URB)	Population Density
	Industrialization (IND)	Industrial output / total GDP
	Globalization (GLO)	Foreign direct investment per capita
	Agricultural (AGR)	Agricultural output / total GDP
	Economic Development (ECO)	GDP per capita
	Employment (EMP)	Persons employed in mining and quarrying
	Distance (DIS)	Distance to a major water body
	Green land (GRE)	Percentage of green area
	Hydrology (HYD)	Upper, middle and lower reaches
	Heavy metals (HM)	Untreated discharged As, Cr, Hg, Cd and Pb

Note: Chemical element symbols: Arsenic - As, Chromium - Cr, Mercury - Hg, Cadmium - Cd, Lead – Pb

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3.3 Methods and approach

GIS techniques and statistical methods were applied to analyze the spatial patterns of heavy metal water pollution and examine the underlying factors, such as GIS mapping, local indicator of spatial autocorrelation (LISA), kriging, regression, and geographically weighted regression (GWR). In addition, kernel function was used to analyze spatial trends of 2009 cancer patients' distributions.

3.3.1 Spatial-temporal analysis of heavy metal water pollution

GIS techniques and statistical methods were applied to analyze the spatial patterns of heavy metal water pollution and examine the underlying factors concerning China's socioeconomic transitions. Basic GIS and LISA maps were created for five heavy metal pollution levels (As, Cr, Hg, Cd and Pb) from 2004 and 2011 data respectively. Moran's I is a very classic measurement of spatial autocorrelation in areal data (Aneslin, 1995; Li and Wei, 2010a). Global Moran's I analyzes overall pattern and the spatial trend of the entire study area which is assumed to be homogeneous. However, different degrees of spatial autocorrelation (both positive and negative) could exist together in the same dataset. Therefore, Local Indicators of Spatial Association (LISA) was applied to diagnose the association for individual units and identify local clusters (Aneslin, 1995; Fotheringham et al., 2003; Li and Wei, 2010a). Heavy metal pollution (As, Cr, Hg, Cd and Pb) trends can be examined by calculating the indices with the following equation:

$$I = \frac{N}{\sum_i \sum_j w_{ij}} \frac{\sum_i \sum_j w_{ij}(X_i - \bar{X})(X_j - \bar{X})}{\sum_i (X_i - \bar{X})^2}$$

N is the total number of units over the entire observed regions. X_i and X_j are the observed values of the variable at area unit i and j. $\bar{X}$ is the mean value. w_{ij} is the weight function measuring the spatial reciprocal of distance between locations i and j. Ten basic GIS maps and ten LISA maps were created for five heavy metals (As, Cr, Hg, Cd and Pb) for 2004 and 2011 respectively.

3.3.2 Kriging model and pollution analysis

Since the heavy metal pollution data was only available for 159 out of over 284 prefecture-level cities, kriging was used to estimate the values for the 125 cities in ArcGIS. Using the residual kriging model, a spatially continuous surface was generated to estimate the water pollution level and further study the factors behind the patterns. Previous studies have proven that kriging has irreplaceable advantages for data estimation because of its high accuracy and low bias (Yang et al., 2004; Li et al., 2005; Simasuwannarong et al., 2012; Wu and Li, 2013). The residual kriging method has better performance when interpolating observed data with residual values within a large spatial area. For each heavy metal, at least 50 different scenarios and parameters were tested, and the best model was used to create a relevant kriging surface. The predicted heavy metal water pollution level was obtained by summing the regression results and the estimated residuals. The difference between observed and predicted data is residual, which can be estimated through the Semivariance equation seen below. (Liu et al., 2008; Wu and Li, 2013):

$$\hat{\gamma}(h) = \frac{1}{2} \cdot \frac{1}{n(h)} \sum_{i=1}^{n(h)} (z(x_i + h) - z(x_i))^2$$

$\hat{\gamma}(h)$ is the experimental semivariogram; h is the lag distance between two locations; N(h) is the number of data locations h distance apart. The value of $\hat{\gamma}(h)$ increases as distance (*h*) increases, and $\hat{\gamma}(h)$ infinitely approaches the sill, or the maximum variance. $z(x_i)$ is the measured pollution level at location *i*. $z(x_i + h)$ is the measured pollution level at a location *h-distance* away from location *i*. The predicted water pollution data is validated by comparing the observed values using a paired t test and root-mean-square-error (RMSE) (Wu and Li, 2013). The optimal estimation data set for each heavy metal is identified based on the results of the RMSE and paired t test. RMSE reflects the difference between predicted and observed values of sample data's standard deviation.

$$\text{RMSE} = \sqrt{\frac{1}{N} \sum_{i=1}^{N} \left(Y_i - \hat{Y}_i\right)^2}$$

These data sets were used to run regressions and geographically weighted regressions (GWR) for examining the underlying trend. GWR has the ability to analyze non-stationary data through changing parameters of regression models across the study area (Li and Wei, 2014). Two sets of results from GWR and ordinary least squares (OLS) for each heavy metal were compared.

Five multiple regression models are formulated as:

$$P = a + bP(2004) + cTRA + dURB + eIND + fGLO + gAGR + hECO + iDIS + jGRE + kHYD + lEMP + \varepsilon$$

The dependent variable *P* represents the pollution level of one of the five heavy metals (As, Cr, Hg, Cd and Pb) in 2011. There are two sets of independent variables. The first set of independent variables measures China's socioeconomic transitions and economic growth. Transportation (*TRA)*, indicated by road density, is the ratio of total length of highway and railway to land area. Human activity on transportation, such as pollution caused by accidental spills, is an important influencing factor on water pollution (DeCatanzaro and Cvetkovic, 2009; Feng and Zhao, 2012). Urbanization (*URB)* is indicated by population density. Municipal sewage has significant ecotoxicological effects on water bodies due to the rapidly increasing urbanization (Schwarzenbach et al., 2010). Industrialization (*IND)* is represented by the percentage of the annual industrial output out of the total GDP. Because of rapid industrialization processes in in China, industrial wastewater has become one of the most serious and common sources of water pollution (Wang et al., 2008; Ebenstein, 2012). Globalization (*GLO)* is indicated by foreign direct investment (FDI). Previous studies showed that globalization, along with industrialization, economic growth, agriculture, transportation, etc. all have incumbent responsibilities for water pollution issues in China (Liu and Diamond, 2005; Cao, 2008; O'Bannon et al., 2014). Economic development (*ECO)*, indicated by GDP per capita, has intensified environmental pollution including severe water contamination. Agriculture (*AGR)* is represented by the percentage of the annual agricultural output from the total GDP. Given that China is a large agricultural country, the extensive use of pesticide and fertilizers is of great concern for ground and surface water contamination with biological chemicals (Schwarzenbach et al., 2010; Liu et al., 2011). While the percentage of persons employed in mining and quarrying can indicate the magnitude of mining activities and mineral reserves for one city. It also serves as an index which can be used to reflect pollution levels (Zhai et al., 2008). Positive causal relationships were expected between the above seven variables and the pollution levels.

The second set of variables reflects the impact of physical environments. The distance between a city and closest major water body (DIS) is applied to represent the accessibility of one city to discharge pollutants (Zhai et al., 2008; Bao et al., 2012). Hydrologic properties (HYD) of water bodies, indicated by the location of a city within the range of a nearby major water body, are also influential to city water pollution levels (Zhai et al., 2008). The cities located at upper, middle, and lower reaches are indicated by one, two, and three, respectively. It was assumed that water is more polluted in the lower reaches than the uppers. The percentage of green area per city is also an important factor to improve water quality (Liu et al., 2009).

The heavy metal pollution level of 2004, *P(2004),* was chosen as the control variable. ε represents the error term.

3.3.3 Human health risk assessment

To analyze water pollution impacts on human health, seven cancers were chosen as analytical objects, including esophagus, stomach, colon, rectum, liver, trachea/bronchus/lung (TBL), and brain/nervous system (BN). The total numbers of registered patients with these seven cancers were mapped in ArcGIS to illustrate the spatial distribution patterns and temporal changes between 2004 and 2009. A point shapefile containing patient and cancer information was created for each type of cancer. The adaptive spatial kernel function was then applied to estimate and map patient incident clusters. It was selected because of its adaptable capabilities with dispersed data, and its ability to adjust bandwidths automatically (Li and Wei, 2014).

The interaction between human health and anthropogenic activities, such as socioeconomic development and heavy metal water pollution issues, were further investigated through statistical methods. Following the multiple regression models for heavy metal water pollution analysis, seven regression models for cancer diseases were built using the equation below:

$$C = a + bSTR + cTRA + dURB + eIND + fGLO + gAGR + hECO + iDIS + jGRE + kHYD + lEMP + mHM + \varepsilon$$

The dependent variable C represents the total number of registered cancer disease patients in 2009. Independent variables included all variables used in heavy metal water pollution regression analysis, plus five additional heavy metal (As, Cd, Cr, Hg, and Pb) pollution levels (*HM*). The sewage treatment rate (*STR*) was chosen as a control variable in human health assessment. The indicator of *STR* reflects the effectiveness of pollution water regulation in prefecture-level cities. ε represents the error term.

4. Results and Interpretation

4.1 Spatial and temporal variations of heavy metal water pollution

Spatial-temporal patterns of five heavy metal water pollution levels were mapped for both 2004 and 2011 (Figs. 2-6). Table 3 summarized the top 5 cities with the highest pollution levels of Arsenic, Cadmium, Chromium, Mercury, and Lead in 2011. High Arsenic levels were concentrated in upstream areas of Yellow River, Yangtze River, Liaohe River, and Zhujiang River watersheds. Arsenic is often extracted from by-products of purified copper and commonly used in alloys and pesticides (USGS, 2008). Hydrogeological conditions and anthropogenic factors in China are linked to elevated Arsenic levels in water pollution (Sun, 2004; Xiao et al., 2008; Rodríguez-Lado et al., 2013; Wen et al., 2013). Example sources are deep groundwater wells (e.g., Chifeng on Fig. 2), mining activities (e.g., Baiyin, Chenzhou, Chongqing, Kunming, Yichang), burning of arsenic-rich coal, and pesticides. Based on the 2004 LISA maps, a significant high-low cluster of arsenic pollution was detected in Baiyin City (Gansu Province), upstream of the Yellow River, while, high-high Arsenic clusters appeared in both the Yangtze River and Pearl River basins in 2011 (e.g., Kunming and Chenzhou).

High Cadmium concentrations were detected in the Yangtze River and Yellow River Basins (Fig. 3). Compared to the 2004 map, high clusters of Cadmium in prefecture-level cities are mainly concentrated in the central and western regions in 2011. LISA maps revealed that higher clusters were found in northern China in 2004 than in southern China in 2011. Cadmium is one of the byproducts of zinc production and is typically obtained from cadmium sulfide ore. High concentrations of cadmium are commonly found in zinc and lead ores, phosphate fertilizers (Singh and McLaughlin, 1999), and sewage sludge (Resource Sciences, 1997). Higher Cadmium concentrations in both 2004 and 2011 were mainly associated with lead and zinc smelting industries, located in such cities as Baiyin, Zhuzhou (Hunan Province, South China), Kunming (Yunan Province, Southwest China), Shaoguan (Guangdong Province, South China), and Chenzhou (Hunan Province, South China)(Liu and Ou, 2003; Lei et al., 2008).

The Chromium pollution clusters were widely distributed from coastal areas to the central region, through the Yellow River, Huaihe River, Yangtze River, and Zhujiang Watersheds (Fig. 4). LISA maps showed only high clusters of Chromium pollution in Chongqing and Xiangtan (Hunan Province), South China. Sources of Chromium pollution include electroplating, medicine production, printing, painting, leather tanning, and dyeing (Cheng, 2003; Rong and Zhou, 2010; Jia et al., 2014). Chromium is widely used to strengthen steel. For example, Jia et al (2014) assessed the Chromium pollution at an industrial area of electroplating factories in Chongqing.

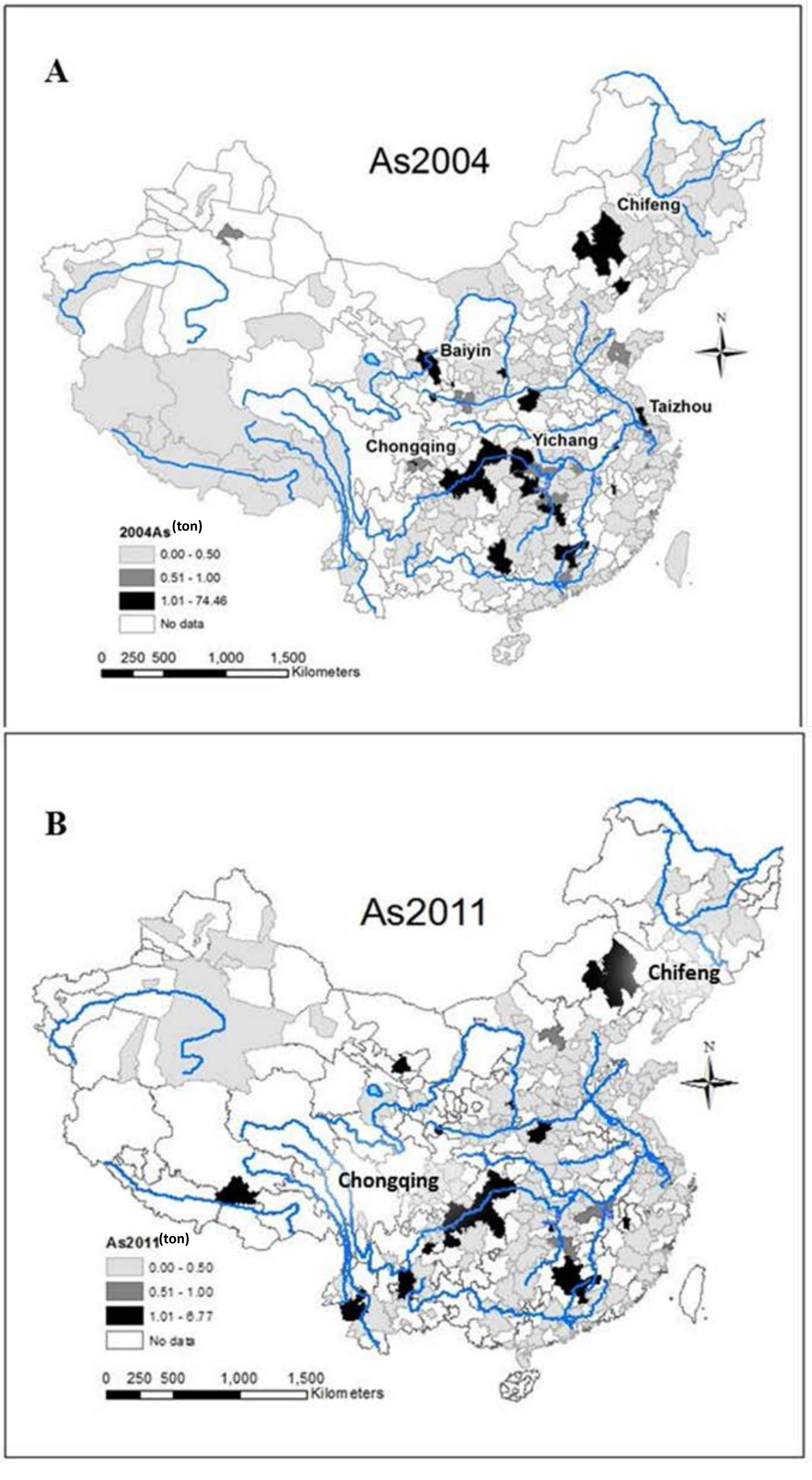
Concentrations of hexavalent Chromium and total Chromium in the wastewater were found to be over 26 and 12 times higher, respectively, at the drain outlets than the national standards. Hu et al. (2014) indicated that the Chromium discharges from the leather industry in Shandong Province accounted for 41.70 percent of the province's total Chromium discharges. Chromium salts have been manufactured since 1958 in over 70 factories in China, including Shanghai, Guangzhou, Chongqing, Xining, and Qingdao. These cities all have high population densities, and produced over 600 million tons of chromium slag (Rong and Zhou, 2010). Chromium slag contamination from the slag stacks causes widespread Chromium pollution, consistent with the results revealed in the maps from this research.

Cities with high Mercury pollution levels are distributed throughout northeast and southeast parts of China, mainly in the Liaohe River and Yangtze River watersheds (Fig. 5). Mining (gold and mercury), chemical industry, and coal combustion are important anthropogenic sources of mercury pollution in China (Jiang et al., 2006). Mercury may contaminate the environment through burning fuel. For example, coal combustion was responsible for the high Mercury levels in Chifeng (Inner Mongolia, North China) (Hu et al., 2011) and in Chongqing (Wu et al., 2006). Zhuzhou Smelting Plant and Zhuzhou Chemical Plant lead to high mercury pollution levels in Zhuzhou, Hunan Province (Chu and Tong, 2004). Shaoguan's elevated mercury levels (particularly highlighted from the LISA maps) were related to both Shaoguan Smelting Plant and Dabaoshan Mining Co., Ltd (Ding et al., 2010).

Central and southern China also suffered from severe lead pollution. Cities with high lead concentration were found upstream of the Yellow River and Yangtze River drainage areas (Fig. 6). Major sources of lead pollution in China are lead/zinc mining, smelting activities, and lead-acid battery industries (Zhang et al., 2005; Zhang et al., 2012). For example, Yunnan Province in Southwestern China is rich in mineral resources, especially lead-zinc deposits. Kunming, the capital city of Yunnan, is one of the most important lead production industrial bases in China (http://www.china.org.cn/e-xibu/2JI/3JI/yunnan/yunnan-ban.htm). Other lead and zinc smelting industries responsible for high Lead pollution are mainly located in Baiyin, Zhuzhou, Shaoguan, and Chenzhou (Liu and Ou, 2003; Lei et al., 2008; Ding et al., 2010; Zhang et al., 2012). These cities were specifically identified on the LISA maps. One report indicated that blood lead levels of some children in Chifeng were as high as 162 ug/L, much higher than the upper threshold level of 99 ug/L. This phenomenon was linked to the emissions from the Jinjian Copper Smelting Plant, established in 1990 (http://hj.ce.cn/gdxw/201309/12/t20130912_1091113.shtml). Lead is commonly used in battery production. Highest producing lead-acid battery industries, mainly located in then Jiangsu, Zhejiang and Guangdong provinces, contributed to higher lead pollution levels in those areas (http://hjj.mep.gov.cn/zdhy/xqdc/201111/t20111130_220782.htm).

Table 3. Pollution levels of top 5 cities

	As	Cd	Cr	Hg	Pb
1	Chenzhou	Kunming	Jiaozuo	Zhuzhou	Kunming
2	Kunming	Luoyang	Xiangtan	Nantong	Shaoguan
3	Lhasa	Chenzhou	Nanchang	Chifeng	Chenzhou
4	Jinchang	Shaoguan	Quanzhou	Chenzhou	Luoyang
5	Chifeng	Jinchang	Guangzhou	Shenzhen	Zhuzhou


A
As2004
Chifeng
Baiyin
Taizhou
Chongqing
Yichang
2004As(ton)
0.00 - 0.50
0.51 - 1.00
1.01 - 74.46
No data
0 250 500 1,000 1,500
Kilometers
N
B
As2011
Chifeng
Chongqing
As2011(ton)
0.00 - 0.50
0.51 - 1.00
1.01 - 6.77
No data
0 250 500 1,000 1,500
Kilometers

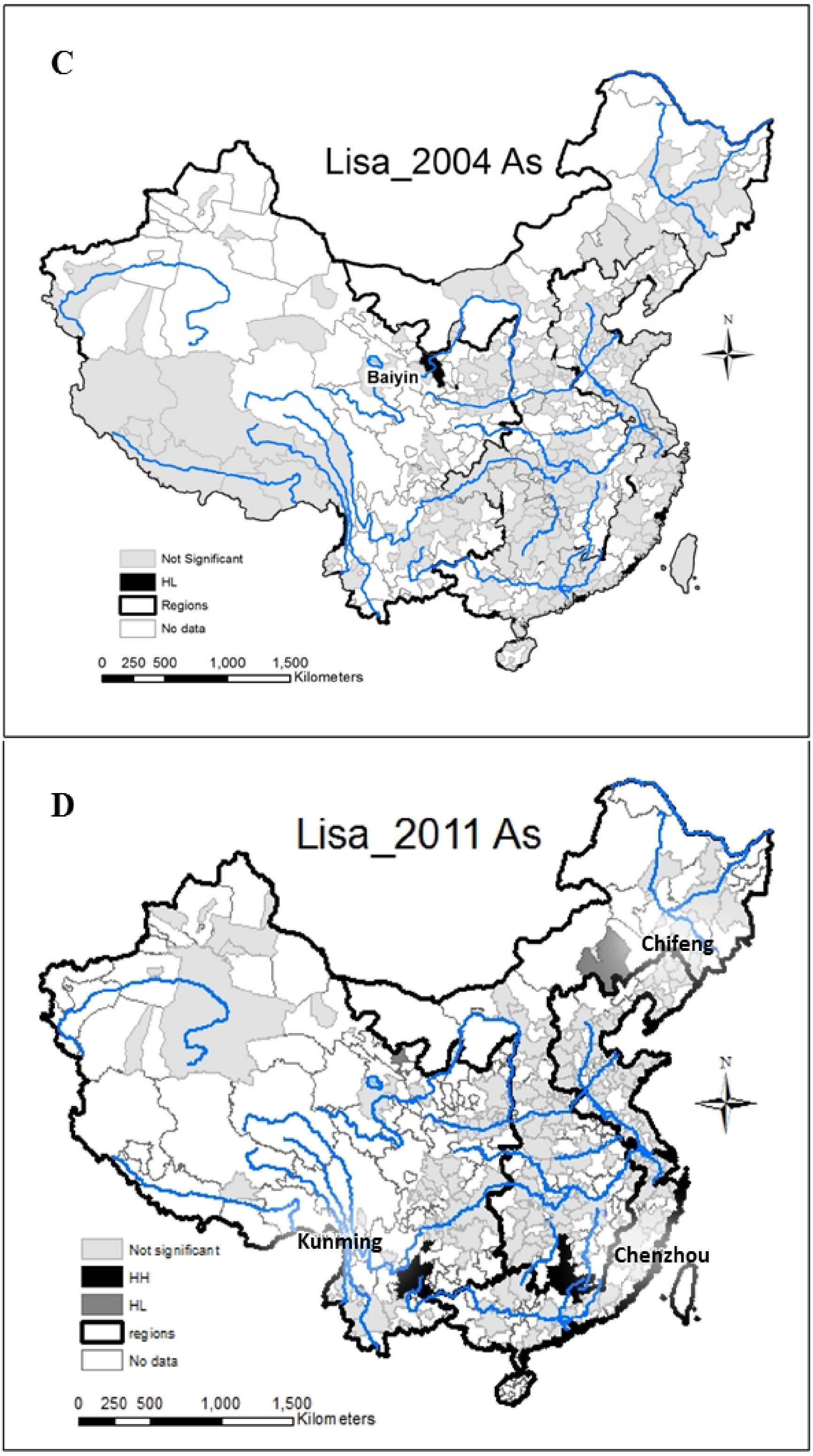
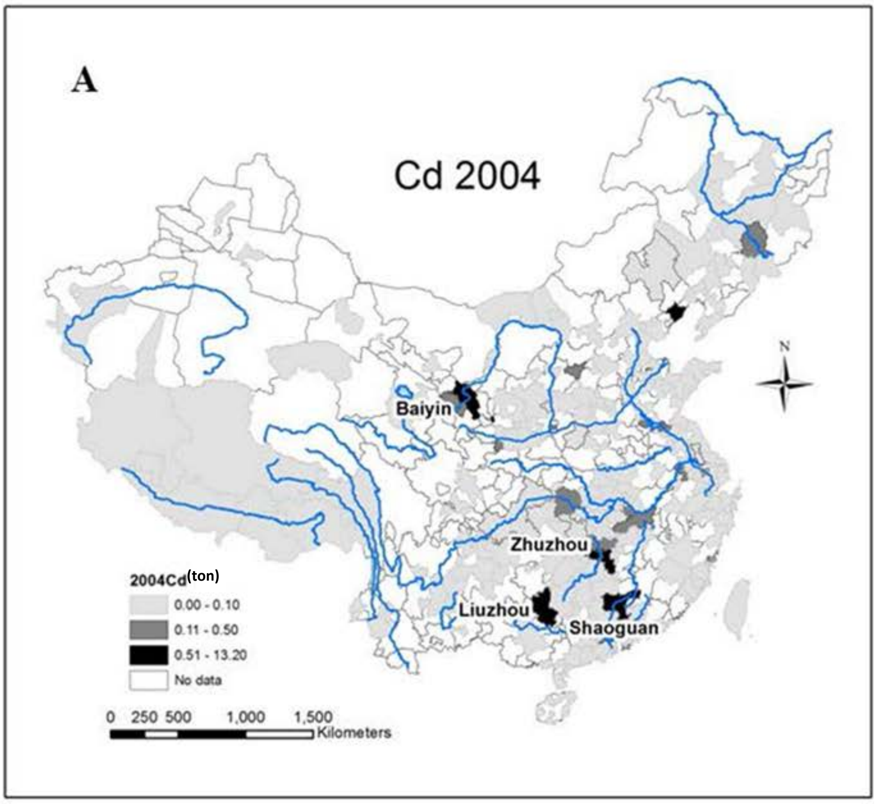
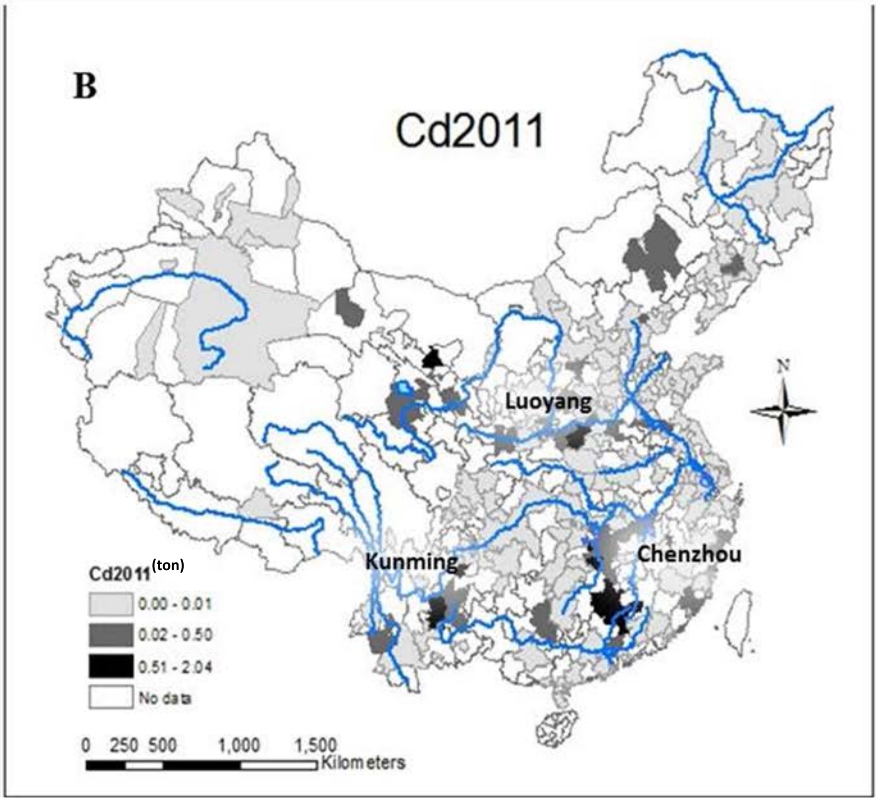


Fig 2. Spatial distribution and LISA maps of Arsenic water pollution (A) 2004 Arsenic water pollution level (B) 2011 Arsenic water pollution level (C) 2004 Arsenic LISA clusters (D) 2011 Arsenic LISA clusters


A
Cd 2004
Baiyin
Zhuzhou
Liuzhou
Shaoguan
2004Cd(ton)
0.00 - 0.10
0.11 - 0.50
0.51 - 13.20
No data
0 250 500 1,000 1,500
Kilometers
N


B
Cd2011
Luoyang
Kunming
Chenzhou
Cd2011(ton)
0.00 - 0.01
0.02 - 0.50
0.51 - 2.04
No data
0 250 500 1,000 1,500
Kilometers
N

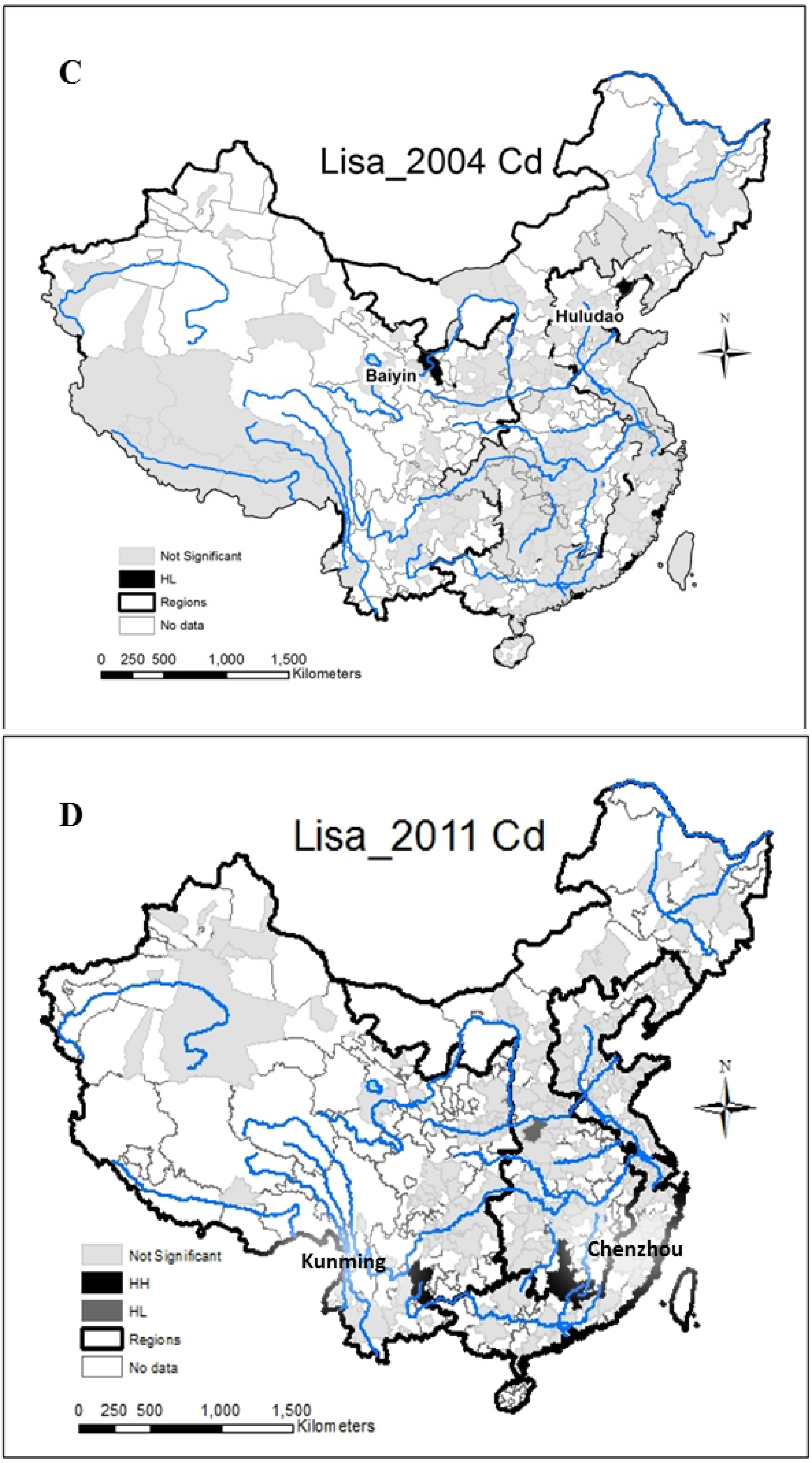
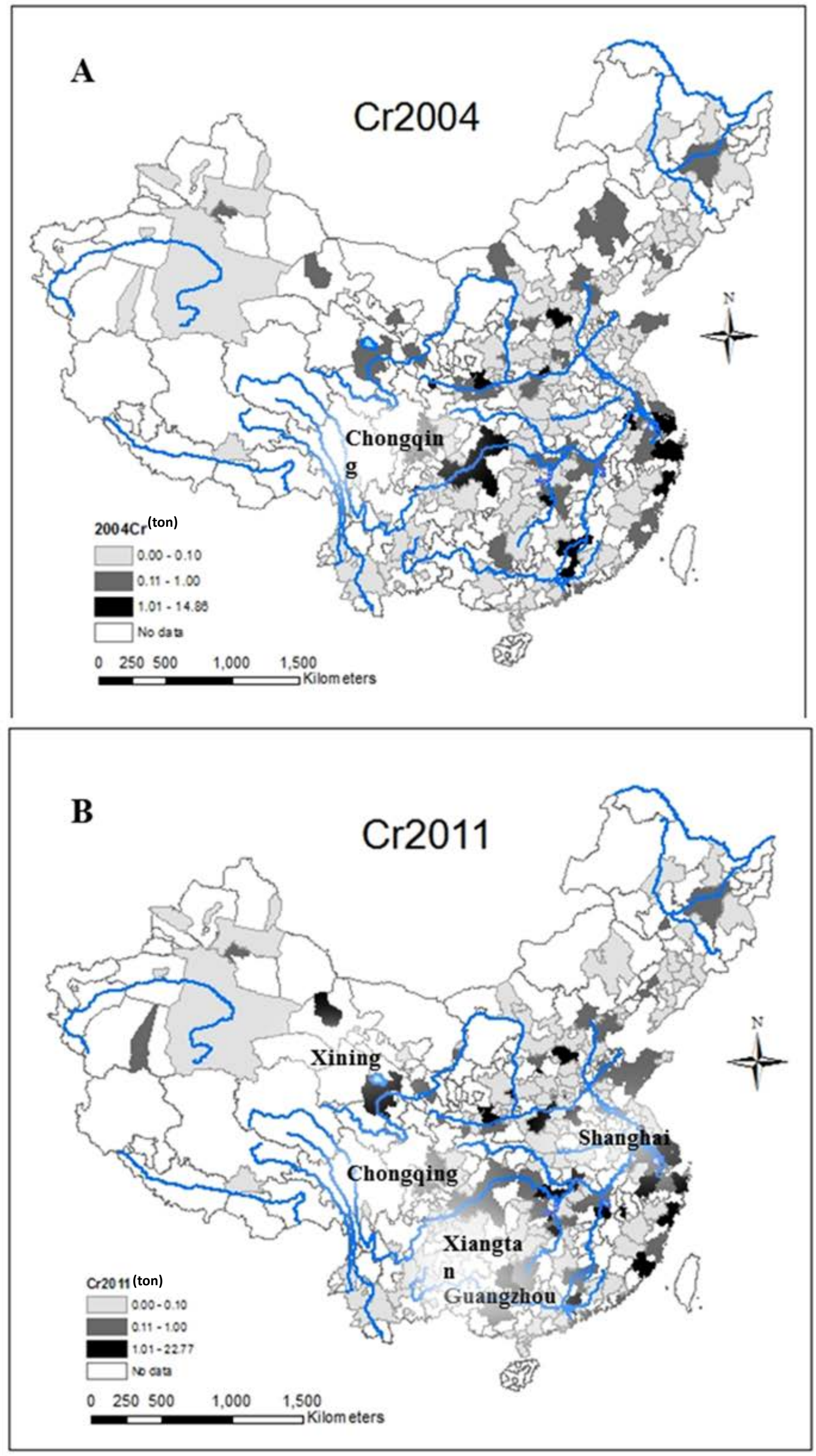


Fig 3. Spatial distribution and LISA maps of Cadmium water pollution
(A) 2004 Cadmium water pollution level (B) 2011 Cadmium water pollution level
(C) 2004 Cadmium LISA clusters (D) 2011 Cadmium LISA clusters


A
Cr2004
Chongqin
g
2004Cr(ton)
0.00 - 0.10
0.11 - 1.00
1.01 - 14.86
No data
0 250 500 1,000 1,500
Kilometers
N
B
Cr2011
Xining
Shanghai
Chongqing
Xiangta
n
Guangzhou
Cr2011 (ton)
0.00 - 0.10
0.11 - 1.00
1.01 - 22.77
No data
0 250 500 1,000 1,500
Kilometers
N

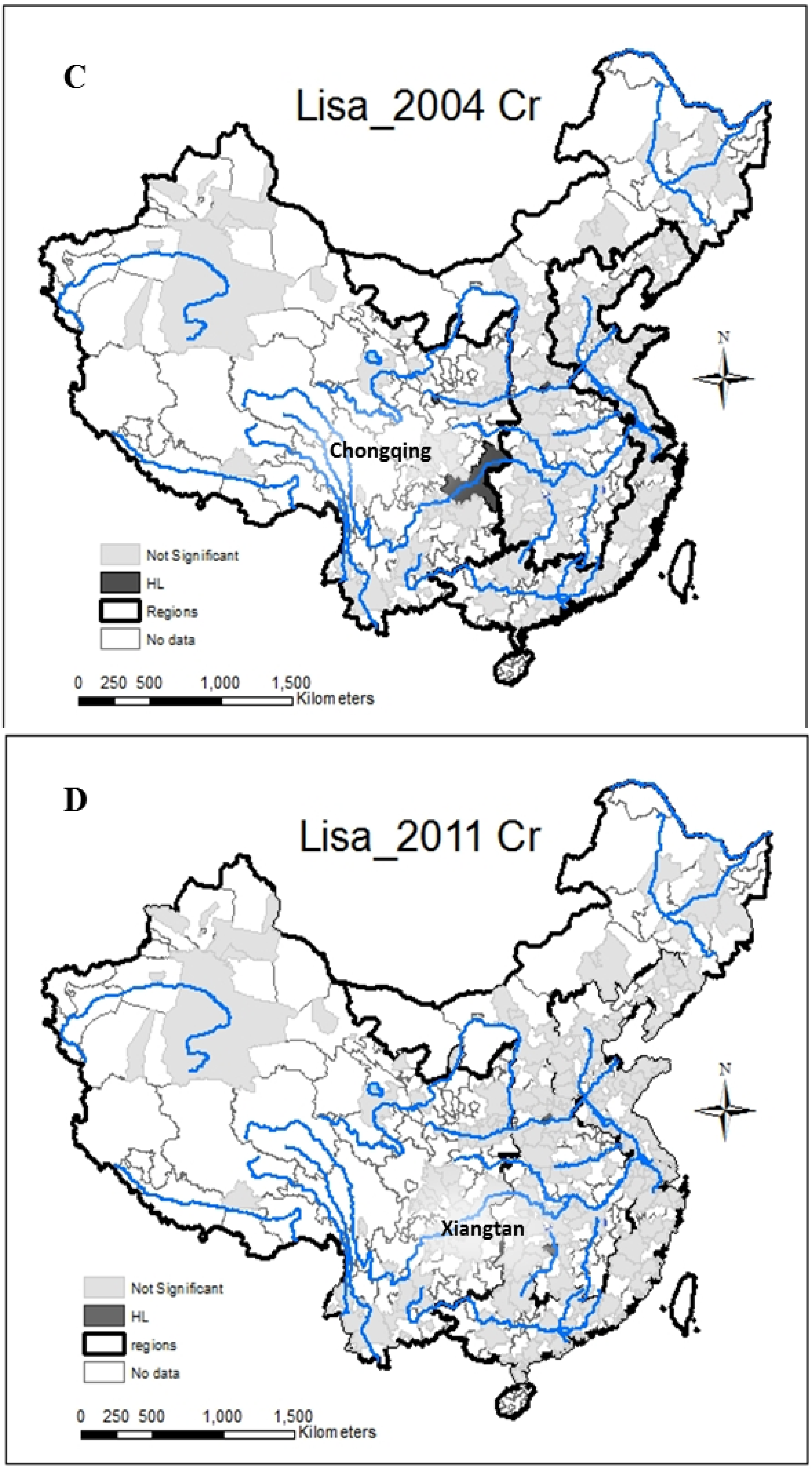
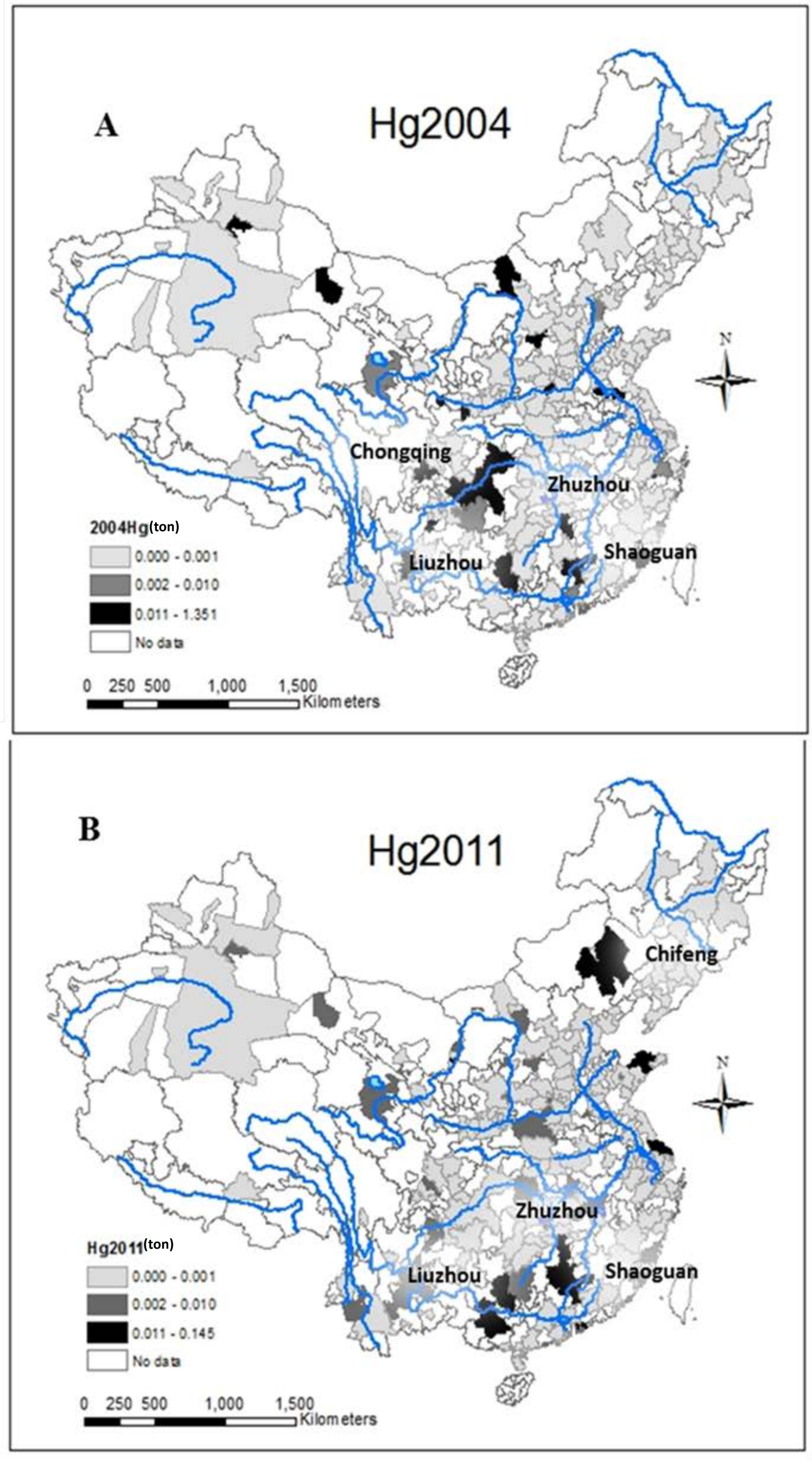
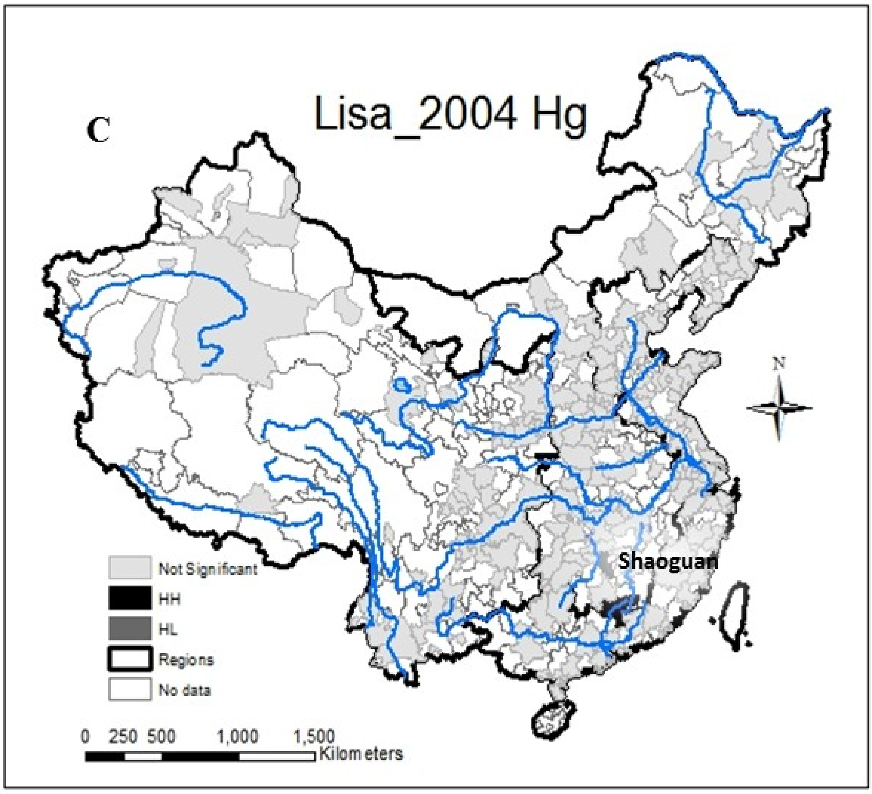


Fig 4. Spatial distribution and LISA maps of Chromium water pollution
(A) 2004 Chromium water pollution level (B) 2011 Chromium water pollution level
(C) 2004 Chromium LISA clusters (D) 2011 Chromium LISA clusters


A
Hg2004
Chongqing
Zhuzhou
Liuzhou
Shaoguan
2004Hg(ton)
0.000 - 0.001
0.002 - 0.010
0.011 - 1.351
No data
0 250 500 1,000 1,500
Kilometers
N
B
Hg2011
Chifeng
Zhuzhou
Liuzhou
Shaoguan
Hg2011(ton)
0.000 - 0.001
0.002 - 0.010
0.011 - 0.145
No data
0 250 500 1,000 1,500
Kilometers
N



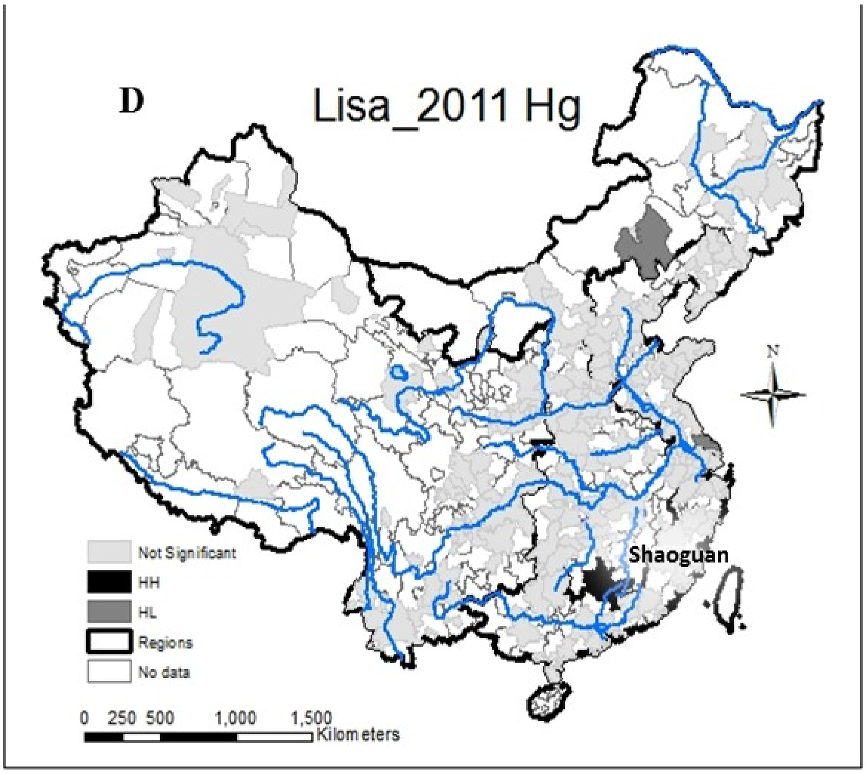
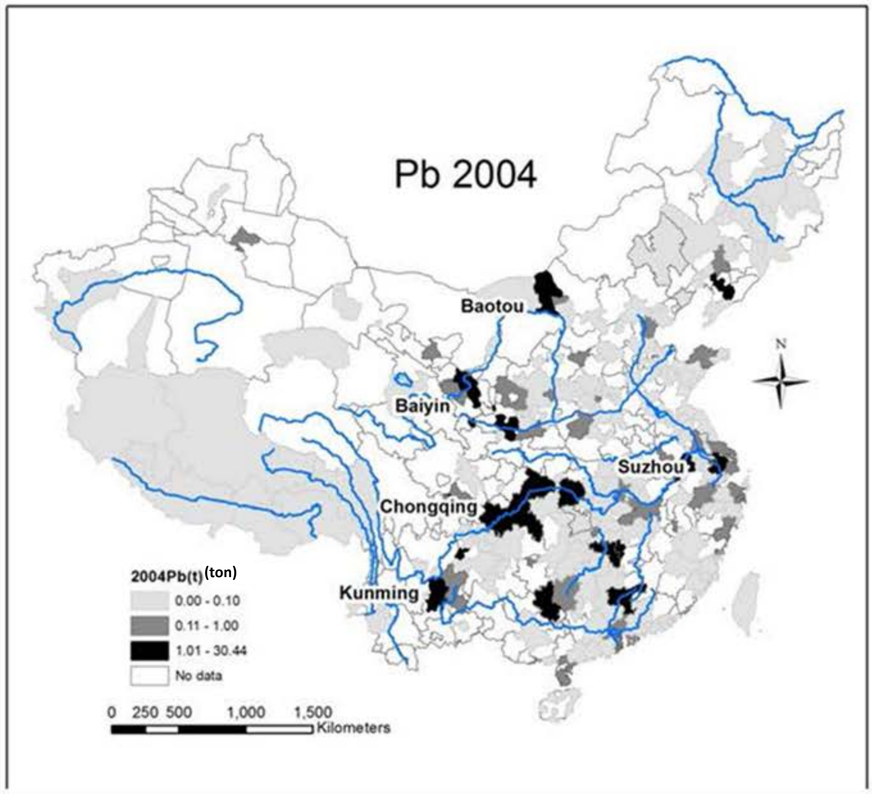
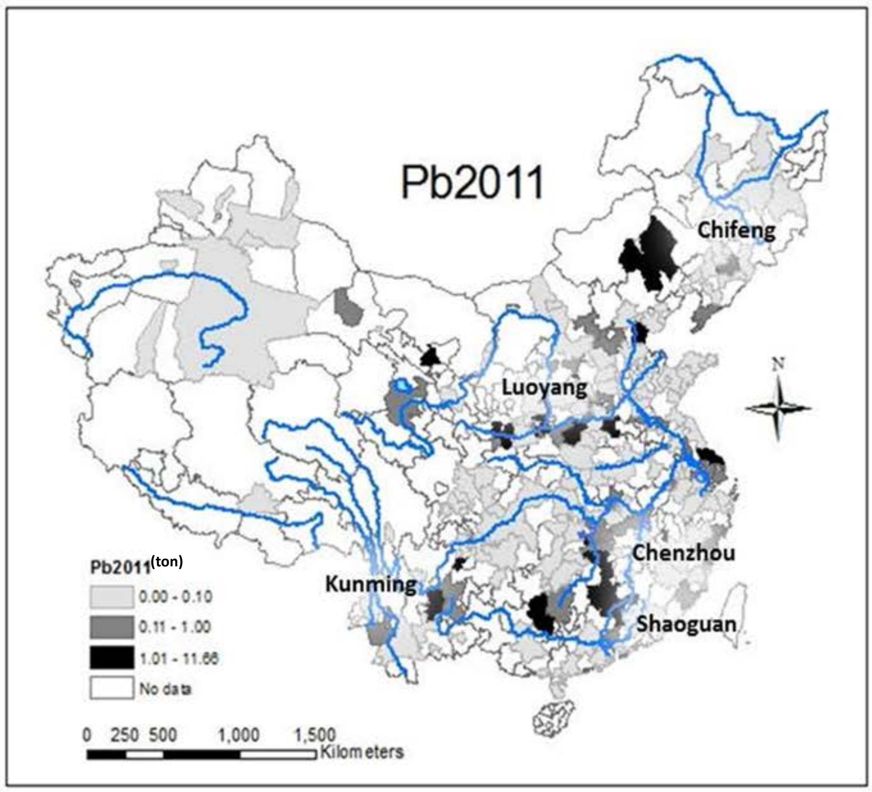


Fig 5. Spatial distribution and LISA maps of Mercury water pollution
(A) 2004 Mercury water pollution level (B) 2011 Mercury water pollution level
(C) 2004 Mercury LISA clusters (D) 2011 Mercury LISA clusters



Pb 2004
Baotou
Baiyin
Suzhou
Chongqing
Kunming
2004Pb(t)(ton)
0.00 - 0.10
0.11 - 1.00
1.01 - 30.44
No data
0 250 500 1,000 1,500
Kilometers
N



Pb2011
Chifeng
Luoyang
Chenzhou
Kunming
Shaoguan
Pb2011(ton)
0.00 - 0.10
0.11 - 1.00
1.01 - 11.66
No data
0 250 500 1,000 1,500
Kilometers
N

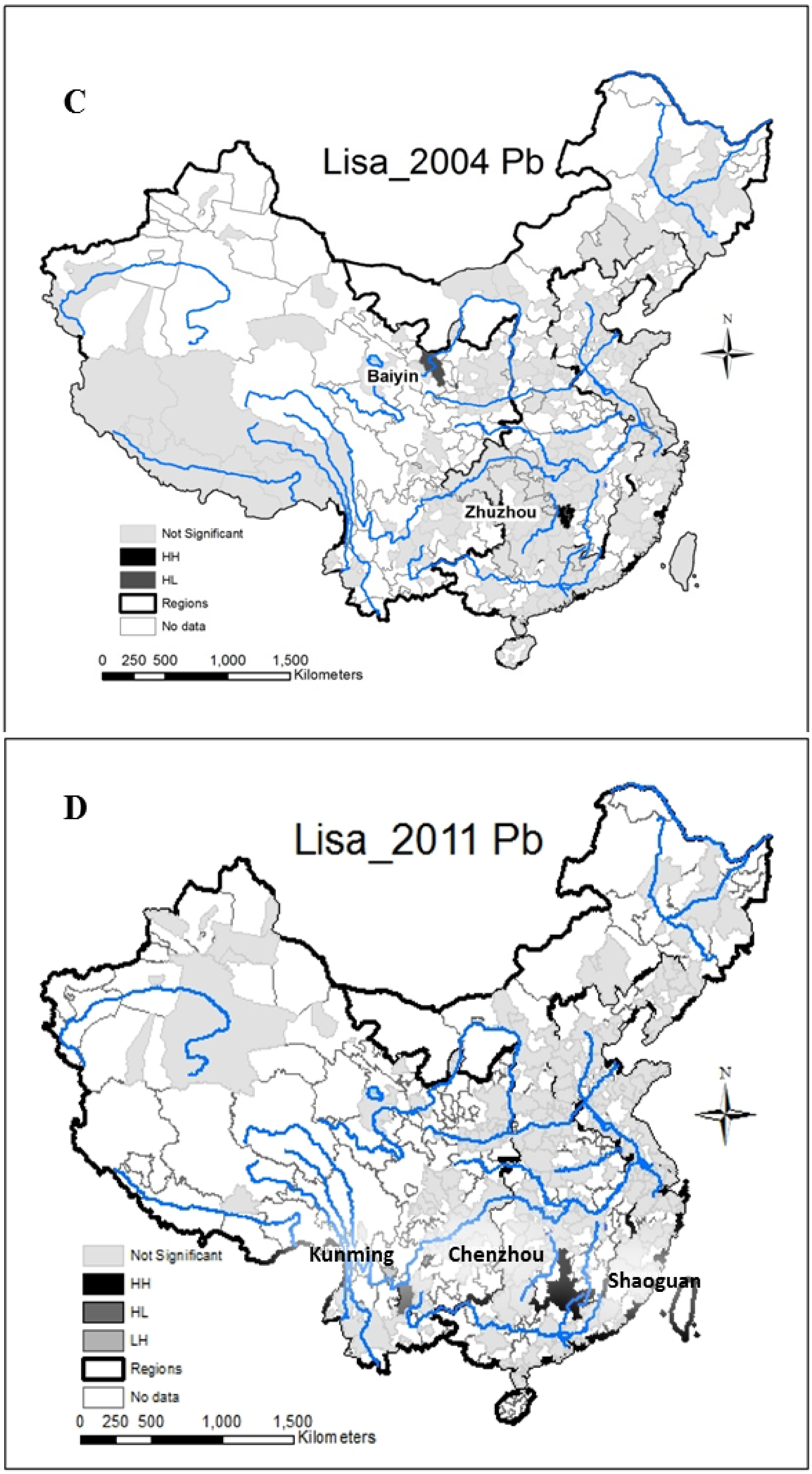


Fig 6. Spatial distribution and LISA maps of Lead water pollution (A) 2004 Lead water pollution level (B) 2011 Lead water pollution level (C) 2004 Lead LISA clusters (D) 2011 Lead LISA clusters

4.2 Socioeconomic transitions, physical conditions, and heavy metal water pollution

Heavy metal water pollution estimation using Kriging and model validation

Because heavy metal water pollution data is not available for all cities for this research, kriging models were used to predict water pollution levels for 159 cities in 2011. Different scenarios and parameters were modeled for kriging and semivariogram methods, including the search radius, number of points, and maximum distance for each heavy metal. The kriging surfaces were created and predicted, values were extracted via ArcGIS. Results showed optimal cell size and search radius, using ordinary kriging methods with a spherical semivariogram to have the best performance. RMSE values were calculated in excel. Values closest to 0 indicate the best prediction. After extensive calibration, the optimal scenario was found for each heavy metal model based on the smallest RMSE values. A paired t-test was applied to further assess kriging model validation. All p-values (two tailed) were larger than 0.05 (p values were between 0.89-0.98), indicating that model calibrations were reliable and that there were no statistical differences between observed and predicted values at the 5 percent significance level. The RMSE and t-test results of the optimal scenario for all five heavy metals are summarized below in Table 4. Based on the kriging estimation and model validation, the fittest and most reliable predicted pollution data was then used to construct GWR and OLS models. The kriging raster surfaces of these optimal models were presented in Figures 7-11.

Table 4. RMSE and paired t-test

Heavy metals	RMSE	p-Value
As	0.78	0.94
Cd	0.32	0.97
Cr	3.36	0.92
Hg	0.03	0.89
Pb	1.10	0.98

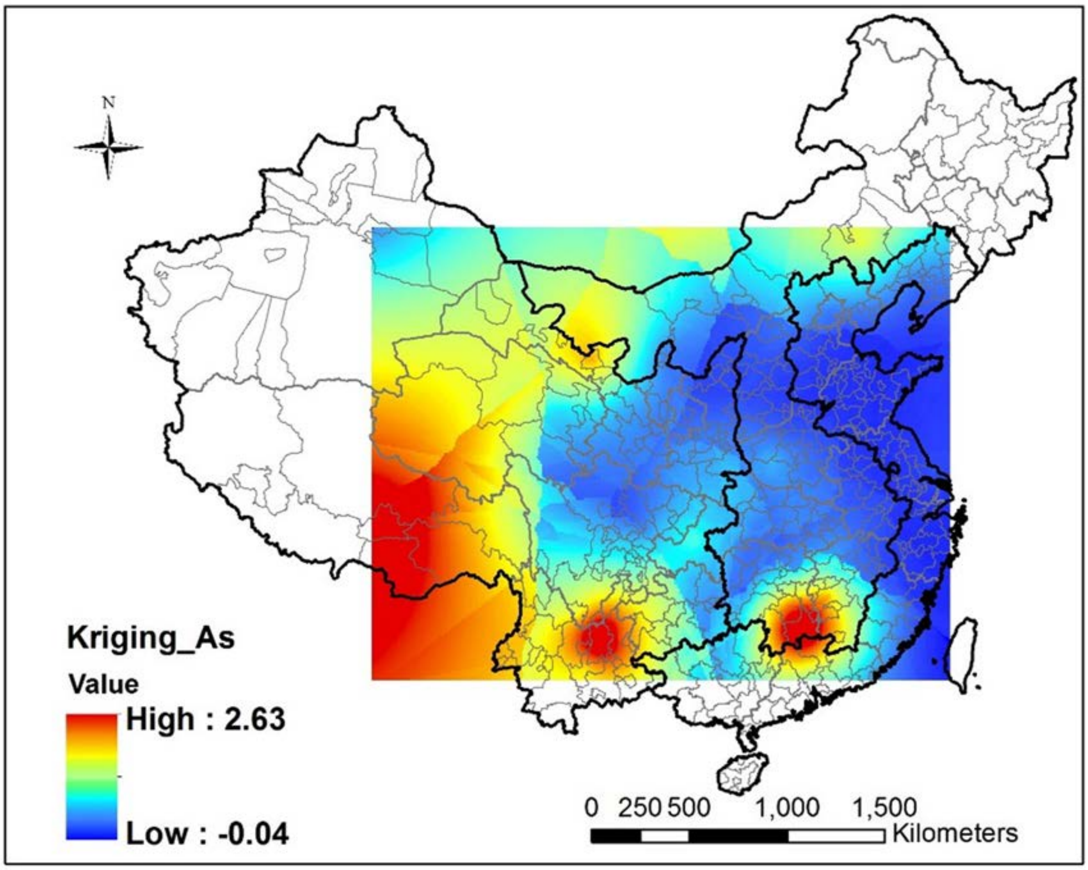


Fig. 7. Arsenic kriging surface

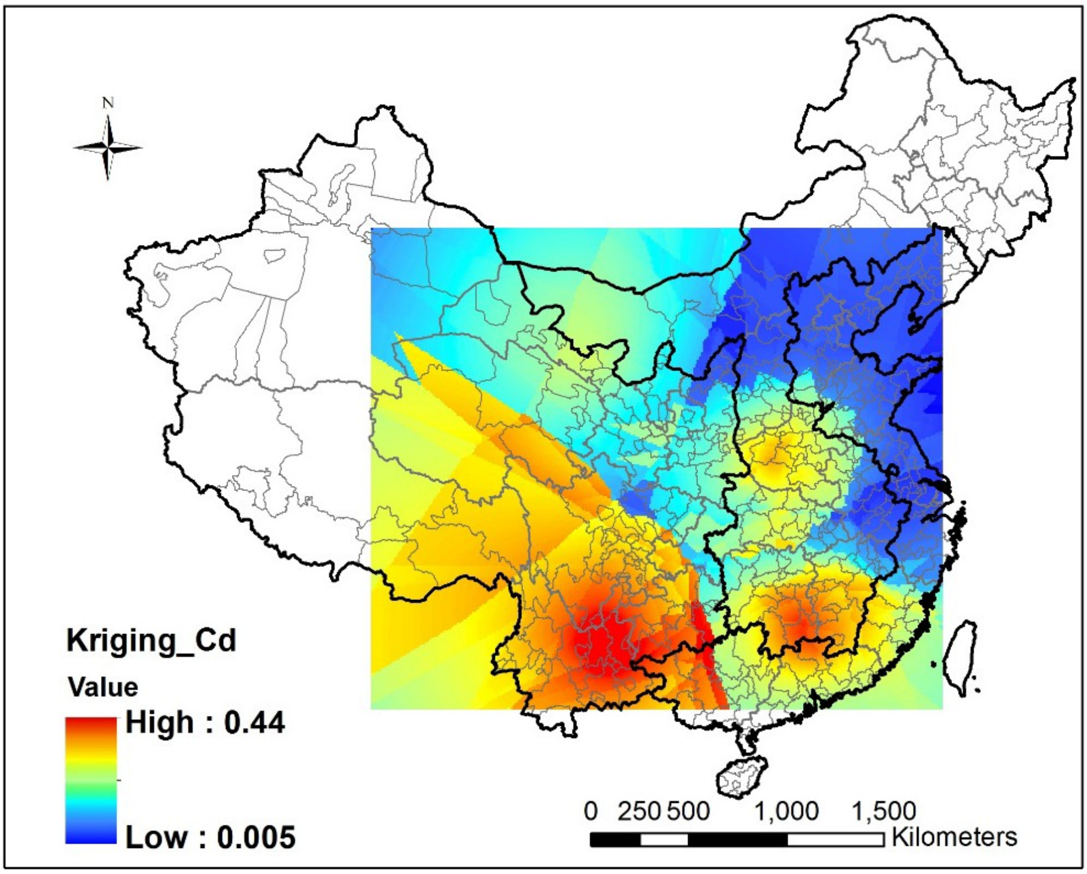


Fig. 8. Cadmium kriging surface

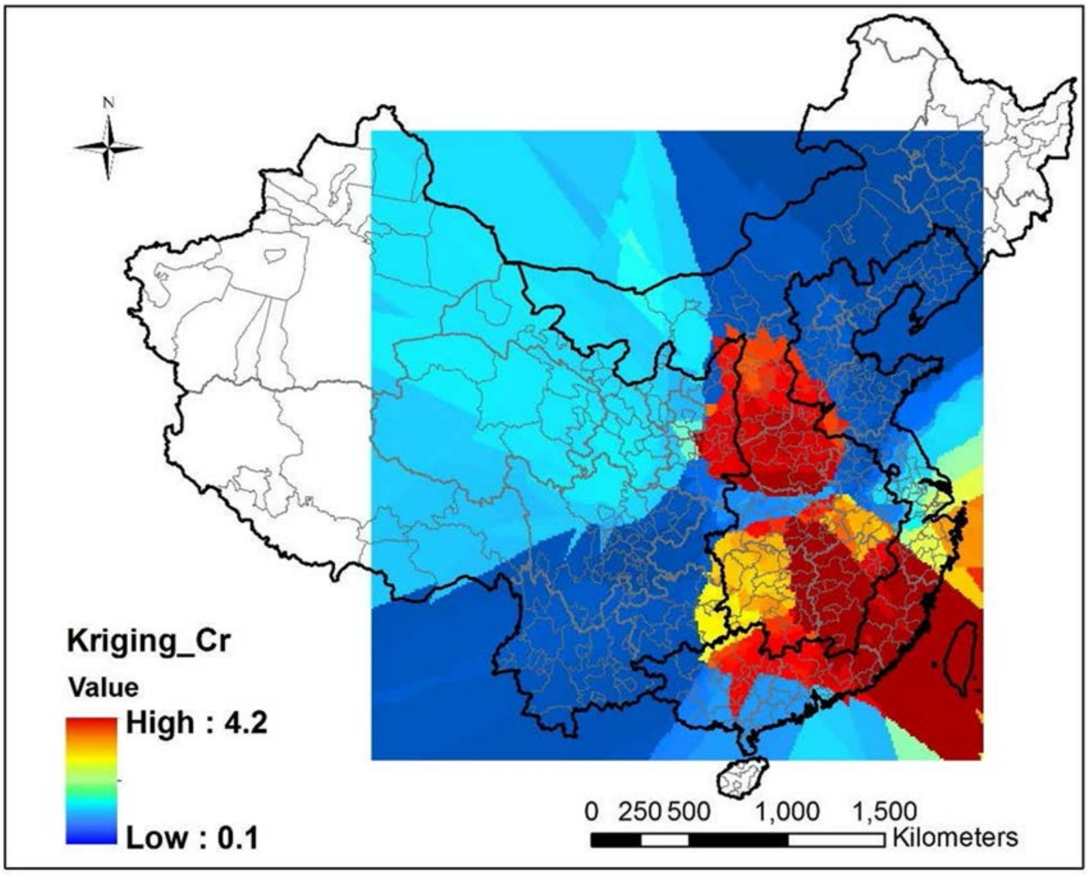


Fig. 9. Chromium kriging surface

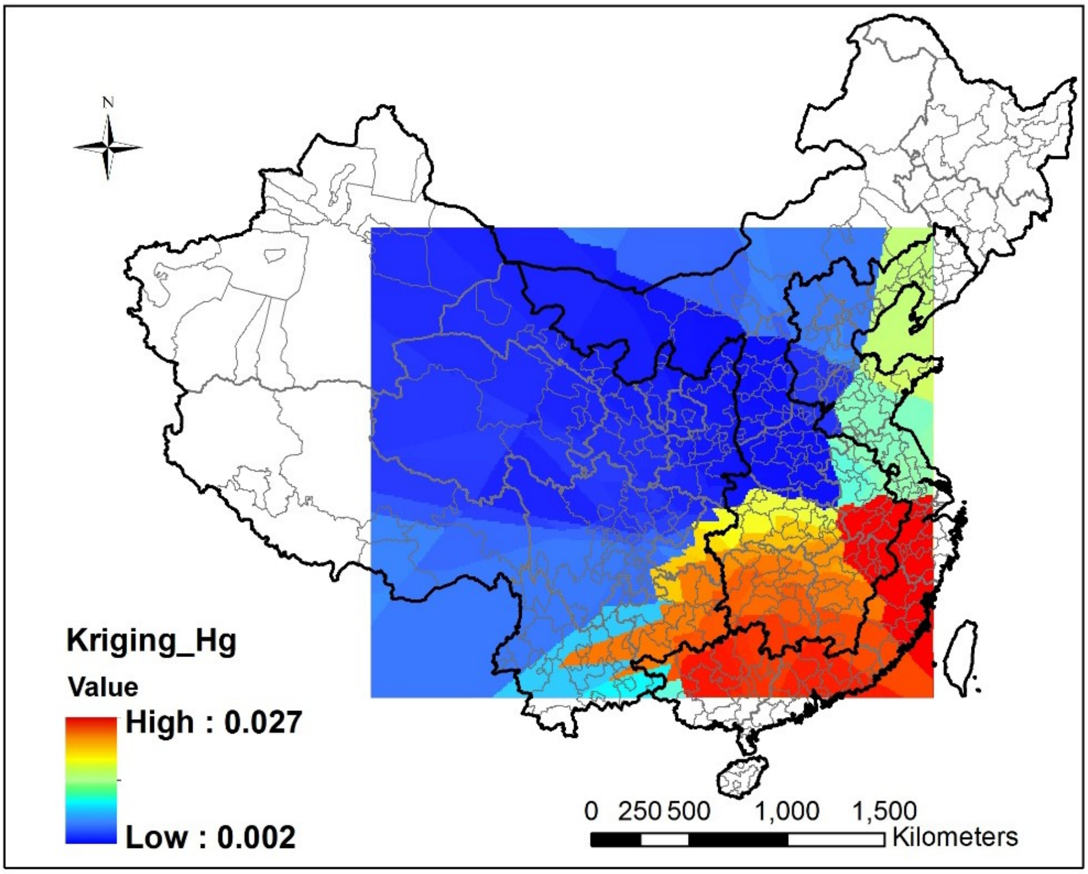


Fig. 10 Mercury kriging surface

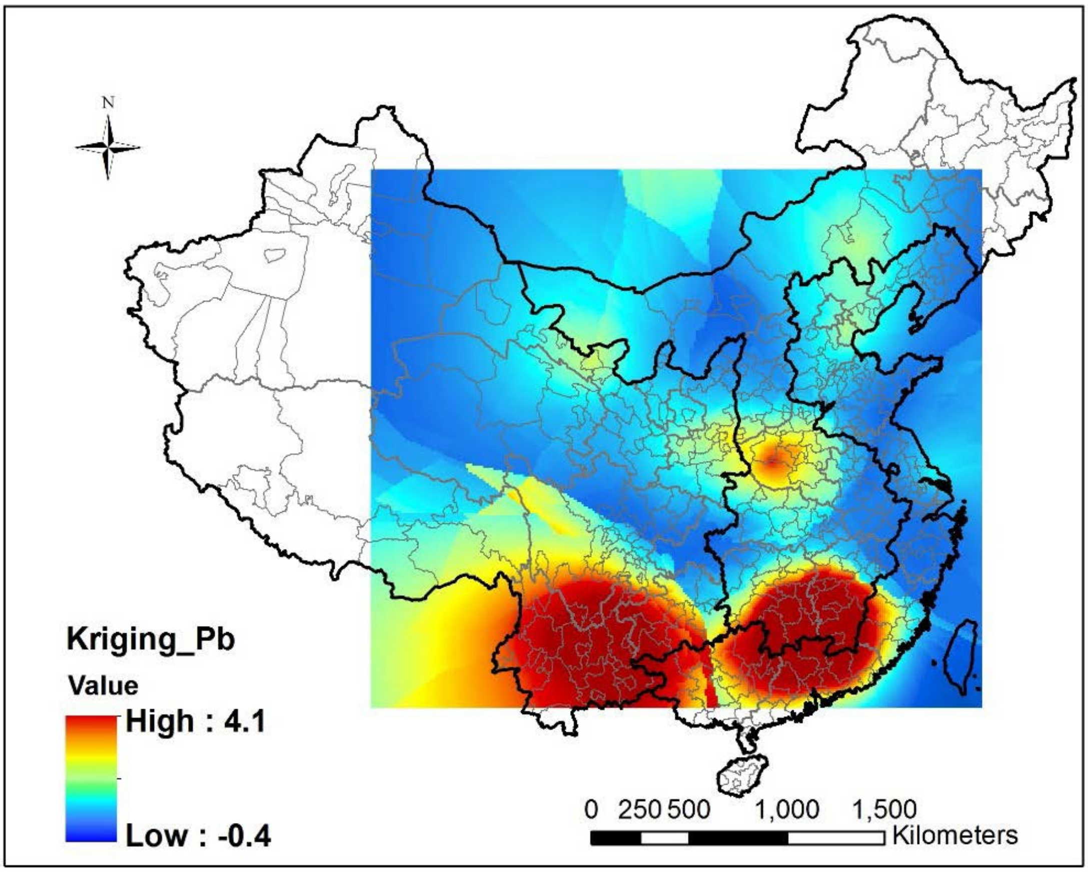


Fig. 11 Lead kriging surface

OLS Regression and GWR results

Results of the multiple regression models are presented in table 5. Results of GWR and OLS regression models are reported and compared in Table 6 Multiple regression models of arsenic, cadmium, and lead are significant at the 1 percent significance level; while chromium and mercury are significant at 5 percent level. GWR models had better performance than OLS models in investigating all five heavy metal pollution problems. All GWR R-square values were significantly higher than OLS values, while the AIC and residual squares were lower in the GWR models than in OLS. Heavy metal pollution connects tightly with human activities, mineral deposits and metallic characters and applications.

The model used for Arsenic in this study explained 30 percent of the variance through eleven independent variables (Table 5), in which urbanization, agriculture, economic development, and arsenic pollution levels in 2004 were significant. Urbanization and agriculture had negative influences on arsenic pollution levels in 2011, while economic development and 2004 arsenic pollution levels had positive impacts. Both urbanization and 2004 Arsenic pollution levels had the greatest impact in the eastern and central regions, while agricultural and economic development had the most impact in the western areas (Fig 12). More specifically, urbanization influenced mainly the Liaohe River, Haihe River, and Huaihe River Watersheds in addition to the Yangtze Delta. Heilongjiang and Zhemintai Watersheds were mainly affected by pollution levels of Arsenic in 2004. Agriculture-dominated areas had larger potential consumptions of pesticide containing Arsenic pollutants. The GWR results showed that highly urbanized areas along with higher developed economies may have relatively lower agricultural to industrial proportions from east to west, resulting in lower pesticide consumption, thereby reducing arsenic pollution. However, there is another potential cause. Natural mining resources and geologic features also have profound influences on heavy metal pollution. For example, naturally formed arsenic in soil is the culprit for the water pollution in India, Bangladesh, and the western part of China; especially in Tibet (Schwarzenbach et al., 2010). Foxconn is a case in point. Many manufacturing bases have been set up in the south and east rural areas of China. Many of these production centers have formed matured production line and independent social systems, which highly disturbs the local population structure of prefecture level cities, when using population density as a measure of urbanization.

A positive correlation was observed between urbanization and chromium pollution levels, mainly influencing the eastern and central coastal areas. Higher population densities, were related to higher pollution areas. The impacted regions include Heilongjiang, Liaohe River, Haihe River, Minzhetai Watersheds, and the Yangtze Delta. Transportation was another significant variable for the chromium GWR model. The overall impact of transportation on cadmium and chromium pollution was negative. One explanation could be that high road density indicates areas of busy traffic, thereby increasing vehicle emissions. Nonetheless, higher road density could result in less traffic on a single route, perhaps reducing the likelihood of an accidental spill due to a vehicle accident. This could result in a broader polluted area but lower contamination levels (Wen and Yang, 2010). Transportation had the strongest positive association with both chromium and cadmium pollution in the western corner of China.

Similar to arsenic, agriculture had a negative association with both cadmium and lead, while economic development had a positive relationship. GDPPC was used to indicate the level of economic development. Gross Domestic Product value includes almost every aspect in people's lives, including agriculture, industry, etc. As expected, the multiple regression results showed that the higher GDPPC levels yielded higher pollution levels of arsenic, cadmium and lead. GWR models showed 53 percent and 40 percent of respective cadmium and lead level changes in 2011. However, GDP from enterprises may increase due to budget savings from water pollution treatment and facilities control. The overall spatial patterns of agriculture were similar, despite its negatively relationship with Cadmium and Lead. All have the greatest positive influence in the western corner of China. Because agricultural data was calculated using the total annual agricultural output over the total GDP, the agricultural results were inconsistent with previous findings. Therefore, the percentage of agricultural output does not necessarily correspond positively with the absolute value of agricultural output. For instance, Nanyang and Shijiazhuang had almost the same agricultural output in 2011 (41.5 billion Yuan). While the agriculture was 20 percent over the total GDP in Nanyang, it was only 10 percent of the total in Shijiangzhuang. The 2004 lead pollution level positively influenced the lead level in 2011 the most, compared with the other two variables (coefficient is 0.069 at 1% significant level). This finding is particularly reflected in southern China (Fig 6). Also, the main source of heavy metal pollution data came from industrial discharged wastewater.

The GWR model accounted for 38 percent of mercury contamination variations in 2011. The mercury regression model was very sensitive to two variables: GRE and EMP. The influence of green area percentage (GRE) and the percentage of persons employed in mining and quarrying (EMP) showed spatial variations. Except in the northeast corner, green area coverage had a positive association with mercury pollution, especially in South China. Mercury resources in China are mainly concentrated in the south. It is also the most populated area with a relatively highly developed economy with extensive uses of automobiles. Pollution problems and human activities were strongly related. Human activities, including coal, gas, and oil combustion emit mercury into air, which then contaminates soil and large water bodies. This can lead to indirect ingestion by humans though fish. The percentage of persons employed in mining and quarrying reflects the magnitude of mining activities and mineral reserves of one city, indirectly indicating pollution levels (Zhai et al., 2008). This variable was negatively associated with mercury levels for the whole country. This negative relationship was particularly strong in northwest China. One possible explanation is that government statistics on persons employed in mining and quarrying might may be inaccurate when disregarding the influence of strong migration forces. Another factor could be that the Chinese government signed the Minamata Convention treaty and followed the regulations to prevent emissions and limit usage of mercury, (UNEP, 2013).

Summary

Natural mineral resources and human activities are the main source of heavy metal pollution. For instance, Chenzhou and Changde (in Hunan Province, in Southern China) are classified as high level Arsenic pollution cities, shown in the 2011 LISA high-high clusters map (Fig. 2D). Chenzhou has abundant tungsten and bismuth ores. Hundreds of minerals have been ascertained in Changde, especially rich realgar ore, which is one of the main resources of arsenic production (Land and Resources Bureau of Changde, 2011). Luoyang, part of Henan Province in central China, is a populated city with the largest Molybdenum reserve in the country. Not surprisingly, it had very high chromium and lead pollution levels. These highly polluted industrial based cities all have many different types of abundant mineral resources and heavy mining activities. Geographic location (i.e. coastal areas or local enclosed water bodies, and hydrologic characteristics) plays an important role in diagnosing pollution levels (Liu et al., 2011). Taizhou, Wenzhou and Suzhou and surrounding coastal areas also had relatively high chromium pollution concentration in 2011.

Because of the high toxicity of these heavy metals, the Chinese government has established strict pollution control regulations. In 2002, the Chinese government issued environmental quality standards for surface water (GB 3838-2002) regulations to help control water pollution. Therefore, many companies introduced more environmentally friendly technologies and equipment. However, bias may exist in raw data due to deliberate false reports of lower discharged pollutant values by enterprises to avoid penalties. Other influential forces such as water quality regulations of local government forces, migration, and landscapes etc. were not taken into consideration but may also contribute to heavy metal water pollution.

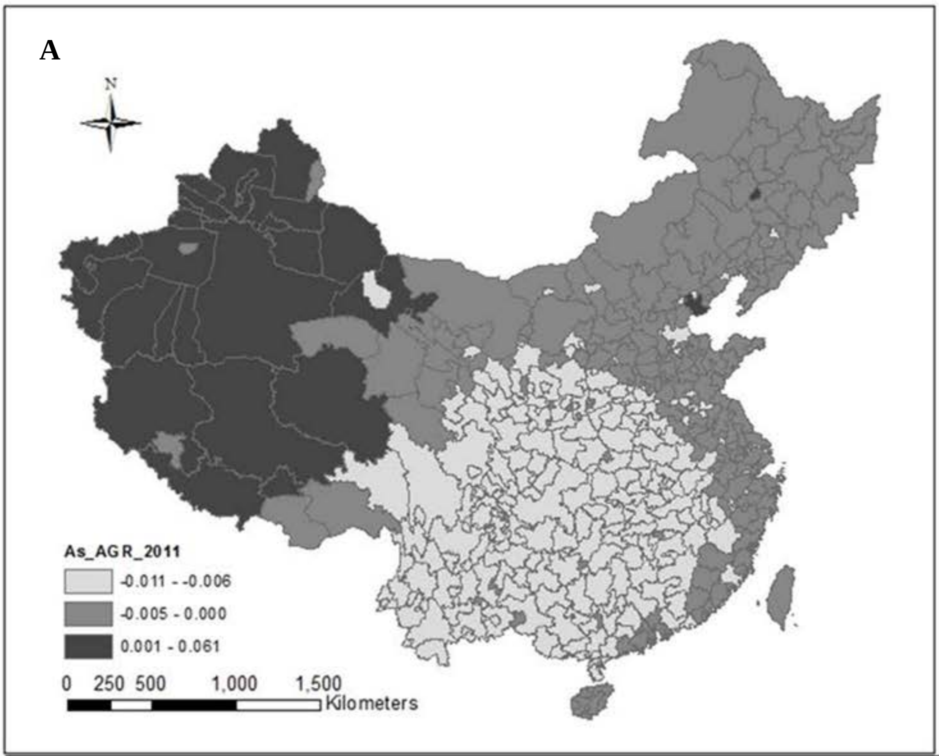
Table 5. Heavy metal multiple regression results

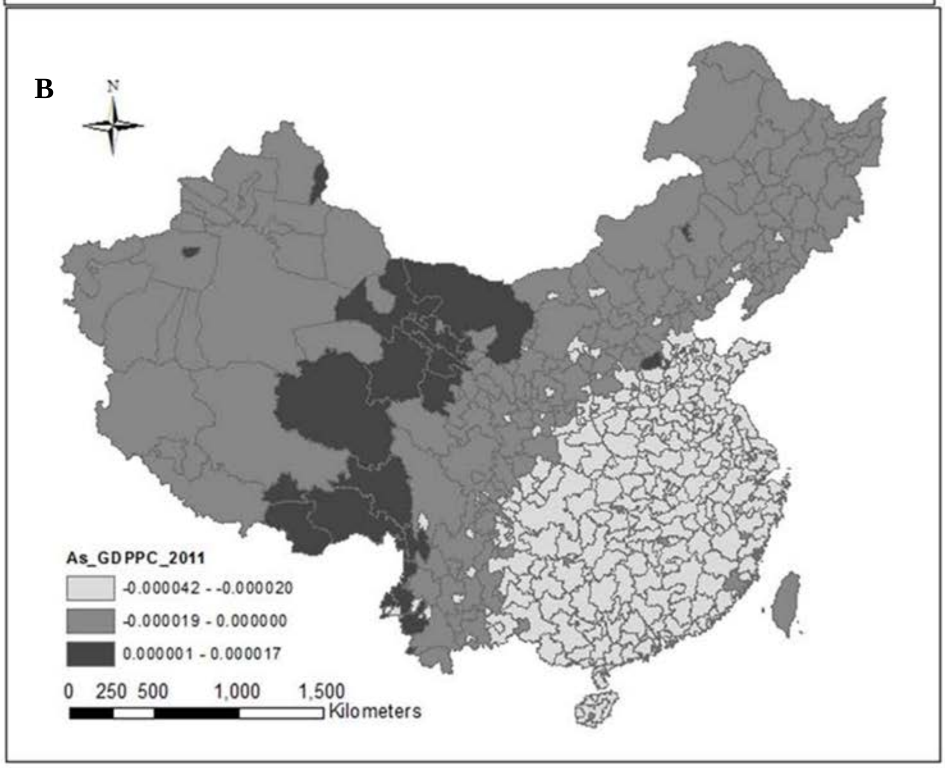
Independent variables	Coefficients (As)	Coefficients (Cd)	Coefficients (Cr)	Coefficients (Hg)	Coefficients (Pb)
Intercept	1.005***	0.284***	1.052	0.005	1.051***
Transportation (TRA)	-0.068	-0.033**	-0.272*	0.0004	-0.075
Urbanization (URB)	-0.00026**	0.000	0.0007***	0.000	-0.0003
Industrialization (IND)	0.00093	0.0002	0.0002	0.000	0.001
Globalization (GLO)	0.000	0.000	-0.0002	0.000	0.000
Agricultural (AGR)	-0.012***	-0.002**	0.0005	-0.0001	-0.009*
Economy (ECO)	0.000**	0.000***	0.000	0.000	0.000***
Heavy metals in 2004	0.039*	0.027	0.038	0.006	0.067***
Distance (DIS)	-0.12	-0.013	0.235	-0.0003	-0.013
Green coverage rate (GRE)	-0.0017	0.000	0.015	0.0002*	0.008
Hydrology (HYD)	0.0025	0.001	-0.172	0.001	-0.107
Employment (EMP)	-0.044	-0.007	-0.020	-0.002***	-0.034
Significance F	0.001***	0.00***	0.048**	0.028**	0.0001***
R-square	0.21	0.29	0.12	0.16	0.23

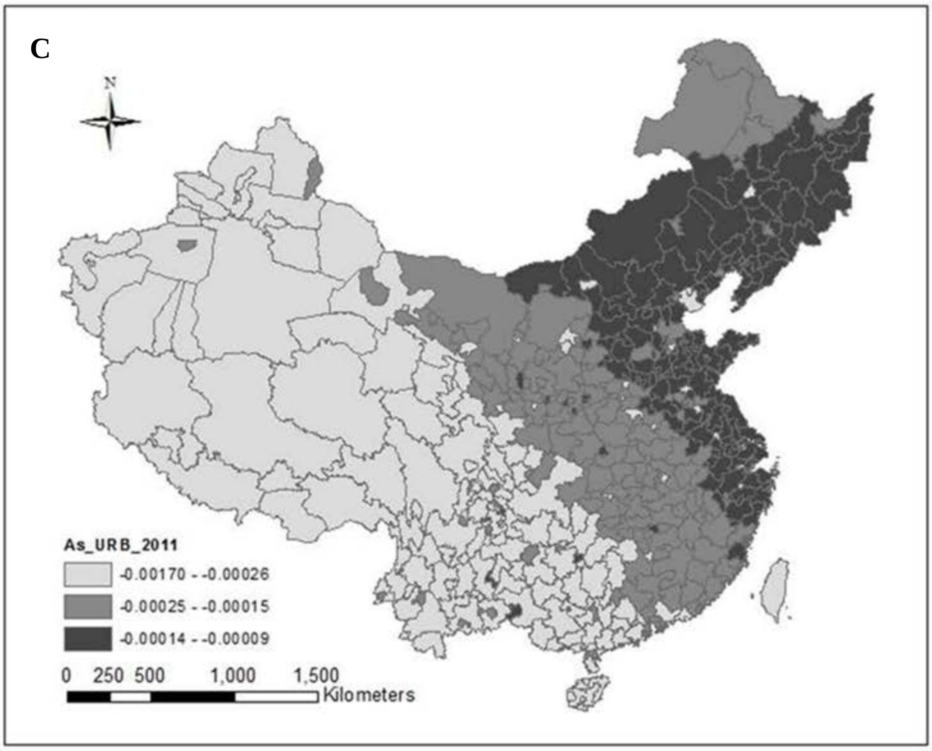
Note: *p-value is significant at 10% significance level **p-value is significant at 5% significance level ***p-value is significant at 1% significance level

Table 6. Comparison between OLS and GWR models (with kriging estimation value in 2011)

	As		Cd		Cr		Hg		Pb	
	OLS	GWR	OLS	GWR	OLS	GWR	OLS	GWR	OLS	GWR
Multiple R^2	0.21	0.30	0.29	0.43	0.11	0.30	0.16	0.21	0.23	0.39
Adjusted R^2	0.14	0.22	0.23	0.35	0.05	0.18	0.08	0.13	0.17	0.29
AICc	161	155	-280	-296	477	462	-915	-918	314	298
Residual Squares	22	19	1	0.8	164	128	0.007	0.006	61	48


A
N
As_AGR_2011
-0.011 - -0.006
-0.005 - 0.000
0.001 - 0.061
0 250 500 1,000 1,500
Kilometers


B
N
As_GDPPC_2011
-0.000042 - -0.000020
-0.000019 - 0.000000
0.000001 - 0.000017
0 250 500 1,000 1,500
Kilometers



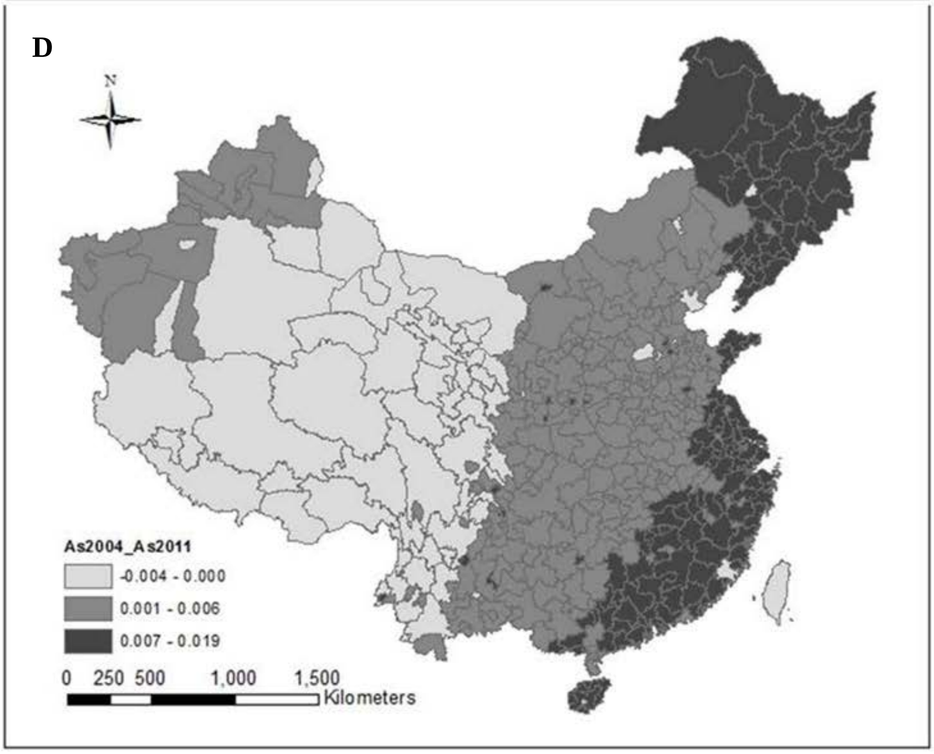
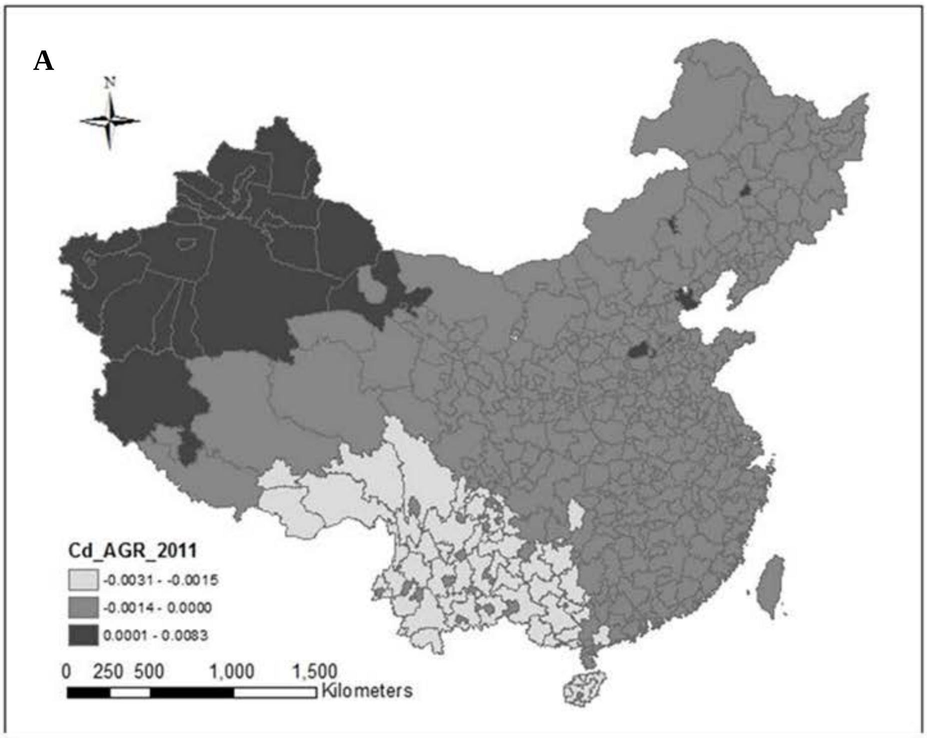
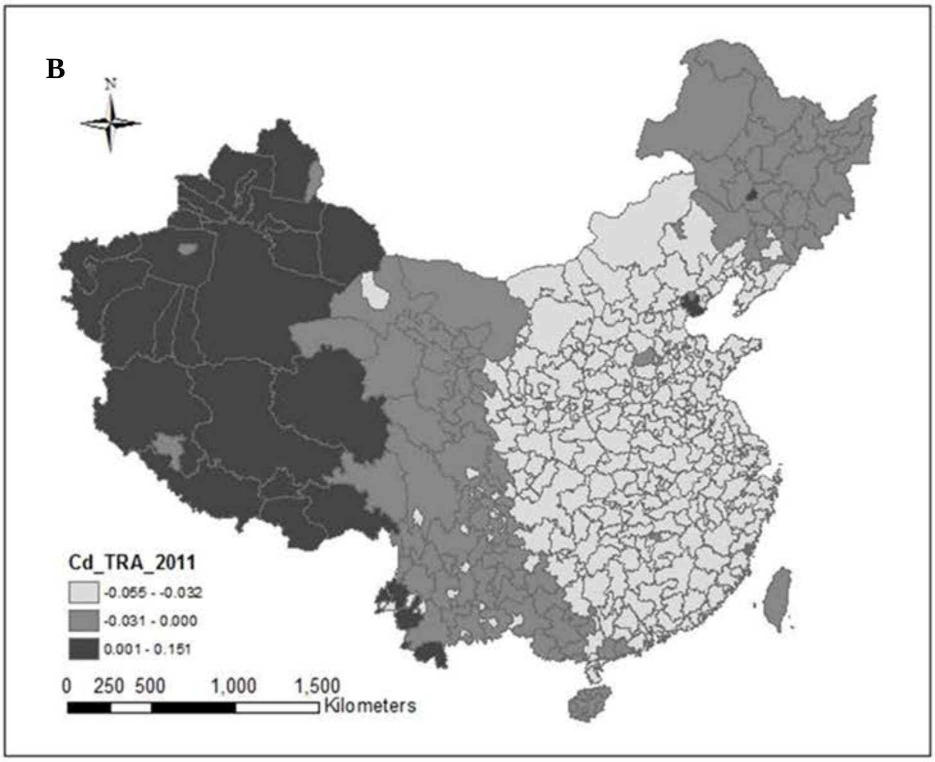


Fig. 12. Spatial variations of Arsenic pollution level in 2011

(A) Arsenic vs. Agriculture
(B) Arsenic vs. Economic Development
(C) Arsenic vs. Urbanization
(D) Arsenic 2011 vs. Arsenic 2004


A
N
Cd_AGR_2011
-0.0031 - -0.0015
-0.0014 - 0.0000
0.0001 - 0.0083
0 250 500 1,000 1,500
Kilometers


B
N
Cd_TRA_2011
-0.055 - -0.032
-0.031 - 0.000
0.001 - 0.151
0 250 500 1,000 1,500
Kilometers

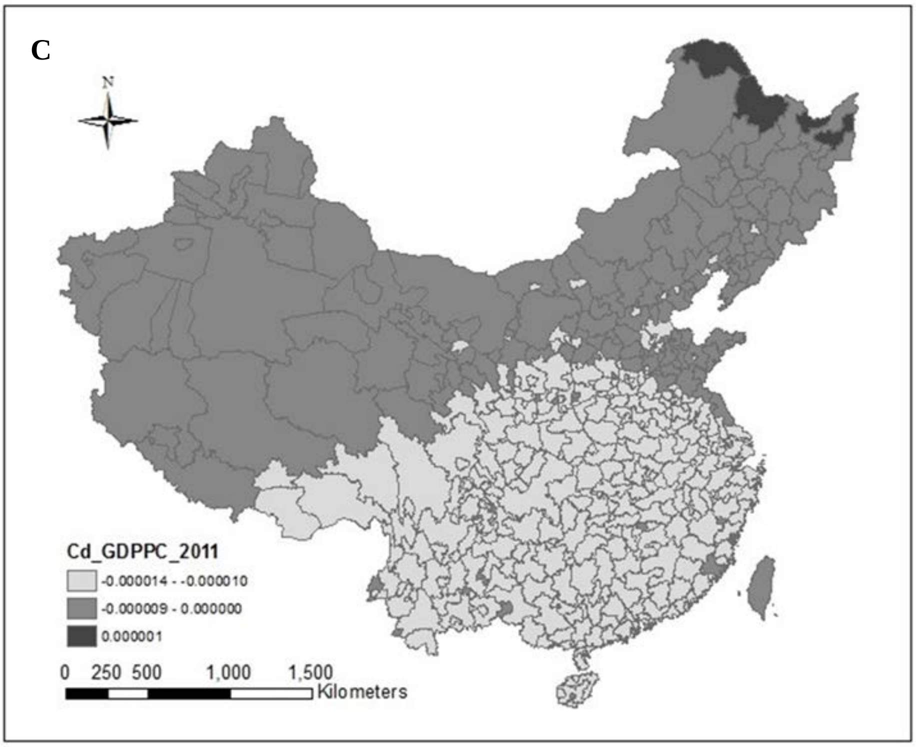


Fig. 13. Spatial variations of Cadmium pollution level in 2011

(A) Cadmium vs. Agriculture
(B) Cadmium vs. Transportation
(C) Cadmium vs. Economic Development

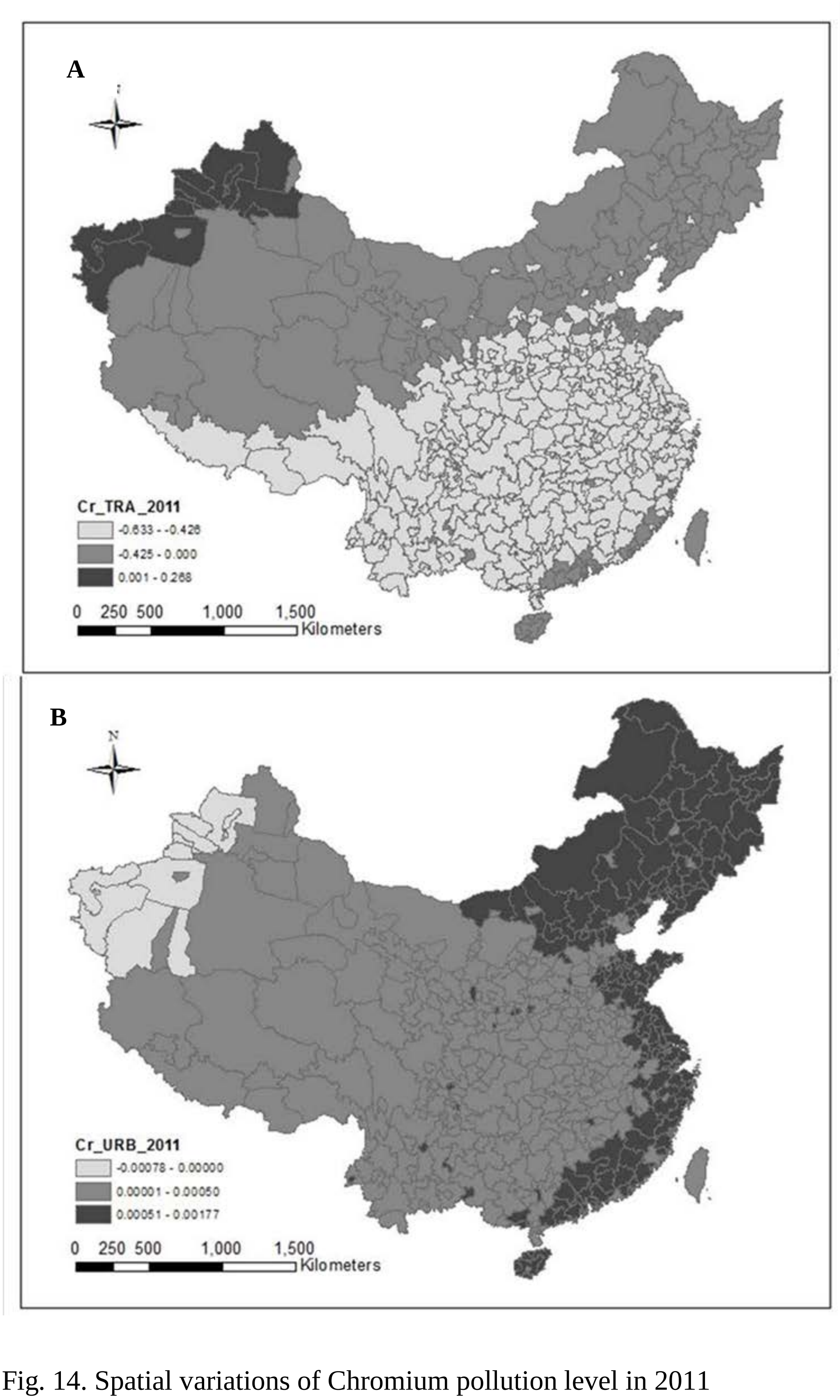


Fig. 14. Spatial variations of Chromium pollution level in 2011

(A) Chromium vs. Transportation

(B) Chromium vs. Urbanization

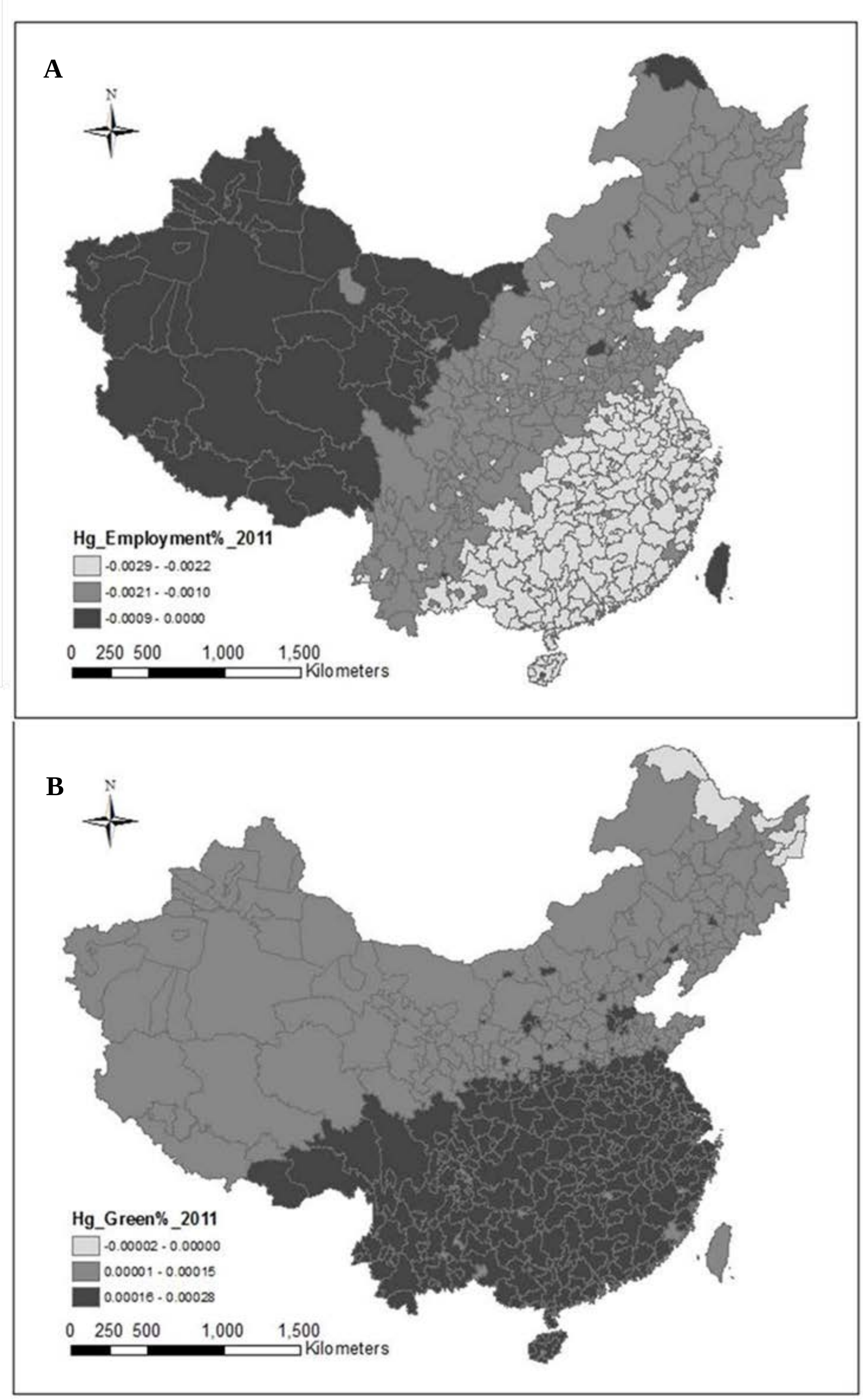
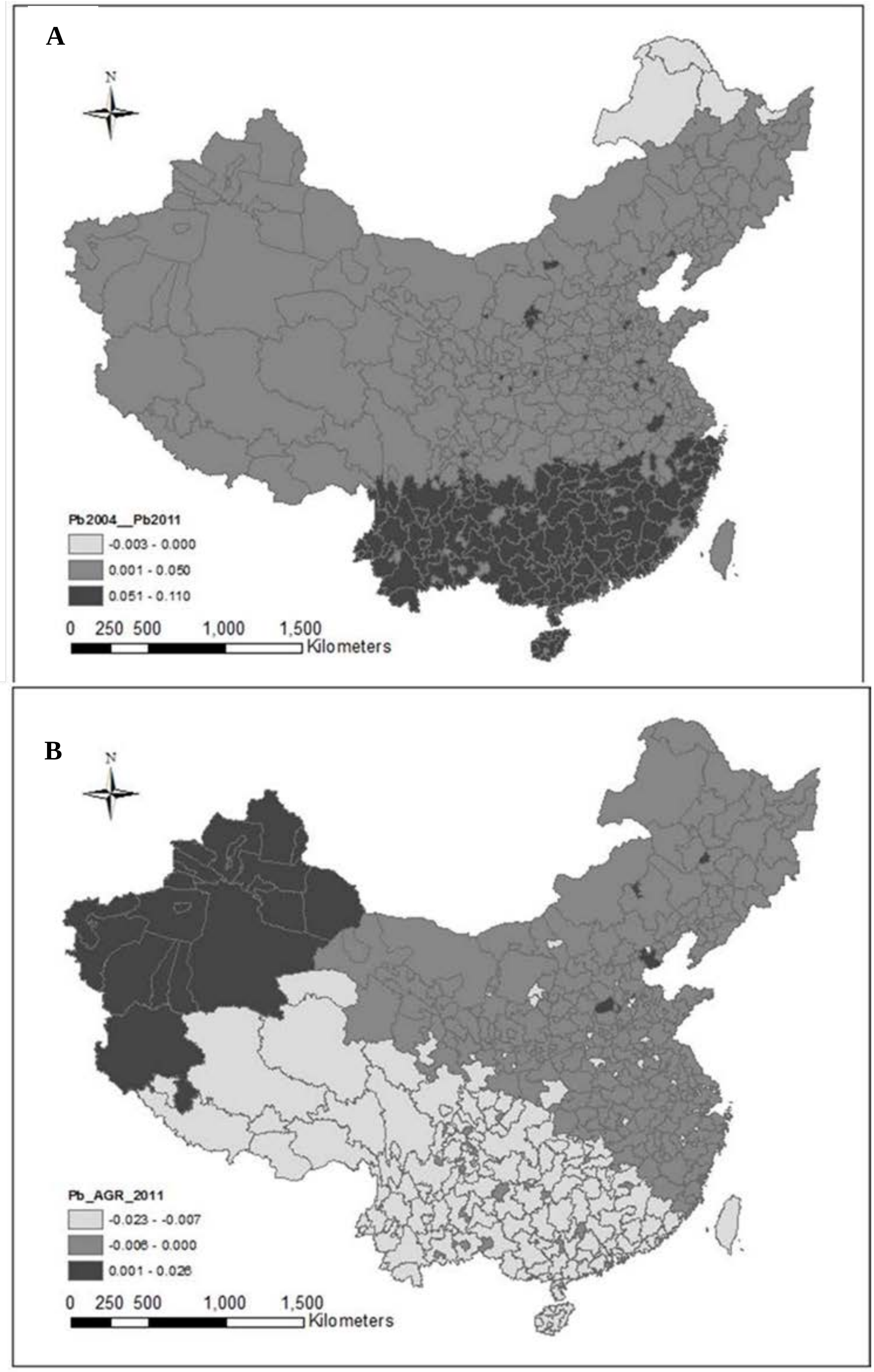


Fig. 15. Spatial variations of Mercury pollution level in 2011

(A) Mercury vs. Employment

(B) Mercury vs. Greenery Coverage


A
N
Pb2004__Pb2011
-0.003 - 0.000
0.001 - 0.050
0.051 - 0.110
0 250 500 1,000 1,500
Kilometers
B
N
Pb_AGR_2011
-0.023 - -0.007
-0.006 - 0.000
0.001 - 0.026
0 250 500 1,000 1,500
Kilometers

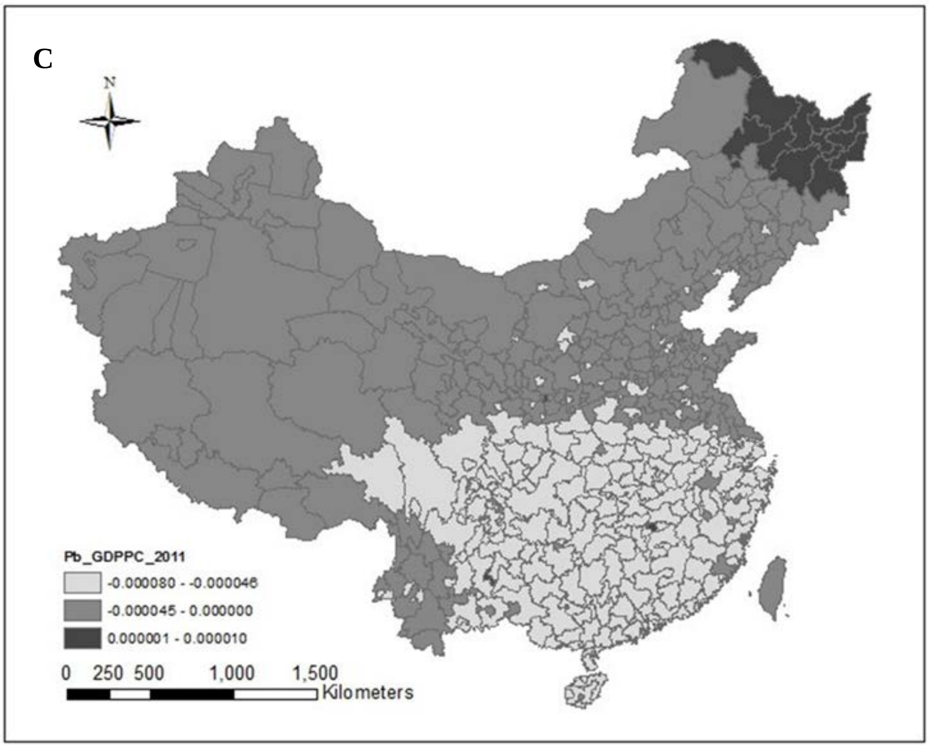


Fig. 16. Spatial variations of Lead pollution level in 2011

(A) Lead 2011 vs. Lead 2004

(B) Lead vs. Agriculture

(C) Lead vs. Economic Development

4.3 Human health risk assessment

4.3.1 Spatial and temporal variations of cancer patients

Based on the cancer maps for both 2004 and 2009, most of the cancer incidences occurred in east and central regions. Kernel estimation results indicated clusters of cancer incidences in Beijing, Shanghai, Guangzhou (capital city of Guangdong Province, South China), Wuhan (capital city of Hubei Province, South China), and Shenyang (capital city of Liaoning, Northeast China). While all areas showed a positive correlation between cancer incidences, water pollution levels, and economic development; an especially strong positive relationship was observed between cancer incidences, industrial expansion, and population boom. All prefecture-level cities with relatively high cancer incidences were located downstream of the Yellow River and the Yangtze River delta. Specifically, Shanghai and surrounding areas documented the highest incidences of all seven cancers, clearly shown in the 2004 and 2009 spatial distribution and kernel estimation maps (Figs 17-23). Clusters of five cancer types, including liver, trachea/bronchus/lung, rectum, colon, and brain/nervous system were identified in the areas corresponding with Beijing, Shanghai, Wuhan, Guangzhou, and Shenyang. One main cluster of esophagus and stomach cancer, developed in the surrounding areas of Shanghai, Suzhou, Nantong, and Yancheng (all in Jiangsu Province, Southeast China). These results are particularly consistent with Liu's study which shows China's cancer villages map (2010).

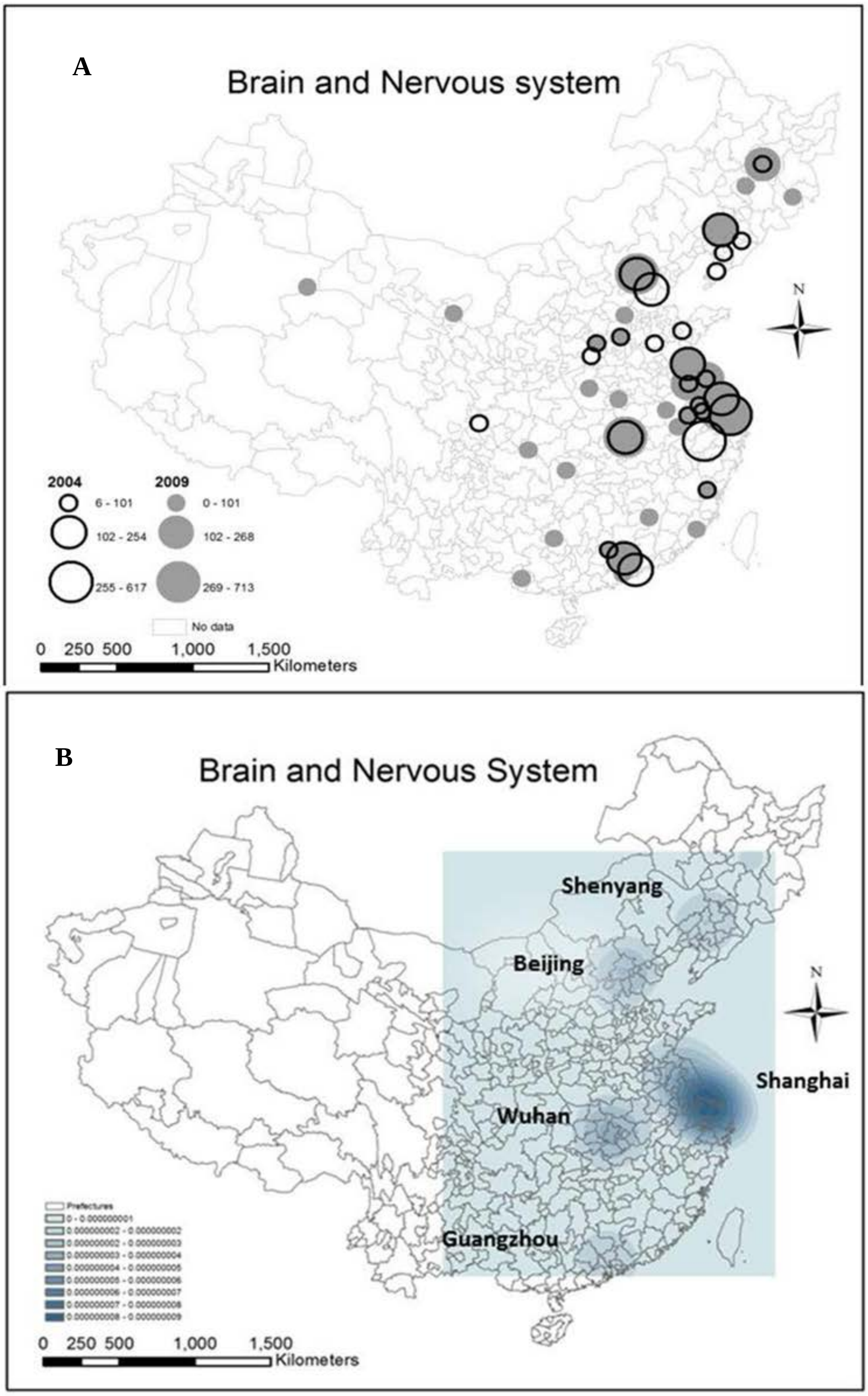


Fig. 17 (A) Spatial distribution of BN between 2004 and 2009

(B) Kernel estimation of BN in 2009

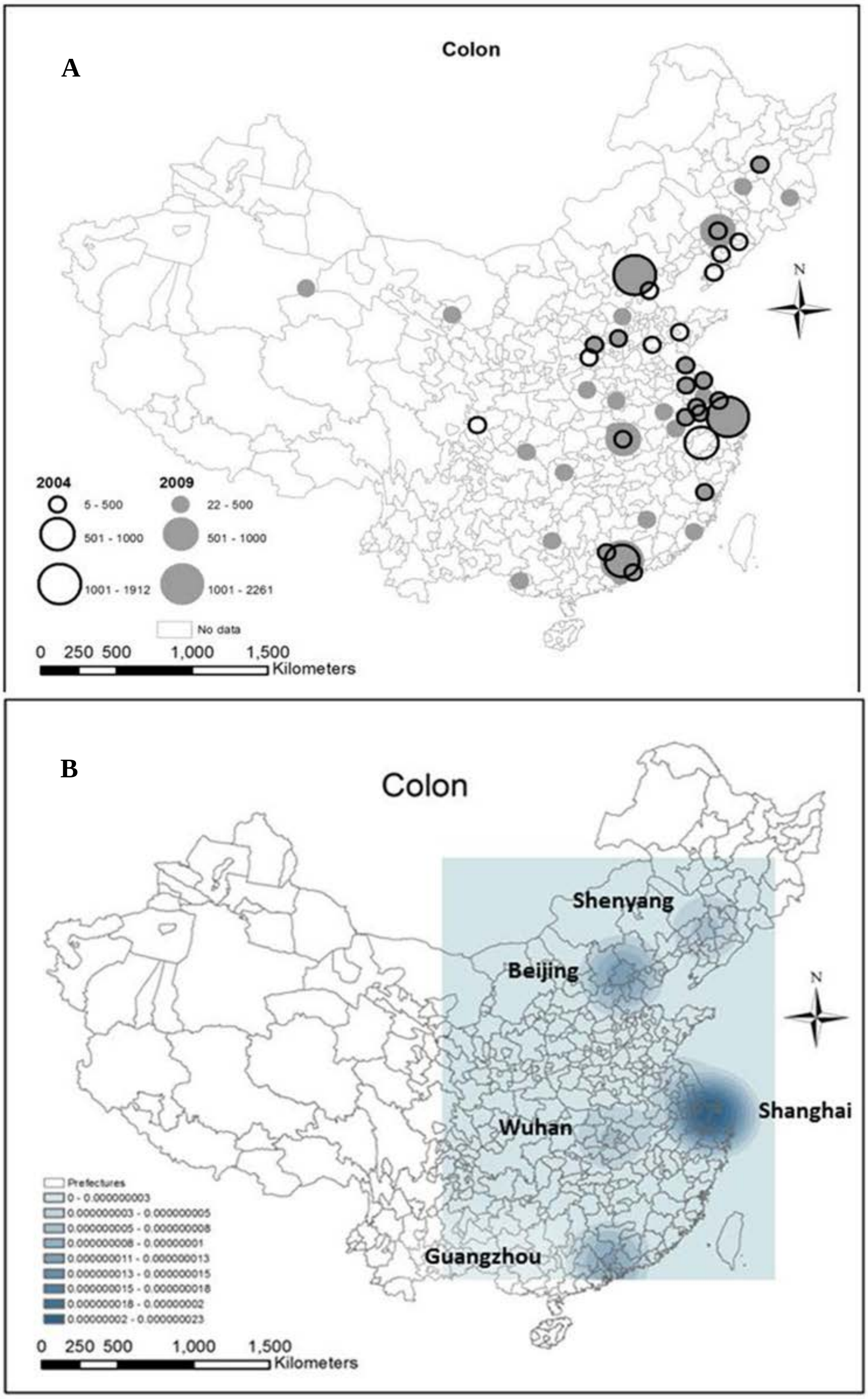


Fig. 18 (A) Spatial distribution of Colon between 2004 and 2009

(B) Kernel estimation of Colon in 2009

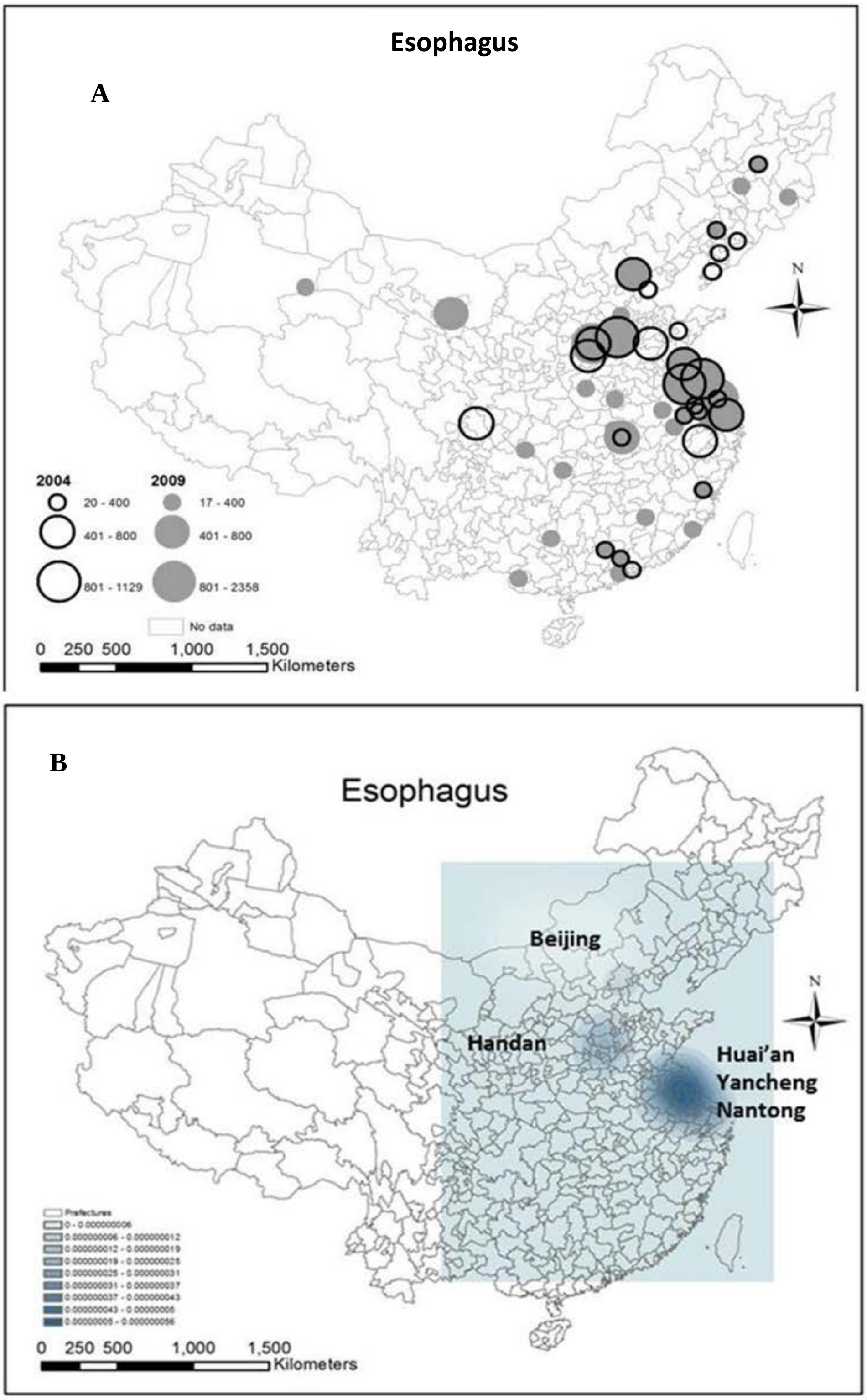


Fig. 19 (A) Spatial distribution of Esophagus between 2004 and 2009

(B) Kernel estimation of Esophagus in 2009

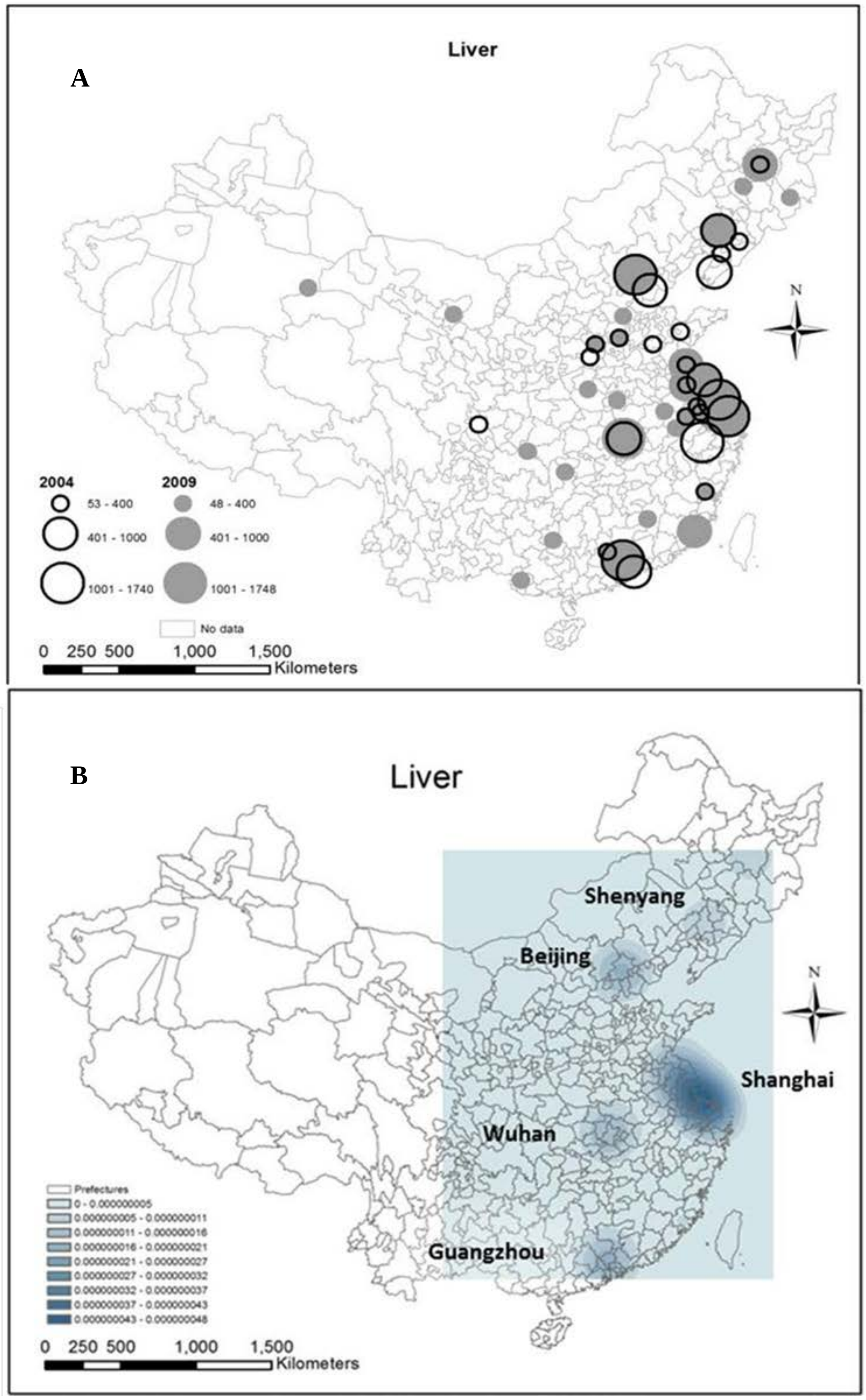


Fig. 20 (A) Spatial distribution of Liver between 2004 and 2009

(B) Kernel estimation of Liver in 2009

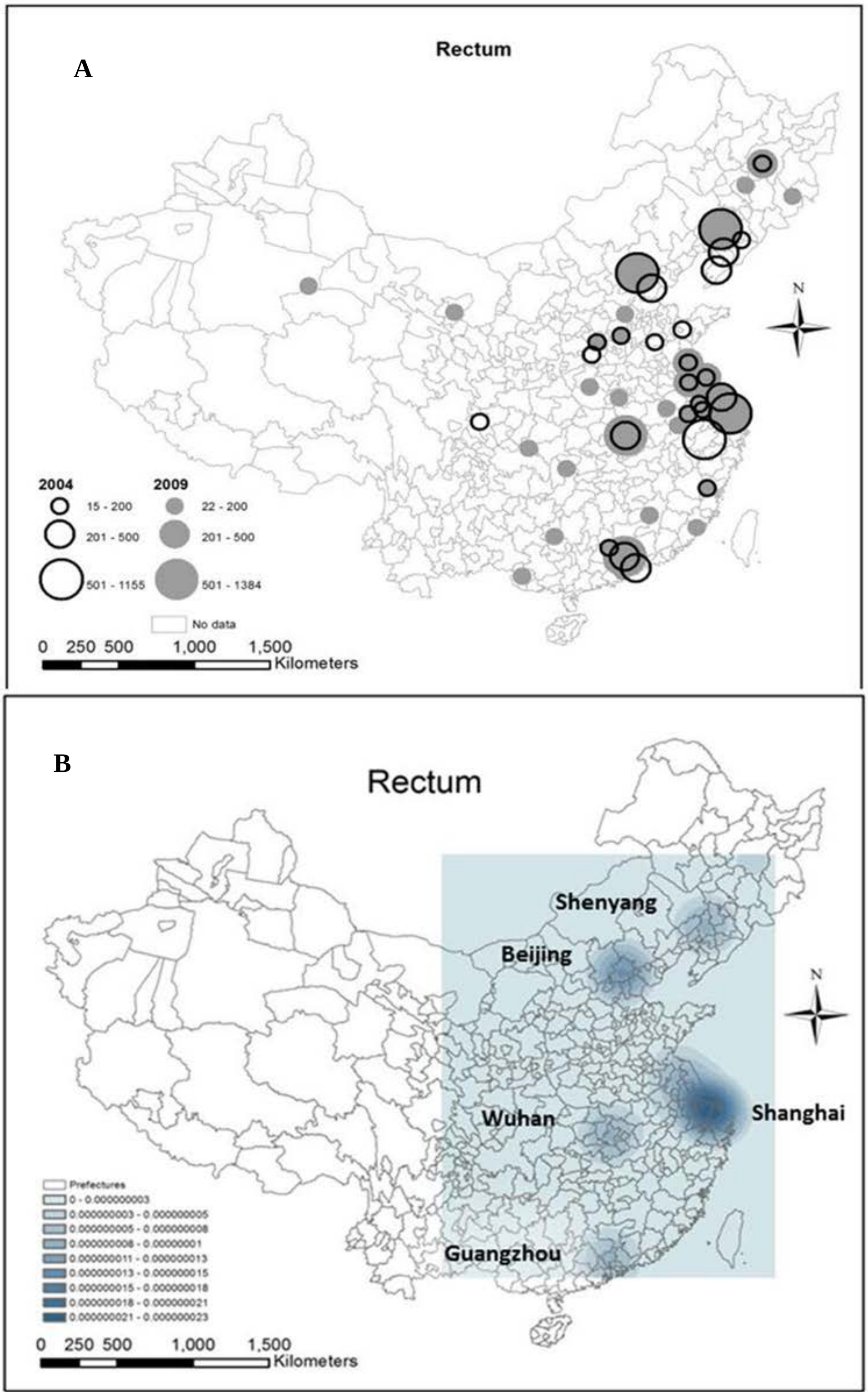


Fig. 21 (A) Spatial distribution of Rectum between 2004 and 2009

(B) Kernel estimation of Rectum in 2009

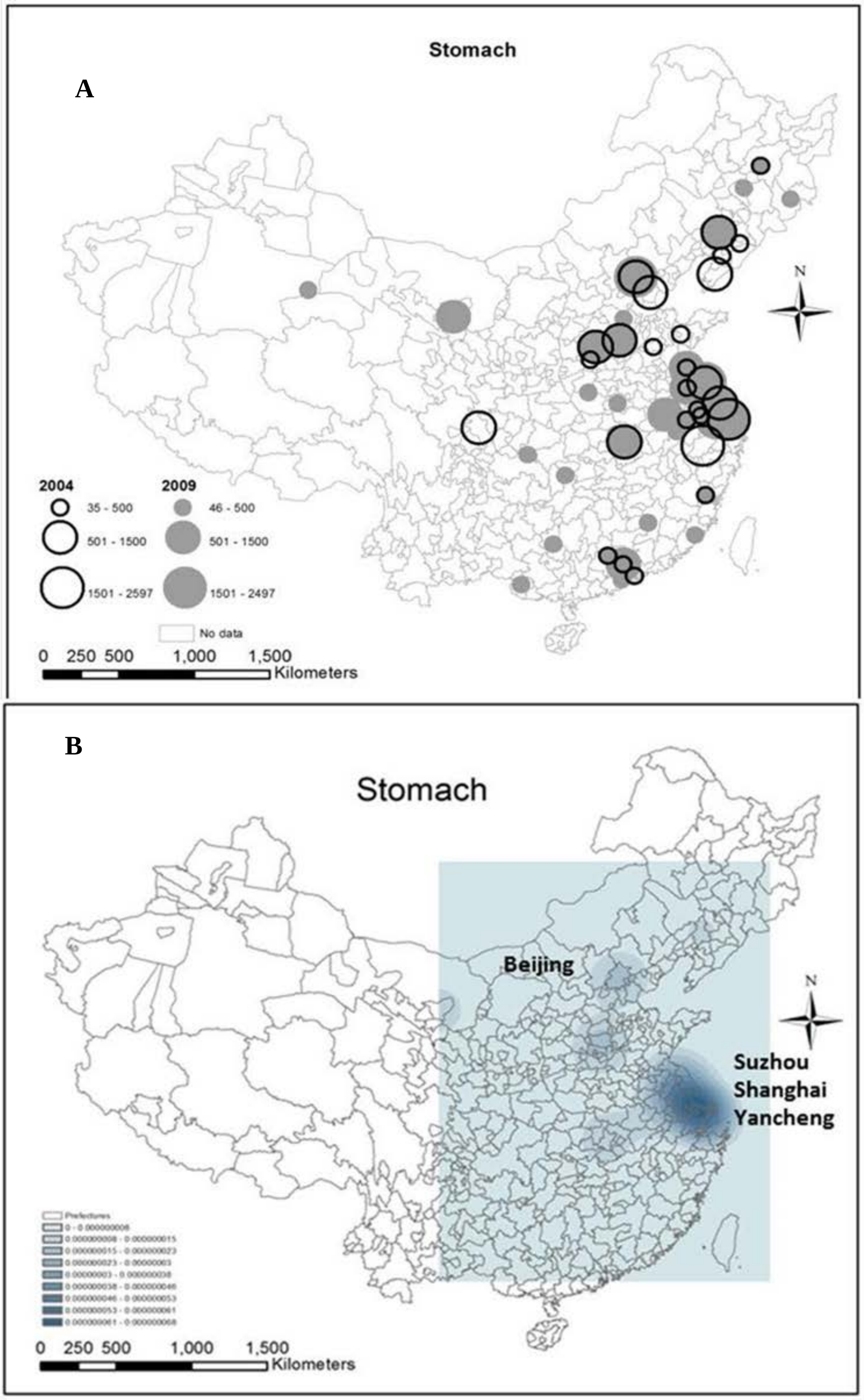


Fig. 22 (A) Spatial distribution of Stomach between 2004 and 2009

(B) Kernel estimation of Stomach in 2009

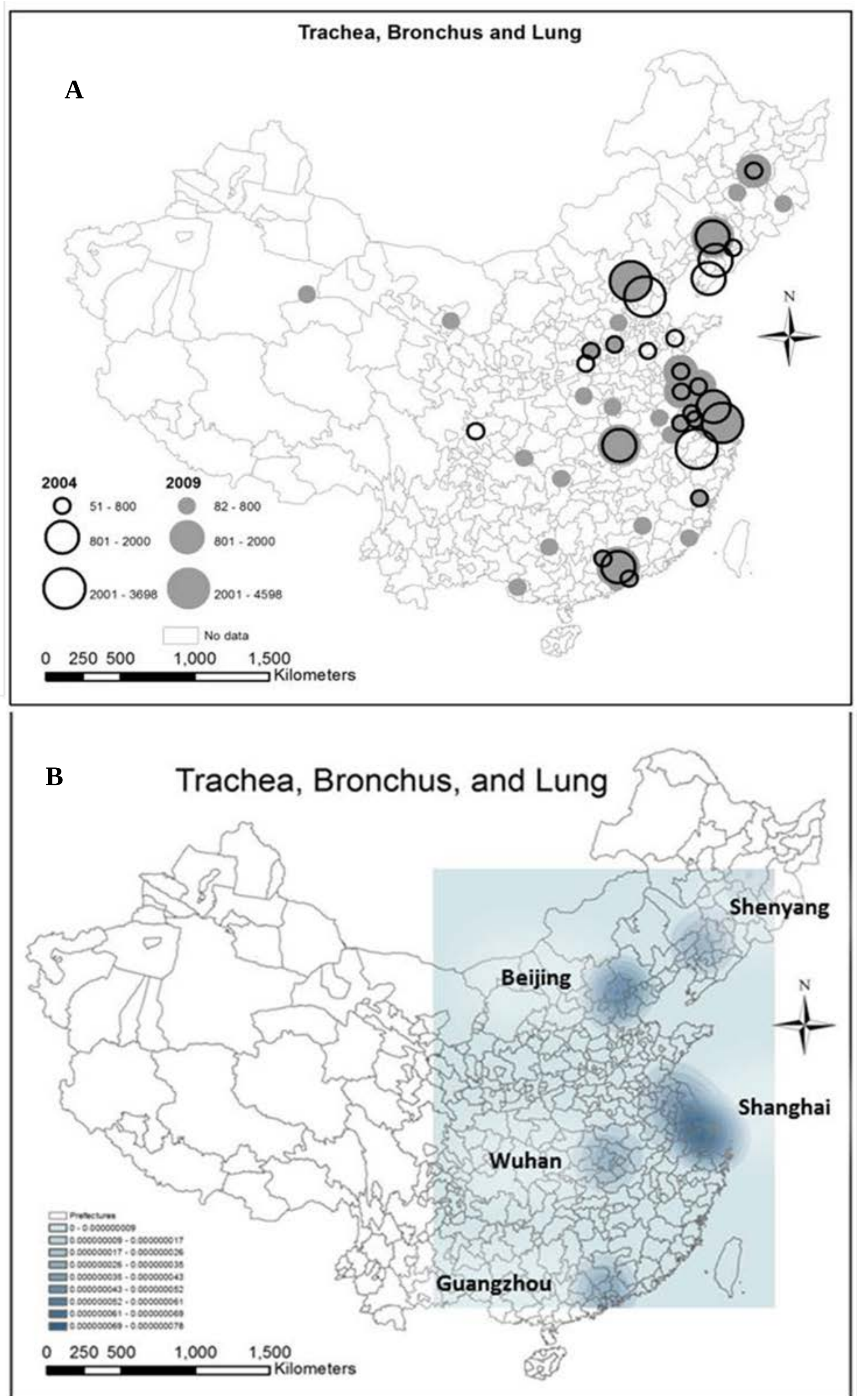


Fig. 23 (A) Spatial distribution of TBL between 2004 and 2009

(B) Kernel estimation of TBL in 2009

4.3.2 Socioeconomic transitions, physical conditions, and human health

Considering the potential multicollinearity problems, the variance inflation factors (VIFs) of the seven models were examined in ArcGIS. According to one criterion, multicollinearity exists when the largest VIF exceeds 10, indicates strong corrections among independent variables (Chatterjee et al., 2000; Wen et al., 2003; Li and Wei, 2010b). Therefore pollution level of lead (VIF = 15.4), and economic development (VIF = 14.9) were dropped since their VIFs were larger than 10. The number of independent variables was thus reduced from 16 to 14 in the health models. All models were run using variables with VIFs smaller than 10. All seven multiple regression models for health analysis were significant at 1 percent confidence level, except for esophagus which was significant at the 10 percent level (Table 7). the R-squared ranged between 0.56 and 0.87 for all seven cancer models, indicating at least 56 percent of variations in cancer incidences were explained by 14 independent variables, reflecting heavy metal water contamination, socioeconomic transitions, and physical conditions of cities. The reliability and performance of these seven cancer models were demonstrated through both significance F and R-squared values.

Based on the results of multiple regression analysis for esophagus cancer, three independent variables significantly influence the incidences of esophagus cancer. Specifically, the mercury pollution level was positively related to the incidences of esophagus cancer, indicating that higher mercury pollution levels increased the risk of esophagus cancer for local residents. Mercury, one of the most toxic heavy metals, poisons the human body directly through the food chain (especially seafood) and long-term exposure (via skin, hair, etc.) to contaminated environments (Jarup, 2003; Morais et al., 2012). Therefore, oral and digestive systems (esophagus, stomach, liver, etc.) face great potential carcinogenic risks (Jarup, 2003; Duruibe et al., 2007). As expected, sewage treatment had a negative impact. Results indicated a higher sewage treatment rate improves water quality, limiting cancer incidences. Downstream areas were more polluted; thus causing more cancer cases, consistent with the spatial distribution maps and kernel estimation results in Fig.19.

Five independent variables significantly influenced stomach cancer incidences. Three of the five, mercury pollution levels, sewage treatment rates, and hydrologic characteristics, also influenced esophageal cancer incidences. Urbanization and industrialization also had a positive correlation with stomach cancer incidences, indicating higher population densities and industrial outputs of cities could result in increased documented cancer incidences.

The colon and rectum both belong to the large intestine, and absorbing water is one of their main functions. Industrialization and distance to the nearest major water body were the two crucial variables regarding colon cancer incidences. Both are positively related to cancer incidence numbers, indicating that, cancer incidence increases with industrialization and shorter distances to major water bodies. Urbanization, transportation and industrialization were all significant variables concerning rectum cancer. Hu et al. (2014) demonstrated that the shift in industrial estates from urban to rural compromised environmental and residential health protection. Urbanized cities are densely populated with better traffic conditions and medical services; they also have stricter pollution prevention. Given the urban versus rural health care inequality issues in China (Li and Wei, 2014), more rural cancer patients could be transferred diagnosed, treated, and documented in urban cities as urban patients.

Mercury pollution levels, population density and distance to the nearest major water body were positively associated with liver cancer. Liver is the most important detoxifying organ for human beings. Therefore it faces great heavy metal poisoning potential, especially from mercury and lead (Cave et al., 2010). The multiple regression results of TBL (trachea, bronchus, and lung) showed similarities to the colon cancer model. Industrialization and the distance between a prefecture-level city and the nearest major water body were the only two positive significant variables found.

The brain and nervous system model is more complicated with six significant variables. Pollution levels of mercury showed the strongest influential power, supported by many previous studies (Risher and DeWoskin, 1999; Jarup, 2003; Duruibe et al., 2007; Morais et al., 2012). Although previous research has proven that lead could seriously damages the human brain and nervous system, this research could not verify that conclusion, unfortunately. Lead pollution levels were removed to avoid multicollinearity problems. Urbanization, industrialization, and distance to nearest major water bodies also caused increased numbers of nervous system and brain cancer incidences, while chromium pollution level and transportation conditions showed negative coefficients. Multiple regression models results are summarized in Table 7.

Table 7. Multiple regression results for health

Independent variables	Coefficients (Esophagus)	Coefficients (Stomach)	Coefficients (Colon)	Coefficients (Rectum)	Coefficients (Liver)	Coefficients (TBL)	Coefficients (BN)
Intercept	322.87	355.15	27.44	45.46	243.59	180.19	25.41
Qualified Rate	-4.39**	-3.19*	0.42	0.20	-0.02	1.05	0.21
As2011	-511.49	-501.04	-130.35	20.58	-144.80	388.03	-47.85
Cr2011	-69.68	-157.04	31.69	-47.26	21.17	-182.10	-32.47*
Hg2011	12477.94*	17770***	-329.97	5262.83	14408.70**	19054.45	4068.7***
Cd2011	438.35	349.75	-315.55	-239.74	-419.86	-1123.35	-42.57
Urbanization (URB)	0.38	0.39*	0.17	0.25*	0.38*	0.82	0.15***
Transportation (TRA)	-29.05	-136.27	-181.17	-205.93**	-218.36	-700.65	-114.53***
Industrialization (IND)	-0.36	1.72*	1.22*	1.03**	0.61	3.74*	0.30*
Agricultural (AGR)	-3.65	-6.83	-2.86	-3.10	-0.37	-6.25	-1.39
Globalization (GLO)	-4.93	0.34	5.62	-0.56	-1.52	-8.79	0.89

Distance (DIS)	-44.66	63.39	368.21**	304.88	347.23*	1101.91**	104.48**
Hydrology (HYD)	321.82**	266.72**	-83.27	2.85	77.53	-24.38	26.48
Green coverage rate (GRE)	1.91	0.71	-0.12	-1.16	-5.27	-3.44	-0.59
Employment (EMP)	9.46	-103.85	-59.44	-54.65	-104.23	-279.21	-33.67
Significance F	0.086*	0.0003***	0.000***	0.0002***	0.003***	0.004***	0.000***
R-square	0.56	0.78	0.83	0.79	0.72	0.70	0.87

Note: *p-value is significant at 10% significance level **p-value is significant at 5% significance level ***p-value is significant at 1% significance level

5. Conclusions and Significance

This research investigated heavy metal water pollution and human health conditions in mainland China from a spatial-temporal perspective while examining the influences of anthropogenic activities on water pollution. Based on the findings, China has faced very serious heavy metal water contamination issues due to the rapid economic development and socioeconomic transitions. From the overall spatial patterns of heavy metal water pollution, severely contaminated areas were concentrated in the Yellow River, Yangtze River and Zhujiang River watersheds, especially within Chongqing, Kunming (Yunnan Province) and Zhuzhou (Hunan Province), in Southern China. As expected, high pollution levels were associated with both anthropogenic activities and natural physical environments, including areas of abundant mineral resources, and heavy mining activities. Economic development and urban green area regulations also play important roles in controlling water pollution problems. Most of the prefecture-level cities are traditional industrial centers and are highly economically developed with large populations.

This research also analyzed cancer clusters as one of the consequences of heavy metal water pollution. Based on the health data analysis, cancer patients are concentrated in highly urbanized areas, corresponding to more advanced, economic development and sophisticated medical services (Li and Wei, 2014). As expected, the analytical results of seven cancer diseases consistently matched the distribution of heavy metal water pollution. Cancer patient incidences were mainly concentrated in central and eastern regions of China. Five clusters of cancer incidences were detected in Beijing, Shanghai, Guangzhou, Shenyang, and Wuhan. Two heavy metals, chromium and mercury and six socioeconomic and physical factors were significant to spatial variations of cancer incidences.

This research will contribute to literature in the following aspects: First, a national-scale investigation was conducted to examine the spatial-temporal patterns of heavy metal water pollution levels in China from 2004 and 2011. Second, a literature search returned few results which employed kriging estimation for water pollution level prediction, combined with multiple regressions to analyze the interactions between China's heavy metal water pollution, socioeconomic transitions, and human health. The comparison between GWR and OLS methods took a step beyond previous studies by further investigating the impacts of anthropogenic activities on environmental pollution (Wang et al., 2013; Li et al., 2014). Third, this research has both scientific and policy implications by providing both macroscopical and detailed information on China's water pollution and human health problems, which may help researchers better understand the complex underlying risk factors. This research made a step forward compared to the previous studies, which neglected the spatial variation of the relationships among heavy metal water pollution, anthropogenic activities, and human health. These research findings will also provide valuable references for governmental agencies to initiate and adjust relevant policies for protecting water resources in addition to improving health conditions and citizens' quality of life of.

Due to the limitation of first-hand data and time constraints, water pollution data from many prefecture-level cities' were not available but still need examination (such as in the west region). Watershed-level water pollution data and analysis could improve this study. Unfortunately, only prefecture-level data are available at the national scale. This research only examined several factors related to water pollution. Other influential forces, such as local governmental policies on water pollution control, could be taken into account in future studies. Given the long term effects of heavy metal pollution on public health, this research could be enhanced by better quantity and quality cancer patient spatial-temporal data as well.

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