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The Spatial Distribution, Contamination Status and Contributing Factors of Heavy Metals in Cropland Soils of Twelve Cities in Shandong Province, China

Weina Xue ^{1,2}, Yanbo Peng ³, Aixia Jiang ¹, Taiyu Chen ² and Jiemin Cheng ^{1,*}¹ College of Geography and Environmental Science, Shandong Normal University, Jinan 250300, China; qingjeep@163.com (W.X.); 110051@sdnu.edu.cn (A.J.)² School of Municipal and Environmental Engineering, Shandong Jianzhu University, Jinan 250101, China; cty0826289@163.com³ Shandong Academy for Environment Planning, Jinan 250101, China; pengyanbo@sdaep.com

* Correspondence: 110087@sdnu.edu.cn; Tel.: +86-0531-86181939

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Abstract: The aims of this study were to analyze the spatial distributions of, contamination statuses of, and factors contributing to, heavy metals in cropland areas of different cities; thus, 55 agricultural soils were collected from 12 cities of Shandong Province, China. Concentrations of copper (Cu), lead (Pb), cadmium (Cd), chromium (Cr), nickel (Ni) and zinc (Zn) were determined. Results showed that average contents of Cu, Pb, Cd, Cr, Ni and Zn were 24.13, 31.77, 0.16, 130.63, 22.13, and 71.19 mg·kg⁻¹, respectively, and Pb and Cd had similar spatial distributions in those cities. Specifically, contents of Cr in cities of Weifang, Weihai, Yantai and Zibo were significantly higher than it in other cities; Weihai and Zibo also had significantly high contents of Zn. Moreover, concentrations of Cr in brown soils and cinnamon soils were significantly higher than that in fluvo-aquic soils, while other metals showed no significant differences among the soil types. Furthermore, Cu, Pb and Zn showed significant concentration decreases with respect to those measured in 2007. The correlation analysis and factor analysis indicated that the contamination of Pb and Cd was mainly caused by economic activities. In addition, the significantly correlated Cu/Ni/Zn and Pb/Cd indicated the inputs from different human activities, while Cr was prone to multiple sources. This study demonstrated that more attention should be given to the contamination by Cr, Pb and Cd, and that the management of human economic activities is vitally imperative for safety of surrounding cropland soils.

Keywords: cropland soils; industrial added value; metal accumulation; soil types

1. Introduction

Soil is a fundamental component of ecosystems and is involved in various human activities, among which, food production is an important function for the survival of mankind [1,2]. However, industrial production and other producing activities usually discharge plenty of pollutants, which would pollute the surrounding cropland [3]. Owing to the rapid industrial and agricultural development, the accumulation of pollutants in agricultural soils in China has become increasingly serious [4,5].

Since the Chinese reform and opening up, economic development in the countryside has greatly increased by governmental effective measures, such as transferring industries from urban areas to the countryside, improving planting technology, increasing employment, etc. [6]. Meanwhile, large quantities of wastewater and pollutants are discharged into surrounding soils and wetlands due to the lack of pollution management and controls in China [7,8]. Industrial sewage, domestic wastewater and agrochemicals contain a great deal of heavy metals, which have the

characteristics of toxicity, abundance and persistence [4,9]. Thus, heavy metal contamination in soils has become a serious problem in China, and even in the world [10]. Heavy metals can be bio-accumulated into vegetation and absorbed by the contactee, and then transferred into human bodies directly or indirectly [11]. Therefore, metal contamination in agricultural soils has threatened human health and should be given more attention [11].

In recent years, cropland soils in many areas of China have been suffering from the contamination of chromium (Cr), copper (Cu), lead (Pb), zinc (Zn), nickel (Ni) and cadmium (Cd) [5,12,13]. For example, Cd and Pb were excessively enriched in cropland soils near the Pearl River Delta, South China [12]. Previous reports showed that distribution and accumulation of heavy metals in agricultural soils of Shunyi, Beijing, Shanghai, Huizhou and many other cities significantly differed [12–16]. However, there are few studies concentrated on the spatial distribution patterns of heavy metals in the agricultural soils of different cities in the same province systematically, which can present the accumulating difference and similarity and contamination status of heavy metals in the areas, and can also provide data references for relevant research to explore the macro trends of metal accumulation among the provinces, China and even the world. Then, municipal management and control measures can be implemented based on the pollution status.

Due to different climates, humidity, etc., the world contains many different soil types, which can hold different contents of heavy metals [17]. For example, yellow brown soil and cinnamon soil show different contents and absorption of Cu, Pb and Cd [18]. Thus, soil types may be an important factor affecting the spatial features of heavy metals, but are rarely studied up to now. Moreover, previous studies indicated that anthropogenic activities most resulted in the metal contamination in cropland soils [12,13]. For example, industrial and agronomic practices, industrial fume, coal burning exhausts and domestic waste reportedly led to various degrees of heavy metal accumulation to the local cropland soils in Northeast China [19], Gan et al. (2019) suggested that human activities contribute 65.8%–86.0% to the metal accumulation in surrounding farmland soils [20]. It is unclear whether human activities can lead to large areas of agricultural soil pollution or not. Economic production dominates the effects of human activities on metal contamination in soils [12,20,21]; thus, the exploration on economic producing and metal contents may indicate their correlations.

Shandong Province is situated in the eastern coast of China. Its total area is 155,800 square kilometers, and the total soil area is 121,100 square kilometers. It has a population of over 100.47 million and governs 16 cities (Figure 1). The topography of Shandong Province is complex and diverse; its eastern area belongs to the Shandong peninsula, the western and northern areas are parts of the North China Plain, and the southern area has various mountains [22]. The Yellow River, Haihe River, Huaihe River and many tributaries are the dominant rivers that flow through Shandong Province from the east to the west and finally discharge into coastal zones. In addition, brown soils, cinnamon soils and fluvo-aquic soils are three dominant soil types which account for 14.09%, 14.66% and 38.53% of the soil areas in Shandong Province, respectively [23,24]. The cultivated rate and agricultural added value in Shandong Province ranked first in China, among which, corn and wheat are the main crops. The gross value of the production of Shandong Province was 7647.0 billion in 2017, 43.99% of which was the added value of secondary industry. Therefore, Shandong Province is a big province for industry and agriculture, which provides an ideal research area to study the spatial distributions and contamination statuses of heavy metals in cropland soils of different cities, and the contributing factors. In addition, the metal accumulation in cropland soils from 2007 to 2017 can also be compared to analyze the metal changes during the decade [5].

Figure 1. The location of sampling sites in the twelve cities of Shandong Province.

Thus, the primary objectives of this study are: (1) to determine spatial variance and contamination status of heavy metals in agricultural soils in cities of Shandong Province, China; (2) to analyze the impacts of soil types on the metal accumulation; (3) to explore the heavy metal changes from 2007 to 2017 and the possible industrial sources to metal contamination. We believe the study on cropland soils of Shandong Province could provide essential information for relevant research.

2. Materials and Methods

2.1. Study Area

Shandong Province has a warm, temperate monsoon climate, and sufficient light resources, with a hot and a rainy season. The precipitation and light is mainly concentrated in summer, and the average annual precipitation is 550–950 mm, decreasing from southeast to northwest. The annual average temperature is 11–14 °C. The average frost-free period is 180 days in northern and eastern areas, and 220 days in the southwest of Shandong Province. Shandong Province has high grain yield, and wheat and corn are grown in summer and autumn, respectively.

2.2. Soil Sampling

Among the 16 cities in Shandong Province, 55 soil samples from cropland soils of Qingdao, Weihai, Yantai, Linyi, Tai'an, Zaozhuang, Weifang, Zibo, Heze, Liaocheng, Dezhou and Jinan were collected in December, 2017 in this study (Figure 1). Surface soils in each site were collected into sealed bags by five-point sampling method in 1 m². The specific sampling sites were the same with the study sites of Zhang et al., and the specific longitude and latitude were listed in Table S1 [5]. In addition, the primary soil types and background values of certain heavy metals in the 16 cities of Shandong Province were listed in Table 1.

Table 1. The background values of the metals in different cities of Shandong Province ($\text{mg}\cdot\text{kg}^{-1}$).

Soil Types	Cu	Pb	Cd	Cr	Ni	Zn	Related Cities
Brown soil	22.4	25.1	0.092	64.5	26.5	68.5	Qingdao, Weihai, Yantai, Dongying, Linyi, Tai'an, Zaozhuang, Weifang, Zibo, Rizhao, Binzhou, Heze, Liaocheng, Dezhou, Jinan, Jining
Cinnamon soil	24.3	21.3	0.1	64.8	30.7	74.1	
Fluvo-aquic soil	22.1	21.9	0.103	66.6	29.6	71.1	

2.3. Soil Analysis

After air-drying at 20 °C, soil samples were ground and then sifted through a 2-mm nylon sieve to get rid of plant debris and gravels. Then, soils were sifted through a 0.149-mm nylon sieve to obtain fine particles. After weighing 0.2000 g soil samples by electronic balance, they were transferred into polytetrafluoroethylene crucibles. Then, crucibles were placed on the electric platen for digestion. With the addition of 10 mL of HNO_3 , 5 mL of HF and 2 mL of HClO_4 , soils were heated in 130 °C for 12 h. Then, we continued heating in 180 °C till the residues in crucibles were less than 1 mL, and residues were collected by dissolving in 1 mL HNO_3 (50%) and transferred into volumetric flasks, with 25 mL ultrapure water [25]. The solutions were filtered into glass tubes and stored at 4 °C.

The elements of Cr, Cu and Zn were examined by inductively coupled plasma atomic emission spectrometry (ICP-AES; IRIS Advantage OPTIMA 7000DV, Thermo Fisher Scientific Incorporated Company, Shanghai, China). Graphite furnace atomic absorption spectrometry (AANALYST800, Perkin Elmer Limited Company, Waltham, MA, USA) was used to determine Cd and Pb. For pH, about 10 g of soils and 25 mL distilled water were mixed, stirred, left for 4 h and determined by the pH meter (Rex PHS-3E, Shanghai INESE Scientific Instrument Limited Company, Shanghai, China).

2.4. Statistical Analysis

Gross product, industrial and agricultural added values of the studied cities were searched from cities' yearbooks; the websites are displayed in Tables S2 and S3. Concentrations of heavy metals were statistical analyzed by using the SPSS (version 21.0, IBM Company, New York, NY, USA, 2012). Essentially, the means, standard errors, and the maximum and minimum values of heavy metals were calculated. For the factors of different cities, soil types and economic indicators, one factor analysis of variance, Pearson correlation analysis and cluster analysis were conducted. We used a Duncan test for post hoc multiple comparisons. The spatial distribution of the sampling sites was generated by using ArcGIS (version 10.2, Environmental Systems Research Institute Incorporated Company, Redlands, CA, USA, 2014) and Adobe Illustrator (version CS6, Adobe Systems Incorporated Company, San Jose, CA, USA, 2012).

3. Results and Discussion

3.1. The Spatial Distribution Patterns of Heavy Metals in the Twelve Cities

The average values of Cu, Pb, Cd, Cr, Ni and Zn were 24.13, 31.77, 0.16, 130.63, 22.13 and 71.19 $\text{mg}\cdot\text{kg}^{-1}$, respectively, suggesting that most cropland soils were suffering from the contamination of Cr, Pb and Cd. Concentrations of Cr ranged from 31.2 to 335.0 $\text{mg}\cdot\text{kg}^{-1}$; that indicated the significantly high spatial disparity in these cities. Contents of Cr in Liaocheng and Dezhou were below the background value, and significantly lower than that in other cities (Figure 2). Moreover, cluster analysis of Cr among the twelve cities showed that three coastal cities (Weifang, Weihai and Yantai) and one inland city (Zibo) were gathered into one cluster, with the significantly high concentrations of

181.13, 177.8, 161.62 and 172.78 $\text{mg}\cdot\text{kg}^{-1}$, respectively (Figures 2 and 3). Similarly, the gross values of industrial output of Weifang, Yantai and Zibo were 230.75, 330.94 and 218.31 billion in 2017, which were also significantly higher than other cities, except Qingdao (tourism dominated the gross product). The higher accumulation of Cr and higher gross value of industrial output in these cities than in other cities may indicate that industrial producing can aggravate the accumulation of Cr in the agricultural soils, and the specific relations need further research. Both Albanese et al. [26] and Lei et al. [27] showed that certain metals can migrate from the inland to the coast, especially Cr and Ni, and in the areas along the flowing rivers. They showed that concentrations of Cr was gradually higher from inland to coast in this study, while the spatial distribution of Ni was not significant (Figure 2). This trend of Cr in this study may attribute to the impact of water flows and the high mobility of Cr [28].

With a slight fluctuation from 8.1 to 40.6 $\text{mg}\cdot\text{kg}^{-1}$, average concentrations of Ni were lower than the background values, indicating the relatively low and stable accumulation of Ni in agricultural soils of the studied cities. Though the contents of Ni in Zibo and Heze (28.65 and 27.8 $\text{mg}\cdot\text{kg}^{-1}$) were approaching the background values, the contamination of Ni was the slightest compared to other metals. Previous reports showed that the contamination of Ni is not serious in cropland, which may attribute to the relative scarcity of Ni in China and the low human input [29,30]. The content of Cu in Weihai (42.87 $\text{mg}\cdot\text{kg}^{-1}$) was significantly higher than in other cities, and was followed by that in Zibo (33.98 $\text{mg}\cdot\text{kg}^{-1}$), suggesting that agricultural soils in the two cities faced high input of Cu. It was deduced that the primary input of Cu was from the industrial activities, as Weihai and Zibo are cities of heavy industry [31,32]. In addition, contents of Cu in Heze and Zaozhuang also exceeded the background values. The mean concentration of Zn was 71.19 $\text{mg}\cdot\text{kg}^{-1}$, and half of the cities showed higher contents of Zn than the background value. Moreover, Weihai and Zibo showed significantly higher accumulation of Zn than other cities, which was similar with the spatial distribution of Cu. The finding further indicated that Zn and Cu had similar industrial input [31].

Figure 2. The spatial distribution patterns and concentrations of heavy metals in cropland soils of the cities in Shandong Province. Bars sharing the same color of each metal are not significant at $\alpha = 0.05$ (Duncan test).

The dendrogram based on the mean Euclidean distance between groups

Figure 3. The cluster analysis of studied cities on the concentration of Cr.

All the studied cities were polluted by Pb and Cd, except for Cd in Jinan and Liaocheng (Figure 2). Though the mean contents of Cd and Pb were $0.162 \text{ mg}\cdot\text{kg}^{-1}$ and $31.77 \text{ mg}\cdot\text{kg}^{-1}$, respectively, the concentrations of Cd in Yantai and Linyi reached $0.8 \text{ mg}\cdot\text{kg}^{-1}$ and $0.6 \text{ mg}\cdot\text{kg}^{-1}$, and the maximal Pb content was up to $73.9 \text{ mg}\cdot\text{kg}^{-1}$. The high standard deviation also suggested that point-source pollution commonly existed in these cities. Deposition of fly ash and irrigation can transfer exhaust gas and industrial waste to surrounding cropland soils directly, which would also lead to the high accumulation of heavy metals [33,34]. Thus, the point-source pollution and the similar spatial distributions of Cd and Pb may also attribute to the industrial activities [35,36]. Cluster analysis to the metals showed that Cu, Ni, Pb and Cd were gathered together, and then clustered the Zn, while Cr had no significant association with other metals, indicating that sources of Cr may be very complex (Figure 4). Shan et al. [37] reported that Cd, Pb and Zn in arable soils were mostly from the human factor, while Sun et al. [19] indicated the lithogenic origin of Cr. The significantly higher contents of Cr in certain cities in this study suggest multiple sources from human activities, not domination by natural sources.

The dendrogram based on the mean Euclidean distance between groups

Figure 4. The cluster analysis of heavy metals among the studied cities.

3.2. The Distribution Pattern and Source Analysis of Heavy Metals in Different Soil Types

Based on the second national soil census in China, brown soils, cinnamon soils and fluvo-aquic soils are the main soil types in Shandong Province [8]. The sampling area in this study involved 26 sites of cinnamon soils, 14 sites of fluvo-aquic soils and 15 sites of brown soils. Previous studies showed that heavy metals had different accumulating features in different soil types, as the soils had different values of organic matter, pH, structural parameter, soil fertility, etc. [5,38,39]. In this study, concentrations of Cr in brown soils and cinnamon soils were $152.06 \text{ mg}\cdot\text{kg}^{-1}$ and $149.38 \text{ mg}\cdot\text{kg}^{-1}$, respectively, which were significantly higher than Cr in fluvo-aquic soils ($72.86 \text{ mg}\cdot\text{kg}^{-1}$; Figure 5 and Table 2). Moreover, the distribution pattern of Cr was the opposite of the pH in the soil types, which was significantly higher in fluvo-aquic soils (8.16) than in brown and cinnamon soils (6.59 & 6.98). Cr is easily migratory and sensitive to pH; thus, the significantly contradictory pattern suggested that the low pH was prone to the migration and bio-accumulation of Cr in the cropland [40,41]. In addition, Yang et al. [39] also suggested that yearly cultivation acidized the farmland, which was more vulnerable to metal accumulation. Furthermore, the higher contents of Cr in brown and cinnamon soils may also be attributed to the different soil fertilities—a soil's ability to supply and coordinate nutrients, moisture, air and heat for biological growth [42,43].

Figure 5. The distribution pattern of heavy metals in the three soil types. Bars sharing the same letter are not significant at $\alpha = 0.05$ (Duncan test).

Table 2. The concentrations of heavy metals in 2007 and 2017 (mg·kg^{−1}) [5].

Soil types (2017)	Cu	Pb	Cd	Cr	Ni	Zn	pH
Brown soil	24.06	32.2	0.38	152.76	20.02	65.66	6.47
Cinnamon soil	26.39	33.21	0.25	150.2	24.12	80.65	6.98
Fluvo-aquic soil	22.36	28.04	0.14	80.64	21.36	68.43	8.10
Soil types (2007)	Cu	Pb	Cd	Cr	Ni	Zn	pH
Brown soil	73.87	99.05	0.08	—	—	85.93	6.70
Cinnamon soil	62.81	130.21	0.32	—	—	143.12	6.78
Fluvo-aquic soil	62.53	140.76	0.26	—	—	141.63	7.82

The concentrations of Cu, Ni, Cd and Pb in the soil types were ordered as: cinnamon soils > brown soils > fluvo-aquic soils. The differences were not significant, indicating that the three soil types had no prominent and differentiated impact on the accumulation of these metals (Figure 5). The Pearson correlation analysis indicated that metals of Cu, Ni and Zn were significantly related, and Pb and Cd were also significantly associated ($p < 0.01$; Table 3). Du et al. [44] showed that discharge of Cu, Zn and Cd were closely correlated with the industrial distribution. Cr, Ni and Pb were from parent sources in Daye City, China, and some reports indicated that Cd and Pb were mostly enriched from industrial effluent, such as coal burning exhausts in Northeast China [19,37]. Thus, the spatial distribution of and contamination by heavy metals greatly differed among cropland areas of China, as did the metal correlations; this was confirmed in previous reports [12,45]. Crustal materials and anthropic pollution were the main sources. The metal contamination in farmland in China is mainly affected by local background values of heavy metals and human activities [45]. In this study, the contamination of Cu, Ni and Zn, and the accumulation of Pb and Cd in the cropland soils in Shandong Province were from different human activities and industrial producing. The contents of Cr were high and the correlation of Cr and other metals was not significant; thus it is suggested that sources of Cr may be various and complex. Thus, regulation and management on the discharge of industrial waste and on the agricultural practices need to be improved.

Table 3. The Pearson correlation analysis of heavy metals and economic indicators.

Parameters	Cr	Ni	Cu	Zn	Cd	Pb
Cr	1					
Ni	−0.005	1				
Cu	0.104	0.313 *	1			
Zn	0.280 *	0.292 *	0.530 **	1		
Cd	0.058	0.320 *	0.033	0.248	1	
Pb	0.099	0.226	−0.016	0.165	0.788 **	1
Gross product	0.15	−0.055	−0.114	−0.284 *	−0.01	0.034
The primary industrial added value	0.048	−0.199	−0.187	−0.307 *	0.001	−0.08
The secondary industrial added value	0.035	0.007	−0.08	−0.242	−0.024	0.032
The industrial added value	0.179	0.021	0.305	−0.187	0.520 *	0.432
The tertiary industrial added value	0.144	0.009	−0.018	−0.155	0.064	0.092

*: significance on levels of $p < 0.05$; **: significance on levels of $p < 0.01$.

3.3. The Decadal Changes of Heavy Metals and Relations with Industrial Producing

The mean value of pH in cinnamon soil and fluvo-aquic soil in 2017 was relatively higher than that in 2007, which suggested that yearly farming changed soil physical feature and grain yield, as pH was significantly and positively correlated with crop production [46]. The concentrations of Cu, Pb,

Zn and Cd in 2017 were significantly decreased with respect to the accumulation in 2007, except for the insignificant change of Cd in brown soils (Table 2); the changes of Cd may attributable to the continuous input, leaching, migration and accumulation procedure of Cd in agricultural soils [19,44]. The significant reduction of above-mentioned metals from 2007 to 2017 suggests decreased human input, such as by decreasing the utilization of agrochemicals and the discharge of industrial waste and increasing the metal remediation and soil management [47,48]. Therefore, environmental management and ecological protection is vitally imperative for heavy metal remediation in agricultural soils.

The development of industry, agriculture, tourism and services brings about rapid growth of economy and living standards [49,50]. Meanwhile, the damage to ecological environment is also profound and lasting, which includes soil contamination around the industries, reduction of grain yield, destruction of natural habitat, etc. [51,52]. Thus, the relationship between economic indicators and heavy metals in agricultural soils should be researched. Factor analysis of heavy metals and economic indicators indicated three dominant factors, which explained 38.04%, 22.54% and 20.97% of the total factors, respectively (Table 4). The first factor mainly explained the Cd, Pb and all economic indicators, indicating the close relation among Cd, Pb and economic production. Factor 2 explained Cr, Ni, Zn and Cu, which may suggest their common sources. Factor three explained the Cr and primary industrial added value, which indicated a certain industrial input. The remarkable impacts of economic production on Cd and Pb are in accordance with many previous studies [53,54]. In addition, concentrations of heavy metals presented certain relations with some economic indicators. Specifically, the primary industrial added value showed a significant and negative correlation with concentration of Zn, while industrial added values indicated a significant and positive relationship with the contents of Cd (Table 3). That may suggest that industrial production aggravated the accumulation of Cd, which can also be confirmed by the high accumulation of Cd in this study [36,37]. The closely related Zn and the primary industrial added value may attributable to the bio-accumulation, as primary industry was mainly involved in biological yield [55]. With the crop growth, Bhatti et al. [56] showed high bioaccumulation of Zn in the crops, while Xu et al. [37] indicated the high enrichment of Cd in the soils. Thus, the high primary industrial added value simultaneously boosted the bio-accumulation of Zn into aboveground vegetation and the enrichment of Cd into rhizosphere soils [57,58].

Table 4. Factor analysis of heavy metals and economic indicators.

Parameters	1 (38.04%)	2 (22.54%)	3 (20.97%)
Cr	0.497	0.67	0.441
Ni	0.185	0.569	−0.47
Cu	0.606	0.7	−0.053
Zn	0.087	0.826	0.384
Cd	0.801	0.03	−0.358
Pb	0.681	0.08	−0.565
The primary industrial added value	0.615	−0.341	0.508
The secondary industrial added value	0.885	−0.349	0.098
The industrial added value	0.863	−0.36	0.085
The tertiary industrial added value	0.913	−0.202	0.071

4. Conclusions

This study presented the spatial distribution patterns and contamination statuses of heavy metals in cropland soils of the twelve cities in Shandong Province, North China. Cropland soils of most studied cities are suffering from the contamination of Cr, Pb and Cd. Specifically, Pb and Cd had similar spatial distributions in those cities; the distribution of Cr may be affected by flowing orientation of basins. Concentrations of Zn and Cu were relatively high in Zibo and Weihai, whereas Weifang, Weihai, Yantai, and Zibo showed significantly high accumulation of Cr. Moreover, contents of Cr in brown soils and cinnamon soils were significantly higher than in fluvo-aquic soils, while other metals showed no significant difference. This study also indicated that accumulation of Pb and Cd

was mainly caused by economic activities, while Cr was prone to multi-sources. Contents of the metals were remarkably decreased with respect to their contents in 2007, except for Cd in brown soils. In conclusion, this study is significant to agricultural management on heavy metals contamination, and waste management and technology improvement are imperative to the cities that suffering from serious metal pollution.

Supplementary Materials: The following are available online at <http://www.mdpi.com/2076-3417/10/6/1963/s1>. Table S1: The longitude and latitude of the total sampling sites in Shandong Province. Table S2: The websites of cities' yearbooks in the statistical bureau. Table S3: The retrieved economic indicators from cities' yearbooks during 2017 (billion).

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