

Spatial trends and pollution assessment for mercury in the surface soils of the Nansi Lake catchment, China

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Abstract Surface soil samples collected from Nansi Lake catchment were analyzed for mercury (Hg) to determine its spatial trends and environmental impacts. Results showed that the average soil Hg contents were 0.043 mg kg^{-1} . A positive correlation was shown between TOC and soil Hg contents. The main type of soil with higher TOC contents and lower pH values showed higher soil Hg contents. Soil TOC contents and CV values were both higher in the eastern catchment. The eastern part of the catchment, where the industry is developed, had relatively high soil Hg contents and a banding distribution of high Hg contents was corresponded with the southwest-northeast economic belt. Urban soils had higher Hg contents than rural soils. The urbanization pattern that soil Hg contents presented a decreasing trend from city center to suburb was shown clearly especially in the three cities. Soil Hg contents in Jining City showed a good consistency with the urban land expansion. The spatial trends of soil Hg contents in the catchment indicated that the type and the intensity of human activities have a strong influence on the distribution of Hg in soils. Calculated risk indices showed that the western part of the catchment presented moderately polluted condition and the

eastern part of the catchment showed moderate to strong pollution level. The area with high ecological risk appeared mainly along the economic belt.

Keywords Mercury · Spatial trend · Anthropogenic disturbance · Environmental impact · Surface soils · Nansi Lake catchment

Introduction

Mercury (Hg) is toxic to humans (NAS 1978). Since industrialization, large amounts of Hg have been released to the environment by human activities such as operations of mining, fuel combustion, industrial discharges, and application of chemical fertilizers (Feng et al. 2006). Accumulation of Hg that came from local pollution sources had a significant impact on local environment (Feng and Qiu 2008). In the transfer of Hg in different environment medium, soils are the most important sinks of Hg in terrestrial ecosystems (Asubiojo et al. 1991). When deposited in soils, chemical and biological transformation processes could happen and many factors such as organic matter, pH, grain size, and the presence of other elements in soils can influence the speciation, distribution, accumulation, and bioavailability of Hg (Chakraborty et al. 2015). China's Hg emission to the atmosphere increased quickly since the beginning of the reform and opening up in the late 1970s and total Hg content in soils shows an obvious urbanization pattern (Fang et al. 2004). Levels of Hg pollution in urban soils can be variable depending on different urban land-use types, the intensity of human activities, pollution history, distance to emission sources, and other factors (Zhao and Li 2013). Urban soil contamination by Hg in China has become increasingly a serious problem (Streets et al. 2005).

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Increased industry dramatically enhances local economic development which may lead to an excessive discharge of contaminants to the environment. Industrial wasted water, gas, and residue containing Hg can enter atmosphere and soils directly. In addition, municipal refuses and agricultural irrigation can make Hg into water and soils. Since the 1980s, Hg content in surface sediments of the Nansi Lake has grown quickly with the rapid development of regional economy (Yang et al. 2003) which indicates that Hg accumulation in the catchment were attached to urbanization effect. Hg accumulation in urban soils, streams, and lake in the catchment has become a widely concerned problem and has been attracting extensive attention by local managers.

The main goals of this study were the following: (1) to describe Hg content in the surface soils quantitatively and to explore the spatial distribution of Hg, (2) to identify the influences and sources of Hg distribution, (3) to evaluate the level of soil Hg contamination in the catchment.

Materials and methods

Study area

The Nansi Lake catchment ($34^{\circ} 26' - 35^{\circ} 57' N$, $114^{\circ} 45' - 117^{\circ} 49' E$) is situated southwest of Shandong with an area of $27.9 \times 10^3 \text{ km}^2$. Three cities and 19 counties in the catchment are shown in Fig. 1. To the eastern part of

the catchment is mainly an alluvial plain, with a slope of $1/1000 \sim 1/10000$, and major inflow rivers, such as the Sihe River and Guangfu River which originate in the eastern mountain, flow into the lake from east to west. The main types of soil in the east of the catchment are cinnamon soil and brunisolic soil. To the western part of the catchment is the Yellow River Floodplain, with a slope of $1/5000 \sim 1/20000$, and main inflow rivers, such as the Zhuzhaoxin River and Dongyu River which originate in the southern margin of the Yellow River levee, flow into the lake from west to east. The main type of soil on the west of the catchment is fluvo-aquic soil.

Agriculture is developed in the western part of the catchment. Industry is developed in the eastern part of the catchment where large enterprises such as Jining power plant (located in Jining City), Shanshui cement group (located in Jining City), Yancon group (located in Zoucheng County), Zouxian power plant (located in Zoucheng County), Shi Li-quan Power Plant (located in Zaozhuang City), and Zaozhuang mining group (located in Zaozhuang City) are mainly concentrated. The economic belt including Jining City, Yanzhou County, and Qufu County is the dominant developing area in the eastern part of the catchment.

Sampling

The Nansi Lake catchment was divided into square sampling grids with sides of 1 km, and then four randomly collected samples at each site were mixed to form a composite sample

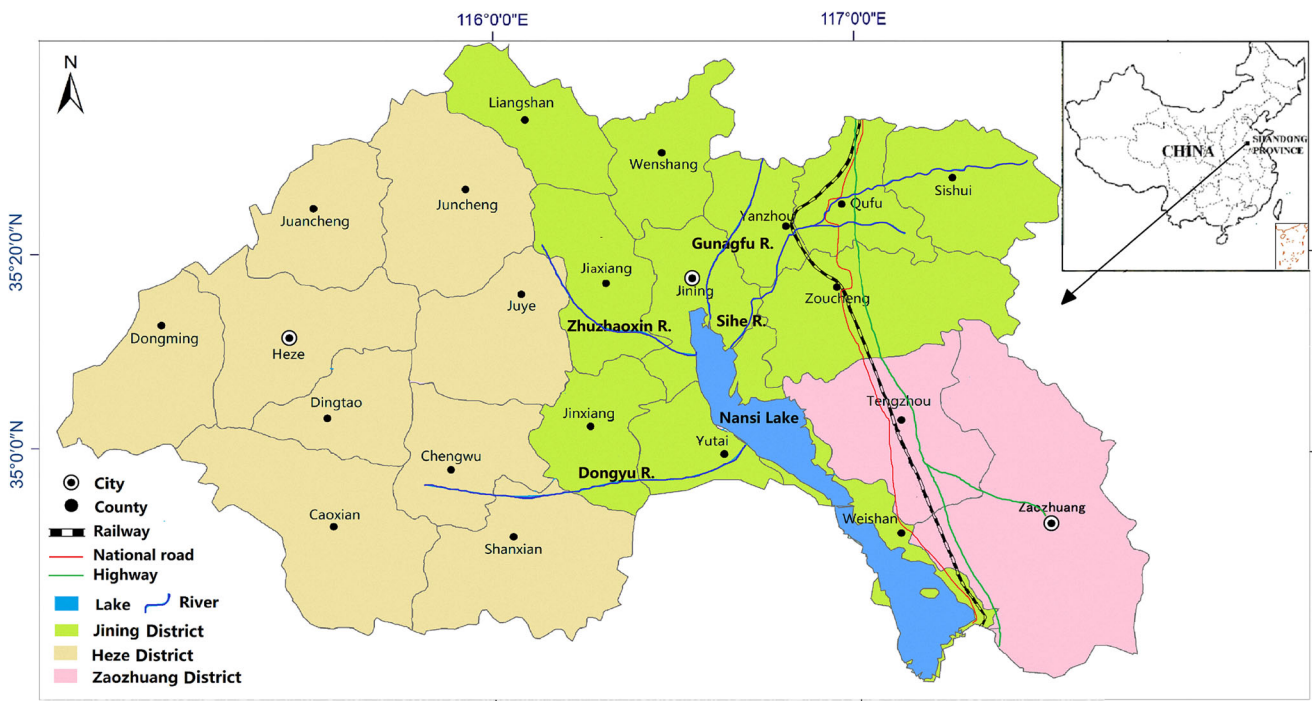


Fig. 1 Study area in Shandong Province, China

(Liang et al. 2017). All total 5000 samples of surface soil in the catchment (Fig. 1) were collected from 0- to 20-cm depths in May 2012. All samples were collected using a luoyang shovel, sealed in polyethylene bags and transported to the laboratory. The quantity of the original samples was more than 1 kg. All samples were air-dried at room temperature, sieved through a 2-mm polyethylene sieve after removing gravel and bricks or other debris and stocked for further analyses.

Chemical analysis

Total Hg content analysis was completed in the lab of the Wuhan Inspection and Testing Center of Geology and Mineral Resources, which is a subordinate research institute of the Ministry of Land and Resources of China. About 0.25 g samples were digested in Teflon vessels with a HNO₃–HClO₄ mixture in a microwave oven (Li et al. 2010). The sample solutions were filtered and adjusted to 25 mL with double deionized water. The total concentrations of Hg were analyzed by an atomic fluorescence spectrometer (AFS, AFS-230E, Beijing, China). Al₂O₃ and Fe₂O₃ were measured by Graphite Furnace Atomic Absorption Spectrometry (GF-AAS, M6, Thermo Elemental, USA), TOC were measured by potassium dichromate oxidation spectrophotometry and the pH were measured by potentiometry. The measurements of Hg, pH, and TOC were all in accordance with the geological survey technical standards of the China Geological Survey.

To ensure experimental accuracy, the system blank was measured before the experiment to minimize the impact of experimental apparatus on element contents. Parallel samples were also added to reflect the precision of the test.

Assessment methods

The background values play an important part in assessing pollution of trace elements. The catchment is located in southwest of Shandong province; for this reason, the soil background values in Shandong province (0.019 mg/kg) were selected as the background values of soil Hg in the catchment (Kong et al. 2013).

Geo-accumulation index (I_{geo})

The geo-accumulation index (I_{geo}), which was developed by Müller (1981), is calculated as follows:

$$I_{geo} = (\log_2 C_n / 1.5 B_n) \tag{1}$$

where C_n is the content of the studied soil samples and B_n is the background content of a given metal. The geo-accumulation index was divided into seven levels by Forstner et al. (1993). I_{geo} ≤ 0, background concentration; 0 < I_{geo} < 1, unpolluted to moderately polluted; 1 ≤ I_{geo} < 2,

moderately polluted; 2 ≤ I_{geo} < 3, moderate to high pollution; 3 ≤ I_{geo} < 4, heavily polluted; 4 ≤ I_{geo} < 5, heavily to extremely polluted; 5 ≤ I_{geo}, extremely polluted.

Potential ecological risk index

The potential ecological risk index, proposed by Hakanson (1980), was applied to assess the ecological risk caused by heavy metals. RI is defined by the following equation:

$$RI = \sum E_r^i = \sum T_r^i (C_s^i / C_n^i) \tag{2}$$

where E_rⁱ is the potential ecological risk index of an individual element; RI describes the potential ecological risk for all heavy metals. T_rⁱ is the toxic-response factor and values for Hg are 40 (Xu et al. 2008). C_sⁱ is the concentration of analyzed metals and C_nⁱ is the reference values of heavy metals. Grading standards of potential ecological risk for heavy metals are divided into five levels by Hilton et al. (1985). E_rⁱ < 40, low ecological risk; 40 ≤ E_rⁱ < 80, moderate ecological risk; 80 ≤ E_rⁱ < 160, considerable ecological risk; 160 ≤ E_rⁱ < 320, high ecological risk; 320 ≤ E_rⁱ, very high ecological risk.

Results and discussion

Soil properties

Soil parent material has a close relationship with Hg contents in surface soils. Basic statistics of soil properties in the catchment are shown in Table 1. Contents of Al₂O₃, Fe₂O₃, and SiO₂ can reflect soil texture. SiO₂ was an important part of the coarse particles in soils. There is a negative correlation between the SiO₂ and soil Hg contents (P < 0.05, R² = 0.65) in the catchment. Compared with the results of multi-target regional geochemical survey in the downstream area of the Yellow River, relatively low SiO₂ and high Al₂O₃, Fe₂O₃ content reflect that the soil texture is biased toward clay. Clay has a relatively strong adsorption capacity for heavy metals. Soil

Table 1 Basic statistics of soil properties

	Max	Min	Average	CV (%)	Average contents in the downstream area of the Yellow River ^a
pH	9.08	4.62	7.75	9.51	–
TOC (%)	11.43	0.15	0.93	64.03	–
SiO ₂ (%)	73.98	25.77	60.22	8.96	61.26
Al ₂ O ₃ (%)	20.78	6.59	12.96	9.80	11.95
TFe ₂ O ₃ (%)	11.6118	2.36	4.66	20.22	4.20

–: not available, CV coefficient of variance

^a Sun et al. (2008)

was slightly alkaline and soil pH had a small fluctuation with low CV values. Soil pH has a close relationship with the stability of Hg and influences the adsorption of organic matter to Hg. For example, one consequence of the increase in dissolved soil organic matter caused by the increase of pH is a corresponding decrease in organic-based surface adsorption sites, which may contribute to the decrease in Hg (II) adsorption (Yin et al. 1996). The main type of soil in the east of the catchment is cinnamon soil, and the main type of soil on the west of the catchment is fluvo-aquic soil. Cinnamon soils had a higher average TOC content and a lower average pH value than fluvo-aquic soils. When compared soil Hg contents in the two types of soils, Hg contents in cinnamon soils (0.065 mg/kg) were slightly higher than that in fluvo-aquic soils (0.036 mg/kg). There is a positive correlation between the TOC and soil Hg contents ($P < 0.01$, $R^2 = 0.72$) in the catchment. The average TOC content was 1.07% in the eastern catchment and was 0.73% in the western catchment. In addition, CV values of TOC were 69% in the eastern and were much higher than that in the western catchment (19%). Soil organic matter has a strong adsorption effect on Hg, so Hg can accumulate easily in surface soils and remain in the top soil layer. As the important metal complexes in soils, the complexation of Hg with TOC may influence Hg contents in soils and their migration behavior (Chakraborty and Babu 2015). Hg contents in surface soils reflect the combined effect of atmospheric deposition for a long history. Density of human activities such as discarding organic microbial products and discharging industrial waste and sewage may lead to the wide range of TOC content and then influence the Hg distribution in surface soils (Liu et al. 2010).

Spatial distribution of Hg in the catchment

The spatial distribution of Hg in the catchment was carried out in ArcGIS 10.0. The distribution of soil Hg contents had a large spatial difference (Fig. 2). Soil Hg contents in the catchment were in the range 0.005–2.27 mg/kg (Table 2). Average soil Hg contents were 0.043 mg kg⁻¹ and were approximately 2.3-fold higher than the soil element background values in Shandong province but lower than Chinese soil element background values (Wei et al. 1991). Variation coefficient (CV) values of Hg were higher than 1. The CV of elements affected by human activities is higher than that from natural sources (Han et al. 2006). When comparing the CV values of Hg in surface soils (> 100%) with the values in surface sediments (31%) from the Nansi Lake (Wang et al. 2014), concentrations of Hg had a higher fluctuation than those in surface sediments, which is a result of a large amount of samples covering broad ranges in surface soils. Additionally, urban soils could be more directly affected by human activities and have a more rapid accumulation rate for pollutants.

Statistics indicated that the average soil Hg content in the eastern part of the catchment was 0.047 mg/kg, and the average soil Hg content in the western catchment was 0.036 mg/kg. Counties in the eastern part of the catchment generally had higher average soil Hg contents than that in the western part of the catchment. The eastern catchment, where the economy is more developed, was rich in coal and had a relatively large population density. The western catchment where its economy dominated by agriculture are mostly flat lands and are less-developed areas. The difference of soil Hg contents between the eastern and western part of the catchment indicated that the level of economic development had a significant impact on soil Hg contents.

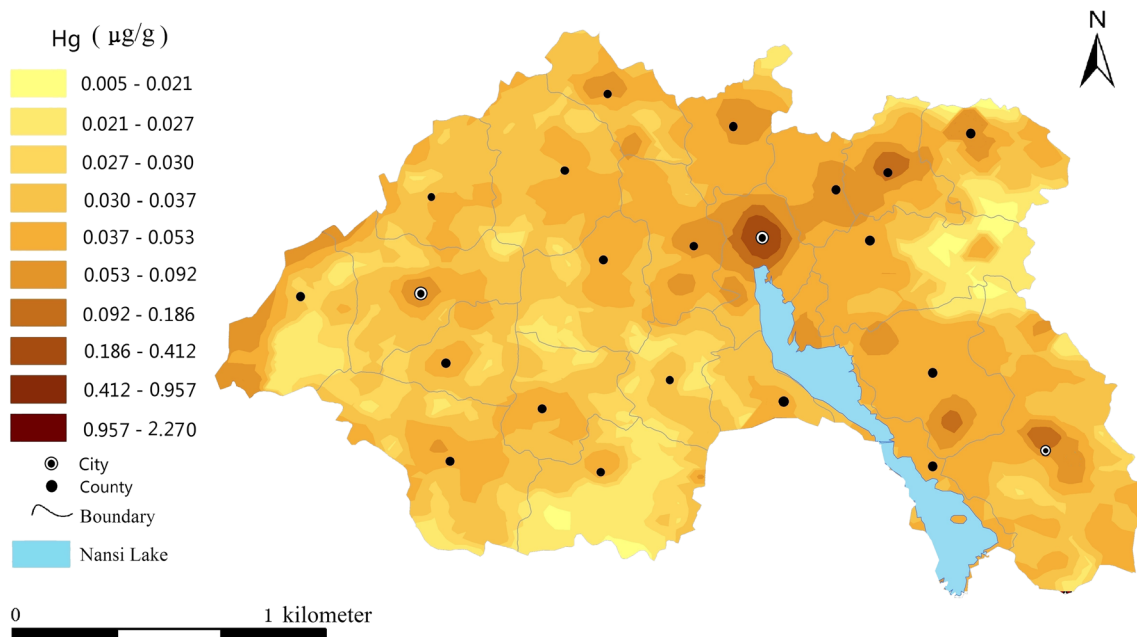


Fig. 2 The distribution of mercury in surface soils of the Nansi Lake catchment

Table 2 Basic statistics and mercury contents in surface soils

	Max	Min	Average	CV (%)
Jining City (mg·kg ⁻¹)	1.360	0.019	0.092	> 100%
Zaozhuang City (mg·kg ⁻¹)	0.645	0.009	0.039	> 100%
Heze City (mg·kg ⁻¹)	0.230	0.010	0.036	> 100%
Eastern catchment (mg·kg ⁻¹)	2.270	0.005	0.047	> 100%
Western catchment (mg·kg ⁻¹)	0.311	0.006	0.036	> 100%

CV coefficient of variance

In the eastern part of the catchment, a banding distribution of relatively high Hg contents was corresponded with the southwest-northeast economic belt including Jining City, Yanzhou County, and Qufu County. Statistics indicated that compared with other counties in the catchment, Yanzhou County and Qufu County had the highest average soil Hg contents (0.055 mg/kg). The economic belt was located in the most economic developed area of the catchment. The rapid growth of GDP after the 1990s in the area was closed linked to the development of the coal mining industry. Coal distribution area accounted for 35% of the land area of Jining City and coal mines were widely distributed in Yanzhou County and Qufu County. It has been reported that Hg contents in the surface sediments of heavily polluted Guangfu River and Sihe River, which flowed through the economic belt, were 0.111 and 0.173 mg/kg (Kong et al. 2013), respectively, and were both higher than that in the surface sediments of the northern part of Nansi Lake. Emission of industrial sewage is a major source of pollution for these heavily polluted rivers.

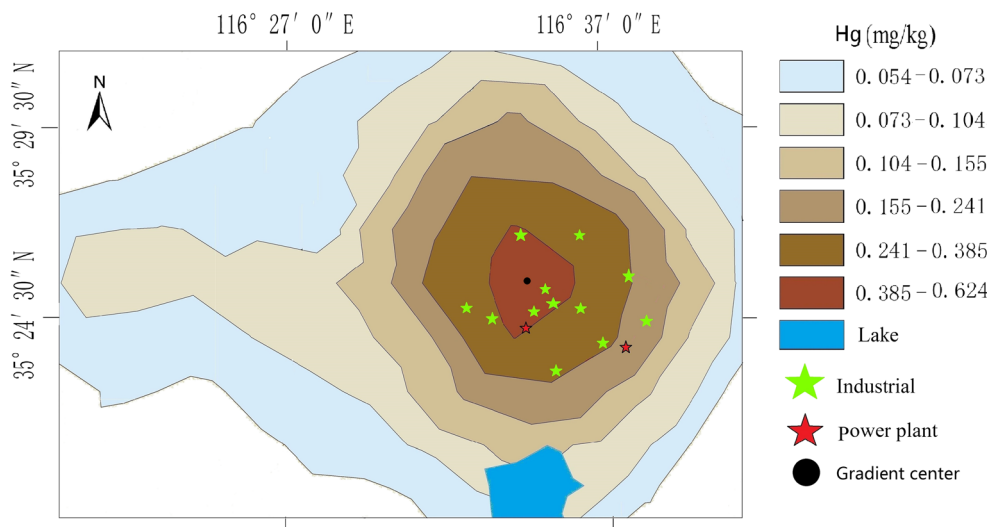
In addition, relatively low soil Hg contents in the eastern part of the catchment were shown in the east of Zoucheng County. Eastern area of Zoucheng County is a mountainous and hilly area with a low population density. The area with high altitude, large slope, and low vegetation coverage is the

most serious area suffering from soil and water loss in the catchment. Topsoil losses were probably the main reason for the low Hg content in soils. However, the western area of Zoucheng County had relatively high soil Hg contents compared with the eastern part. Beijing-Shanghai Railway, 104 National Road, and Beijing-Fuzhou Highway run through the western part of Zoucheng County and Tengzhou County. As the important coal development base of China, the area with convenient transportation was densely populated and had many township enterprises. There are many large coal mines, thermal power plants, and coal washery concentrated in the area. The large difference of soil Hg contents between the western and eastern area of Zoucheng County indicated that civil coal burning and coal mining were the major causes of soil Hg accumulation in the western part of Zoucheng County and Tengzhou County.

In the western part of the catchment, the distribution of soil Hg contents was non-uniform. Generally low soil Hg contents were widely distributed in most rural areas and relatively high soil Hg contents were shown in the county areas especially in Dongming County which had the highest average soil Hg contents in the western part of the catchment. The urban-rural difference revealed an effect of urbanization on soil Hg contents. Dongming County is rich in crude oil and natural gas deposits and the petrochemical industry was developed quickly especially in the southwestern part of the county. In the process of developing oil and gas fields, Hg can be discharged into atmosphere or directly into surrounding soils. It can be the possible reason for high soil Hg contents in the southwestern Dongming County.

Urban soils had higher Hg contents than rural soils in the catchment. This pattern was shown especially in Jining, Heze, and Zaozhuang City and was presented by an urbanization gradient that soil Hg contents showed a decreasing trend from city center to suburb. The accumulation and distribution of Hg

Fig. 3 Gradient change of mercury contents in surface soils of Jining City



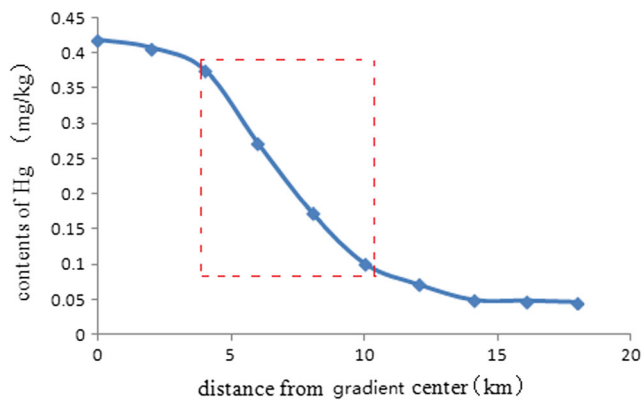


Fig. 4 Fluctuation curve of soil mercury contents with distance from gradient center

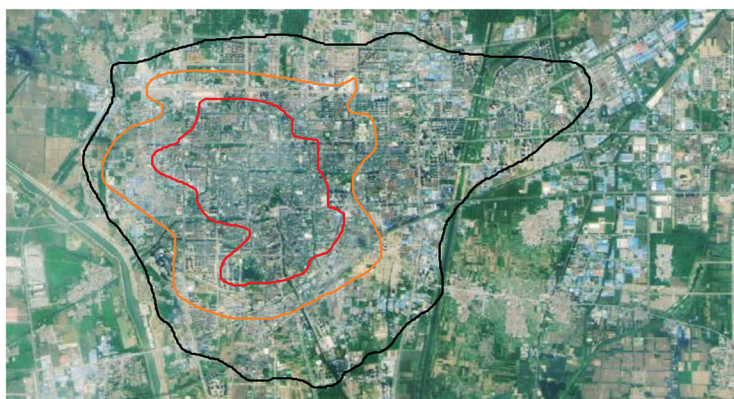
in urban soils were affected by comprehensive factors. The major cause of soil Hg accumulation in different area can be various. It has been proven that pulverized coal ash from power industry and coal-burning boilers and waste released from industrial production can make Hg enter into soils directly (Hu and Cheng 2016). In addition, coal combustion in low capacity domestic heating units should be rather considered as a Hg source of local range impact compared with industrial boilers (Hlawiczka et al. 2003). Most of Hg from coal burning was released to atmosphere and then enter into soils by atmospheric deposition. In urban areas, industrial coal-burning boilers and civil coal-burning boilers are concentrated which is the reason of much accumulation of Hg in urban soils than in rural soils.

Highest soil Hg contents were shown in Jining City which had the highest average values 0.092 mg/kg in the catchment and average soil Hg contents in Jining City were much higher than that in Zaozhuang City and Heze City. Jining City is located at the intersection of the canal development belt and the southwest-northeast economic belt with convenient transportation. It is the economic center of the catchment with the population density 1280 people per square mile and it has a long history. An obvious gradient change of soil Hg contents were shown in the center of Jining City.

Soil Hg distribution changing with an urbanization gradient in Jining City is shown in Fig. 3. Each annular zone from center to the most outer has an area of 14.5, 62.1, 69.3, 75.1, and 165.3 km². Average soil Hg contents in the central zone were 0.4 mg/kg and were approximately 21 times higher than the background values. Average soil Hg contents in the most outer annular zone were 0.08 mg/kg and were approximately four times higher than the background values. Large industries such as chemical plants, chlor-alkali plants, boiler factories, and machinery factories were distributed in a 4-km radius of the gradient center. Highest soil Hg contents were shown in the range of 4 km from the gradient center in Jining City. The area with dense industry and population was an old urban district. Combustion of civil coal especially for home heating and cooking is the major cause of Hg accumulation in the area.

The fluctuation curve of soil Hg contents with distance from the gradient center had two inflection points at the distance of 4 and 10 km (Fig. 4). The rate of decline in Hg contents was slow in the range of 4 km from the gradient center. In the range of 4 to 10 km, Hg contents in surface soils dropped quickly and had a significant negative correlation ($R = -0.995$) with the distance. Per 1 km increase in distance was associated with a 0.05 mg/kg drop in Hg contents in this range. The decreasing rate of soil Hg contents was slow in the range of 10 to 14 km and soil Hg contents remained stable beyond the distance of 14 km from the gradient center.

Soil Hg contents in Jining City have a close relationship with regional economic development. Jining's GDP increased quickly since 1987 and annual coal yield increased from 928 million tons to 8809 million tons between 1978 and 2010. It has been reported that Hg contents in surface sediment of the Nansi Lake increased rapidly since 1981 because of the economic recovery and the increasing demand for energy (Yang et al. 2003). Moreover, the change of Hg contents in lake sediment of the catchment was accorded with the local economic condition. In addition, accelerated urbanization since the 1980s had a significant impact on the accumulation of Hg in soil.



- The urban land scale of 1987
- Expansion range from 1987 to 2000
- Expansion range from 2000 to 2010

Fig. 5 Urban land expansion of Jining City

Table 3 Evaluation of mercury pollution in the surface soils of the Nansi Lake catchment

	Average of I_{geo}	Pollution level	Average of E_r	Ecological risk
Jining City	1.1	Moderate pollution	193.9	High risk
Zaozhuang City	0.3	None to moderate pollution	79.9	Moderate risk
Heze City	0.2	None to moderate pollution	76.4	Moderate risk
Eastern catchment	0.4	None to moderate pollution	100.3	Considerable risk
Western catchment	0.2	None to moderate pollution	76.9	Moderate risk
Catchment	0.3	None to moderate pollution	89.8	Considerable risk

The urban land expansion of Jining City from 1987 to 2010 is shown in Fig. 5. Urban construction after the 1990s formed a structure of circle layer of one old urban district and decreasing progressively to the periphery region and urban extension, which was limited by the Beijing-Hangzhou Canal and the unexploited coal zone, mainly expanded eastward. The distribution of soil Hg contents showed a good consistency with the urban land expansion. Highest soil Hg contents were shown in the old urban district and considerable higher soil Hg contents were shown in its urban area than in its suburban area with a suburban-urban gradient. In addition, the banding distribution of relatively high Hg contents which was corresponded with the economic belt was found stretching along the expansion of urban land in Jining City. The consistency between soil Hg distribution and urban land extension indicated that the distribution of soil Hg contents was heavily influenced by human activities. Large accumulation of soil Hg in the old urban district was a result of rapid urbanization with the fast economic development and a long history of domestic coal uses. Despite the fact that factories were relocated and farming lands were converted to residential or commercial uses, high soil Hg contents in the earlier developed area may reflect the legacy left behind since the 1980s.

Pollution assessment

Metal pollution assessment

I_{geo} values are shown in Table 3. Mean I_{geo} values of Hg were 0.3, specifying an unpolluted to a moderately polluted condition. Statistics indicated that 34.9% of all soil samples had I_{geo} values less than 0 and 53.5% of all soil samples had I_{geo} values between 0 and 1. As shown in Fig. 6, the western part of the catchment presented moderately polluted condition (I_{geo} between 1 and 2) and the eastern part of the catchment showed moderate to strong pollution level (I_{geo} between 2 and 3). The strongly polluted areas (I_{geo} between 3 and 4) for Hg were observed near the northern margin of the northern sublake especially the Jining City and in Qufu County, Wenshang County, and Dongming County. These areas were needed to focus on and preferentially treated.

Potential ecological risk

Statistics of E_r for Hg based on Eq. (2) are presented in Table 3. Mean E_r values of Hg were 89.8, specifying a considerable risk

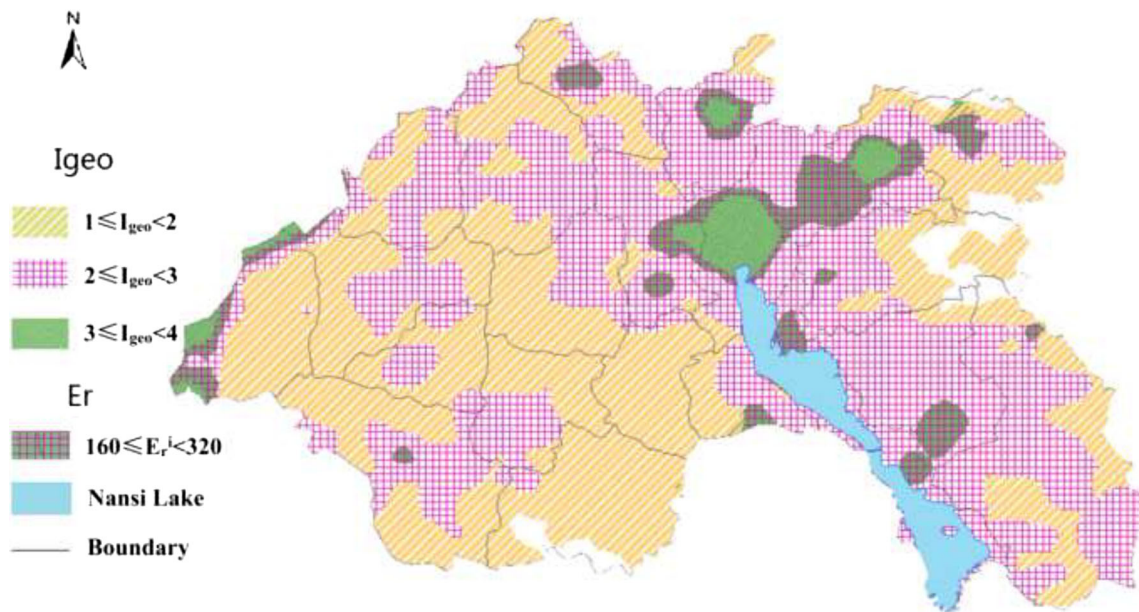


Fig. 6 The distribution of I_{geo} between 1 and 4 and E_r between 160 and 320 for mercury

condition. Statistics indicated that 59.5% of the sampling sites presented a moderate ecological risk, and 29.2% of the sites showed a considerable risk for Hg. As shown in Fig. 6, areas with high ecological risk (E_r , between 160 and 320) appeared mainly along the main economic belt in Jining City and in south of Tengzhou County and west of Dongming County, which indicated that the biodiversity of Hg in these areas was high and Hg could easily be released from soils to have bad influence on the environment and humans.

Conclusion

The average soil Hg contents of the Nansi Lake catchment were 0.043 mg/kg and were 2.3-fold higher than their background values. The eastern part of the catchment had higher soil Hg contents than the western part of the catchment. Urban soils had higher Hg contents than rural soils in the catchment. This pattern was shown especially in Jining, Heze, and Zaozhuang City. Highest soil Hg contents were shown in Jining City. Relatively high soil Hg contents were shown in Yanzhou County, Qufu County, and Tengzhou County. The reason of the Hg distribution characteristics in Nansi Lake catchment is because of the regional economic development and regional urbanization. Civil coal burning and industrial coal combustion were the major causes of soil Hg accumulation in the catchment. Pollution assessment results showed that the eastern part of the catchment showed strong pollution level and areas around the economic belt in the eastern part of the catchment had high ecological risk.

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