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Risk assessment and source apportionment of heavy metals pollution from atmospheric deposition in Nanjing, China

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ABSTRACT

Heavy metals (HMs) are the toxic pollutants in urban environment, and their sources are complex. Dust might be a good carrier of HMs into ecosystem and human. In this study, 48 dust samples were collected in Nanjing, an industrial city and transportation hub in the Yangtze River Delta, China. The concentrations, spatial distribution, sources and risks of heavy metals (Cr, Ni, Cu, Zn, Cd and Pb) in dust were determined and analyzed. The results showed that although the health risks of some HMs had decreased, Cr, Zn, and Cd had high concentrations and high risks on ecosystem/human. And thus, the total risks of the target HMs were higher than the threshold of non-risks. Especially, children may face the highest possible risks due to the frequent hand-oral ingestion when children play. The hot spot regions of the HMs were mainly in the industrial district in the north, urban, and rural region in south, relating with the industrial, traffic, and agricultural sources, respectively. The analysis for the risks of individual sources further confirmed these sources should be further controlled. The results in this study will provide information on the priority of HMs' monitoring and source management.

1. Introduction

Dust is the large size solid particles with short-range transport in air. Due to its weak mobility, dust is often used as the indicator of local air pollution [1,2]. In addition, dust is also a carrier of pollutants, among which heavy metal pollution is particularly prominent [3]. Heavy metals (HMs) are absorbed mostly on the surface of dust particles, and then deposit to the surface ground via dry and wet deposition, causing ecological risks [4]. Moreover, direct exposure on skin and possible weak inhalation of the heavy metals carried by dust could cause the risks of human health [5,6]. These risks of heavy metals were focused widely. Many studies reported that the atmospheric dusts can reveal that the mega-cities in China were polluted heavily by heavy metals due to industrial and traffic emissions [7,8]; [9]). Similar results were also reported in the cities of developing countries (e.g. Kermansha, Iran, [10]).

As one of the major cities in the Yangtze River Delta region, Nanjing has developed economy and large-scale heavy industry such as petrochemical industry and iron and steel smelting. In recent years, many scholars have studied the characteristics of heavy metal pollution in Nanjing, but most of them focus on the heavy metal pollution in soil or farmland in local areas, and the research on heavy

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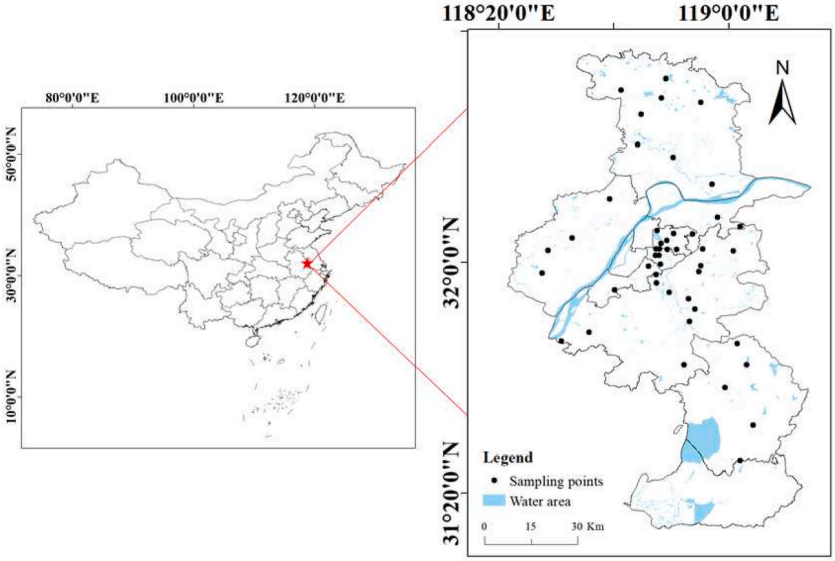

Fig. 1. Sampling sites of atmospheric dust in Nanjing.

metal pollution in atmospheric dust fall is relatively lacking [11–13]. In this study, the content characteristics, spatial distribution characteristics, pollution sources and contribution rates of heavy metals in atmospheric dust fall in Nanjing, as well as the ecological risks and health risks of different pollution sources were discussed, and an assessment system was established to provide theoretical support for the prevention and control of heavy metal pollution in the city.

In this paper, the concentrations and spatial distribution of HMs in dust in Nanjing were observed, and the risks of HMs in dust on ecosystem and human were assessed. Absolute principal component-multiple linear regression (APCS-MLR) model was used to analyze the sources and their contribution on HMs' pollution.

2. Materials and methods

2.1. Study area

Nanjing (31.23–32.62 N, 118.36–119.24 E) is one of the main cities in the Yangtze River Delta region, East China, and is also an important traffic and industrial hub. The most widely distributed representative soils in Nanjing are yellow brown soil, yellow sols and lime-tide soil. Huanggang soil was developed in the Pleistocene Xiashu clay, not only the parent material source is single, but also the texture is uniform. The lime-tidal soils are all developed in the modern alluvial of the Yangtze River, the parent material is the same, but the texture is different. The parent material of yellow brown soil, however, includes all kinds of rock weathering with very different ages and properties [14]. The heavy industry of Nanjing is mainly located in the north of the city, including chemical/steel/power plants. As a typical industrial city, the air quality in Nanjing is quite poor [15].

2.2. Sample collection

The dust samples were collected in Nanjing in July 17–20, 2021. Sampling points are more distributed in densely populated areas and less distributed in sparsely populated areas. Five points sampling method was used for sampling. The sampling sites (Fig. 1) are chosen far away from obvious emissions and generally located at the building above at least 1 m from ground. Three samples were collected at each sampling site. The dust were swept into plastic bag with a clean soft brush, and large stones/plastics were removed from the plastic bag. The samples were then brought back to the laboratory for analysis.

2.3. Sample analysis

The pretreatment methods of dust samples were described by Ref. [16]. Briefly, 100 mg dust sample were dissolved by mixed acids (nitric acid, hydrofluoric acid, and perchloric acid) with heating for 1–2 h. Two blanks were added in every 10 samples. The heavy metals were measured by inductively coupled plasma mass spectrometry (ICP-MS) [17], and the target elements included cadmium (Cd), chromium (Cr), copper (Cu), lead (Pb), nickel (Ni), and zinc (Zn).

Table 1
Concentrations of heavy metal in dust.

	Cr	Ni	Cu	Zn	Cd	Pb
Range/(mg·kg^{-1})	65.93–10848.23	37.74–7404.02	25.26–940.91	181.35–30483.96	0.33–4.00	30.48–797.47
Mean/(mg·kg^{-1})	528.94	322.83	167.40	3175.36	1.28	141.84
Standard deviation/(mg·kg^{-1})	1525.70	1048.18	152.81	5826.14	0.77	156.90
CV/%	288.00	325.00	91.00	183.00	60.00	111.00
Background value/(mg·kg^{-1})	83.00	40.00	31.00	79.00	0.14	23.00
The ratio of the mean to the background value	6.37	8.07	5.40	40.19	9.17	6.17

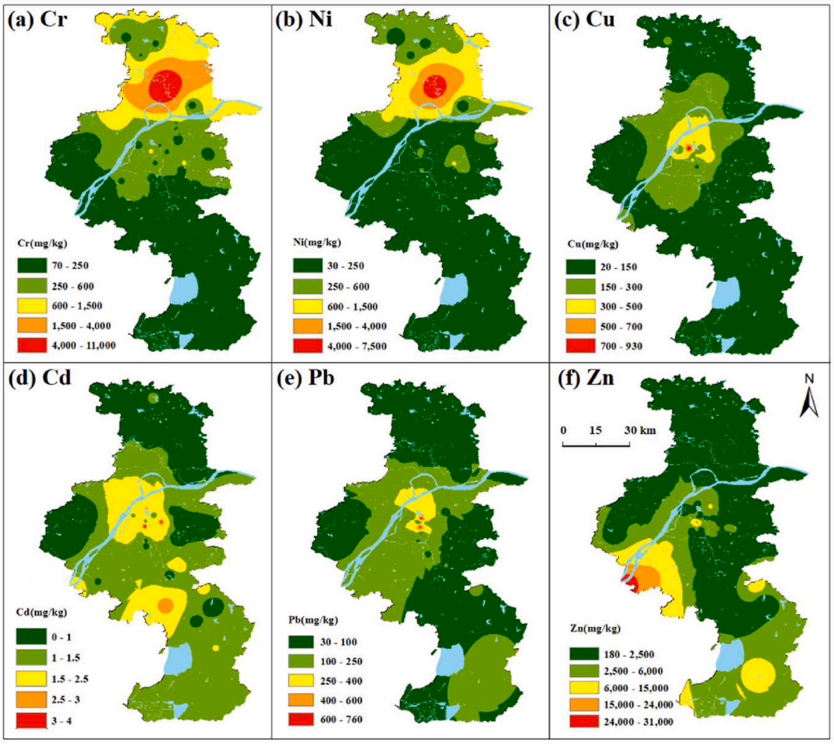

Fig. 2. Spatial distribution of heavy metals in atmospheric dust.

2.4. *Spatial interpolation, source diagnostic, and risk assessment*

The geostatistical analysis with inverse distance weight interpolation (IDW) was run in ArcGIS10.3 to describe the distribution of heavy metals in atmospheric dust. Compared with other interpolation methods such as Kriging, the inverse distance weight interpolation method has certain advantages in analyzing the spatial distribution characteristics of heavy metals and their related attributes because it does not have clear assumptions about the statistical properties of the input data [18]. The absolute principal component score, based on principal component analysis (PCA), and multiple linear regression (APCS-MLR) receptor model was used to quantitatively explore the sources of heavy metal pollution, conducted by IBM SPSS Statistics 26. Land accumulation index and ecological damage index were applied for assessing ecological risk ([19]; [20], and health risk assessment model provided by US Environment Protection Agency was also applied [21]. We further combined APCS-MLR receptor model with the above indexes to evaluate the risks of heavy metals from individual sources. The detailed calculations were in the supplementary materials (Text S1–S3).

3. Result and discussion

3.1. *Overview of concentrations*

The concentrations of Cr, Ni, Cu, Zn, Cd and Pb in dust in Nanjing are 65.93–10848.23 mg kg^{-1}; 37.74–7404.02 mg kg^{-1}; 25.26–940.91 mg kg^{-1}; 181.35–30483.96 mg kg^{-1}; 0.33–4.00 mg kg^{-1}; 30.48–797.47 mg kg^{-1}, respectively (Table 1), which are 40, 6.4, 8.1, 5.4, 6.2 and 9.2 times of the background values of these elements in the soil of Nanjing [22]. The concentrations of Zn and Cr are relatively high, accounting for more than 85% of the total concentrations of the six elements. The coefficient of variation of the six

Table 2
Contribution of heavy metal pollution sources to atmospheric dust.

	Fossil fuel combustion	Agricultural usage	Industrial emission	Traffic source
Cr	81%	15%	2%	2%
Ni	84%	8%	2%	6%
Cu	2%	92%	3%	3%
Zn	1%	31%	60%	8%
Cd	1%	82%	10%	6%
Pb	2%	43%	6%	49%
average	29%	45%	14%	12%

heavy metal elements is Ni (325%) > Cr (288%) > Zn (183%) > Pb (111%) > Cu (91%) > Cd (60%), meaning that the spatial distributions of these elements varied greatly.

The content and distribution of heavy metals in soil are affected by many environmental factors. This study only analyzes the content of heavy metals in atmospheric dust fall, and the actual sampling work is limited. More factors can be considered for in-depth and detailed exploration in subsequent studies.

3.2. Spatial distribution

The spatial distributions of HMs are shown in Fig. 2. The HMs were classified into three groups: 1) Cr and Ni showed the peak concentrations in the northern parts of Nanjing (Fig. 2a–b), which matched with the distribution of industrial parks and traffic hub. 2) The highest concentrations of Cu, Cd, and Pb occurred in the urban regions of Nanjing (Fig. 2c–e), relating possibly with the domestic activities in Nanjing; while, another polluted hot-spot of Cd located in the south of Nanjing (Fig. 2d), which may relate with agricultural emission. 3) Zn had the unique pattern with scattered high concentrations in the regions near the upstream of Yangtze River in the Nanjing (Fig. 2f).

Pearson correlation (Table S5) further verified the trends of spatial distribution: the relationship between Cr and Ni was significantly positive ($p < 0.01$), and significant relationships occurred among Cu, Cd, and Pb ($p < 0.05$). Generally, significant relationships are usually explained by similar emission sources [23]. Therefore, the spatial patterns, as well as the relationships between HMs, suggested that the at least three types of sources happened for the HMs in Nanjing dust.

3.3. Source diagnostic

The PCA extracted four principal components (PCs) from the data series, and the cumulative contribution rate reached 93.4% (Table S6). PC1 explained 33.5% variations of the HMs with high loading of Cr and Ni (Table S6). Cr and Ni mainly come from the fossil fuel combustion during metal smelting, oil production, and waste incineration [24,25]. The hot spots of Cr and Ni were mainly located in the northern part of Nanjing (Fig. 2a–b), where is the main industrial district. PC2, relating with Cu and Cd, explained 26.4% of the variances (Table S6). Cd is a marker of chemical fertilizer usage [26], suggesting that PC2 stand for the agricultural emission. One element, Zn, had high loading in PC3 (Table S6). The hot pots of Zn were in the upstream of Nanjing (Fig. 2f), near Maanshan, a famous city for steel production. PC3 might relate with the industry of mining and smelting (Table S6). Finally, Pb is in the PC4 (Table S6), with the probability from traffic emission.

Based on the multiple linear regression, the contributions of the four sources for each element were estimated. Briefly, more than 80% of Cr and Ni come from the industrial district in Nanjing, and more than 80% of Cu and Cd are from agriculture or the corresponding activities (Table 2). Approximate 60% of Zn may be from the industry of the upstream city, and others were from farmland. An exception is that two sources, traffic and agriculture, contributed the Pb emission. The contribution of the four pollution sources to the total heavy metal content in atmospheric dust fall was as follows: agricultural usage (45%) **>** fossil fuel combustion (29%) **>** industrial emission (14%) **>** traffic source (12%). Agricultural usage contributes the most, and it is necessary to strengthen the control of agricultural emissions.

3.4. Risk assessment

Three methods were applied to assess the risks of HMs in Nanjing dust. Generally, based on geo-cumulative pollution index and potential ecological risk index, the Zn and Cd could be the primary pollutants, which had apparent risks to influence the health of ecosystem (Text S4). Especially, the Cd in more 90% sampling sites had high risks. However, the most toxic HMs for human is Cr, which results in that the non-carcinogenic risks of the total HMs for children are above the threshold values of non-risks (Figure S3). The hand-oral ingestion is the primary pathway of the HMs in dust into children body (Table S8).

In order to make the evaluation more comprehensive and accurate, we choose two index methods to evaluate the risk of the study area. There are differences in the conclusions drawn by different evaluation methods. The risk evaluation results obtained by the ground accumulation index method are higher than the evaluation results of potential ecological risks on the whole. Comprehensive analysis shows that this may be related to the selection of parameters or its own evaluation principle. The ground accumulation index shows that Zn is highly polluted, while the evaluation results of potential ecological risk index show that Cd is the main pollution risk, because Cd has a large toxicity response coefficient and the overall harm to the environment is greater. The results of using multiple

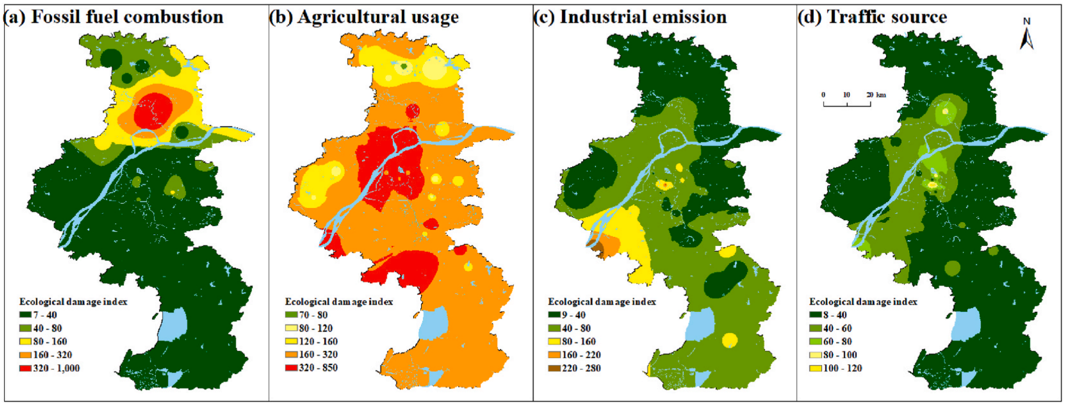

Fig. 3. Ecological risks of individual sources.

index evaluation methods are better than single evaluation methods, and different methods can complement each other and make the results more scientific and reasonable.

Now that the sources were recognized by APCS-MLR, the risks of individual sources were further assessed (Fig. 3 and Text S5). The high ecological risks were from industrial and agricultural sources (Fig. 3). Also, it can be found that the high ecological risks of industrial sources located in the northern Nanjing and the regions near Maanshan (Fig. 3a and c), and the agricultural sources impacted the safety of ecosystems in the southern regions of Nanjing (Fig. 3b). In addition, the high risks of traffic sources were all in the urban regions (Fig. 3d). These patterns followed the source regions of HMs (Fig. 2), indicating that emission could be the most important factor to threat the health of ecosystem and human. As other mega-cities, the primary task of environmental management in Nanjing should be to control possible emissions.

4. Conclusion

This study reported the concentrations, sources, and risks of HMs in Nanjing dust, and found that Cd, Ni, and Cr, which were from industrial emission, could be the primary HMs threaten the health of ecosystem and human. It will provide useful information on the priority of pollution control in Nanjing. However, the results of current study based on the total concentrations, but didn't consider the speciation and bioavailability of HMs in dust, which could introduce uncertainties. Future studies should be focused on the cycling of HMs across environmental medium and their chemical transformation during the cycling.

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Author contribution statement

Dike Feng: Ping Gong: Yan Li: Conceived and designed the experiments; Analyzed and interpreted the data; Contributed reagents, materials, analysis tools or data; Wrote the paper.

Ning Li: Zhen Dong: Ziyue Zhu: Ruihao Jiang: Siyun Deng: Performed the experiments.

Data availability statement

Data will be made available on request.

Additional information

Supplementary content related to this article has been publish online at [URL].

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Appendix A. Supplementary data

Supplementary data related to this article can be found at https://doi.org/10.1016/j.heliyon.2023.e18858.

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