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Anthropogenic Contribution and Migration of Soil Heavy Metals in the Vicinity of Typical Highways

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Abstract: To reveal the secondary anthropogenic contribution and accumulation rate of heavy metals in highway-side soils, we studied soil heavy metals on three representative highways in Southeast China, with different traffic intensities, service years, land use patterns and distances from roads, with high-resolution sampling of soil profiles. Concentrations of soil Cu, Zn, Pb and Cd were measured. The comparison of concentrations in surface soils with original values and their vertical distributions shows that soils within 150 m of the highway side are contaminated by heavy metals, with surface accumulation and possible movement down the profiles. The transferring depth of heavy metals was 10–30 cm. The contribution ratios of heavy metals were 1.0–30.5% in the surface at 30 cm, with the sequence of Cd >> Cu > Zn > Pb. The accumulation rates were 1.27–2.03 kg Cu ha^{-1} y^{-1}, 2.44–5.27 kg Zn ha^{-1} y^{-1}, 0.71–1.40 kg Pb ha^{-1} y^{-1} and 0.010–0.018 kg Cd ha^{-1} y^{-1} in soils within 50 m range. Furthermore, the accumulation of these metals varied with the traffic intensity, service years and land use patterns. Soils under forest have less heavy metal accumulation, which suggests a protective forest to set beside highways at a distance of at least 50 m to prevent soils from being contaminated.

Keywords: accumulation rate; anthropogenic contribution; element migration; heavy metals; highway-side soil; protective forest

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1. Introduction

Highways are indispensable in modern industrialized societies. Besides their obvious economic benefits, highways are the source of some environmental problems [1–7]. The initial problem is direct damage to the ecosystem at the time of highway construction, and by habitat fragmentation [8]. As soon as the highway enters service, emissions from internal combustion engines cause air, soil and water pollution, mostly in the immediate vicinity of the highway [9,10]. Some emitted particles with heavy metal precipitate onto highway-side soil, while others' direct deposits on the roadway surface are washed onto adjacent soils by rainfall runoff [11–14]. Plants growing in contaminated environments can accumulate trace elements at high concentrations [15–18]. Thus, soil is the sink and source of heavy metals from traffic atmospheric deposition and highway runoff, and it is an important medium between traffic pollution and human beings via the food cycle [16,19–21]. However, soil pollution of heavy metals is more covert than atmospheric and water pollution, and is hence not easy to determine. Therefore, many crops are planted at the sides of highways. It induces heavy biological toxicity and is subjected to enrichment and magnification via the food chain; consequently, it becomes a serious threat to human health [22,23]. Over the last several decades, there has been increased attention given to heavy metal contamination associated with highways [1,5,7,16,24–26]. However, research on the anthropoenic contribution and accumulation rate of heavy metals in highway-side soils has not yet been conducted.

Generally, the heavy metals monitored in roadside soils include copper (Cu), zinc (Zn), lead (Pb), cadmium (Cd), chromium (Cr), cobalt (Co), nickel (Ni), arsenic (As) and iron (Fe) [23,27–29]. However, according to the published data, Cu, Zn, Pb and Cd are the main polluting metals from vehicles [3,12,23,29–33].

Most research on heavy metals in highway-side soils has been limited to surface soils [16,25,27,34–36] and seeks to relate metal contamination to the distance from roads [16,25,27,36]. Ruan et al. [37] found that high-resolution sampling in soil profiles could be used to observe the clear migration track and accumulation characteristics of heavy metals. Thus, high-resolution sampling is necessary for heavy metal research. However, only a few papers report on heavy metal mobility within the soil profile beside highways [28,38–40]. Most of these studies were based on genetic horizons or large sampling intervals; usually, the studied thicknesses were more than 10 cm. This sampling design could not appropriately identify the accumulation and migration characteristics of heavy metals because of the slow migration rates and shallow migration depth of heavy metals.

Jiangsu Province of China is located in the Yangtze Delta, Southeast China, and is characterized by a flat terrain and fertile land, and thus it is a major agricultural and industrial base and is one of China's most developed regions. Consequently, the development of its highways has also been rapid. In 2009, the expressways at a province scale amounted to 3725 km, and they were 4443 km in 2013. Due to the high population density and strong industrial economy, the traffic intensity in this region is the highest of the whole country. Thus, the aim of this study was to reveal the anthropogenic contribution and its ratio, the accumulation rate of heavy metals in highway-side soils and also the effect factors in the rapidly developing regions of China. These will provide a scientific basis for soil management beside highways for human health.

2. Materials and Methods

2.1. Study Area

Three highways, the Ningtong, Jinghu and Yanjiang expressways, located in Jiangsu Province, Southeast China (Figure 1), were selected for sampling in 2007, differing in their highway service years, vehicle flow and land use patterns (Table 1). The sampling sites were far from industrial areas and other pollution sources. The parent materials of all sampled soils originated from Yangtze Delta alluvium, which was heterogeneous. Since previous studies showed that heavy metal contamination was concentrated within 300 m from roads [1,41–44], we sampled at two distances from roads: 50 and 150 m.

Table 1. Characteristics of studied highways.

Highway	Service Yeas	Vehicles per Day #	Distance from Roads (m)	Land Use	Profile	Samples (n)
Ningtong expressway	10	40,000	50	Cropland	NT1, NT4, NT6	42
			150	Cropland	NT2, NT3, NT5, NT7	56
Jinghu expressway	6	34,000	50	Forest and cropland	JH1, JH3, JH5, JH7	56
			150	Cropland	JH2, JH4, JH6, JH8	56
Yanjiang expressway	3	25,000	50	Forest and cropland	YJ1, YJ3, YJ6	42
			150	Cropland	YJ2, YJ4, YJ5	42

Data derived from Jiangsu Transportation Bureau in 2007.

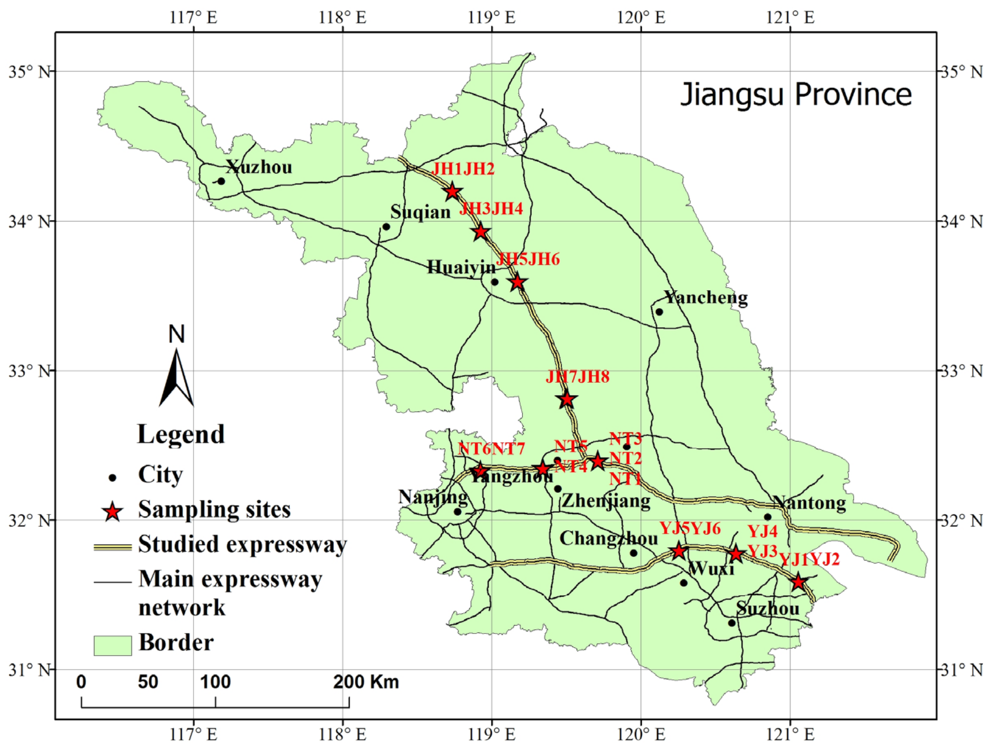

Figure 1. The study areas and sampling sites.

2.2. Soil Sampling and Preparation

There were 21 profiles along different highways at 50 and 150 m (Figure 1). Samples were collected every 5 cm from the surface to 40 cm, and then every 10 cm at the sections 40 cm to 100 cm from soil profiles. The substrate was a layer of parent materials. Thus, there were 14 sampled layers in each soil profile. The same expressway and same distance from the road had 3–4 profiles in one repetition. In total, we obtained 294 soil samples. To avoid heavy metal contamination in the sampling process, a stainless-steel shovel was used to excavate profiles and to collect samples. The samples were air-dried, cleaned of stones and plant debris and ground with an agate mortar, and all of the soils were passed through 0.149 mm nylon sieves for analysis. Soil samples were not in contact with heavy metals.

2.3. Chemical Analysis

Soil samples (0.1 $\pm$ 0.0001 g) were digested using a mixture of acids (6 mL of 90% HNO_3, 3 mL of 75% $HClO_4$ and 3 mL of 78% HF) in a polypropylene vessel. The obtained residuals were dissolved in 10 mL HCl and diluted to 25 mL with distilled water to measure the concentrations of metals. The total concentrations of Cu, Zn, Pb and Cd were determined using an inductively coupled plasma atomic emission spectrometer (ICP-AES) [45]. Blanks were analyzed during the same procedure. Measurement quality was controlled with national standard samples (GSS-3 and GSS-8). Soil pH was measured with a 1:2.5 soil:water ratio. Soil organic matter (SOM) was determined with wet oxidation, digested by $K_2Cr_2O_7$ and quantified by distillation and titration. Undisturbed soils were sampled with standard core rings of 100 cm^3 in volume (height, 52 mm; diameter, 70 mm) to determine the soil bulk density.

2.4. Data Analysis

The data were statistically analyzed via the Statistical Program for Social Sciences (SPSS 13.0 for Windows). Analysis of variance was used to study statistical differences (95% confidence level) in the concentrations of heavy metals in soil profiles and at different distances from roads. The figures were generated using the Excel 2013 and OriginPro 8 software.

3. Results

3.1. Soil Basic Characteristics

Soil pH was 4.2–9.0, with the mean of 7.6. In the surface soil, pH was 4.2–8.1, with the mean of 7.0, while it was 6.5–9.0, with the mean of 7.8, in the subsoils. It was clear that soil beside the highway was weakly alkaline, but some acidity was noted in the topsoil.

The content of soil organic matter was 1.6–38.5 g kg^{-1}, with the mean of 10.4 g kg^{-1}. The consistent pattern for organic matter decreased from the surface to lower levels, with the mean of 23.7 g kg^{-1} in the topsoil and 4.5 g kg^{-1} in the subsoils. Therefore, the soil was fertile, with high organic matter at the surface.

3.2. Vertical Distribution of Heavy Metal Concentrations

The concentrations of Cu, Zn, Pb and Cd were 8.7–32.2, 26.4–90.2, 8.3–34.2 and 0.007–0.213 mg kg^{-1}, respectively. The average concentrations of the metals were 19.1 $\pm$ 4.6 mg Cu kg^{-1}, 58.4 $\pm$ 14.2 mg Zn kg^{-1}, 19.8 $\pm$ 6.2 mg Pb kg^{-1}, 0.080 $\pm$ 0.037 mg Cd kg^{-1} in the three highway-side soils, which were lower than the corresponding geochemical background values (22.7 mg Cu kg^{-1}, 62.9 mg Zn kg^{-1}, 24.9 mg Pb kg^{-1} and 0.118 mg Cd kg^{-1}) [46], respectively.

Regarding the vertical distribution of Cu, Zn, Pb and Cd, the highest average concentrations were found in the surface at both 50 and 150 m from highways (Figure 2). Moreover, they decreased from surface to subsoils with depth. There were large standard deviations (STD) for each element in the profiles (Figure 2), which were ascribed to the heterogeneous parent material, i.e., alluvium originating from the Yangtze Delta.

Although there was no significant difference in the heavy metal concentrations at 50 and 150 m (Figure 2), the data showed some distribution characteristics between them. For the Ningtong expressway, Zn and Pb concentrations in the soil 50 m from roads were higher than those at 150 m. For the Yanjiang expressway, the concentrations of Cu, Zn, Pb and Cd in the soil 50 m from roads were higher than those at 150 m (Figure 2). These data show that the effects of heavy metals from traffic on soils are more severe at 50 m than 150 m from expressways. It is surprising that the concentrations of Cu, Zn, Pb and Cd in the Jinghu expressway at 150 m are slightly higher than those at 50 m in most cases (Figure 2b). A possible reason is the high concentrations of heavy metals in the parent material. However, in the soil surface of these profiles, the Zn and Pb concentrations are higher at 50 m than that at 150 m, and the Cu and Cd concentrations are similar (Figure 2b), which further confirms the addition of heavy metals in the soil, especially in the surface soil at 50 m. Therefore, the exogenous added heavy metals are not sufficient to lead to significant differences in concentrations between 50 and 150 m over ten years of highway service, but the data at the soil surface reflect a slight difference.

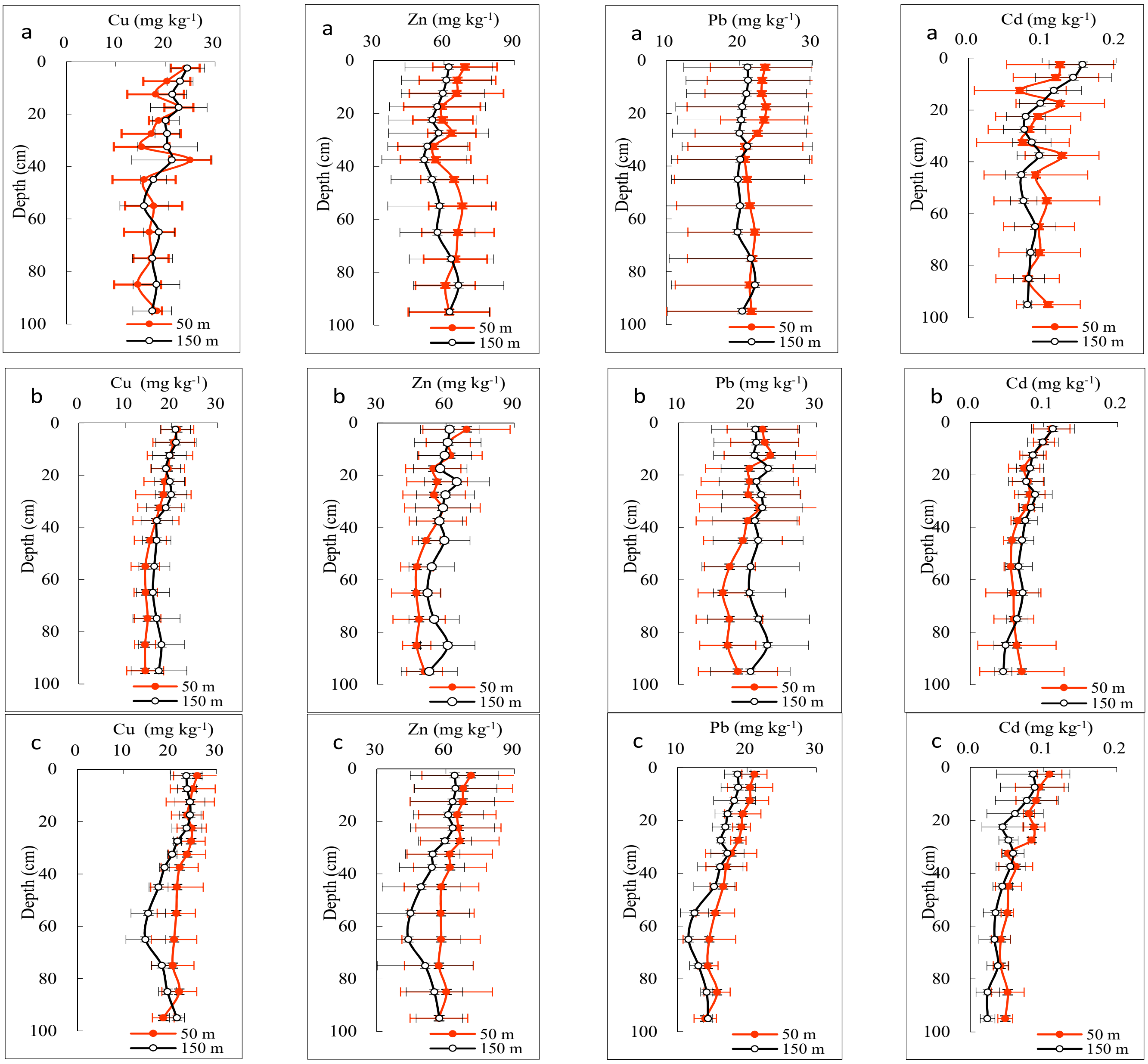

Figure 2. Vertical distributions of heavy metal concentrations for the average values and standard deviations (STD) in the vicinity of three highways: (**a**) Ningtong expressway; (**b**) Jinghu expressway; (**c**) Yanjiang expressway. The concentrations are the average values of repetitions. n = 3–4, the number of repetitions, as shown in Table 1.

3.3. Heavy Metal Concentrations in Surface Soils

The average values of Cu at the Jinghu expressway; Pb at the Ningtong, Jinghu and Yanjiang expressways; and Cd at the Jinghu and Yanjiang expressways in the surface soil (0–5 cm) are lower than the corresponding elemental background values (22.7 mg Cu kg^{-1}, 24.9 mg Pb kg^{-1} and 0.118 mg Cd kg^{-1}) (Figure 3). However, some concentrations are higher than their background values, such as 57.1% Cu, 42.9% Zn, 28.6% Pb and 71.4% Cd samples along the Ningtong expressway; 37.5% Cu, 66.7% Zn, 25.0% Pb and 37.5% Cd samples along the Jinghu expressway; and 66.7% Cu, 66.7% Zn and 16.7% Cd samples along the Yanjiang expressway. Only soil Pb concentrations along the Yanjiang expressway are lower than the background value (Figure 3). Clearly, the surface soils are contaminated by Cu, Zn, Pb and Cd at the three expressways, especially along the Ningtong expressway.

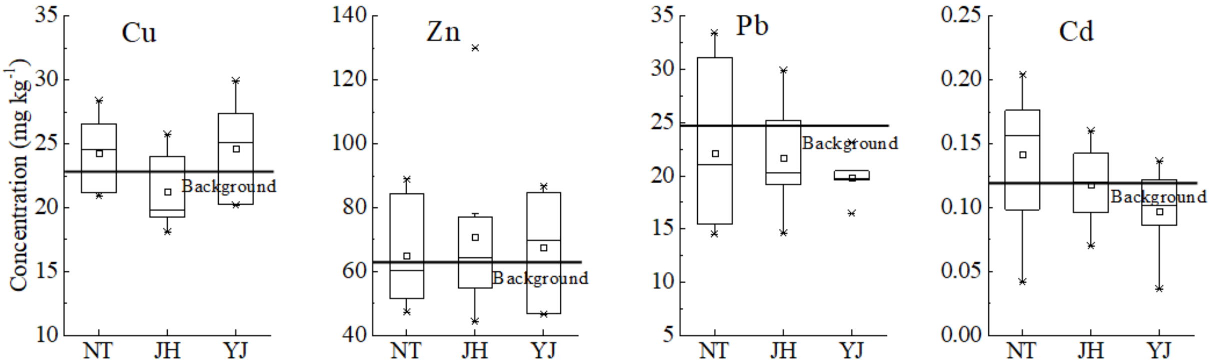

Figure 3. Heavy metal concentrations at the depth of 0–5 cm in highway-side soils and the corresponding background values. (The black bold lines are the background values of corresponding elements. NT, Ningtong expressway; JH, Jinghu expressway; YJ, Yanjiang expressway. The interquartiles from bottom to top in the box are 25%, 50% and 75% of the data, respectively. The small square is the average value. The two endpoints of the bar are minimum and maximum values, respectively. The asterisks are the outliers).

3.4. Heavy Metal Concentrations in Subsoils

Generally, the pollution risk for deep soil is low because of the slow translocation rate of heavy metals. In this study, all average concentrations of Cu, Zn, Pb and Cd in highway-side soils at a 90–100 cm depth for all three highways are lower than the background values (22.7 mg Cu kg^{-1}, 62.9 mg Zn kg^{-1}, 24.9 mg Pb kg^{-1} and 0.118 mg Cd kg^{-1}) (Figure 4). These demonstrate that the subsoils are at natural levels and are not polluted by Cu, Zn, Pb and Cd. The comparison of Figures 3 and 4 shows that the maximum and average metal concentrations in subsoils are all lower than those of corresponding surface soils, which implies that surface soils have received additional metals, and further implying that this is from traffic. However, the paired-samples *t*-test (Table 2) could only prove a significant difference for Cu and Cd for all three expressways, and for Pb at the Yanjiang expressway. No significant difference for Zn and Pb is ascribed to the large variance in surface soils and subsoils (Table 2).

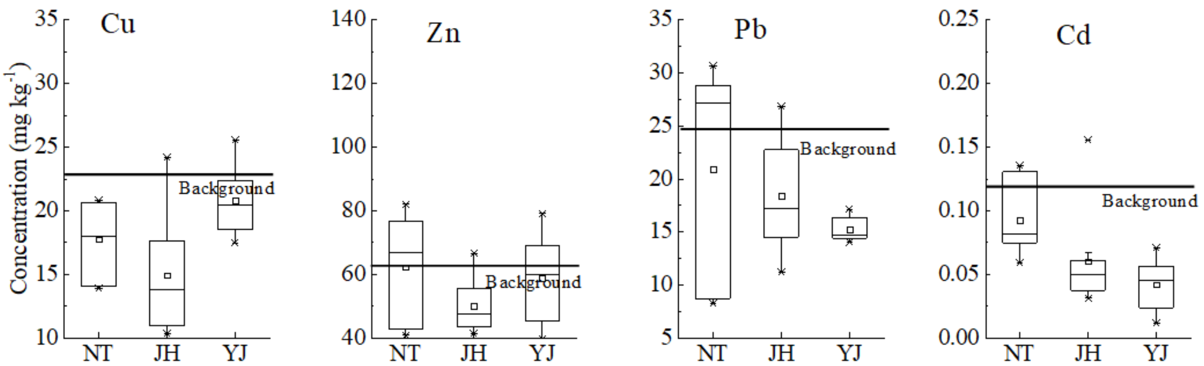

Figure 4. Heavy metal concentrations at the depth of 90–100 cm in highway-side soils and the corresponding background values. (The black bold lines are the background values of corresponding elements. NT, Ningtong expressway; JH, Jinghu expressway; YJ, Yanjiang expressway. The interquartiles from bottom to top in the box are 25%, 50% and 75% of the data, respectively. The small square is the average value. The two endpoints of the bar are minimum and maximum values, respectively. The asterisks are the outliers).

Table 2. Comparison between heavy metals in surface and in subsoils at three highways.

Highway	Layer	Cu	Zn	Pb	Cd
		mg kg^{-1}			
Ningtong expressway	Surface	24.2a * ± 2.99 #	65a ± 15.89	22.2a ± 7.63	0.142a ± 0.055
	Substrate &	17.8b ± 2.87	62.2a ± 15.72	20.9a ± 9.90	0.092b ± 0.029
Jinghu expressway	Surface	21.2a ± 3.02	70.9a ± 26.78	21.7a ± 5.36	0.118a ± 0.032
	Substrate	15.0b ± 4.77	50.1a ± 9.13	18.4a ± 5.72	0.060b ± 0.040
Yanjiang expressway	Surface	24.7a ± 4.06	67.5a ± 18.69	19.9a ± 2.12	0.097a ± 0.036
	Substrate	20.8b ± 2.94	58.8a ± 14.70	15.2b ± 1.26	0.042b ± 0.022

& Parent material, 90–100 cm. * The different letters for the same highway indicate significant difference at 95% confidence level based on paired *t*-test (n = 7 in Ningtong expressway, n = 8 in Jinghu expressway, n = 6 in Yanjiang expressway). # Standard deviation.

3.5. Distribution Mode of Heavy Metals

The distributions of heavy metals in the profiles imply that Cu, Zn, Pb and Cd accumulated in the surface soil and moved down to deeper layers (Figure 2). In the metal accumulation layers, the heavy metal concentrations decrease exponentially with the increase in soil depth in the profile. Moreover, in soils under metal accumulation layers, the heavy metal concentrations are identical to the background values. Therefore, we obtained conceptual sketches of heavy metal translocation (Figure 5). In homogeneous materials, the heavy metal concentrations are constant in the unpolluted soil layers (Figure 5A). However, in heterogeneous parent materials, such as the soils studied here, developed from alluvium, the heavy metal concentrations are fluctuant in the unpolluted soil layers and within the range of background values (Figure 5B). The obvious accumulation and migration of metals are found at a 20–30 cm depth in locations with a distance of 50 m from roads (Figure 2). Similarly, the accumulation and migration of metals are found at approximately a 10–20 cm depth in locations with a distance of 150 m from roads (Figure 2).

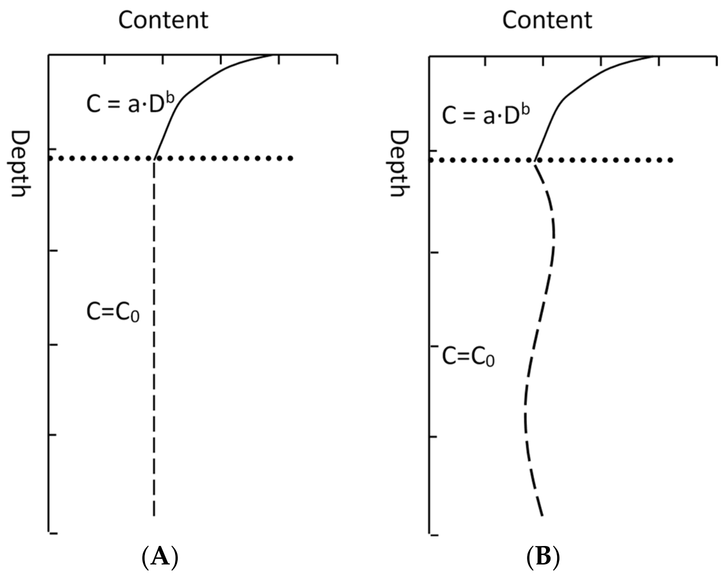

Figure 5. Heavy metal distribution conceptual mode in homogeneous material (**A**) and alluvium material (**B**). C, heavy metal concentration; C_0, heavy metal concentration of parent material; D, soil depth; a and b, the equation coefficient.

3.6. Accumulation Amount of Heavy Metals

In the present study, a new evaluation method different from previous research is introduced. The soil heavy metal concentration is the sum of that in the soil parent material and that of metal input from the environment. Consequently, many studies have estimated soil heavy metal pollution by comparing the heavy metal concentrations studied with the national or local soil background value, or they have estimated metal accumulation by calculating the differences between the concentrations at the study time and those in the past [30,39]. In our study, because the parent material was alluvium in all cases, their heavy metal concentrations were heterogeneous, even in the same highway (Figure 5B). Thus, we took the concentrations of the below-soil layers as the original values, into which metals from the surface have migrated. The calculation formula is as follows:

$$G = S \times D \times P \times C \quad (1)$$

G is the total amount of heavy metal; S represents the land surface area; D is the soil depth; P is the soil's bulk density; and C is the heavy metal concentration. Thus, the heavy metal j accumulation (ΔG_{ji}), i.e., the input amount from the environment, in the ith layer soil is as follows:

$$\Delta G_{ji} = S \times D_i \times P_i \times (C_{ji} - C_{j0}) \quad (2)$$

D_i is the ith layer soil depth; P_i is the ith layer soil bulk density; C_{ji} is the heavy metal j concentration in the ith layer soil. C_{j0} is the heavy metal j concentration in the parent material. Thus, the heavy metal j accumulation (G_{jn}) at a certain depth (n layers) is as follows:

$$G_{jn} = \sum_{1}^{n} \Delta G_{ji} = \sum_{1}^{n} S \times D_i \times P_i \times (C_{ji} - C_{j0}) \quad (3)$$

We established the depths of heavy metal migration and the original background value, so Formula (3) could be applied. The original values were the content of heavy metals in the subsoils of every profile. The depths of heavy metal migration were obtained by a comparison of the heavy metal content in all soil layers to that in the subsoils. The average accumulation amounts were 5.12–12.7 kg Cu ha^{-1}, 15.8–24.4 kg Zn ha^{-1}, 4.20–7.12 kg Pb ha^{-1} and 0.055–0.101 kg Cd ha^{-1} in soils with a distance of 50 m from roads along all three highways according to Formula (3) (Table 3). The accumulation sequence is Zn > Cu > Pb > Cd. The same sequence but at lower accumulations for 1.72–8.46 kg Cu ha^{-1}, 8.02–13.2 kg Zn ha^{-1}, 0.82–5.95 kg Pb ha^{-1} and similar 0.051–0.140 kg Cd ha^{-1} was found at the 150 m distance.

Table 3. Accumulation amount of heavy metals in soil profiles in vicinity of highways.

Expressway	Distance from Roads (m)	Cu	Zn	Pb	Cd
		kg ha^{-1}			
Ningtong expressway	50	12.7	24.4	7.12	0.101
	150	8.46	13.2	5.95	0.140
Jinghu expressway	50	12.2	22.6	4.79	0.067
	150	4.15	8.09	0.82	0.064
Yanjiang expressway	50	5.12	15.8	4.20	0.055
	150	1.72	8.02	3.29	0.051

There are different types of land use in the vicinity of highways. The accumulation amount of heavy metals was calculated in cropland and forests with a distance of 50 m from two highways, the Jinghu expressway and Yanjiang expressway. In the Jinghu expressway, JH3 was located under a forest, but JH1, JH5 and JH7 were located in cropland used for maize, wheat and paddy. The accumulation amounts of Cu, Zn, Pb and Cd in the forest were 5.31, 11.75, 1.51, 0.02 kg ha^{-1}, respectively, while those in the cropland were 5.94–20.0, 15.8–37.6, 2.17–7.81, 0.04–0.13 kg ha^{-1}, respectively (Table 4). In the Yanjiang expressway, YJ1, YJ3 and YJ6 were within 50 m of the edge of expressway; of these, YJ1 and YJ3 were in cropland, while YJ6 was under a forest. The accumulation of Cu, Zn, Pb and Cd in the forest was 1.80, 5.03, 1.23, 0.02 kg ha^{-1}, respectively, while that in the cropland was 6.14–7.41, 15.8–26.5, 4.07–7.29, 0.07–0.08 kg ha^{-1}, respectively (Table 5). It is clear that land use affects the accumulation of heavy metals in the soil.

Table 4. Accumulation amounts of heavy metals for different land uses with a distance of 50 m from Jinghu expressway.

Profile	Land Use	Cu	Zn	Pb	Cd
		kg ha^{-1}			
JH1	Cropland	20.0	25.3	7.81	0.08
JH3	Forest	5.31	11.75	1.51	0.02
JH5	Cropland	17.4	37.6	7.67	0.13
JH7	Cropland	5.94	15.8	2.17	0.04

Table 5. Accumulation amounts of heavy metals for different land uses with a distance of 50 m from the edge of the Yanjiang expressway.

Profile	Land Use	Cu	Zn	Pb	Cd
		kg ha^{-1}			
YJ1	Cropland	6.14	15.8	4.07	0.08
YJ3	Cropland	7.41	26.5	7.29	0.07
YJ6	Forest	1.80	5.03	1.23	0.02

3.7. Contribution of Anthropogenic Components

The soils located in the vicinity of the highways were polluted by the heavy metals Cu, Zn, Pb and Cd, especially the surface soils. Because surface soils are acidic, with high organic matter, the heavy metals are easily migrated in the profiles. The migrated depths of heavy metals were approximately 10–30 cm. The anthropogenic contribution ratios of heavy metals could be obtained by the comparison of accumulation, as shown in Table 3, and the total amount calculated from Formula (1). The results show that the secondary anthropogenic contribution ratios of Cu, Zn, Pb and Cd were 1.0–30.5% in the surface 30 cm soils (Table 6). Although Cd had the lowest accumulation amount, it had the highest anthropogenic contribution ratio of 14.9–30.5% among the four types of heavy metals. Cu was also a high-accumulation element, especially at a distance from roads of 50 m for the Ningtong and Jinghu expressways, with anthropogenic contribution ratios of more than 10%. Even for the Yanjing expressway with only three years of service, 1.9–5.3% Cu, 3.3–6.0% Zn, 4.8–5.4% Pb and 15.5–19.1% Cd from anthropogenic components are accumulated in the highway-side soils. Cd contamination is the most severe among the heavy metals considered in this study. Roadside dust is one of the heavy metal sources in highway-side soils. Therefore, there are anthropogenic components from traffic activities in the vicinity of expressways within 150 m of roads.

Table 6. Anthropogenic contribution ratios of heavy metals in the surface 30 cm soils in the vicinity of highways.

Expressway	Distance from Roads (m)	Cu	Zn	Pb	Cd
		%			
Ningtong expressway	50	16.2	9.8	7.9	25.3
	150	9.9	5.8	7.4	30.5
Jinghu expressway	50	13.4	7. 9	4.7	14.9
	150	5.3	3.4	1.0	18.2
Yanjiang expressway	50	5.3	6.0	5.4	15.5
	100	1.9	3.3	4.8	19.1

According to the service years of every expressway (Table 1) and the amount of accumulation (Table 3), the accumulation rates of heavy metals are calculated as 1.27–2.03 kg Cu ha^{-1} y^{-1}, 2.44–5.27 kg Zn ha^{-1} y^{-1}, 0.71–1.40 kg Pb ha^{-1} y^{-1} and 0.010–0.018 kg Cd ha^{-1} y^{-1} in soils at the highway side at 50 m (Table 7). These rates of

Cu, Zn and Pb generally decline to one third or half at 150 m from roads, while Cd has similar accumulation rates in the soils at 50 m and 150 m from roads.

Table 7. Accumulation rates of heavy metals in soil profiles in vicinity of expressways.

Highway	Distance from Roads (m)	Cu	Zn	Pb	Cd
		kg ha^{-1} y^{-1}			
Ningtong expressway	50	1.27	2.44	0.71	0.010
	150	0.85	1.32	0.60	0.013
Jinghu expressway	50	2.03	3.77	0.80	0.011
	150	0.69	1.35	0.14	0.011
Yanjiang expressway	50	1.71	5.27	1.40	0.018
	100	0.57	2.67	1.10	0.017

4. Discussion

4.1. Effects of Traffic Intensity and Service Years of Highway on Soil Heavy Metal Accumulation

Wheeler and Rolfe [47] found that the Pb and Cd levels in vegetation increased linearly with the traffic density and proximity to roads. Other researchers also showed that heavy metal levels in soil and plants in the vicinity of highways were related to the traffic intensity [16,18,35,48], while Olajire and Ayodele [41] reported that there was no obvious correlation between traffic density and heavy metal concentrations in highway-side soil and herbage plants in Ibadan, Nigeria. In this study, the accumulation amount (Table 3) and anthropogenic contribution ratios (Table 6) of Cu, Zn, Pb and Cd in soils in the vicinity of three highways were in the order Ningtong expressway > Jinghu expressway > Yanjiang expressway. Then, it was found that the above sequence was consistent with the traffic intensity (vehicles per day) and service years of the highways (Table 1). Furthermore, the accumulation rates of heavy metals in the soil profiles in the vicinity of expressways (Table 7) were increased with the number of service years. Therefore, traffic intensity and service years are important factors that affect heavy metal accumulation in highway-side soils. These rates of heavy metal accumulation will help to assess the evolution and environmental risk of highway-side soils.

4.2. Effects of Distance from Roads on Soil Heavy Metal Accumulation

Many researchers have shown that the heavy metal concentration decreases with the distance from the highway [1,44,49,50]. The distance at which heavy metals are affected by the highway is not consistent [14,29,41–44]. For example, the polluting distance of heavy metals is often found to be less than 150 m [17,36,48,50] and even less than 30 m [14] or 10 m [29]. However, others showed that it was more than 150 m [1,30,35]. In this study, there was no statistically significant difference (at the 95% level) between the Cu, Zn, Pb and Cd concentrations in surface (0–5 cm) soils with a distance of 50 m from roads and those at 150 m, although, in most cases, the amounts were slightly larger. A possible reason is that the parent material was Yangtze Delta alluvium, with great variation in element concentrations [46]. However, the accumulation amounts and rates of Cu, Zn and Pb in soils with a distance of 50 m from roads were higher than those at 150 m in all three highways (Tables 3 and 7). The accumulation amount and rate of Cd were similar at 50 m and 150 m. Therefore, heavy metals accumulate and move in the surface soils at locations with a distance of at least 150 m, although some elements would decrease with increased distance from roads.

4.3. Effects of Land Use Patterns on Soil Heavy Metal Pollution

There are forests and cropland at both sides of the selected highways. Heavy metal accumulation in the forest is the lowest among those of all land use patterns with identical distance from a certain highway side (Tables 4 and 5). Of course, it is undeniable that fertilization might contribute some heavy metals in the cropland. However, Tables 3 and 6 show that the accumulation amounts and anthropogenic contribution ratios at 50 m are greater than those at 150 m, indicating the contribution of traffic to heavy metal pollution in roadside soil, because the land use type was cropland in all sampling sites at 150 m. Wang et al. [51] also showed that the Pb concentrations in surface soils without forest cover were clearly higher than those with forest cover along highways. Therefore, the forest may obstruct particles with heavy metals beside forest edges and prevent some heavy metals from reaching the soil in the forest and other areas.

Some particles from engine exhaust, tire friction and goods such as coal and cement scattered from uncovered cargo may contain metals that are distributed by wind and airflow to the highway side [16,49]. Tree leaves could adsorb and absorb both particulate and gaseous contaminants. Further, Mori et al. [52] showed that trees near the road had a higher contribution of filtrating pollution elements as compared to those from greater distances. Therefore, a protective forest belt may be useful to prevent soils from being contaminated. However, some studies showed that the affected area of traffic-related metals was <30 m from the highway [14]. The safest distance to minimize metal pollution for agricultural production is proposed to be greater than 10 m from the road edge [29]. At present, many crops are planted within 50 m of highways. According to the results from this study, we strongly suggest that crops should be planted at least 50 m away from the highway, and trees should be planted within 50 m.

5. Conclusions

This study reveals that heavy metal pollution in soils has some concealment, especially in heterogeneous materials with large variance in element concentrations. In this case, the comparison of heavy metals in vertical distribution is very useful to determine the migration depth and calculate the accumulation amount.

Although the migration rate of heavy metals is slow, the migration depth of Cu, Zn, Pb and Cd in highway-side soils could be observed. The secondary contribution ratios of heavy metals by traffic are high, with the sequence of Cd >> Cu > Zn > Pb. Cadmium has the lowest accumulation amount compared to Cu, Zn and Pb, but it has the highest anthropogenic contribution ratio among these elements. The traffic intensity and service years of highways are the main effect factors related to heavy metal accumulation in highway-side soils. With increasing traffic intensity and service years, the highway-side soils are increasingly polluted by heavy metals. The distance of heavy metal pollution from traffic could reach 150 m. Soil under forest cover has less Cu, Zn, Pb and Cd accumulation, possibly due to its barrier effect against road dust. Therefore, a protective forest belt is strongly suggested to be implemented beside highways at a distance of at least 50 m in order to attenuate soil heavy metal pollution.

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