



## Enrichment and sources of trace metals in roadside soils in Shanghai, China: A case study of two urban/rural roads

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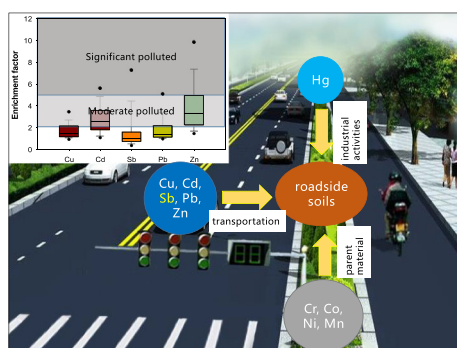
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### HIGHLIGHTS

- Traffic-derived Zn, Cd, Sb, Pb and Cu enriched in 78.5% of the roadside soil samples.
- Accumulation of emerging metal-Sb has been found in both young and old roads.
- Metal enrichment in soils were affected by level of urbanization and road age.

### GRAPHICAL ABSTRACT



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### ABSTRACT

The road traffic has become one of the main sources of urban pollution and could directly affect roadside soils. To understand the level of contamination and potential sources of trace metals in roadside soils of Shanghai, 10 trace metals (Sb, Cr, Co, Ni, Cu, Cd, Pb, Hg, Mn and Zn) from two urban/rural roads (Hutai Road and Wunign-Caoan Road) were analyzed in this study. Antimony, Ni, Cu, Cd, Pb, Hg and Zn concentrations were higher than that of soil background values of Shanghai, whereas accumulation of Cr, Co and Mn were minimal. Significantly higher Sb, Cd, Pb contents were found in samples from urban areas than those from suburban area, suggesting the impact from urbanization. The concentrations of Sb and Cd in older road (Hutai) were higher than that in younger road (Wunign-Caoan). Multivariate statistical analysis revealed that Sb, Cu, Cd, Pb and Zn were mainly controlled by traffic activities (e.g. brake wear, tire wear, automobile exhaust) with high contamination levels found near traffic-intensive areas; Cr, Co, Ni and Mn derived primarily from soil parent materials; Hg was related to industrial activities. Besides, the enrichment of Sb, Cd, Cu, Pb and Zn showed a decreasing trend with distance to the road edges. According to the enrichment factors ( $EF_n$ ), 78.5% of Sb, Cu, Cd, Pb and Zn were in moderate or significant pollution, indicating considerable traffic contribution. In particular, recently introduced in automotive technology, accumulation of Sb has been recognized in 42.9% samples of both roads. The accumulation of these traffic-derived metals causes potential negative impact to human health and ecological environment and should be concerned, especially the emerging trace elements like Sb.

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## 1. Introduction

With rapid development, intensive anthropogenic activities have led to a large amount of pollutants emitted into urban environment. Toxic trace metal has attracted great concern in environmental studies, due to their non-degradability and long residual time (Mwesiye et al., 2016; Peng et al., 2017). Many studies have demonstrated the importance of traffic being a source of trace metals to the urban environment, especially in roadside soils (Wang et al., 2009; Atkinson et al., 2011; Yang et al., 2011; Ordóñez et al., 2015; Botsou et al., 2016). Roadside soils, as main reservoir of the traffic-derived pollutants, expose people to trace metals through inhalation, direct ingestion, and dermal contact, thereby affecting human health, such as sanitation workers, residents nearby roads, and pedestrians (Li et al., 2014; Roy and McDonald, 2015). With the rise of green travel mode, the influence of trace metals in roadside soils on people should be given attention. Furthermore, as an important part of urban ecosystem, metal contaminated roadside soils may affect the ecological environment of the city through animals or plants uptake and leached by rainwater into water body (Calace et al., 2012). Therefore, it is very important to explore the accumulation and source of trace metals in urban roadside soils.

In recent years, many scholars have carried out researches on trace metals in roadside soils in different countries, e.g. Greece (Christoforidis and Stamatis, 2009), Saudi Arabia (Kadi, 2009), Germany (Kluge and Wessolek, 2012), Canada (Wiseman et al., 2015),

Australia (De Silva et al., 2016), Spain (Ordóñez et al., 2015), most of which were focused on the pollution characteristics including contamination level, distribution and migration of metals. Elevated concentrations were generally found due to the traffic. High contents of trace metals have raised concerns about the safety of vegetables grown on roadside soils (Nabulo et al., 2006). Trace metals generated by traffic activities could spread 100–200 m on both sides of roads (Dan-Badjo, 2008; Zhang et al., 2015), with most remained in surface soil (Boivin et al., 2008). Various factors including road age, traffic density, etc. had great impact on contamination level and distribution of trace metals (Perez et al., 2008; Werkenthin et al., 2014; De Silva et al., 2016). However, the number of researches on trace metals in roadside soils in China is relatively limited and the focus of previous studies was mainly on the conventional metals of Cd, Cu, Pb, Zn and Cr. There is little known on “new” metals introduced into automotive technology in recent decades, such as Sb in brake pads (Hjortenkrans et al., 2007), Mn in fuel as a replacement for Pb (De Silva et al., 2016), and Pt group elements in vehicle emission catalysts (VEC) to reduce air pollution (Almécija et al., 2015). Accumulations of these metals have occurred in the urban environment (Zereini et al., 2007; Wiseman et al., 2013; Ordóñez et al., 2015) and threaten human health (Ole Von Uexküll et al., 2005; Sanchez-Rodas et al., 2017).

After 30 years of rapid urbanization, by 2015, the population of Shanghai has reached 24.1527 million, following the increase of highway mileage to 13,195 km and number of cars to 282.32 million (Shanghai Statistic Bureau, 2016). Fast development has caused various environmental pollutions, even though many industries have moved from urban area since 1990s. Previous studies has demonstrated that the urban soils of Shanghai were highly contaminated by Cd, Cu, Pb, Zn, and the traffic emission may be the main source of Cu, Pb, Zn (Shi et al., 2008; Liu et al., 2012). However, little was known about metals in roadside soils (Fang et al., 2009), especially the contribution from emerging metals-Sb. To the author's best knowledge, the research presented here represents the first study to discuss pollution level of Sb in roadside soils of Shanghai. By intensive sampling along two typical roads in Shanghai, this study was aiming to (1) analyze the contents of trace metals and their spatial distribution in roadside soils; (2) identify the potential sources of the metals; (3) assess the contribution from traffic to metal enrichment in roadside soils. The results would help us to understand the pollution status of trace metals in roadside soils of Shanghai and to control the risk to human and environmental exposure.

## 2. Materials and methods

### 2.1. Study area

Shanghai situates in the east coast of China, with its center located at 31.14°N, 121.29°E. Adjacent to the Pacific Ocean, the city is subject to a subtropical monsoon climate, with an annual rainfall of 1122 mm and an average temperature of 15.8 °C (Shi et al., 2008). The city is normally divided into four zones according to the urban highway rings: the central urban core (CUC, inside the Inner-ring Highway), developed urban (DDU, between the Inner- and Middle-ring Highways), developing urban (DIU, between the Middle- and Outer-ring Highways), and sub-urban (SU, outside the Outer-ring Highway) areas (Li et al., 2011). In this study, two urban/rural roads, with similar traffic volume but different road ages, were selected as the research objects. Hutai Road (HT), reconstructed in 1994, is a north-south main road of Shanghai, through the central urban core, developed urban and developing urban. Wuning-Caoan Road (WC) is an east-west main road of Shanghai, through the central urban core, developed urban, developing urban and suburban, which was broadened in 2012. Therefore, both roads reflect the influence of rapid urbanization and industrial relocation on environment in the past two decades.

### 2.2. Sample collection

In this study, 15 and 20 sampling sites (at intervals about 1–2 km) were selected from both sides of HT and WC respectively in May 2016. The sites were mainly located at high traffic volume areas, such as road intersection, gas station and coach station and some sites near park and school were also selected (Fig. 1). Each sampling site represented 2 topsoil samples (0–10 cm) from both sides of road and each sample was a mixture of 4 sub-samples. Therefore a total of 70 topsoil samples were collected from the two roads within a 10 m distance from road edges. Moreover, additional soil samples were collected at distances of 1 m, 2 m, 5 m, 10 m, 20 m, 30 m and 40 m from road edges in a park (121°24'22"E, 31°18'46"N) near sampling site of Hutai Road (HT 11). All samples were collected using a clean stainless steel spade and placed in zip-lock polythene bags (approximately 1 kg). They were then taken back to laboratory immediately and stored at –20 °C prior to further treatment.

In order to investigate the effect of traffic volume on metal content in roadside soils, the number of vehicles was counted and classified using loop counts and video recording at 5 sampling sites of HT located in different areas. The recorded time period was 8: 00–18: 00, 15 min per hour.

### 2.3. Sample preparation and analysis

Samples were freeze-dried with stones and other debris removed. Soil pH and particle size distribution were analyzed on 2 mm sieved samples. The pH of soil samples were measured in deionized water with a 1:2.5 (w:v) ratio (Thomas, 1996). A laser diffraction particle size analyzer (Mastersizer 3000) was used to measure the soil particle size distribution. The samples were further pestled into sizes < 0.15 mm for total organic carbon (TOC) and trace elements analyses. TOC was determined using a total organic carbon analyzer (TOC-VCPN, Shimadzu, Japan) (Angeles Munoz et al., 2015). Total metal concentration was measured on an aliquot (0.2 g) of each sample digested by HCl-HNO<sub>3</sub>-HF-HClO<sub>4</sub> in a Teflon beaker (Chen et al., 2010a). The digestion was diluted to 10 ml using 2% (v:v) HNO<sub>3</sub>, and filtered through nylon membranes with pore size 0.45 μm. The concentrations of Sb, Cu, Zn, Pb, Cd, Ni and Co were determined using Inductively Coupled Plasma-Mass Spectrometry (ICP-MS: Nexlon 300×, Perkin Elmer), and the concentrations of Mn, Zn, Al were determined using Inductively Coupled Plasma-Atomic Emission Spectroscopy (ICP-AES: Agilent720ES,

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