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Heavy metals in the finest size fractions of road-deposited sediments☆

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ABSTRACT

The concentration of heavy metals in urban road-deposited sediments (RDS) can be used as an indicator for environmental pollution. Thus, their occurrence has been studied in whole road dust samples as well as in size fractions obtained by sieving. Because of the limitations of size separation by sieving little information is available about heavy metal concentrations in the road dust size fractions <20 μm. In this study air classification was applied for separation of dust size fractions smaller than 20 μm from RDS collected at different times during the year. The results showed only small seasonal variations in the heavy metals concentrations and size distribution. According to the Geoaccumulation Index the pollution of the road dust samples deceased in the following order: Sb » As > Cu ≈ Zn > Cr > Cd ≈ Pb ≈ Mn > Ni > Co ≈ V. For all heavy metals the concentration was higher in the fine size fractions compared to the coarse size fractions, while the concentration of Sr was size-independent. The enrichment of the heavy metals in the finest size fraction compared to the whole RDS <200 μm was up to 4.5-fold. The size dependence of the concentration decreased in the following order: Co ≈ Cd > Sb > (Cu) ≈ Zn ≈ Pb > As ≈ V » Mn. The approximation of the size dependence of the concentration as a function of the particle size by power functions worked very well. The correlation between particle size and concentration was high for all heavy metals. The increased heavy metals concentrations in the finest size fractions should be considered in the evaluation of the contribution of road dust re-suspension to the heavy metal contamination of atmospheric dust. Thereby, power functions can be used to describe the size dependence of the concentration.

1. Introduction

The composition of urban road-deposited sediments (RDS) is an indicator for environmental pollution. For this reason, numerous studies have been carried out to determine heavy metals contents in RDS. An overview of the concentrations of Cu, Pb and Zn in the RDS of more than fifty cities worldwide was published recently (Hwang et al., 2016). In another review summarizing data from twenty cities in China, the concentrations of Cd, Cr and Ni were additionally included (Wei and Yang, 2010). The number of studies reporting the concentrations of further heavy metals such as As, Co, Hg, Mn, Sb, V, etc. is comparatively small.

The main sources of traffic related particulate matter (PM) emissions are vehicle tailpipe exhausts, wear products from tires, brakes and the road surface and re-suspension of RDS. In sub-arctic regions in the cold season the use of sanding for traction control contributes to the amount of RDS (Kupiainen, 2007). Important factors that influence the PM emissions are the vehicle type, technology and fuel used, maintenance and the speed of the vehicle (Wrobel et al., 2000; Sadler et al., 1996). In a study it was demonstrated that PM10 concentrations in the air strongly correlate with traffic density. During daytime, a considerable amount of PM mass was generated by vehicles. During night-time the PM10 concentration decreased because of reduced traffic. The average PM2.5 and PM1 concentrations showed much less dependence on the traffic flow because of the slower settling of fine particles (Srimuruganandam and Nagendra, 2010).

Besides local industry, traffic emissions contribute significantly to the heavy metal contamination of RDS (Ordonez et al., 2003; Shi et al., 2008). In a study it was found that traffic-generated emissions account for more than 50% of the heavy metals content of airborne PM in urban areas (Wrobel et al., 2000). The highest concentrations of heavy metals in RDS were found in samples from highways, while in downtown areas the concentrations are considerably lower (Shi et al., 2010; Duong and Lee, 2011).

Various heavy metals have been classified as being part of traffic emissions. Brake pads contain Cu, Mn, V and Zn (Apeagyei at al., 2011; Straffelini et al., 2015) and tires contain Zn (Fujiwara et al.,

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2011; Kreider et al., 2010). Wear of brakes and tires releases these components into the environment as fine particles. Zn is also an oil additive (Zn dialkyldithiophosphate) for wear protection (Fujiwara et al., 2011). In a study in Great Britain (Harrison et al., 2012) approximately one third of Zn in atmospheric dust was attributed to automobile emissions. Sb is contained in brake linings and organic Sb compounds are used as additives in grease and oil (Fujiwara et al., 2011). After the phase-out of leaded gasoline, vehicular Pb emissions are now caused mainly from loss of Pb wheel weights, which is considered as the main current source of this element in the urban environment (Fujiwara et al., 2011). Nevertheless, Pb is also present at trace levels as a natural component in un-leaded automotive fuels (Harris and Davidson, 2005). Mn is used as an anti-knock additive (methylcyclopentadienyl manganese tricarbonyl) in motor vehicle fuels (Fujiwara et al., 2011).

In several studies the size dependence of the heavy metal concentrations in RDS has been investigated (Table 1). For this purpose, the collected RDS was separated into various size fractions by dry sieving. However, as the cut size of the separation by dry sieving is limited the finest size fraction in these studies was typically 0–63 µm. Only in two studies was the finest size fraction smaller (0–20 µm and 0–37 µm). These studies have shown that concentrations of heavy metals decrease with increase in particle size. The reasons for this are an increase in the surface area, surface charges, organic matter content, and metal sorption capacity and coatings on the particles (Loganathan et al., 2013). In most studies only the heavy metals Cd, Cu Pb and Zn were investigated. Only limited data are available for other heavy metals (As, Co, Mn, Sb and V). With few exceptions the highest concentrations of the heavy metals Cd, Cu, Pb and Zn were found in the finest RDS fractions. However, because of the limitations of the applied classification method (sieving) the finest size fractions investigated were much bigger than the dust size fractions commonly used in air pollution assessment (PM10 or PM2.5).

Results for the size dependence of the concentrations of Cd, Cr, Cu, Ni, Pb and Zn were also found in some other studies (Duong and Lee, 2011; German and Svensson, 2002; Sutherland, 2003; Viklander, 1998; Bian and Zhu, 2009; Zhao et al., 2010). However, these results could not be included into Table 1 because the data are available in diagrams only. The general trend of higher heavy metal concentrations in the finer fractions was also noticed in these studies. Nevertheless, the finest size fraction investigated in these studies was also significantly bigger then PM10 (0–44 µm or bigger).

In a study it was concluded that re-suspended RDS is one of the most important sources of aerosol pollution in Beijing (Han et al., 2007). In another study a similar conclusion was drawn for Lisbon (Almeida et al., 2006). Re-suspension of RDS can be caused by passing vehicle-induced turbulence and shear stress of the tires or wind (Nicholson and Branson, 1990). The coarser part of PM10 becomes re-suspended more easily than finer material. This can be attributed to the fact that drag force increases faster than adhesive force with increasing particle diameter (Patra et al., 2008). In a

Table 1
Overview of RDS composition by particle size (in mg/kg).

Country	Year	Sample characterization	Size in µm	Cd	Cu	Pb	Zn	As	Co	Mn	Sb	V	Sr	Source
Argentina	2010 (s)	Average for two residential areas	<37	3.4	822	313	1000	5.4		776	11			Fujiwara et al. (2011)
			37–50	1.9	349	326	813	2.6		679	11			
			50–75	1.7	281	235	635	4.6		484	4			
			75–100	2.0	425	320	846	4.4		540	5			
Egypt	2012 (s)	Average residential area and road	<125	0.7		163	435	1.6	5.1			62		Abdel-Latif and Saleh (2012)
			125–200	0.3		103	239	1.1	3.0			30		
Poland	2016 (s)	Urban road dust	<20	1.1	389	789	2565						129	Adamic et al. (2016)
			20–56	0.8	353	743	2267						98	
			56–90	0.6	189	569	1768						95	
			90–250	0.2	103	309	1879						67	
			>250	0.1	89	387	946						89	
Malaysia	2013 (s)	Residential area	<63	0.7	84	88	394			311				Han et al. (2014)
			63–125	0.6	50	40	243			203				
			125–250	0.3	22	32	153			147				
Scotland	1998 –2000 (m)	Average 2 point in residential area	<63	1.7	500	1265	1070							Deletic and Orr (2005)
			63–250	0.6	325	305	345							
			250–500	0.3	150	145	245							
			>500	0.2	55	55	120							
China	1998 –2001 (m)	One sample as mixed sample from 5 sampling points	<63	0.6	72	117	253	12.1						Han et al. (2008)
			63–80	0.6	88	141	286	12						
			80–88	0.6	97	149	289	12.3						
			88–101	0.6	102	154	296	11.6						
			101–121	0.6	97	153	275	11.7						
			121–152	0.6	71	150	268	11.2						
			152–196	0.5	61	133	223	9.4						
			196–357	0.3	87	135	203	9.06						
			357–681	0.3	41	95	157	7.62						
			681–920	0.3	40	84	157	7.95						
Korea	2006 (m)		<75	2.3	345	223	1271							Duong et al. (2006)
			75–180	1.0	236	118	752							
			180–850	0.5	139	57	430							
			850–2000	0.3	265	52	258							
Spain	2010 (s)	Average for two residential zones	<63	38	124	350	630		51	374				Zafra et al. (2011)
			63–125	23	91	280	399		31	285				
			125–250	19	104	273	309		26	285				
			250–500	22	141	297	268		33	292				
			500–1000	11	47	210	139		20	170				
			1000–2000	8	36	154	83		14	100				

(s) Year paper submitted to journal, date of RDS sampling not reported; (m) Year RDS sampling performed.