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Spatial distribution patterns of four traffic-emitted heavy metals in urban road dust and the resuspension of brake-emitted particles: Findings of a field study

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ABSTRACT

This paper presents the results of a field campaign that was aimed to assess the total amount and the spatial variation in the cross-sectional surface loads of four selected heavy metals (Zn, Cu, Pb and Sb) at three different locations of a downtown street at Budapest, Hungary. The distribution of surface loads were found to be determined by their prevailing emission source: brake wear dominated metals show distinctive cross-sectional gradients (depending on traffic dynamics) and are only above the background values at the curbside, while tire wear dominated ones feature only a weak gradient and are consistently higher than the background values, over the entire road surface. The relative strength of post-precipitation resuspension was further investigated in the case of principally airborne brake-emitted particles by studying the changes in the ratios of selected metals in dry and wet roadside dust samples, in comparison with the typical values for urban road runoff. The results suggest that roughly 70% of brake-emitted metals that are left behind by the curb after a rainfall event will return into the atmosphere by resuspension as the wet dust dries up. The improved insight into this process has important implications on the effectiveness of stormwater treatment technologies.

1. Introduction and aims

Nowadays a substantial portion of anthropogenic heavy metal emissions, especially from diffuse (non-point) sources, originates from road transportation. Traffic-borne heavy metals, emitted mainly as solid particles, reach all three environmental compartments, causing a complex air, water and soil pollution problem (Wåhlin et al., 2009; Salma and Maenhaut, 2006; Councell et al., 2004; Bergbäck et al., 2001; Boller, 1997). Due to their persistency, these contaminants tend to accumulate in their final recipients: usually in the topsoil and in the sediment of surface waters, but also in sewage sludge (in the case of urban areas served by combined sewer systems). A number of studies have warned that traffic-emitted heavy metals also accumulate in the biosphere and might have adverse effects on the health of humans, as well as other organisms (Quiroz et al., 2009; Sandahl et al., 2007; Zechmeister et al., 2006; von Üexküll et al., 2005; Councell et al., 2004).

Road traffic is emitting a range of heavy metals, among which Cu and Zn require attention because of their quantity, while some others, like Pb and Sb, despite of being emitted in lower levels, are of concern because of their elevated health risks. Although in varying quantities, Cu and Sb emissions are known to originate almost entirely from brake wear (which is also a dominant source of Pb); while Zn is emitted mainly by tire wear, yet in lower quantities it can also be found in brake linings and thus, their wear products

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as well (Hulskotte et al., 2014; Thorpe and Harrison, 2008; Hjortenkrans et al., 2007). These two abrasion processes produce solid particles that range in size over at least three orders of magnitude, which strongly influences the transport pathways of the associated heavy metals. Brake wear is characterized by particles spanning in diameter from submicron sizes up to a few microns (Kukutschová et al., 2011; Wahlström et al., 2010; Sanders et al., 2003), which settle out from the atmosphere rather slowly, spreading and depositing over extended areas. Compared to the former, tire wear emits larger particles, typically reaching several tens of micrometers or even more, which tend to remain rather close to their source (Thorpe and Harrison, 2008). The budget of heavy metals deposited on the road is further influenced by wet deposition during precipitation events and resuspension by vehicle and wind induced turbulence in dry periods: the former increases the flux of airborne particles falling out on the surface, while the latter acts in the opposite direction (Gunawardena et al., 2013; Amato et al., 2013; Vaze and Chiew, 2002). The attachment of some particles to various solid surfaces in the environment (vehicle parts, road surface, roadside buildings, vegetation) and the spraying and splashing of stormwater by the moving vehicles during the precipitation events complicate the transport processes of traffic-emitted heavy metals even more (Schipper et al., 2007; Sanders et al., 2003).

Several studies have investigated the particle size distribution and the heavy metal content of roadside dust (Zafra et al., 2011; Fujiwara et al., 2011; Brown and Peake, 2006; Herngren et al., 2006; Deletic and Orr, 2005; Lau and Stenstrom, 2005; Lee et al., 2005; Zanders, 2005), as well as the accumulation of traffic-generated heavy metals in roadside topsoil (De Silva et al., 2016; Guney et al., 2010; Hjortenkrans et al., 2008; Viard et al., 2004; van Bohemen and van de Laak, 2003; Sutherland and Tolosa, 2001) for different geographical locations and adjacent land use categories. However, there is still little quantitative information available about the deposition and resuspension fluxes of particular traffic-related heavy metals. These dynamic processes are key factors in determining the partitioning of loads between the different environmental compartments, and therefore, they need to be paid more attention in order to estimate atmospheric and surface loads more precisely.

In this paper, we first present the results of a measurement program that was aimed to characterize the total amount and the spatial variation in the cross-sectional surface loads of four selected heavy metals (Zn, Cu, Pb and Sb) at an average downtown street of Budapest, Hungary. This is followed by an analysis of the ratios of metals specific to brake wear (Cu, Sb) and tire wear (Zn) in dry and wet roadside dust samples, and their comparison with the value of the same ratios measured in the urban road runoff of the same city. The outcomes provide an estimation about the scale of post-precipitation resuspension of traffic-emitted heavy metals associated with brake wear particles.

2. Materials and methods

Road dust particles were sampled from the surface of a 2 × 1 lane downtown street, sided by tall buildings. Such an "urban canyon" was favorable for this study, as the buildings shield the road effectively from natural winds perpendicular to the street. The road is bordered by a 10 cm high curb on both sides, allowing particles to accumulate within an approximately 0.5–1 m wide strip by the curb, a phenomenon that is well known from the literature (Deletic and Orr, 2005; Sartor and Boyd, 1972). The average daytime hourly traffic at the studied sections of the street varies between 500 and 600 vehicles per lane, consisting predominantly of light vehicles.

Three sampling sites were selected along the road (Fig. 1-a). These locations (S1, S2 and S3) are fairly close to each other, yet they are characterized by different traffic dynamics. S1 lies three meters ahead of a junction equipped with traffic lights, where vehicles have to stop in 40% of the time. S2 lies likewise ahead of a pedestrian crossing, without traffic lights, where vehicles have to yield for pedestrians. Due to the stochastic nature of crossings, it is hard to estimate the frequency of braking. However, it can be assumed that even if there might be less braking at this place compared to S1, the number of sudden and strong decelerations is higher. At S3, the traffic flow is much more even and less disturbed than at the other two locations. Furthermore, this section of the street has a smoother surface compared to S1 and S2, offering less room for particle accumulation within the road texture. In addition to the street sites, a set of samples were obtained from two nearby impervious surfaces located within a park (BG). As these places are not exposed to traffic, they were utilized to represent the urban background dry deposition, and served as a comparison base for the loads that were measured on the road surface.

Each time, three samples were obtained from each of the three sampling sites in order to examine the variation of surface loads along the cross-section of the road (Fig. 1-b). The first sampling point (A) was taken from the edge (0.2 m away from the curb), which has very little direct interaction with the moving vehicles. This point represented the narrow roadside strip, where the majority of the particles accumulate. The remaining two sampling points (B and C) were located 1 and 1.7 m from the curb, respectively, both experiencing frequent contact with the moving vehicles. The latter two points represented areas of the road surface where traffic-induced turbulent air movements are dominating over the dry deposition processes.

Sampling was executed with a portable high-pressure washer device, whose piston was fitted into a rigid, sealed rubber dome used for fencing off the circle-shaped sampling area, measuring 14 cm in diameter. The washing water that detached and suspended the surface dust particles, was continuously drawn out from the enclosed area through a perforated tube torus using a vacuum pump, and collected in glass flasks (Fig. 1-c). The advantage of this novel method over other commonly utilized techniques, such as hoovering or sprinkling, is the improved efficiency in terms of pollutant mobilization. While the usual methods rather mimic particle wash-off that is caused by rainfall runoff alone, the high-pressure washer also mimics the effects of the intensive pressure and suction forces exerted by the rolling vehicle wheels during wet weather. High-pressure washing thus provides more relevant information about the potentially removable pollutant amounts under real-life circumstances.

The study lasted over one month, with paying attention to avoid taking consecutive samples from the very same spots. For checking the uncertainties, two batches of longitudinal sample series (each one consisting of five samples taken in an even layout