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# Contribution of coarse particles from road surfaces to dissolved and particle-bound heavy metal loads in runoff: A laboratory leaching study with synthetic stormwater

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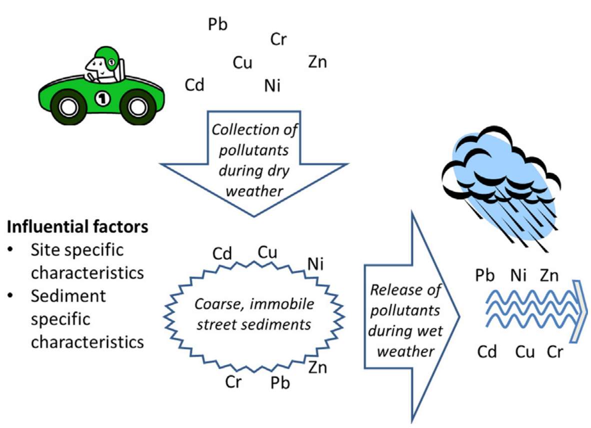
## HIGHLIGHTS

- Coarse street sediments released fine particles in experiments simulating runoff.
- Release of particulate bound heavy metals increased with organic matter content.
- Street cleaning affected releases of heavy metals from coarse street sediments.

## GRAPHICAL ABSTRACT

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## ABSTRACT

Laboratory leaching experiments were performed to study the potential of coarse street sediments (i.e. >250 μm) to release dissolved and particulate-bound heavy metals (i.e. Cd, Cr, Cu, Ni, Pb and Zn) during rainfall/runoff. Towards this end, street sediments were sampled by vacuuming at seven sites in five Swedish cities and the collected sediments were characterized with respect to their physical and chemical properties. In the laboratory, the sediments were combined with synthetic rainwater and subject to agitation by a shaker mimicking particle motion during transport by runoff from street surfaces. As a result of such action, coarse street sediments were found to release significant amounts of heavy metals, which were predominantly (up to 99%) in the particulate bound phase. Thus, in dry weather, coarse street sediments functioned as collectors of fine particles with attached heavy metals, but in wet weather, metal burdens were released by rainfall/runoff processes. The magnitude of such releases depended on the site characteristics (i.e. street cleaning and traffic intensity), particle properties (i.e. organic matter content), and runoff characteristics (pH, and the duration of, and energy input into, sediment/water agitation). The study findings suggest that street cleaning, which preferentially removes coarser sediments, may produce additional environmental benefits by also removing fine contaminated particles attached to coarser materials.



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## 1. Introduction

Urban street and highway runoff contains high levels of pollutants, such as heavy metals, nutrients and trace organics (Kayhanian et al., 2012), and is regarded as a very significant diffuse source of pollution in the urban environment (Gobel et al., 2007; Opher and Friedler, 2010). Much of the heavy metal content of street runoff originates from vehicular traffic, and sources of individual metals (i.e. Cu, Zn, Pb, Cr, Cd and Ni) can be traced to specific components of vehicles, or their operation (brake pad abrasion, tire wear, body corrosion, and pavement marking and abrasion). While some of the pollutants emitted by traffic are dispersed in the atmosphere, large quantities accumulate on street surfaces and are subsequently washed off and transported by stormwater runoff to the receiving waters, where they may cause environmental problems (US EPA, 1983; Legret and Pagotto, 1999; Crabtree et al., 2008).

Particles collected from street surfaces usually contain elevated concentrations of heavy metals (Zafra et al., 2011; Loganathan et al., 2013; Huber et al., 2016), so their presence in street runoff increases its heavy metal content. The environmental effects of these metals depend on several factors. For example, the nature of their binding to the sediments determines how they are released under various ambient conditions, which ultimately determines their bioavailability (Stone and Marsalek, 1996; Sutherland et al., 2012; Zhang et al., 2015). It has also been shown that the sediment particle size is important; smaller particles typically contain higher metal concentrations, as shown e.g. by Viklander (1998), who reported the highest metal concentrations (with few exceptions) in street particles smaller than 75 μm. However, coarser particles with smaller metal concentrations account for a much greater proportion of the total street sediment mass and, therefore, contain high proportion of the total metal burden in sediment particles. For example, Sansalone and Ying (2008) reported that 90% of the total metal mass was associated with coarser particles >75 μm. However, coarse particles may not be washed off street surfaces or may settle out quickly (Andral et al., 1999).

In spite of their impaired transport, coarse particles can contribute to the heavy metal loads in stormwater runoff by collecting pollutants via dry deposition and adsorption of exhaust emissions during dry periods, and the release of such accumulations during rain events. Experimental evidence of such processes was provided by Sansalone and Ying (2008) who performed metal leaching with rainwater (pH 3–7) on sediments from trafficked areas and concluded that coarse particles (>250 μm) contributed between 45 and 80% of the total leached metal mass. However, those tests were performed on particles collected by passive samplers placed adjacent to street surfaces; such samples are likely to differ from street sediments. Other authors, e.g. Gunawardana et al. (2015), applied leaching tests to street sediments and noted that heavy metals (i.e. Zn, Cu, Pb and Ni) were preferentially adsorbed to Fe, Al and Mn oxides. Individual metals exhibited differing levels of mobility in the desorption tests, with Zn having the greatest likelihood of release. Similarly, Joshi et al. (2009) conducted metal dissolution tests on street sediments using various kinds of leaching media (a riverine water, deionized water, and acidified deionized water at pH 2). It was concluded that metal dissolution was enhanced by reducing the pH and the properties of the riverine water. A limitation of all the above studies is that they only considered release of dissolved heavy metals; no studies have yet explored the possibility that coarse particles may also release particulate bound metals during wet weather. Viklander (1998) provided some evidence for the occurrence of such a process by comparing the particle size distributions (PSD) of street sediments determined by dry and wet sieving. The wet sieving yielded a finer PSD than the dry sieving, which was explained by suggesting that the collected sediments featured aggregates of smaller and larger particles that became separated under the wet conditions. Therefore, it seems necessary to consider both dissolved and particle-bound heavy metals when assessing the contribution of coarse particles to the heavy metal content of stormwater runoff.

To advance the understanding of the role of coarser street sediments in heavy metal transport, leaching experiments were performed on coarse sediments collected from various urban streets and the release of dissolved and particle-bound heavy metal loads, during rainfall-runoff events, was assessed. A secondary objective was to identify the factors that may influence the release of heavy metals from coarse particles.

## 2. Materials and methods

Street sediments were sampled at seven sites in five Swedish cities and analyzed for various physical and chemical parameters. After discarding the fine fractions (≤250 μm), laboratory leaching experiments were performed to determine heavy metals release from the coarse fraction exposed to synthetic rainwater under various ambient conditions occurring on urban streets during wet weather. Finally, statistical analyses were applied to determine the influence of ambient conditions on heavy metals release.

### 2.1. Study sites

Seven street sites in five Swedish cities were sampled between August 2013 and August 2015. All sites were two-lane streets with curbs, but differed with respect to the surrounding land use, street type, average daily traffic (ADT), and street cleaning practices. The samples also differed with respect to the antecedent dry period (ADP) before the sample collection. The site characteristics are presented in Table 1.

### 2.2. Sediment sampling

Sampling was done along the curb lengths of 3 to 5 m, depending on the allowed working time for the site. The width of the sampled area was 1.5 m from the curb towards the street crown. Sediments were collected with a domestic vacuum cleaner (Dyson DC-51), which applies cyclonic separation to remove the particles from the air stream. The minimum diameter of the recovered particles was 3 μm, as determined by laser diffraction. Vacuum cleaning was repeated multiple times, in perpendicular directions, so that near-complete collection of the sediments could be assumed. Such sampling techniques were tested previously and shown to achieve sampling efficiencies >90% (Ball et al., 1998; Egodawatta, 2007). To prevent contamination of samples by the vacuum cleaner, an adjacent section of the curb was vacuumed first and the collected sediments were discarded prior to sampling. This was done twice. The collected samples were dried overnight in the laboratory at room temperature and stored in a cold and dark environment.

**Table 1**
Characteristics of the sampled street sites.

| City | Malmö | | Stockholm | | Kiruna | Luleå | Umeå |
|---|---|---|---|---|---|---|---|
| Designation | A | B | C | D | E | F | G |
| Land use[a] | R, C | R | C | R | C, I | C, I | C |
| Street type | Arterial street | Main street | Main street | Arterial street | Main street | Arterial street | Arterial street |
| Month | May 2015 | May 2015 | Apr 2015 | Apr 2015 | Aug 2015 | Aug 2013 | Jun 2015 |
| ADT[b] | 10600[c] | 7800[c] | 15000[d] | 5900[d] | 5800[e] | 6000[e] | 7700[e] |
| Heavy traffic [%] | 11[c] | 10[c] | 9[d] | 13[d] | 9[e] | 8[e] | 5[e] |
| ADP [days] | 3[f] | 3[f] | 10[f] | 10[f] | 8 | 10 | 11 |

[a] Surrounding land use: R = residential, C = commercial, I = industrial.
[b] The traffic loads were measured in one flow direction (vehicles/day).
[c] Source: malmo.se (accessed August 2015).
[d] Source: Traffic Office Stockholm (personal communication, April 2015).
[e] Source: http://vtr.trafikia.se (accessed August 2015).
[f] Source: 6 days since last street cleaning.