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Legacy (Pb, Cu, Zn) and emerging (Sb) trace metal contaminants in the road - highway stormwater pond continuum: a focus on geochemistry and speciation of antimony

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In urbanized watersheds of the Paris area, an increase in antimony (Sb) concentrations in suspended particulate matters is observed along an increasing urbanization gradient, from 2 to 5 $mg.kg^{-1}$, indicating that urban environment suffers from Sb accumulation [1]. A main hypothesis is that antimony is emitted by traffic through brake lining [2]. However, the biogeochemistry of Sb along the continuum between the highway and stormwater ponds is unknown. Speciation and isotopic tools may provide useful information to elucidate the Sb cycle in this type of environment.

Here, three highway stormwater ponds of different ages/environments are studied within the aim to document the road-derived Sb contamination. Lead isotopic analyses allow to classify the ponds as (1) semi-urban, with lead isotopic signature around the urban signature [3],[4] (sewer particles 2.107-2.109 $^{208}Pb/^{206}Pb$, 1.1542-1.1566 $^{206}Pb/^{207}Pb$; waste incinerators 2.106-2.107 $^{208}Pb/^{206}Pb$, 1.1540-1.1550 $^{206}Pb/^{207}Pb$) and (2) rural, with lead isotopic signatures dispersed between pre-2000 leaded-gasoline (2.18 $^{208}Pb/^{206}Pb$, 1.08 $^{206}Pb/^{207}Pb$) and the local geochemical background endmembers (2.049-2.053 $^{208}Pb/^{206}Pb$, 1.2007-1.2087 $^{206}Pb/^{207}Pb$). We observe that enrichment factors (EF) for road-related trace metals (Pb, Cu, Zn) were 4 to 6 times higher for semi-urban ponds than for rural ponds. Traffic $\delta^{123}Sb$ signatures of road-contaminated samples range between 0.02‰ and 0.11‰, which allows to hypothesize diverse geological/material sources for Sb but also that there is no unique road-source $\delta^{123}Sb$ signature in the study area. Fe-(oxyhydr)oxides containing traces of S, Cu, Zn, and Ba are observed as discrete carrier phases by SEM-EDX, which points to the Sb brake pad source. X-Ray-Absorption-Near-Edge-Structure spectroscopy (XANES) data obtained at the Sb K-edge show a speciation evolution between samples collected as dry deposits in the road-to-pond continuum and those collected under water. Dry samples show mostly Sb(V)-O (44-100%) and Sb(III)-O species (0-56%) while underwater samples contain mostly Sb(III)-S (48-81%) and Sb(V)-O species (19-52%), showing that road-derived (or -sourced) Sb is highly sensitive to variations in the local redox conditions, which could impact its mobility and $\delta^{123}Sb$ isotopic signature in urban surface environments.

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[**3**]Ayrault *et al.*(2014) *Environ.Sci.Pollut.Res. 21*,4134-4148]
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