

Isotopic (Cu and Zn) and elemental fingerprints for tracking anthropogenic sources in urbanized coastal environments

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Non-exhaust emission (NEE) sources, such as road dust, brake pads, tires, asphalts, curbs, and road/antifouling paints, are considered significant metal contamination sources in urban environments due to their high levels of harmful metals. These contaminants are discharged into the coastal environment through rivers, sewers, atmospheric deposition, and urban runoff, and also affect surrounding biological organisms. Metal stable isotopes, such as Cu and Zn, have been utilized in environmental forensic research to offer objective scientific evidence and trace pollution sources in coastal environments. Access to isotope data for anthropogenic contaminants is crucial since most countries import and utilize both raw materials and final products. Here, this work presents isotopic (Cu and Zn) and elemental fingerprints of common urban sources and their potential use for tracking anthropogenic emissions in sediment, biota, coastal environments (Korea, Brazil, and Europe). Combining isotopic compositions with the elemental ratio (Zn/Cu) of anthropogenic contaminants enables the distinction of different sources. Total suspended solids in rainfall runoff ($\delta^{65}\text{Cu}_{\text{AE647}}$: +0.01‰ and $\delta^{66}\text{Zn}_{\text{IRMM3702}}$: -0.11‰) are isotopically similar to those of road dust, particularly for fine particle size fractions (<63 μm ; $\delta^{65}\text{Cu}_{\text{AE647}}$: +0.03‰ and $\delta^{66}\text{Zn}_{\text{IRMM3702}}$: -0.13‰). Antifouling paints were heavier Cu ($\delta^{65}\text{Cu}_{\text{AE647}}$: -0.16 to +0.36‰) and lighter Zn ($\delta^{66}\text{Zn}_{\text{IRMM3702}}$: -0.34 to +0.03‰) isotopic compositions compared to other NEE sources. Urban runoff (e.g., road dust) affects most metal contents and bulk isotope signatures of surface sediments in a highly urbanized Korean coastal area, while antifouling paints do not appear to be the primary sources of these metals. Isotope ratios of biomonitoring organisms vary geographically suggesting different bioaccumulation gradients of anthropogenic Cu and Zn. Our study demonstrates that elemental and isotope fingerprints of various NEE sources can help improve anthropogenic source tracking in natural archives/environmental media of coastal environments.