Goldschmidt 2021 Abstract
https://doi.org/10.7185/gold2021.6818

A top-down emissions estimation in the Boston urban region suggests an underestimation of small point and/or non-point mercury emissions

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Mercury (Hg) is an environmental toxicant dangerous to human health and the environment. Anthropogenic emissions are regulated by global, regional, and local policies. Here, we report atmospheric Hg concentrations from August 2017 to April 2019 and investigate Hg sources in the coastal city of Boston, the third largest metropolitan area in the Northeastern United States. Ambient Hg concentrations (median of 1.37 ng/m3) were at the low end of the range reported in the Northern Hemisphere likely due to aggressive emission reductions under a regional Hg action plan. Despite relatively low ambient concentrations, we calculate anthropogenic emissions to be 3-7 times higher than in current emission inventories using a measurement-model framework, suggesting an underestimation of small point and/or non-point emissions. Legacy emissions accounting for ~60% of Hg emitted to the atmosphere annually and much of this recycling taking place through the ocean, we also test the hypothesis that a legacy Hg source from the nearby ocean contributes to atmospheric Hg concentrations in the region. We find that elevated concentrations observed during easterly oceanic winds can be fully explained by low wind speeds and recirculating air allowing for accumulation of land-based emissions. This result suggests a relatively low contribution of oceanic evasion to atmospheric concentrations in Boston. Our study further highlights the need for top-down approaches to better constrain urban emissions.