Goldschmidt 2023 Abstract
https://doi.org/10.7185/gold2023.17245

Stable mercury isotope revealing the sources of dissolved gaseous mercury in lake water

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We present the first measurements of the isotopic composition of dissolved gaseous mercury (DGM) at the three lakes (HFL (Hongfeng lake, Guizhou province); HGL (Hugangyan maar lake, Guangdong province); Nam Co, (Nam Co Lake, Tibet)). The surface DGM samples at the three lakes exhibit significantly negative δ^{202}Hg values (means = -1.01 to -0.43‰, n = 3) and slightly negative Δ^{199}Hg values (means = -0.14 to -0.07‰, n = 3). We demonstrate that production of DGM at lakes could lead an enrichment light isotope, as well as enrichment odd isotopes of atmospheric Hg(0). The odd-MIF signatures of DGM were consistent across HFL, HGL and Nam Co, with Δ^{199}Hg/Δ^{201}Hg slope of 1.07, 1.19 and 1.13, respectively, consistent with photoreduction of aqueous Hg(II), highlighting the important of photoreduction mechanisms in controlling the odd-MIF isotope composition of water surfaces DGM production. The mean Δ^{200}Hg of DGM displayed positive values at Nam Co (0.08‰) and near-zero values at HFL and HGL (-0.01 to 0.00‰, n = 2). Nam Co DGM samples exhibits even-MIF signatures (Δ^{200}Hg) generally consistent with our observations of water Hg(II) (mean = 0.08‰, n = 9), while HFL and HGL were lower than Hg(II) (means = 0.05 and 0.13‰, n = 2) but were higher than values typically observed of the atmospheric Hg(0) (means = -0.02 and -0.04‰, n = 2). We therefore use a Δ^{200}Hg mass balance to estimate that water Hg(0) uptake contributed 74.8% and 81.0%, while aqueous Hg(II) photoreduction contributed 25.2% and 19% to production of DGM at HFL and HGL, respectively. Photochemical process is main driver for DGM production at Nam Co, since its highest elevation and low Hg(0) deposition flux. The DGM Δ^{200}Hg composition suggests that current understanding for the source of DGM is incomplete. This indicates that photochemical Hg(0) production in water surfaces is overestimated, and atmospheric Hg(0) deposition indeed play an important role in Hg cycling between atmosphere and water bodies.