

Hg isotopes in gaseous and particulate atmospheric mercury species above a coastal suburban environment (Pensacola, Florida, USA)

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The fate of mercury in the atmosphere is controlled by multiple physical, (photo)chemical and meteorological processes. Assessing Hg cycling in the atmosphere via the use of its stable isotopes remains complicated due to the high reactivity of this element and the Hg isotope fractionation that has thus far been reported. In this study we measured the Hg isotopic composition of gaseous elemental Hg ($\text{Hg}^0_{(g)}$, GEM), reactive gaseous Hg ($\text{Hg}^{\text{II}}_{(g)}$, RGM) and particulate aerosol Hg ($\text{Hg}^{\text{II}}_{(p)}$) in the atmosphere above a coastal area located near the city of Pensacola, Florida. Gold-coated quartz sand traps (1 LPM) were used to collect total gaseous Hg. Aerosol Hg was collected on pre-baked quartz fiber (Whatman QMA) filters deployed in a Tisch Environmental high-volume aerosol sampler (~1100 LPM). Reactive gaseous Hg was collected on a QMA filter that had been soaked in a KCl solution (then dried and pre-baked) that was deployed underneath the aerosol QMA filter. These sampling methods collected from 1 to 30 ng of the different atmospheric Hg species after 1 to 3 days of deployment during the summer 2012. Both mass-dependent (MDF) and mass-independent (MIF) signatures of GEM were significantly different than those of Hg(II) species. GEM, which represented more than 99% of the total Hg at this site, displayed positive $\delta^{202}\text{Hg}$ values (0.7 to 1.2‰, average $1.0\pm 0.2\%$, $n=14$) whereas both RGM and Hg(p) had negative $\delta^{202}\text{Hg}$ values (-2.4 to -0.1‰, $-0.7\pm 0.4\%$, $n=34$). On the other hand, GEM ($n=14$) had slightly negative $\Delta^{199}\text{Hg}$ and $\Delta^{200}\text{Hg}$ average values ($-0.26\pm 0.09\%$ and $-0.07\pm 0.03\%$, respectively) whereas Hg(p) ($n=17$) exhibited slightly positive values ($0.33\pm 0.13\%$ and $0.10\pm 0.04\%$, respectively). RGM displayed no significant MIF ($\Delta^{199}\text{Hg}=0.02\pm 0.14\%$ and $\Delta^{200}\text{Hg}=-0.02\pm 0.10\%$, respectively) of both even and odd isotopes. We suggest that both oxidation and reduction processes are responsible for the observed patterns in the isotopic composition of RGM and Hg(p). This suggests that preferential scavenging of RGM and Hg(p) (rather than GEM) in wet and dry deposition may result in significant variability in the isotopic fractionation of Hg that is deposited in coastal environments as a function of seasonal and sporadic changes in air mass trajectories and atmospheric physico-chemistry.

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