Isotopic compositions of atmospheric total gaseous mercury in ten Chinese cities and implications for land surface emissions

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Total gaseous mercury(TGM) concentrations and isotopic compositions were measured in ten Chinese cities and one rural site in summer and winter 2018. Mean TGM concentrations at the urban sites during the study periods ranged from 2.34 to 4.56 $ng m^{-3}$ (n = 10) with a mean (±1 σ) of 3.08 ± 0.79 ng m⁻³ (Figure 1). Large variations in daily TGM isotopic composition were observed with value ranging from -1.68 to 0.63% for δ^{202} Hg and from -0.23 to 0.10% for Δ 199Hg. The mean TGM concentrations and Δ^{199} Hg values were relatively higher in summer than winter in most cities except for the two (Guiyang and Guangzhou) in the low latitudes that showed an opposite trend. Soil emissions are potentially an important source of atmospheric TGM in urban areas. The mean δ^{202} Hg and Δ^{199} Hg values of GEM emitted from soils were -2.16 \pm 0.60‰ and -0.27 \pm 0.15‰ (1 σ , n = 5), respectively, in Guiyang, and were -1.07 \pm 0.86‰ and -0.01 \pm 0.52‰ (1 σ , n = 3), respectively, in Wuhan. These values suggest that the isotopic compositions of soil GEM emissions in urban areas of China likely have highly negative δ^{202} Hg values. Thus, high surface GEM emissions should shift TGM δ^{202} Hg towards negative values. A weak positive correlation was observed between mean TGM Δ^{199} Hg and simulated soil GEM emission fluxes (ANOVA, $R^2 = 0.21$, p < 0.05), suggesting that high surface GEM emissions led to a slightly positive shift of TGM Δ^{199} Hg. Seasonal amplitudes of δ^{202} Hg (or Δ^{199} Hg) in the ten cities were significantly negatively (or positively) correlated with seasonal amplitudes of simulated soil GEM emission flux (ANOVA, R^2 of 0.54 or 0.63, p < 0.01 for both, Figure 2), suggesting the dominant role of surface GEM emissions on the seasonal variations in TGM isotopic compositions.



