

Sequential samples revealing significant variation of mercury isotopes during single rainfall events

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Previous studies have reported both mass-dependent fractionation (MDF) and mass-independent fractionation of odd and even Hg isotopes (odd-MIF and even-MIF) in natural samples, and demonstrated the potential of Hg isotopes in investigating atmospheric Hg sources, transport and deposition. Prior studies on Hg isotopic compositions of the precipitation samples (collected in weeks to months) from the North America and China largely improved our knowledge on the sources and transformation of Hg in the atmosphere. To be noted, these samples were all integrated samples collected from at least one entire precipitation or from multiple precipitation events, due to the low Hg concentration. This would obscure some key and detailed information about physiochemical transformation and dynamic deposition of Hg (and its isotopes) encountered in the intra precipitation event.

In this study, the sequential rain samples were collected in three single rainfall events in Guiyang, China, and Hg isotope compositions were analyzed, together with other geochemical parameters, for both the “dissolved” (Hg_D) and for the first time the particulate phases (Hg_{PM}) of precipitation. For all samples, the total Hg concentration and also Hg_D decreased progressively from the beginning to the end of each rainfall event. The acquired Hg isotopic composition of high temporal resolution in single events showed large variation of both MDF and MIF and was consistent with those previously reported for the same region. Our data suggested that the large variation was triggered by variable contribution from different sources, due to the change of the meteorological condition, rather than the atmospheric processes. While the Hg_D was primarily a mixture of local sources and long-range transport Hg, Hg_{PM} with relatively lower odd-MIF was inclined to be derived from the local anthropogenic emissions. Moreover, our study demonstrated that Hg isotopic composition in precipitation was very sensitive to the feedback of momentous anthropogenic activities, as demonstrated for examples by the rapid response of MDF and odd-MIF to the incense burning in Tomb Sweeping Day.