Towards an advanced description and modelling of the atmospheric multiphase chemistry of mercury: CAPRAM HG module 1.0

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Mercury is a neurotoxic element emitted predominantly in its less-reactive form as gaseous elemental mercury (GEM) into the atmosphere by various natural and anthropogenic processes. Once emitted it undergoes chemical processing in the atmospheric gas and aqueous phase. There, GEM is oxidised into gaseous oxidised mercury (GOM), which partitions into aerosol particles residing there as particulate bounded mercury (PBM) due to its much higher solubility. The faster deposition of GOM and PBM compared to GEM is of special environmental importance, because they can be converted into more toxic organic mercury in aquatic environments and then take serious place in the food web. Thus, it is crucial for models to understand the transformation of GEM into GOM and PBM and vice versa. To date, numerous gas-phase chemistry simulations were performed, but reveal missing oxidation and reduction processes. However, only few models exist that investigate the multiphase mercury chemistry in a detailed manner.

Therefore, a comprehensive multiphase mercury chemistry mechanism, the CAPRAM HG module 1.0 (CAPRAM-HG1.0), has been developed. The HG1.0 includes 74 gas-phase reactions, 22 phase transfers and 77 aqueous-phase reactions. It was coupled to the multiphase chemistry mechanism MCMv3.2/CAPRAM4.0 and the extended CAPRAM halogen module 3.0 (CAPRAM-HM3.0) for investigations of multiphase Hg redox under Chinese polluted conditions. Simulations were performed for summer conditions in 2014 using the air parcel model SPACCIM to investigate the performance of the model to simulate typical concentrations and patterns of GEM, GOM and PBM.

Under non-cloud conditions, model results reveal good coincides with concentrations and patterns for GEM, GOM and PBM measured in China. However, the simulations also show that there are still high uncertainties in atmospheric mercury chemistry. Especially, the complexation with HULIS within aerosol particles needs evaluation as the simulations indicate this process as key process driving concentrations and patterns of both GOM and PBM. Further, the present study demonstrates the need of a better understanding of continental concentrations of reactive halogen species and particle bounded halides as well as their link to the multiphase chemistry and atmospheric cycling of mercury.