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A numerical modelling study on regional mercury budget for eastern North America

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Abstract. In this study, we have integrated an up-to-date physio-chemical transformation mechanism of Hg into the framework of US EPA's CMAQ model system. In addition, the model adapted detailed calculations of the air-surface exchange for Hg to properly describe Hg re-emissions and dry deposition from and to natural surfaces. The mechanism covers Hg in three categories, elemental Hg (Hg^0), reactive gaseous Hg (RGM) and particulate Hg (HgP). With interfacing to MM5 (meteorology processor) and SMOKE (emission processor), we applied the model to a 4-week period in June/July 1995 on a domain covering most of eastern North America. Results indicate that the model simulates reasonably well the levels of total gaseous Hg (TGM) and the specific Hg wet deposition measurements made by the Hg deposition network (MDN). Moreover, results from various scenario runs reveal that the Hg system behaves in a closely linear way in terms of contributions from different source categories, i.e. anthropogenic emissions, natural re-emissions and background. Analyses of the scenario results suggest that 37% of anthropogenically emitted Hg was deposited back in the model domain with 5155 kg of anthropogenic Hg moving out of the domain during the simulation period. Overall, the domain served as a net source, which supplied ~a half ton of Hg to the global background pool over the period. Our model validation and a sensitivity test further rationalized the rate constant for gaseous oxidation of Hg^0 by hydroxyl radical OH used in the global scale modelling study by Bergan and Rodhe (2001). A further laboratory determination of the reaction rate constant, including its temperature dependence, stands as one of the important issues critical to improving our knowledge on the budget and cycling of Hg.

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