Geophysical Research Abstracts
Vol. 19, EGU2017-5085-1, 2017
EGU General Assembly 2017

Measurement-based modeling of daytime and nighttime oxidation of atmospheric mercury

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Accurate characterization of gaseous elemental mercury (GEM) chemical oxidation pathways and their kinetics is critically important for assessing the transfer of atmospheric mercury to bioaquatic systems. Recent comprehensive field measurements have suggested that the nitrate radical (NO_3) plays a role in efficient nighttime oxidation of GEM, and that the role of the hydroxyl radical (OH) as a GEM oxidant has been underestimated. We used the CAABA/MECCA chemical box model and additional kinetic calculations to analyze these measurement results, in order to investigate the nighttime and daytime oxidation of GEM. We assumed a second-order reaction for the NO_3 induced nighttime oxidation of GEM. Our analysis demonstrated that nighttime oxidation of GEM has to be included in the model to account for the measured variations in nighttime reactive gaseous mercury (RGM) concentration. A lower limit and best-fit rate constant for GEM nighttime oxidation are provided. To the best of our knowledge, this is the first time that a rate for nighttime oxidation of GEM has been determined based on field measurements. Our analysis further indicates that OH has a much more important role in GEM oxidation than commonly considered. A lower-limit rate constant for the OH–RGM reaction is provided.