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Parametric sensitivity and uncertainty analysis of dimethylsulfide oxidation in the clear-sky remote marine boundary layer

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Abstract. Local and global sensitivity and uncertainty methods are applied to a box model of the dimethylsulfide (DMS) oxidation cycle in the remote marine boundary layer in order to determine the key physical and chemical parameters and sources of uncertainty. The model considers 58 uncertain parameters, and simulates the diurnal gas-phase cycles of DMS, SO_2, methanesulfonic acid (MSA), and H_2SO_4 for clear-sky summertime conditions observed over the Southern Ocean. The results of this study depend on many underlying assumptions, including the DMS mechanism, simulation conditions, and probability distribution functions of the uncertain parameters. A local direct integration method is used to calculate first-order local sensitivity coefficients for infinitesimal perturbations about the parameter means. Key parameters identified by this analysis are related to DMS emissions, vertical mixing, heterogeneous removal, and the DMS+OH abstraction and addition reactions. MSA and H_2SO_4 are also sensitive to numerous rate constants, which limits the ability of using parameterized mechanisms to predict their concentrations. Of the chemistry, H_2SO_4 is highly sensitive to the rate constants for a set of nighttime reactions that lead to its production through a non-SO_2 path initiated by the oxidation of DMS by NO_3. For the global analysis, the probabilistic collocation method is used to propagate the uncertain parameters through the model. The concentrations of DMS and SO_2 are uncertain (1-σ) by factors of 3.5 and 2.5, respectively, while MSA and H_2SO_4 have uncertainty factors that range between 4.1 and 8.6. The main sources of uncertainty in the four species are from DMS emissions and heterogeneous scavenging, but the uncertain rate constants collectively account for up to 59% of the total uncertainty in MSA and 43% in H_2SO_4. Of the uncertain DMS chemistry, reactions that form and destroy $CH_3S(O)OO$ and CH_3SO_3 are identified as important targets for reducing the uncertainties.

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