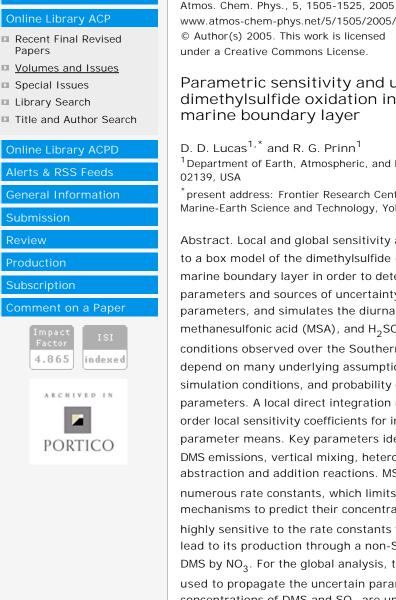
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Parametric sensitivity and uncertainty analysis of dimethylsulfide oxidation in the clear-sky remote marine boundary layer

D. D. Lucas^{1,*} and R. G. Prinn¹ ¹Department of Earth, Atmospheric, and Planetary Sciences, MIT, Cambridge, MA

present address: Frontier Research Center for Global Change, Japan Agency for Marine-Earth Science and Technology, Yokohama, Japan

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, SO₂, methanesulfonic acid (MSA), and H₂SO₄ 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 firstorder 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 H₂SO₄ 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 $_{\rm 2}$ path initiated by the oxidation of DMS by NO3. For the global analysis, the probabilistic collocation method is used to propagate the uncertain parameters through the model. The concentrations of DMS and SO₂ are uncertain $(1-\sigma)$ by factors of 3.5 and 2.5, respectively, while MSA and H₂SO₄ 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 CH₃S(0)00 and CH₃SO₃ are identified as important targets for reducing the uncertainties.

■ Final Revised Paper (PDF, 656 KB) ■ Discussion Paper (ACPD)

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