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Atmospheric
Chemistry
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Discussions

Interactive comment on "Consistent simulation of bromine chemistry from the marine boundary layer to the stratosphere, Part I: model description, sea salt aerosols and pH" *by* A. Kerkweg et al.

Anonymous Referee #2

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Recent model simulations and measurements confirm the essential influence of bromine chemistry on tropospheric as well as stratospheric processes. The two main sources are the release of bromine from sea salt and the decomposition of bromocarbons caused by mainly oceanic sources. The paper as first part of a series of articles, focussing on atmospheric bromine chemistry, addresses on the simulated sea salt aerosol size distribution and the aerosol pH, especially in the marine boundary layer. The applied atmospheric chemistry general circulation model ECHAM5 / MESSy is state-of-the-art for long-term hindcasts of aerosol and gas phase distributions from the boundary layer to the stratosphere. The gas and aerosol phase chemistry are prognostically calculated in great detail. The approach allows also a prognostic determination of the aerosol pH which strongly affects the efficiency of the bromine release. A comparison with available field data and an extensive discussion of the results are given.

The paper is sound and informative. It contributes to the actual discussion on the climate impact of atmospheric halogen chemistry and should be published after revision. A first global simulation attempting a detailed description of the marine aerosol chemistry is performed by considering prognostic pH values. However, due to numerical instabilities this explicit chemistry was calculated for only the coarse mode particles with a liquid water content larger than $10^{-12}\ m^3(aq)$ / $m^3(air)$. This restriction is the main critical point in the presented results. In my feeling, it is not evident that the influence of aerosol chemistry in the accumulation mode and over the continents can be neglected. Chemical transformations involving the accumulation mode have also an effect on the aerosol pH of the coarse mode, especially in coastal regions where freshly emitted sea salt particles are mixed with aged continental aerosol. Although I see the numerical problems, the arguments given by the authors are not fully convincing. These issues should be discussed in more detail.

Furthermore, the authors should consider the following recommendations:

(i) The readability of Section 2 should be improved. Detailed descriptions of the model system ECHAM5/MESSy and the main MESSY submodels are specified in several papers of a special ACP issue. In principle, the authors should refer to the corresponding papers. Only special choices and the key processes (e. g., sea salt emissions, and aerosol chemistry in MECCA-AERO) should be described specifically. In the current manuscript, a brief process description is often added to the MESSy reference (e. g., for the sub-models DRYDEP, SEDI, and SCAV). Nevertheless, reading of the cited paper is required for understanding the used process parametrization. Therefore, the additional comments are redundant and can be omitted. Furthermore, repeated references to the same submodel or paper should avoid.

(ii) The implementation of the explicit aerosol chemistry in MECCA-AERO and its coupling to other processes should be described in more detail: Is the liquid water content of the coarse mode aerosol the sole criteria for calculating the aerosol chemistry? Is this check performed independently for each grid cell in the whole domain or is the liquid water profile considered? What is the sequence of the processes in the operator splitting approach? How is the KPP solver initialized in the restart case (grid cell with aerosol chemistry calculations in the last coupling time step) and in the new start case (no aerosol chemistry in the last step)?

(iii) The thickness of the ground layer and the boundary layer is of essential importance for the interpretation of the results given in the figures. The authors should specify the vertical model resolution especially in the boundary layer? Fig. 1 of the electronic supplement is not helpful in this context.

(iv) Annually and seasonally averaged boundary layer aerosol pH values are presented in Figs. 8 and 9. The authors should briefly explain the applied averaging procedure which has to be taken into account the logarithmic scale of the pH value, the strong non-linear dependency on the liquid water content as well as the expected temporal variability. Furthermore, the vertical structure and the magnitude of temporal fluctuations of the aerosol pH values should be shortly analyzed in the paper.

Interactive comment on Atmos. Chem. Phys. Discuss., 8, 7217, 2008.