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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Modeling the evolution of aerosol particles in a ship plume using PartMC-MOSAIC" *by* J. Tian et al.

Anonymous Referee #1

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Ship-emitted aerosol particles are important contributors to the climate change. These aerosols directly and/or indirectly affect the Global radiation budget. This manuscript presents the evolution of the aerosol the aerosol mixing state of ship-emitted particles using PartMC-MOSASIC (Rimmer et al., 2009). Such information is important for current research on global climate change. The purpose of this paper fits well within the scope of ACP. The manuscript is generally well-structured and the research methodology appears to be acceptable. However, some parts need to be logically strengthened to provide readers with a better understanding of ship-emitted aerosols.

Specific comments: 1. The initial condition for HCs is higher than general emission factors. Several previous studies have reported lower NMVOCs emissions compared to those presented in this manuscript. For instance, Eyring et al. (2005) reported

the global annual emission of NOx and HCs from ocean-going ships to be 21.38 Tg and 1.96 Tg, respectively. Thus, the author should re-check the initial conditions for NMVOCs. In addition, the author has ignored the turbulent dispersion or dilution of emitted HCs with background HCs. The neglecting of background HCs can underestimate the mixing ratios of HCs. This can affect the model-estimated concentrations of gas-phase species and particulate matters, since the ship-plume photochemistry is highly non-linear. Hence, turbulent dilution with background HCs should be included in the model simulations.

2. In general, the mixing ratios of H2SO4 can be highly elevated due to the active oxidation cycle within ship-plumes. This elevation can be particularly conductive to nucleation or new particle formation. In this study, the ship-plume release time for the case study was 2 pm, and hence, the increase in aerosol number density by new particle formation can be negligible (owing to the less active oxidation cycle within the ship-plume). However, in general ship-plumes, nucleation by H2SO4 should be included in the main equation (i.e., the main equation of PartMC-MOSAIC has limitation for general ship-plumes released before noon time). Therefore, a detailed discussion on the uncertainties caused by neglecting nucleation process should be addressed in the manuscript.

3. As shown in Sec. 2, the turbulent dispersion parameters (alpha and beta) and meteorological conditions (mixing height) for this study were fixed values (0.75, 0.6, and 400 m). In general, the magnitude of these lateral and vertical dispersion parameters is a function of atmospheric stability classes or metrological conditions, and mixing height is a time-variant value. Since the model-estimated aerosol number concentration is mainly governed by the turbulent dispersion process, the author should discuss this in more detail or address sensitivity tests for alpha, beta, and z to estimate the uncertainties caused by fixed meteorological parameters.

4. The evolution of gas and bulk aerosol species for two cases was summarized in Fig. 1. The results of the base case and sensitivity case well represent the general trends of

the ship-plume aging. However, the 0-d Gaussian model simulation cannot represent the evolution of these species throughout the entire ship-plume. Also, if the model-estimated mixing ratios of the species can be compared with the observed values, the performance of the model for gas-phase chemistry and heterogeneous transformation can be verified more clearly.

5. In the comparisons presented in Sec. 4.2, the model simulation conditions and the gas-phase and aerosol initial conditions were obtained from HERCULES and the study by Pfaffenberger et al. (2013). Although information for model simulation conditions has been illustrated in the cited references, it is necessary to clearly present the relevant information such as observed species, research domain, and metrological condition in this manuscript for the benefit of the readers. Further, since the article by Pfaffenberger et al. (2013) has not been published yet, this reviewer cannot verify the information for QUANTIFY. Even if the same initial conditions are assumed, the model-estimated number concentration in the present study may include some level of uncertainty since the initial conditions for HERCULES can be different from those for QUANTIFY.

6. In Figs. 3 and 4, the measured aerosol number distribution for the background condition, initial condition, and ship corridor is compared with the model results. The model-estimated number distribution is lower than the background data for sensitivity case when the aging time is more than 5 h and the particle diameter is smaller than 0.25 μm (from Fig. 4(b)). A reason for this underestimation needs to be provided.

7. In Fig 5, two-dimensional number distributions were compared for cases with and without coagulation. Evaluations for aerosol size distribution and aerosol constituents have not been carried out here, and hence, the reviewer believes that further comprehensive analysis or discussion clearly demonstrating the effect of coagulation on the aerosol mixing state should be included in Sec. 4.4.

8. Figs. 5 and 6 (i.e., Sec 4.4) indicate that the analysis for two-dimensional number

distributions was based on dry diameter. Why was this done even though this model includes water transfer as shown in Eq. (2)? Analyses based on the wet diameter can provide more useful information.

9. In Fig. 7, two-dimensional number distributions are compared. However, it is not clear what type of aerosol mixing state of chemical composition was assumed in the model simulations. According to my understanding, the author assumed the aerosol mixing state as external mixture. The value of the hygroscopicity (k) varies with as chemically aged. Although fresh ship-emitted aerosol is externally mixed, background sea-salt aerosol may be internally mixed. The analysis therefore requires additional discussion since the chemical mixing state of aerosols can affect hygroscopicity.

Minor comments: 1. As summarized in Table 3, for model simulation, one initial condition and two boundary conditions (i.e., Background and Ship corridor) were used. In order to clearly express the model prediction methodology, it is necessary to clarify how these background conditions were considered in the model predictions.

2. In Fig. 2, since the scale of y-axis is logarithmic, it is difficult to distinguish aerosol number concentration at each point. Therefore, the y-axis should be changed to a linear scale for the benefit of readers. In addition, statistical analysis (e.g., error and bias) should be included in the model evaluation to scientifically quantify the model simulation performance.

3. In the conclusion, the author states that “the model results agreed well with observed particle number concentration and size distributions.” However, the comparison study (i.e., Figs 3 and 4) did not sufficiently verify model simulation performance for aerosol size distribution; the comparison was not carried out. Therefore, the above sentence should be replaced by “the model results agreed well with observed particle number concentration.”

4. In order to clearly express the results of this study, the labels of figures should be uniform. - p. 32, in the Fig. 2: “Number Concentration” should be replaced by

“Number concentration”. - p. 33-37, in the Figs. 3-5: “No Coag” and “With Coag” should be replaced by “Without coag.” and “With coag.”. - p. 33-34, in the Figs. 3-4: “Dry Diameter” should be replaced by “Dry diameter D”. - p. 36-39, in the Figs. 5-7: “Dry Diameter D” should be replaced by “Dry diameter D”. - p. 36-39, in the Figs. 5-7: “number conc.” should be replaced by “Number concentration”. - p. 40, in the Fig. 8: “CCN Concentration” and “Local Time” should be replaced by “CCN concentration” and “Local time”.

Interactive comment on Atmos. Chem. Phys. Discuss., 13, 16733, 2013.