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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 #3**

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This manuscript investigated aerosol processes in ship plume, developing a ship-plume model. For the development of the ship-plume model, authors combined three model components of particle-resolved Monte Carlo (PartMC), MOSAIC (WRF-CHEM gas-phase chemistry and aerosol dynamic/thermodynamic package), and ship-plume dispersion scheme. The research framework appears to be reasonable. The manuscript is well written and the scope of the study is appropriate to ACP. However, the manuscript also has several major weak points (see below). I recommend that authors should address the following issues and comments properly, before the publication of the manuscript on ACP.

Major points Main concerns of this study are as follows: The largest concern is model validation. Fig. 2 appears to be only the direct validation of the ship-plume model, in



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terms of total aerosol number concentrations (also just over “70 minutes” after ship-plume release). But, for this comparison, there are also many “knobs” available in the model. For example, it appears that the model does not include “nucleation” parameterization (based on Eq. 2). The nucleation is an important process for the “total aerosol number concentrations”. Although I understand that it is extremely difficult to correctly simulate the aerosol number concentrations generated from the nucleation process, it is still of great significance, because inside the ship plume H2SO4 mixing ratios are so high (authors also mentioned the presence of high levels of NH3 in the background air and high relative humidity of 90%). These conditions would create very favorable condition for nucleation. Therefore, without the consideration of the nucleation process, I do not think that the model can produce realistic total aerosol number concentrations.

Another knob would be dilution rates. Authors incorporated von Glasow et al.’s parameterizations in this study. But, the turbulent dispersion of the ship plumes is dictated primarily by the stability conditions of the marine boundary layer. The changes in the stability conditions would greatly affect the aerosol number concentrations by the orders of magnitude. This model validation issue is related to the omission of sensitivity analysis to the abovementioned factors such as nucleation, stability condition of the marine boundary layer and emission rates of particles from ship. Authors have to show some results of these sensitivity studies. Otherwise, there would be huge uncertainty (or possible errors) in keeping track of the aerosol number density inside the ship plume, e.g., in the analysis like Fig. 2.

I wonder if the QUANTIFY aircraft measurement data did not include chemical composition such as the concentrations of sulfate, nitrate, ozone, HNO3, NO2, SO2 etc.... If these are available, authors should compare this composition with the modeled ones for the model verification.

Another issue is condensation. This manuscript greatly focused on coagulation, and then investigated particle-based/resolved chemical (e.g. internal-external mixing) and



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physical (e.g. CCN activities) aging. However, the dominant factor determining the mixing state may not be coagulation but condensation, since coagulation is slow but condensation is much faster process. For instance, H2SO4 condensation onto BC and sea-salt particles would be faster and more effective than sulfate particle coagulation. Being related to this issue, Fig. 5 only shows the coagulation effects and pp.16737-16740 concentrates on coagulation too much. How about the impacts of HNO3 & N2O5 condensation onto particles (i.e. nitrate formation), e.g. on CCN activity?

Other issues 1. P. 16734: 6-10, Again, this conclusion could be changed, if the nucleation parameterization and other dilution schemes were introduced to ship-plume modeling. 2. P. 16739: Eqs. 3&4, In Eq. 2, authors have to explain how to calculate "water transfer". I guess that the thermodynamic aerosol module (MTEM & MESA) may have a calculation component, but the important thing is again that the water condensation (particulate water formation) would be one of the main processes that can determine "aerosol size distribution" (such as Fig. 3), particularly under such a high relative humidity condition at RH=90%. In addition, in Eq. 3, authors have to explain more about gas-particle transfer (i.e. condensation of atmospheric species), because this is very important process in determining the aerosol mixing state and aerosol size distribution. 3. P.16740: 6-12 & p.16741: 1-3, Authors should show some sensitivity analysis about ïĄąïĂăand ïĄć. In the same context, the initial ship-plume cross section (wo and ho) can also be important parameters. 4. P.16742:9-27, This is a bit lengthy explanation about particle-resolved MC model. I understand this is a stochastic model, but is there such a big computation burden, because this is a "box model" simulation? 5. P. 16743: 2-5, Coagulation may be important (or fast) only between very small particles and large particles. However, again nucleation governs the number concentrations of the very small particles (nucleation-mode particles). 6. p.16747: 14-19, Again, more important process for aerosol chemical evolution is condensation (of H2SO4, HNO3, N2O5, SOA precursors etc. . .) than coagulation. This chemical modification is important further for CCN activity, as discussed by authors. 7. P. 16748: 16-20, Denitrogen pentoxide (N2O5) is NOT reservoir species in NOy. N(V) species



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such as HNO3 and nitrate are reservoirs in NOy. 8. P.16748: 24, Sulfate and BC were "diluted", not "depleted". Words should be used more carefully. 9. P.16749: 1-4, SOAs were formed via the oxidation of alkenes with ozone and NO3, too. 10. P.16752: 16-20, Particle growth due to condensation of atmospheric species and water can also alter the aerosol number-size distribution greatly. 11. P.16764 Table2, The level of paraffin carbon (PAR) is almost the same level of sulfur dioxide (SO2) and is even higher than that of CO. This is really high level (or large emission). NMVOC emission is usually from evaporative and fugitive emissions (from engines, pump, joints, and storages. . ..), not from combustion. It appears that this level is too high. Obviously, this will affect ozone and OH mixing ratios, SOA formation, and particle growth and chemical evolution inside the ship plumes. Therefore, authors have to consider this factor more seriously. Regarding the NMVOC emissions from ships, there are numerous articles available. 12. P.16766 Fig. 1, In the modeling results, very near the ship stack, OH is depleted, but HNO3 is not depleted. Could you explain this? Because HNO3 is produced by NO2+OH reaction in the presence of the third body (M). 13. P.16770 Fig. 5, Again, the composition change can be due to condensation of many gas species such as H2SO4, HNO3, N2O5 SOA precursors. . ...

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