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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Microphysical simulations of new particle formation in the upper troposphere and lower stratosphere" *by* J. M. English et al.

J. M. English et al.

jayenglish@gmail.com

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Dear Jeff: Please see your comments below with our responses. We have attached the modified paper as a pdf supplement.

This paper describes the coupling of the CARMA microphysics model to the WACCM GCM. Since WACCM extends to higher altitudes than typical GCMs, this combination makes for an ideal model for UT/LS aerosol studies. The authors compare predictions from this model with a large and diverse set of observations. The extent of the comparison is impressive and commendable, and it proved useful for ïňĄnding biases in the model that could be corrected (e.g. with VDW coagulation correction, and the

meteorites). Finally the authors show the sensitivity of the model results to different nucleation schemes. They ïňĄnd that particle number concentrations and size distributions above 10 nm are not sensitive to the nucleation scheme. The paper is well in the scope of ACP, provides good background info on the model, has the detailed evaluation and gives several new scientiïňĄc ïňĄndings (insensitivity of size dist to nucleation in the UT/LS, need for VDW coagulation ïňĄx etc.). I suggest this paper be published in ACP once the following minor corrections (mostly for clariïňĄcation) have been addressed. P 12442, L 6: I was confused when I ïňĄrst read that IMN is 25% higher than BHN, but the two BHN schemes were 2-orders of magnitude apart. I was thinking "How did they arbitrarily choose which BHN scheme to compare IMN to?". It made more sense when I realized that Yu's BHN scheme is related to Yu's IMN scheme when reading the paper. Maybe rewrite the sentence in the abstract.

Authors' response: We have clarified the sentence on line 15, 16: "(two binary homogeneous schemes and an ion-mediated scheme related to one of the binary schemes)." And "Simulations suggest that ion-mediated nucleation rates in the UTLS are 25% higher than its related binary scheme.."

P 12443, L 24: Should be changed to (changes in allcaps): "Pierce and Adams (2009) AND SNOW-KROPLA ET AL. (2011) calculated CHANGES IN IMN from solar cycle using a SECTIONAL model..." Snow-Kropla, E. J., Pierce, J. R., Westervelt, D. M., and Trivitayanurak, W.: Cosmic rays, aerosol formation and cloud-condensation nuclei: sensitivities to model uncertainties, Atmos. Chem. Phys., 11, 4001-4013, doi:10.5194/acp-11-4001-2011, 2011.

Authors' response: We have changed the text to your suggested wording (line 60-61) and added the reference (lines 717-719).

P 12443, L 26: Yu did compare to BHN (and several other mechanisms) in the following paper: Yu, F., G. Luo , T. Bates , B. Anderson , A. Clarke , V. Kapustin , R. Yantosca , Y. Wang , S. Wu, Spatial distributions of particle number concentrations in the global

troposphere: Simulations, observations, and implications for nucleation mechanisms, J. Geophys. Res., 115, D17205, doi:10.1029/2009JD013473, 2010.

Authors' response: We have added to line 65-68: "Yu et al. (2010) compared nucleation rates and number concentration from IMN and two different BHN schemes in the troposphere to aircraft observations, but did not study the aerosol evolution (size, mass, effective radius) and did not study stratospheric properties."

P 12443, L 29: In Kazil et al. (2010) they used activation nucleation in the CONTINENTAL boundary layer (not the entire boundary layer). Also, they said its likely because of the roll of organics, but activation nucleation is does not necessarily involve organics.

Authors' response: We have changed the sentence to lines 68-71 : "Kazil et al. (2010) found that simulations agree best with observations in the lower and mid-troposphere when IMN and BHN are included across the entire model domain and organic cluster formation is included but limited to the continental boundary layer.."

P 12445, L 17: Can you give more details on size-dependent wet deposition? Are all particles removed with the same efĩňĄciency (even particles too small to activate)?

Authors' response: Yes. We have added line 114-115: "All of the aerosol bins are assumed to have a constant 0.3 solubility parameter."

P 12445, L 24: Is achieving stability good enough to achieve accuracy. I'd worry that if my timestep was long enough to have H2SO4 go negative, that only halving my timestep would still result in biased results. Which process is calculated ĩňĄrst, nucleation or condensation? You could check the accuracy of your scheme by switching the order and running one more simulation to see if the results change.

Authors' response: We have looked at higher substep rates and found that increasing the number of timesteps past the point at which negative gas amounts were found did not significantly change results. We've added the following at line 121: "Nucleation and growth are treated simultaneously in the model." And at lines 124-126: "We have found

that increasing the number of timesteps past the point at which negative gas amounts were found did not significantly change results. Additionally, we limited nucleation to 40% of the sulfuric acid available."

P12446, L7 : Do you think the roll of organics in condensation/coagulation sink would affect your sulfate results?

Authors' response: We don't expect organics to impact stratospheric aerosol– observations suggest they do not cross the tropopause (Murphy et al., "Carbonaceous material in aerosol particles in the lower stratosphere and tropopause region," JGR, 2007). In the upper troposphere, organics may influence mass but probably not number; regardless, we wanted to isolate the impacts of sulfates. At lines 134-137 we added "Although other aerosols, such as organics, are known to compose a significant fraction of the sulfate aerosol mass in the UTLS (Froyd et al., 2009, Murphy et al., 2007), sulfates are believed to be the primary source of new particles in this region, and the primary aerosol in the lower and middle stratosphere (Murphy et al., 2007)."

P12446, L14: What do you mean by brownian diffusion of aerosols is important above 100 km? Is it important for diffusion between grid boxes there? Brownian diffusion is important everywhere for coagulation. Were you referring to diffusion between gridboxes, or diffusion on the microscale.

Authors' response: At line 141 we added "However, CARMA treats Brownian diffusion of aerosols, which becomes important above 100 km as the heterosphere is approached, and which is not well treated by algorithms in WACCM." You are correct that Brownian diffusion is important for aerosol coagulation everywhere in the model as stated on line 142. CARMA treats Brownian diffusion of all gases and aerosols.

P12449, L6: Why not use the ion formation rates from Usoskin, I. G. and Kovaltsov, G. A.: Cosmic ray induced ionization in the atmosphere: Full modeling and practical applications, J. Geophys. Res., 111, doi:10.1029/2006JD007150, 2006. However, I don't think it will change your results much (it will be a smaller change than the change

between nucleation schemes), so probably not a big deal.

Authors' response: We used a constant value of 10 cm-3 s-1 because ionization rate does not vary much in the tropical UTLS, where most new particle formation takes place. We have stated and did not change lines 211-212: ". . ..it is relatively constant in the UTLS and is estimated to be between 5 and 20 ion-pairs per cm3".

P12450, L9 and Figure 3: What is causing the 3rd SO2 max at the top of 3b?

Authors' response: We have added at line 239-240: ". . .and SO2 increases again in the upper stratospheric due to photolytic conversion of H2SO4 back to SO2 (Mills et al., 2005)." And added it to the list of references.

P12451, L7: Figure 5c shows H2SO4 concentrations, not mixing ratios.

Authors' response: We have changed "mixing ratios" to "concentrations" on line 266.

P12452, L9: I don't think this is an ironic situation. Maybe say "interesting" instead.

Authors' response: We have changed "ironic" to "interesting" at line 295.

P12454, L18: Please add Snow-Kropla et al. 2011 to this list.

Authors' response: Done at line 360.

Section 4.2 and Figures 13 and 15. Did you pick individual grid boxes that corresponded to where ïňĆights were (at the same time of day as the ïňĆights) or did you average over all time and space in the regions? The averaging could lead to differences from the observations.

Authors' response: We averaged over all time and space, as stated in the paper: "Calculated 1-day averages of the third year are checked for NPF conditions and segregated into two sets of data (with and without recent NPF). Simulation "data" points include values for 360 days in the third simulation year. The model outputs daily averages, so these criteria will not provide instantaneous indicators of recent NPF." We

agree this could lead to differences, so we've added at line 396: "Additionally, since the model output is across the entire year, while the aircraft data are obtained on specific days, differences may be due to temporal variability."

Page 12459, L17: VDW improves number concentrations. I'd like to see its affects on the size distributions too. Section 4.2: Did you compare the size distributions when VDW corrections were turned on?

Authors' response: We assume you mean "off"; because Figure 13 shows results with VDW corrections on. Yes, we compared size distributions with VDW turned off, and the results were slightly worse (high number concentrations, especially at smaller sizes, compared to observations). We've added the "Zhao no VW" line to Figure 14 and added a short discussion in the paper at line 425: "On the other hand, the Zhao no VW curve has higher number concentrations than any of the nucleation schemes. This reinforces the conclusion that coagulation, not nucleation is the dominant process determining aerosol number at atmospherically relevant sizes. "

Page 12459, L10: Pierce and Adams (2009) would be a better citation here than (2007).

Authors' response: We've made the change at line 489, and removed the Pierce and Adams (2007) reference from the list.

Figure 2b: What are all the different green lines?

Authors' response: We've added the statement to the figure caption: "Each simulation line represents model output at a specific latitude at 4° increments between 30° and 70° at all longitudes."

Figures 7, 8, 9, 10, 11, 16: Can you make the blue and green colors lighter. They are hard to differentiate from the black lines in some instances.

Authors' response: Done.

Figure 11: VDW appears to have a large affect on aerosol surface area than number. This is not what I would have expected. Do you know why this is?

Authors' response: The no VW simulation predicts higher nucleation rate in places with higher CO (30 N near 150 mb), but lower nucleation rates outside this region. Since the particles grow faster with VW correction, the Area plot for no VW shows higher area. We've added at line 337-339: "Additionally, the Zhao no VW predicts higher nucleation at higher CO but lower nucleation at lower CO, as shown in Fig. 11b. The Zhao no VW area plot (Fig. 11b) is higher than with the VW correction due to slower growth rates." Also, the y-axis for the Number plot spans one additional order of magnitude, which can explain some of the difference.

Thanks for helping us improve this paper.

Sincerely, Jason English, Brian Toon, Michael Mills, and Fangqun Yu

Please also note the supplement to this comment:
http://www.atmos-chem-phys-discuss.net/11/C6793/2011/acpd-11-C6793-2011-supplement.pdf

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