

***Interactive comment on* “Effects of boundary layer particle formation on cloud droplet number and changes in cloud albedo from 1850 to 2000” by J. Merikanto et al.**

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Review of “Effects of boundary layer particle formation on cloud droplet number and changes in cloud albedo from 1850 to 2000” by Merikanto et al.

This article explores the sensitivity of pre-industrial and present-day cloud droplet number to the inclusion of activation-type nucleation to the boundary layer in the GLOMAP-TOMCAT model. The Nenes and Seinfeld activation parameterization was used to map the predicted aerosol size distributions to CDNC by assuming updraft velocities. The corresponding sensitivity of the change in cloud albedo from 1850-present due

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to activation-type nucleation in the boundary layer is presented based on the CDNC changes.

This article presents useful new information to the aerosol and climate communities, is of high quality and deserves to be published in ACP. However, I do have several comments that should be addressed before publication.

I have two major comments:

1. In the introduction you discuss the Spracklen (2006) paper and how uncertainties in primary emissions can greatly affect nucleation rates. There are thus large uncertainties in the nucleation rates of both the 1850 and present-day simulations. Primary emissions are even more uncertain in your 1850 simulation than in the present-day simulations. I would assume that if you could do a large number of 1850 and present-day simulations, spanning the uncertainty range of primary emissions in both, you could come up with many combinations of 1850 and present-day simulations where activation-type nucleation in the boundary layer clearly increased the global albedo change. This would occur for 1850 simulations with higher primary emissions and present-day simulations with lower primary emissions. You would also find many combinations that clearly decreased the global albedo change (due to the opposite pairings). It is not clear that the simulations chosen here are the perfect middle ground.

Two recent articles published related results to this. In Wang and Penner (Atmos. Chem. Phys., 9, 239-260, 2009), they find the 1st aerosol indirect forcing (present day - pre-industrial) when boundary-layer nucleation is included/removed in either time period and when primary-sulfate emissions are included in the present day. The sensitivity of the AIE to nucleation changes greatly when primary sulfate is included. In Pierce and Adams (Atmos. Chem. Phys., 9, 1339-1356, 2009), we found similar results to you in our base case simulations. The sensitivity of

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CCN (not CDNC) to nucleation was similar in the pre-industrial and present-day simulations. However, the sensitivity of CCN to nucleation changed when we changed either our present-day primary emissions or our SOA.

I understand how expensive these simulations are, so I don't recommend running any new simulations; however, the sensitivity of the nucleation/CDNC connection to primary emissions must be made clear in the results section and particularly in the conclusions/discussion section. The global near cancellation of changing 1850 and present-day cloud albedo may just be a coincidence, and any regional trends found might not occur if other emissions were used.

2. The A-factor in the activation-type nucleation used in this paper is tuned to observations, and often very different values are found for different regions (e.g. Riipinen et al., *Atmos. Chem. Phys.*, 7, 1899-1914, 2007). Therefore, it is not obvious that this A-factor would have remained constant since 1850. This is a fundamental assumption of this paper, and it should be addressed clearly during the background of the nucleation parameterization as well as in the conclusions. Obviously, it is impossible to say right now how the A-factor would have changed (and the approach you've taken is as good as you can do); however, it must be made clear that this a fundamental assumption.

Minor comments:

1. Page 5265, Line 18: There has been several new papers showing global modelling results with new particle formation within the last 6 months. If I remember correctly, this paper was originally submitted several months ago, so the omission of these papers is understandable. Please add a brief discussion somewhere in the introduction about Makkonen et al. (*Atmos. Chem. Phys. Discuss.*, 8, 10955-10998, 2008) along with the Wang and Penner (2009) and Pierce and Adams

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- (2009) papers cited above. It would also be useful to compare your results to the Wang and Penner (2009) and Pierce and Adams (2009) results later in the paper.
2. Page 5269, Line 7: How consistent are the activation diameters used in GLOMAP for wet chemistry and wet removal with the activation diameters found using Nenes and Seinfeld for CDNC? There may be sizable uncertainties if large numbers of Aitken-mode particles that are not susceptible to wet removal are counted as CDNC. This might deserve a brief discussion if the diameters are inconsistent.
 3. All tables and figures: I assume that the values shown in the tables and figures are boundary-layer values and not tropospheric averages, but it doesn't appear to be mentioned. This should be explicitly stated in the captions of each table and figure.
 4. Figure 1: Why use N(3-10nm) rather than J(3nm)? It seems like J(3nm) would be a more straightforward comparison here rather than N(3-10nm). N(3-10nm) depends not only on J(3nm), but also on the rate of particles growing out of this range and their removal rate, and thus it is a convolution of terms.
 5. Page 5270, Line 8: "Since the condensation sink represents the effective coagulation sink for the smallest particles, a smaller portion of the newly formed marine particles is able to grow to large sizes in 2000 than in 1850." The fraction of new particles that will grow to larger sizes is also a function of the growth rate.
 6. Page 5270, Line 13: 0.4 m/s updraft might be high for large-scale clouds that would be most susceptible to albedo changes and that cover the largest areas of the earth (e.g. Guo et al., Environ. Res. Lett. 3 (October-December 2008) doi:10.1088/1748-9326/3/4/045020). This would cause smaller activation diameters and would increase the importance of nucleation. The connection between the choice of updraft velocity and the CDNC results should be mentioned.

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7. Figure 2: It might be preferable to have the two lower panels showing the percent change rather than the difference. This is just a preference though.
8. Page 5271, Line 4: “...indicated in Fig. 2.” Add, “by gray boxes in the upper two panels.” You should also mention what the gray boxes are in the caption of Figure 2
9. Page 5274, Line 5: “Boundary layer particle formation is not included in current GCMs.” The Makkonen et al. (2008), Wang and Penner (2009) and Pierce and Adams (2009) papers all include simulations with activation-type nucleation in the boundary layer.
10. Page 5274, Line 5: Activation-type nucleation is not the only scheme that predicts nucleation in the boundary layer (e.g. Yu et al., Atmos. Chem. Phys., 8, 2537–2554, 2008 and Napari et al. 2002... though I am aware that Merikanto et al., 2007 updates this and the BL nucleation tends to go away). It is probably more appropriate here to say activation-type nucleation in the boundary layer rather than boundary layer nucleation.
11. Page 5274, Line 20: This sentence should be rewritten to reflect the Wang and Penner (2009) paper.
12. Page 5274, Line 8, 10, 12, 24... others: Throughout this paper (and especially in this conclusions section), you mention the effects of “particle formation” on CDNC, albedo etc.. If I understand correctly, you are not adding and removing particle formation altogether. Rather, you are adding and removing the activation-type nucleation for the boundary layer (leaving the Vehkamäki and Merikanto parameterizations for the entire atmosphere). It would be more precise then to say that you these are the effects of the additional nucleation scheme in the boundary layer, rather than particle formation altogether.

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13. Page 5275, Line 15: This paragraph is a good and necessary discussion of uncertainties. However, the two points I described in the major comments need to be addressed in your conclusions. The main conclusions of your paper might easily change if different emissions or varying A-factors were used.

Interactive comment on Atmos. Chem. Phys. Discuss., 9, 5263, 2009.

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