

***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.**

J. Merikanto et al.

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We would like to thank Jeffrey Pierce (referee) for his insightful and constructive comments. The issues raised in the comments are well justified. Below we will reply to every comment made by the referee.

Replies to Major comments:

1. The referee points out that uncertainties in the primary particle emissions can greatly affect boundary layer nucleation rates both in 1850 and present day simulations. The connection between primary particle emissions and the aerosol production capability of BL nucleation that has been addressed in previous stud-

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ies is now discussed in the revised paper. There are significant uncertainties in the emissions of primary particles and aerosol precursor gases, and historical change in global cloud albedo may be both enhanced or suppressed due to BL particle formation depending on the selected emission scheme. Nevertheless, including activation-type BL particle formation changes the regional CCN (and CDNC) distributions considerably, and climate models that do not include BL particle formation are likely to get the regional climate forcing from anthropogenic aerosol incorrect.

We add to Introduction: "Wang and Penner (2009) studied the effect of activation BL nucleation on present day and pre-industrial CCN and on the first indirect effect. They found that this mechanism increased present day global boundary layer CCN by 31.4% when no primary emitted sulfate was included and 5.3 % when primary sulfate was included. Overall, activation BL nucleation reduced the anthropogenic fraction of CCN and decreased the first aerosol indirect effect. In another recent model study, Pierce and Adams (2009) also obtained a 5% increase in global CCN due to activation nucleation, but also pointed out that the sensitivity of CCN to different nucleation scenarios depended on the selected primary emission and secondary organic aerosol schemes."

We will say in Results: "The global mean albedo change is predicted to be 3.97% with particle formation and 3.85% without, so the globally averaged impact of BL particle formation is negligible. However, the negligible effect of BL particle formation on average global albedo change may be coincidental and related to the selected representation of emissions, as discussed in Introduction. Using different but plausible combinations of emissions could lead to both increase or decrease in the average global albedo change due to BL particle formation."

We will add to Conclusions and discussions: "Recent studies by Wang and Penner (2009) and Pierce and Adams (2009) have showed that historical change in the impact of BL particle formation on clouds depends on the selected represen-

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tation of primary particle emissions. There are uncertainties in the emitted total mass and size distribution of primary particles both in 1850 and 2000. Wildfires represent a major but uncertain component in 1850 continental primary emissions. Recent charcoal records suggest that global wildfire activity was much higher in 1850 than today (Marlon et al., 2008). For 1850 we have used a population-weighted average of AEROCOM estimates for years 1750 and 2000 (Dentener et al., 2006) where wildfire emissions in the high latitudes in the northern hemisphere (Europe, N. America, Russia) are higher in 1850 than in 2000 due to less fire suppression, but total emissions are lower in 1850 than in 2000 due to changes in population and land use. We show that the regional contribution of BL particle formation to CDNC is not the same over the period 1850 to 2000. Therefore, climate model studies that do not include BL particle formation are likely to get the regional climate forcing from anthropogenic aerosol incorrect. Both this work and that of Pierce and Adams (2009) show that on a global scale BL particle formation contributes equally, to CDNC and CCN concentrations respectively, over this period. There are significant uncertainties in the emissions of primary particles and aerosol precursor gases. With different emission assumptions historical changes in BL particle formation may also be important at the global scale."

2. The referee notes that throughout the paper we assume a constant A-factor both in different regions and in 1850 and 2000 simulations, but that observed A-factors in the activation-type nucleation can have different values in different regions. The referee also points out that it is not obvious that the A-factor has remained constant since 1850. These points are now mentioned in description of the nucleation parameterization and in the discussion, as suggested by the referee. To the description of the nucleation parameterization we will add:

"Calculated A-factors for observed nucleation events vary temporally and regionally between $A=10^{-8} - 10^{-4}s^{-1}$ (Riipinen et al., 2007). The reasons for the varia-

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tion in A-factor are not known."

We will add to Conclusions and Discussion: "In this study we have assumed a universally constant activation nucleation rate coefficient $A=2\times 10^{-6}$ 1/s, but observed rate coefficients vary between $10^{-8} - 10^{-4}\text{s}^{-1}$ for different nucleation events (Riipinen et al., 2007). It is not clear if a single rate coefficient can be applied in different regions. We also assume the same rate coefficient for 1850 to 2000, but it is possible that some unknown factors contributing to the rate coefficient have also changed since 1850. However, our other work (Spracklen et al., 2008; Merikanto et al., 2009) has shown that CCN (and therefore CDN) are fairly insensitive to large changes in the A-factor."

Replies to Minor Comments:

1. Previous studies that are connected to the current paper are now discussed in much more detail. We will add to Introduction:

"The importance of activation nucleation in the BL has been recently studied also with other global models. Makkonen et al. (2009) used ECHAM5-HAM to show that particle formation is likely to enhance global present day CDNC significantly, but also noted that the uncertainties in the mechanism lead to large uncertainties in the obtained CDNC. Wang and Penner (2009) studied the effect of activation BL nucleation on present day and pre-industrial CCN and on the first indirect effect. They found that this mechanism increased present day global boundary layer CCN by 31.4% when no primary emitted sulfate was included and 5.3 % when primary sulfate was included. Overall, activation nucleation reduced the anthropogenic fraction of CCN and decreased the first aerosol indirect effect. In another recent model study, Pierce and Adams (2009) also obtained a 5% increase in global CCN due to activation boundary layer nucleation, but also pointed out that the sensitivity of CCN to different nucleation scenarios depended on the selected primary emission and secondary organic aerosol schemes."

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We will add to results: "BL particle formation increases global mean CDNC by 16.0% in 1850 and 13.5% in 2000, indicating that global relative contribution to CDNC has been quite similar. This is in line with the result of Pierce and Adams (2009), who found that the sensitivity of cloud condensation nuclei to activation-type particle formation was similar in their pre-industrial and present day simulations."

We will also add to results: "In their recent study Wang and Penner (2009) found that activation boundary layer particle formation decreases the first aerosol indirect effect especially over northern hemisphere oceans. Also in our simulations the albedo change is decreased in these regions. Over northern hemisphere land regions Wang and Penner (2009) found a modest decrease in the first indirect effect due to boundary layer particle formation when primary sulfate was included, or a modest increase in the first indirect effect when primary sulfate was not included. According to our results the albedo change is decreased over most northern hemisphere land areas (except spring and summer seasons in Canada and Siberia due to reductions in wildfire emissions since 1850 in these regions). Our results suggest an enhanced albedo change over most of southern hemisphere, while Wand and Penner (2009) obtained a modest decrease or no change at all in this region depending on the applied primary sulphate scheme."

2. The reviewer makes a valid point that the differences in the activation diameters used in GLOMAP and those obtained from Nenes and Seinfeld parameterization can lead to uncertainties in the results.

We will add to model description: "Cloud droplet activation in clouds is modeled using a constant activation dry diameter of 50 nm. In an earlier GLOMAP study the 50 nm activation diameter was found to correspond to a minimum updraft velocity of approximately 0.4 m/s in the Nenes and Seinfeld parameterization both in polluted and clean conditions (Figure 8 in Korhonen et al. (2008))."

We will also add to Results: "Here, we have used a value of 0.4 m/s for the cloud
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- updraft velocity, which approximately corresponds to the constant cloud activation nuclei dry diameter of 50 nm used in the simulations. Measured updraft velocities in BL clouds vary greatly, but range typically between 0.3–0.5 m/s (Nenes and Seinfeld, 2001)."
3. Will be done.
 4. The purpose of Figures 1 and 2 is to show the difference of the impact of boundary layer nucleation to concentrations of freshly nucleated particles and CDNC. We agree that $N(3-10\text{nm})$ does not describe the actual nucleation rates. In our opinion comparing two concentrations is more illustrative than comparing a rate and a concentration. $J(3\text{nm})$ would be interesting on its own right, but unfortunately it was not recorded during simulations.
 5. The sentence is modified to read: "A smaller proportion of newly formed marine particles is able to grow to large sizes in 2000 than in 1850 because the changes in condensation sink are larger than changes in condensing sulfuric acid as shown in Figure 1."
 6. The referee asks us to comment more about the results obtained with different updrafts. We will added to results: "We note that results are similar with different updraft velocities, although the regional differences are more pronounced with higher updrafts. This is because the importance of activation boundary layer nucleation on CDNC decreases with smaller updrafts due to increasing cloud droplet activation radius. Clouds that are most susceptible to albedo change may have smaller characteristic updrafts than 0.4 m/s, but larger impact of BL nucleation on high updraft clouds would partly compensate this."
 7. We prefer to show a difference rather than percentage change in the lower panels of Figure 2, so that comparison with lower panels of Figure 1 is more straightforward.

8. Will be added.
9. The sentence will be removed.
10. We will use the term activation boundary layer particle formation, rather than just boundary layer nucleation.
11. The sentence will be removed.
12. We will use terms "BL particle formation" or "activation nucleation in the BL". In our model activation nucleation is the only significant nucleation mechanism in the BL, so we use these terms interchangeably.
13. Discussion about the use of constant A-factor will be included (see reply to major comment 2).

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

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