

Anonymous Referee #1

This is an interesting study of the role of dust in droplet nucleation. Although some of the conclusions are compromised by neglect of droplet collision, I don't think those concerns need to be addressed in this study. There might even be value in neglecting droplet collisions, although that raises questions about the evaluation.

We would like to thank the reviewer for his/her positive response. Indeed, as discussed in the text, the CDNC shown in this study is equal to the nucleated droplet number concentration before the collision and coalescence processes, which we acknowledge as an upper limit in clouds. This may result on the overestimation of CDNC in some areas. However, over polluted regions, where the model overestimates CDNC, the sensitivity of cloud albedo (R_c) to CDNC is low. For typical values of cloud albedo ($0.28 \leq R_c \leq 0.72$) $\Delta R_c = 0.075 \Delta \ln(\text{CDNC})$ (Seinfeld and Pandis, 2006). Therefore, Cloud albedo sensitivity to CDNC decreases with increasing CDNC. Based on the typical properties of stratus clouds, a 30% overestimation of CDNC results in 2.25% increase to cloud albedo and in a perturbation of -1.1 W m^{-2} in the global mean cloud radiative forcing (Schwartz, 1996). Below is a point by point response to the reviewer's comments.

Minor comments

- 1. Lines 65-58. Confusing text. I suggest instead "Reports of hygroscopic growth measurements of dust particles indicate solubility to be very low, so that activation of observed cloud condensation nuclei (CCN) has been attributed to soluble ions present in the particles".*

We adopted the reviewer's suggestion and changed the text accordingly.

- 2. Line 71. Wouldn't the "fraction of soluble material on the particles" correspond to the soluble ions referred to above? Or is the critical distinction between soluble material within and on the surface of the particles? And between the fresh dust and aged dust? Perhaps even fresh dust is coated with soluble ions. This is not to say that adsorption or condensation of secondary soluble material are not important, but why neglect soluble material in the emitted dust. Surely some types of dust (clays?) must contain soluble material.*

Yes, in Line 71, the "fraction of soluble material on the particles" corresponds to the soluble ions referred in line 66. In this study we have implicitly taken into account the presence of soluble material in the freshly emitted dust by assuming that the emitted mineral particles are a mixture of inert material (i.e., bulk dust) with reactive components (i.e., Ca^{2+} , Mg^{2+} , K^+ , and Na^+) that form soluble salts.

- 3. Line 90. Start new paragraph with "Hatch".*

Done.

- 4. Line 96. Start new paragraph with "Based".*

Done.

5. *Lines 117. Start new paragraph with “Soluble”, as the previous text describes mechanism while the following text describes conclusions above dust activity sampled in the atmosphere.*

Done.

6. *Line 143. Drop “Only”, as “few” implies it.*

Corrected.

7. *Line 159. I think you mean “aged dust can substantially deplete in-cloud supersaturation”, and replace “eventually” with “hence”.*

Corrected.

8. *Line 179. Replace “which “ with “that”.*

Done.

9. *Line 201. Replace “is” with “are”.*

Done.

10. *Section 3.1. This discussion never mentions the role of droplet collision in depleting droplet number concentration. Droplet activation is not the only process that determines droplet number concentration. Please consider the role of collision in your discussion, or show that it is not important (perhaps in thin warm clouds).*

In this study, droplet depletion by collision, coalescence and collection are not taken into account. Therefore, CDNC values presented in this section can be considered as an upper limit. This is now pointed out at the beginning of the section.

11. *Line 336. Are these in-cloud means?*

Yes, in this study, CDNC is referred to the number concentration of droplets nucleated in-cloud. We added this information in the text.

12. Line 338. Replace “are” with “is”.

Corrected.

13. Line 381. *The grid cell mean is typically less than 1 cm/s in global models. How large is the mean velocity over the central Asian deserts?*

The large scale updraft velocity over the central Asian deserts (e.g., over Gobi) ranges from -0.4 cm s^{-1} to 0.3 cm s^{-1} throughout the year with an annual mean value of 0.01 cm s^{-1} .

14. Line 384. *Is 113 the global annual mean?*

Yes, it is the annual mean over all oceans.

15. Section 3.2 *I’m not sure what the purpose of this section is, since the aerosol and updraft velocity are not evaluated. Are you trying to show that the activation process is realistic, or just that droplet numbers are realistic? I’m not sure that you can achieve the former without validating the aerosol and updraft velocity too (or stratifying droplet number by aerosol and updraft velocity), and the latter is of limited value because EMAC neglects collision (as we learn later).*

Aerosol fields produced by EMAC have been evaluated against in-situ observations in previous studies (Pozzer et al., 2012; Tsimpidi et al., 2014; Karydis et al., 2016). The cloud droplet formation parameterization used in this work has been also extensively evaluated by comparing computations of CDNC and S_{max} and their sensitivity to aerosol properties against detailed numerical simulations of the activation process by a parcel-model (Betancourt and Nenes, 2014a). Furthermore, the cloud-averaged CDNC for stratocumulus clouds, which are described by EMAC, is well captured by the cloud droplet formation parameterization used in this study (Morales et al., 2011). Considering the influence of droplet collision and coalescence processes may, in part, reduce CDNC prediction biases, however, these processes are becoming important in the presence of clouds with substantial amount of drizzle. The purpose of this section is actually to provide a qualitative evaluation of the model’s ability to capture the spatial and temporal variations of CDNC. The model is able to reproduce the increasing CDNC in air masses from clean marine regions to polluted marine and continental regions, though are biased somewhat high over the latter. However, a quantitative evaluation

of the model is not currently feasible since the observations span over a decade (in contrast to the simulation which represents one year) and typically do not represent monthly means over 1.9° grid squares (as sampled from the model results). Furthermore, the model tendency to overestimate the high values of CDNC has small impact on the overall cloud radiative forcing since cloud albedo sensitivity to CDNC decreases with increasing CDNC. Part of this discussion has been added in the revised manuscript.

16. Line 408. In-cloud values?

Yes, in this study, CDNC is referred to the number concentration of droplets nucleated in-cloud. We added this information in the text

17. Line 441. Spatial and/or temporal variability?

Here we refer to spatial variability. We have now clarified this in the text.

18. Line 468. Now we finally learn that collision is neglected in the simulations. This should be noted before the comparisons are presented.

In the revised manuscript, we have also included this information at the beginning of section 3.1.

19. Line 493. This gets confusing. Please be explicit about whether you are referring to addition or subtraction of mineral dust.

We refer to changes caused by the addition of mineral dust particles. This is now explicitly stated in the sentence.

20. Line 502-503. This is the first time we learn about nudging. This should be reported in the experiment design.

We included this information in section 2.1.

21. Section 4.2. This is written very clearly and is quite interesting.

We thank the reviewer for his/her positive comment.

22. Line 607. Over over.

Corrected.

23. *Lines 665-667. Should note again that the simulation neglects droplet collision.*

We noted again that we have neglected the collision and coalescence processes, which can lead to an overestimation of CDNC.