Reviewer 1 comments

Summary

This manuscript performs global model simulations with a simplified dust-specific ice nucleation parameterization, which relates the activation temperature for immersion freezing to dust number concentrations, to investigate the low bias in outgoing shortwave radiation fluxes over the Southern Ocean (SO). After implementing the parameterization into the Met Office's Unified Model, more LWP and less IWP are simulated in the Southern Ocean (SO), along with an increase in cloud albedo. However, the outgoing shortwave radiation fluxes in SO are found to decrease, likely due to a reduction in cloud fraction, which makes the bias over the SO even worse. The authors conduct sensitivity experiments to investigate the cloud fraction decrease.

The question that the authors investigate is important and very interesting. However, unfortunately, the authors seem to have conceptual misunderstanding on the impact of aerosols on ice nucleation process, and thereby the dust-specific ice nucleation parameterization proposed and used in this manuscript is not valid. Besides, I have some concerns related to the interpretation of the results and experiments performed in the discussion. I therefore recommend rejection of this work.

Specific Points

The statement that higher (lower) dust number density results in higher (lower) nucleation temperature is incorrect. It has been well established that the activation temperature for immersion freezing is related to aerosol species, instead of aerosol concentrations. As found by many observational studies, organic and biogenic aerosols tend to nucleate at warmer temperatures, while dust particles have lower activation temperatures. Therefore, the parameterization proposed in this paper that relates the activation temperature to dust concentrations is not valid. Even if the parameterization is valid, the authors should explain why they choose this formula and evaluate it against observations. This is the major reason for my rejection of this work.

We would like to thank the reviewer for the very helpful review. After going through the comments, we realised that the message we were trying to convey has not been captured well in the submitted version of the manuscript. So, we have restructured the manuscript to give more clarity to the focus of this study. We hope that this will address many of the reviewer comments as well. We would like to clarify that we were not trying to include a new INP parametrisation as such in the Unified Model rather introducing a workaround for the lack of INP functionality in the global version of our model. This study is more like a follow up to the Varma et al., 2020 in the sense that Varma et al., 2020 had a hemispherical impact in terms of supercooled liquid content and cloud brightness. We wanted to make a targeted response over the SO region alone. As now mentioned and emphasized in the revised manuscript, if there already was an INP recognised in the model, this would have been relatively straightforward. Since the Unified Model (at the resolution we use) does not have an INP parametrisation presently available, we just implemented the prognostic dust approach so that there is more regional distribution of heterogeneous nucleation temperatures and hence ice/liquid cloud formation. As a result of this parametrisation, we now have more targeted response on SCL and in-cloud albedo over the SO region. We now have restructured the manuscript to take these into account. Most importantly, we have also added an interim comparison of our parametrisation with that of Demott et al., 2010 to show that they are agreeable.

It is also not clear to me why the authors link the dust concentrations and the activation temperature of heterogeneous ice nucleation to the detrainment temperature in convection scheme. In other words, how is the detrainment process related to primary ice formation in the convection system?

As noted in Section 3, since in-cloud micro-physics and convective clouds are treated separately in almost all of the low-resolution GCMs, immersion freezing is also parametrized separately in micro-physics and convection schemes. While in-cloud micro-physics predicts the cloud phase, the convection scheme provides a temperature dependent threshold for the detraining of ice (e.g. Kay et al 2016). As a result, along with the micro-physics scheme, the convection scheme also plays a role in determining the ice formation in the model through detrainment temperatures.

Actually, there are many dust-specific ice nucleation parameterizations that are ready to use (e.g., Atkinson et al., 2013; DeMott et al., 2015; Hoose et al., 2010; Knopf and Alpert, 2013; Niemand et al., 2012; Ullrich et al., 2017; Wang et al., 2014). These parameterizations are derived based on either observational or theoretical evidences. They have also been implemented into regional and global models. The authors may want to use these parameterizations in their future work.

As added now in the Introduction section, in order to implement and thoroughly examine the impact of dust as INP on cloud radiation properties as per these existing parametrisations, ideally state-of-the-art atmospheric models with extensive double-moment bulk micro-physics schemes or comprehensive aerosol models that allow the identification of aerosol species and number densities etc are desired. However, for low-resolution GCMs (like ours), this is not currently available. Our model does not identify the dust species or number densities but rather provide the mass mixing ratios based on representative diameters belonging to 6 size bins. This makes any direct comparison practically impossible. Also, we have clarified that our motive hence is not a new INP parametrisation. However, there are currently ongoing developments on the implementation of a GLOMAP dust scheme (which allows the speciation of dust and use/comparison of some of the existing dust INP parametrisations feasible in the future). Also, please see the response to 'Specific Points'.

The authors should evaluate the modeling results against observations, before concluding if the new parameterization leads to any improvements in the model. For example, the authors can use MODIS LWP, CloudSat IWP, and MODIS cloud fraction. It would also be interesting to compare the simulated shortwave and longwave cloud forcing (SWCF and LWCF) with CERES-EBAF dataset. If possible, the authors may also evaluate the simulated dust and INPs in SO. For dust, the authors can use CALIPSO dust extinction vertical profiles. For INPs, a lot of field measurements are available in SO, e.g., CAPRICORN campaign (McCluskey et al., 2018).

We have now added comparison of SW TOA/LWCF/SWCF with CERES data in the Supplementary material.

To investigate the cloud fraction decrease in exp_{dust} over SO, the authors include the comparisons between expcap and control in their discussion. However, expdust and expcap are two experiments with different modifications in the microphysical processes. What happened in expcap should not be expected in expdust. Therefore, such comparisons do not help to understand the cloud fraction decrease in expdust. The authors should instead look into the changes in RH, precipitation, and probably lower-tropospheric stability (LTS) in expdust.

We have now removed the results from the capacitance experiment in the Discussion section. We have also moved the additional experiment to the Supplementary material.

The sensitivity experiment, expeff, is not carefully designed. Why do you assume the liquid clouds are equally spread as the ice cloud in the convection scheme? Does this assumption make the model more physically correct? Are there any previous literatures that can support your assumption? Also, it is not fair to compare the DJF results in expeff with the annual mean results in expdust.

We have now completely removed the mention of exp_{eff}. As the model tuning is an ongoing process, we have added another sensitivity study with results included in the Supplementary material.

Other comments

Line 38: "... can proceed quicker ...". It should be "proceed at warmer temperatures".

Modified

Section 2: It would be better to include how dust is parameterized in this section.

Section 3: The word "prognostic-dust parameterization" in the title of this section sounds like a dust transport parameterization. Please consider to replace it by something like "dust-specific ice nucleation parameterization".

We have changed the section heading to "Nucleation temperature as function of dust distribution: Experimental design"

Eq (1). How do you get the ice nucleation concentrations or the immersion freezing rate from thetrn.

As mentioned earlier, we have now made it clear that we are not introducing an INP parametrisation. Also, please see the additional section in the Supplementary material showing comparison with Demott et al., 2010.

Line 162: "..., probably accounting for ... than before". This sentence is not clear to me.

The sentence has been modified.

Line 164: Why do you show the IWP and LWP for stratocumulus boundary layer clouds only? Why not show those for the whole column?

The focus of IWP/LWP in this study is on the stratocumulus boundary layer type clouds (in lines with Varma et al., 2020 study). We have now made it clear in the manuscript. The IWP/LWP plots through the entire cloud types/column are now included in the Supplementary material.

Line 211-213: How do you know the liquid cloud fraction is smaller than the ice cloud fraction? What about the mixed-phase clouds? Also, the explanation in the second sentence does not make any sense.

This has been modified.

Figure 6. It would be better to give a subtitle for each panel.

Added

References

Atkinson, J. D., Murray, B. J., Woodhouse, M. T., Whale, T. F., Baustian, K. J., Carslaw, K. S., Dobbie, S., O'Sullivan, D., Malkin, T. L. (2013). The importance of feldspar for ice nucleation by mineral dust in mixed-phase clouds. Nature, 498(7454), 355–358. https://doi.org/10.1038/nature12278

DeMott, P. J., Prenni, A. J., McMeeking, G. R., Sullivan, R. C., Petters, M. D., Tobo, Y., Niemand, M., Möhler, O., Snider, J. R., Wang, Z., Kreidenweis, S. M. (2015). Integrating laboratory and field data to quantify the immersion freezing ice nucleation activity of mineral dust particles. Atmospheric Chemistry and Physics, 15(1), 393–409. https://doi.org/10.5194/acp-15-393-2015

Hoose, C., Kristjánsson, J. E., Chen, J.-P., Hazra, A. (2010). A Classical-Theory-Based Parameterization of Heterogeneous Ice Nucleation by Mineral Dust, Soot, and Biological Particles in a Global Climate Model. Journal of the Atmospheric Sciences, 67(8), 2483– 2503. https://doi.org/10.1175/2010JAS3425.1

Knopf, D. A., Alpert, P. A. (2013). A water activity based model of heterogeneous ice nucleation kinetics for freezing of water and aqueous solution droplets. Faraday Discussions, 165, 513. https://doi.org/10.1039/c3fd00035d

McCluskey, Hill, T. C. J., Humphries, R. S., Rauker, A. M., Moreau, S., Strutton, P. G., Chambers, S. D., Williams, A. G., McRobert, I., Ward, J., Keywood, M. D., Harnwell, J., Ponsonby, W., Loh, Z. M., Krummel, P. B., Protat, A., Kreidenweis, S. M., DeMott, P. J. (2018). Observations of Ice Nucleating Particles Over Southern Ocean Waters. Geophysical Research Letters, 45(21), 11,989-11,997. https://doi.org/10.1029/2018GL079981

Niemand, M., Möhler, O., Vogel, B., Vogel, H., Hoose, C., Connolly, P., Klein, H., Bingemer, H., DeMott, P., Skrotzki, J., Leisner, T. (2012). A Particle-Surface-Area-Based Parameterization of Immersion Freezing on Desert Dust Particles. Journal of the Atmospheric Sciences, 69(10), 3077–3092. https://doi.org/10.1175/JAS-D-11-0249.1

Ullrich, R., Hoose, C., Möhler, O., Niemand, M., Wagner, R., Höhler, K., Hiranuma, N., Saathoff, H., Leisner, T. (2017). A New Ice Nucleation Active Site Parameterization for Desert Dust and Soot. Journal of the Atmospheric Sciences, 74(3), 699–717. https://doi.org/10.1175/JAS-D-16-0074.1

Wang, Y., Liu, X., Hoose, C., Wang, B. (2014). Different contact angle distributions for heterogeneous ice nucleation in the Community Atmospheric Model version 5. Atmospheric Chemistry and Physics, 14(19), 10411–10430. https://doi.org/10.5194/acp- 14-10411-2014