Atmospheric Chemistry and Physics Discussions

Atmos. Chem. Phys. Discuss., author comment AC1
https://doi.org/10.5194/acp-2021-509-AC1, 2021

Reply on RC1

Manu Anna Thomas et al.

Author comment on "Marine aerosol properties over the Southern Ocean in relation to the wintertime meteorological conditions" by Manu Anna Thomas et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2021-509-AC1, 2021

Response to Reviewer #1

This study presents a detailed investigation into sea spray aerosol properties over the Southern Ocean. Sea spray aerosol is an important contributor to total aerosol radiative forcing and poorly constrained in climate models; hence how it may change in a warming world is not well understood. The analysis is thorough and gives a comprehensive picture of sea spray aerosol properties (which are assumed to be the dominant source of marine aerosol during austral winter).
We thank the referee for the encouraging words and constructive suggestions. Please find below point by point reply to your comments.

Where I would like to see improvement (which I think will also enhance the impact of the paper), is in thinking about how the results are meaningful. They are nicely summarised in the conclusions, but a discussion/synthesis regarding importance and outlook is missing. The authors discuss that climate models represent SSA poorly in the introduction (lines 90-109) – can the work be linked back to that discussion, for example? Do the results shed new insight into what climate models are doing wrong and how SSA parametrizations could be improved? The end of the introduction (lines 121-127) could also be modified to address the significance of the objectives.
The following text will be added to the end of the 'Conclusions' section as a synthesis of the work carried out here and the outlook.
"This study elaborates on the relative importance of RH on the optical properties of sea salt over the Southern oceans during SH winter months when other natural emissions and long range transport of anthropogenic emissions are negligible. Although a handful of in situ measurements have been carried out using ship cruises and aircrafts, this is the first study of its kind that has attempted a detailed mapping of the optical properties of sea salt in its natural environment on the basis of the three fundamental parameters that the emissions depend upon - RH, wind speed and sea surface temperature and their vertical distribution purely from satellites. The generation of sea salt aerosols in the models are described in terms of source functions that depend on either one or more of the factors mentioned above for varying size ranges and these are derived from lab or ship/aircraft measurements. These source functions are adapted depending on the model to get the emissions in the correct order so as to accurately estimate the total aerosol optical depth (for example, some models introduce a temperature dependency or alter the power factor so as to

increase/decrease the wind speed dependency or include the super micron size particles in the calculations). Also, due to the high hygroscopicity of sea salt, the parameterization schemes simulate the emissions at 80% RH (Monahan, et al., 1986). To introduce the variability of the water uptake by dry sea salt particles, recent models use hygroscopic growth factors at varying RH that are prescribed (Chin et al., 2002) or that are based on empirical equations or are explicitly calculated (Ghan et al., 2001; Vignati et al., 2004). The dependency of the aerosol optical properties on RH is of profound importance for climate forcing estimates, particularly, in the case of sea salt as they can co-exist in both phases at the same time between 50% and 70% RH. And this dependency changes when sea salt particles are contaminated by other chemical species. This study here shows the variability in the sea salt aerosol optical properties on a range of observed relative humidities, water temperatures and wind speeds over the Southern Oceans. In this study, we also generate fitting functions for aerosol extinction and backscatter based on wind speed and relative humidity for naturally occurring sea salt which can in turn, help improve the source functions used in the sea salt parameterization schemes. Further insights are needed to map the behaviour of the optical properties of sea salt when they are coated with other naturally occurring chemical components as is in the case of the rest of the oceans. "

Methods – I was interested to know how often cloud-free pixels were found in the data – after fulfilling all the selection criteria, what fraction of data are included in the analysis?
The total cloud cover over the Southern Ocean ranges from 75%-85% depending on the winter month and region. However not all clouds are optically thick and the CALIOP lidar is able to see through the thin clouds providing retrievals in nearly 40-50% of the cases. Here we have therefore used all-sky retrievals. The criterion of using only those retrievals that successfully converge without the change in initial lidar ratio (i.e. Extinction quality flag == 0) results in retaining nearly all aerosol retrievals. The cases when the lidar ratio was changed were less than 1% in total. Among all the retrievals available to compute mean climatological features, the scenarios representing the enhanced cyclonic and anticyclonic conditions represented about 27% and 17% of those cases respectively. We have added this text in the revised manuscript.

Lines 158: A reference is needed to justify the use of the 700 hPa GPH. Eg
https://agupubs.onlinelibrary.wiley.com/doi/full/10.1029/2005GL022419
or https://journals.ametsoc.org/view/journals/clim/18/5/jcli-3284.1.xml
These references are added in the revised manuscript.

In the paper's present form, the readers are supposed to take it as read that the 25th and 75th percentile thresholds of GPH correspond to ascending and descending air masses; further explanation/references are needed here, which links to my next point.
We have now clarified in the manuscript that 25th/75th percentile corresponds to ascending/descending airmasses. The figures and text are adapted to follow only one set of terminology - ascending and descending airmasses.

Line 184. I follow the logic here, but because cyclonic conditions are defined as less than the 25th percentile in GPH, it's slightly confusing that they are also representative of the mean state. A plot of the GPH distribution (either in the Methods or Supplementary info) would help.
Southern Oceans are predominantly a region of intense extratropical cyclonic activity. More than half of the winter cyclones have a structure that extends through to the lower atmospheric levels (Simmonds and Keay 2000a; Hougthon et al. 2001; Lim and Simmonds 2002; Lim and Simmonds, 2007). Hence, it is not surprising that the 25th percentile of GPH that corresponds to cyclonic

conditions is representative of the mean state.

A plot of the GPH distribution is added to the revised manuscript.

Figures: the use of the red/green colormap cannot be interpreted by those with color blindness; see e.g. https://www.scientificamerican.com/article/end-of-the-rainbow-new-map-scale-is-more-readable-by-people-who-are-color-blind/
A different colormap is used and all the figures are modified accordingly.
Instead of labelling the figures with 'P25' and 'P75' in the legend, using the labels 'ascending' and 'descending' (or similar) would make interpretation easier.
The labels are re-written in the revised manuscript.

Minor comments:
L45. Aerosol radiative forcing has been updated toward more negative values, and is estimated to be between -2 to -0.4 W m-2 (90% likelihood).
https://agupubs.onlinelibrary.wiley.com/doi/full/10.1029/2019RG000660
Replaced the sentence by: "A recent study reports the global mean total effective aerosol radiative forcing to be in the range of -2.0 to -0.35 W/m2 when constrained by observations, with estimates of aerosol-cloud interactions in the range of -2.65 to -0.07 W/m2 (Bellouin et al., 2020)"

L203-204. Definitions of efflorescence and deliquescence are given here; it would be helpful to include these in the introduction also, around lines 59-65.
The definitions are added in the introduction.

L 80-81. Unclear – at what wind speeds are 26% of the accumulation mode particles sea salt?
This information is added and the sentence is rephrased as: "It was observed that 26% of the accumulation mode particles below 200 m was sea salt and this fraction increases with wind speed by 11% for wind speeds less than 4 m/s and respectively, 20% and 30% at 4-8 m/s and 8-12 m/s."

L146-147. I assume that avoiding the sea-ice zone further reduces the risk of contamination from DMS-derived sulfate aerosol (which should have low concentrations during austral winter anyway).
Yes, that is one of the reasons to avoid the sea-ice zones. The other reason is to avoid potential contamination and misclassification due to wind-blown snow/ice crystals in CALIOP-CALIPSO retrievals. This is already mentioned in the manuscript under Section 2.

Line 198. Further explanation needed – why is it remarkable that linear depolarization is not sensitive to varying winds?
We find it remarkable from two perspectives: First, as the wind speed increases, one would expect drying of sea salt particles and hence, higher linear depolarization. This is especially true above the boundary layer where not only the winds are stronger but the humidity is lower. Second, models for optical properties of marine aerosols show that the linear depolarization ratio is often increasing with the volume-equivalent particle radius - see, e.g. https://agupubs.onlinelibrary.wiley.com/doi/full/10.1029/2020JD033674 Correspondingly, one would expect a generally increasing trend of the depolarization ratio with wind speed.
We have clarified these points in the revised manuscript.

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