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Interactive comment on "The value of remote marine aerosol measurements for constraining radiative forcing uncertainty" *by* Leighton A. Regayre et al.

Anonymous Referee #2

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The authors present an interesting approach by which large PPEs of the UK Hadley Centre General Environment Model (HadGEM3) are performed in order to sample the parametric uncertainty of nearly 30 model parameters to address the following questions:

i) To what extent can measurements of aerosols in pristine (natural) environments help to constrain model simulations and thereby reduce the large uncertainty in aerosol forcing? ii) What is the relative importance of measurements in remote and polluted environments for constraining the forcing uncertainty?

Using Southern Ocean ship based measurements (CCN0.2%; CCN1%; mass concen-

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Discussion paper

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trations of non-sea salt sulfate in PM10 and N700) as model constraints, the authors demonstrate a reduction in model parametric uncertainty (most notably: sea spray emissions and dry deposition velocity) that in turn reduces the aerosol forcing uncertainty from the original PPE.

The paper is generally well written, however, the methodology I find to be lacking detail in key areas. The results are generally well presented, however, given the focus of the paper is on using remote marine measurements as constraints, I would like to see a more rigorous presentation of these measurements in the main study or SI.

Finally, the conclusions presented in this study are in places not supported by the data or references. I believe that this is in part due to a lack of clarity in presentation of the methodology and assumptions therein, and in part due to limitations associated with applying pre-existing PPEs that were perhaps not specifically designed to investigate the two key questions listed above.

Due to the computationally demanding nature of running such aerosol PPEs using one set to probe a number of interesting questions associated with aerosol uncertainty is understandable, as are the potential limitations. The discussion and conclusions presented by the authors could be improved by linking to any such limitations or assumptions.

General comments:

1. Fig. 1(b/d), please overlay measured standard deviation as dots, as performed for the average of the measurements (a/c).

2. The authors are focussing on natural aerosol. How were any ship measurements influenced by anthropogenic pollution eliminated from the analysis?

3. Is each measurement used given equal weighting in constraining the model?

4. SI: The authors state: “The variance terms in the denominator of Eq. (1) are calculated uniquely for each measurement. Following Johnson et al., (2019), we use a mea-

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surement uncertainty of 10%”. Are the measurement errors for the constraints used in this study homoscedastic or heteroscedastic? Do they correspond with the definition of the implausibility metric (eq. 1, SI)? How does the variability in the measurements compare to the uncertainty chosen (10%)?

5. CCN0.2% and CCN1% are used as observational constrains in the study. The measurement study in which these constraints were taken from measured CCN at more than two supersaturations. Why was a CCN spectra (or measured aerosol size distribution) not used from the observations to provide a tighter constraint on the model?

6. Please provide more detail on the observations used as constraints in the SI, linking clearly to Fig. 1 in the main article. For example, demonstrate a time-series of one of the observation dots in Fig. 1 graphically, including the variability (bars representing standard deviation), and colour of dotted time-series representing position. Clearly link this graphic to the mathematical construction of the model constraint e.g. implausibility metric in the SI.

7. The authors use four measurements as a constraint (listed above). Which measurements provided the highest information content for model constraint? I would like to see some discussion on the relative constrain the individual measurement parameters provided o the model. This would help inform future measurement campaigns in this region on key measurement parameters. For example, the authors state (SI): “Non-sea-salt sulfate was calculated by subtracting this fraction from the total particulate sulfate”. How much extra constraint on the parameters (Fig. 3) is provided by using both N700 and Nss-sulfate as constraints, over just one of these.

8. The authors provide the unconstrained and constrained model PDFs of the aerosol properties. A uniform prior range is assumed in this method. How does this represent the observations? Please show a PDF of the observed distributions to see if this is a true representation of the ship observations.

9. The authors have shown how the aerosol parameters are constrained using obser-

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vations, and subsequently the reduction in forcing uncertainty from the original PPEs. The paper is missing some discussion on the linkage between the constraint of these parameters and forcing. Inclusion of this would be very beneficial to the community. For example, how has average cloud microphysical properties –e.g. cloud droplet concentrations been constrained following the constraints shown in Fig. 2? Do they compare better, or worse with satellite observations in the region? This would help inform whether the constrain on forcing represents a true constraint on the aerosol processes (i.e. is the constraint of CCN by scaling sea salt right for the right reasons, or should the results be presented/interpreted as a tuning...?).

10. What is the average supersaturation over the Southern Ocean simulated by the model? How does this correspond with the selected value of CCN0.2% as representative for (cloud-active aerosol, SI) in the region?

11. The authors make clear that they are targeting parametric uncertainty, and the method does not address model structural uncertainty. However, some of the conclusions presented rely too heavily on the information provided by the parametric uncertainty analysis alone, specifically in the comparison to Revell et al., (2019) (Line 166 and thereafter). The differences in conclusions related to over/underestimation of sea spray aerosol are attributed to a lack of sampling of aerosol processes by Revell et al., 2019. A discussion on the role of structural errors in the model used by the author would be is required. What are the key differences between the model configurations with respect to representation of marine aerosol sources and sinks? What is the relevant contribution to aerosol mass from secondary vs. primary marine aerosol sources in the two model configurations?

12. Given the use of an older configuration of the model HadGEM by the authors, the results should be presented in light of the latest configuration. Stars showing the values for the parameters overlaid on Fig.3/5 that represent the configuration used by Revell et al., 2019 should be included to aid the reader in understanding differences found between the two studies with regard to sea salt emissions.

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13. How much of the constraints found in Fig.3 are due to compensating parameters across the multi-dimensional marginal probability distributions? For example, what is the relationship between the marginal distributions between dry deposition and sea salt? Could the authors also provide an investigation of the joint marginal histograms between DMS and sea salt emission.

14. It is stated that the "model-measurement comparison is improved when aerosol number concentrations and mass are relatively high". Does the model configuration used have the same total sources of aerosol number/mass compared to the configuration of the model used by Revell et al., 2019? This could be included in the SI. Are there any other potential marine aerosol sources currently missing in the model configuration used by the authors that would increase aerosol number/mass by a similar magnitude than scaling sea salt emissions to 3 times the default value? This requires discussion, in particular in light of the conclusions presented by the study cited for the source of the observations (Schmale et al., 2019) used by the authors, e.g.:

Schmale et al., 2019: "The regions of highest underestimation are close to the coast of Antarctica during leg 2, close to South Africa and around 45°E during leg 1. These regions coincide with the highest concentrations of gaseous MSA . . . This preliminary model–measurement comparison suggests that the model may be missing an important source of high-latitude CCN."

15. SI: The authors state that the wind speed discrepancies do not affect the results presented. This is an important statement that deserves more detailed justification as I currently do not see how this is supported by the data or Korhonen et al., 2010. How do the differences in simulated and observed wind-speeds relate to the scaling of sea salt required to constrain CCN?

16. SI: The authors nudge the models to 2008 meteorology from reanalysis data. A comparison between the meteorological data between the measurement years and that used in the model simulation should be provided in the SI, comparing both monthly

averages and variability.

17. SI “Marginal parameter distributions are constrained consistently when we remove measurements with average wind speed differences larger than 50% of the measured value from the model-measurement comparison.” How many results does this effect? Please show a global map where the grid-box colour represents a measure of how often this threshold is exceeded.

Technical comments:

1. Line 178: “The constraint on the scaled DMS emission flux is two-sided, 179 reducing the credible range of DMS emission scaling from 0.5 to 2.0 down to 0.54 to 1.9.” Could the authors please make clear what in the figure 0.54/1.9 corresponds to.

2. SI, Line 95: Grammar - “pdfs with centralised tendencies will by heavily weighted”. Change by to be.

3. SI, Line 63: “We make use of the ATM and AER-ATM perturbed parameter ensembles (PPEs)”. Following this the authors refer only to AER and AER-ATM. Should this read: “We make use of the AER and AER-ATM”?

4. Fig. 2: Should y-axis density not be labelled 0-1? Or are these not normalised marginal densities.

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