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The value of remote marine aerosol measurements for constraining radiative forcing uncertainty

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Abstract. Aerosol measurements over the Southern Ocean are used to constrain aerosol-cloud interaction radiative forcing uncertainty in a global climate model. Aerosol forcing uncertainty is quantified using one million climate model variants that sample the uncertainty in nearly 30 model parameters. Ship-based measurements of cloud condensation nuclei, particle number concentrations and sulfate mass concentrations from the Antarctic Circumnavigation Expedition: Study of Preindustrial-like Aerosols and Their Climate Effects (ACE-SPACE) are used to identify observationally implausible variants and thereby reduce the spread in the simulated forcing. Southern Ocean measurements strongly constrain natural aerosol emissions: default sea spray emissions in the model need to be increased by around a factor of 3 to be consistent with measurements. Aerosol forcing uncertainty is reduced by around 7% using these measurements, which is comparable to the 8% reduction achieved using an extensive set of over 9000 predominantly Northern Hemisphere measurements. The radiative forcing due to aerosol-cloud interactions (RF_{aci}) is constrained to -2.61 to -1.10 W m^{-2} (95% confidence) and the effective radiative forcing from aerosol-cloud interactions (ERF_{aci}) is constrained to -2.43 to -0.54 W m^{-2}. When Southern Ocean and Northern Hemisphere measurements are combined, the uncertainty in RF_{aci} is reduced by 21% and the strongest 20% of forcing values are ruled out as implausible. In this combined constraint the observationally plausible RF_{aci} is around 0.17 W m^{-2} weaker (less negative) with credible values ranging from -2.51 to -1.17 W m^{-2} and from -2.18 to -1.46 W m^{-2} when using one standard deviation to quantify the uncertainty. The Southern Ocean and Northern Hemisphere measurement datasets are complementary because they constrain different processes. These results highlight the value of remote marine aerosol measurements.

1 Introduction

The uncertainty in the magnitude of the effective radiative forcing caused by aerosol-cloud interactions (ERF_{aci}) due to changing emissions over the industrial period is around twice that for CO_2 (Stocker et al., 2013). It is essential to reduce this uncertainty if global climate models are to be used to robustly predict near-term changes in climate (Andreae et al., 2005, Myhre et al., 2013, Collins et al., 2013, Tett et al., 2013, Seinfeld et al., 2016).

Aerosol forcing uncertainty has persisted in climate models since the 1990s partly because there are no measurements covering the industrial period that can be used to directly constrain simulations of long-term changes in aerosol and cloud properties (Gryspeerdt et al., 2017; McCoy et al., 2017). Estimates of aerosol

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forcing over the industrial period therefore rely on models that have been evaluated against measurements made in the present-day atmosphere. However, it is known that the aerosol forcing (in particular the component caused by aerosol-cloud interactions) depends sensitively on the state of aerosols in the pre-industrial period (Carslaw et al., 2013; Wilcox et al. 2015) when natural aerosols were dominant (Carslaw et al., 2017). Observations of natural aerosols in the present-day atmosphere are therefore expected to help constrain the simulated forcing unless there have been significant changes in natural aerosol processes over the industrial period, for which there is little evidence (Carslaw et al., 2010).

In this paper we address the 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? It is known that the abundance of natural aerosols affects the magnitude of forcing in a model (Spracklen and Rap, 2013; Carslaw et al., 2013). However, to assess the effect on the *uncertainty* in forcing it is necessary to explore how the spread of predictions of a set of models changes when constrained by measurements. The 5th Coupled Model Intercomparison Project is inadequate for this purpose because of insufficient aerosol diagnostics (Wilcox et al., 2015). Here we use large perturbed parameter ensembles (PPEs) of the UK Hadley Centre General Environment Model HadGEM3 (Hewitt et al, 2011). The PPEs were created by systematically perturbing numerous model parameters related to natural and anthropogenic emissions and physical processes. The simulated aerosol forcings have uncertainty ranges that exceed those of multi-model ensembles (Yoshioka et al., 2019; Johnson et al., 2019). Instantaneous radiative forcing (RF) is quantified using the 26-parameter AER PPE in which just aerosol-related parameters were varied, and the effective radiative forcing (ERF) is quantified using the 27-parameter AER-ATM PPE in which aerosol and physical atmosphere parameters were varied (Yoshioka et al., 2019). We use these PPEs to quantify how the constraint provided by pristine aerosol measurements affects the spread of aerosol forcings simulated by the ensembles.

Previous analysis of HadGEM3 PPEs showed that measurements of the present-day atmosphere in regions affected by anthropogenic emissions have limited impact on the uncertainty in simulated aerosol forcing. For example, Regayre et al., (2018) showed that top-of-the-atmosphere shortwave radiation flux measurements reduce ERF_{aci} uncertainty by only around 10%, despite the fluxes in the present-day and early-industrial environments sharing multiple causes of uncertainty. Johnson et al. (2019) showed that a much larger dataset of over 9000 (predominantly Northern Hemisphere) aerosol measurements constrained the global, annual mean aerosol RF uncertainty by only around 8%. These measurements reduce the uncertainty in a small number of parameters related to anthropogenic emissions and aerosol processing in polluted environments. However, important causes of uncertainty in RF_{aci}, such as natural aerosol emission fluxes, were largely unconstrained.

The Southern Ocean is one of the few regions on Earth (along with some boreal forests) in which the same processes are expected to affect cloud-active aerosol concentrations in the present-day and early-industrial atmospheres (Hamilton et al., 2014). In this study we make use of aerosol measurements from the Antarctic Circumnavigation Expedition: Study of Preindustrial-like Aerosols and Their Climate Effects (ACE-SPACE) campaign (Schmale et al., 2019). They offer a unique opportunity to constrain the early-industrial aspects of aerosol forcing uncertainty because the Southern Ocean is a source of natural aerosols that are relevant at the global scale and remains largely unaffected by anthropogenic aerosol and precursor emissions.

We use near-surface measurements of cloud condensation nuclei concentrations at 0.2% and 1.0% supersaturations ($CCN_{0.2}$ and $CCN_{1.0}$; Tatzelt et al., 2019), as well as mass concentrations of non-sea-salt sulfate in PM_{10} and number concentrations of particles larger than 700 nm (N_{700}; Schmale et al., 2019a). The measurements are compared to output from 1 million variants of the HadGEM3 model that sample combinations of parameter settings in the model. These model variants are used to represent aerosol forcing uncertainty in our model using probability density functions (pdfs) and were generated by sampling from Gaussian Process emulators that were trained on the PPE model outputs (see SI Methods). Model variants that were judged to be observationally implausible against the measurements were rejected, resulting in a set of plausible variants from which the uncertainty in aerosol forcing could be computed (see SI Methods). In the results shown below, we retained approximately 3% of model variants (following Johnson et al., 2019) that best match all four measured aerosol properties.

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2 Results

Fig. 1 shows the $CCN_{0.2}$ mean and standard deviation from the unconstrained and constrained model variants to exemplify the effect of constraint on model output. The mean concentrations in the unconstrained sample are much smaller than measured concentrations. However, the range of $CCN_{0.2}$ values in the unconstrained sample spans the measurements in most locations (Fig. 1b). The measurement constraint increases $CCN_{0.2}$ concentrations (more than double the unconstrained mean in many locations; Fig. 1c) and greatly reduces the $CCN_{0.2}$ uncertainty (by more than half everywhere to less than 50 cm^{-3}; Fig. 1d).

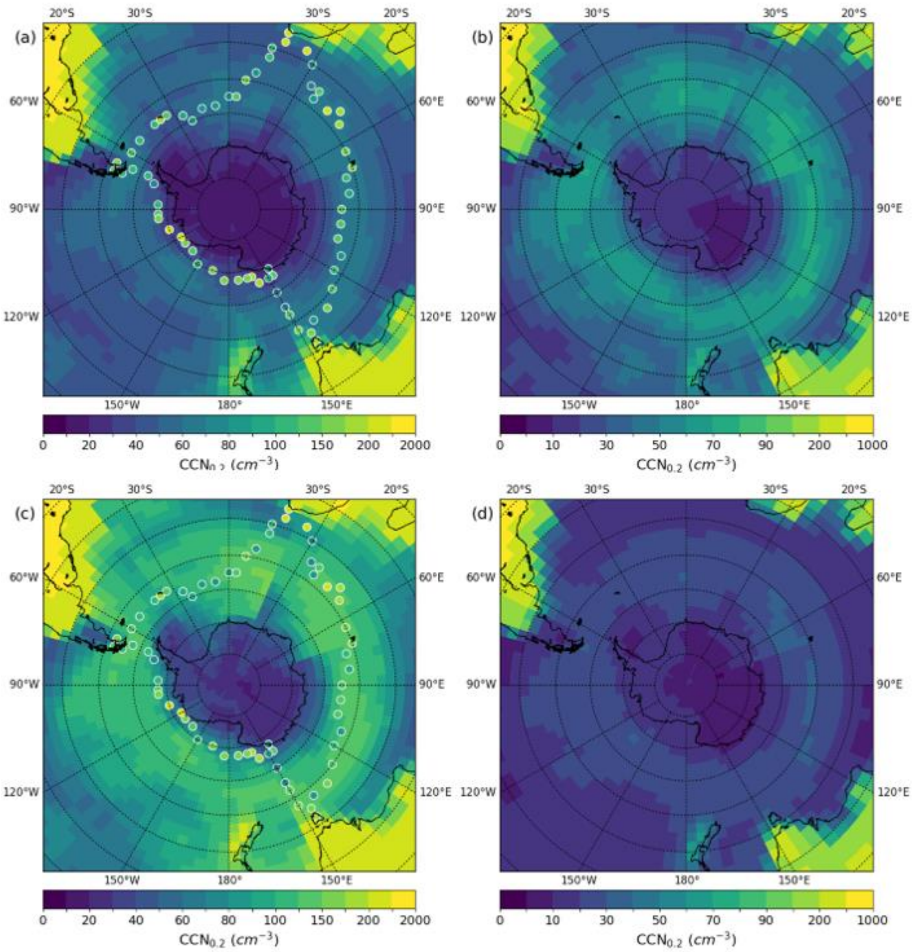

Fig. 1. a,c) Mean and b,d) standard deviation of $CCN_{0.2}$ concentrations from the a,b) unconstrained sample and c,d) the sample constrained using concentration measurements of $CCN_{0.2}$, $CCN_{1.0}$, non-sea-salt sulfate and particle numbers larger than 700 nm. Measured $CCN_{0.2}$ values are plotted as dots. Means and standard deviations were calculated using samples taken from emulators trained using monthly mean values. December to March sample values were combined based on longitudinal agreement with measurements.

Fig. 2 shows pdfs of the output from the model for the four variables used as constraints, calculated as means over the locations where measurements were taken. The constraint reduces the uncertainty in all measurement types (narrower pdfs) and the central tendency of the pdfs is closer to the regional mean of measurements after

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constraint. Rejecting around 97% of model variants as implausible compared to measurements greatly improves the model-measurement comparison.

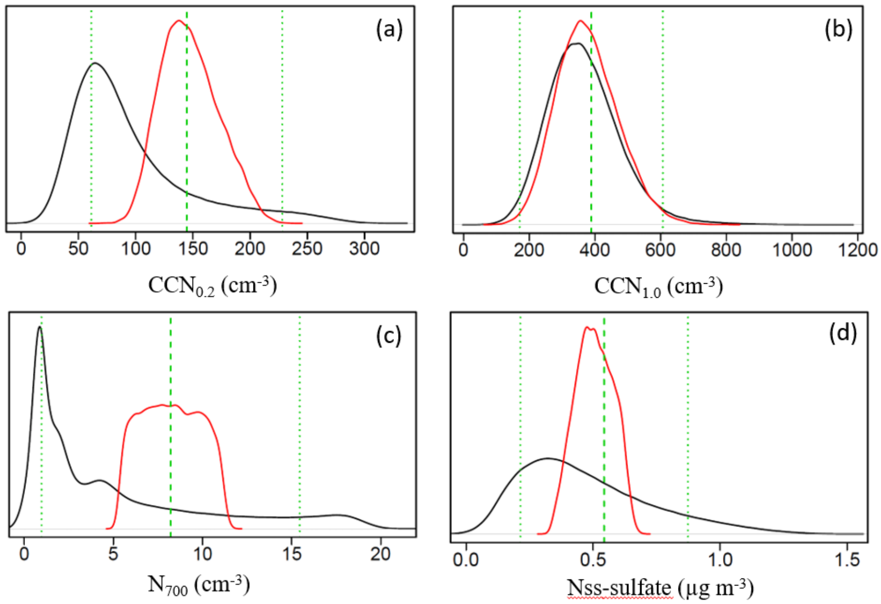

Fig. 2. Unconstrained (black) and observationally constrained (red) pdfs of aerosol properties: a) $CCN_{0.2\%}$, b) $CCN_{1.0\%}$, c) N700 and d) aerosol sulfate. The pdfs were calculated at locations where measurements were used for constraint across the months December to March. The green dashed line shows the median of the measurements and the dotted green lines show the approximate uncertainty ranges that were accounted for in the constraint (See SI Methods).

After constraint, the remaining model variants inhabit specific parts of the 26-dimensional parameter uncertainty space used to quantify the model uncertainty. We explore the effect of constraints on parameter values using 1-dimensional marginal probability distributions (described in detail in Johnson et al., 2019) – see Fig. 3 and Fig. S1 for equivalent AER-ATM results. The magnitude of the marginal probability distribution after constraint reflects the number of ways in which a particular value of a parameter can be combined with settings of all the other parameters to produce an observationally plausible model. The white space in the marginal pdfs shows where parameter value density has decreased.

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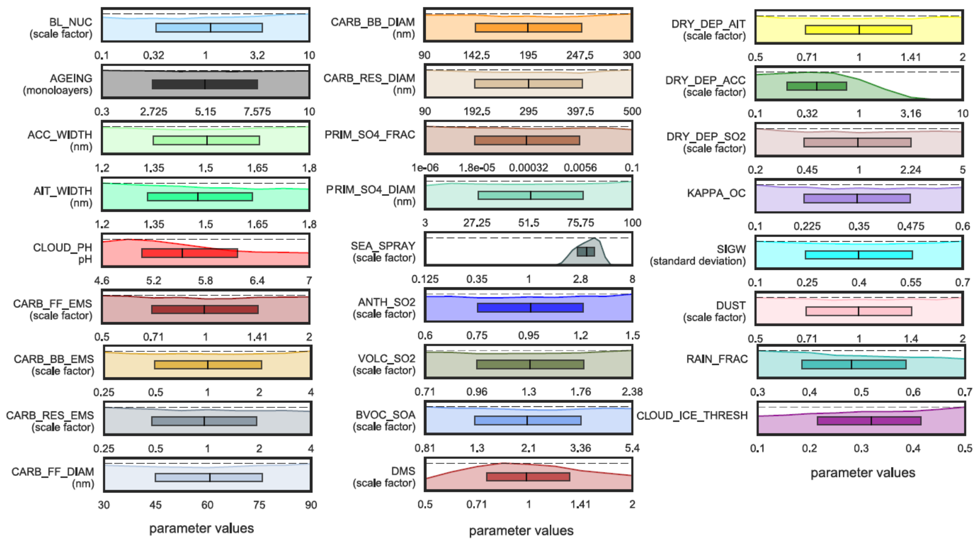

Fig. 3. Marginal probability distributions for the 26 aerosol parameters after constraint using ACE-SPACE measurements. The density of parameter values in the unconstrained sample are shown as dashed lines. Densities of constrained samples are shown in colour and are scaled so that the maximum densities in the constrained and unconstrained samples are aligned. The 25th, 50th and 75th percentiles of each marginal distribution are shown in the central boxes. Parameter values on the x-axes correspond to values used in the model (Yoshioka et al., 2019).

The relative simplicity of aerosol emissions and processes over the Southern Ocean (compared to polluted continental regions) means that measurements can be used to tightly constrain uncertainty in the associated parameters. Two parameters, sea spray emissions and dry deposition velocity, are tightly constrained such that some parameter values are ruled out as implausible. Several other parameters (related to cloud droplet pH, DMS emissions and wet deposition) are more modestly constrained. These constraints suggest the model-measurement comparison is improved when aerosol number concentrations and mass are relatively high.

Sea spray emissions are tightly constrained to be around 3 times larger than the default model value. Observationally plausible values of the sea spray scaling parameter range from around 1.6 to 5.1 and all other values (including the default emission calculated in the model) are ruled out as implausible. This suggests that sea spray emissions in our model need to be significantly higher than those calculated using the wind speed dependent Gong (2003) parametrisation. We do not make any assumptions about the composition of these additional sea spray particles. They may be rich in organic material as proposed by Gantt et al., (2011) which would alter the CCN activity of emitted particles. However, the consistency of constraint of CCN0.2 and N700 towards higher values (Fig. 1) implies that a general scaling of the existing sea spray flux is consistent with the measurements without the need for an additional source of fine-mode, organic-rich particles.

These results conflict with the findings of Revell et al. (2019) who suggest the relatively simple wind speed dependent nature of the Gong (2003) parametrisation produces too much sea spray aerosol over the Southern Ocean from December to February. If Revell et al. (2019) had sampled a wider range of processes (such as deposition) as we have here, our results might be brought into agreement. A better understanding of these conflicting results could be achieved using a multi-model experiment that sampled a range of atmospheric process representations.

The dry deposition velocity of accumulation mode aerosols (Dry_Dep_Acc) has an 84% likelihood of being lower than the default model value after applying the constraint. Furthermore, deposition velocities larger than around 3 times the default value are effectively ruled out. This constraint is consistent with the higher aerosol concentrations implied by constraint of the sea spray emission parameter.

Other parameters are more modestly constrained. The constraint on the scaled DMS emission flux is two-sided, reducing the credible range of DMS emission scalings from 0.5 to 2.0 down to 0.54 to 1.9. This constraint suggests the default emission inventory is consistent with measurements and doesn't benefit from being scaled.

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Furthermore, ACE-SPACE measurements are consistent with less efficient aerosol scavenging (55% likelihood of Rain_Frac, the parameter that controls the proportion of cloudy model grid boxes where rain occurs, being below 0.5) and less aqueous phase sulfate production (pH of cloud droplets has a 62% likelihood of being lower than the unconstrained median value). These combined constraints suggest, in agreement with sea spray and deposition parameter constraints, higher aerosol number and mass concentrations are consistent with measurements.

The effects of measurement constraint on pdfs of RF_{aci} and ERF_{aci} are shown in Fig. 4. Removing implausible model variants has reduced the uncertainty in several parameters including natural aerosol emission fluxes, which translates into a reduction in RF_{aci} uncertainty (Carslaw et al., 2013). The measurement constraints have two important effects on aerosol forcing. Firstly, the magnitude of median RF_{aci} weakens from -1.99 W m^{-2} to -1.88 W m^{-2} (-1.64 to -1.49 W m^{-2} for ERF_{aci}). A weaker forcing is consistent with higher natural aerosol emissions and increased aerosol load in the early-industrial period. Secondly, the constrained forcing pdfs are approximately symmetric but have shorter tails (lower kurtosis). This suggests the constraints are selectively ruling out model variants that are outliers. The 95% credible range of RF_{aci} values is reduced by around 9% (from -2.84 to -1.15 W m^{-2} down to -2.64 to -1.10 W m^{-2}) and around 9% for ERF_{aci} (from -2.69 to -0.62 W m^{-2} down to -2.43 to -0.54 W m^{-2}). The consistency of forcing constraint across two distinct PPEs suggests the results are insensitive to differences in meteorology, parameters perturbed in the PPEs, and the inclusion of rapid atmospheric adjustments. These results are also insensitive to additional constraint to ensure energy balance at the top of the atmosphere (Fig. S2).

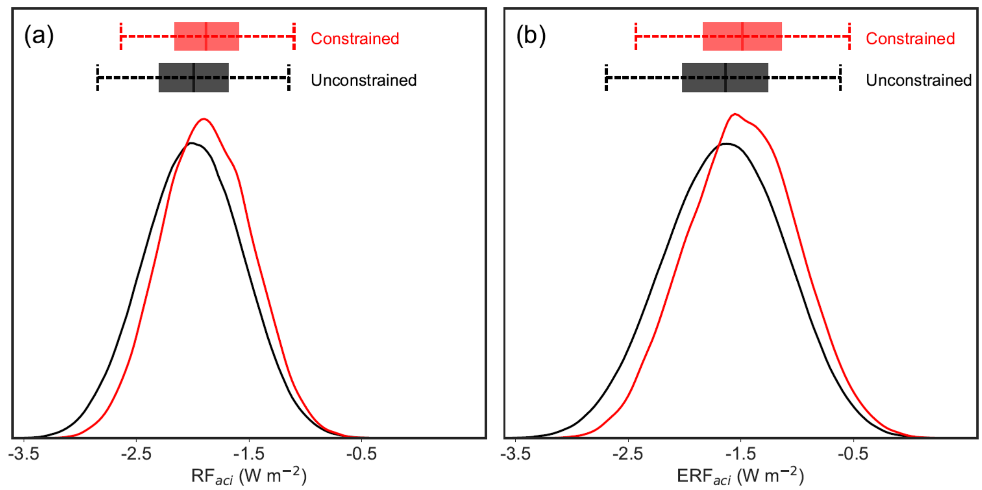

Fig. 4. Probability distributions of a) RF_{aci} and b) ERF_{aci}. The distributions of the unconstrained sample of one million model variants from statistical emulators of each PPE are in black. Red lines show the distributions after constraint using ACE-SPACE measurements (around 3% of the unconstrained sample). The 25th, 50th and 75th percentiles of each sample are shown as shaded boxes and dashed lines span the 2.5th and 97.5th percentiles.

Johnson et al. (2019) reduced the global, annual mean RF_{aci} uncertainty by constraining multiple anthropogenic emission and model process parameters (as well as some natural aerosol parameters) using over 9000 predominantly Northern Hemisphere measurements of aerosol optical depth, $PM_{2.5}$, particle number concentrations and mass concentrations of organic carbon and sulfate. We used the same methodology as Johnson et al. (2019) to rule out implausible model variants from the same original sample of one million model variants, so we can readily combine these constraints. Around 700 model variants (0.07%) are observationally plausible in both the Southern Ocean (ACE-SPACE) and Johnson et al. (2019) constraints. The marginal parameter pdfs from this 700-member sample are shown in Fig. 5. Because Johnson et al. studied only the AER PPE (from which RF_{aci} can be computed) we are unable to explore the effect of the combined constraint on ERF_{aci}.

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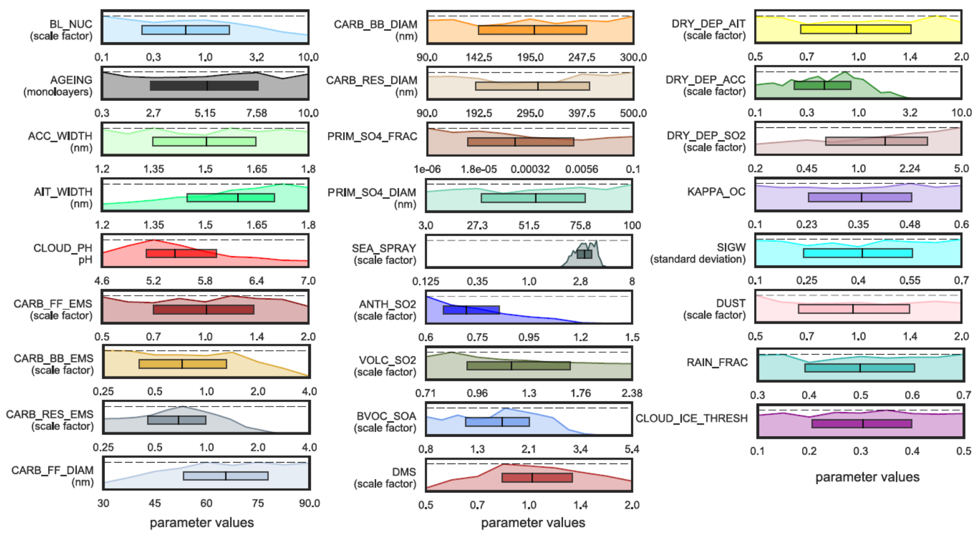

Fig. 5. Marginal probability distributions for the 26 aerosol parameters after constraint using around 250 Southern Ocean measurements and more than 9000 aerosol measurements in Johnson et al. (2019). Plotting features of this figure are identical to Fig. 3.

The two measurement datasets constrain distinct groups of parameters. There are a few cases where the same parameters are constrained by both datasets and in these cases the parameter values are constrained consistently (e.g. cloud droplet pH) or more strongly through ACE-SPACE (e.g. sea spray emissions). The complementary nature of these constraints means that the combined constraint marginal parameter pdfs (Fig. 5) are remarkably similar to those in our Fig. 3e (for sea spray and DMS emission fluxes, as well as deposition and pH parameters) and in figure 6 of Johnson et al. (2019) for other parameters.

The Johnson et al. (2019) constraint reduced the RF_{aci} uncertainty by around 6% and our ACE-SPACE measurement constraint reduced the uncertainty by around 9%. However, the RF_{aci} uncertainty is reduced by around 21% (Fig. 6a) after applying both constraints, meaning the combined constraint is stronger than the sum of individual constraints.

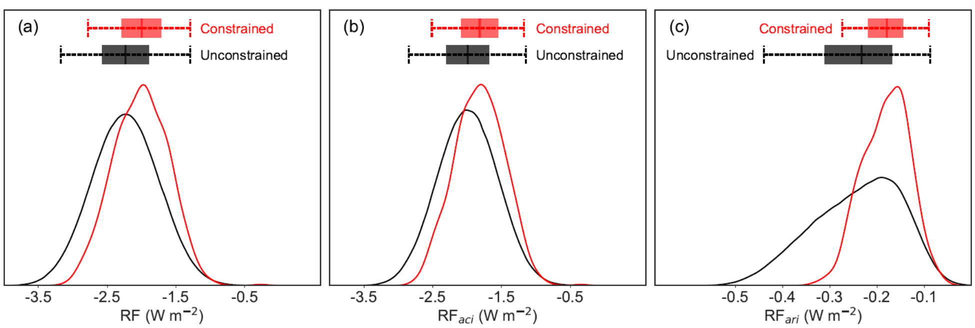

Fig. 6. Probability distributions of a) RF, b) RF_{aci} and c) RF_{ari} from the unconstrained (black line) and constrained (red line) samples of model variants. The constrained sample includes model variants that agree with our ACE-SPACE measurement constraint and the Johnson et al. (2019) constraint. Plotting features are identical to Fig. 4.

The Johnson et al. (2019) constraint strengthened the RF_{aci} by around 0.3 W m^{-2} because the largest sea spray emission flux scaling and largest new particle formation rates were ruled out. Our ACE-SPACE constraint rules

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out the same large sea spray emission fluxes, but also rules out all emission flux scale factors lower than around 1.6, which increases the baseline aerosol concentration in the early-industrial atmosphere. The ACE-SPACE measurements also constrain several other parameters that collectively weaken RF_{aci} weaken the median RF_{aci} by around 0.18 W m^{-2}. Therefore, using the combined measurement dataset, the highest and lowest RF_{aci} values have been ruled out as implausible and the credible range of observationally plausible RF_{aci} values is reduced to around -2.51 to -1.17 W m^{-2} (-2.18 to -1.46 W m^{-2} , when using one standard deviation to quantify the uncertainty). Uncertainty in RF_{ari} is reduced by around 48% with observationally plausible values ranging from -0.27 to -0.09 W m^{-2} (-0.23 to -0.13 W m^{-2}, when using one standard deviation), because the strongest RF_{ari} values are ruled out as observationally implausible.

3 Discussion

Our results show, as hypothesised from previous sensitivity analyses, that remote marine measurements are valuable for constraining the natural aerosol state of the atmosphere (Carslaw et al., 2013; Regayre et al., 2014; Regayre et al., 2018). Remote marine aerosol measurements provide new information about plausible model behaviour because they are closely related to model emissions and processes that measurements in polluted environments do not constrain.

For the first time we have achieved a meaningful reduction of 21% in the RF_{aci} uncertainty by constraining the aerosol properties in the model. The reduction in forcing uncertainty can still be improved by considering the following: Firstly, there are several causes of RF_{aci} uncertainty that are not constrained by a combination of Northern Hemisphere and pristine Southern Ocean measurements. Identifying measurements associated with primary particle emission diameters (BB_diam and Prim_SO4_diam), Aitken mode aerosol removal rates (Dry_Dep_Ait) and model process parameters related to cloud droplet activation (Kappa_OC, Ait_width, Sig_W) and using them as additional constraints should further reduce the forcing uncertainty. Secondly, even within the considerably reduced volume of multi-dimensional parameter space there still exist many compensating parameter effects, which limit the constraint on individual parameter ranges (Lee et al., 2016; Regayre et al., 2018). The impact of these compensating effects could be greatly reduced by perturbing uncertain emissions regionally rather than globally as we do here.

Our results are based on uncertainty in a single climate model. Model inter-comparison projects (such as CMIP6) can be used to quantify the diversity of RF (or ERF) output from models, but they lack information about single model uncertainty. Ideally, multi-model ensembles would contain a perturbed parameter component, but the computational cost prevents many modelling groups from engaging with this important aspect of uncertainty quantification, limiting our shared knowledge about the causes of aerosol forcing uncertainty. Studies like ours that quantify the remaining uncertainty in aerosol forcing and its components after constraint using multiple measurement types fill an important knowledge gap. This knowledge can be used to form a more complete understanding of the importance of historical and near-term aerosol radiative forcing which would reduce the diversity in equilibrium climate sensitivity across models.

Data availability

The ACE-SPACE data are accessible from: https://zenodo.org/communities/spi-ace. Simulation output data in both AER and AER-ATM PPEs are available on the JASMIN data infrastructure (http://www.jasmin.ac.uk). Some of the climate-relevant fields are derived and stored in netCDF files (.nc) containing data for all ensemble members and made available as a community research tool as described in Yoshioka et al. (2019). Model data and analysis code can be made available from the corresponding author upon request.

Author Contribution

LR applied the statistical methodology and generated results. LR and MY created the PPEs. LR and JJ designed the experiments and elicited probability density functions of all aerosol parameters. KC and MY participated in the formal elicitation process. JS, AB, MG, CT, SH and FS collected and processed the ACE-SPACE measurements. LR, KS, JS and JJ analysed the results. LR and KS wrote the manuscript with contributions from all authors.

Competing Interests

Author KC is an executive editor of ACP.

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