@AGUPUBLICATIONS

Geophysical Research Letters

RESEARCH LETTER

10.1002/2014GL062029

Key Points:

- Forcing sensitivity to aerosol parameters is strongly period dependent
- Understanding near-future climate is limited if a single period is considered
- In recent decades, parametric uncertainty is smaller than model diversity

Supporting Information:

- Readme
- Table S1
- Table S2
- Table S3
- Figure S1
- Figure S2
- Figure S3

Correspondence to:

L. A. Regayre, L.A.Regayre11@leeds.ac.uk

Citation:

Regayre, L. A., K. J. Pringle, B. B. Booth, L. A. Lee, G. W. Mann, J. Browse, M. T. Woodhouse, A. Rap, C. L. Reddington, and K. S. Carslaw (2014), Uncertainty in the magnitude of aerosol-cloud radiative forcing over recent decades, *Geophys. Res. Lett.*, *41*, 9040–9049, doi:10.1002/2014GL062029.

Received 26 SEP 2014 Accepted 17 NOV 2014 Accepted article online 24 NOV 2014 Published online 16 DEC 2014

This is an open access article under the terms of the Creative Commons Attribution License, which permits use, distribution and reproduction in any medium, provided the original work is properly cited.

Uncertainty in the magnitude of aerosol-cloud radiative forcing over recent decades

L. A. Regayre¹, K. J. Pringle¹, B. B. B. Booth², L. A. Lee¹, G. W. Mann^{1,3}, J. Browse¹, M. T. Woodhouse⁴, A. Rap¹, C. L. Reddington¹, and K. S. Carslaw¹

¹School of Earth and Environment, University of Leeds, Leeds, UK, ²UK Hadley Centre Met Office, Exeter, UK, ³National Centre for Atmospheric Science, University of Leeds, Leeds, UK, ⁴CSIRO Oceans and Atmosphere, Aspendale, Victoria, Australia

Abstract Aerosols and their effect on the radiative properties of clouds are one of the largest sources of uncertainty in calculations of the Earth's energy budget. Here the sensitivity of aerosol-cloud albedo effect forcing to 31 aerosol parameters is quantified. Sensitivities are compared over three periods; 1850–2008, 1978–2008, and 1998–2008. Despite declining global anthropogenic SO₂ emissions during 1978–2008, a cancelation of regional positive and negative forcings leads to a near-zero global mean cloud albedo effect forcing. In contrast to existing negative estimates, our results suggest that the aerosol-cloud albedo effect was likely positive (0.006 to 0.028 W m⁻²) in the recent decade, making it harder to explain the temperature hiatus as a forced response. Proportional contributions to forcing variance from aerosol processes and natural and anthropogenic emissions are found to be period dependent. To better constrain forcing estimates, the processes that dominate uncertainty on the timescale of interest must be better understood.

1. Introduction

Aerosols directly reflect sunlight and affect cloud properties such as albedo [*Twomey*, 1977]. Other rapid adjustments to cloud properties in response to changes in aerosol concentrations can also occur, yet these remain poorly understood and poorly represented in global climate models (GCMs) [*Boucher et al.*, 2013]. Uncertainty in the magnitude of aerosol-cloud interaction (ACI) forcing in response to changing anthropogenic emissions is the dominant source of uncertainty in net aerosol radiative forcing within current GCMs [*Skeie et al.*, 2011; *Stocker et al.*, 2013]. The cloud albedo effect (CAE) [*Boucher et al.*, 2013], an effect characterized by a decrease in cloud drop effective radius that results from an increase in cloud droplet number concentration for a fixed amount of liquid water [*Twomey*, 1977], remains the largest component of the ACI.

The greatest source of uncertainty in global CAE forcing between the preindustrial and the present-day is the state of the preindustrial atmosphere [*Carslaw et al.*, 2013a]. This arises because cloud albedo responds, to a first-order approximation, logarithmically to increasing aerosol concentrations, so a large proportion of the uncertainty in cloud radiative change over the industrial period is associated with low aerosol concentrations in the preindustrial [*Schmidt et al.*, 2012; *Carslaw et al.*, 2013a; *Ghan et al.*, 2013]. *Carslaw et al.* [2013a] found that 45% of CAE forcing variance, calculated between 1750 and 2000, was attributable to uncertain and potentially unconstrainable natural aerosol emissions, suggesting a substantial component of climate model forcing uncertainty may be irreducible.

 CO_2 concentrations are the main source of uncertainty in radiative forcing of future climate, when calculated to 2100, because CO_2 is a long-lived greenhouse gas for which emissions vary substantially in emission scenarios [*van Vuuren et al.*, 2011]. By the end of the century, aerosol forcing is likely to be negligible compared to CO_2 forcing [*Smith and Bond*, 2014]. On decadal timescales however, uncertainty in the change in aerosol forcing due to the representation of aerosol processes and emissions is comparable to the change in CO_2 forcing and can strongly influence radiative forcing calculations [*Hawkins and Sutton*, 2009; *Kirtman et al.*, 2013].

It is important to know the sign and magnitude of changes in CAE forcing in recent decades because changes in near-term historical forcing will inform the interpretation of near-future climate changes. Reducing forcing uncertainty in near-future projections is critical, yet the sources of CAE forcing uncertainty within global models are unknown, which limits individual model development and hinders the interpretation of

model intercomparison studies. Here we quantify uncertainty in CAE forcing attributable to 31 uncertain aerosol parameters, within a single global model for the periods 1850–2008, 1978–2008, and 1998–2008. This statistical analysis allows for areas of research to be prioritized for further model development, so that uncertainty in near-term climate projections may be reduced and makes an analysis of the role of CAE forcing in near-term historical climate change possible.

2. Methods

2.1. Time Periods

The three periods 1850–2008, 1978–2008, and 1998–2008 were chosen to provide the greatest contrast in changing anthropogenic aerosol emissions. By considering CAE forcing sensitivities across the three periods simultaneously, it is assumed that all parameters with the potential to strongly influence near-future climates will be identified.

Historical radiative forcing is usually calculated from preindustrial to present day, where an overall increase in global emissions of anthropogenic aerosols occurs. The period 1850–2008 is therefore included in this study for consistency. Global SO₂ emissions peaked in the late 1970s at approximately 120 Tg per year in 1978 [*Lamarque et al.*, 2010], then experienced several periods of decline, with Asian emissions causing further increases in the early part of this century [*Smith et al.*, 2011] resulting in approximately 103 Tg being emitted in 2008 [*Lamarque et al.*, 2010]. The overall decline in global anthropogenic emissions since the late 1970s coincides with a period of relatively rapid warming of surface temperatures [*Hartmann et al.*, 2013]. The period 1978–2008 can therefore be considered as distinct from 1850 to 2008, with the potential to produce CAE forcing values that are influenced by a unique set of parameters.

The 10 year period 1998–2008 can also be considered as a distinct period of anthropogenic emissions. In 1998, approximately 108 Tg of SO₂ was emitted globally [*Lamarque et al.*, 2010] hence between 1998 and 2008 global SO₂ emissions declined more gradually than in previous decades [*Granier et al.*, 2011]. The multidecadal trend in declining SO₂ emissions eased in Europe, yet became stronger in North America, during the 1998–2008 period. Asian emissions increased more rapidly than in the 1978–2008 period. The 1998–2008 period is also of interest because of the hiatus in global surface temperature rise which has been noted in the observational record since the late 1990s [*Brohan et al.*, 2006]. Identifying the sign and magnitude of CAE forcing, along with the associated variance, will shed light on the role of CAE forcing during the hiatus period.

The choice of 2008 as the end point for each period is based on an interest in evaluating decadal forcings and is thus constrained by our choice to use 1978 and 1998 as the start of the most recent periods. The experimental design outlined in section 2.2 is such that the choice of end year is largely arbitrary and is not expected to affect the results or conclusions.

2.2. Perturbed Parameter Ensemble

Thirty-one parameters related to aerosol processes as well as natural and anthropogenic aerosol emissions were identified and perturbed simultaneously within the GLObal Model of Aerosol Processes (GLOMAP) [*Spracklen et al.*, 2005; *Mann et al.*, 2010, 2012], at a horizontal resolution of $2.8^{\circ} \times 2.8^{\circ}$ with 31 vertical levels between the surface and 10 h Pa. GLOMAP is an extension to the TOMCAT three dimensional chemical transport model [*Stockwell and Chipperfield*, 1999]. Maximin Latin Hypercube sampling was used to create a parameter combination design, of 187 points, that spans the uncertain parameter space.

The parameters perturbed in this ensemble are similar to those used in *Lee et al.* [2013] and *Carslaw et al.* [2013a] with some new or adjusted parametrisations that relate to uncertain aspects of a newer version of the model. Particle formation within the continental boundary layer now uses a parametrisation that is enhanced in the presence of organic material [*Metzger et al.*, 2010]. Parametrisations for the dry deposition of SO₂, the emission flux of dust aerosol and two parameters relating to the wet removal of aerosols in low-level drizzling clouds have been included. The probability distributions for the uncertain parameters were identified through expert elicitation updated from *Lee et al.* [2013].

In this version of the model, three-dimensional meteorological fields and distribution of clouds obtained from the European Centre for Medium-Range Weather Forecasts (ECMWF) ERA-Interim reanalysis for 2008 are used for all years. Low-level stratiform clouds are prescribed from the International Satellite Cloud Climatology Project (ISCCP) D2 data [*Rossow and Schiffer*, 1999]. Modeled aerosols do not affect the

meteorology, transport, and presence of cloud, although the aerosols themselves are affected by cloud processing and precipitation. Changes in simulated CAE forcing across the ensemble, for each period, can therefore be attributed solely to the parameter perturbations.

Emission scenarios prepared for the Atmospheric Chemistry and Climate Model Intercomparison Project (ACCMIP) [*Lamarque et al.*, 2010] and prescribed in some of the Coupled Model Intercomparison Project Phase 5 (CMIP5) experiments [*Taylor et al.*, 2012] were used here to prescribe anthropogenic aerosol emissions for the years 1850, 1978, 1998, and 2008.

Pairs of simulations were used in the calculation of radiative forcing. Identical model configurations were used for each pair with the exception of anthropogenic aerosol emissions which were prescribed according to the years at either end of the periods examined. CAE forcing is defined here to be the difference in top of atmosphere net radiative fluxes between years and was quantified by modifying the cloud drop effective radius (r_e) for low- and middle-level clouds up to 6×10^2 h Pa, within the offline version of the Edwards and Slingo radiative transfer model [*Edwards and Slingo*, 1996]. This is the same approach used to calculate forcing in *Carslaw et al.* [2013a]. Surface albedo and cloud optical depths from ISCCP D2 for the year 2000 were used, and therefore, r_e was modified relative to values derived for that year, denoted here using the "ref" superscript:

$$r_e = r_e^{\text{ref}} \times \left(\frac{\text{CDNC}^{\text{ref}}}{\text{CDNC}}\right)^{1/3},\tag{1}$$

where CDNC is the monthly mean cloud drop number concentration within each model grid box. A fixed value of $r_e^{\text{ref}} = 10 \,\mu\text{m}$ was used to ensure consistency with the ISCCP retrievals, and CAE forcing over a given period was taken as the difference between forcings for each year relative to the year 2000. The net cloud radiative effect from our year 2000 reference simulation is $-25.7 \,\text{W}\,\text{m}^{-2}$. *Calisto et al.* [2014] use 10 years of satellite retrievals to calculate an average cloud radiative effect of $-18.8 \,\text{W}\,\text{m}^{-2}$. While our reference cloud radiative effect is higher than determined from satellite retrievals, it is in agreement with CMIP5 models [*Calisto et al.*, 2014].

A cloud droplet activation parametrisation [Fountoukis and Nenes, 2005; Barahona et al., 2010] was used to calculate CDNCs using the monthly mean aerosol distribution and composition in each grid box. Global, annual averages of CAE forcing for the 187 ensemble members were used to construct a statistical approximation to the model output and perform a sensitivity analysis.

2.3. Sensitivity Analysis

A variance-based sensitivity analysis [*Saltelli et al.*, 2000] of CAE forcing is made possible using validated Bayesian emulators [*Oakley and O'Hagan*, 2002] that are conditioned on the 187 member ensemble for each period to provide a statistical approximation of model output at any point in the 31-dimensional parameter space. The Bayesian emulation approach has been successfully applied to GLOMAP model output by *Lee et al.* [2011, 2012, 2013] and *Carslaw et al.* [2013a, 2013b].

The generation of a complete parametric response surface allows for contributions to variance, from each parameter and interactions between parameters, to be quantified explicitly across the entire surface. Nonlinear variations within the response surface are accounted for automatically and can be examined as required. These advantages cannot be obtained using one-at-a-time parameter perturbations, as is standard practice in climate model development. The results of the CAE forcing sensitivity analyses are provided in section 3.2. Sensitivity analyses were conducted using probability distributions of forcing that were obtained using the extended-FAST sampling method [*Saltelli et al.*, 1999], with 10⁴ emulator sample points per parameter. These samples were also used to calculate 90% credible intervals of CAE forcing that account for variation across the parameter space.

3. Results

3.1. Ensemble Mean Aerosol-Cloud Radiative Forcing

Figure 1 shows the mean CAE forcing of the 187 member ensemble, for each period, within each model grid box. In Figure 1a, the well-documented negative anthropogenic aerosol forcing between the early-industrial to present-day period can be seen. The CAE forcing is strongest in the Northern Hemisphere where anthropogenic aerosol emissions increased significantly since 1850. The majority of the atmosphere changes from clean to polluted during this period.

AGU Geophysical Research Letters



Figure 1. Average CAE radiative forcing for (a) 1850–2008, (b) 1978–2008, and (c) 1998–2008 in W m⁻². Forcing values for all ensemble members are averaged within individual grid boxes. The global means are calculated using 10^4 values sampled from the emulator for each period.

In recent decades, the effect of the regional changes in emissions outlined in section 1 become evident. There are regions of strong positive forcing where the CAE is smaller in 2008 than in 1978 in response to declining anthropogenic aerosol emissions. The patterns of regional forcing in the 1978-2008 period closely resemble those detected by Andrews [2013], in aerosol effective radiative forcing and changes in aerosol optical depth, over a similar period using a GCM with dynamic meteorology. Regional CAE forcings of opposite sign cancel out when calculating the global mean forcing. In the 1978-2008 period, ensemble mean CAE forcings in individual model grid boxes range from -1.8 to 3.0 W m⁻² and from -0.8 to 1.1 W m^{-2} in the 1998–2008 period.

The spatial pattern of CAE forcing is similar between 1978-2008 and 1998–2008 (Figures 1b and 1c). The regions of positive and negative forcing are considerably smaller in the most recent decade, which is to be expected since the 1998-2008 period is much shorter and also because the rates of change in SO₂ emissions in these regions during this period are generally smaller than in previous decades. Differences in the spatial patterns of ensemble mean CAE forcing confirm that the magnitude and sign of changes in anthropogenic emissions lead to distinct cloud albedo responses in the three periods.

3.2. Sensitivity Analysis ofEach Period3.2.1. Magnitude and Diversity of

CAE Forcing Estimates

Effective radiative forcing due to aerosol-cloud interactions for the 1750–2011 period is calculated by *Myhre et al.* [2013], using a multimodel ensemble, to be -0.45 W m⁻² with a credible interval of (-1.2 to 0.0 W m⁻²). Our mean emulated global CAE forcing

for 1850–2008 is -1.01 W m^{-2} with a credible interval of (-1.235 W m^{-2} , -0.782 W m^{-2}). Individual ensemble members produce values ranging from -1.817 to -0.461 W m^{-2} . The breadth of CAE forcing values is sufficient to provide a useful framework for exploring parametric sources of uncertainty.

The strong negative global mean forcing in the 1850–2008 period is not present in 1978–2008 where the mean emulated CAE forcing is zero with a credible interval of (-0.035 W m⁻², 0.033 W m⁻²). The global



Contributions to CAE radiative forcing variance

Figure 2. Contributions to globally averaged CAE forcing variance from aerosol process parameters and natural and anthropogenic emissions, for the three periods 1850–2008, 1978–2008, and 1998–2008. Percentages are obtained by Monte Carlo sampling from independent emulators of forcing for each period. Note that the total variance changes substantially between periods.

mean CAE forcing magnitude and credible range for the 1978–2008 period are both small compared to a CO_2 forcing of 0.7 W m⁻² over the same period [Myhre et al., 2013], resulting from compensating positive and negative regional forcings of up to 3 W m⁻². The relatively small uncertainty around zero that we calculate does not alter the conclusion that increasing concentrations of CO_2 and other well-mixed greenhouse gases produced the observed rapid warming of global mean surface temperature, which started in the late 1970s [Hartmann et al., 2013].

The small credible forcing range calculated here suggests a confidence in the zero 1978–2008 emulated mean CAE forcing which is in contrast with other studies. *Skeie et al.* [2011] calculate a CAE forcing during the

1978–2008 period of approximately -0.093 W m⁻² and *Shindell et al.* [2013] use three CMIP5 GCMs to calculate combined CAE and rapid adjustment forcings of approximately -0.04, -0.15, and -0.67 W m⁻² between 1980 and 2000. The magnitude of global CAE forcing diversity between models is the same order of magnitude as the CO₂ forcing over recent decades [*Myhre et al.*, 2013]. Our results isolate the uncertainty attributable to aerosol parameters and emissions within a global model and suggest that these factors are a smaller source of CAE forcing uncertainty than the uncertainty arising from the representation of atmospheric physics within models and the structural choices of aerosol and atmospheric physics parametrisations, at least for the globally averaged CAE forcing. Our small global mean CAE forcing uncertainty for the 1978–2008 period is the result of compensating uncertainties in positive and negative regional forcings. A large part of the difference between our small parametric uncertainty and model diversity in global CAE forcing over recent decades may be caused by differences in the extent to which regional forcings truly cancel within models.

In the 1998–2008 period, there is an overall positive forcing of 0.018 W m⁻² with a credible interval of (0.006 W m⁻², 0.028 W m⁻²). The continued decline in global anthropogenic emissions during this period, although smaller per decade than the period 1978–2008, produces a positive CAE forcing. The small, likely positive CAE forcing suggests that CAE forcing is unlikely to be the cause of the hiatus in global surface temperature rise, which would require a forcing of the order of -0.35 W m⁻² [Solomon et al., 2007]. The positive global CAE forcing calculated here contrasts with the -0.06 W m⁻² potential contribution of CAE forcing to recent changes in surface temperatures calculated by *Schmidt et al.* [2014]. The positive 1998–2008 global CAE forcing is a small but nonnegligible 2–11% of the 0.25 W m⁻² CO₂ forcing over the same period [*Myhre et al.*, 2013]. When CAE forcing and its parametric sensitivities are accounted for the magnitude of forcing per decade that would need to be explained by other external forcings increases. Approximately half of current GCMs exclude the CAE [*Wilcox et al.*, 2013] and are therefore unable to account for this important process and its inherent uncertainty when calculating forcing over recent decades.

3.2.2. Changes in the Sources of Uncertainty

Uncertainty in global mean CAE forcing can be decomposed into uncertainty arising from aerosol process parameters and from natural and anthropogenic emissions, using a variance-based sensitivity analysis [*Saltelli et al.*, 2000]. Proportional reductions in total variance that can be expected if all parameters within a group were known precisely, are presented for each period in Figure 2. The CAE forcing variance is smaller in the two most recent periods than in the 1850–2008 period, and the contributions to variance should be interpreted in this context. Because the emulators produce results in a fraction of the time required for

AGU Geophysical Research Letters



Forcing (W m⁻²)

Figure 3. Box-and-whisker plots of CAE radiative forcing for individual parameters and groups of parameters during the periods (a) 1850–2008, (b) 1978–2008, and (c) 1998–2008. The probability density of each parameter is obtained by Monte Carlo sampling from the emulators for each period. The 25th, 50th, and 75th percentiles are used to create each box, and the whiskers extend to the most extreme sample point within 1.5 times the interquartile range. All box-and-whisker plots are centered on the mean emulated forcing value for that period. The range of emulated CAE forcing changes within each period.

the global model, sufficiently large samples can be taken from the multidimensional response surface to produce meaningful statistical summaries, such as those provided in Figure 2.

Each of the anthropogenic aerosol emission periods produces a distinct mix of contributions to variance, with substantial changes in the influence of parametric uncertainties on global CAE forcing variance between periods. Natural aerosol contributions to forcing variance decline from 34.4% in 1850-2008 to only 6.8% and 1.9% in 1978-2008 and 1998-2008, respectively. This decline was predicted by Carslaw et al. [2013a], who showed that using a relatively polluted baseline in the forcing calculation reduces the importance of uncertainty in natural aerosol emissions. Note that the parametric contributions presented here differ from those in Carslaw et al. [2013a] because a different early-industrial period start year was used and furthermore model results are influenced by the structural changes implemented in this version of the model, as described in section 2.2.

Uncertainties in anthropogenic emissions dominate forcing uncertainty during the 1978-2008 period, contributing 49.8% of the global CAE forcing variance, compared to a 30.2% contribution from aerosol process parameters. Anthropogenic emission uncertainties determine both the magnitude and the sign of CAE forcing during the 1978-2008 period. Strong regional forcings of opposite sign have the potential to be canceled out, and the anthropogenic emission uncertainties control the relative importance of regional contributions to the global CAE forcing.

Aerosol process parameters dominate the 1998–2008 forcing uncertainty, contributing 46.4% of the global CAE forcing variance, with anthropogenic emissions accounting for only 18.5%. Global CAE forcing during this period is controlled by those regions experiencing a decline in anthropogenic emissions, leading to an overall positive forcing. Aerosol process parameter uncertainties take on a larger role in controlling global CAE forcing during this period, because uncertainties in parameters controlling the growth and removal of aerosol are more important than the uncertainties in the emissions themselves when changes in emissions are small. The uncertainties in aerosol process parameters contribute to the forcing variance in each period, although their influence is strongest in the absence of large changes in anthropogenic aerosol emissions. The dominance of aerosol process parameters as a source of global mean CAE forcing variance in the near-term suggests that aerosol model structural uncertainty is likely to make an important contribution to near-future climate projection uncertainty.

The changing proportional contributions to global CAE forcing variance from the three sources of parametric uncertainty suggest that the periods examined here are diverse enough to enable the identification of those uncertain aerosol parameters which may influence future CAE forcing uncertainty. Changes in the contributions from individual parameters are discussed further in section 3.2.3.

3.2.3. Changes in Contributions From Individual Parameters

Individual aerosol parameter contributions to CAE forcing variance over the three periods are summarized using box-and-whisker plots in Figure 3. Each box and whisker plot is generated by sampling from the emulator 10⁴ times with the parameter in question allowed to vary across the parameter space and all other parameters held fixed at their median values, making these plots comparable to those in *Carslaw et al.* [2013a].

Some parameters such as the drizzle rate (Drizz_rate), defined as the precipitation rate in low-level stratocumulus clouds within a 6 h period [*Browse et al.*, 2012], contribute to CAE forcing variance regardless of emission period. Other parameters make small contributions to variance in some periods and substantial contributions in others. For some parameters, such as the magnitude of anthropogenic SO₂ emissions (Anth_SO₂) and global fossil fuel and global biofuel emission fluxes (FF_Ems and BF_Ems), the parametric contributions to forcing variance are correlated with the magnitude of emissions. Anthropogenic emission fluxes change less dramatically in 1998–2008 than in the other periods [*Bond et al.*, 2007; *Lamarque et al.*, 2010], and as such the associated parameters make smaller contributions to forcing variance.

The period dependence of individual parametric contributions to CAE forcing variance suggests that studies that are designed to quantify aerosol model uncertainty from a single time period, such as the preindustrial to present-day, may not be informative of model sensitivities in near-future climates of interest.

4. Conclusions

The most striking result of our study is that the parametric uncertainty of global mean CAE forcing over recent decades is much smaller than the range predicted by other climate models. Regional positive and negative forcings of up to 3 W m⁻² cancel each other in calculations of global CAE forcing over recent decades. Our analysis suggests that uncertainties in aerosol processes and emissions in a single global model (if spatially and temporally correlated as we assume) are less important than structural differences between models over recent decades. In contrast, over the 1850–2008 period, the forcing is almost everywhere negative, and aerosol parameters and emissions account for a substantial fraction of the multimodel range [*Carslaw et al.*, 2013a]. Thus, in historical or future periods in which regional patterns of forcing of opposite sign can occur, the true uncertainty in global mean forcing may be largely determined by the extent to which regional forcing cancelation occurs. That is, an understanding of regional patterns of forcing across multiple models becomes paramount.

The small global mean CAE uncertainty attributable to aerosol parameters and emissions over recent decades hides much larger regional forcing uncertainties. Causes of regional forcing uncertainty are likely to be highly variable, and an analysis of them may provide insight into the cause of model diversity. One method for understanding the sources of multimodel diversity in global and regional CAE forcing over recent decades would be to incorporate a perturbed parameter framework into multimodel intercomparison projects. The design of such experiments would benefit from being informed by the results here, which highlight those aerosol processes and emissions that are likely to influence multimodel global CAE forcing diversity on different timescales.

Despite the small calculated uncertainties in global mean CAE forcing, our analysis clearly shows that the causes of uncertainty in forcing over recent decades are very different to those associated with forcing

referenced to the preindustrial state [*Carslaw et al.*, 2013a]. Natural emission uncertainty was shown by *Carslaw et al.* [2013a] to substantially influence CAE forcing uncertainty since the preindustrial era. Here natural emission uncertainty has been shown to play only a minor role in controlling global CAE forcing variance in recent decades, where the atmosphere at both the start and end of the forcing period can be considered polluted relative to the preindustrial atmosphere. In order to identify parameters that have the potential to influence the near-future climate forcing, it is essential to perform sensitivity analyses using a range of diverse anthropogenic aerosol emission periods. Reducing uncertainty in these parameters will likely lead to improved near-future climate projections. The implication of the period dependence of CAE forcing sensitivity to uncertain aerosol parameters and emissions is that multimodel intercomparison projects may benefit from a design structure that utilizes historic periods over which anthropogenic emissions most closely resemble those used in the near-future climates they are to inform.

If the historical period under consideration contains relatively large regional and global changes in anthropogenic aerosol emissions, such as the 1978–2008 period, then uncertainties in these emissions contribute most to global CAE forcing uncertainty. Addressing the causes of anthropogenic emission inventory diversity during the 1978–2008 period is shown here to be a priority for reducing uncertainty in CAE forcing calculations over recent decades and therefore warrants further research.

Aerosol process parameters influence global CAE forcing variance much more strongly in periods where the forcing is controlled by relatively small changes in anthropogenic aerosol emissions, as is the case in the 1998–2008 period. This decade contrasts with the 1978–2008 period when there were relatively large regional increases and decreases in anthropogenic emissions. The CAE forcing uncertainty for the 1998–2008 period can be viewed as the lower limit that could be expected in near-future climates because first, the anthropogenic aerosol emission changes during the most recent decade are so small, and second, our results highlight the importance of aerosol process parameters over recent decades suggesting that uncertainties in the structural representation of aerosols between models may also have the greatest impact on uncertainty during these periods. Furthermore, we assume that anthropogenic aerosol emission fluxes are perfectly correlated, both temporally and spatially, so that anthropogenic emissions are systematically scaled high/low and regional positive and negative forcings can cancel in the calculation of the global mean forcing. If anthropogenic aerosol emissions were underestimated at one end of a period and overestimated at the other, for example, then anthropogenic emission uncertainty would be a larger source of CAE forcing variance during that period which would inflate the credible ranges of CAE forcing.

Acknowledgments

Data can be made available upon request from the corresponding author. L.A. Regayre is funded by a Doctoral Training Grant from the Natural Environment Research Council (NERC) and a CASE studentship with the UK Met Office Hadley Centre. Ben Booth was supported by the Joint UK DECC/Defra Met Office Hadley Centre Climate Programme (GA01101). Ken Carslaw acknowledges funding from the Royal Society Wolfson Award. We acknowledge funding from NERC under grants AEROS and GASSP (NE/G006172/1 and NE/J024252/1). This work made use of the facilities of N8 HPC provided and funded by the N8 consortium and EPSRC (grant EP/K000225/1). The Centre is coordinated by the Universities of Leeds and Manchester. Thanks to those experts who participated in the elicitation exercise and L. Rotstayn at CSIRO Oceans and Atmosphere for an insightful review of this article. The authors thank two anonymous reviewers for their helpful comments.

The Editor thanks two anonymous reviewers for their assistance in evaluating this paper.

Here we show that the credible range of CAE forcing during the 1998–2008 period is (0.006 W m⁻², 0.028 W m⁻²), indicating that a positive CAE forcing is likely during the 1998–2008 period. In contrast to existing negative estimates of aerosol indirect forcing, our results suggest that the aerosol-cloud albedo effect was likely positive during the last decade, indicating that the hiatus in surface warming cannot be attributed to CAE forcing. A likely positive CAE forcing during this period reframes the role of CAE forcing in explaining model overestimation of recent warming using external forcings. The attribution of the present pause in surface warming as a forced response [*Kaufmann et al.*, 2011; *Estrada et al.*, 2013; *Haywood et al.*, 2013; *Kosaka and Xie*, 2013; *Santer et al.*, 2014] is more difficult given the present results. The existing role of aerosols in explaining the hiatus therefore needs to be reevaluated.

References

- Andrews, T. (2013), Using an AGCM to diagnose historical effective radiative forcing and mechanisms of recent decadal climate change, J. Clim., 27, 1193–1209, doi:10.1175/JCLI-D-13-00336.1.
- Barahona, D., R. E. L. West, P. Stier, S. Romakkaniemi, and A. Nenes (2010), Comprehensively accounting for the effect of giant CCN in cloud activation parameterizations, *Atmos. Chem. Phys.*, *10*, 2467–2473, doi:10.5194/acp-10-2467-2010.
- Bond, T. C., E. Bhardwaj, R. Dong, R. Jogani, S. Jung, C. Roden, D. G. Streets, and N. M. Trautmann (2007), Historical emissions of black and organic carbon aerosol from energy-related combustion, 1850–2000, *Global Biogeochem. Cycles*, 21, GB2018, doi:10.1029/2006GB002840.
- Boucher, O., et al. (2013), Clouds and aerosols, in Climate Change 2013: The Physical Science Basis. Contribution of Working Group I to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change, edited by T. F. Stocker et al., pp. 574, 578, 609–610, Cambridge Univ. Press, Cambridge, U. K., and New York.
- Brohan, P., J. J. Kennedy, I. Harris, S. F. B. Tett, and P. D. Jones (2006), Uncertainty estimates in regional and global observed temperature changes: A new data set from 1850, *J. Geophys. Res.*, 111, D12106, doi:10.1029/2005JD006548.
- Browse, J., K. S. Carslaw, S. R. Arnold, K. J. Pringle, and O. Boucher (2012), The scavenging processes controlling the seasonal cycle in Arctic sulphate and black carbon aerosol, *Atmos. Chem. Phys.*, *12*, 6775–6798, doi:10.5194/acp-12-6775-2012.
- Calisto, M., D. Folini, M. Wild, and L. Bengtsson (2014), Cloud radiative forcing intercomparison between fully coupled CMIP5 models and CERES satellite data, Ann. Geophys., 32, 793–807, doi:10.5194/angeo-32-793-2014.

Carslaw, K. S., et al. (2013a), Large contribution of natural aerosols to uncertainty in indirect forcing, *Nature*, 503, 67–71, doi:10.1038/nature12674.

Carslaw, K. S., L. A. Lee, C. L. Reddington, G. W. Mann, and K. J. Pringle (2013b), The magnitude of uncertainty in global aerosol, *Faraday Discuss.*, 165, 495–512, doi:10.1039/C3FD00043E.

Edwards, J. M., and A. Slingo (1996), Studies with a flexible new radiation code: I. Choosing a configuration for a large scale model, Q. J. R. Meteorol. Soc., 122, 689–719, doi:10.1256/smsqj.53106.

Estrada, F., P. Perron, and B. Martínez-López (2013), Statistically derived contributions to diverse human influences to twentieth-century temperature change, Nat. Geosci., 6, 1050–1055, doi:10.1038/ngeo1999.

Fountoukis, C., and A. Nenes (2005), Continued development of a cloud droplet formation parameterization for global climate models, J. Geophys. Res., 110, D11212, doi:10.1029/2004JD005591.

Ghan, S. J., S. J. Smith, M. Wang, K. Zhang, K. J. Pringle, K. S. Carslaw, J. Pierce, S. Bauer, and P. Adams (2013), A simple model of global aerosol indirect effects, J. Geophys. Res. Atmos., 118, 6688–6707, doi:10.1002/jgrd.50567.

Granier, C., et al. (2011), Evolution of anthropogenic and biomass burning emissions of air pollutants at global and regional scales during the 1980–2010 period, *Clim. Change*, *109*, 163–190, doi:10.1007/s10584-011-0154-1.

Hartmann, D. L., et al. (2013), Observations: Atmosphere and surface, in Climate Change 2013: The Physical Science Basis. Contribution of Working Group I to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change, edited by T. F. Stocker et al., p. 193, Cambridge Univ. Press, Cambridge, U. K., and New York.

Hawkins, E., and R. Sutton (2009), The potential to narrow uncertainty in regional climate predictions, *Bull. Am. Meteorol. Soc.*, 90, 1097–1107, doi:10.1175/2009BAMS2607.1.

Haywood, J. M., A. Jones, and G. S. Jones (2013), The impact of volcanic eruptions in the period 2000–2013 on global mean temperature trends evaluated in the HadGEM2-ES climate model, Atmos. Sci. Lett., 15, 92–96, doi:10.1002/asl2.471.

Kaufmann, R., H. Kauppi, M. Mann, and J. Stock (2011), Reconciling anthropogenic climate change with observed temperature 1998–2008, *Proc. Natl. Acad. Sci. U.S.A.*, *108*, 11,790–11,793, doi:10.1073/pnas.1102467108.

Kirtman, B., et al. (2013), Near-term climate change: Projections and predictability, in Climate Change 2013: The Physical Science Basis. Contribution of Working Group I to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change, edited by T. F. Stocker et al., pp. 1002–1007, Cambridge Univ. Press, Cambridge, U. K., and New York.

Kosaka, Y., and S. P. Xie (2013), Recent global-warming hiatus tied to equatorial Pacific surface cooling, *Nature*, *501*, 403–407, doi:10.1038/nature12534.

Lamarque, J. F., et al. (2010), Historical (1850–2000) gridded anthropogenic and biomass burning emissions of reactive gases and aerosols: Methodology and application, *Atmos. Chem. Phys.*, *10*, 7017–7039, doi:10.5194/acp-10-7017-2010.

Lee, L. A., K. S. Carslaw, K. J. Pringle, G. W. Mann, and D. V. Spracklen (2011), Emulation of a complex global aerosol model to quantify sensitivity to uncertain parameters, *Atmos. Chem. Phys.*, *11*, 12,253–12,273, doi:10.5194/acp-11-12253-2011.

Lee, L. A., K. S. Carslaw, K. J. Pringle, and G. W. Mann (2012), Mapping the uncertainty in global CCN using emulation, Atmos. Chem. Phys., 12, 9739–9751, doi:10.5194/acp-12-9739-2012.

Lee, L. A., K. J. Pringle, C. L. Reddington, G. W. Mann, P. Stier, D. V. Spracklen, J. Pierce, and K. S. Carslaw (2013), The magnitude and causes of uncertainty in global model simulations of cloud condensation nuclei, *Atmos. Chem. Phys.*, 13, 8879–8914, doi:10.5194/acp-13-8879-2013.

Mann, G. W., K. S. Carslaw, D. V. Spracklen, D. A. Ridley, P. T. Manktelow, M. P. Chipperfield, S. J. Pickering, and C. E. Johnson (2010), Description and evaluation of GLOMAP-mode aerosol microphysics model for the UKCA composition-climate model, *Geosci. Model Dev.*, 3, 519–551, doi:10.5194/gmd-3-519-2010.

Mann, G. W., et al. (2012), Intercomparison of modal and sectional aerosol microphysics representations within the same 3-D global chemical transport model, *Atmos. Chem. Phys.*, *12*, 4449–4476, doi:10.5194/acp-12-4449-2012.

Metzger, A., et al. (2010), Evidence for the role of organics in aerosol particle formation under atmospheric conditions, Proc. Natl. Acad. Sci. U.S.A., 107, 6646–6651, doi:10.1073/pnas.0911330107.

Myhre, G., et al. (2013), Anthropogenic and natural radiative forcing, in *Climate Change 2013: The Physical Science Basis. Contribution of Working Group I to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change*, edited by T. F. Stocker et al., p. 677, Cambridge Univ. Press, Cambridge, U. K., and New York.

Oakley, J. E., and A. O'Hagan (2002), Bayesian inference for the uncertainty distribution of computer model outputs, *Biometrika*, 89, 769–784, doi:10.1093/biomet/89.4.769.

Rossow, W. B., and R. A. Schiffer (1999), Advances in understanding clouds from ISCCP, Bull. Am. Meteorol. Soc., 80, 2261–2288, doi:10.1175/1520-0477(1999)080<2261:AIUCFI>2.0.CO;2.

Saltelli, A., S. Tarantola, and K. P. S. Chan (1999), A quantitative model-independent method for global sensitivity analysis of model output, *Technometrics*, 41, 39–56, doi:10.2307/1270993.

Saltelli, A., K. Chan, and E. M. Scott (2000), Sensitivity Analysis, Wiley, Hoboken, N. J.

Santer, B. D., et al. (2014), Volcanic contribution to decadal changes in tropospheric temperature, *Nat. Geosci.*, 7, 185–189, doi:10.1038/ngeo2098.

Schmidt, A., K. S. Carslaw, G. W. Mann, A. Rap, K. J. Pringle, D. V. Spracklen, M. Wilson, and P. M. Forster (2012), Importance of tropospheric volcanic aerosol for indirect radiative forcing of climate, *Atmos. Chem. Phys.*, *12*, 7321–7339, doi:10.5194/acp-12-7321-2012.

Schmidt, G. A., D. T. Shindell, and K. Tsigaridis (2014), Reconciling warming trends, *Nat. Geosci.*, 7, 158–160, doi:10.1038/ngeo2105.
Shindell, D. T., et al. (2013), Radiative forcing in the ACCMIP historical and future climate simulations, *Atmos. Chem. Phys.*, 13, 2939–2974, doi:10.5194/acp-13-2939-2013.

Skeie, R. B., T. K. Bernsten, G. Myhre, K. Tanaka, M. M. Kvalevåg, and C. R. Hoyle (2011), Anthropogenic radiative forcing time series from pre-industrial times until 2010, Atmos. Chem. Phys., 11, 11,827–11,857, doi:10.5194/acp-11-11827-2011.

Smith, S. J., and T. C. Bond (2014), Two hundred fifty years of aerosols and climate: The end of the age of aerosols, Atmos. Chem. Phys., 14, 537–549, doi:10.5194/acp-14-537-2014.

Smith, S. J., J. van Aardenne, Z. Klimont, R. J. Andres, A. Volke, and S. D. Arias (2011), Anthropogenic sulfur dioxide emissions: 1850 to 2005, Atmos. Chem. Phys., 11, 1101–1116, doi:10.5194/acp-11-1101-2011.

Solomon, S., D. Qin, M. Manning, Z. Chen, M. Marquis, K. B. Averyt, M. Tignor, and H. L. Miller (Eds.) (2007), IPCC, 2007: The Physical Science Basis. Contribution of Working Group I to the Fourth Assessment Report of the Intergovernmental Panel on Climate Change, Cambridge Univ. Press, Cambridge, U. K., and New York.

Spracklen, D. V., K. J. Pringle, K. S. Carslaw, M. P. Chipperfield, and G. W. Mann (2005), A global off-line model of size-resolved aerosol microphysics: I. Model development and prediction of aerosol properties, *Atmos. Chem. Phys.*, *5*, 2227–2252.

- Stocker, T. F., D. Qin, G. K. Plattner, M. Tignor, S. K. Allen, J. Boschung, A. Nauels, Y. Xia, V. Bex, and P. M. Midgley (2013), Summary for policymakers, in *Climate Change 2013: The Physical Science Basis. Contribution of Working Group I to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change*, edited by T. F. Stocker et al., pp. 13–14, Cambridge Univ. Press, Cambridge, U. K., and New York.
- Stockwell, D. Z., and M. P. Chipperfield (1999), A tropospheric chemical-transport model: Development and validation of the model transport schemes, Q. J. R. Meteorol. Soc., 125, 1747–1783, doi:10.1256/smsqj.55713.
- Taylor, K. E., R. J. Stouffer, and G. A. Meehl (2012), An overview of CMIP5 and the experiment design, Bull. Am. Meteorol. Soc., 93, 485–498, doi:10.1175/BAMS-D-11-00094.1.

Twomey, S. (1977), Influence of pollution on shortwave albedo of clouds, J. Atmos. Sci., 34, 1149–1152.

- van Vuuren, D. P., et al. (2011), The representative concentration pathways: An overview, *Clim. Change*, *109*, 5–31, doi:10.1007/s10584-011-0148-z.
- Wilcox, L. J., E. J. Highwood, and N. J. Dunstone (2013), The influence of anthropogenic aerosol on multi-decadal variations of historical global climate, *Environ. Res. Lett.*, 8, 024033, doi:10.1088/1748-9326/8/2/024033.