Constraining uncertainty in aerosol direct forcing

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Abstract

The uncertainty in present-day anthropogenic forcing is dominated by uncertainty in the strength of the contribution from aerosol. Much of the uncertainty in the direct aerosol forcing can be attributed to uncertainty in the anthropogenic fraction of aerosol in the present-day atmosphere, due to a lack of historical observations. Here we present a robust relationship between total present-day aerosol optical depth and the anthropogenic contribution across two multi-model ensembles and a large singlemodel perturbed parameter ensemble. Using observations of aerosol optical depth, we determine a reduced likely range of the anthropogenic component and hence a reduced uncertainty in the direct forcing of aerosol.

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12	Key Points:					
13 14	• A strong relationship is demonstrated between total present-day aerosol loading and the anthropogenic contribution, across a variety of models.					
15 16	• Observations of the total present-day aerosol loading are thus used to constrain uncertainty in anthropogenic aerosol and aerosol direct forcing					
17 18	• Applying these constraints to one million samples of a 26-parameter perturbed parameter ensemble leads to a clear-sky RF_{ari} estimate of -0.67 ± 0.13 Wm ⁻² .					

20 Abstract

- 21 The uncertainty in present-day anthropogenic forcing is dominated by uncertainty in the strength
- of the contribution from aerosol. Much of the uncertainty in the direct aerosol forcing can be
- attributed to uncertainty in the anthropogenic fraction of aerosol in the present-day atmosphere,
- 24 due to a lack of historical observations. Here we present a robust relationship between total
- 25 present-day aerosol optical depth and the anthropogenic contribution across two multi-model
- 26 ensembles and a large single-model perturbed parameter ensemble. Using observations of
- aerosol optical depth, we determine a reduced likely range of the anthropogenic component and
- 28 hence a reduced uncertainty in the direct forcing of aerosol.

29 Plain Language Summary

- 30 Despite the impacts of global warming already being felt around the world it is still unclear how
- 31 much of the effect of greenhouse gasses is being offset by the cooling effect of atmospheric
- 32 aerosol through the scattering of incoming sunlight and the modification of clouds. A large part
- of the difficulty in determining the effect of aerosol is in understanding the proportion of present-
- day aerosol that is due to human activity. In this work we demonstrate a strong relationship
- between the total amount of aerosol in the present-day atmosphere (something we can measure)
- and the amount due to human activity (something we cannot). We further show that this allows
- 37 us to reduce the uncertainty in the cooling effect of aerosols.

38 **1 Introduction**

- 39 Aerosols affect the climate both directly by scattering and absorbing incoming solar radiation,
- and indirectly by providing the nuclei on which cloud droplets form. Anthropogenic
- 41 perturbations to the natural background aerosol population can therefore change the balance in
- 42 radiation at the top of the atmosphere and hence provide a forcing, which offsets some of the
- 43 forcing due to anthropogenic greenhouse gasses. Despite a concerted effort since the last IPCC
- 44 assessment (Myhre et al., 2013) the magnitude of anthropogenic aerosol forcing remains highly
- 45 uncertain.
- 46 One of the main sources of uncertainty in aerosol forcing is the lack of reliable observational
- 47 estimates of the amount of natural, or pre-industrial aerosol (Charlson et al., 1992; Carslaw et al.,
- 48 2013; Carslaw et al., 2017). Discerning the anthropogenic contribution to present-day aerosol
- 49 from present-day observations directly is challenging though. While some aerosol species, such
- as sea-salt, are easy to attribute to natural processes, others, such as sulfate and organic carbon,
- 51 can have a variety of natural and anthropogenic sources. Further, non-linearities in some aerosol
- 52 processes mean that anthropogenic perturbations can affect the production and removal of
- 53 natural aerosol (Stier et al., 2006).
- 54 Aerosol optical depth (τ) is a measure of the extinction of solar radiation by aerosol and is
- directly related to the direct aerosol effect. Satellite remote sensing retrievals of τ in the present
- 56 day (τ_{PD}) are available from a wide range of sensors, on different platforms and using different
- retrieval algorithms. While satellite-based retrievals of $\boldsymbol{\tau}$ require accurate models of surface
- albedo and can suffer from biases due to the need to accurately screen for clouds, they are the
- only aerosol datasets available which provide near-global coverage over land and ocean. τ is also
- a common GCM diagnostic, and a large ensemble of model values are available from the
- 61 AeroCom modelling community (Myhre et al., 2013), as well as some contributions to the
- 62 CMIP5 ensemble, making it an ideal observational constraint.
- 63 A number of cases have been found recently where relatively simple relationships between
- observable and unobservable quantities can be discerned, belying the apparent complexity of the
- underlying system (Allen & Ingram, 2002; Hall & Qu, 2006). These relationships can be
- 66 exploited using observations to constrain model ensemble values of the unobservable quantity.
- 67 Correlation does not imply causation, however, and so these 'emergent constraints' must be
- treated with care to ensure that the relationship is physically based and observationally robust.
- 69 Further care should be taken that the ensemble of models used to sample the uncertainty
- adequately reflects the uncertainty in the properties in question. Similarly, perturbed parameter
- ensembles (PPEs) enable exploration of the parametric uncertainty, and with sufficient
- observations, a constrained estimate of a quantity from a given model. This method however can
- say little of the structural deficiencies of a model and large inter-model differences may be un-
- accounted for. Here we look to combine the strengths of each of these approaches in a
- 75 complimentary way. An emergent constraint from a large multi-model ensemble is
- demonstrated, but not relied on. Instead a PPE is used to explore, and constrain, the parametric
- ⁷⁷ uncertainty quantitively in one model, in the context of the larger multi-model ensemble.

- In this work we demonstrate, and explain, a robust relationship between τ_{PD} and the
- anthropogenic aerosol optical depth, τ_{ant} (defined as the difference between present-day and
- 80 pre-industrial aerosol optical depth, denoted τ_{PD} and τ_{PI} respectively) across three different
- 81 model ensembles. We use satellite-based observations of τ_{PD} to constrain the uncertainty in τ_{ant}
- so in a large PPE, and in turn the clear-sky aerosol forcing (RF_{ari}) .

83 **2** A constraint emerges

- 84 As common GCM diagnostics, both τ_{PI} and τ_{PD} are available from a wide range of models.
- 85 Here we consider the CMIP5 (Taylor et al., 2012) models which participated in the fixed sea
- surface temperature aerosol experiments (Zelinka et al., 2014) and the AeroCom Phase II models
- 87 (Myhre et al., 2013). One drawback in using these ensembles to represent uncertainty in aerosol
- forcing however is that many of these simulations share anthropogenic emissions inventories and
- use the same or similar parameterizations for natural aerosol emissions, potentially leading to a
- 90 lack of representativity within and across these ensembles. To sample these uncertainties, we use
- Gaussian Process (GP) emulators¹ (O'Hagan, 2006) trained on a perturbed parameter ensemble
 (PPE) of 183 simulations of HadGEM3-UKCA corresponding to distinct combinations of 26
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 physical parameters relating to aerosol processes and emissions, for both present-day (2008) and
- physical parameters relating to aerosol processes and emissions, for both present-day (2008) and
 preindustrial (1850) emissions (Yoshioka et al., 2019; Carslaw et al., 2017). We are then able to
- explore the full parametric uncertainty attributable to the chosen parameter perturbations in
- global mean τ_{PI} and τ_{PD} by sampling the emulators at 1,000,000 parameter combinations from
- across the 26-dimensional parameter space of the PPE, drawn using a set of expert-elicited
- marginal distributions on the parameters (Yoshioka et al., 2019). Note that the three parameters
- relating to carbonaceous emissions were left unperturbed in this experiment as τ_{PD} provides little
- 100 constraint on these and they are only of secondary importance for RF_{ari} .
- In order to account for the uncertainty in satellite observations of τ_{PD} we use the standard
- deviation in the global mean value obtained from 7 distinct datasets covering 5 platforms, 3
- sensors and 5 retrieval algorithms, as listed in Table 1. These observational datasets then provide
- 104 well characterized global estimates of τ , although the possibility of remaining systematic biases 105 cannot be entirely discounted. Due to the orbits of the platforms and the difficulty in retrieving τ
- cannot be entirely discounted. Due to the orbits of the platforms and the difficulty in retrieving
 over snow and ice these values represent averages only between 60S and 60N.... For the PPE
- values reported here we consider the same latitudinal range. While using only the global yearly
- average of τ_{PD} as a constraint allows considerable freedom in the spatial and temporal
- distribution in the model, we find it still provides a robust constraint, and minimizes observation
- and sampling uncertainties.

¹ The emulators are created using GPflow (Matthews et al. 2017) using a Gausian Process Regression model with a Radial Basis Function kernel and hyper-parameters optimized using the Broyden, Fletcher, Goldfarb, and Shanno (BFGS) algorithm (Nocedal and Wright 2006).

- 111 We consider all the sampled emulator variants whose τ_{PD} is outside one standard deviation of
- the mean in the observed values implausible, and hence reject that parameter combination. The
- observed values are not normally distributed (since they are not truly independent, sharing
- instruments and retrieval algorithms) and this uncertainty range almost spans the entire observed values. In effect we are using a wide top-hat (or box-car) distribution for the observed τ_{PD} (see
- Figure S1) and explore the sensitivity of our results to the width of this distribution below. This
- 117 'history matching' approach produces a 'constrained' set of model variants which is now
- compatible with the observations (see e.g. Lee et al. 2016). The parameter combinations
- 119 corresponding to these plausible simulations are then used to predict unobservable quantities
- such as τ_{ant} and RF_{ari}, providing the new, observationally constrained distributions.
- 121

122 **Table 1**: The satellite products and global mean values used to estimate the observational

Platform	Sensor	Retrieval	AOD	Reference
Aqua	MODIS	DarkTarget	0.159	Levy et al. 2013
Terra	MODIS	DarkTarget	0.175	Ibid.
ENVISAT	AATSR	ADV	0.168	Kolmonen et al. 2016
ENVISAT	AATSR	ORAC	0.174	Thomas et al. 2009
ENVISAT	AATSR	SU	0.136	North et al. 2002
Noaa18	AVHRR	DeepBlue & SOAR	0.146	Hsu et al. 2013
Seastar	SeaWiFS	DeepBlue & SOAR	0.130	Ibid.
Mean			0.155	
Standard deviation			0.018	

123 uncertainty in τ_{PD}

- 124 Figure 1 shows the relationship between τ_{ant} and τ_{PD} in the CMIP5 and Aerocom multi-model
- ensembles, as well as the joint probability distribution for both the unconstrained HadGEM-
- 126 UKCA PPE (contour lines) and the PPE constrained by the observations (as a hex-density plot²).
- 127 The marginal distributions of τ_{ant} and τ_{PD} for each of the ensembles are shown along the top
- and right-hand-side. The distribution of τ_{PD} in both the CMIP5 and AeroCom ensembles peaks just below the lower observational bounds, while the unconstrained PPE shows a larger spread
- and higher mean value above the upper observational bound. Aerosol models are typically
- 131 'tuned' to have a plausible τ_{PD} (although they appear to be biased low compared to these
- observations), while the PPE was designed to sample the full parametric uncertainty, and so this
- difference is not surprising. The higher range of values for τ_{PD} shown in the PPE is due to the
- large base τ_{PD} produced by the model (labelled 'default' in Fig.1) and the large range of
- 135 uncertainty in sea-salt emissions elicited during the construction of the experiment.

136 A clear relationship between global annual mean τ_{ant} and τ_{PD} is evident in each of the

137 ensembles and can be understood in simple physical terms. Firstly, it can be shown that the

138 covariance between X and X + Y, where X and Y are two normally distributed random

139 variables, must be positive. The fact that both τ_{ant} and τ_{PD} are not independent and co-vary

through shared removal mechanisms only increases this covariance. This is demonstrated by

141 drawing samples from the simple analytic model for τ_{ant} described by Charlson et al., 1992:

$$\tau_{ant} = \alpha_{SO_4} f(RH) Q_{SO_2} Y_{SO_4} L_{SO_4} / A,$$

- 142 where the molar scattering cross section (α_{SO_4}), enhancement in scattering due to humidification
- 143 (f), anthropogenic sulfur dioxide source strength (Q_{SO_2}), sulfate yield
- 144 (Y_{SO_4}) , sulfate lifetime (L_{SO_4}) , and global area (A) all have the same values and uncertainty
- 145 estimates. We then extend this to estimate τ_{PD} as:

$$\tau_{PD} = \alpha_{SO_4} f(RH) (Q_{SO_2} Y_{SO_4} + Q_{SO_4}^n) L_{SO_4} / A + \tau_{SS} + \tau_D + \tau_{OC},$$

- by including a natural sulfate source term ($Q_{SO_4}^n$, with the same lifetime) and including natural
- 147 sea-salt (τ_{SS}), dust (τ_D) and organic carbon (τ_{OC}) components from the distributions described
- in Bellouin et al., 2013. As shown in gray-scale contours of Figure 1, this distribution shows a
- 149 very similar relationship to both ensembles and is in remarkably good agreement with the
- observations. While this simple model does not include many of the processes and uncertainties
- as the more complex PPE, it demonstrates the basis for the relationship between τ_{ant} and τ_{PD} .

² A hex-density plot represents a 2D histogram using hexagonal bins – avoiding visual artefacts sometimes associated with square bins

- Indeed, the parameters which were found to affect the shape of the PPE joint distribution the 152
- most were the scaling of sea-salt and anthropogenic sulfate emissions, and the parameter scaling 153 removal through dry deposition (not shown) - the three key uncertainties in the simple model
- 154 (where all removal terms are represented by an SO4 lifetime). While the anthropogenic 155
- emissions primarily affect τ_{ant} and the sea salt emissions only affect τ_{PD} , the dry deposition 156
- affects both, providing the basis of the relationship in the PPE (see Figure S2). This allows an 157
- observational constraint on τ_{PD} (0.14 to 0.17) to be translated into a constraint on τ_{ant} (a 1 σ 158
- range of approximately 0.03 to 0.05). Visually inspecting the overlap between the multi-model 159
- 160 relationships and the observational range of τ_{PD} provides a very similar range of τ_{ant} of
- approximately 0.03-0.04. 161



Figure 1: Distributions of the present-day aerosol optical depth, τ_{PD} against the industrial-era 164 change in aerosol optical depth at 0.55 μ m, τ_{ant} , between pre-industrial and present-day, from 165 AeroCom Phase II and CMIP5 sstClimAerosol models. The full joint-probability distribution 166

sampled from the emulated HadGEM-UKCA 26 aerosol parameter PPE is shown as contour 167

lines, and the constrained distribution as a hex-density. The default and median model runs of the 168

PPE are also shown for completeness. The grey-scale contour lines show the joint distribution 169

- sampled from an analytic approximation described in the main text. The horizontal lines show
- the 1 σ observational uncertainty range in globally-averaged τ_{PD} , while the vertical lines show the resulting 1 σ range in τ_{ant} of the constrained PPE.
- 173
- 174 Clear-sky RF_{ari} against τ_{ant} for the same model ensembles is shown in Fig. 2. The full joint-
- probability distribution from the emulated PPE is shown with contour lines, while the values
- 176 constrained by the τ_{PD} observations are shown as a hex-density. The effect of the constraint of
- 177 the τ_{PD} on the spread in uncertainty in both τ_{ant} and RF_{ari} is immediately obvious. The full 178 distribution of RF_{ari} in the model variants sampled from the emulated PPE peaks at -0.8 W m⁻²
- distribution of RF_{ari} in the model variants sampled from the emulated PPE peaks at -0.8 W m⁻² and is non-negligible even at -1.2 W m⁻². After applying the constraint, the remaining variants
- provide a 1 σ range in clear-sky RF_{ari} of -0.54 -0.8 W m⁻². This is very similar to the original
- 181 AeroCom range $(-0.47 -0.84 \text{ W m}^{-2})$, and a similar range to that which would be provided by
- assuming a linear relationship in both the AeroCom and CMIP5 ensembles $(-0.7 -0.85 \text{ W m}^{-2})$
- using the values of τ_{ant} derived above. The RF_{ari} is directly proportional to τ_{ant} (Charlson et al.,
- 184 1992) and this is demonstrated by the excellent agreement in clear-sky forcing efficiencies $\frac{RF_{ARI}}{\tau_{ant}}$
- among all three ensembles. All of which are in-line with the values for the AeroCom models
- reported by Myhre et al., 2013 of -23.7 \pm 3.1 Wm⁻² τ^{-1} (neglecting an anomalous outlier).
- 187



Figure 2: Clear-sky radiative forcing of aerosol-radiation interactions, RFari in Wm-2, as a 190 function of the industrial-era change in aerosol optical depth at 0.55 μ m, τ_{ant} in AeroCom 191 models (green) and CMIP5 models (purple). The slopes of the lines of best fit for each dataset 192 are -19.1 and -21Wm⁻² τ^{-1} , respectively. The joint-distribution of the full emulated PPE is shown 193 with contours, while the samples consistent with τ_{PD} is shown as a hex-density. The default and 194 median model runs are also shown for completeness. The 1σ uncertainty in the fits are shaded 195 and the correlation coefficients are indicated in the parentheses in the legend. The 1σ ranges in 196 τ_{ant} and RFari from the constrained PPE are indicated by the vertical and horizontal lines 197 198 respectively. 199

Within this framework we can ask what effect a reduction in the observational uncertainty in τ_{PD} 201 would have on reducing the uncertainty in RF_{ari} from the PPE even further. Figure 3 shows the 202 uncertainty (1 σ spread in the constrained emulated samples) in RF_{ari} as a function of lower and 203 upper observational bounds on τ_{PD} . Naively, the uncertainty might be expected to decrease 204 closer to the diagonal where the observational uncertainty is smallest. However, the largest 205 206 control on the uncertainty in the RFari as constrained through τ_{PD} is actually the magnitude of the upper bound. This reflects the shape of the joint probability distribution in Fig.1 where the 207 lower bound is already on the edge of the plausible values from the PPE, whereas the upper 208 209 bound intersects the peak of the probability distribution. Intuitively this can be understood as the τ_{PD} containing less information about the anthropogenic aerosol as the magnitude increases, as 210 there is more flexibility in how the total is partitioned between anthropogenic and seasalt. 211







215 PPE for different lower and upper bounds on the corresponding emulated τ_{PD} .

217 **3 Discussion**

The anthropogenic contribution to the present-day aerosol loading has been a key source of 218 uncertainty in both bottom-up and top-down estimates of the aerosol forcing. We have 219 demonstrated a robust relationship between present-day total and anthropogenic aerosol loading 220 across two multi-model ensembles and a perturbed parameter ensemble. Combined with 221 observational bounds on the total present-day aerosol loading we estimate τ_{ant} to be in the 1σ 222 range 0.031 - 0.048 (or a 95% credible range of 0.026 - 0.054). For comparison, by determining 223 the anthropogenic contribution to fine mode aerosol in the Monitoring Atmospheric Composition 224 and Climate (MACC) Reanalysis (Benedetti et al. 2009), Bellouin et al. 2013 determine τ_{ant} as 225 0.06. This is slightly higher than our estimate, potentially due to relying on the MODIS retrieved 226 τ which are two of the largest values used in our observational dataset. The Max Planck Institute 227 Aerosol Climatology (MAC) (Kinne et al., 2006) combines AERONET climatologies with 228 aerosol properties from AeroCom models (Kinne et al., 2006) and report τ_{ant} of 0.03, which is 229

230 in good agreement with our estimate.

231 The plausible range in τ_{ant} translates into a constrained clear-sky RF_{ari} of -0.67 ± 0.13 Wm⁻² (or

a 95% credible range of -0.91 - -0.47 Wm⁻²). Although this range is not significantly smaller than the original ranges demonstrated in the AeroCom (Myhre et al., 2013) and CMIP5 (Zelinka

et al., 2014) ensembles, these were not sampling the full model uncertainty, as demonstrated by

the large uncertainty in the unconstrained PPE forcing for HadGEM-UKCA (-0.91 \pm 0.23 Wm⁻²,

or 95% range of $1.32 - 0.56 \text{ Wm}^{-2}$). While performing similar PPE experiments for each of the

237 models in the AeroCom and CMIP5 ensembles would reveal the full, presumably larger,

238 uncertainty in RF_{ari} , the robust relationship between the base models suggests this constraint may

hold. We have also explored the sensitivity of this constraint to the observational uncertainty in

240 τ_{PD} . The constraint is much more sensitive to the upper bound, rather than the spread and so a

robust estimate of this upper bound should be the focus for future investigation. It is also

242 possible that other retrieved properties, such as fine mode AOD or multiple-wavelength AOD

243 would provide a tighter constraint.

244 While all the model ensembles are in good agreement on the clear-sky forcing efficiencies, there

is still uncertainty around the magnitude of aerosol absorption, and in particular the absorptivity

- of black carbon (BC). Indeed, this PPE did not explore the large uncertainty in the imaginary part
- of the refractive index of BC. By combining the AOD constraint with observations of absorbing

AOD it may be possible to better constrain the magnitude of aerosol absorption. Future work will

use absorbing AOD measurements to constrain the RFari, including the (large) uncertainty in

these particles. The radiative forcing due to aerosol-cloud interactions (RF_{aci}) depends

logarithmically on τ_{ant} and so is even more sensitive to its uncertainty than RF_{ari} (Carslaw et al.,

252 2013). Despite AOD being an unreliable proxy for cloud condensation nuclei (Stier, 2016), a

constraint on anthropogenic fraction should be expected to constrain the anthropogenic

contribution to CCN, and hence help constrain RF_{aci}. Future work will explore this possibility.

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279 Data availability

The CMIP5 values are available as a supplementary table to Zelinka et al., 2014. The AeroCom

II data is read directly from the tables in Myhre et al. 2013. Raw simulation output data from the

HadGEM-UKCA PPE ensembles are available as Met Office postprocessing data format (.pp;

Met Office, 2013) from 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. The total data sizes are

286 around 85 TB.

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