The role of anthropogenic aerosols in the anomalous cooling from 1960 to 1990 in the CMIP6 Earth System Models

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Abstract The Earth System Models (ESMs) that participated in the 6th Coupled Model Intercomparison Project (CMIP6) tend to simulate excessive cooling in surface air temperature (TAS) between 1960 and 1990. The anomalous cooling is pronounced over the Northern Hemisphere (NH) midlatitudes, coinciding with the rapid growth of anthropogenic sulfur dioxide (SO_2) emissions, the primary precursor of atmospheric sulphate aerosols. Structural uncertainties between ESMs have a larger impact on the anomalous cooling than internal variability. Historical simulations with and without anthropogenic aerosol emissions indicate that the anomalous cooling in the ESMs is attributed to the higher aerosol burden in these models. The aerosol-forcing sensitivity, estimated as the outgoing shortwave radiation (OSR) response to aerosol concentration changes, cannot well explain the diversity of PHC biases in the ESMs. The relative contributions to aerosol-forcing-sensitivity from aerosol-radiation interactions (ARI) and aerosol-cloud interactions (ACI) can be estimated from CMIP6 simulations. We show that even when the aerosol-forcing-sensitivity is similar between ESMs, the relative contributions of ARI and ACI may be substantially different. The ACI accounts for between 64 to 87% of the aerosol-forcing-sensitivity in the models, and is the main source of the aerosol-forcing sensitivity differences between the ESMs. The ACI can be further decomposed into a cloud-amount term (which depends linearly on cloud fraction) and a cloud-albedo term (which is independent of cloud fraction, to the first order) with the cloud-amount term accounting for most of the inter-model differences.

1. Introduction

Surface air temperature (TAS) variation is an essential indicator of climate change, and reproducing the evolution of historical TAS is a crucial criterion for model evaluation. However, the historical TAS anomaly simulated by the models in the 6th Coupled Model Intercomparison Project (CMIP6) is on average colder than that observed in the mid-twentieth century, whereas the CMIP5 models tracked the instrumental TAS variation quite well (Flynn and Mauritsen, 2020). This is surprising because the transient climate response in CMIP6 models is generally higher than in CMIP5 models (e.g., Flynn and Mauritsen, 2020; Meehl et al., 2020).

As a result of anthropogenic emissions, atmospheric aerosol concentrations increased along with rising greenhouse gases, but with greater decadal variability. Aerosols are generally not evenly distributed around the planet as greenhouse gases, and they have relatively short lifetimes of the order of a week. Aerosols increased rapidly in the mid-twentieth century, predominantly due to US and European emissions. The rate of change of global aerosol emissions slowed down in the late 20th century (Hoesly et al., 2018), and the trend of global emission has been negative since the mid-2000s (Klimont et al., 2013). There has also been a shift in emission source regions. European and US emissions have declined following the introduction of clean air legislation since the 1980s, while Asian emissions have risen due to economic development. East Asian emissions clearly increased from 2000 to 2005, followed by a decrease with large uncertainties (Aas et al., 2020). The decade long emission reduction since 2006 over East China is not well represented by the CMIP6 emission (Wang et al., 2021).

Although greenhouse warming was concluded to be the dominant forcing for long-term changes (e.g., Weart, 2008; Bindoff et al., 2013), multidecadal variability in TAS and the reduced rate of warming in the mid-twentieth century in particular, has been attributed to aerosol forcing (e.g., Wilcox et al., 2013). Ramanathan and Feng (2009) noted that the aerosol cooling effect might have masked as much as 47% of the global warming by greenhouse gases in the year 2005, with an uncertainty range of 20~80%.

The aerosol cooling effect is mainly attributed to the ability of sulphate particles to reflect incoming solar radiation and modify the microphysical properties of clouds (e.g., Charlson et al., 1990; Mitchell et al., 1995; Lohmann and Feichter, 2005). The increase in anthropogenic aerosols was also responsible for weakening the hydrological cycle between the 1950s and the 1980s (Wu et al., 2013).

Previous work has suggested that the anomalous mid-twentieth century cooling in the CMIP6 models is the result of excessive aerosol forcing. Flynn and Mauritsen (2020) suggested that aerosol cooling is too strong in many CMIP6 models because there is no apparent relationship between the warming trends simulated by models and their transient climate responses (TCRs) before the 1970s. Dittus et al. (2020) found that historical simulations can better capture the observed historical record by reducing the aerosol emissions in HadGEM3-GC3.1, demonstrating an overly strong aerosol cooling effect. In this study we characterize the mid-twentieth century excessive cooling in CMIP6 ESMs. In order to quantify the role of aerosol processes in this anomalous cooling, historical experiments with and without anthropogenic aerosol emissions are employed. The remainder of the paper is organized as follows. Section 2 introduces the models, data, and a quantitative method to separate the aerosol forcing components. The major features of anomalous cooling in CMIP6 ESMs are examined in section 3. Section 4 investigates the possible reasons for the anomalous cooling. The relative importance of aerosol-radiation interactions and aerosol-cloud interactions in each ESM is quantified and discussed in section 5. Conclusion is given in Section 6.

2. Model, data, and method
2.1 CMIP6 ESMs

CMIP6 includes an unprecedented number of models with representations of aerosol-cloud interactions. Many also have interactive tropospheric chemistry and aerosol schemes. Six such ESMs are employed in this study: BCC-ESM1 (Wu et al., 2020; Zhang et al., 2021), EC-Earth-AerChem (van Noije et al., 2020), GFDL-ESM4 (Dunne et al., 2020), MPI-ESM-1-2-HAM (Neubauer et al., 2019), NorESM2-LM

89   (Seland et al., 2020), and UKESM1-0-LL (Sellar et al., 2019). The surface air

90   temperature simulated in corresponding models with lower-complexity are also

91   examined: BCC-CSM2-MR (Wu et al., 2019b), EC-Earth3 (Döscher et al., 2021), and

92   MPI-ESM1-2-LR (Mauritsen et al., 2019) with prescribed tropospheric chemistry and

93   aerosol; GFDL-CM4 (Held et al., 2019), NorCPM1 (Bethke et al., 2019), and

94   HadGEM3-GC31-LL (Williams et al., 2017) with prescribed tropospheric chemistry

95   and interactive aerosol scheme. BCC-CSM2-MR, EC-Earth3, and MPI-ESM1-2-LR

96   prescribe the anthropogenic aerosol forcings using the MACv2-SP parameterization

97   (Stevens et al., 2017). MACv2-SP approximates the observationally constrained spatial

98   distributions of the monthly mean anthropogenic aerosol optical properties and an

99   associated Twomey effect. A brief summary of the ESMs and the lower-complexity

100   models is introduced in Table 1.

Table 1. Information of the ESMs with interactive chemistry and aerosol scheme, as

well as the corresponding lower-complexity models.

Modeling group	ESM (Atmospheric Resolution)	Lower-complexity models (Atmospheric Resolution)	Prescribed tropospheric chemistry	Prescribed aerosol	Number of members	References
Beijing Climate Center (BCC)	**BCC-ESM1:** the BCC Earth System Model version 1 (T42, 26 layers to 2.914 hPa)	**BCC-CSM2-MR:** the median resolution BCC Climate System Model version 2 (T106, 46 layers to 1.459 hPa)	Y	Y	3	Wu et al.(2019b, 2020); Zhang et al. (2021)
European consortium of meteorological services, research institutes, and high-performance computing centres	**EC-Earth-AerChem:** the EC-Earth configuration with interactive aerosols and atmospheric chemistry (T255, 91 layers to 0.01 hPa)	**EC-Earth3:** the EC-Earth version 3 (T255, 91 layers to 0.01 hPa)	Y	Y	1	von Noije et al. (2020); Döscher et al. (2021)
US Department of Commerce/NOAA / Geophysical Fluid Dynamics Laboratory (GFDL)	**GFDL-ESM4:** the GFDL Earth System Model version 4 (C96, 49 layers to 1 hPa)	**GFDL-CM4:** the GFDL Climate Model version 4 (C96, 33 layers to 1 hPa)	Y	N	1	Dunne et al. (2020); Held et al. (2019)
Max Planck Institute for Meteorology (MPI)	**MPI-ESM-1-2-HAM:** the HAMMOZ-Consortium of MPI Earth System Model (T63, 47 layers to 0.01 hPa)	**MPI-ESM1-2-LR:** the lower-resolution version of MPI Earth System Model (T63, 47 layers to 0.01 hPa)	Y	Y	3	Neubauer et al.(2019); Mauritsen et al. (2019);
Norwegian Climate Center (NCC)	**NorESM2-LM:** the lower-resolution of Norwegian ESM version 2 (About 2º, 32 layers to 2 hPa)	**NorCPM1:** the Norwegian Climate Prediction Model version 1 (About 2º, 26 layers to 3 hPa)	Y	N	3	Seland et al. (2020); Bethke et al. (2019)
Met Office's Hadley Centre for Climate Prediction and Research (MOHC)	**UKESM1-0-LL:** U.K. Earth System Model version 1 (N96, 85 layers to 85 km)	**HadGEM3-GC31-LL:** the Hadley Centre Global Environment Model in the Global Coupled configuration 3.1 (N96, 85 layers to 85km)	Y	N	3	Sellar et al. (2019); Williams et al. (2017)

104 **2.2 Data**

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106 <center>**Table 2** Variables used in this study.</center>

Variable name	CMIP6 diagnostic label	Description	Units
TAS	tas	Surface air temperature	^{o}C
OSR	rsut	All-sky outgoing shortwave radiation at the top of atmosphere (TOA)	W m^{-2}
OSRclr	rsutcs	OSR assuming clear sky	W m^{-2}
mmrso4	mmrso4	Mass mixing ratio of sulphate aerosol in the atmosphere	kg kg-1
CLT	clt	Total cloud amount	%
r_{eff}	reffclwtop	cloud-top effective droplet radius	μm
loadSO4		Sulphate loading in the atmosphere, calculated from mmrso4	mg m^{-2}
OSRclr_hist		Mean OSRclr in the historical simulation from 1850 to 1990	W m^{-2}
CLT_hist		Mean CLT in the historical simulation from 1850 to 1990	%

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108   The CMIP6 historical experiment and hist-piAer experiment are employed. The

109 historical experiment is forced by time-evolving, externally imposed natural and

110 anthropogenic forcings, such as solar variability, volcanic aerosols, greenhouse gases,

111 and aerosol emissions (Eyring et al., 2016). The hist-piAer experiment is designed by

112 the CMIP6-endorsed Aerosol Chemistry Model Intercomparison Project

113 (AerChemMIP; Collins et al., 2017). It is run in parallel with the historical experiment

114 but fixes aerosol and aerosol precursor emissions to pre-industrial conditions.

115 Therefore, the differences between these two experiments are attributable to

116 anthropogenic aerosol emissions. The design of the hist-piAer simulation means that it

can also capture any nonlinearities resulting from GHG-driven changes in clouds. This is in contrast to the hist-aer simulations available from the Detection and Attribution Model Intercomparison Project (DAMIP; Gillett et al., 2016), which resembles the historical simulations but are only forced by transient changes in aerosol.

The monthly outputs from historical and hist-piAer simulations for ESMs are used, including TAS, all-sky outgoing shortwave radiation at the top-of-atmosphere (OSR), OSR assuming clear sky (OSRclr), mass mixing ratio of sulphate aerosol in the atmosphere (mmrso4), total cloud amount (CLT), and cloud-top effective droplet radius (r_{eff}). These variables are summarized in Table 2. The corresponding lower-complexity models have conducted the historical but not the hist-piAer simulations, and only the monthly TAS output from the historical simulations are used. Therefore, we focus on the ESMs when identifying the main aerosol processes contributing to the anomalous cooling.

The verification data used in this study is HadCRUT5, the monthly 5°lat by 5°lon gridded surface temperature (Morice et al., 2021), a blend of the Met Office Hadley Centre SST data set HadSST4 (Kennedy et al., 2019) and the land surface air temperature CRUTEM5 (Osborn et al., 2021).

2.3 Method

By comparing the TAS anomalies in ESMs and the lower-complexity models with HadCRUT5, our study found that TAS anomalies from 1960 to 1990 relative to 1850-1900 in ESMs and most of the lower-complexity models are on average much lower than observed, resembling a "pot-hole" shape. The magnitude of this anomalous cooling, i.e., the "pot-hole" cooling (PHC), is quantified as the near-global mean (60°S to 65°N) difference in the TAS anomaly between models and HadCRUT5 from 1960 to 1990. The variations over the polar regions (north of 65°N and south of 60°S) are not considered due to the lack of long-term reliable observations (Wu et al., 2019a).

The aerosol cooling   is dominated by the contribution of sulphate aerosol as

estimated by models and observations (Myhre et al., 2013; Smith et al., 2020). We use the evolution of sulphate loading (loadSO4) through the historic simulation as a proxy for total aerosol concentration changes to link estimates of the impact of aerosol forcing. Whilst the overall impact of aerosol forcing will also depend on other aerosol species, we adopt this approach because the sulphates dominate estimates of aerosol forcing during this period and other aerosols species can be assumed (as a 1st order approximation) to have covaried with the SO_2 emissions during this period as presented by the Community Emissions Data System (CEDS) inventory adopted by CMIP6 models (Hoesly et al, 2018). As such when we present estimates of the aerosol impact/loadSO4 we are presenting the impact of all aerosol species (including absorbing aerosols such as black carbon) as they covary with the sulphate concentrations during the historic period. The motivation for presenting it in this way, is we can separate differences in ESM responses to changes in aerosol amount from the differences in aerosol amount (represented by loadSO4) simulated by the ESMs.

We can estimate the impact of anthropogenic aerosol by using the difference in OSR between the historical and hist-piAer simulations, *ΔOSR*. *ΔOSR* of course involves any differences in natural variability and planetary albedo between the two simulations, including clear-sky albedo changes and any adjustments in the microphysical or macroscopic properties of clouds. The sensitivity of the OSR-response to aerosol changes, i.e., the aerosol-forcing-sensitivity, can be measured by the linear fit slope between the annual mean globally averaged OSR differences and loadSO4 differences between the historical and hist-piAer simulations:

Aerosol-forcing-sensitivity = *ΔOSR* /*ΔloadSO4*.   (1)

In this study, we diagnose the OSR differences from historical simulations that also capture the temperature response. As such the OSR differences do not represent a measure of only the aerosol forcing impact but combine OSR differences arising from both the aerosol forcing and the temperature response to this forcing, which we refer to in this manuscript as the aerosol-forcing-sensitivity. It presents a measure of the

174   importance of aerosol changes in simulated temperature changes that can be easily

175   calculated for existing transient simulations. The aerosol-forcing-sensitivity is different

176   from the commonly used aerosol effective radiative forcing (ERFaer), which is the

177   change in net TOA downward radiative flux after allowing adjustments in the

178   atmosphere, but with sea surface temperatures and sea ice cover are fixed at

179   climatological values. The ERFaer for each ESM except MPI-ESM-1-2-HAM is listed

180   in Table 3 and compared with the aerosol-forcing-sensitivity in section 4.

181   The aerosol-forcing-sensitivity can be further partitioned into a contribution from

182   aerosol-radiation interactions (ARI), and aerosol-cloud interactions (ACI). ARI and

183   ACI can be readily estimated from the CMIP6 output because annual mean cloud

184   amount (CLT), OSR, and the OSR *assuming only clear-sky* (OSRclr), are available for

185   all the CMIP6 ESMs. For each model, the clear-sky part OSR, OSRclr_p, can be

186   estimated as (1-CLT/100.)*OSRclr. The aerosol-forcing-sensitivity in clear-sky part

187   can therefore be estimated as:

188   *ΔOSRclr_p/ Δ loadSO4 =   (1-CLT_hist/100.)*ΔOSRclr/ΔloadSO4*

189   *- OSRclr_hist/100*ΔCLT/ΔloadSO4+residual_clr,*   (2)

190   where CLT_hist and OSRclr_hist are the mean CLT and OSRclr in the historical

191   experiment. The aerosol-forcing-sensitivity in cloudy part are relative to cloud amount

192   response to aerosol loading and cloud radiative effect changes and can be estimated as:

193   *ΔOSRcld_p/ Δ loadSO4 = A*ΔCLT/ Δ loadSO4 + cloud-albedo term*

194   *+residual_cld.*   (3)

195   Therefore, the aerosol-forcing-sensitivity can be decomposed as:

196   *ΔOSR/ΔloadSO4*   *= (1-CLT_hist/100)* ΔOSRclr/ΔloadSO4*

197   *Aerosol-forcing-sensitivity*   *+ (A-OSRclr_hist/100.)* ΔCLT/ΔloadSO4*

198   *+ cloud-albedo term   +   residual*

199   *= (1-CLT_hist/100)*M   +   (A-OSRclr_hist/100.)*N*

200   *Aerosol-rad. Interactions (ARI)   cloud-amount term*

$$+ \textit{cloud-albedo term} \quad + \quad \textit{residual}, \tag{4}$$

where M, N and A are empirically determined parameters. The parameter M is the slope of a linear fit of ΔOSRclr to ΔloadSO4, and therefore measures the strength of the aerosol-radiation interactions in each model. The first term on the right-hand side of Eq. (4), *(1-CLT_hist/100.)*M,* can therefore be identified with ARI. The parameter A is the slope of a linear fit of ΔOSRcld to ΔCLT, and therefore measures the correlation of the shortwave radiation reflected by clouds with changes in cloud amount. That is, the parameter A represents the baseline cloud albedo which is sensitive to the cloud parameterizations via Cloud Droplet Number Concentration (CDNC), cloud-droplet effective radius, and other factors. The parameter N is the slope of a linear fit of ΔCLT to ΔloadSO4, and therefore measures the sensitivity of cloud amount to aerosols. Note that changes in cloud amount by definition also affect the fraction of clear-sky, hence increases in OSRcld due to increases in CLT (i.e., *A*N*) can be partly offset by changes in area of clear-sky containing aerosols (OSRclr_hist/100.*N). The second term on the right-hand side of Eq. (4), *(A-OSRclr_hist/100.)*N,* can therefore contribute to the ACI. Specifically, it is the part of ACI that is linearly proportional to changes to cloud fraction, which we will refer to in this manuscript as the cloud-amount term. It is therefore sensitive to *any* aerosol-induced cloud fraction changes (Lohmann and Feichter, 2005), including any slow adjustments in clouds due to feedbacks within the Earth System.

In addition to depending on ΔCLT, ACI is also influenced by any changes in cloud-albedo that might occur independently of cloud-amount changes. Such adjustments would include increases in cloud droplet number concentration and increases in simulated cloud-droplet effective radius without accompanying changes in cloud cover. Changes purely in the brightness of clouds, without changes in macroscopic properties of clouds, are difficult to identify from the CMIP6 output because all the bulk-properties of clouds co-vary over the course of the projections. However, subtracting ARI and the cloud-amount term from the aerosol-forcing-sensitivity gives a residual that is, by definition, linearly independent of cloud fraction

differences (since by construction these have been regressed out). This residual can then be interpreted as due to differences in the albedo of clouds between the historical and hist-piAer, and will be called the "cloud-albedo term". Note that this method of calculation implies that purely albedo effects cannot be distinguished from general residual terms that result from the linear approximation made.

Decomposition of the ARI, the cloud-amount term and cloud-albedo term of ACI are detailed further in the Appendix. The aerosol-cloud feedbacks are mainly in the ACI term which includes cloud spatial extent (amount), cloud albedo on radiative fluxes, and cloud particle swelling by humidification (Christensen et al., 2017; Neubauer et al., 2017). There is also a (smaller) effect of feedback on the ARI term that is also affected by cloud amount changes insofar as increased/decreased cloud cover can obscure/reveal clear-sky radiative fluxes. We acknowledge that the linear approximation in our method doesn't explicitly account for the absorption above clouds, or the adjustments to aerosol-radiation interactions (e.g., Carslaw et al., 2013) that are known to be locally important. Our formulation explicitly assumes that there is a broadly linear relationship between loadSO4 and emissions, and aerosol radiation with loadSO4 (and non-linearity due to cloud albedo or amount or any interaction is small at global scale as suggested in Booth et al. (2018)). Should these interaction terms be non-negligible in this analysis, we still expect the broader attribution of the reasons for the model diversity in temperature response over the PHC period, either how they simulate aerosol concentrations or how they simulate the response to this, to generally hold.

This decomposition method is an approximate approach designed to be used with existing simulations, rather than a strict decomposition by dedicated simulations/output variables not included in CMIP6. It can't tell us precise information about each interaction and adjustment, but it can give us an indication of why models behave differently.

3. The "pot-hole" bias in CMIP6 ESMs
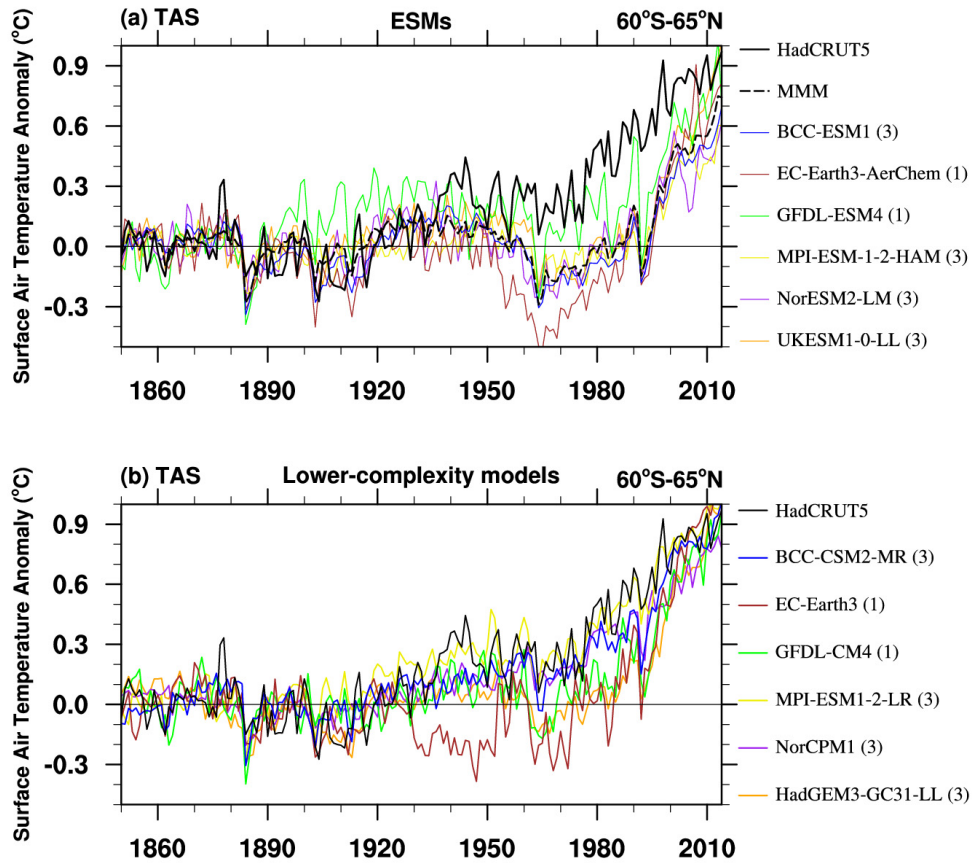
Figure 1. (a) Historical near-global mean (60ºS to 65ºN) surface air temperature (TAS) anomalies relative to 1850-1900 mean from HadCRUT5 (thick black line), the ensemble mean for each ESM (solid color lines), and multi-model mean (MMM, dashed black line). (b) is the same as (a), but for the lower-complexity models. Units: ºC. Value in bracket is the number of available members for each model.

Figure 1a shows the near-global averaged time series of annual mean TAS anomaly relative to 1850 to 1900 in HadCRUT5 during the historical period from 1850 to 2014, and the ensemble means for each model except for EC-Earth3-AerChem and GFDL-ESM4 (where only a single realization is available for the hist-piAer experiment). The unforced, long-term drifts in TAS may occur in some of the ESMs, as estimated by their control simulation under pre-industrial conditions (Yool et al., 2020). We have not accounted for long-term control simulation drifts in our study as we are assuming that our focus on inter-decadal scale variability of TAS anomalies is likely to be fairly insensitive to any century scale drifts.

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The TAS anomaly in HadCRUT5 is generally above the baseline climate from the 1940s onwards, and warms fastest from the 1980s to 1990s. Compared with the observations, all the ESM simulations have negative TAS anomaly biases after the 1940s, which are also evident in the ensemble-mean historical TAS of 25 CMIP6 models with and without interactive chemistry schemes (Flynn and Mauritsen, 2020). In the ESMs and their ensemble mean (MMM), the cold anomaly biases resemble a "pot-hole" shape (Fig.1a), which is relatively small before the 1950s and after the 2000s but prominent from the 1960s to 1990s. To reduce the impact of the change in the spatial pattern of the emissions in the late 20th century, and the Pinatubo eruption in the early 1990s, we mainly focus on the excessively cold anomaly from 1960 to 1990 in this study. The impacts from the Agung (1963) and El Chichon (1982) eruptions have been left in the PHC period as their effect on the simulated temperature is not as pronounced as the response to Pinatubo and are short-lived in time compared to the period we study. The period of anomalous cold in the global mean from 1960 to 1990 in model simulations is defined as the "pot-hole" cooling (PHC). Table 3 shows the TAS anomaly biases in two periods, the pre-PHC period (1929~1959) and the PHC period (1960~1990). The cold bias in the MMM is -0.14 in the pre-PHC period and intensified to -0.40 in the PHC period. The PHC bias ranges from -0.20°C to -0.58°C among the ESMs with a standard deviation of 0.11°C. Intra-model spread of PHC is relatively smaller. That is, model structural uncertainty is more responsible for PHC than internal climate variability.

Table 3. Biases in near-global averaged TAS anomalies relative to 1850-1900 from the ensemble mean and standard deviation (SD) for each ESM and the corresponding lower-complexity model in the pre-PHC (1929~1959) and the PHC period. Biases are relative to the HadCRUT5. The MMM and the SD of the ESMs are shown in the bottom row. The aerosol effective forcing (ERFaer) is also shown for each ESM. Note that the relevant fixed-SST simulations to calculate ERF were not available for MPI-ESM-1-2-HAM.

| ESMs	pre-PHC Ensemble mean (SD)	PHC Ensemble mean (SD)	ERFaer	Lower-complexity models	pre-PHC Ensemble mean (SD)	PHC Ensemble mean (SD)
BCC-ESM1	-0.12 (0.01)	-0.45 (0.07)	-1.47	BCC-CSM2-MR	-0.09 (0.01)	-0.10 (0.01)
EC-Earth-AerChem	-0.27	-0.58	-1.1	EC-Earth3	-0.37	-0.37
GFDL-ESM4	-0.02	-0.20	-0.70	GFDL-CM4	-0.12	-0.26
MPI-ESM-1-2-HAM	-0.16 (0.01)	-0.39 (0.03)	—	MPI-ESM1-2-LR	0.03 (0.03)	0.01 (0.01)
NorESM2-LM	-0.16 (0.04)	-0.41 (0.04)	-1.21	NorCPM1	-0.10 (0.03)	-0.08 (0.04)
UKESM1-0-LL	-0.10 (0.09)	-0.38 (0.08)	-1.1	HadGEM3-GC31-LL	-0.16 (0.02)	-0.33 (0.03)
MMM	-0.14 (0.08)	-0.40 (0.11)				

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The PHC bias is generally smaller in the corresponding lower-complexity models (Fig.1b and Table 3). For models with prescribed chemistry and aerosol (BCC-CSM2-MR and MPI-ESM1-2-LR), the TAS anomaly are reasonably reproduced during the pre-PHC period and the PHC period. The PHC bias are large (-0.37°C) in EC-Earth3, which has prescribed chemistry and aerosol. The large bias may be a reflection of the large internal variability on TAS in EC-Earth3 (Döscher et al., 2021), for which we have only one member. For models with prescribed chemistry and interactive aerosol scheme (GFDL-CM4 and HadGEM3-GC31-LL), the cold biases during the PHC period are comparable with that in the corresponding ESMs.

The spatial and temporal evolution of annually averaged TAS anomalies are further examined (Fig.2). In HadCRUT5, TAS anomalies are generally positive after the 1940s. The most significant TAS anomalies are evident in the late 20th Century and at the beginning of the 21st Century, especially over the NH midlatitudes. The results

319 from BCC-CSM2-MR and MPI-ESM1-2-LR agree well with the observations.
320 However, the ESMs and the other lower-complexity models simulate pronounced cold
321 anomalies over NH subtropical-to-high latitudes during the PHC period. The
322 overestimated tropical and southern hemispheric warming in NorCPM1 offsets most of
323 the cooling biases over NH subtropical-to-high latitudes.

324 Surface anthropogenic SO_2 emissions rapidly increase during the PHC period (the
325 line contours in Fig.2). The latitudes of the cooling centers in the ESMs and lower-
326 complexity models with interactive aerosol scheme are spatially co-located with the
327 SO_2 emission sources – North America and East Asia (at around 30ºN) and Western
328 Europe (at around 50ºN). Generally, the different behaviours seen in Fig.1 and Fig.2
329 suggest that aerosol forcings may be overestimated in the ESMs and lower-complexity
330 models with interactive aerosol scheme, and the anomalous cooling is a result of the
331 extra complexity associated with aerosol processes.
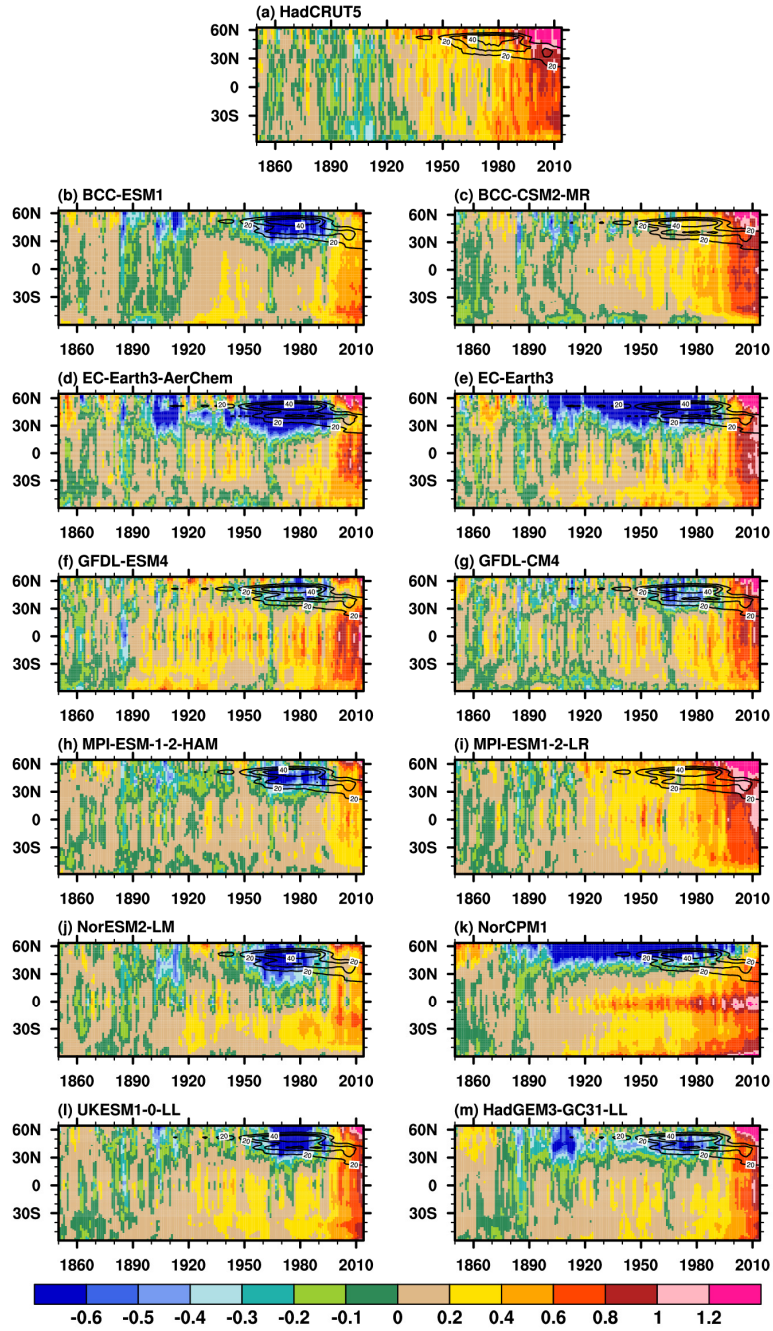
Figure 2. Time-latitude cross-section for annual-mean TAS anomalies (shaded) from (a) HadCRUT5, the ensemble mean for each ESM (left panel), and the corresponding lower-complexity model (right panel). The anomalies are related to the 1850 ~ 1900 mean. Units: °C. Note that the color scale intervals in the positive and negative directions are 0.2 °C and -0.1 °C, respectively. Line contours range from 20 to 40 ng m^{-2} s^{-1} with an interval of 10 ng m^{-2} s^{-1} show the zonal mean anthropogenic surface SO$_2$ emission provided by CMIP6.
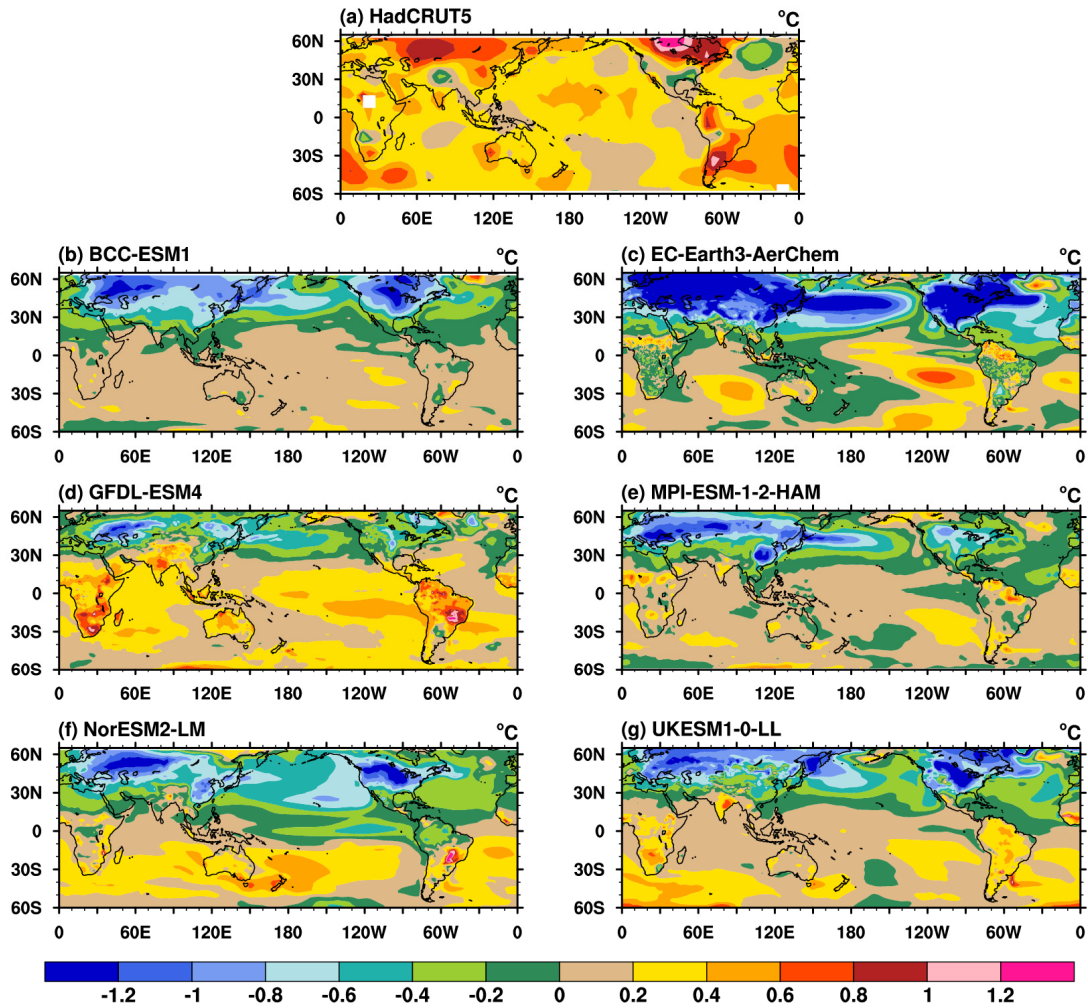
Figure 3. The TAS anomalies during the "pot-hole" period (1960 ~ 1990) from (a) HadCRUT5 and (b-g) the ensemble mean for each ESM. The anomalies are relative to the 1850~1900 mean. Units: °C.

During the PHC period. The TAS anomalies in HadCRUT5 are generally positive and are the largest over Eurasia and North America (Fig.3a). The warm anomalies are on average more than 0.4 °C along the 30°N ~ 60°N latitudinal belt. However, the ESMs show anomalies with the opposite sign (Fig.3b-3g) as do the lower-complexity models with interactive aerosol scheme (figures not shown). The PHC is pronounced over major SO_2 emission centers (Western Europe, East Asia, and the east US) and their downstream regions. The cold anomalies over Eurasia and North America are lower than -0.6°C in the ESMs. The PHC biases are strongest at lower levels (Figures not shown), which is distinct from the amplified upper-tropospheric warming response to

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4. Possible reasons for the excessive cooling

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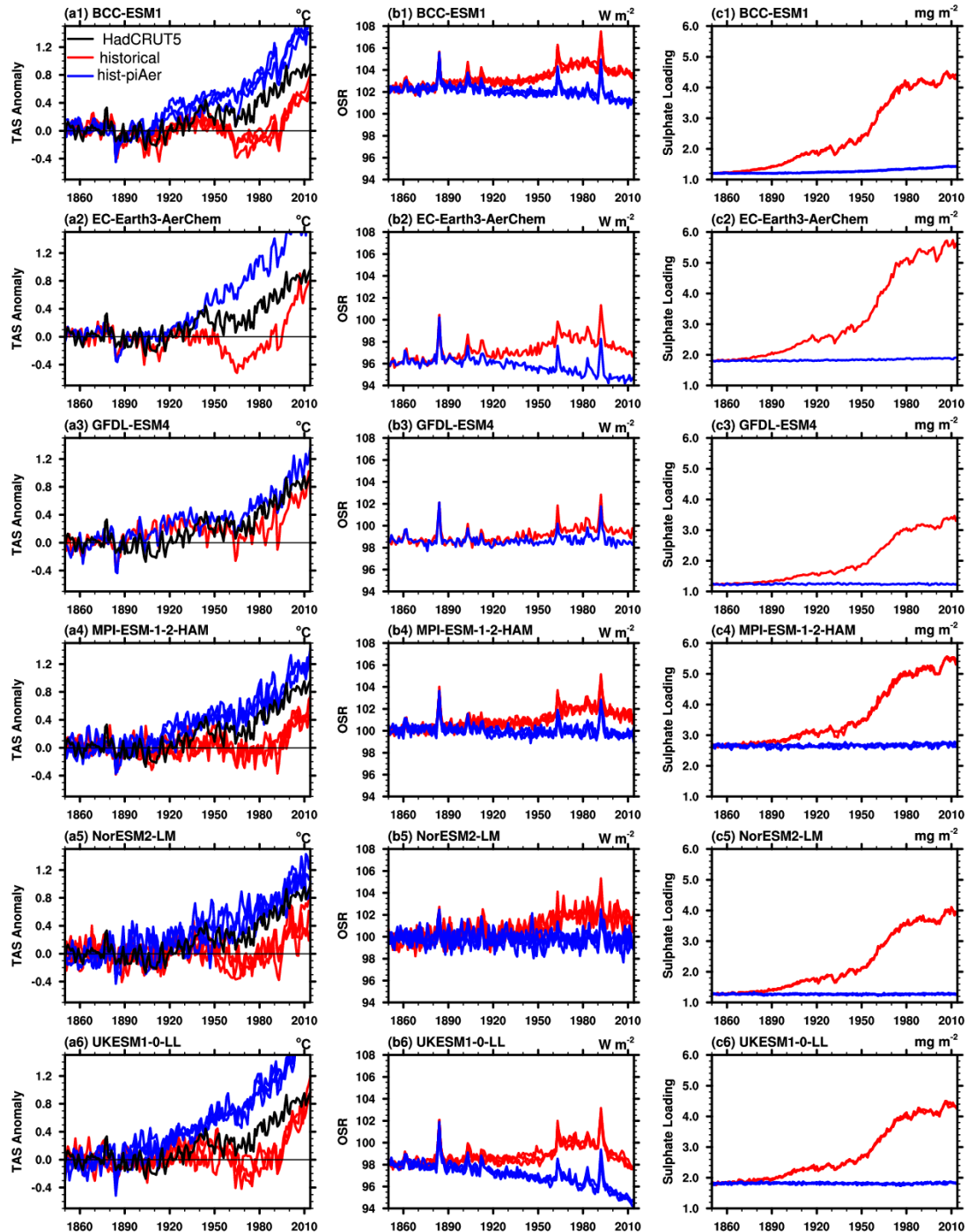
Figure 4. Evolutions of global annual means of (a1-a6) TAS anomalies (left panel, units: °C.), (b1-b6)

outgoing shortwave radiation at TOA (OSR, middle panel, units: W m^{-2}), and (c1-c6) sulphate loading

(right panel, units: mg m^{-2}) in HadCRUT5 (black line), each ESM member of the historical (red lines),
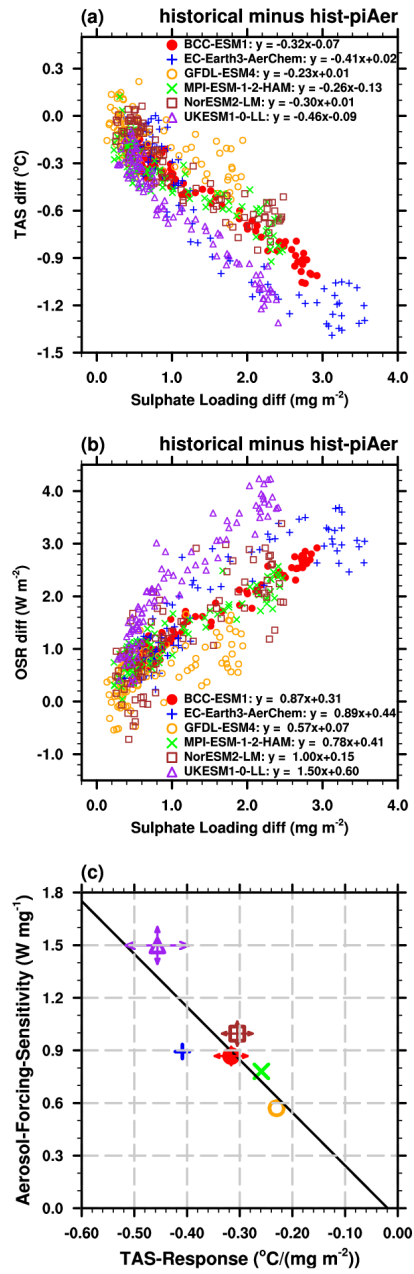
363   and hist-piAer experiments (blue lines). The TAS anomalies are relative to the 1850~1900 mean.

364

365   The differences between the historical and hist-piAer simulations help to

366   investigate the impact of anthropogenic aerosol emissions and its possible contribution

367   to the PHC biases. In this section, we examine the TAS, OSR, and sulphate loading

368   differences, and look in detail at their relationship. As shown by the evolution of TAS

369   anomalies in the two experiments (Fig.4, left panel), during the PHC period TAS

370   anomalies in HadCRUT5 (black line) are higher than those in the historical members

371   but lower than those in the hist-piAer members in all ESMs. That is, the model

372   responses to anthropogenic aerosol emissions are larger than the amplitude of the PHC.

373   The temporal evolution of the OSR corresponds with that of the TAS but occurs in the

374   opposite direction (middle panel). The sulphate loading differences are relatively small

375   in the 19th Century, mildly increase in the first half of the 20th Century, grow most

376   rapidly during the PHC period, and remain high afterward (right panel). The growing

377   sulphate loading during the PHC period corresponds with the increase in northern-

378   hemisphere anthropogenic surface SO_2 emissions (line contours in Fig.2). In

379   comparison with the TAS and OSR differences, the intra-model spread of sulphate

380   loading for each ESM is relatively small. However, the inter-model diversity of

381   sulphate loading is large. For example, the sulphate loading difference between the

382   historical and hist-piAer experiments around the year 2000 is about 4 mg m^{-2} in EC-

383   Earth3-AerChem, almost twice of that in GFDL-ESM4. With similar anthropogenic

384   SO_2 emission rates, the lower sulphate loading difference in GFDL-ESM4 indicates it

385   has a shorter sulphate aerosol residence time than that in EC-Earth3-AerChem, which

386   may be due to their different sulphate production and deposition schemes. The sulphate

387   loading diversity is also evident in CMIP5 models and is partly responsible for the

388   diversity in modeled radiative forcing (Wilcox et al., 2015).

389   The latitudinal movement of the SO_2 emission center from the 1990s affects the

390   relative strength of aerosol forcing. Due to the more rapid oxidation and higher

391   incoming solar flux at lower latitudes, an equatorward shift in SO_2 emissions around

392   1990s result in a more efficient production of sulphate and stronger aerosol forcing

393   (Manktelow et al., 2007). The northern mid-latitude temperature is also more sensitive

394   to the distribution of aerosols, which is approximately twice as large as the global

395   average (Collins et al., 2013; Shindell and Faluvegi, 2009). Therefore, we focus on the

396   relationships between TAS, OSR and sulphate loading after 1900 when SO_2 emissions

397   changes are dominated by its anthropogenic component, and before 1990.. As shown

398   in Fig.6a, the TAS differences between the historical and hist-piAer simulations vary

399   approximately linearly with the differences in the sulphate loading. The OSR

400   differences are approximately linearly correlated with sulphate loading differences

401   (Fig.6b). In both cases, the approximation of linearity holds less well for UKESM1-0-

402   LL, especially at small sulphate loadings. This reflects the behaviour of HadGEM2, a

403   predecessor of UKESM1 (Wilcox et al., 2015), and is likely to be due to the strong

404   aerosol-cloud albedo effect in these models. The global mean annual mean reff

405   decreases by about 0.7 μm since pre-industrial era, more than twice the magnitude of

406   change seen in the other models (Fig.1b in Wilcox et al., 2015 and Fig.9b in this study).

(a) historical minus hist-piAer

BCC-ESM1: y = -0.32x-0.07
EC-Earth3-AerChem: y = -0.41x+0.02
GFDL-ESM4: y = -0.23x+0.01
MPI-ESM-1-2-HAM: y = -0.26x-0.13
NorESM2-LM: y = -0.30x+0.01
UKESM1-0-LL: y = -0.46x-0.09

TAS diff (°C) vs Sulphate Loading diff (mg m⁻²)

(b) historical minus hist-piAer

BCC-ESM1: y = 0.87x+0.31
EC-Earth3-AerChem: y = 0.89x+0.44
GFDL-ESM4: y = 0.57x+0.07
MPI-ESM-1-2-HAM: y = 0.78x+0.41
NorESM2-LM: y = 1.00x+0.15
UKESM1-0-LL: y = 1.50x+0.60

OSR diff (W m⁻²) vs Sulphate Loading diff (mg m⁻²)

(c) Aerosol-Forcing-Sensitivity (W mg⁻¹) vs TAS-Response (°C/(mg m⁻²))

Figure 5. Scatter plots of 1900-1990 yearly sulphate loading differences between the historical and hist-piAer simulations (x-axis) versus (a) TAS differences and (b) OSR (y-axis). Results are from the ensemble mean for each ESM. The captions are the linear fitting equations. (c) shows the TAS response (x-axis) and aerosol-forcing-sensitivity (y-axis) which is equal to slope of linear fitting for each ESM (markers), and the corresponding intra-model spread (arrows).

The slope of the linear fitting equation between TAS (OSR) and sulphate loading as shown in the captions in Fig.5a (Fig.5b) is a measure of the sensitivity of TAS (aerosol forcing) to perturbations in atmospheric aerosol. Moreover, TAS-response and

21

417   aerosol-forcing-sensitivity are linearly correlated across the ESMs (Fig.5c). That is, the

418   strength of the TAS-response can be understood as the magnitude of aerosol-forcing-

419   sensitivity within each ESM. The TAS-response and aerosol-forcing-sensitivity is the

420   lowest in GFDL-ESM4. The TAS-response and aerosol-forcing-sensitivity in

421   UKESM1-0-LL (the purple marker in Fig.6c) are the strongest, as well as their intra-

422   model spread (the length of arrows), indicating that TAS and aerosol forcing in this

423   model are relatively more susceptible to changes in aerosol. The aerosol-forcing-

424   sensitivity is not correlated with the aerosol effective radiative forcing (ERFaer, Table

425   3), largely due to the strong influence of UKESM1-0-LL on the result.
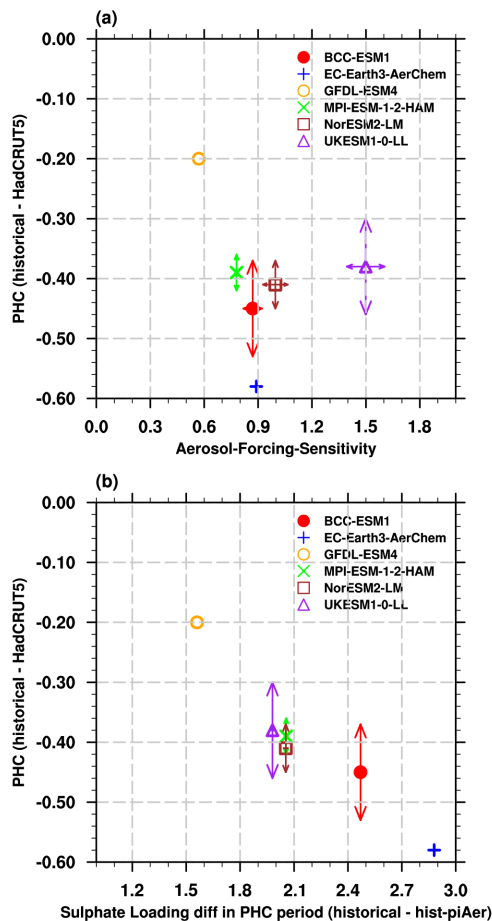
426

427   **Figure 6.** Pot-hole Cooling (PHC) bias in ESMs (°C) versus (a) the aerosol-forcing-sensitivity (W mg^{-1})

428   and (b) sulphate loading differences (mg m^{-2}) during the PHC period. The arrows show the uncertainty

429   ranges among the members in each ESM.

430

431   Considering the close relationship between TAS anomalies and aerosol loading

432 (Fig.5a), and the impact of aerosol-forcing-sensitivity on the TAS response in ESMs

433 (Fig.5c), their relative contributions to the PHC biases are examined. Figure 6a shows

434 the PHC biases versus the aerosol-forcing-sensitivity (markers) and their intra-model

435 spread (arrows). GFDL-ESM4 has the weakest aerosol-forcing-sensitivity (~0.60 W

436 mg^{-1}) and the smallest PHC (-0.20 °C). However, the relationship between the PHC

437 biases and the aerosol-forcing-sensitivity among the ESMs is not clear: ESMs have

438 similar PHC biases (MPI-ESM, NorESM2, and UKESM1) show large differences in

439 the aerosol-forcing-sensitivity, ranging from 0.78 to 1.5 W mg^{-1}; the aerosol-forcing-

440 sensitivity in EC-Earth3-AerChem is close to that in BCC-ESM1, but the PHC is more

441 than 0.1°C lower; the aerosol-forcing-sensitivity in UKESM1-0-LL is the strongest

442 (~1.5 W mg^{-1}) but not the PHC bias. Therefore, the aerosol-forcing-sensitivity is not

443 able to explain the different PHC biases among ESMs.

444 As shown in Fig.6b, the sulphate loading differences between the historical and hist-

445 piAer experiments during the PHC period are large among ESMs (the X-axis), which

446 are about 1.5 mg m^{-2} in GFDL-ESM4 but approximately 2.9 mg m^{-2} in EC-Earth3-

447 AerChem. The sulphate loading differences during the PHC period and PHC biases

448 shows a negative correlation: the PHC bias is generally larger in models with higher

449 sulphate loading over this period; the ESMs with similar PHC biases (MPI-ESM,

450 NorESM2, and UKESM1) show similar aerosol loading differences. Therefore, the

451 excessive cooling during the PHC period and the inter-model diversity in ESMs are

452 attributed to the higher aerosol burden in these models.

453 **5. Discussion**

454 **5.1 The proportions of ARI and ACI**

455 Although the aerosol-forcing-sensitivity is not responsible for the anomalous

456 cooling biases in ESMs, it is a good way to identify model differences in the response

457 to aerosol changes. As shown in Fig.5b, there are significant differences in the aerosol-

458 forcing-sensitivity among ESMs. The aerosol-forcing-sensitivity in UKESM1-0-L is

459 almost three times of that in GFDL-ESM4. Due to the uncertainties in physical

processes and cloud parameterizations, the dominant component (ARI or ACI) of aerosol-forcing-sensitivity may also vary among ESMs. Here, we separate the different components of the aerosol-forcing-sensitivity in each ESM by the method introduced in the section 2.3 and the Appendix. Sulphate loading is used as a proxy of aerosol amount for all aerosol components in the quantification of the total effect because of its dominant contribution to anthropogenic aerosol load during this period and its covariation with the other aerosol species.
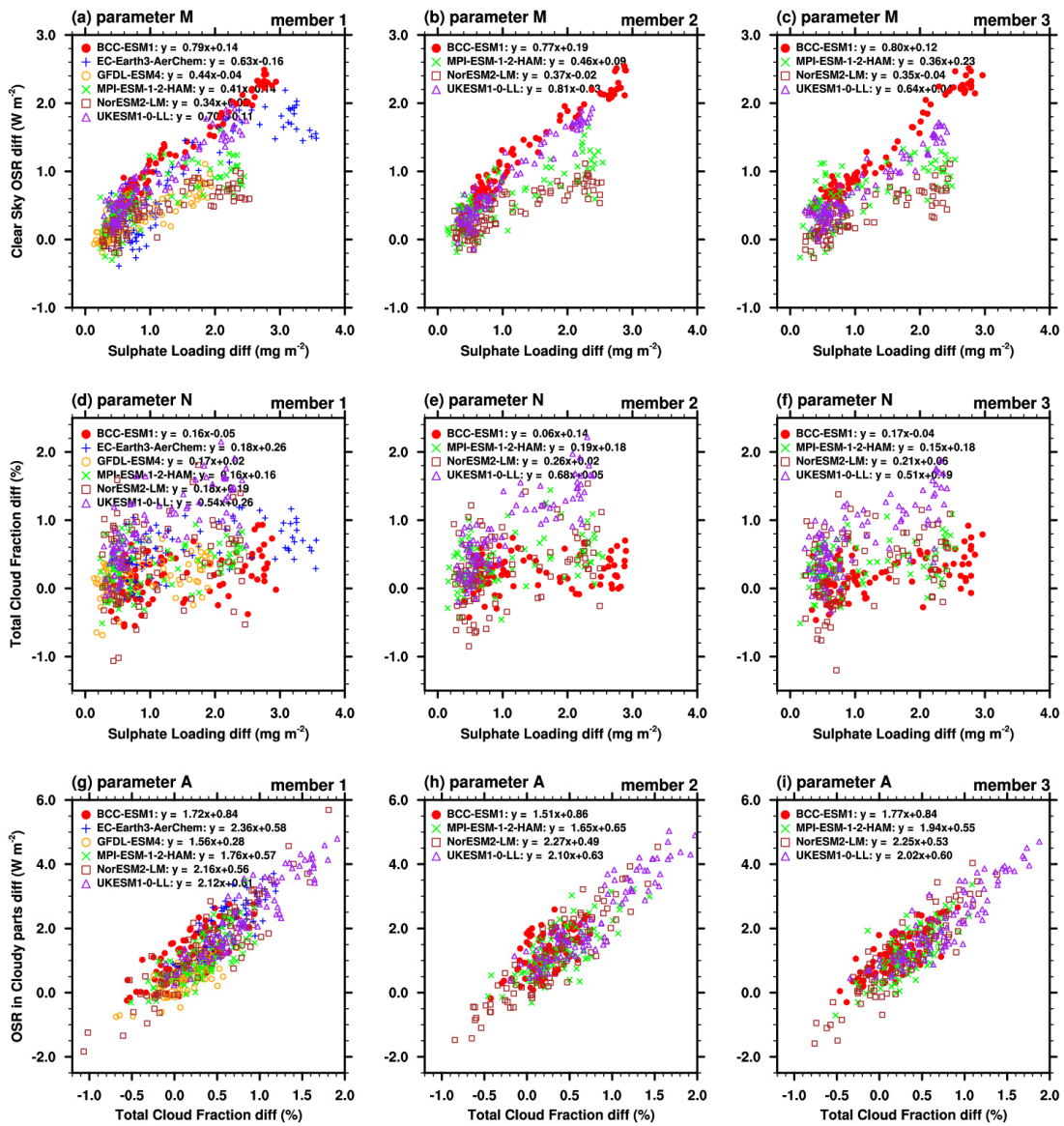
Figure 7. Annual mean differences between the historical and hist-piAer simulations in the ESM members during 1900 to 1990 period for (a-c) sulphate loading (mg m^{-2}) versus clear-sky OSR (OSRclr,

471   W m^{-2}), (d-f) sulphate loading versus total cloud fraction (%), and (g-i) total cloud fraction versus OSR

472   in cloudy parts (W m^{-2}). Slopes of the linear fitting equations from the top row to the bottom row refer

473   to the parameters M, N, and A, respectively.

474

475   The ARI can be approximated to (1-CLT_hist/100.)*M, where CLT_hist is cloud

476   amount in the historical simulation and parameter M is a measure of the strength of

477   aerosol-radiation interactions (*ΔOSRclr/ΔloadSO4)*. Parameter M varies widely from

478   about 0.35W mg^{-1} in NorESM2-LM to about 0.79 W mg^{-1} in BCC-ESM1 (captions in

479   Fig.7a-7c). Since parameter M does not change much among ensemble members in

480   each ESM, their ARI is similar across members. That is, the impact of internal climate

481   variability on the ARI is relatively small, which is consistent with the quantitative

482   analysis in Fig.8 (Red bars).

483
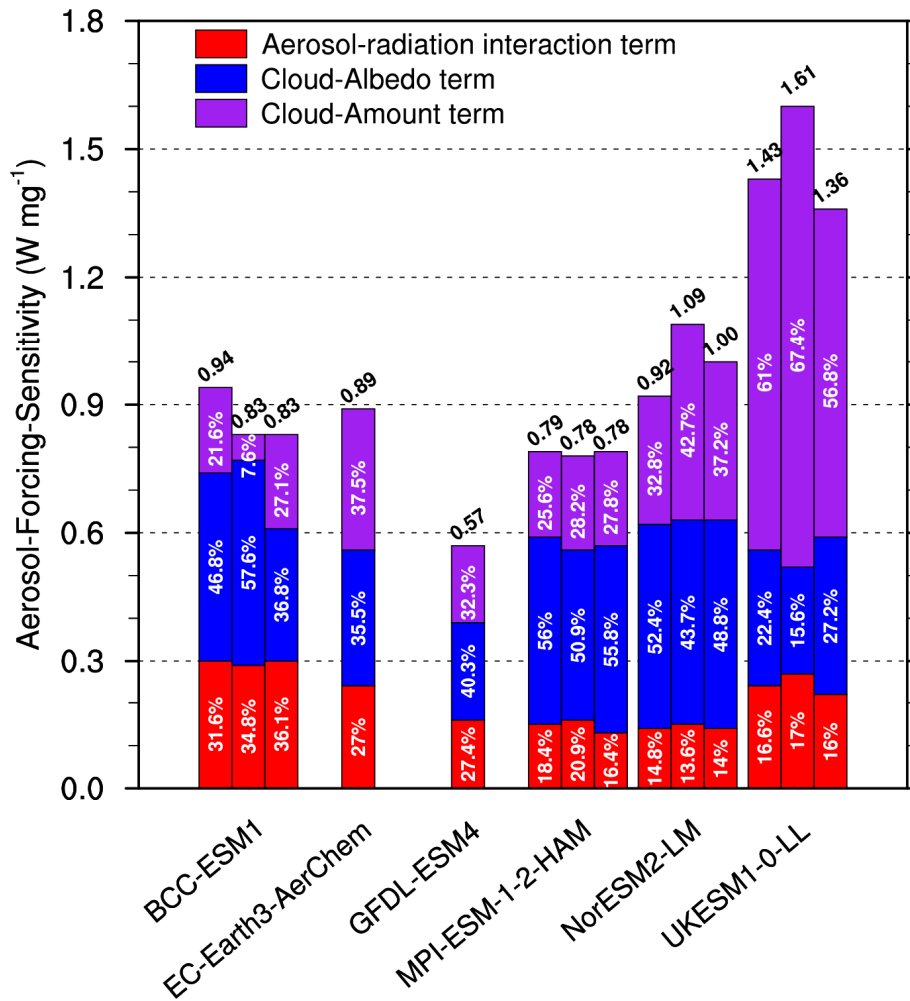
Figure 8. Total aerosol-forcing-sensitivity from each member in ESMs. The number marked on the top is the total aerosol-forcing-sensitivity. Partition of aerosol-radiation interaction term, cloud-albedo term, and cloud-amount term are marked in the corresponding color bars. Unit: W mg[-1]. Where multiple realizations are available for a model, a bar is shown for each member.

The ACI can be estimated from the difference between the aerosol-forcing-sensitivity and the ARI. The proportion of the aerosol-forcing-sensitivity arising from the ACI is higher than 64% in all ESMs (Fig.8). The inter-model variation of the ACI (0.37 W mg[-1]) is much larger than that for the ARI (0.09W mg[-1]). For example, the ACI in UKESM1-0-LL (~1.2W mg[-1]) is higher than all the others and is about three times of that in GFDL-ESM4 (0.41 W mg[-1]). This demonstrates that differences in the aerosol-forcing-sensitivity across the ESMs are dominated by the differences in their

individual representation of ACI. Chen et al. (2014) also suggested that ACI is the main contribution to the Aerosol radiative forcing uncertainty and the response of marine clouds to aerosol changes is paramount. The intra-model variations in the ACI are also larger than that for the ARI. That is because the intra-model variations of the ACI are influenced by the effects of climate system internal variability on aerosol-induced cloud microphysics, with cloud radiative properties and cloud lifetimes varying regionally. The intra-model variations are also attributable to the differences in atmospheric circulation among different ensemble members, which may affect the geographical distributions of aerosols and clouds and lead to a different magnitude of interactions.

The quantitative analysis in Fig.8 also indicates that ESMs with similar aerosol-forcing-sensitivity may have different contributions from ARI and ACI. The aerosol-forcing-sensitivity is similar in BCC-ESM1, EC-Earth3-AerChem, MPI-ESM-1-2-HAM and NorESM2-LM, but the fractional contribution from the ACI is the largest in NorESM2-LM and its ARI is less than half of that in BCC-ESM1. Generally, BCC-ESM1 has the largest fractional ARI contribution (34%), whereas NorESM2-LM has the largest fraction of ACI contribution (86%).

5.2 The proportions of cloud-amount and cloud-albedo terms

Our ACI metric includes several mechanisms by which aerosols can alter cloud properties. This includes the cloud-albedo effects (or 'Twomey' effect), referred to as the radiative forcing part of ACI, and effects of aerosols on the macroscopic properties of clouds (for example, cloud extent and lifetime), referred to as the adjustments part of ACI. However, it is complicated to separate these two parts of ACI directly using available CMIP6 diagnostics, because the former is most accurately defined as a change in cloud albedo with all other cloud properties held constant (i.e., a change in cloud-droplet number concentration only), whilst the latter allows cloud properties to respond.
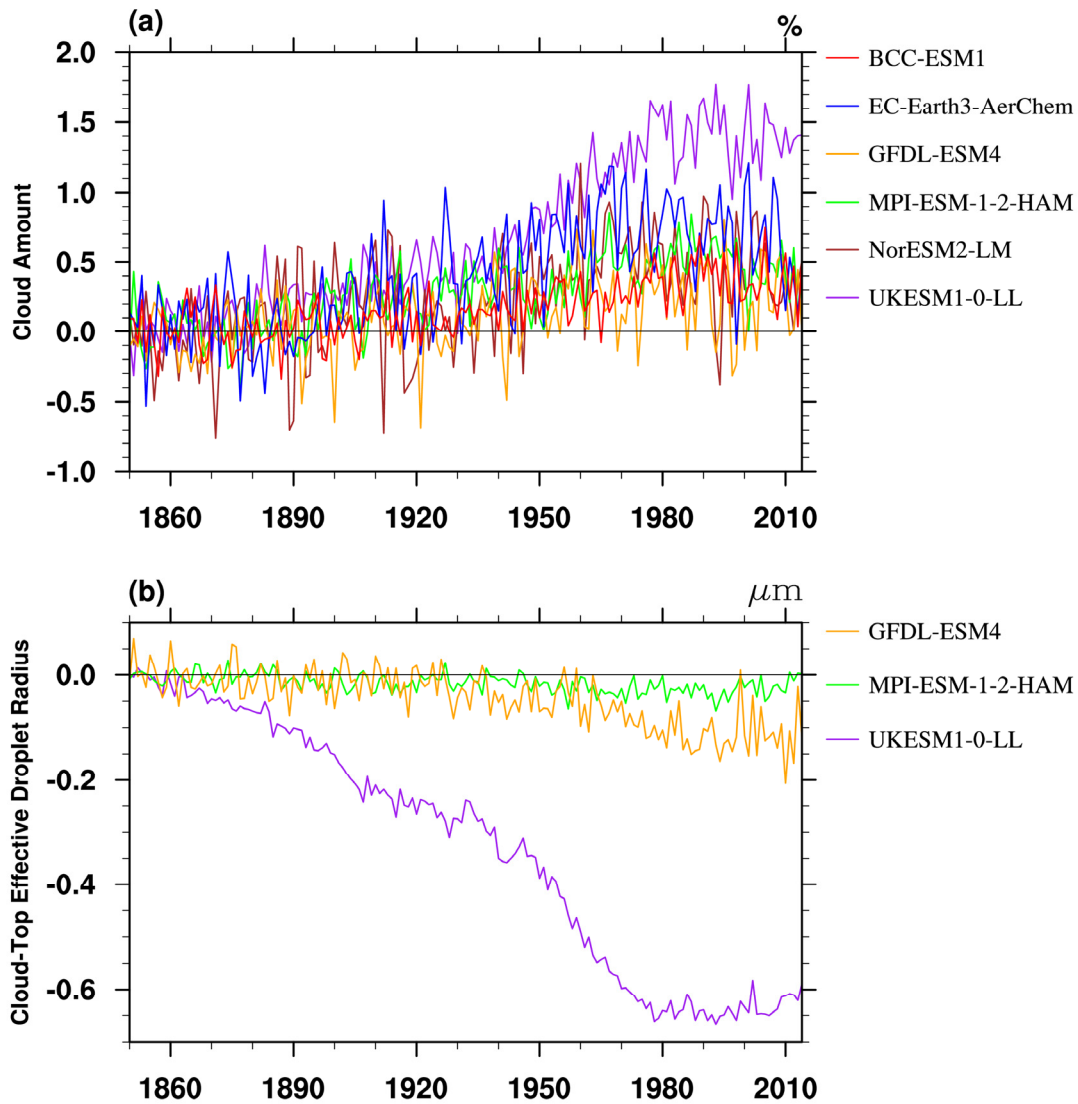
Figure 9 (a) Evolutions of global mean cloud amount differences between the historical and hist-piAer simulations in ensemble mean for each ESM, units: %. (b) is the same as (a), but for cloud-top effective droplet radius (r_{eff}, μm). The r_{eff} data is only available for GFDL-ESM4, MPI-ESM-1-2-HAM, and UKESM1-0-LL.

Figure 9 shows the evolution of global-mean differences in total cloud amount (ΔCLT) and cloud-top effective droplet radius (Δr_{eff}) between the historical and hist-piAer experiments. The ΔCLT and Δr_{eff} in UKESM1-0-LL are the largest and highly correlated with each other (with a correlation coefficient of -0.92 during the 1900 to 1990 period). For the other two ESMs for which Δr_{eff} was archived, the correlation

536 coefficient is -0.40 for MPI-ESM-1-2-HAM and insignificant for GFDL-ESM4 (-0.09).

537 The ΔCLT and Δr_{eff} differences are smaller in MPI-ESM-1-2-HAM and GFDL-ESM4

538 than in UKESM1-0-LL, especially for the Δr_{eff} differences. Δr_{eff} is generally related to

539 the cloud-optical depth and cloud water path, and ΔCLT is related to adjustments in

540 cloud cover due to ACI. Therefore, the radiative forcing part and adjustments part of

541 ACI may be closely coupled in UKESM1-0-LL and are hard to separate statistically.

542 The strong correlation between cloud amount and r_{eff} response in UKESM1-0-LL

543 indicates that this model is sensitive to aerosol-cloud interactions, which likely

544 contributes to it having the strongest aerosol-forcing-sensitivity and intra-model spread

545 of all the CMIP6 models (Fig.5c). MPI-ESM-1-2-HAM and UKESM1-0-LL have

546 similar ensemble mean PHC biases and close sulphate burden, but the aerosol-forcing-

547 sensitivity differences in UKESM1-0-LL is almost twice of that in MPI-ESM-1-2-

548 HAM (Fig.5). That is, the overestimated sulphate burden dominates the PHC biases,

549 but the ACI sensitivity may partly affect the amplitude and uncertainty ranges of PHC

550 biases.

551 Despite of the closely coupled radiative forcing part and adjustments part of ACI

552 in UKESM1-0-LL, it is still possible to split the ACI into a part that is correlated with

553 cloud amount differences and a residual term. This can be done statistically by

554 regressing-out the approximate linear dependence of the differences between historical

555 and hist-piAer simulations of the cloudy part of OSR (OSRcld_p) on cloud fraction in

556 each ESM (parameter A in Fig.7g-7i). We call the degree of linear correlation of

557 ΔOSRcld_p with ΔCLT the "cloud-amount term", and the residual will be referred to

558 as the "cloud-albedo term". However, we reiterate that the so-called "cloud-amount

559 term" may also include changes in the reflectivity of clouds if these are correlated with

560 changes in cloud amount. Similarly, the cloud-albedo term will contain any sources of

561 cloud amount changes which have not been removed by linearly regressing OSRcld_p

562 against cloud amount. As such, we do not intend this nomenclature to indicate a precise

563 separation of the radiative forcing part and adjustments part of ACI. Our decomposition

564 allows first order assessment of these terms from historical simulations without the need

for extra simulations or calls, and also allows estimates from observations and intermodel comparisons.

As described in the section 2.3 and the Appendix, the cloud-amount term is sensitive to two parameters: the cloud amount response (parameter N in Fig.7d-7f) and the sensitivity of OSR reflected from clouds to cloud amount changes (parameter A, Fig.7g-7i). As shown in Fig.8, UKESM1-0-LL has the largest contribution of the cloud-amount term to aerosol-forcing-sensitivity (62%, 0.91W mg^{-1}); the cloud-amount term is the smallest in GFDL-ESM4 (~0.18W mg^{-1}). The cloud-albedo term is defined to be linearly independent of cloud-amount changes (adjustments). For the CMIP6 ESMs, it can only be estimated as the residual after subtracting the cloud-amount term from the ACI. The cloud-albedo term is similar in BCC-ESM1, MPI-ESM-1-2-HAM, and NorESM2-LM. The inter-model variation for the cloud-amount term is about twice of that for the cloud-albedo term (0.29W/mg v.s. 0.16W/mg). That is, the variations of cloud-amount term are the major source of inter-model ACI (and the aerosol-forcing-sensitivity) differences between ESMs. Therefore, difference in the cloud-amount terms, across the ESMs, dominates the uncertainties in the aerosol-forcing-sensitivity.

Note that, our definitions do not correspond to the effects measured by using multiple calls to the radiation scheme of a model, with and without aerosols, which measure instantaneous radiative effects; multiple calls give a measure of the fast response of clouds to aerosol perturbations in a fixed thermodynamic and dynamical background, allowing for a clear separation between ACI and rapid adjustments (e.g., Bellouin et al., 2013). This differs from aerosol forcing diagnosed by differencing climate projections with different aerosol forcings, which include the slow effects of other feedbacks. For example, differences in climate forcings can lead to different SST patterns, which in turn alter the location and characteristics of clouds. Despite these differences, an advantage of our classification is that it provides a possible method for model evaluation since the variables used are also, in principle, available from the observations.

6. Conclusion

This study focuses on the reproduction of historical surface air temperature anomalies in six CMIP6 ESMs. The ESMs systematically underestimate TAS anomalies relative to 1850 to 1900 in the NH midlatitudes, especially from 1960 to 1990, the "pot-hole" cooling (PHC) period. Previous studies suggested that aerosol cooling is too strong in many CMIP6 models. Our study more specifically found that the PHC is concurrent in time and space with anthropogenic SO_2 emissions, which rapidly increase in the PHC period in NH. Models with larger aerosol burdens have larger PHC biases. The primary role of aerosol emissions in these biases is further supported by the differences between ESMs and the lower-complexity models with prescribed aerosol.

Differences between historical simulations and simulations with aerosol emissions fixed at their preindustrial levels (hist-piAer) are used to isolate the impacts of industrial aerosol emission. We propose that the overestimated aerosol concentrations in the ESMs are responsible for the spurious drop in TAS in the mid-twentieth century, rather than a high sensitivity of the models to aerosol forcing. Although the aerosol-forcing-sensitivity differences in ESMs cannot explain the PHC biases, it is a good measurement of aerosol effects that can be used to explore structural differences between models. A simple metric is derived for determining the dominant contribution to the aerosol-forcing-sensitivity in any specific model: ARI or ACI. The ACI accounts for more than 64% of the aerosol-forcing-sensitivity in all analyzed ESMs. The considerable inter-model variation in the aerosol-forcing-sensitivity is mainly attributable to the uncertainty in the ACI within models. The ACI can be further decomposed into a cloud-amount term and a cloud-albedo term. The cloud-amount term is found to be the major source of inter-model diversity of ACI. Considering the crucial role of cloud properties on the inter-model spread in aerosol-forcing-sensitivity, the aerosol-cloud interactions should be a focus in development of aerosol schemes within ESMs.

Appendix: Decomposition of the Aerosol-radiation interaction and aerosol-cloud interaction

Considering the dominant role of sulphate aerosol on anthropogenic aerosol forcing, we use the sulphate loading (loadSO4) as a proxy for all aerosol in our analysis. The aerosol-forcing-sensitivity (as determined by the difference between the historical and hist-piAer experiments) is estimated by the all-sky OSR differences per sulfate burden unit (ΔOSR/ΔloadSO4) and it is the combination of OSR differences in the clear-sky parts (ΔOSRclr_p/ΔloadSO4) and the cloudy parts (ΔOSRcld_p/ΔloadSO4):

$$\Delta OSR/\Delta \text{loadSO4} = \Delta OSRclr\_p/\Delta \text{loadSO4} + \Delta OSRcld\_p/\Delta \text{loadSO4}.$$

(A1)

The OSRclr_p for a particular experiment can be calculated as:

$$OSRclr\_p = (1-CLT/100.)*OSRclr, \qquad (A2)$$

where CLT is the total cloud amount (unit: %), and OSRclr is the OSR assuming all clear sky (unit: W/m^2). The cloud amount changes (ΔCLT) will modify the proportion of clear-sky and then affect the OSR changes attributed to the clear-sky part by covering or uncovering aerosols in clear sky. Therefore, based on equation (A2), ΔOSRclr_p/ΔloadSO4 can be decomposed into the OSRclr-response ($\Delta OSRclr/\Delta loadSO4$) and CLT-response ($\Delta CLT/\Delta loadSO4$):

$$\Delta OSRclr\_p/\Delta loadSO4 = (1-CLT\_hist/100.)*\Delta OSRclr/\Delta loadSO4$$

$$- OSRclr\_hist/100*\Delta CLT/\Delta loadSO4+residual\_clrp$$

$$= (1-CLT\_hist/100.)*M - OSRclr\_hist/100*N + residual\_clr, \quad (A3)$$

where CLT_hist and OSRclr_hist are the mean CLT and OSRclr in the historical experiment. Residual_clr is the residual term that is non-linear in ΔOSRclr and ΔCLT. The parameter M= $\Delta OSRclr/\Delta loadSO4$ is related to strength of aerosol-radiation interaction and can be estimated by linear fitting of ΔOSRclr on ΔloadSO4. The parameter N= $\Delta CLT/\Delta loadSO4$ is related to CLT-response and estimated by linear fitting of ΔCLT on ΔloadSO4. Therefore, the first term on the right-hand side of Eq. (A3), $(1-CLT\_hist/100.)*M$, corresponds to the aerosol radiative effect; the second term, $- OSRclr\_hist/100*N$, corresponds to the impact of changes in clear-sky area.

The OSRcld_p is the cloudy part of OSR, accounting for the difference between OSR and OSRclr_p. The cloudy part of the OSR differences (ΔOSRcld_p) can be

655   generally estimated as:

656    $\Delta OSRcld\_p = A*\Delta CLT + cloud\text{-}albedo\ relative\ changes + residual\_cld,$

657   where the parameter A $=\Delta(OSR\text{-}OSRclr\_p)/\Delta CLT$ is the sensitivity of the shortwave

658   flux reflected by clouds to changes in cloud amount. The parameter A depends on the

659   baseline cloud albedo (radiative flux per cloud amount unit) and can be estimated by

660   linear fitting of ΔOSRcld_p on ΔCLT. Hence,

661    $\Delta OSRcld\_p/\Delta loadSO4 = A*\Delta CLT/\Delta loadSO4 + cloud\text{-}albedo\ term$

662                                $+residual\_cld,$

663                                $= A*N + cloud\text{-}albedo\ term + residual\_cld,$ (A4)

664

665   where N is the parameter defined above. Therefore, the first term on the right-hand side

666   of equation (A4), A*N, corresponds to the impact of cloud amount changes on the cloud

667   radiation; and the cloud-albedo term can be obtained as a residual after subtracting A*N

668   from ΔOSRcld_p/ΔloadSO4, thereby eliminating any linear dependence of the cloudy-

669   sky shortwave flux response on cloud-amount changes.

670   As with the clear-sky decomposition, *residual_cld* is a possible non-linear term and

671   is assumed to be small. This term cannot in fact be distinguished from the cloud-albedo

672   term, in this analysis: we must therefore accept that cloud-albedo changes could be

673   accompanied by non-linear changes in macroscopic cloud properties (in this

674   framework).

675   The total aerosol-forcing-sensitivity can be measured by substituting the derived

676   values of $\Delta OSR/\Delta loadSO4$ from both the clear sky (equation A3) and cloudy

677   (equation A4) parts back into equation (A1):

678    $\Delta OSR/\Delta loadSO4$ $= (1\text{-}CLT\_hist/100.)*M - OSRclr\_hist/100*N$

679                                $+A*N + cloud\_albedo\_term + residual$

680                                $= (1\text{-}CLT\_hist/100)*M + (A - OSRclr\_hist/100.)*N$

681                                $+ cloud\_albedo\_term + residual\_osr.$ (A5)

682

683   Based on equation (A5), the total aerosol-forcing-sensitivity can therefore be

684   decomposed to the aerosol-radiation interaction term (ARI), (1-CLT_hist/100.)*M,

685   cloud-amount term as (A - OSRclr_hist/100.)*N including the impacts of cloud amount

686   changes on aerosol radiation (-OSRclr_hist/100.*N) and cloud radiation (A*N), and

687   cloud-albedo term (defined as a residual).

688

689

690 **Data Availability.** All the model data can be freely downloaded from the Earth System
691 Federation Grid (ESGF) nodes (https://esgf-node.llnl.gov/search/cmip6/). The global
692 historical surface temperature anomalies HadCRUT5 dataset is freely available on
693 https://www.metoffice.gov.uk/hadobs/hadcrut5/data/current/download.html.

694

695 **Author contributions**
696 The main ideas were developed by JZ, KF, STT, JPM, and TW. JZ, KF, and STT wrote
697 the original draft, and the results were supervised by LJW, BBB, and DS. All the
698 authors discussed the results and contributed to the final manuscript.

699

700 **Competing interests**
701 The authors declare that they have no conflict of interest.

702

710

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