

Quantifying sources of inter-model diversity in the cloud albedo effect

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1 Auxiliary material for “Quantifying sources of 2 inter-model diversity in the cloud albedo effect”

L. J. Wilcox,¹ E. J. Highwood,² B. B. B. Booth,³ and K. S. Carslaw⁴

3 Analysis focusses on four models, where the underlying equations are known, and key
4 variables are available in the CMIP5 archive: HadGEM2-ES, CSIRO-Mk3.6.0, IPSL-
5 CM5A-LR, and NorESM1-M. In each of these models, the first indirect effect is repre-
6 sented by an equation for cloud droplet effective radius in terms of cloud droplet number
7 concentration. Cloud droplet number concentration (N_d) is a function of either aerosol

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8 mass concentration, or aerosol number concentration. Both relationships introduce a
9 source of inter-model diversity. Analysis of the influence of the N_d calculation has been
10 the topic of many previous studies, some of which analyse the underlying equations of
11 the models we consider here [Storelvmo *et al.*, 2009]. We focus on the influence of the
12 relationship between N_d and cloud droplet effective radius.

1. Full-model and simple functional forms

13 We aim to create a simple functional form with which to test the sensitivity of the full
14 climate model to perturbations of various parameters. We assume that sulfate accounts
15 for most of the changes in effective radius over the industrial era, even though other
16 aerosol species can act as CCN. We find a linear correlation between global-mean annual-
17 mean vertically integrated sulfate load and N_d ($r^2 \geq 0.98$ for HadGEM2-ES, CSIRO-
18 Mk3.6.0, and NorESM1-M; data was not available for IPSL-CM5A-LR), which allows the
19 substitution of N_d for sulfate load in our analysis. Linear regression is then used to create a

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20 simple equation for effective radius in terms of sulfate load, where sulfate load and effective
21 radius have the same power relationship as the aerosol-dependent term and effective radius
22 in the full model. Constants from the regression analysis are regionally dependent due to
23 different regional trends in cloud liquid water path. Our simple equations underestimate
24 the interannual variability in effective radius relative to the full model output as they do
25 not account for inter-annual variability in liquid water path.

26 The four models, their underlying equations, and the simple equations based on these
27 are introduced below.

1.1. HadGEM2-ES

28 The Met Office Hadley Centre Global Environment Model 2 - Earth System, HadGEM2-
29 ES, is a coupled AOGCM with an atmospheric resolution of N96 ($1.875^\circ \times 1.875^\circ$), and 38
30 vertical levels. It includes an interactive tropospheric chemistry scheme, interactive land
31 and ocean carbon cycles, and dynamic vegetation [Jones *et al.*, 2011]. Seven aerosol species
32 are represented in HadGEM2-ES: sulfate, fossil-fuel black and organic carbon, sea salt,
33 mineral dust, biomass burning and biogenic aerosols [Collins *et al.*, 2008]. HadGEM2-ES
34 accounts for both anthropogenic sources of sulphur, and natural sulphur from DMS and
35 continuously degassing volcanoes.

36 *Bellouin et al.* [2007] give details of the updates to the aerosol scheme between
37 HadGEM1 and HadGEM2, and their effects. Key changes introduced in HadGEM2 in-
38 clude improvements to the sulfate and biomass burning schemes, and the representation
39 of new aerosol species: mineral dust and secondary organic aerosol.

40 The number concentration of hydrophilic aerosols is given by:

$$A = A_{SO_4} + A_f + A_j \quad (1)$$

$$A_{SO_4} = 5.125 \times 10^{17} \cdot m \quad (2)$$

$$A_f = \begin{cases} 3.856 \times 10^6 (1 - e^{-0.736u}), & 0 \text{ m s}^{-1} \leq u \leq 2 \text{ m s}^{-1} \\ 10^{(0.095u+6.283)}, & 2 \text{ m s}^{-1} \leq u \leq 17.5 \text{ m s}^{-1} \\ 1.5 \times 10^8 (1 - 97.87e^{-0.313u}), & u > 17.5 \text{ m s}^{-1} \end{cases} \quad (3)$$

$$A_j = \begin{cases} 0.671 \times 10^6 (1 - e^{-1.351u}), & 0 \text{ m s}^{-1} \leq u \leq 2 \text{ m s}^{-1} \\ 10^{(0.0422u+5.7122)}, & 2 \text{ m s}^{-1} \leq u \leq 17.5 \text{ m s}^{-1} \\ 3.6 \times 10^6 (1 - 103.926e^{-0.353u}), & u > 17.5 \text{ m s}^{-1} \end{cases} \quad (4)$$

41 where A is the aerosol number concentration, A_{SO_4} is the number concentration of am-
 42 monium sulfate particles, A_f and A_j are the number concentrations of sea salt aerosol
 43 particles originating from film and jet droplets respectively, m is the total mass concen-
 44 tration of aerosol sulfur, and u is the 10m wind speed [Jones *et al.*, 2001].

45 The aerosol number concentration is used to find the cloud droplet number concentra-
 46 tion, N_d :

$$N_d = \max\{3.75 \times 10^8 (1 - e^{-2.5 \times 10^{-9} A}), N_{min}\} \quad (5)$$

$$N_{min} = \begin{cases} 3.5 \times 10^7 & \text{over ice-free land} \\ 5 \times 10^6 & \text{otherwise} \end{cases} \quad (6)$$

Effective radius is then found from:

$$r_e = \left(\frac{3q_c \rho_0}{4\pi \rho_w k N_d} \right)^{\frac{1}{3}} \quad (7)$$

where r_e is the cloud droplet effective radius, q_c is the cloud liquid water content, ρ_0 and ρ_w are the densities of air and water respectively, and k is a constant whose values depend on whether the clouds are over land or sea in the model [Jones *et al.*, 2001].

$$k = \begin{cases} 0.67, & \text{continental} \\ 0.80, & \text{marine} \end{cases} \quad (8)$$

47 Following this, the HadGEM2-ES simple equation has the form:

$$r_e = a + b.load^{-0.33} \quad (9)$$

48 where the global and regional values of constants a and b are found by linear least squares
49 regression of global- and regional-mean time series of $load^{-0.33}$ onto global- and regional-
50 mean time series of r_e . The global and regional values of the constants a and b are shown
51 in Table 2.

1.2. CSIRO-Mk3.6.0

52 The Commonwealth Scientific and Industrial Research Organisation model 3.6, CSIRO-
53 Mk3.6.0, is a coupled AOGCM with dynamical sea ice and soil canopy schemes. The
54 atmosphere has a horizontal resolution of T63 ($\approx 1.875^\circ \times 1.875^\circ$), and 18 vertical levels.
55 The main difference between CSIRO-Mk3.6.0 and Mk3.5 is the inclusion of an interactive
56 aerosol scheme. This explicitly treats sulfate, carbonaceous aerosol, dust, and sea salt.
57 Mk3.6 also includes an updated radiation scheme, and other changes to the atmospheric
58 physics component [Syktus *et al.*, 2011].

59 Prescribed anthropogenic and biomass burning sources of sulfur, black carbon, and
60 organic aerosol are based on Lamarque *et al.* [2010], but with emissions of black carbon
61 and organic aerosol uniformly increased by 25% and 50% respectively in order to improve
62 the agreement between modelled and observed carbonaceous aerosol. Natural sources of
63 sulfur are continuously degassing volcanoes, and biogenic emissions of DMS [Rotstayn
64 *et al.*, 2012].

65 Rotstayn *et al.* [2012] note that CSIRO-Mk3.6.0 burdens of sulfate, organic aerosol, and
66 dust in 2000 are close to the top of their reference range. The relatively large sulfate

67 burdens can be seen in Supplementary Figures 3 and 4. *Rotstayn et al.* [2012] find that
 68 large DMS emissions are a contributing factor to the relatively large sulfate burden in the
 69 model.

70 The number concentration of hydrophilic aerosol is given by:

$$A = A_S + A_{SS} + A_C \quad (10)$$

$$A_S = 5.1 \times 10^{17} m_S \quad (11)$$

$$A_C = 3.0 \times 10^{17} m_C \quad (12)$$

71 where A is the number concentration of hydrophilic aerosols, A_S is the sulfate concentra-
 72 tion, A_{SS} is the sea salt concentration, A_C is the concentration of hydrophilic carbona-
 73 ceous aerosol, and m_S and m_C are the mass concentrations of sulfate and hydrophilic
 74 carbonaceous aerosol respectively. A_{SS} is provided directly by the windspeed-dependent
 75 diagnostic for sea salt [*Rotstayn et al.*, 2012].

76 Cloud droplet number concentration, N_d , is given by:

$$N_d = \max\{3.75 \times 10^8 (1 - e^{-2.5 \times 10^{-9} A}), N_{min}\} \quad (13)$$

$$N_{min} = 10 \times 10^6 \quad (14)$$

77 The calculation of cloud droplet effective radius includes a parameterization of increased
 78 droplet dispersion with increased cloud droplet number concentration, such that:

$$r_e = 0.07 r_v \left(\frac{L}{N} \right)^{-1.14} \quad (15)$$

$$r_v = \left(\frac{3L\rho_0}{4\pi\rho_w N_d} \right)^{\frac{1}{3}} \quad (16)$$

$$r_e = 0.07 \left(\frac{3\rho_0}{4\pi\rho_w} \right)^{\frac{1}{3}} \left(\frac{L}{N_d} \right)^{0.19} \quad (17)$$

79 where L is the cloud liquid water content, r_v is the volume-averaged mean droplet radius,
 80 and ρ_0 and ρ_w are the densities of air and water respectively [Rotstayn *et al.*, 2012].

81 Following this, the CSIRO-Mk3.6.0 simple equation has the form:

$$r_e = a + b.load^{-0.19} \quad (18)$$

82 where the global and regional values of constants a and b are found by linear least squares
 83 regression of global- and regional-mean time series of $load^{-0.19}$ onto global- and regional-
 84 mean time series of r_e . The global and regional values of the constants a and b are shown
 85 in Table 2.

1.3. IPSL-CM5A-LR

86 The Institut Pierre Simon Laplace Climate Model 5A (low resolution), IPSL-CM5A-
 87 LR, is an AOGCM with an interactive carbon cycle, representation of tropospheric and
 88 stratospheric chemistry, and a comprehensive representation of aerosol processes [Dufresne
 89 *et al.*, 2013]. It has a horizontal resolution of $3.75^\circ \times 1.875^\circ$, and 39 vertical levels. IPSL-
 90 CM5A-LR treats sulfate, black carbon, particulate organic matter, sea salt, and dust. The
 91 model represents a substantial improvement over its predecessor, which only considered
 92 sulfate aerosol [Dufresne *et al.*, 2013].

The total mass of soluble aerosol is given by:

$$m_{soluble} = m_{SO_4} + m_{BC,soluble} + m_{POM,soluble} \quad (19)$$

93 where m_{SO_4} , $m_{BC,soluble}$, and $m_{POM,soluble}$ are the masses of sulfate, soluble black carbon,
 94 and soluble particulate organic matter respectively [Szopa *et al.*, 2012].

Cloud droplet number concentration, N_d , is given by:

$$N_d = 10^{1.7+0.2\log(m_{soluble})} \quad (20)$$

Effective radius is then found from:

$$r_e = 1.1 \left(\frac{L\rho_{air}}{\frac{4}{3}\pi\rho_{water}N_d} \right)^{\frac{1}{3}} \quad (21)$$

where r_e is the cloud droplet effective radius, L is the cloud liquid water content, and ρ_{air} and ρ_{water} are the densities of air and water respectively [Boucher and Lohmann, 1995].

The IPSL-CM5A-LR simple equation has the form:

$$r_e = a + b.load^{-0.33} \quad (22)$$

where the global and regional values of constants a and b are found by linear least squares regression of global- and regional-mean time series of $load^{-0.33}$ onto global- and regional-mean time series of r_e . The global and regional values of the constants a and b are shown in Table 2.

1.4. NorESM1-M

The Norwegian Earth System Model, NorESM1-M, is based on CCSM4 (Community Climate System Model version 4). It's atmospheric component is CAM4-Oslo, a modified version of CAM4 (Community Atmosphere Model 4), which includes advanced chemistry-aerosol-cloud-radiation interactions [Bentsen *et al.*, 2013]. It has a horizontal resolution of $2.5^\circ \times 1.9^\circ$, and 26 vertical levels.

NorESM1-M includes sea salt, mineral dust, particulate sulfate, black carbon, and primary and secondary organic aerosols [Kirkevåg *et al.*, 2013]. Key updates from the previous version of the model include: modified prognostic sea salt emissions; updated

109 treatment of precipitation scavenging and gravitational settling; increased abundance of
 110 organic matter relative to black carbon; and the inclusion of biogenic primary organic and
 111 methanesulfonic acid from the oceans [Kirkevåg *et al.*, 2013].

112 The activation of CCN in NorESM1-M follows the parameterization of *Abdul-Razzak*
 113 *and Ghan* [2000] [Kirkevåg *et al.*, 2013]. In an update over the previous version, cloud
 114 droplet spectral dispersion is represented, so that:

$$r_e = \beta r_v \quad (23)$$

$$r_v = \left(\frac{3L\rho_0}{4\pi\rho_w N_d} \right)^{\frac{1}{3}} \quad (24)$$

$$\beta = \frac{(1 + 2\epsilon^2)^{\frac{2}{3}}}{(1 + \epsilon^2)^{\frac{1}{3}}} \quad (25)$$

$$\epsilon = 1 - 0.7e^{-0.003N_d} \quad (26)$$

$$r_e = \left(\frac{3L\rho_{air}}{4\pi\rho_w N_d} \right)^{\frac{1}{3}} \frac{(1 + 2\epsilon^2)^{\frac{2}{3}}}{(1 + \epsilon^2)^{\frac{1}{3}}} \quad (27)$$

115 Following this, the NorESM1-M simple equation is more complex than that for the
 116 other models we consider:

$$r_e = a + b.load^{-0.33} \frac{(1 + 2(1 - 0.7e^{3000load})^2)^{0.66}}{(1 + (1 - 0.7e^{3000load})^2)^{0.33}} \quad (28)$$

where the global and regional values of constants a and b are found by linear least squares
 regression of global- and regional-mean time series of:

$$load^{-0.33} \frac{(1 + 2(1 - 0.7e^{3000load})^2)^{0.66}}{(1 + (1 - 0.7e^{3000load})^2)^{0.33}} \quad (29)$$

117 onto global- and regional-mean time series of r_e . The global and regional values of the
 118 constants a and b are shown in Table 2.

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Table 1. CMIP5 models used in this study.

Institute	Model	1 st	2 nd	Ant	Reference
		Indirect	Indirect		
CCCma	CanESM2	Y	N	E1	<i>von Salzen et al.</i> [2013]
CNRM-CERFACS	CNRM-CM5	Y	N	E1	<i>Szopa et al.</i> [2012] <i>Voldoire et al.</i> [2012]
CSIRO-QCCCE	CSIRO-Mk3.6.0	Y	Y	E1a	<i>Rotstayn et al.</i> [2012]
NOAA GFDL	GFDL-CM3	Y	Y	E1	<i>Donner et al.</i> [2011] <i>Levy et al.</i> [2013]
MOHC	HadGEM2-CC	Y	Y	E1	<i>Bellouin et al.</i> [2007] <i>Collins et al.</i> [2011]
MOHC	HadGEM2-ES	Y	Y	E1	<i>Bellouin et al.</i> [2007] <i>Collins et al.</i> [2011]
IPSL	IPSL-CM5A-LR	Y	N	E1	<i>Dufresne et al.</i> [2013]
IPSL	IPSL-CM5A-MR	Y	N	E1	<i>Dufresne et al.</i> [2013]
NCC	NorESM1-M	Y	Y	E1	<i>Iversen et al.</i> [2012]
MIROC	MIROC5	Y	Y	E1	<i>Watanabe et al.</i> [2010]
MIROC	MIROC-ESM	Y	Y	E1	<i>Watanabe et al.</i> [2011]
MIROC	MIROC-ESM-CHEM	Y	Y	E1	<i>Watanabe et al.</i> [2011]
MRI	MRI-CGCM3	Y	N	E1	<i>Yukimoto et al.</i> [2012] Pers. Comm., S. Yukimoto [2013]

Table 2. Values of the constants in the simple equations for cloud droplet effective radius in terms of vertically integrated sulfate load.

	HadGEM2-ES		CSIRO-Mk3.6.0		IPSL-CM5A-LR		NorESM1-M	
	a($\times 10^{-6}$)	b($\times 10^{-8}$)	a($\times 10^{-6}$)	b($\times 10^{-7}$)	a($\times 10^{-7}$)	b($\times 10^{-9}$)	a($\times 10^{-6}$)	b($\times 10^{-8}$)
Globe	9.24	2.73	8.11	2.32	21.6	4.70	10.1	1.12
Europe	5.15	5.70	6.96	2.62	7.86	2.28	9.01	3.48
N. Atlantic	7.66	4.14	7.96	2.00	28.8	10.1	10.4	1.24
China	6.28	3.85	6.68	3.15	8.80	4.41	8.62	3.82
US	6.57	2.28	8.86	0.45	6.04	1.93	10.1	1.49

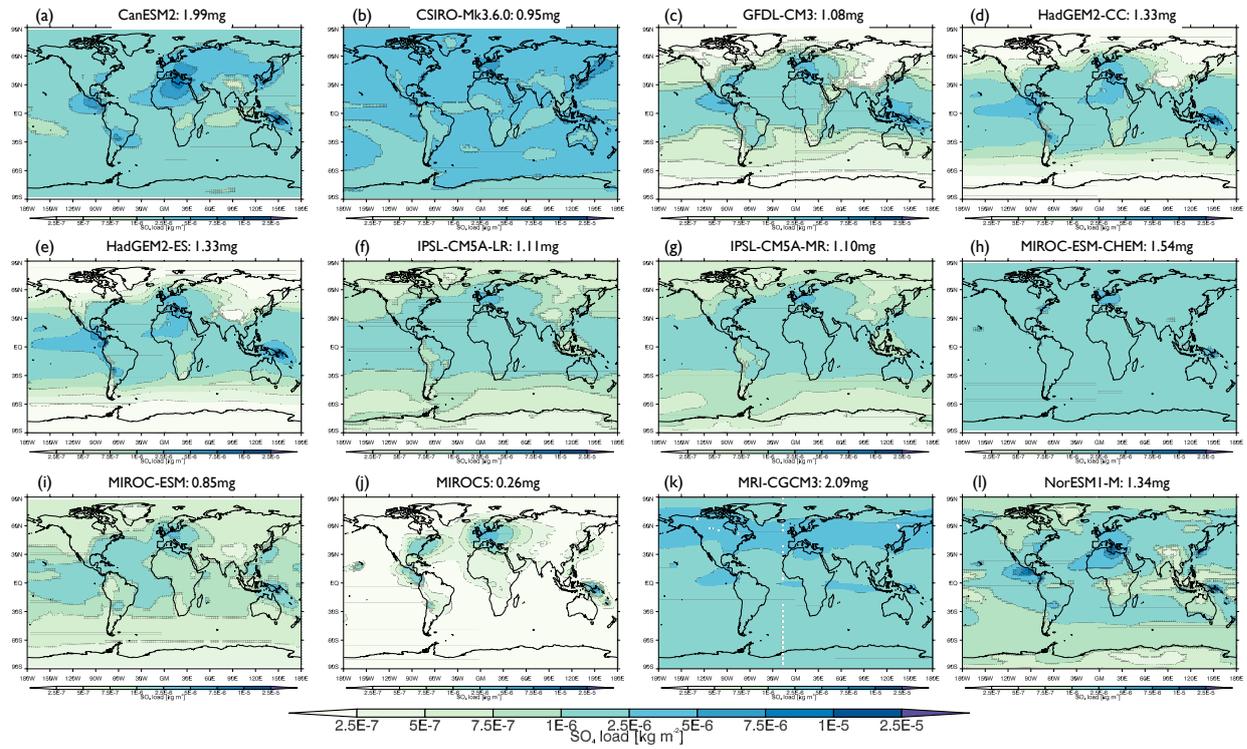


Figure 1. 1860-1900 column total sulfate load for (a): CanESM2, (b): CSIRO-Mk3.6.0, (c): GFDL-CM3, (d): HadGEM2-CC, (e): HadGEM2-ES, (f): IPSL-CM5A-LR, (g): IPSL-CM5A-MR, (i): MIROC-ESM-CHEM, (j): MIROC-ESM, (k): MIROC5, (l): MRI-CGCM3, (m): NorESM1-M.

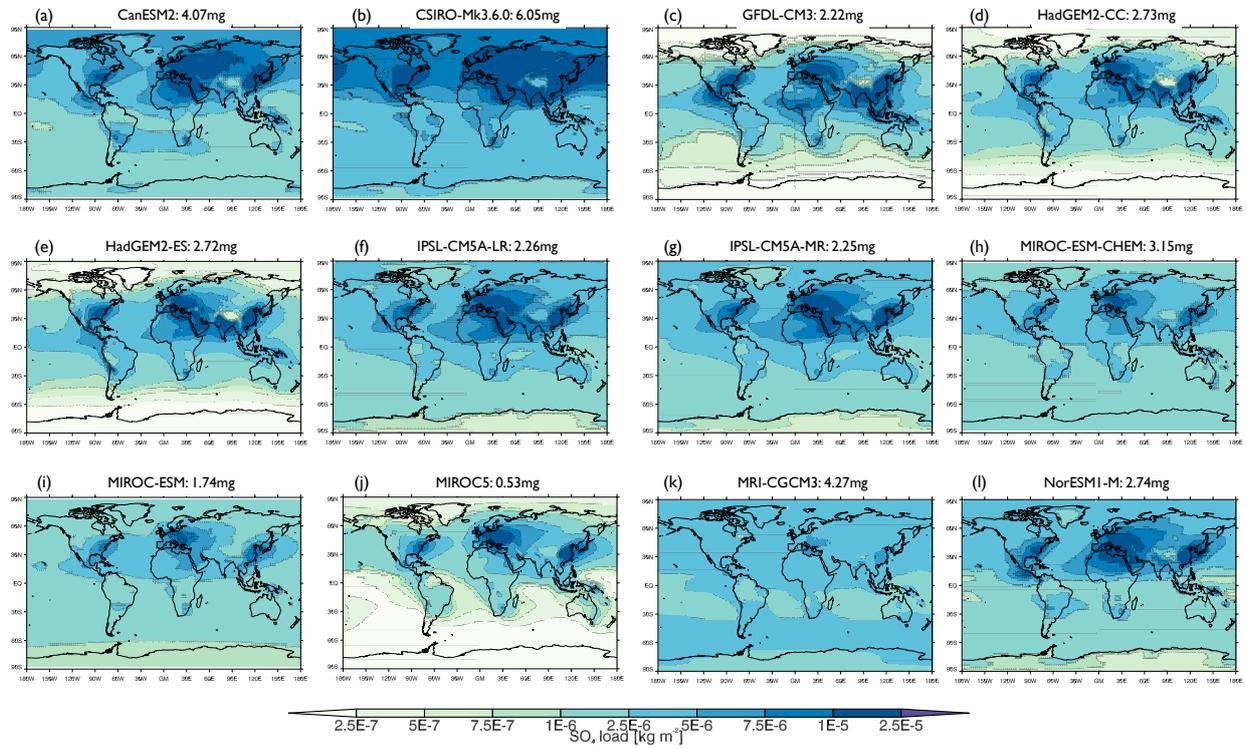


Figure 2. 1986-2005 mean column total sulfate load for (a): CanESM2, (b): CSIRO-Mk3.6.0, (c): GFDL-CM3, (d): HadGEM2-CC, (e): HadGEM2-ES, (f): IPSL-CM5A-LR, (g): IPSL-CM5A-MR, (i): MIROC-ESM-CHEM, (j): MIROC-ESM, (k): MIROC5, (l): MRI-CGCM3, (m): NorESM1-M.

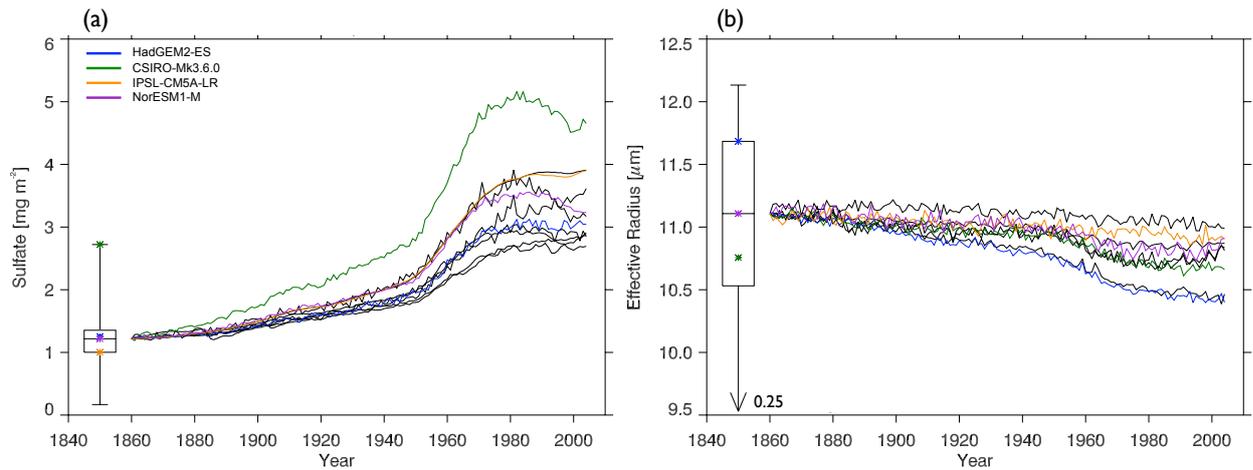


Figure 3. Annual-mean global-mean (a): sulfate load and (b): cloud-top effective radius for CMIP5 models. Time series are adjusted to the 1860 CMIP5 median value. The box and whisker shows median, interquartile range, and absolute range. Colours pick out the models focussed on in this study. Crosses show the location of these models within the CMIP5 range.

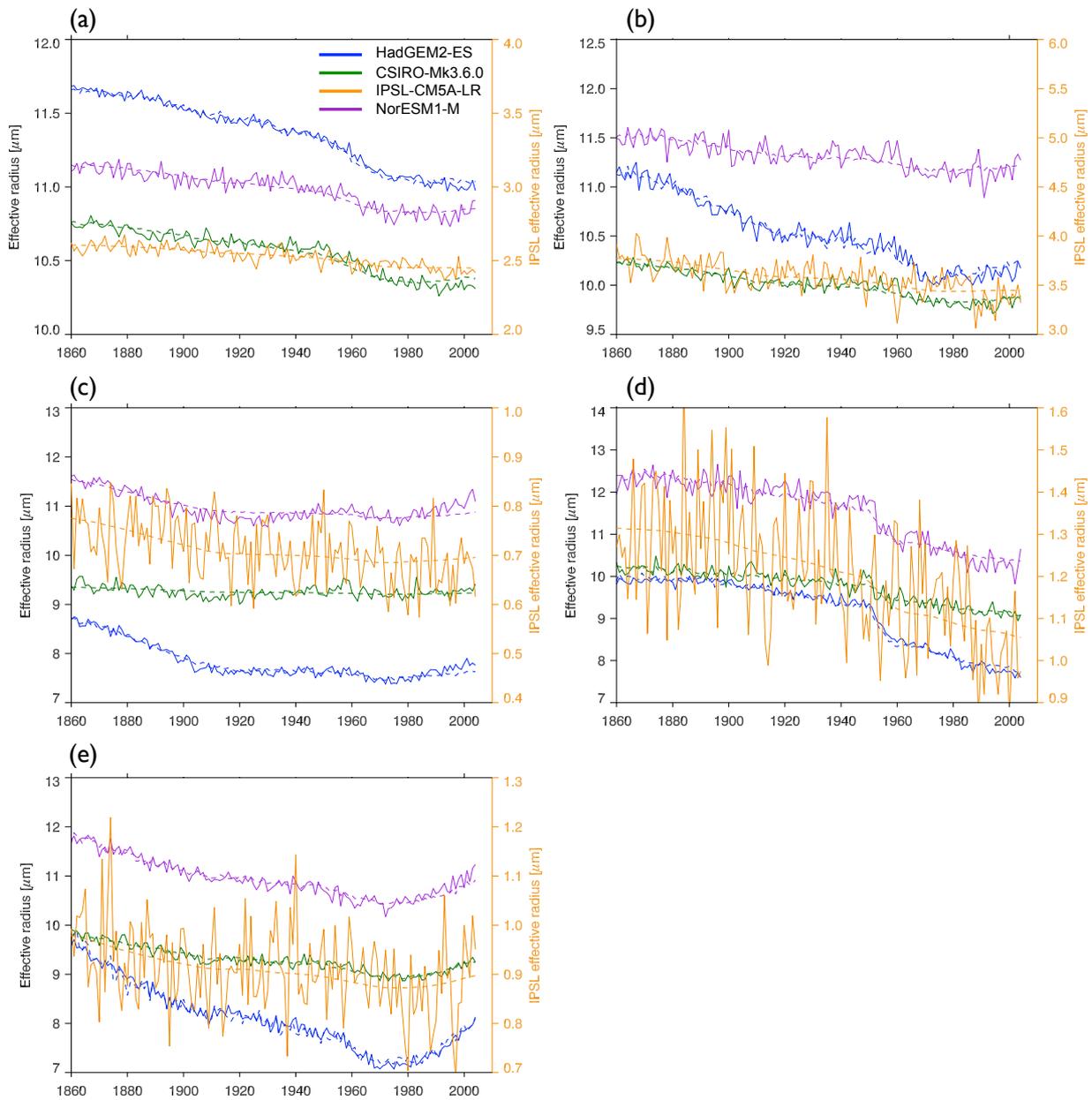


Figure 4. Annual-mean cloud-top effective radius output from CMIP5 models (solid lines), and produced using simplified equations in terms of sulfate load (dotted lines) for (a): Global, (b): China, (c): Europe, (d): North Atlantic, (e): continental United States mean. Note that all results for IPSL-CM5A-LR are shown on a separate axis.

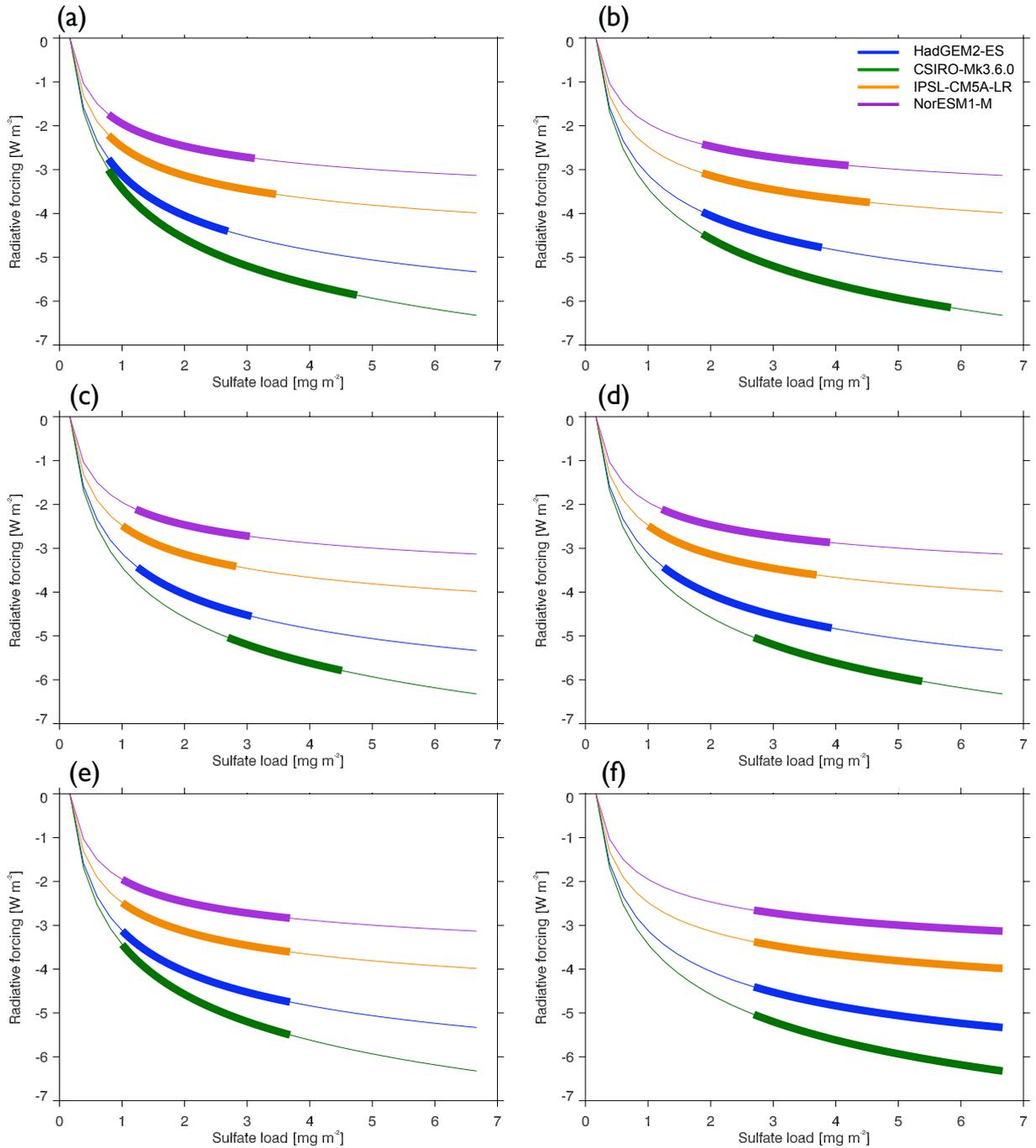


Figure 5. Schematic showing the perturbations to sulfate load made in the sensitivity experiments, and their impact on radiative forcing. Thin lines use the functional forms to show cloud albedo for the whole CMIP5 range of global mean sulfate load. Thick lines highlight the sulfate loads used in each model in each experiment. (a): Minimum pre-industrial load, (b): maximum pre-industrial load, (c): minimum load change, (d): maximum load change, (e): IPSL-CM5A-LR