

## Supplemental Material

**Table S1** Sources of non-BB aerosol in the models

<b>MODEL</b>	<b>Anthropogenic emissions</b>	<b>Dust</b>	<b>Volcanic</b>	<b>Sea salt</b>	<b>Default model emission injection height</b>
<b>CAM5</b>	SO <sub>2</sub> , BC, and OC from fossil fuel and biofuel combustion: AeroCom2-ACCMIP (Lamarque et al., 2010)	Calculated using friction velocity, soil erodibility, soil moisture.	Non-eruptive volcanic SO <sub>2</sub> emissions from AeroCOM (Dentener et al., 2006)	Calculated using 10-m wind speed, sea surface temperature.	Anthrop. emissions: model surface layer. Energy and industry: 100-300 m above surface. BB: evenly distributed from 0 to 1 km.
<b>ECMWF-IFS (CY45R1)</b>	SO <sub>2</sub> , OC and BC from non BB source, from MACCity BB BC and OC: GFAS (Kaiser et al., 2012)	Calculated using surface topography, surface bareness, 10 m wind speed, and ground wetness (Ginoux et al., 2001). Updated with the use of wind gusts.	none	Calculated using 10-m wind speed (Monahan & O’Muircheartaigh, 1986; Morcrette et al., 2009)	BB: Daily injection heights computed by a Plume Rise Model (Paugam et al., 2015).
<b>ECHAM6-SALSA (6.1)</b>	SO <sub>2</sub> , BC, and OC from fossil fuel and biofuel combustion: AeroCom2-ACCMIP (Lamarque et al., 2010)	Dust flux calculated on-line using 10 m wind speeds, soil clay content and soil moisture from ECHAM6 (Bergman et al., 2012; Tegen et al., 2002)	Emissions from volcanic sources are based on GEIA inventory (Andres & Kasgnoc, 1998)	Calculated using 10-m wind speed (Bergman et al., 2012)	Anthrop. emissions: model surface layer. Energy and industry: 100-300 m above surface. BB: prescribed ecosystem-specific emission profiles from 0 to 6 km. Volcanic: estimated from magnitude of eruption.
<b>GEOS</b>	SO <sub>2</sub> , NH <sub>3</sub> , BC, and OC from fossil fuel and biofuel combustion: HTAP2 dataset	5-bins calculated as a function of surface topography, surface bareness, 10 m wind	Global volcanic SO <sub>2</sub> emission inventory by Carn et al. (2017)	5-bins calculated based on met fields	BB emission: evenly distributed within the boundary layer. The rest (du, ss, anthropogenic: oc, bc, su) are injected in the lowest model layer.

		speed, and ground wetness (Ginoux et al., 2001)			Volcanic SO <sub>2</sub> is distributed evenly within a layer defined in Carn's dataset (2017).
<b>GEOS-CHEM (v9-02)</b>	CO, NO <sub>x</sub> , and SO <sub>2</sub> from EDGAR v3.2-FT2000 (Olivier et al., 2005) NMVOCs: RETRO monthly global inventory for 2000 implemented after (Hu et al., 2015); (Xiao et al., 2008); (Yevich & Logan, 2003); Global anthrop. BC/OC: (Bond et al., 2007) GEIA ammonia emissions (Bouwman et al., 1997) the US NEI05, Canada (CAC), Mexico (BRAVO), Europe (EMEP), East Asian emissions from (Streets et al., 2006) and (Q. Zhang et al., 2009) <b>Aircraft emissions</b> BC/OC, sulfur emissions calculated from fuel parameters (Stettler et al., 2011)	Fairlie et al. (2007)	Eruptive and non-eruptive volcanic SO <sub>2</sub> emissions for individual years are from the AEROCOM, (Fisher et al., 2011)	Jaegle et al. (2011)	Species are emitted in the lowest model level, and mixed homogeneously up to the mixing height recalculated in the model.
<b>GISS ModelE OMA</b>	Emissions are taken from Lamarque et al. (2010)	Prognostic (friction velocity, soil erodibility, soil moisture)	Stratospheric AOD prescribed, continous SO <sub>2</sub> AeroCom	Prognostic (10-m wind speed, sea surface temperature)	Anthropogenic emissions from agriculture, domestic, transportation, waster, and shipping sectors are distributed to the model surface layer while those from energy and industry

					sectors are emitted at 100-300 m above surface ; fire emissions are evenly distributed in the boundary layer.
<b>GISS ModelE MATRIX</b>	Emissions are taken from Lamarque et al. (2010)	Prognostic (friction velocity, soil erodibility, soil moisture)	Stratospheric AOD prescribed, continous SO <sub>2</sub> AeroCom	Prognostic (10-m wind speed, sea surface temperature)	Anthropogenic emissions from agriculture, domestic, transportation, waster, and shipping sectors are distributed to the model surface layer while those from energy and industry sectors are emitted at 100-300 m above surface ; fire emissions are evenly distributed in the boundary layer.
<b>GOCART (5 rev. 32)</b>	SO <sub>2</sub> , BC, and OC from fossil fuel and biofuel combustion: A2-ACCMIP dataset (Diehl et al., 2012)	Calculated using surface topography, surface bareness, 10-m wind speed, and ground wetness (Ginoux et al., 2001) Update including NDVI (Kim et al., 2013)	Volcanic SO <sub>2</sub> from (A2-MAP), + continuous volcanoes (Diehl et al., 2012)	Calculated based on met fields (Reference?)	BB: evenly distributed within the BL. Volcanic: estimated from magnitude of eruption and volcanic SO <sub>2</sub> index. The rest: injected in the lowest model layer.
<b>OsloCTM2</b>	ECLIPSE V5 emissions for year 2010 (Stohl et al., 2015)	Interactive, (Grini et al., 2005)	Volcanic emissions from AEROCOM (Frank Dentener et al., 2006)	Interactive, (Grini et al., 2005)	Anthropogenic: distributed in the four lowest model layers dependent on emission sectors.
<b>SPRINTARS</b>	SO <sub>2</sub> , BC, and OC from fossil fuel and biofuel combustion: HTAP2.	See Appendix A in (T. Takemura et al., 2009)	(Andres & Kasgnoc, 1998)	See Appendix B in (T. Takemura et al., 2009)	BB emission are evenly distributed within the sigma level larger than 0.74. The rest: lowest model layer.

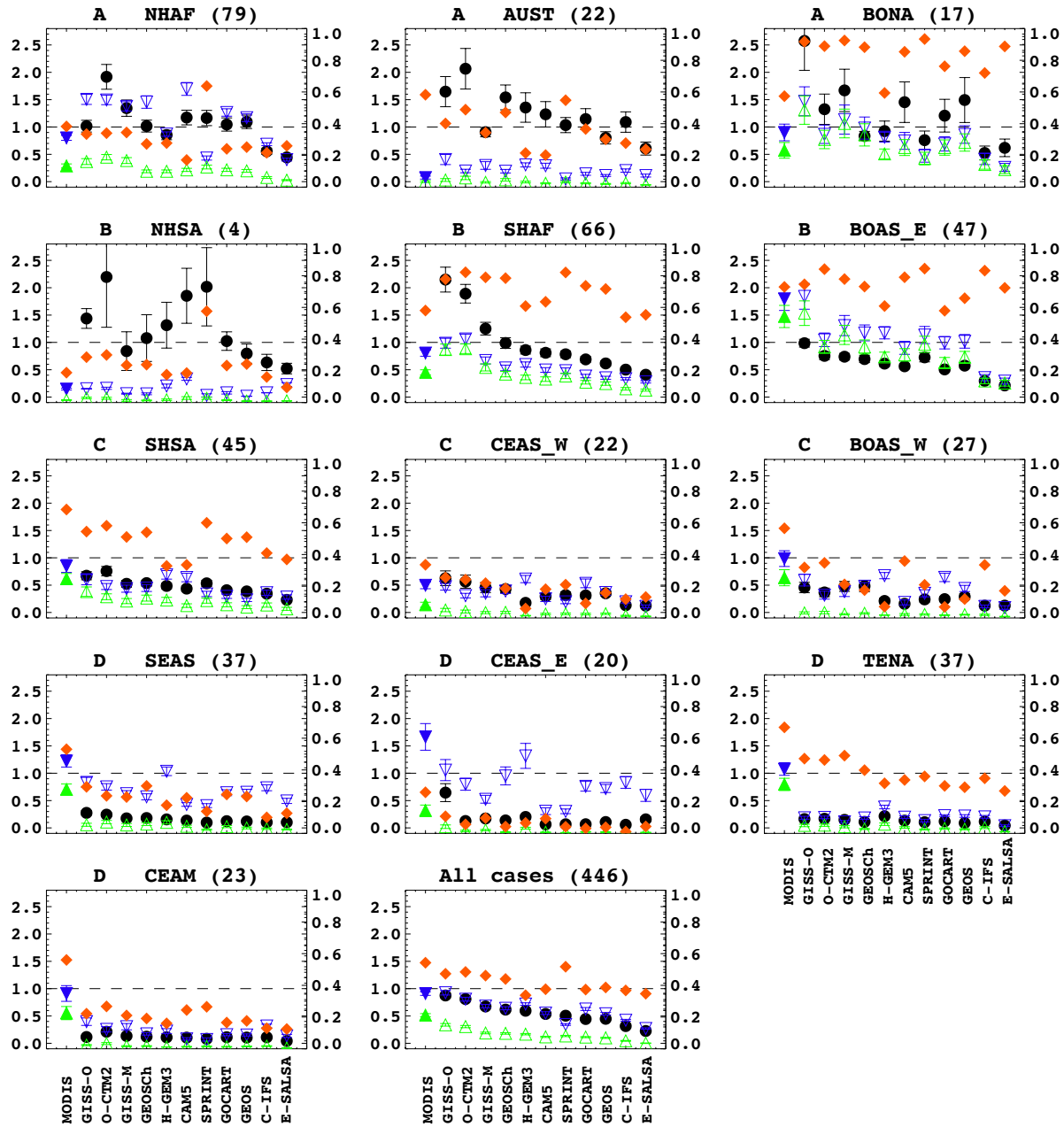
**Table S2** Aerosol properties in the models

Model	Aerosol species considered in the model	Aerosol mixing state	Size distributions (size bins if int. mixed)	Aerosol mass extinction efficiency at 550 nm	Aerosol refractive index at 550 nm	Hygroscopicity assumptions for BB-related species	BB-related aerosol aging assumptions	OM/OC conversion factor
<b>CAM5</b>	du, ss, BC, primary organic matter (POM), secondary organic aerosol (SOA and precursor gases), sulfate (and its gaseous precursors SO <sub>2</sub> and DMS)	Internal in the same mode, external between different modes; carbonaceous aerosols are all in the accumulation mode for the three-mode standard representation.	Log-normal size distributions for 3 aerosol modes with the prescribed standard deviation and dry diameter size range for each mode can be found in Table 1 of (Liu et al., 2012)	Wet RI in each mode is calculated based on the volume-weighted means of individual aerosol species. The wet surface mode radius of particles in the mode is calculated based on the Köhler theory. These two parameters are used to parameterize the optical properties of particles in the mode based on (Ghan & Zaveri, 2007).	(1.95,0.79i) for BC; OPAC (Hess et al., 1998) for other species.	0 for BC and 0.1 for POM; SO <sub>4</sub> =0.507	No aging assumptions applied; upon emission BC and POM are internally mixed with more hygroscopic species in the same accumulation mode, and thus subject to wet scavenging.	1.4
<b>ECMWF-IFS (CY45 R1)</b>	du, ss, sulfate, OM, BC	External	See Remy et al. (2019)	See Bozzo et al. (2020)	See Bozzo et al. (2020)	Considered only for optical properties (Remy et al. 2019)	original 80% of BC and 50% of OC are hydrophilic with e-folding time of 1.16 days	1.5 (for this study)
<b>ECHA M6-</b>	du, ss, OC, BC, sulfate (and its	External	See Figure 1 in	Look-up-tables	Volume weighted	Hygroscopicity: ZSR	no ageing	1.4

SALSA (6.1)	gaseous precursors SO <sub>2</sub> and DMS)		Kokkola et al. (2018)	based on mie calculations for the extinction cross section, asymmetry factor, and single scattering albedo as a function of size parameter and refractive index.	means of individual aerosol species (refractive indices are given by Bergman et al. (2012), Table 5.	method (Jacobson, 2005). BC completely insoluble, 15% of OC is soluble taking as an ideal solute, SO <sub>4</sub> hygroscopicity is according to Jacobson (2005).		
GEOS5	du, ss, OC, BC, sulfate, nitrate, and ammonium	External	See table 2 from Chin et al. (2009) except a new sea salt bin (0.03-0.1 um in radius). Nitrate has 3-bins, mean radius 0.27, 2.1, 7.6 um.	See Fig. 2 in Chin et al, 2002	Table 2 from Chin et al. (2009)	Hygroscopic growth factors - see table 3 from Chin et al. (2002)	80% of BC and 50% of OC are hydrophilic with e-folding time of 2.5 days	1.4
GEOS-CHEM (v9-02)	du, ss, OC, BC, sulfate, nitrates, ammonium, MSA	External	Bulk mass aerosol model, except dust and sea salt. Dust: 4 bins, radius 0.1-1; 1-2; 2-3; 3-6 um. Sea Salt: fine (0.01-0.5 um, radius) and coarse (0.5-8.0 um, radius).	From GADS/OPAC. Data in m <sup>2</sup> /g: sulfate, ammonium and nitrate 2.2; BC 8.0; OC 2.8; SS fine 2.4; SS coarse 0.9; dust from 3.1 to 0.16 from finer to coarser bin	From GADS/OPAC. Sulfate-ammonium-nitrate: 1.43 + 10 <sup>-8</sup> i; BC 1.75+0.45 i; OC 1.53+0.006 i; SS 1.5 + 10 <sup>-8</sup> i; dust 1.56 + 0.0014 i	Hygroscopicity factors are assigned in 7 RH bin for each species, and Mie scattering and absorption efficiencies recalculated at those bins, and the interpolated at ambient RH online.	Primary hydrophobic fraction of carbonaceous aerosol is converted to hydrophilic with an e-folding time of 1.2 days	2.1 (Turpin & Lim, 2001)
GISS ModelE OMA	du, su coated dust, nitrate coated dust, ss, BC, POM, SOA and precursor gases, sulfate (and its gaseous precursors	Externally mixed, except for sulfate	Effective radii [in um] of bulk aerosol species: SO <sub>4</sub> (0.3),		Sulfate (1.528-1.e-7i), nitrate (1.528-1.e-7i), OC (1.527-	80% solubility assumed	No aging	1.4

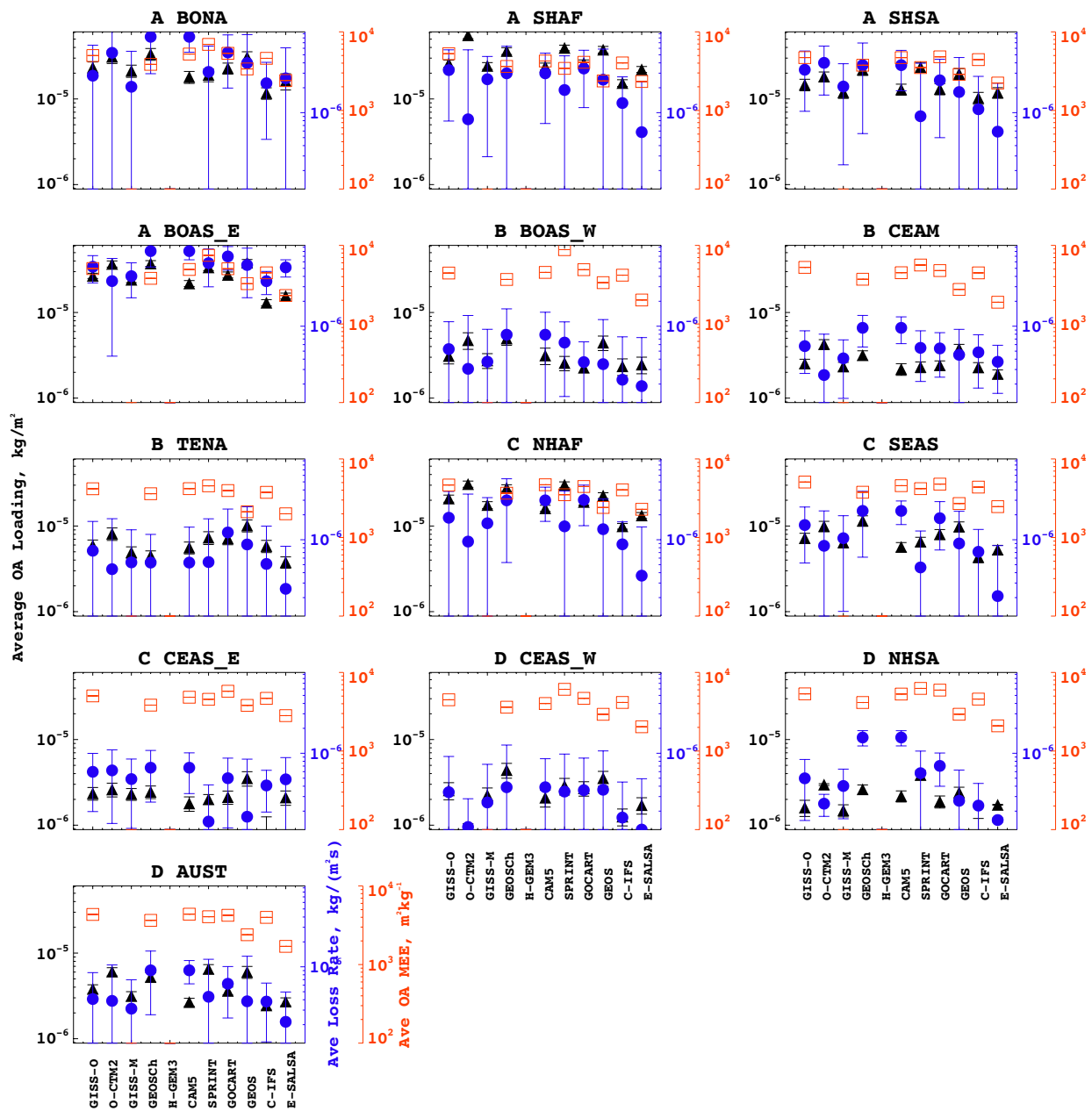
	SO2 and DMS), ammonium, nitrate, MSA	coated dust	BC (0.1), OC (0.3), NO3 (0.3), seasalt(0.44, 5.), dust (0.46, 1.47, 2.94, 5.88,11.77)		0.014i), BC (1.85-0.71i), sea salt (1.45-0.i), dust (1.564-0.002i)			
GISS ModelE MATRI X	16 mixing state classes (for definition see Bauer et al 2008) containing: sulfate,nitrate,amm inum,organics,BC, sea salt, mineral dust and aerosol water.	Prognostic mixing state	Dynamic, Size distribution s evolve with microphysical processes		Sulfate (1.528-1.e-7i), nitrate (1.528-1.e-7i), OC (1.527-0.014i), BC (1.85-0.71i), sea salt (1.45-0.i), dust (1.564-0.002i) and water (1.334-3.91e-8i)See Bauer et al 2010	Prognostic, determined by mixing state		1.4
GOCA RT (5 rev. 32)	du, ss, OC, BC, sulfate (and its gaseous precursors SO <sub>2</sub> and DMS)	External	See table 2 from (Chin et al., 2009)	See Fig. 2 in Chin et al, (2002)	Table 2 from (Chin et al., 2009)	Hygroscopic growth factors - see table 3 below (Chin et al., 2002)	80% of BC and 50% of OC are hydrophilic . with e-folding time of 2 days	1.8
<b>OsloCT M2</b>	du, ss, OC, BC, sulfate, secondary organic aerosols (SOA), nitrate	For fossil fuel and biofuel hydrophilic BC internal mixing, hydropobic BC external mixing. For BB internal mixing of BC and OC.	See Table 2 in (Myhre et al., 2007)	All values in m <sup>2</sup> /g at 30% RH: Sulfate 3.70, BC fossil fuel external mixture 9.24, OC fossil fuel 4.92, BB(BC + OC) 5.04, nitrate 3.71, sea salt and mineral dust is size dependent	See Table 2 in (Myhre et al., 2007)	Based on (Magi & Hobbs, 2003) $(1+1.31*(RH/100)^{4.88})$	80% of BC and 50% of OC are hydrophilic at the time of emissions. Aging time is dependent on season and latitude (Skeie, Berntsen, Myhre, Tanaka, et al., 2011) part a; (Skeie, Berntsen, Myhre,	1.6 for fossil and biofuel burning; 2.6 for BB

							Pedersen, et al., 2011) part b.	
SPRINTARS (5.5)	du, ss, OC, BC, sulfate (and its gaseous precursors SO <sub>2</sub> and DMS)	external except a half of BC from fuel is internally mixed with OC.	See Table A1 and B1 in Takemura et al. (2009) for dust and sea salt, respectively. The others are assumed to be the log-normal distribution (Toshihiko Takemura et al., 2002)	See (Toshihiko Takemura et al., 2002)	See (Toshihiko Takemura et al., 2002)	See (Toshihiko Takemura et al., 2002)	None	1.6 for fuel; 2.6 for BB



**Figure S3** Ratios of model BB AOD to MODIS BB AOD ( $R_{BB\_AOD}$ ) for 11 models, aggregated over each of 13 regions (black dots and left Y axes), plus ratios when aggregated over all regions in the bottom row, right. Shown on the right Y axes are the mean model and MODIS total AOD (blue triangles) and BB AOD (green triangles) for all cases in the region, and the mean BB fraction of total AOD for each model in the region (red diamonds). The group identifiers defined in Section 3.1 are given in the upper left of each panel. Note that the models are ordered in these plots based on the overall  $R_{BB\_AOD}$  from highest to lowest, i.e., all individual case averages in a region were used to calculate one arithmetic mean for each model.





**Figure S4** Plots showing the OA Mass Loading ( $\text{kg}/\text{m}^2$ ; left Y axis and filled black triangles), OA Loss Frequency ( $\text{s}^{-1}$ ; right blue Y axis and filled blue circles), and OA Mass Extinction Efficiency ( $\text{m}^2/\text{kg}$ ; right orange Y axis and open orange squares) for 10 models in each of 13 region-specific panels. Note that Loss Frequency values are missing for the CAM5 model and MEEs are lacking for GISS-M and O-CTM2.

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