

S1 GAW in-situ site details

The following 2 tables contain information about the individual GAW sites used, both for absorption and scattering coefficient measurements. The data were accessed through the EBAS database (<http://ebas.nilu.no/>).

S1.1 Absorption coefficient data

Table S1: GAW site details for in-situ dry absorption coefficient data. Start and Stop indicate first and last available months for computation of climatology in the 2005–2015 time frame. λ provides measurement wavelengths (multiple wavelengths can occur, e.g. in case instruments were changed / updated over time). Cov. indicates the percentage loss of daily average values when requiring 25% coverage in the provided hourly data. The following sites did not meet the coverage constraints for the computation of the climatology: Egbert, Finokalia.

#	Station name	Lat.	Lon.	Alt. [m]	Start	Stop	λ [nm]	Cov. [%]
1	Alert	82.50	-62.34	420	2005/01	2014/12	528; 520; 530	96
2	Anmyeon-do	36.54	126.33	56	2008/01	2014/12	520; 880	99
3	Annaberg-Buchholz	50.57	13.00	549	2012/01	2013/12	637	100
4	Aspvreten	58.80	17.38	20	2008/05	2013/12	550	99
5	Barrow	71.32	-156.61	21	2005/01	2014/12	528; 530; 520	93
6	Birkenes II	58.39	8.25	223	2008/05	2014/12	522	99
7	Bondville	40.05	-88.37	223	2005/01	2014/12	528; 530	99
8	Bösel	53.00	7.94	40	2009/01	2013/06	670	100
9	Cabauw Zijdeweg	51.97	4.93	1	2008/01	2013/12	670	100
10	Cape Point	-34.35	18.49	230	2005/11	2013/09	530	98
11	Cape San Juan	18.38	-65.62	75	2006/05	2014/11	528; 530	97
12	DEM_Athens	37.99	23.82	280	2012/01	2014/12	520	100
13	Danum Valley	4.98	117.85	427	2011/01	2012/12	670	100
14	El Arenosillo	37.10	-6.73	51	2012/07	2014/12	528	100
15	Gosan	33.29	126.16	81	2011/10	2014/12	528	99
16	Gual Pahari	28.43	77.15	320	2007/12	2010/01	670	74
17	Harwell	51.57	-1.32	137	2009/11	2010/12	370	100
18	Hohenpeissenberg	47.80	11.01	1000	2005/01	2014/12	660	100
19	Hyytiälä	61.85	24.28	185	2006/05	2014/12	520; 637; 530	99
20	Ispra	45.80	8.63	213	2005/01	2014/12	520; 670	100
21	K-puszta	46.97	19.58	135	2008/04	2014/12	528	97

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Table S1: GAW site details for in-situ dry absorption coefficient data. Start and Stop indicate first and last available months for computation of climatology in the 2005–2015 time frame. λ provides measurement wavelengths (multiple wavelengths can occur, e.g. in case instruments were changed / updated over time). Cov. indicates the percentage loss of daily average values when requiring 25% coverage in the provided hourly data. The following sites did not meet the coverage constraints for the computation of the climatology: Egbert, Finokalia.

#	Station name	Lat.	Lon.	Alt. [m]	Start	Stop	λ [nm]	Cov. [%]
22	Kosetice (NOAK)	49.57	15.08	539	2012/09	2013/12	520	99
23	Leipzig	51.35	12.43	131	2009/01	2014/12	670	100
24	Leipzig-Mitte	51.34	12.38	115	2010/06	2014/12	670	100
25	Leipzig-West	51.32	12.30	126	2013/01	2014/12	670	99
26	Mace Head	53.33	-9.90	5	2009/01	2010/12	670	100
27	Melpitz	51.53	12.93	90	2007/01	2014/12	670	100
28	Montseny	41.77	2.35	710	2009/01	2014/12	637; 670; 520	100
29	Neumayer	-70.67	-8.27	50	2006/03	2014/12	637; 670	100
30	Observatoire Perenne de l'Environnement	48.56	5.51	396	2012/09	2014/12	520	99
31	Pallas (Sammaltunturi)	67.97	24.12	572	2007/09	2014/12	637	100
32	Preila	55.38	21.03	5	2010/03	2010/12	520	100
33	SIRTA Atmospheric Research Observatory	48.71	2.16	168	2012/01	2012/12	520	99
34	Southern Great Plains E13	36.60	-97.48	328	2005/01	2014/12	530	96
35	Tiksi	71.59	128.92	18	2007/05	2014/12	520	98
36	Trinidad Head	41.05	-124.15	117	2005/01	2014/12	528; 530	98
37	Vavihill	56.02	13.15	175	2008/05	2011/12	520	98
38	Waldhof	52.80	10.76	78	2013/01	2014/09	670	100
39	Zeppelin mountain (Ny-Ålesund)	78.91	11.89	478	2005/01	2014/12	520; 525	100

5 S1.2 Scattering coefficient data

Table S2: GAW site details for in-situ dry scattering coefficient data. Start and Stop indicate first and last available months for computation of climatology in the 2005–2015 time frame. λ provides measurement wavelengths (multiple wavelengths can occur, e.g. in case instruments were changed / updated over time). Cov. indicates the percentage loss of daily average values when requiring 25% coverage in the provided hourly data. The following sites did not meet the coverage constraints for the computation of the climatology: Columbia River Gorge, East Trout Lake, Great Gulf Wilderness, Preila.

#	Station name	Lat.	Lon.	Alt. [m]	Start	Stop	λ [nm]	Cov. [%]
1	Acadia National Park-McFarland Hill (ME98)	44.38	-68.26	150	2005/01	2014/09	550	56
2	Alert	82.50	-62.34	420	2005/01	2014/12	550	96
3	Anmyeon-do	36.54	126.33	52	2008/01	2014/10	550	92
4	Barrow	71.32	-156.61	21	2005/01	2014/12	550	85
5	Birkenes II	58.39	8.25	223	2010/01	2014/12	550	98
6	Bondville	40.05	-88.37	223	2005/01	2014/12	550	96
7	Cabauw Zijdeweg	51.97	4.93	1	2008/01	2012/12	550	99
8	Cape Point	-34.35	18.49	260	2006/01	2013/09	550	88
9	Cape San Juan	18.38	-65.62	75	2005/03	2014/10	550	72
10	Cedar Bluff	38.77	-99.76	665	2005/01	2007/07	550	65
11	Cohutta	34.79	-84.63	735	2005/01	2007/03	550	64
12	DEM_Athens	37.99	23.82	280	2012/01	2014/12	525;	98
							520	
13	El Arenosillo	37.10	-6.73	51	2006/01	2014/12	550	85
14	Finokalia	35.32	25.67	255	2013/01	2014/12	550	99
15	Glacier National Park-Fire Weather Station (MT05)	48.51	-114.00	980	2008/03	2012/06	550	55
16	Gosan	33.29	126.16	81	2011/09	2014/12	550	98
17	Great Smoky Mountains NP	35.63	-83.94	813	2005/01	2014/12	550	53
18	Hohenpeissenberg	47.80	11.01	985	2006/01	2014/11	550	97
19	Hyytiälä	61.85	24.28	185	2010/12	2014/12	550	100
20	Ispra	45.80	8.63	214	2005/01	2014/12	550	94
21	K-puszta	46.97	19.58	135	2008/04	2014/12	550	98
22	Kosetice (NOAK)	49.57	15.08	539	2012/08	2014/12	550	94
23	Mace Head	53.33	-9.90	15	2005/01	2013/12	550	96
24	Mammoth Cave National Park-Houchin Meadow	37.13	-86.15	236	2005/01	2014/11	550	56
25	Melpitz	51.53	12.93	90	2012/01	2012/12	550	100
26	Montserrat	41.77	2.35	710	2010/01	2014/12	525	97
27	Mount Rainier National Park-Tahoma Woods (WA99)	46.76	-122.12	427	2005/03	2014/11	550	47
28	Neumayer	-70.67	-8.27	50	2013/01	2014/12	550	100
29	Observatoire Perenne de l'Environnement	48.56	5.51	396	2012/09	2014/12	525	92

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Table S2: GAW site details for in-situ dry scattering coefficient data. Start and Stop indicate first and last available months for computation of climatology in the 2005–2015 time frame. λ provides measurement wavelengths (multiple wavelengths can occur, e.g. in case instruments were changed / updated over time). Cov. indicates the percentage loss of daily average values when requiring 25% coverage in the provided hourly data. The following sites did not meet the coverage constraints for the computation of the climatology: Columbia River Gorge, East Trout Lake, Great Gulf Wilderness, Preila.

#	Station name	Lat.	Lon.	Alt. [m]	Start	Stop	λ [nm]	Cov. [%]
30	Organ Pipe Cactus National Monument	31.95	-112.80	506	2005/01	2010/05	550	92
31	Pallas (Sammaltunturi)	67.97	24.12	572	2005/01	2014/12	550	97
32	SIRTA Atmospheric Research Observatory	48.71	2.16	168	2012/05	2013/12	525; 450	91
33	Seney NWR	46.29	-84.05	216	2005/03	2006/06	550	47
34	Southern Great Plains E13	36.60	-97.48	328	2005/01	2014/12	550	94
35	Trinidad Head	41.05	-124.15	117	2005/01	2014/12	550	98
36	Upper Buffalo Wilderness	35.83	-93.20	722	2005/02	2009/09	550	54
37	Vavihill	56.02	13.15	180	2012/01	2014/03	520	94
38	Wichita Mountains	34.73	-98.71	509	2005/01	2007/08	550	77
39	Zeppelin mountain (Ny-Ålesund)	78.91	11.89	474	2010/07	2013/12	550	100

S2 Evaluation of satellite products at AERONET stations

All satellite data sets (see Table 1 in main manuscript) that were used for the model evaluation were evaluated themselves against the ground based AERONET data. This was done in order to establish an estimate of the relative differences (biases, correlation coefficients) between the different data sets when comparing them with the models (e.g. a difference in model bias

10 against AERONET and a satellite product may be due to differences in spatio-temporal sampling, but may also resemble a systematic bias in the satellite dataset due to retrieval errors). The evaluation of the gridded satellite level 3 products against AERONET was performed in the same manner as the evaluation of the models (see Sect. 2.3 in main manuscript). Note that for this analysis the satellite data were used in the original $1^\circ \times 1^\circ$ resolution.

The results of this analysis are available online on the AeroCom satellite evaluation website (Mortier et al., 2020), allowing 15 interactive exploration of the data and evaluation metrics (see Sect. 2.3 in the main manuscript for details regarding the web-interface). Note that the website contains results from more satellites than were used in this article.

In addition, the retrieved network averaged normalised-mean-biases (NMBs) from this assessment of the 2010 satellite records are provided in Table 1 in the main manuscript.

The retrieved correlation (metric R) against the AERONET observations are generally high ($R > 0.80$) with highest correlation (R = 0.89) being found for the merged satellite dataset (Sogacheva et al., 2020), suggesting that the satellites are capable 20 of monitoring spatio-temporal variations in the AOD signal. In terms of NMB, AATSR-SU and the MERGED-FMI product show slight underestimations ($NMB \approx -5\%$) while MODIS Aqua and Terra yield slight overestimates AODs, approximately +9 % and +17 %, respectively (see product names M6.1-Aq-DTDB, M6.1-Tr-DTDB in Mortier et al., 2020).

This analysis is biased by the uneven distribution of AERONET sites (highest density in Europe and North America) and 25 thus some problematic regions in the satellite retrievals (e.g., regions with high surface albedo, such as the Sahara desert) may not be well represented in this comparison. In addition, this assessment provides little insights about the satellite performance over oceans (sea salt dominated), since the AERONET site coverage is land dominated.

In the case of the AATSR-SU data, the retrieval includes a conservative cloud mask utilising thermal channels in addition to optical, and thereby avoids retrieval near cloud edges. Evaluation under aerosol CCI of six data sets showed AATSR and 30 SeaWifs exhibited the lowest bias (with SeaWifs) with respect to ocean and coastal sun photometers Popp et al. (2016).

S3 Spatio-temporal sensitivity of satellite evaluation results

The established metrics (e.g., NMB, R) retrieved when comparing models with level 3 gridded satellite datasets (see e.g. Fig. 6 in main manuscript) are sensitive to the temporal and spatial resolution used. Since many AP3-CTRL model outputs were provided only in monthly resolution fields, all statistics were computed in monthly resolution. Further, as described

35 above, the satellite products were spatially re-gridded to a latitude / longitude resolution of $5^\circ \times 5^\circ$ in order to increase the temporal coverage and to aggregate enough measurements to meet the required 25 % time sampling constraint in each grid point, for monthly averaging (for details see main manuscript). In order to investigate the impacts of this choice of spatio-temporal resolution, a further sensitivity study was performed. For all models (and variables) that have submitted data also in daily (or higher) resolution, the analysis was repeated in the highest available resolution (i.e. $1^\circ \times 1^\circ$ for models with higher
40 resolution, else, native model resolution). Table S3 summarizes the differences from this study. In most cases NMBs are shifted towards more positive values for the higher resolution dataset, with differences of up to +13 % (AOD ECMWF-IFS vs. MODIS satellites). This positive shift usually conducts to lower absolute biases as most models underestimate the considered variables.
45 In general, it appears that the bias shift is larger in the higher resolution models, however, correlation coefficients are slightly worse in high resolution dataset. The latter is expected due to the smoothing effect that temporal and spatial averaging has on the results (i.e. it is easier for a model to match a monthly average within a 500x500km grid box than a daily average in a 100x100km grid box).

Table S3. Comparison of statistics (NMB and R) retrieved when co-locating models with satellite data a) in *Low* resolution, that is, using monthly means and re-gridding to $5^\circ \times 5^\circ$ horizontally (with 25 % temporal sampling coverage requirement, as done in this study) and b) in *High* resolution, that is, using daily means and highest available horizontal resolution (*High*).

Model (Lat./Lon. res.)	Satellite	Variable	Statistic:	NMB [%]		R	
			Resolution:	Low	High	Low	High
CAM5-ATRAS (1.9 x 2.5)	AATSR4.3-SU	AOD	-17.65	-18.31	0.66	0.50	
	MODIS6.1-aqua	AOD	-36.42	-30.67	0.57	0.35	
	MODIS6.1-terra	AOD	-42.16	-36.70	0.55	0.34	
	ECMWF-IFS (0.4 x 0.4)	AE	-38.80	-28.63	0.70	0.59	
		AOD	-18.15	-18.73	0.82	0.70	
		AOD _c	14.51	5.69	0.78	0.66	
EMEP (0.5 x 0.5)	AATSR4.3-SU	AOD _f	-39.84	-36.20	0.81	0.66	
		MODIS6.1-aqua	AOD	-34.25	-21.79	0.71	0.57
		MODIS6.1-terra	AOD	-40.18	-27.55	0.69	0.56
	MODIS6.1-aqua	AE	36.57	42.61	0.67	0.50	
		AOD	-34.40	-30.38	0.73	0.58	
		AOD _c	-69.81	-69.25	0.64	0.54	
OsloCTM3 (2.2 x 2.2)	MODIS6.1-terra	AOD _f	-10.88	-2.56	0.74	0.57	
		AOD	-44.31	-36.49	0.68	0.51	
		MODIS6.1-aqua	AOD	-49.31	-41.25	0.66	0.50
	AATSR4.3-SU	MODIS6.1-terra	AOD	-31.88	-30.32	0.74	0.53
		AE	-12.35	-13.43	0.83	0.69	
		MODIS6.1-aqua	AOD	-25.14	-23.65	0.76	0.55
SPRINTARS (0.6 x 0.6)	AATSR4.3-SU	MODIS6.1-terra	AOD	-31.88	-30.32	0.74	0.53
		AE	-8.13	0.88	0.68	0.55	
		AOD	2.92	8.71	0.74	0.62	
	TM5 (2.0 x 3.0)	AOD _c	-1.84	-3.93	0.75	0.55	
		AOD _f	-9.76	-14.64	0.64	0.41	
		MODIS6.1-aqua	AOD	3.42	3.42	0.81	0.66
	AATSR4.3-SU	MODIS6.1-terra	AOD	-18.78	-15.83	0.79	0.55
		AOD	-26.09	-22.84	0.76	0.53	

S4 Models

The following sub-sections briefly introduce each of the individual models used in the paper (see also Tab. 2 in the main manuscript and supplement 1 for further information).

50 S4.1 CAM5-ATRAS

The Community Atmosphere Model version 5 (CAM5) with the Aerosol Two-dimensional bin module for foRmation and Aging Simulation (ATRAS, Matsui, 2017; Matsui and Mahowald, 2017) calculates the following atmospheric aerosol and chemistry processes: emissions, gas-phase chemistry, new particle formation, condensation of sulphate, nitrate, and organic aerosols, coagulation, cloud activation, aqueous-phase chemistry, dry and wet deposition, and aerosol-radiation and aerosol-cloud interactions. Aerosol particles from 1 nm to 10000 nm in dry diameter are represented with a two-dimensional sectional representation with 12 size bins and 8 BC mixing state bins. Meteorological nudging was used for temperature and wind fields in the free troposphere (<800 hPa) using data from the Modern-Era Retrospective Analysis for Research and Applications version 2 (MERRA2, Gelaro et al., 2017). Aerosol optical properties are calculated based on the Mie theory with the core-shell (BC core and non-BC shell) treatment for internally-mixed BC particles and the well-mixed treatment for pure BC and BC-free particles (Matsui, 2017).

S4.2 EC-Earth3-AerChem and TM5

Two configurations of the atmospheric composition model TM5 (Tracer Model 5) are included in this study (van Noije et al., 2014): a standalone version of TM5, and an atmosphere-only version of the CMIP6 climate model EC-Earth3-AerChem (van Noije et al., 2020). The standalone model is driven by meteorological and surface fields from the ERA-Interim reanalysis (Dee et al., 2011), whereas in the climate model there is online interaction between TM5 and the atmospheric general circulation model, which is based on model cycle 36r4 of ECMWF's Integrated Forecasting System (IFS). The sets of meteorological and surface variables that drive TM5 are similar in both configurations. In the EC-Earth simulations analyzed in this study, sea surface temperatures and sea ice concentrations were prescribed using forcing fields from the Atmospheric Model Intercomparison Project (AMIP, Gates et al., 1999) provided for CMIP6; in addition, vorticity, divergence and surface pressure fields were nudged to ERA-Interim, using a Newtonian relaxation scheme with a time constant of 8 h and 15 min in the whole atmosphere. TM5 uses the aerosol scheme M7 (Vignati et al., 2004), which represents sulphate, black carbon, organic aerosols, sea salt and mineral dust with seven lognormal size distributions or modes. Aerosol components are assumed to be internally mixed inside the modes. The formation of secondary organic aerosols in the atmosphere is described following Bergman et al. (2020). Ammonium-nitrate and methane sulphonic acid (MSA) are described by their total mass, and assumed to be present only in the soluble accumulation mode (see van Noije et al., 2020, for more details). TM5 has an interactive tropospheric chemistry scheme (Williams et al., 2017), which also describes the aqueous-phase oxidation of dissolved sulphur dioxide in clouds. When calculating the dust source, TM5 does not include particles with dry diameter larger than 16 μm . The aerosol optical properties in TM5 are calculated based on Mie theory, where the mixing rules of Bruggeman and Maxwell Garnett are applied as approximations of the refractive index of the internally mixed modes.

80 S4.3 ECHAM-HAM

The global aerosol-climate model ECHAM6.3-HAM2.3 (ECHAM-HAM in the following) is part of the fully coupled aerosol chemistry climate model ECHAM–HAMMOZ (Tegen et al., 2019; Schultz et al., 2018). ECHAM-HAM uses the PSrad two-stream radiation scheme which uses 16 shortwave and 14 longwave wavelength bands (Pincus and Stevens, 2013). Aerosol microphysical processes in ECHAM-HAM are described with the modal M7 aerosol model (Vignati et al., 2004) in contrast to ECHAM-SALSA which employs the sectional aerosol scheme SALSA (Kokkola et al., 2018). The aerosol size distribution is represented by seven lognormal modes (four soluble and three insoluble modes) and the aerosol components include sulphate, black carbon, particulate organic matter, sea salt, mineral dust and aerosol water. The aerosol optical properties are computed for each mode using volume weighted mixing of the refractive indices of the aerosol components. Pre-computed values based on Mie theory of the aerosol optical properties are read from a look-up table using the Mie size-parameter and the volume

90 weighted refractive index (real and imaginary part) of each mode for each wavelength band. Aerosol water uptake follows
κ-Köhler theory (Petters and Kreidenweis, 2007) and is computed from the chemical composition of each mode and ambient
relative humidity of the cloud-free part of the grid box (O'Donnell et al., 2011). The aerosol representation in ECHAM-HAM
has been evaluated in Tegen et al. (2019) but using different aerosol emissions (different inventories for anthropogenic and
biomass burning emissions as well as a different sea salt emission parameterisation). For the AP3-CTRL experiment the sea
95 salt emission parameterisation from Guelle et al. (2001) was chosen, firstly because the one proposed by Long et al. (2011)
(with temperature correction of Sofiev et al., 2011) resulted in an underestimation of the sea salt concentrations (Tegen et al.,
2019) and secondly, to be consistent with the AP3-CTRL setup of ECHAM-SALSA (Sect. S4.4). However, this comes at the
100 price of larger sea salt particles (on average), resulting in a slightly decreased correlation against AERONET compared to
Tegen et al. (2019). Mineral dust emissions are computed online from the 10 m wind speed, only in predefined potential dust
105 source areas (Tegen et al., 2002; Cheng et al., 2008; Heinold et al., 2016). Dust emission fluxes are computed internally well
size-resolved and are subsequently remapped and emitted to the two largest M7 insoluble modes using mass median emission
radii of $0.37 \mu\text{m}$ and $1.75 \mu\text{m}$ with standard deviations of 1.59 and 2.00 respectively. The experiment analysed in this study
110 was performed at T63 spectral horizontal resolution ($1.875^\circ \times 1.875^\circ$) with 47 vertical hybrid-sigma levels with a model top
at 0.01 hPa. Vorticity, divergence and surface pressure were nudged towards ERA-Interim (Dee et al., 2011) using relaxation
115 times of 6 h, 48 h and 24 h respectively and sea surface temperature and sea ice cover are prescribed from AMIP data for the
respective year (Taylor et al., 2000).

S4.4 ECHAM-SALSA

110 SALSA is the sectional aerosol microphysics module within the ECHAM-HAMMOZ aerosol-chemistry-climate model (Kokkola
et al., 2018) alongside the modal aerosol module M7 (Tegen et al., 2019). The implementation of SALSA to ECHAM-
HAMMOZ and its evaluation against satellite retrievals, ground-based remote sensing retrievals, and in situ observations has
been described by Kokkola et al. (2018). One change in these model simulations compared to those in Kokkola et al. (2018)
are, in addition to using anthropogenic emissions required for AeroCom phase III simulations, is using sea salt emission
115 parameterisation of Guelle et al. (2001) for the reasons described in the previous Section S4.3.

S4.5 ECMWF-IFS

115 As part of the Copernicus Atmosphere Monitoring Service (CAMS; <https://atmosphere.copernicus.eu/>), ECMWF runs a version
of the IFS model that includes prognostic aerosol and tropospheric chemistry schemes to produce global forecasts of
atmospheric composition. The underlying meteorological model is essentially identical to that used for operational medium-
range weather forecasting and is documented at <https://www.ecmwf.int/en/forecasts/documentation-and-support>, but at a lower
120 resolution of 40 km to offset the cost of the extra schemes. The results presented here are from a “cycling forecast” configu-
ration, that is, a forecast with free-running aerosols and chemical species (no assimilation of atmospheric composition), with
meteorology reinitialised at 00 UTC each day from operational ECMWF analyses. The model version used in this paper is
125 cycle 46R1.¹

The aerosol component is described in Rémy et al. (2019) and based on the earlier work of Morcrette et al. (2009). This is an
externally-mixed hybrid bin / bulk scheme, consisting of three size bins each for desert dust (up to $20 \mu\text{m}$ dry radius) and sea salt
130 (up to $20 \mu\text{m}$ radius at 80 % relative humidity), and bulk tracers for organic matter, black carbon, sulphate, nitrate (from gas-
particle partitioning and heterogeneous reactions at the surface of dust and sea-salt aerosol particles) and ammonium aerosol.
For organic matter and black carbon, there are separate hydrophobic and hydrophilic tracers, with a fixed ageing timescale for
conversion of the former to the latter. Sulphate production rates are provided by the global chemistry component of the IFS,
IFS-CB05. There is no primary sulphate aerosol emission. The tropospheric chemistry scheme is described in Flemming et al.
135 (2015); in the model version used for AP3-CTRL simulations it is directly coupled to the aerosol scheme by providing the
precursor gases for the reactions that produce nitrate and ammonium as well as the sulphate production rates.

¹Note that the ECMWF-IFS model version was updated during revision (i.e., cycle 45R1 was used in the discussion paper, Gliß et al., 2020).

S4.6 EMEP MSC-W

The EMEP MSCW model is a chemical transport model, designed for policy related applications to combat acid deposition, eutrophication and health adverse air pollution (Simpson et al., 2012). It accounts for all main anthropogenic (SO_4 , NO_3 , NH_4 , BC and OC) and natural (SS, DU, biogenic SOA) aerosols, contributing to the health related indicators $\text{PM}_{2.5}$ and PM_{10} . The results presented in the paper were obtained in a model run at $0.5 \times 0.5^\circ$ grid, driven by 3-hourly ECMWF-IFS meteorology and using (differently from the other models) ECLIPSE6b annual anthropogenic emissions with EMEP default monthly profiles, ECLIPSE6a for international shipping, and FINN forest fire emissions (Wiedinmyer et al., 2011), all for the year 2010. The model includes aerosols with diameters up to $10 \mu\text{m}$ and calculates the mass concentrations of aerosols in fine (below $\text{PM}_{2.5}$) and coarse $\text{PM}_{2.5-10}$ mode. Based on these, the extinction and absorption coefficients are calculated for the individual aerosol components using mass extinction/absorption coefficients, implicitly accounting for aerosol hygroscopic growth (aerosol effective radii, growth factors and specific extinction efficiencies are tabulated, Schulz et al., 2012). The results in this paper indicate an inconsistency between MEC and MAC for BC (see Tab. 3 in main manuscript). This is likely due to the fact that MAC is a constant value in EMEP, selected from a range of values reported in (Bond et al., 2013). MEC, on the other hand, is computed online based on extinction efficiencies from (Hess et al., 1998) and (Chin et al., 2002) to account for hygroscopic growth.

S4.7 GEOS

GEOS is a global Earth system model, containing components for atmospheric circulation and composition, ocean circulation and biogeochemistry, land surface processes, and data assimilation (Rienecker et al., 2008). The version of GEOS Earth System Model (with a GOCART aerosol module) used for this study is *Icarus-3_3_p2*. The simulations run at a latitude/longitude resolution of $1.0^\circ \times 1.0^\circ$ and 72 vertical levels from surface up to 0.01 hPa (~ 85 km) with “replay” mode, denoting simulations driven by the reanalysis meteorological fields from MERRA2.

The GOCART module includes major aerosol types of BC, organic carbon (OC, see supplement 1 for assumed OC to OA ratios), brown carbon (BrC), SO_4 , NO_3 , NH_4 , DU, and SS (Chin et al., 2002; Colarco et al., 2010; Bian et al., 2019). The emissions of dust, sea salt, DMS, and biogenic VOCs are model calculated time-varying fields. All other aerosol emissions used in this study follow the AeroCom phase III protocol. The major updates on this GOCART version include newly implemented nitrate and ammonium (Bian et al., 2017), anthropogenic and biomass burning SOAs, as well as separate treatment of optical properties for BrC (from biomass burning source) and OC (from all other sources).

S4.8 GFDL-AM4

The Geophysical Fluid Dynamics Laboratory Atmospheric Model version 4 has cubed-sphere topology with 96×96 grid boxes per cube face (approximately 100 km grid size) and 33 levels in the vertical, contains an aerosol bulk model that generates mass concentration of aerosol fields (sulphate, carbonaceous aerosols, sea salt and dust) from emissions and a “light” chemistry mechanism designed to support the aerosol model but with prescribed ozone and radicals (Zhao et al., 2018). The model is driven by time-varying boundary conditions, and natural and anthropogenic forcings developed in support of CMIP6 (Eyring et al., 2016), except for ship emission of SO_2 (BC ship emission is included), which has unintentionally not been included. The dust is emitted from constant sources with their erodibility expressed as a function of surrounding topography (Ginoux et al., 2001). The sea salt emissions are based on Mårtensson et al. (2003) and Monahan et al. (1986) for fine and coarse mode particles, respectively. Aerosols are externally mixed except for black carbon, which is internally mixed with sulphate. The optical properties of the mixture are calculated by volume weighting of their refractive indices using a Mie code. In the present configuration, the model is run with observed sea surface temperatures (SSTs) and sea-ice distribution (Taylor et al., 2000). In addition, the wind components are nudged, with a 6-hour relaxation time, towards the NCEP-NCAR re-analysis provided on a T62 Gaussian grid with 192 longitude equally spaced and 94 latitude unequally spaced grid points (Kalnay et al., 1996). This resolution is lower than in GFDL-AM4, which may create a low bias of aerosol emission depending on surface winds.

175 S4.9 GISS-OMA

GISS-OMA is the short name of the GISS ModelE Earth system model (Kelley et al., 2020), coupled with the One-Moment Aerosol scheme (OMA, Bauer et al., 2020). In OMA, all aerosols are externally mixed and tracked by their total mass only, except for sea salt and dust where 2 and 5 size-resolved sections are used, respectively. OMA tracks sulphate, nitrate, ammonium, carbonaceous aerosols (black and organic carbon), dust (up to $16\text{ }\mu\text{m}$) and sea salt (up to $4\text{ }\mu\text{m}$).

180 Relevant to this work, a random maximum cloud overlap is calculated in the column, which is then used to define a totally cloudy or totally cloud-free state in radiation, using a pseudo-random number generation. This is described in Hansen et al. (1983).

185 For AS AOD calculations RH = 100 % is used, while for CS ambient is used. This applies to the whole atmospheric column, as dictated by the random maximum cloud overlap calculation. As a result, AS diagnostics show unrealistically high values when compared to observations. Therefore, only the provided CS diagnostics from GISS-OMA fields are presented in this paper.

S4.10 INCA

190 The INCA (INteraction with Chemistry and Aerosols) and ORCHIDEE land surface modules have been coupled to LMDZ dynamical core to conform the LMDZORINCA model. It has been run with forced sea-surface temperatures, sea-ice concentrations and with nudged monthly wind-fields from ERA-Interim. Comparisons with climatological simulations without nudged winds show slightly larger emissions in the un-nudged runs of those aerosols driven interactively by wind at the surface (Balkanski et al., 2004; Schulz et al., 2009). The aerosol modelling in INCA relies on a modal approach to represent the size distribution of DU, SS, BC, NO_3 , SO_4 , SO_2 and OA with a combination of accumulation and coarse log-normal modes (both soluble and insoluble). Since these runs use a simplified chemistry scheme, DMS emissions are prescribed and 195 not interactively calculated, and the secondary organic aerosols are not simulated.

200 The current version is modelling BC as internally mixed with sulphate (Wang et al., 2016), where the refractive index is estimated using the Maxwell-Garnett method. This results in an increased and more accurate BC absorption. On the other hand, the dust refractive index is deduced from dedicated experiments (Biagio et al., 2017, 2019) showing a marked impact on the longwave part of the spectrum. This results in a less absorbing dust aerosol. BC emissions are derived from inventories and are equally partitioned between surface and altitude.

S4.11 NorESM2

205 The atmosphere module in NorESM2 (NorESM2-MM, see Selander et al., 2020), CAM6-Nor (Olivié et al., 2020), is an updated version of CAM5.3-Oslo, for which optical properties have been described and validated by Kirkevåg et al. (2018). The grid latitude/longitude resolution for the atmosphere model is $0.9^\circ \times 1.25^\circ$, with 32 layers in the vertical and model top at 3.6 hPa. The aerosol components represented are sulphate, black carbon, primary and secondary OM, sea salt and mineral dust. Processes taken into account include: primary emission; gas- and aqueous-phase chemistry; nucleation; growth by coagulation, condensation (including water uptake) and cloud processing; in-cloud and below-cloud scavenging; dry deposition and gravitational settling. The aerosol module, OsloAero6, makes use of a “production tagged” method (Kirkevåg et al., 2018) to calculate aerosol size and chemical composition. It describes a number of “background” log-normal modes that can change their size 210 distribution due to condensation, coagulation, and cloud processing. A detailed offline size-resolving model (AeroTab6, see AeroTab user guide, 2020) carries out the corresponding aerosol micro-physical calculations, from which results are stored in lookup tables for use in OsloAero6 (in CAM6-Nor). Hygroscopicity is estimated for each particle size and type by use of the volume mixing rule for internal mixtures, adding water as a function of RH according to Köhler theory.

S4.12 OsloCTM3

215 The OsloCTM3 is a global, offline chemical transport model (CTM) driven by 3-hourly meteorological data from the European Centre for Medium Range Weather Forecast (ECMWF) Integrated Forecast System (IFS) model, and is an updated version of the OsloCTM2 used in previous AeroCom phases (Søvde et al., 2012; Lund et al., 2018). The model is run in a

2.25° x 2.25° horizontal resolution, with 60 vertical levels (the uppermost centered at 0.1 hPa). OsloCTM3 includes the aerosol species sulphate, black carbon, organic aerosols, nitrate, sea salt, dust. The treatment of transport and scavenging, as well as individual aerosol modules, is described in detail in Lund et al. (2018) and references therein. In OsloCTM3, the absorption properties have been updated, with BC mass absorption coefficient (MAC) following formula in Zanatta et al. (2016) and a weak absorption implemented for OA (Lund et al., 2018). Only all-sky (AS) AOD is provided from OsloCTM3. This is done because a reliable sub-grid scale parameterisation for RH is unavailable, in order to avoid the AOD used in the radiative transfer calculations to be biased low or high.

225 **S4.13 SPRINTARS**

SPRINTARS (Takemura et al., 2005, 2009), coupled with an atmosphere-ocean general circulation model, MIROC (Tatebe et al., 2019), is used in this study. SPRINTARS predicts the mass mixing ratios of major tropospheric aerosol components (SO_4 , BC, OA, soil DU, SS, and precursor gases of SO_4 , including SO_2 and dimethyl sulphide (DMS)) by simulating the aerosol transport processes. It also simulates aerosol–radiation and aerosol–cloud interactions by connecting atmospheric radiation and cloud/precipitation processes, respectively, with MIROC. The 6-hourly ECMWF ERA5 data (Hersbach et al., 2020) are used for nudging temperature and horizontal wind. The horizontal resolution is T85 (approximately 1.4° x 1.4° in longitude and latitude), and the vertical resolution of the hybrid sigma-pressure coordinate was 40 layers.

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