

## Evaluation of global simulations of aerosol number and cloud condensation nuclei, and implications for cloud droplet formation

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## 15 **Appendix S1. Model and measurements description**

### **Details on aerosol microphysics representation in the participating models (Tables S1, S2, S3):**

The original M7 scheme (Vignati et al., 2004) uses seven log-normal distributions to represent aerosol populations. Four of these are describing mixed (water-soluble) particles, in the nucleation, Aitken, accumulation and coarse modes, and the  
20 remaining three modes include insoluble particles in the Aitken, accumulation and coarse modes. Each mode is represented by the total number concentration of particles and the mass mixing ratio of each aerosol component. Soluble particles are formed from insoluble ones by coagulation and condensation.

With respect to the original M7 scheme, ECHAM5.5-HAM2-ELVOC\_UH and TM4-ECPL are using an improved numerical scheme (Kokkola et al., 2009) to compute the formation of sulfuric acid by oxidation of SO<sub>2</sub>, and its removal by  
25 nucleation and condensation on pre-existing particles. The EMAC model uses the Global Modal-aerosol eXtension (GMXe) microphysics model (Pringle et al., 2010a), which is also based on M7 and allows the treatment of additional aerosol species (sodium, chloride, magnesium, calcium, and potassium).

The ECHAM5.5-HAM2-ELVOC\_UH, ECHAM6-HAM2-AP and ECHAM6-HAM2 models are using the same aerosol module HAM2 (Zhang et al., 2012; Tegen et al., 2018). Secondary organic aerosols (SOA) produced by oxidation of  
30 isoprene, monoterpene, and anthropogenic precursors and primary organic aerosols (POA) are treated as different tracers within the HAM2-ELVOC-UH module, while in ECHAM6-HAM2 and ECHAM6-HAM2-AP SOA is added to POA (see Tables S2 and S3). ECHAM6-HAM2-AP is identical to ECHAM6-HAM2 with the difference that the aerosol processing (AP) scheme in stratiform clouds by Neubauer et al. (2014) is used, which is adapted from Hoose et al. (2008a, 2008b). In

addition to the seven modes in ECHAM6-HAM2, ECHAM6-HAM2-AP has an explicit representation of aerosol particles in cloud droplets and ice crystals in stratiform clouds, which are represented by 5 tracers (sulfate – SO<sub>4</sub>, BC, OA, dust- DU and sea-salt- SS). Nucleation and impact scavenging of aerosols, freezing and evaporation of cloud droplets, and melting and sublimation of ice crystals are treated explicitly. Aerosol particles from evaporating precipitation are released to modes, which correspond to their size and can therefore act as CCN or INP again.

The TM4-ECPL model also treats POA and SOA separately. The latter are produced from semi-volatile organics formed by oxidation of isoprene, terpene (a- and b- pinenes) and aromatic (benzene, toluene and xylene) compounds (Tsigaridis and Kanakidou, 2003; Tsigaridis and Kanakidou, 2007) as well as by aqueous phase chemistry (Myriokefalitakis et al., 2011). For this study, although TM4-ECPL uses ISORROPIA II thermodynamic model (Fountoukis and Nenes, 2007) to calculate the portioning between HNO<sub>3</sub> and NO<sub>3</sub><sup>-</sup>, the contribution of particulate NO<sub>3</sub><sup>-</sup> to aerosol formation and growth is not taken into account.

TM5 uses a similar host model as TM4-ECPL. TM5 considers SOA formation from isoprene and monoterpenes. SOA formation mechanism is implemented according to Jokinen et al. (2015). Both monoterpenes and isoprene produce extremely low volatility organic compounds (ELVOCs) and semi-volatile products with prescribed yields (from Jokinen et al., 2015). Oxidation by OH and O<sub>3</sub> is considered. The ELVOCs participate in aerosol nucleation, which is parameterized according to Riccobono et al. (2014) accounting for sulfuric acid and organic vapors. In addition, ELVOCs in TM5 provide early growth for nucleation mode particles, as they are distributed to particle phase according to condensation sink. The semi-volatile products are distributed to particle phase according to particle-phase organic mass, as in Jokinen et al. (2015). Hence, after oxidation, no SOA products remain in the gas-phase, but immediate condensation to aerosol-phase is assumed. The approach provides dynamics for aerosol mass and number formation, but does not consider e.g. particle-phase reactions, evaporation of organic material, or aqueous-phase SOA formation. More detailed description can be found in Bergman et al. (in prep.)

CAM5-MAM3 employs the three-mode modal aerosol module MAM3 (Liu et al., 2012). Within each of the three modes (Aitken, accumulation, and coarse mode) all types of aerosols are internally mixed, while particles between different modes are externally mixed. SO<sub>4</sub>, BC, POA, SOA, SS, DU are the aerosol components considered using a total of 15 aerosol tracers. CAM5-MAM4 has an extra primary-carbon mode to treat freshly emitted POA and BC, amounting to in total 18 aerosol tracers (Liu et al., 2016). The MAM3 and MAM4 aerosol modules describe the following microphysical processes: nucleation, condensation, coagulation, ageing, dry (gravitational and turbulent) deposition, wet (in-cloud and below-cloud scavenging) deposition, in-cloud activation, and release from the evaporation of clouds and raindrops.

Two of the global models (GEOS-Chem-APM and CAM5-Chem-APM) are using the sectional Advanced Particle Microphysics (APM) package (Yu and Luo, 2009). The APM package is optimized to accurately simulate secondary particles (composed of SO<sub>4</sub>, NO<sub>3</sub>, NH<sub>4</sub>, and SOA) formation and their growth to CCN sizes, with a higher size resolution for the size range of importance (1.2–120 nm: 30 bins, 10 additional bins for 120 nm–12 μm). 20 sectional bins represent sea salt, covering dry diameters from 0.012 to 12 μm, and 15 sectional bins represent dust for 0.03–50 μm. Two modes (Aitken

mode and accumulation mode) are used in this study to represent hydrophobic and hydrophilic BC and POC in the model. APM also considers the aging of BC and OA that turns the hydrophobic BC and OA to hydrophilic. Coating of SO<sub>4</sub> and other secondary aerosols on primary particles such as BC, OA, SS, and DU are considered. The contribution of nitrate, ammonium, and SOA to SO<sub>4</sub> particle growth are considered. Via the coating process caused by coagulation, condensation, and in-cloud oxidation, some secondary species attach to primary particles and are transported and scavenged with these primary particles. The kinetic condensation of low volatile secondary organic gas (LV-SOG) in addition to sulfuric acid on nucleated particles is calculated based on a scheme that considers the SOG volatility changes arising from the oxidation aging (Yu, 2011).

Two-Moment Aerosol Sectional (TOMAS) microphysics package (Adams and Seinfeld, 2002; Lee and Adams, 2012) was used by the GISS-E2-TOMAS (Lee et al., 2015) and GEOS-Chem-TOMAS (Kodros et al., 2016) global models. Both models simulate SO<sub>4</sub>, SS, OA, BC and DU. In GEOS-Chem-TOMAS, TOMAS tracks two independent moments (number and mass) within each of 15 size sections for size-resolved condensation, coagulation, nucleation, dry deposition, wet deposition, emissions, and aqueous chemistry. Condensation includes sulfuric acid and SOA (including both biogenic SOA and anthropogenically enhanced SOA as described in D'Andrea et al., 2013). GEOS-Chem-TOMAS accounts for binary (Vehkamäki et al., 2002) and ammonia ternary (Napari et al., 2002) nucleation, where ternary nucleation rates are scaled down by a factor of 105 to match observed nucleation rates (Westervelt et al., 2013). TOMAS has also been integrated into GISS-E2 (Lee et al., 2015), a fully coupled Earth System Model with interactive atmosphere, land, ocean, and sea ice components (Schmidt et al., 2014). TOMAS in GISS-E2 consists of 15 size sections covering 3 nm to 10  $\mu\text{m}$ . Condensation in GISS-E2-TOMAS includes both sulfuric acid and a simpler treatment of SOA than is present in GEOS-Chem-TOMAS, in which 10% of terpene emissions are condensed as hydrophilic organic aerosol, resulting in a production rate of 17.1 Tg a<sup>-1</sup>. GISS-E2-TOMAS uses a physically-based aerosol-cloud activation parameterization from Nenes and Seinfeld (2003), using both modeled large-scale and sub-grid updraft velocity.

The Multiconfiguration Aerosol TRacker of mIXing state (MATRIX) module (Bauer et al., 2008; 2010; Bauer and Menon, 2012) is used by GISS-E2.1-MATRIX. MATRIX is an aerosol microphysics scheme based on the quadrature method of moments (QMOM; McGraw, 1997). MATRIX represents new particle formation (binary and ternary nucleation), particle emissions, gas-particle mass transfer, aerosol phase chemistry, condensational growth, and coagulation within and between particle populations. Condensational growth is calculated for the sulphate-ammonium-nitrate-water system. Unique to MATRIX is the ability to explicitly simulate aerosol mixing state (Bauer et al., 2013). For this study MATRIX is set up with 16 aerosol populations, as defined by their mixing state. The tracked variables vary by population but can include number concentration and mass concentration of SO<sub>4</sub>, NO<sub>3</sub>, NH<sub>4</sub>, aerosol water, BC, OA, mineral dust, and SS. MATRIX is coupled to the NASA GISS climate model, GISS-E2.1, an updated version form model GISS-E2 (Schmidt et al., 2014). Sea-salt, dust and isoprene emission fluxes are calculated interactively, and additional natural and anthropogenic fluxes are from the CMIP6 inventory (Hoesly et al., 2017; Marle et al., 2017).

The CAM5-chem-ATRAS2 is using the Aerosol Two-dimensional bin module for foRmation and Aging Simulation version 2 (ATRAS2) (Matsui, 2017; Matsui and Mahowald, 2017). The ATRAS2 model uses 12 size bins from 0.001 to 10  $\mu\text{m}$  in aerosol dry diameter. BC mixing state is resolved with 8, 3, or 1 bins for each size bin. In this study, we perform simulations with a single mixing state representation. Mass concentrations of eight aerosol species (SO<sub>4</sub>, NO<sub>3</sub>, NH<sub>4</sub>, DU, SS, OA (POA+SOA), BC, and water) and number concentrations are calculated for each bin in the model. The number of advected species (chemistry and aerosol) is 215 in this study. The CAM5-chem-ATRAS2 model considers emissions, gas-phase chemistry, condensation and evaporation of SO<sub>4</sub>, NO<sub>3</sub>, NH<sub>4</sub>, and OA, coagulation, nucleation, activation of aerosol and evaporation from cloud, aerosol formation in clouds, dry and wet deposition, aerosol optical properties, aerosol-radiation interactions, and aerosol-cloud interactions. OA formation is calculated by a volatility basis-set approach which considers oxidations of POA (gas-phase), alkanes, alkenes, aromatics, isoprene, and monoterpene (Matsui et al., 2014; Matsui, 2017). Nucleation is calculated by the activation-type theory (Kulmala et al., 2006) within the planetary boundary layer and by a binary homogeneous nucleation scheme (Vehkamäki et al., 2002) within the free troposphere.

CAM5.3-Oslo (Kirkevåg et al., 2018; Karset et al., 2018) includes a “production tagged” aerosol life-cycle module (OsloAero5.3), and comes with an offline microphysics scheme (AeroTab5.3), which is used to make look-up tables for aerosol optics and dry sizes. In AeroTab5.3, the size distributions of number and mass concentrations are estimated by solving the respective continuity equations, using 44 size bins with radii ranging from 0.001 to 20  $\mu\text{m}$ . The size modes are assumed to be log-normally distributed at the point of emission or nucleation, but are modified by condensation, coagulation, and cloud processing. The term “production-tagged” is used since the tracers that change the aerosol size distributions are tagged according to their production pathway (e.g., condensation). In the aerosol life-cycle module, scavenging and dry deposition also change the total size distribution, but not the shape/radius-dependence of each of the size modes. The size modes are here assumed to be log-normal, either with prescribed typical sizes, or with log-normal fits (through look-up tables, for use in the CCN activation) to the respective size distributions calculated offline. In total 21 aerosol tracers are taken into account, distributed onto 10 internally mixed and 2 externally mixed (except with water through hygroscopic growth) size-modes, consisting of SO<sub>4</sub>, SS, OC, BC, and DU.

HadGEM3-UKCA includes GLOMAP modal aerosol scheme, which is a two-moment scheme representing seven (7) lognormal modes; soluble nucleation, Aitken, accumulation and coarse modes and insoluble Aitken, accumulation and coarse modes. Each of these modes carries particle numbers and masses for 5 aerosol components; SO<sub>4</sub>, BC, OC (including both primary and secondary aerosols), DU and SS. Insoluble mode particles can be aged and become soluble when soluble secondary aerosol materials condense on them and form coating. All particles in the lowermost atmosphere are subject to dry deposition, all particles are subject to impaction scavenging by rainfall, and soluble particles are additionally subject to nucleation scavenging in large scale and convective rainfall.

**Table S1.** Host model description

| Model                   | Horizontal resolution<br>(latitude × longitude) | Vertical resolution          | Host Model              | Meteorology                                  | Model references   |
|-------------------------|---|------------------------------|-------------------------|--|--|
| CAM5-Chem-APM           | 1.875° × 2.5°                                   | 32                           | GCM                     | Online, Nudged with MERRA2                   | (Luo and Yu, 2018 in prep.)  |
| CAM5-Chem-ATRAS2        | 1.875° × 2.5°                                   | 30 up to 3.6 hPa             | GCM (nudged)            | MERRA U and V (6h)                           | (Matsui, 2017) (Matsui and Mahowald, 2017)   |
| CAM5_MAM3               | 1.875° × 2.5°                                   | 30 up to 3.6 hPa             | GCM (nudg)              | MERRA U and V (6h)                           | (Neale et al., 2012) (Wang et al., 2013), (Yang et al., 2017)  |
| CAM5_MAM4               | 1.875° × 2.5°                                   | 56 up to 40 Km               | GCM (nudged)            | MERRA  | (Neale et al., 2012) (Wang et al., 2013), (Yang et al., 2017)  |
| CAM5.3-Oslo             | 0.938° × 1.25°                                  | 30 up to 3.6 hPa             | GCM (nudg)              | ERA-interim U, V and PS (6h)                 | (Kirkevåg et al., 2018); (Karset et al., 2018)   |
| ECHAM5.5-HAM2-ELVOC_UH  | 1.875° × 1.875°                                 | 31 (hybrid sigma)            | GCM (nudg)              | ERA-Interim, U, V, PS                        | (Zhang et al., 2012); (Jokinen et al., 2015)   |
| ECHAM6-HAM2             | 1.875° × 1.875°                                 | 31 (hybrid sigma) to 10 hPa  | GCM (nudg)              | ERA-interim DIV (48h), VOR (6h) and PS (24h) | (Stevens et al., 2013), (Tegen et al., 2018), (Lohman and Neubauer, 2018)                                  |
| ECHAM6-HAM2-AP          | 1.875° × 1.875°                                 | 31 (hybrid sigma) to 10hPa   | GCM (nudg)              | ERA-interim DIV (48h), VOR (6h) and PS (24h) | (Stevens et al., 2013); (Tegen et al., 2018); (Neubauer et al., 2018)                                      |
| EMAC                    | 1.875° × 1.875°                                 | 31 (hybrid sigma)            | GCM (nudg)              | ERA-interim DIV (48h), VOR (6h) and PS (24h) | (Jöckel et al., 2010); (Karydis et al., 2017)  |
| GEOS-Chem-APM           | 2° × 2.5°                                       | 38                           | CTM                     | MERRA2                                       | (Yu and Luo, 2009), (Yu, 2011)   |
| GEOS-Chem-TOMAS         | 4.0° × 5.0°                                     | 25                           | CTM                     | MERRA  | (Bey et al., 2001) ( <a href="http://acmg.seas.harvard.edu/geos/">http://acmg.seas.harvard.edu/geos/</a> ) |
| GISS-E2.1-MATRIX        | 2° × 2.5°                                       | 40                           | GCM (nudg)              | NCEP   | (Bauer et al., 2008)   |
| GISS-E2-Tomas           | 2° × 2.5°                                       | 40                           | GISS GCM (free-running) | Online                                       | (Lee et al., 2015)   |
| TM4-ECPL and TM4-ECPL/v | 2° × 3°   | 34 (hybrid sigma) to 0.1 hPa | TM4, CTM                | ECMWF interim ERA-                           | (Kanakidou et al., 2012), (Daskalakis et al., 2015)  |
| TM5                     | 2° × 3°   | 34 (hybrid sigma) to 0.1 hPa | TM5, CTM                | ECMWF Interim ERA-                           | (Van Noije et al., 2014), (Huijnen et al., 2010); (Bergman et al., 2018, in prep.)                         |

**Table S2.** Details about the microphysics schemes employed by the models.

| MODEL                  | Microphysics Scheme                                       | Scheme Type  | Number of bins or modes                                | Reference  | Aerosol components  | Nucleation Scheme   |
|------------------------|---|--|--|--|---|---|
| CAM5-chem-APM          | APM   | Sectional (2m)   | 40   | To be published  | SO4, NO3, NH4, SOA, SS, DU, BC, POA   | Ternary ion-mediated nucleation (Yu et al., 2018)   |
| CAM5-chem-ATRAS2       | ATRAS2  | Sectional (2m)   | 12 (47 in maximum)                                     | (Matsui, 2017) (Matsui and Mahowald, 2017)   | SO4, NO3, NH4, OA (POA, SOA), SS, DU, BC  | (Kulmala et al., 2006) within the planetary boundary layer (Vehkamäki et al. 2002) within the free troposphere  |
| CAM5_MAM3              | MAM3  | Modal (2m)   | 3  | (Liu et al., 2012)   | SO4, BC, DU, SS, OA (POA+SOA)   | (Vehkamäki, 2002)   |
| CAM5_MAM4              | MAM4  | Modal (2m)   | 4  | (Liu et al., 2012) (Liu et al., 2016)  | SO4, BC, DU, SS, POA+SOA  | (Vehkamäki, 2002) . The boundary layer parameterization, (only applied in PBL), uses the empirical first order (in H <sub>2</sub> SO <sub>4</sub> ) nucleation rate from (Sihto et al., 2006) |
| CAM5.3-Oslo            | OsloAero5.3 with look-up tables from AeroTab5.3 (offline) | Production-tagged (linked to an offline 2m Sectional scheme) | 44 (offline)   | {Kirkevåg et al., 2018}  | SO4, BC, DU, SS, OA (POA+SOA), distributed over 12 external and internal mixtures (size-modes)  | (Vehkamäki, 2002), (Paasonen et al., 2010)  |
| ECHAM5.5-HAM2-ELVOC_UH | HAM2  | Modal (2m)   | 7  | (Zhang et al., 2012)   | SO4, BC, DU, SS, OA (POA+SOA) SOA as in (Joniken et al., 2015)  | (Vehkamäki, 2002) (Paasonen et al., 2010)   |
| ECHAM6-HAM2            | HAM2  | Modal (2m)   | 7  | (Tegen et al. 2018); (Neubauer et al, 2018), (Zhang et al., 2012)                    | SO4, BC, DU, SS, OA (POA+SOA) SOA: approximated as 15% of monoterpene emissions at the surface. SOA is assumed to condense immediately on existing aerosol particles and to have identical properties to primary organic aerosols   | (Kazil et al., 2010);   |
| ECHAM6-HAM2-AP         | AP  | Modal (2m)   | 7+2  | (Tegen et al., 2018) ; (Neubauer et al., 2014) based on (Hoose et al., 2008a, 2008b) | SO4, BC, DU, SS, OA (POA+SOA) SOA: as in ECHAM6-HAM2  | (Kazil and Lovejoy, 2007); (Kazil et al., 2010); (Kulmala et al., 2006)   |
| EMAC                   | GMXe  | Modal (2m)   | 7  | (Pringle et al., 2010a) (Pringle et al., 2010b)                                      | Thermodynamic equilibrium using ISORROPIA-II (Fountoukis and Nenes, 2007) Species considered: K <sup>+</sup> -Ca <sup>2+</sup> -Mg <sup>2+</sup> -NH <sub>4</sub> <sup>+</sup> -Na <sup>+</sup> -SO <sub>4</sub> <sup>-2</sup> -NO <sub>3</sub> <sup>-</sup> -Cl <sup>-</sup> -H <sub>2</sub> O-BC-OA-DU-SS | (Vehkamäki, 2002)   |
| GEOS-Chem-APM          | APM   | Sectional (1m)   | 40 bins for inorganic aerosols & 2 modes for BC and OA | (Yu and Luo, 2009)   | SO4, NO3, NH4, SOA, SS, DU, BC, POA   | Ternary ion-mediated nucleation (Yu et al., 2018)   |
| GEOS-Chem-TOMAS        | TOMAS   | Sectional (2m)   | 15   | (Trivittayanurak et al., 2008), (Kodros et al.,                                      | SO4, NH4, BC, DU, SS, POA, SOA  | Ternary (+ammonia) (Napari et al., 2002) scaled by 1E-5 If no ammonia   |

|                         |        |                |                  |                        |  |   |
|-------------------------|--------|----------------|------------------|------------------------|--|---|
|                         |        |                |                  | 2016)                  |  | (Vehkamäki, 2002)   |
| <b>GISS-E2.1-MATRIX</b> | MATRIX | Modal (2m)     | 16 mixing states | (Bauer et al., 2008)   | SO <sub>4</sub> , NO <sub>3</sub> , NH <sub>4</sub> , H <sub>2</sub> O, OA, BC, DU, SS   | NAPARI ternary nucleation scheme (Napari et al., 2002)  |
| <b>GISS-E2-Tomas</b>    | TOMAS  | Sectional (2m) | 15               | (Lee et al., 2015)     | SO <sub>4</sub> , BC (hydrophilic), BC (hydrophobic), OC (hydrophilic), OC (hydrophobic), DU, SS, NH <sub>4</sub> , water  | Binary (Vehkamäki, 2002)  |
| <b>TM4-ECPL</b>         | M7     | Modal (2m)     | 7                | (Vignati et al., 2004) | SO <sub>4</sub> , BC, DU, SS, OA (POA+SOA)<br>SOA: Produced from oxidation of pinene, isoprene and aromatics (xylene, benzene and toluene) with O <sub>3</sub> , OH, NO <sub>3</sub> . | Binary (Vehkamäki, 2002) with (Kokkola et al., 2009) scheme for the treatment of the sulfuric acid gas      |
| <b>TM5</b>              | M7     | Modal (2m)     | 7                | (Vignati et al., 2004) | SO <sub>4</sub> , BC, DU, SS, POA, SOA, NO <sub>3</sub> , NH <sub>4</sub> , MSA  | Organic+ sulfuric acid nucleation (Riccobono et al., 2014); binary homogeneous nucleation (Vehkamäki, 2002) |



**Table S3.** Emission inventories and schemes used in the models Anthropogenic emissions are given for the year 2011. For interactive emissions, the references are for the model used to calculate the emissions.

| Model                             | SO <sub>x</sub> (SO <sub>2</sub> +SO <sub>4</sub> )<br>SO <sub>x</sub> (total) Tg a <sup>-1</sup> |                           | SO <sub>4</sub><br>(emit-<br>ted) Tg<br>a <sup>-1</sup>  | BC (Tg a <sup>-1</sup> )                              |      | POA (Tg a <sup>-1</sup> )                             |                         | VOC (Tg a <sup>-1</sup> )  |  | SS (Tg a <sup>-1</sup> )  |                            | DUST (Tg a <sup>-1</sup> )   |                            |
|-----------------------------------|---|---------------------------|--|---|------|---|-------------------------|--|--|---|----------------------------|--|----------------------------|
| <b>CAM5-<br/>chem-<br/>APM</b>    | (Keller et al., 2014)   | 55 Tg S a <sup>-1</sup>   | 0  | (Keller et al., 2014)                                 | 7    | (Keller et al., 2014)                                 | 41 TgC a <sup>-1</sup>  | (Lamarque et al., 2012)  | Isoprene & monoterpenes 500 TgC a <sup>-1</sup>                            | (Gong, 2003)  | 11100                      | (Mahowald et al., 2006)  | 3620                       |
| <b>CAM5-<br/>chem-<br/>ATRAS2</b> | (Lamarque et al., 2010)   | 64.8                      | 1.66   | (Lamarque et al., 2010)                               | 7.8  | (Lamarque et al., 2010)                               | 50.3                    | NMVOC derived from MOZART emissions for year 2000.                                     | 115 (SOA)  | Calculated on line (Mårtensson et al., 2003) (Monahan et al., 1986)   | 5039 (wind dependent flux) | Calculated online (Zender et al., 2003) (Albani et al., 2014)      | 2677 (wind dependent flux) |
| <b>CAM5-<br/>MAM3</b>             | CMIP6 (Hoesly et al., 2017), (van Marle et al., 2017)   | 70.17                     | 1.75   | CMIP6 (Hoesly et al., 2017), (van Marle et al., 2017) | 9.32 | CMIP6 (Hoesly et al., 2017), (van Marle et al., 2017) | 44.66                   | CMIP6 (Hoesly et al., 2017), (van Marle et al., 2017) MOZART-2 (Horowitz et al., 2003) | 75.36 (SOAg)   | Calculated on line (Mårtensson et al., 2003) (Monahan et al., 1986)   | 2581.4                     | Calculated online (Zender et al., 2003)                            | 3117.7                     |
| <b>CAM5-<br/>MAM4</b>             | SO <sub>2</sub> : ACCMIP (Lamarque et al., 2013)<br>SO <sub>4</sub> : 2.5% of SO <sub>2</sub>     | 64.8                      | 1.62   | ACCMIP (Lamarque et al., 2013)                        | 7.76 | ACCMIP (Lamarque et al., 2013)                        | 50.2                    | NMVOC derived from MOZART emissions for year 2000.                                     | SOA created=103.3  | Calculated on line (Mårtensson et al., 2003) for aerosols with geometric diameter < 2.8 µm (Monahan et al., 1986) for aerosols with geometric diameter > 2.8 µm | Wind dependent flux        | Online (Scanza et al., 2015), (Albani et al., 2014)                | Wind dependent flux        |
| <b>CAM5.3-<br/>Oslo</b>           | (Lamarque et al., 2010)   | 66.70                     | 1.67   | (Lamarque et al., 2010)                               | 7.93 | Kirkevåg et al., (2018)                               | 87.03                   | (Guenther et al., 2006)  | 499.5 (isoprene and monoterpene only, i.e. not including MSA contribution) | Calculated online (Salter et al., 2015; Kirkevåg et al., 2018))   | 1956                       | Calculated online (Zender et al., 2003)                            | 3117                       |
| <b>ECHAM6-<br/>HAM2</b>           | (Lamarque et al., 2010)   | 69.7 Tg-S a <sup>-1</sup> | 1.7 Tg S a <sup>-1</sup> (only direct emitted SO <sub>4</sub> ; SO <sub>4</sub> produced in clouds | (Lamarque et al., 2010)                               | 8.0  | (Lamarque et al., 2010)                               | 66.7 Tg a <sup>-1</sup> | (Dentener et al., 2006)  | Included in POA  | (Long et al., 2011); (Sofiev et al., 2011)  | Wind dependent flux        | (Tegen et al., 2002); (Cheng et al., 2008); (Heinold et al., 2016) | Wind dependent flux        |

|                               |  |                           |   |  |      |   |  |   |  |   |                     |   |                     |
|-------------------------------|--|---------------------------|---|--|------|---|--|---|--|---|---------------------|---|---------------------|
|                               |  |                           | by wet chemistry: 51.9 Tg S a <sup>-1</sup> is not included)  |  |      |   |  |   |  |   |                     |   |                     |
| <b>ECHAM5.5-HAM2-ELVOC_UH</b> | (Lamarque et al., 2010)  | 53.9 Tg S a <sup>-1</sup> | 1.3 Tg S a <sup>-1</sup>  | (Lamarque et al., 2010)  | 7.24 | (Lamarque et al., 2010)   | 49.3 Tg C a <sup>-1</sup>                              | (Guenther et al., 2006)   | 27   | (Schultz et al., 2002)  | 5032                | (Tegen et al., 2002), (Cheng et al., 2008)  | 931                 |
| <b>ECHAM6-HAM2-AP</b>         | (Lamarque et al., 2010)  | 70.1 Tg S a <sup>-1</sup> | 1.7 Tg S a <sup>-1</sup> (only direct emitted SO <sub>4</sub> ; SO <sub>4</sub> produced in clouds by wet chemistry: 51.9 Tg S a <sup>-1</sup> is not included) | (Lamarque et al., 2010)  | 8.1  | (Lamarque et al., 2010)   | 47.9 Tg C a <sup>-1</sup> = 67.0 Tg OA a <sup>-1</sup> | (Dentener et al., 2006)   | Included in POA  | Calculated online (Long et al., 2011); (Sofiev et al., 2011)  | 1161                | Calculated online (Cheng et al., 2008); (Heinold et al., 2016)  | 926                 |
| <b>EMAC</b>                   | RCP 8.5 (Riahi et al., 2007)   | 63.6 Tg S a <sup>-1</sup> | 1.6 Tg S a <sup>-1</sup>  | RCP 8.5 (Riahi et al., 2007)   | 6.7  | RCP 8.5 (Riahi et al., 2007)  | 41.1 Tg C a <sup>-1</sup>                              | RCP 8.5 (Riahi et al., 2007)  | Included in POA  | Total Sea salt: (Dentener et al., 2006) chemical Composition of sea salt: (Seinfeld and Pandis, 2006) | 7889                | Total dust emissions: (Astitha et al., 2012) Chemical composition of dust: (Karydis et al., 2016); (Klingmüller et al., 2018) | 4760                |
| <b>GEOS-Chem-APM</b>          | (Keller et al., 2014)  | 55 Tg S a <sup>-1</sup>   | 0   | (Keller et al., 2014)  | 7    | (Keller et al., 2014)   | 41 Tg C a <sup>-1</sup>                                | (Keller et al., 2014)   | Isoprene & monoterpenes 760 TgC a <sup>-1</sup>  | (Gong, 2003)  | 10800               | Calculated online (Zender et al., 2003) (Mahowald et al., 2006)   | 1180                |
| <b>GEOS-Chem-TOMAS</b>        | Mixture of global and regional inventories (described in Kodros et al., 2016; Keller et al., 2014) | 395.36                    | 0.27 Tg S a <sup>-1</sup>   | Anthropogenic: Bond et al. (2007) Biomass burning: (Wiedinmyer et al., 2011) | 6.97 | Anthropogenic: (Bond et al. 2007) Biomass burning: (Wiedinmyer et al. 2011) | 44.72  | Biogenic VOC emissions from (Guenther et al., 2006), Anthropogenically-influenced VOCs from (D'Andrea et al., 2013) | Biogenic SOA created=19 Tg C a <sup>-1</sup> Anthropogenically-enhanced SOA created=100 Tg a <sup>-1</sup> | Calculated online (Jaeglé et al., 2011)   | Wind dependent flux | Calculated online (Zender et al., 2003)   | Wind dependent flux |

|                                |  |                          |                         |   |      |   |                            |  |  |   |         |  |         |
|--------------------------------|--|--------------------------|-------------------------|---|------|---|----------------------------|--|--|---|---------|--|---------|
|                                |  |                          |                         |   |      |   |                            |  | 1  |   |         |  |         |
| <b>GISS-E2.1-MATRIX</b>        | (Lamarque et al., 2010)  | 191.82(SO <sub>2</sub> ) | 5.27                    | (Lamarque et al., 2010)                               | 9.35 | (Lamarque et al., 2010)                               | 61.73                      | Isoprene: (Guenther et al., 1995).<br>Monoterpenes and ORVOC: (Lathiere et al., 2005)                                  | Isoprene: 524<br>Terpenes + ORVOC: 193<br>SOA production 17.1                    | (Schmidt et al., 2014)  | 2662.69 | (Schmidt et al., 2014)                 | 1275.60 |
| <b>GISS-E2-Tomas</b>           | 1 % of SO <sub>2</sub> ACCMIP (Lamarque et al., 2013)                                  | 65.6                     | 0.66                    | ACCMIP and GFEDv3 for biomass burning                 | 7.4  | ACCMIP and GFEDv3 for biomass burning                 | 43.7                       | ACCMIP (Lamarque et al., 2013)   | SOA production rate of 17  | (Gong, 2003)  | 3231.9  | (Ginoux et al., 2001)                  | 705.8   |
| <b>TM4-ECPL and TM4-ECPL/v</b> | 2.5% of SO <sub>2</sub> ACCMIP (Lamarque et al., 2013)                                 | 121.25(SO <sub>2</sub> ) | 4.66 (SO <sub>4</sub> ) | ACCMIP (Lamarque et al., 2013)                        | 8.02 | ACCMIP (Lamarque et al., 2013)                        | 35.12 Tg C a <sup>-1</sup> | Emissions of Biogenic VOCs are taken from (Sindelarova et al., 2014)<br>Anthropogenic VOC ACCMIP, Ocean emissions POET | Isoprene: 525<br>monoterpenes: 104.3<br>other VOC: 572.3<br>SOA production: 32.6 | (Vignati et al., 2010)  | 8521    | (Dentener et al., 2006)                | 1169.5  |
| <b>TM5</b>                     | CMIP6 (Hoesly et al., 2018); (van Marle et al., 2017); GEIA (Andres and Kasgnoc, 1998) | 62.04                    | 1.59                    | CMIP6 (Hoesly et al., 2018); (van Marle et al., 2017) | 9.49 | CMIP6 (Hoesly et al., 2018); (van Marle et al., 2017) | 52.02                      | CMIP6 (Hoesly et al., 2018); (van Marle et al., 2017); MEGAN-MACC (Sindelarova et al., 2014)                           | 109.32   | Calculated online (Gong et al., 2000); (Salisbury et al., 2013) | 5497.08 | Calculated online (Tegen et al., 2002) | 978.88  |

**Table S4.** Measurement sites included in this study. The classification and location of the stations are denoted, the supersaturation values at which CCN concentrations are measured at each site and the aerosol components for which mass is measured in the PM<sub>1</sub>. The followed notation is: CCN<sub>x</sub>: CCN at x % supersaturation, SO<sub>4</sub>: sulfate, OA: organic aerosols, SS: sea salt, NO<sub>3</sub>: Nitrates, NH<sub>4</sub>: ammonium. Details on observations, methods and data coverage can be found in Schmale et al. (2017).

| Station name   | Location/type   | Geoposition              | Observations   |
|----------------|---|--------------------------|--|
| Cabauw         | Cabauw, The Netherlands near coast, rural-background        | 51°58'N, 04°56'E, -1 m   | CCN <sub>0.1</sub> , CCN <sub>0.2</sub> , CCN <sub>0.3</sub> , CCN <sub>0.5</sub> , CCN <sub>1.0</sub> , SO <sub>4</sub> , OA, SS, NH <sub>4</sub> , NO <sub>3</sub>   |
| Finokalia      | Finokalia, Crete, Greece, coastal background, Mediterranean | 35°20'N, 25°40'E, 260 m  | CCN <sub>0.2</sub> , CCN <sub>0.4</sub> , CCN <sub>0.6</sub> , CCN <sub>0.8</sub> , CCN <sub>1.0</sub> , SO <sub>4</sub> , OA, SS, NH <sub>4</sub> , NO <sub>3</sub>   |
| Jungfraujoch   | Jungfraujoch, high alpine, background Switzerland           | 46°33'N, 07°59'E, 3580 m | CCN <sub>0.1</sub> , CCN <sub>0.15</sub> , CCN <sub>0.2</sub> , CCN <sub>0.25</sub> , CCN <sub>0.3</sub> , CCN <sub>0.35</sub> , CCN <sub>0.4</sub> , CCN <sub>0.5</sub> , CCN <sub>0.7</sub> , CCN <sub>1.0</sub> , SO <sub>4</sub> , OA, NH <sub>4</sub> , NO <sub>3</sub> |
| Mace Head      | Mace Head, Ireland, coastal background                      | 53°19'N, -9°54'E, 5 m    | CCN <sub>0.1</sub> , CCN <sub>0.25</sub> , CCN <sub>0.35</sub> , CCN <sub>0.5</sub> , CCN <sub>0.75</sub> , CCN <sub>1.0</sub> , SO <sub>4</sub> , OA, SS, NH <sub>4</sub> , NO <sub>3</sub>   |
| Melpitz        | Melpitz, Germany, continental, background                   | 51°32'N, 12°56'E, 89 m   | CCN <sub>0.1</sub> , CCN <sub>0.2</sub> , CCN <sub>0.3</sub> , CCN <sub>0.5</sub> , CCN <sub>0.7</sub> , SO <sub>4</sub> , OA, SS, NH <sub>4</sub> , NO <sub>3</sub>   |
| Noto Peninsula | Noto Peninsula, Japan, coastal background                   | 37°45'N, 137°36'E, 0 m   | CCN <sub>0.1</sub> , CCN <sub>0.2</sub> , CCN <sub>0.5</sub> , CCN <sub>0.8</sub>  |
| Puy de Dôme    | Puy de Dôme, France, mountain continental background        | 45°46'N, 2°57'E, 1465 m  | CCN <sub>0.2</sub>   |
| Hyytiälä       | Hyytiälä, Finland, background boreal forest rural           | 61°51'N, 24°17'E, 181 m  | CCN <sub>0.1</sub> , CCN <sub>0.2</sub> , CCN <sub>0.3</sub> , CCN <sub>0.5</sub> , CCN <sub>1.0</sub> , SO <sub>4</sub> , OA, SS, NH <sub>4</sub> , NO <sub>3</sub>   |
| Vavihill       | Vavihill, Sweden, background rural                          | 56°01'N, 13°09'E, 172 m  | CCN <sub>0.1</sub> , CCN <sub>0.15</sub> , CCN <sub>0.2</sub> , CCN <sub>0.25</sub> , CCN <sub>0.3</sub> , CCN <sub>0.35</sub> , CCN <sub>0.4</sub> , CCN <sub>0.5</sub> , CCN <sub>0.7</sub> , CCN <sub>1.0</sub> , CCN <sub>1.4</sub>                                      |

## Appendix S2

### S2.1. Particle numbers, CCN calculations

- 5 Number concentrations of particles of various sizes ( $N_{50}$ ,  $N_{80}$  and  $N_{120}$ ) are computed by 14 models for all observational sites. For the modal models, where lognormal distributions are used to describe internally or externally mixed particle populations, the total number of particles with dry diameters larger than a threshold diameter  $D_0$  is given by

$$N(D > D_0) = \sum_{i=1}^k \frac{N_0^i}{2} \left( 1 - \operatorname{erf} \left( \frac{\ln(D_0/\tilde{D}_n^i)}{\sqrt{2}} \right) \right) \quad (1)$$

- where  $k$  is the number of modes,  $N_0^i$  and  $\tilde{D}_n^i$  are the number of particles and the particle number median diameter of mode  $i$ ,  
 10 respectively, while  $\operatorname{erf}$  is the error function. In several models calculation of the CCN number concentration is based on the  $\kappa$ -Köhler theory as developed by (Petters et al, 2007), according to which, for a given supersaturation  $s$ , the critical dry diameter  $D_d$  of activated particles is given by

$$s(D) = \frac{D^3 - D_d^3}{D^3 - D_d^3(1 - \kappa)} \exp \left( \frac{4\sigma_{s/a}M_w}{RT\rho_w} \right) \quad (2)$$

- Here  $D$  and  $D_d$  are the wet and dry diameters of particles, respectively,  $\sigma_{s/a}$  is the surface tension of the solution/air interface,  
 15 which is taken equal to  $0.072 \text{ J m}^{-2}$ ,  $\rho_w$  is the density of water,  $M_w$  is the molecular weight of water,  $R$  is the ideal gas constant and  $T$  is the temperature. After having determined the critical dry diameter from Eq. (2), the number of CCN is calculated from Eq. (1) as the number of particles with dry diameters larger than this critical value.

- The hygroscopicity,  $\kappa$ , for the particle as a whole, or just for the coating layer if it is sufficiently thick for models that take into account coated particles, is determined based on the volume weighted hygroscopicities of the individual components,  $\kappa_j$ ,  
 20 where  $\varepsilon_i$  is the fraction of the dry aerosol volume occupied by the individual components,  $j$ .

$$\kappa = \sum_j \varepsilon_j \kappa_j \quad (3)$$

In the present study, different hygroscopicity parameters are used in each model as summarized in Table 1

### S2.2. Persistence – autocorrelation function

- According to the mathematical definition for a stationary time series of values  $Y_1, Y_2, \dots, Y_N$  at the time instances  $t_1, t_2, \dots, t_N$ ,  
 25 the autocorrelation function (ACF) at time lag  $k$  is given by

$$r_k = \frac{\sum_{i=1}^{N-k+1} (Y_i - \bar{Y})(Y_{i+k} - \bar{Y})}{\sum_{i=1}^{N+1} (Y_i - \bar{Y})^2} \quad (4)$$

where  $\bar{Y}$  is the average value of  $Y_i$ . The large-lag standard error is computed as the square root of the variance given by:

$$Var(r_k) = \frac{1}{N} \left( 1 + 2 \sum_{i \approx 1}^k r_i^2 \right) \quad (5)$$

where  $r_i$  is the ACF at time  $i$ . The persistence time can then be defined as the time lag at which the ACF crosses the curve of the large-lag standard error, i.e. when the ACF is significant compared to the standard error (Schmale et al., 2018).

## 5 Appendix S3. Results

### S3.1 CCN persistence- Individual model's behavior and sensitivity to size of the emitted particles

It is worth noting here that the number concentration of CCN is not directly related to the persistence time. For example, based on the observational data at the Melpitz station, the persistence time of CCN<sub>0.2</sub> during winter is 3.5 times longer than during summer (7 days and 2 days respectively; Fig. 7), although the CCN<sub>0.2</sub> number concentration during winter (~1500 cm<sup>-3</sup>) is only 25% higher than during summer (1200 cm<sup>-3</sup>) (Fig. 3). This is because the ACF and the subsequently computed persistence are related to the rate of change of the CCN number concentration due to the various formation, transport and removal processes, and not to the absolute level of CCN. As such, when the CCN population remains almost unchanged for a long time period, the persistence is high, independent of the CCN concentrations.

For the Vavihill station, in contrast to the observations, all models (except EMAC) calculate somewhat longer persistence during winter. However, the observed ratio at this station has been assumed to reflect a peculiarity of the observation period during 2013 and 2014 (Schmale et al., 2018). The same behavior is also observed at the Noto Peninsula station. Most of the models, in agreement with observations, simulate persistence of ~ 2 days during summer, but none of them is able to simulate the longer persistence observed during winter.

For the remaining stations, the comparisons show similar performances for all models in terms of the winter/summer persistence. For Melpitz, Cabauw and Hyytiälä, longer persistence is observed during winter and can be ascribed to the more stable weather conditions during this season. For these stations, it appears that only four out of the 14 models (GEOS-Chem-APM, CAM5-MAM3, CAM5-MAM4, and CAM5-chem-ATRAS2) correctly simulate the winter/summer relative order of persistence, although the model persistence is smaller than the observationally derived values. These models also all use MERRA, rather than ERA which is used in the other models. At Finokalia, the longer persistence observed during summer is attributed to the more stable weather conditions during summer. This behavior is reproduced by all models but five (CAM5-Chem-APM, CAM5.3-Oslo, EMAC, GISS2.1-MATRIX, TM4-ECPL).

Analyzing the reasons that affect the persistence and then attributing the differences between the observed and the model-derived values to the underlying physical and/or chemical process parameterizations in each model is a demanding

task which most probably is model dependent. Here a first attempt is made by examining the sensitivity of the persistence to the size of the emitted OA and BC particles. For that, we first focus on the Finokalia station and we examine the results obtained from two different simulations using the TM4-ECPL model. The setups of the two simulations are almost identical differing only in the size of the emitted OA and BC particles (details in the caption of Figure S5). Since the total OA and BC emitted masses are kept constant, changing the emitted particle sizes affects the number of emitted particles. Overall, in the new sensitivity simulation larger but fewer particles are emitted compared to the base simulation. The  $CCN_{0.2}$  persistences computed from the results of the two simulations are compared with the observations in Figure S5a. In the sensitivity simulation (green bars, larger particles) an increase of the persistence during summer by a factor  $> 2$  is seen compared to the base simulation (red bars). This increase leads to an inversion of the winter/summer ratio of persistence since during winter the persistence time remained almost unchanged. The results of the sensitivity simulation are in better agreement with observations and demonstrate the importance of the size distribution of the emitted particles for the CCN simulations and also suggest that that size is probably underestimated in the base case simulation of TM4-ECPL.

In order to identify the factors that caused this change in the relative order of the persistence at Finokalia, we have calculated the ACF of the  $CCN_{0.2}$  number concentrations (Figures S5b, S5c). The black lines show the observational data during summer (solid line) and winter (dashed line) and the red lines show the results of the base simulation with TM4-ECPL, obtained for the same time-periods with the observations (Fig. S5b), while the green lines show the results of the sensitivity simulation (Fig. S5c). It is seen that for short time-periods ( $< 15$  h) both the observational and model derived ACF (denoted as  $ACF^{obs}$  and  $ACF^{model}$ , respectively) smoothly decrease. Since the  $ACF^{model} > ACF^{obs}$  during that time ( $t < 15$  h) the simulated CCN population varies less than the observed one. The fact that both  $ACF^{model}$  in the base simulation and  $ACF^{obs}$  vary less during winter than during summer, suggests that both model- and observationally-computed persistence are higher in winter. However, during summer, after the first  $\sim 24$  hours a local maximum is present in both  $ACF^{model}$  and  $ACF^{obs}$  that shows a periodicity of 24 hours. This peak indicates a daily periodic phenomenon and can be attributed to the almost constant weather conditions during summer that drive a diurnal cycle in the photochemical formation of  $H_2SO_4$  in the gas phase in the Finokalia region (Mihalopoulos et al., 2007). During summer a significant difference between the  $ACF^{model}$  from the base simulations and  $ACF^{obs}$  is found at longer correlation times ( $t > 15$  h), when the  $ACF^{obs}$  decays much slower than the  $ACF^{model}$  leading to smaller observed than modeled persistence. This can be attributed to the different production/loss rates ratio of the  $CCN_{0.2}$  particles. Possible explanations are related to the aerosol aging, wet deposition and sedimentation removal, long-range transport, or model resolution that affect the lifetime of aerosols in the atmosphere.

Figure S5c depicts the ACFs of the sensitivity simulation (green lines) and the observations. Comparing the results of the two simulations (panels b and c), it is seen that during winter the two  $ACF^{model}$  are similar, while large differences are found between the two  $ACF^{model}$  during summer. This is due to the slower decay of  $ACF^{model}$  in the sensitivity simulation leading to a higher persistence time, similar to the observed one. For Cabauw station (Fig. S5d) that is affected by fresh primary emissions, the sensitivity simulation shows better agreement in persistence time with the observationally derived values; while for Mace Head station (Fig. S5f) that is received air masses transported over the ocean the two simulations

provide similar results. For Hyytiälä (Fig. S5e), the adopted change in the size of emitted particles is not sufficient to reconcile model-derived and observationally-derived persistence times, indicating that other factors might be also important for this discrepancy.

### 5 S3.2. Statistics for the comparisons between model results and observations

The following statistical variables have been computed here to quantify the agreement of model results with the observations.

Mean Error (ME)

$$ME = \frac{1}{N} \sum_{i=1}^N (P_i - O_i)$$

10

Normalized Mean Bias (NMB)

$$NMB = \frac{\sum_{i=1}^N (P_i - O_i)}{\sum_{i=1}^N O_i} \times 100\%$$

Normalized Mean Error (NME)

$$NME = \frac{\sum_{i=1}^N |P_i - O_i|}{\sum_{i=1}^N O_i} \times 100\%$$

Root Mean Square Error (RMSE)

$$RMSE = \sqrt{\frac{1}{N} \sum_{i=1}^N (P_i - O_i)^2}$$

Index of agreement

$$IOA = 1 - \frac{\sum_{i=1}^N (P_i - O_i)^2}{\sum_{i=1}^N (|O_i - \bar{O}| + |P_i - \bar{O}|)}$$

15 Mean absolute Error (MAE)

$$MAE = \frac{1}{N} \sum_{i=1}^N |P_i - O_i|$$

The results are shown in Table S5 (separate file).

### S3.3. Surface PM<sub>1</sub> composition - Global distributions

The global surface distributions of the multi-model mean of the various chemical compounds (SO<sub>4</sub>, BC, OA, SS and DU) concentrations that contribute to PM<sub>1</sub> as the median of the 15 models are shown in supplementary Figure S15 (left columns)



together with the corresponding model diversities (right column); while Figures S6-S14 show the results of individual models. Note that all simulated PM<sub>1</sub> component diversities maximize south of 60°S and north of 60°N, which reflects the challenges of the models in simulating atmospheric circulation and chemistry close to the poles. Since sulfate is formed by oxidation of sulfur dioxide (SO<sub>2</sub>), which is mainly produced from anthropogenic activities, high concentrations of sulfate are observed in industrialized regions. In Eastern China, where the highest annual concentration of SO<sub>4</sub> is computed, the concentration reaches 12.5 µg m<sup>-3</sup>, in India 7.0 µg m<sup>-3</sup>, around the Mediterranean basin 5.0 µg m<sup>-3</sup> and in the eastern United States 3.0 µg m<sup>-3</sup>. In marine regions, where SO<sub>2</sub> is formed predominantly through oxidation of dimethyl sulfide (DMS), the sulfate concentrations are significantly lower and do not exceed 1.0 µg.m<sup>-3</sup> in the Northern Hemisphere and 0.6 µg m<sup>-3</sup> in the Southern Hemisphere. The highest diversities are calculated for the high latitude oceanic regions in both hemispheres. In continental regions the highest diversities of 5 to 10 are found far from the main SO<sub>2</sub> production sources, in South America, Central Africa and Indonesia. Mann et al. (2014) pointed out that the model diversities are higher in northern Europe than in southern Europe, while in Figure 12b the opposite is seen for the present study.

BC is entirely produced by anthropogenic combustion of fossil fuel, and biomass, and wild-fires and volcanoes (Bycenkiene et al., 2013). Therefore, BC is observed in high amounts over continents and in significantly lower amounts over the ocean. Mass concentrations reach a maximum in Eastern China (~4.5 µgC m<sup>-3</sup>) and in India (~2.0 µgC m<sup>-3</sup>), while in other continental regions the annual average concentrations do not exceed 1.0 µg m<sup>-3</sup> and over the ocean 0.1 µg m<sup>-3</sup>. Besides polar and high-latitude regions, high diversities are also observed over the ocean and in continental regions away from the sources. Model differences can be attributed to the emissions used in the models, which differ in strength, spatial distribution, and the assumed size of the emitted particles, as well as to the removal processes, such as wet and dry deposition. Indeed, high diversity is computed over biomass burning regions in tropical South America, Africa and Indonesia but also in Saudi Arabia where oil extraction plants exist. Differences in the assumed organic mass to organic carbon ratio in the models could explain some of this model diversity. High diversities over the tropical oceans in the outflow of biomass burning regions are also associated with very low BC concentrations, differences in the transport and in the deposition patterns. The highest diversities are, however, computed close to the poles where BC levels are very low. These results are in agreement with Mann et al. (2014) who attributed model differences mainly to removal processes.

Modeling OA is more challenging than SO<sub>4</sub> and BC, since it involves the treatment of both primary emissions as well as the secondary formation of organics from precursor gases. Model diversities are largest over the tropical oceans as well as the southern oceans suggesting that differences in the marine source of OA (or lack of such source) are the major contributors to this diversity. Similar to BC, smaller diversities are found over and downwind of biomass burning regions and in the Arabian Peninsula where oil combustion emissions prevail. Tsigaridis et al. (2014) also revealed large differences between the models in the organics source strength and formation of SOA, which reached almost an order of magnitude. That study also investigated the models' differences in the removal of OA and found large inter-model differences especially in the strength of the wet deposition. They also found that the diversity between the models has maxima over oceanic regions south of 30°S, part of this diversity can be attributed to the marine source representation of OA in the models. In the present

study, the model diversity is slightly higher than in the earlier study over these regions, while the global surface distribution of OA shows a similar pattern.

The SS concentration (Fig. S15g), which is driven by wind speed and temperature, has maxima over the Southern Ocean. It also reveals some advection of SS over land. However, due to differences in the source parameterization and strength as well as in the deposition, large diversities ( $>10$ ) between models (Fig. S15h) are computed for the Southern Ocean as well as at the Polar regions. Similarly high diversity is found over the Caspian Sea, which most probably is not properly resolved by many models, as well as over the northern part of South America where sea salt concentrations are low resulting from little inland transport of SS.

In line with these remarks, submicron dust aerosol (Fig. 15i), which is a minor contributor to the total dust emissions, has maxima over source regions with the highest concentrations over the Sahara and Gobi deserts. However, emissions from South African, Australian, Patagonia, Arizona and Great Basin deserts are also clearly seen in the DU distribution. The largest model diversities (Fig. S10j) are found to be associated with long-range transport and deposition towards the poles and in the tropical Pacific, as well as with emissions from the Patagonia desert, which seem not to be well represented in all models.

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## Supplementary figures captions

The following figures can be found in a separate file:

**Fig. S1.** Monthly averages of the mass concentration of sulfate ( $\text{SO}_4$ ), organics (OA), dust (DU) and sea-salt (SS) of  $\text{PM}_{10}$  particles for the period 2011-2015. The median of all models is shown with blue bold line, while the shaded areas depict the 25%/75 % quartiles of all data. The green-dashed lines show the min/max values of all models. Available observational data are also shown with black dashed-lines and dots. Observations are from Schmale et al. (2017). Note that SS is here derived from  $\text{Cl}^-$   $\text{PM}_{10}$  measurements that underestimate the SS levels.

**Fig. S2.** Comparisons of the monthly mean observations of CCN at various supersaturation ratios,  $N_{50}$ ,  $N_{80}$ ,  $N_{120}$  particle number concentrations, and sulfate ( $\text{SO}_4$ ) and organic aerosol (OA) mass concentrations with the corresponding results of each model. Models are plotted with different colours as indicated in the figure legend. Observations (black circles) are from Schmale et al. (2017).

**Fig. S3.** Comparison between monthly averages of the cloud droplet properties from observations (black circles) and from the individual models (see figure legend for model identification). Figures are drawn per station and for 2 different updraft velocities ( $w=0.3 \text{ ms}^{-1}$  and  $w=0.6 \text{ ms}^{-1}$ ) marked on the y-axis. For each station and updraft velocity the four graphs show (as indicated in the y-axis label), the number of cloud droplets,  $N_d$ , the maximum supersaturation,  $s_{\text{max}}$  (in %), the sensitivity of the  $N_d$  to the total number of aerosol particles,  $(\partial N_d / \partial N_a)$ , and the sensitivity of the  $N_d$  to the wind speed,  $(\partial N_d / \partial w)$ .

**Fig. S4.** Comparison between the persistence times derived from the observations (in black bars) and from the model results of  $\text{CCN}_{0.2}$  during winter and summer for each station. Each pair of bars that follows corresponds to the predictions of each model for winter (left bar) and summer (right shaded bar), respectively. The white bars show the persistence times of the MMM. The persistence times derived from model simulations have been computed at the same time periods as those derived from the observations.

**Fig. S5.** (a) Comparison of the winter and summer persistence times of  $\text{CCN}_{0.2}$  for Finokalia station calculated using observational data (black bars) and the results of TM4-ECPL model for the base simulation (red bars) and the sensitivity simulation assuming the same emission of carbonaceous aerosols with the base case but fewer particles of larger size (green bars). In both simulations a log-normal distribution for the size distribution of emitted particles with a geometric standard deviation  $\sigma=1.59$  has been assumed/used. In the base simulation, the dry median diameters of the emitted particles coming from fossil fuel combustion and from vegetation fires are 30 nm and 80 nm, respectively, while in the sensitivity simulations all particles are emitted at 60 nm. Left bars are for winter and right shaded bars are for summer. (b) Autocorrelation function (ACF) of the  $\text{CCN}_{0.2}$  for Finokalia station calculated for summer (continuous lines) and for winter (dashed lines). Lines in colors are for the bars. The large-lag standard deviation curves are also shown in the graphs. The persistence time is defined

as the time that the large-lag standard error crosses the ACF curve (Schmale et al., 2018). (c) same as panel-b using the results of the sensitivity simulation. (d)-(f) same as the panel-a for Cabauw, Hyytiälä and Mace Head stations, respectively.

**Fig. S6.** Global surface distribution for the year 2011 of the  $N_3$  number concentrations as computed by the fifteen models that participated in this study. At the top of each panel the maximum value of the  $N_3$  simulated by the model is denoted. Units are number of [particles.cm<sup>-3</sup>]

**Fig. S7.** Same as Figure-6 for  $N_{50}$  number concentrations. Units are number of [particles.cm<sup>-3</sup>]

**Fig. S8.** Same as Figure-6 for  $N_{120}$  number concentrations. Units are number of [particles.cm<sup>-3</sup>]

**Fig. S9.** Same as Figure-6 for  $CCN_{0.2}$  number concentrations. Units are number of [particles.cm<sup>-3</sup>]

**Fig. S10.** Global surface distribution of the mass concentration of sulfate ( $SO_4$ ) of  $PM_{10}$  particles as computed by all models. At the top of each map the maximum value of the  $SO_4$  is denoted. Units are  $\mu\text{g-}SO_4.m^{-3}$

**Fig. S11.** Same as Figure-S10 for OA. Units are  $\mu\text{g-OA.m}^{-3}$

**Fig. S12.** Same as Figure-S10 for BC. Units are  $\mu\text{g.m}^{-3}$ .

**Fig. S13.** Same as Figure-S10 for SS. Units are  $\mu\text{g.m}^{-3}$

**Fig. S14.** Same as Figure-S10 for DU. Units are  $\mu\text{g.m}^{-3}$

**Fig. S15.** Global distributions of the annual multi-model median concentrations of the  $SO_4$ , OA, BC, DU and SS (from top to bottom) for the year 2011 (left column) and the corresponding diversities (right column). Model diversities are calculated as the ratio of the standard deviation to the mean of the models.