

## Chapter 2

# GLOMAP-Mode Overview

### 2.1 Introduction

The GLObal Model of Aerosol Processes (GLOMAP) is a comprehensive size-resolving global aerosol model which is embedded in the Toulouse Offline Model of Chemistry and Transport (TOMCAT, Stockwell and Chipperfield 1999; Chipperfield 2006). GLOMAP was initially developed using a two-moment (i.e. carrying information on particle number concentration and on particle mass concentration) bin scheme (GLOMAP-bin; Spracklen et al. 2005a,b). GLOMAP-bin has been used in numerous studies (e.g., Spracklen et al. 2008a,b; Merikanto et al. 2009; Pringle et al. 2009), and shown to perform well when compared to observational data sets of, for example, particle number concentrations and SO<sub>2</sub> mass concentrations (e.g. Spracklen et al. 2005a).

Mann et al. (2010) presented a full description and evaluation of a version of GLOMAP that uses a two-moment modal scheme (GLOMAP-mode). GLOMAP-mode treats the same aerosol microphysical processes as GLOMAP-bin but at greatly reduced computational cost. Throughout this thesis GLOMAP-mode has been used. GLOMAP-mode treats microphysical processes such as binary homogeneous nucleation, hygroscopic growth, coagulation, condensation, cloud processing (oxidation of dissolved SO<sub>2</sub> to SO<sub>4</sub> in cloud droplets), as well as dry and wet deposition.

GLOMAP-mode is benchmarked against the AEROCOM model inter-comparison data set (Textor et al. 2006) and several other observational data sets such as for example EMEP (European Monitoring and Evaluation Programme, Loevblad et al. 2004) or IMPROVE (Interagency Monitoring of Protected Visual Environments, Malm et al. 2002). Mann et al. (2010) have shown that the model is capable of simulating realistic present-day aerosol mass concentrations, number concentrations, particle size distributions, and CCN concentrations in both marine and continental locations.

GLOMAP-mode has been used in several previous studies. Manktelow et al. (2007) have investigated regional and global changes in the sulphur cycle between the years

1985 and 2000. Woodhouse et al. (2008) have investigated the response of CCN number concentrations to a regional, fivefold increase in DMS concentrations, and found only modest changes in CCN number concentrations. In a follow-up study, Woodhouse et al. (2010) have shown that, on a global scale, the sensitivity of CCN to changes in DMS is too low for future changes in DMS to be climatically relevant.

## 2.2 TOMCAT Chemical Transport Model

GLOMAP-mode is run as an extension to the TOMCAT global three-dimensional CTM (Stockwell and Chipperfield 1999; Chipperfield 2006) using a horizontal resolution of  $2.8^\circ \times 2.8^\circ$  and 31 hybrid  $\sigma$ - $p$  levels extending from the surface to 10 hPa in the vertical. Below about 100 hPa, the vertical  $\sigma$  levels are purely terrain-following whereas above  $\sim 100$  hPa they are pure pressure levels. The model is driven by meteorological fields specified from European Centre for Medium-Range Weather Forecasts (ECMWF) analyses at 6-h intervals. TOMCAT uses the tracer advection scheme of Prather (1986) which preserves sharp gradients and minimises numerical diffusion due to the conservation of second-order moments of the spatial distribution of the advected tracers. In order to calculate sub-grid scale moist convection in cumulus clouds, TOMCAT uses the mass flux scheme of Tiedtke (1989).

## 2.3 Gas-Phase Species and Chemistry

### Trace Gas Emissions

Depending on the aim of the study, different combinations of emission inventories have been used throughout this thesis. Each results chapter includes a statement as to what emissions have been used.

Oceanic DMS emissions have been calculated from monthly mean seawater DMS concentrations from Kettle and Andreae (2000) using the sea-air flux parameterisation of Nightingale et al. (2000). According to Kettle and Andreae (2000), DMS fluxes range from 13 to 37 Tg(S) per year depending on the gas flux parameterisations and wind speed prognostics. GLOMAP-mode predicts an annual mean DMS flux of 18.6 Tg(S) (Woodhouse et al. 2010).

In GLOMAP-mode, SO<sub>2</sub> is emitted from anthropogenic, biomass burning and volcanic sources. Anthropogenic SO<sub>2</sub> emissions are from Cofala et al. (2005) and account for 52.4 Tg(S) per year (with 24.1 Tg(S) per year from power stations, 19.6 Tg(S) per year from industrial processes, 5.7 Tg(S) per year from transportation, and 4.6 Tg(S) per year from domestic consumption). Anthropogenic SO<sub>2</sub> emissions used in this thesis are assumed to be constant throughout the year. Monthly varying SO<sub>2</sub> emission from biomass burning sources are from Van der Werf et al. (2003) and account for 2.1 Tg(S) per year. Depending on the vegetation type that is burning, the injection height ranges from 0 to 6 km which follows recommendations by Dentener et al. (2006).

The volcanic SO<sub>2</sub> emission inventory used in GLOMAP-mode is based on data sets by Andres and Kasgnoc (1998) and Halmer et al. (2002). The data set provided by Andres and Kasgnoc (1998) takes into account both continuously erupting volcanoes and sporadically erupting volcanoes and accounts for 10.4 Tg(S) per year. Following recommendations of Dentener et al. (2006) sulphur emissions from continuously erupting volcanoes are released into grid boxes between the height of the volcano and one third below that height in order to account for degassing along the flanks of the volcano. It should be noted, that the Andres and Kasgnoc (1998) data set (also referred to as the GEIA inventory, which includes information on the spatial distribution of the sources required for global modelling studies) is scaled by a factor 1.21 (Dentener et al. 2006, following recommendations of Graf et al. (1998) and Textor et al. (2004))<sup>1</sup> when referred to as AEROCOM data set with the latter being used throughout this thesis. Halmer et al. (2002) estimated that explosive volcanic activity averaged over the last 100 years accounts for 2.0 Tg(S) per year. In GLOMAP-mode sulphur emissions from explosively erupting volcanoes are released into grid boxes between 500 and 1500 m above the height of each volcano that has been active over the last 100 years. In order to account for gas-to-particle conversion in the volcanic plume, 2.5 % of the total sulphur flux is assumed to be emitted as primary sulphate into the accumulation and coarse mode (see also Sect. 2.5). Note that, this partitioning follows Stier et al. (2005) based on observations of anthropogenic sulphur emissions from power plants. In GLOMAP-mode, the total volcanic sulphur flux (i.e. using the inventories of Andres and Kasgnoc (1998) and Halmer et al. (2002)) equates to 14.22 Tg(S) per year; that is after partitioning into SO<sub>2</sub> and primary SO<sub>4</sub>. For the purpose of the study presented in Chap. 3 the Halmer et al. (2002) inventory was excluded whereas all remaining chapters used both the Andres and Kasgnoc (1998) and Halmer et al. (2002) inventories.

Monthly mean emission of monoterpenes are from Guenther et al. (1995) which is the only source of secondary organic carbon treated in this work.

### Gas Phase Chemistry

The sulphur chemistry scheme in GLOMAP-mode includes eight sulphur species: DMS, DMSO (dimethyl sulphoxide), MSA (methane sulphonic acid), SO<sub>2</sub>, H<sub>2</sub>S, H<sub>2</sub>SO<sub>4</sub>, COS (carbonyl sulphide), and CS<sub>2</sub> (carbon disulphide). The original sulphur reaction scheme was based on Pham et al. (1995) and used “offline oxidant fields” (i.e. by means of a look-up table) (e.g., Manktelow et al. 2007; Woodhouse et al. 2010). This “offline-scheme” has been updated as part of a PhD project in order to fully couple the oxidants and sulphur species (i.e. allowing for interactions between the sulphur species and oxidants) which is referred to as “coupled-chemistry scheme” (Breider et al. 2010; Breider 2010). Using a fully coupled chemistry scheme is crucial when simulating volcanic eruptions as high concentrations of SO<sub>2</sub> significantly impact oxidant concentrations which in turn feeds back onto the chemistry. For the work presented in this thesis the coupled chemistry version of GLOMAP-mode has been used which is coupled to the full background chemistry

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<sup>1</sup> Dentener et al. (2006) wrongly stated that the GEIA data set is scaled using a factor 1.5.

**Table 2.1** Sulphur reaction scheme in the coupled TOMCAT-GLOMAP-mode model

Reactants		Products	Reference
DMS + OH	→	SO <sub>2</sub> + CH <sub>3</sub> O <sub>2</sub> + HCHO	Atkinson (2000)
DMS + OH	→	0.6 SO <sub>2</sub> + 0.4 DMSO + 0.6 CH <sub>3</sub> O <sub>2</sub> + 0.4 HO <sub>2</sub>	Pham et al. (1995)
DMSO + OH	→	0.6 SO <sub>2</sub> + 0.4 MSA + 0.6 CH <sub>3</sub> O <sub>2</sub>	Pham et al. (1995)
DMS + NO <sub>3</sub>	→	SO <sub>2</sub> + HNO <sub>3</sub> + CH <sub>3</sub> O <sub>2</sub> + HCHO	Atkinson (2000)
H <sub>2</sub> S + OH	→	SO <sub>2</sub> + OH	Pham et al. (1995)
CS <sub>2</sub> + OH	→	SO <sub>2</sub> + COS + OH	Pham et al. (1995)
COS + OH	→	SO <sub>2</sub> + OH	Pham et al. (1995)
SO <sub>2</sub> + OH + M	→	H <sub>2</sub> SO <sub>4</sub> + HO <sub>2</sub> + M	Pham et al. (1995)

(O<sub>x</sub>–NO<sub>y</sub>–HO<sub>x</sub>, C<sub>1</sub>–C<sub>3</sub> NMHCs (non-methane hydrocarbons), isoprene) in TOMCAT (Breider et al. 2010). The sulphur reaction scheme is presented in Table 2.1.

### Aqueous Chemistry

Aqueous phase oxidation denotes the processes by which soluble gases such as SO<sub>2</sub> condense onto cloud droplets and subsequently become oxidised through the reaction with H<sub>2</sub>O<sub>2</sub> or O<sub>3</sub> (see also Reactions 1.5 and 1.6 in Chap. 1). GLOMAP-mode treats aqueous phase oxidation (on aerosol that are considered to be activated into drops) in grid boxes that contain ISCCP-derived low-level stratiform clouds (see Sect. 2.6 for details). The dissolved sulphate mass produced by aqueous phase oxidation is then added to the soluble accumulation and coarse modes according to their fractional contribution (in terms of number) with respect to the total number concentration within these two modes (Mann et al. 2010).

### Sulphuric Acid Vapour

Sulphuric acid vapour concentrations are crucial in determining the binary nucleation rate, and thus the rate of new particle formation. GLOMAP-mode accounts for the direct competition of nucleation and condensation (see Sect. 2.6) for the available H<sub>2</sub>SO<sub>4</sub> vapour by introducing short competition sub-steps calling these routines within the normal model time-step (see Sect. 2.6 and Fig. 2.2, Spracklen et al. 2005a; Mann et al. 2010).

## 2.4 Aerosol Size Distribution

GLOMAP-mode uses a modal aerosol dynamics approach (e.g., Whitby and McMurry 1997) by parameterising the shape of the aerosol size distribution as a series of log-normal modes. GLOMAP-mode uses a two-moment aerosol scheme with the particle number concentration and mass concentration of each component prognosed in each mode (the width of each mode is fixed). The scheme can be run with any number of modes and components but generally (in its standard configuration) follows the framework of the M7 model (Vignati et al. 2004; Stier et al. 2005) carrying sulphate (SU), sea-salt (SS), black carbon (BC), organic carbon (OC) and dust (DU)

**Table 2.2** Standard aerosol configuration for GLOMAP-mode with  $d_p$  = particle dry diameter; SU = sulphate; POM = particulate organic matter (i.e. POM = OC mass multiplied by 1.4 in order to account for molar mass of non-carbon atoms contributing to POM); BC = black carbon; SS = sea-salt; DU = mineral dust, and *sigma* referring to the geometric standard deviation of each of the respective modes

Mode	Size range	Sigma	Composition	Soluble?
Nucleation soluble	$d_p < 10 \text{ nm}$	1.59	SU, POM	Yes
Aitken soluble	$10 < d_p < 100 \text{ nm}$	1.59	SU, BC, POM	Yes
Accumulation soluble	$100 < d_p < 1 \mu\text{m}$	1.59	SU, BC, POM, SS, DU	Yes
Coarse soluble	$d_p > 1 \mu\text{m}$	2.0	SU, BC, POM, SS, DU	Yes
Aitken insoluble	$10 < d_p < 100 \text{ nm}$	1.59	BC, POM	No
Accumulation insoluble	$100 < d_p < 1 \mu\text{m}$	1.59	DU	No
Coarse insoluble	$d_p > 1 \mu\text{m}$	2.0	DU	No

Table modified after Mann et al. (2010)

in 4 soluble and 3 insoluble internally mixed modes. Table 2.2 lists properties and composition of each mode. For the work presented here, the component and mode setup used is stated in the methods section of each results chapter.

## 2.5 Primary Aerosol Emissions

Following recommendations of Dentener et al. (2006) a small fraction (2.5 %) of the total  $\text{SO}_2$  flux is emitted as primary sulphate in order to account for sub-grid scale nucleation processes. The formation of new sulphate particles is known to occur in both volcanic and industrial plumes (Allen et al. 2002; Brock et al. 2002). In the model, primary sulphate originating from anthropogenic sources is emitted (in equal proportions) at two mean mode radii of 0.075 and 0.75  $\mu\text{m}$  following Stier et al. (2005). Primary sulphate originating from volcanic and biomass burning sources is emitted at mean mode radii of 0.03 and 0.075  $\mu\text{m}$  following Stier et al. (2005).

Wind speed dependent sea-salt emissions in GLOMAP-mode are calculated using the parameterisation of Gong (2003) which produces a size-resolved sea-salt flux into the soluble accumulation and soluble coarse modes. The version of GLOMAP-mode used here does not account for sub-micron sea-salt fluxes; however this is not regarded as a limitation given the aim of this study.

Annual mean BC and OC emissions from fossil fuel and biofuel sources are from Bond et al. (2004), and monthly-varying BC and OC emissions from vegetation fires are from Van der Werf et al. (2003). Fossil fuel and biofuel emissions are injected into grid boxes below 100 m and emissions from wildfires are injected into grid boxes between the surface and 6 km (depending on the vegetation type that is burning) following Dentener et al. (2006).

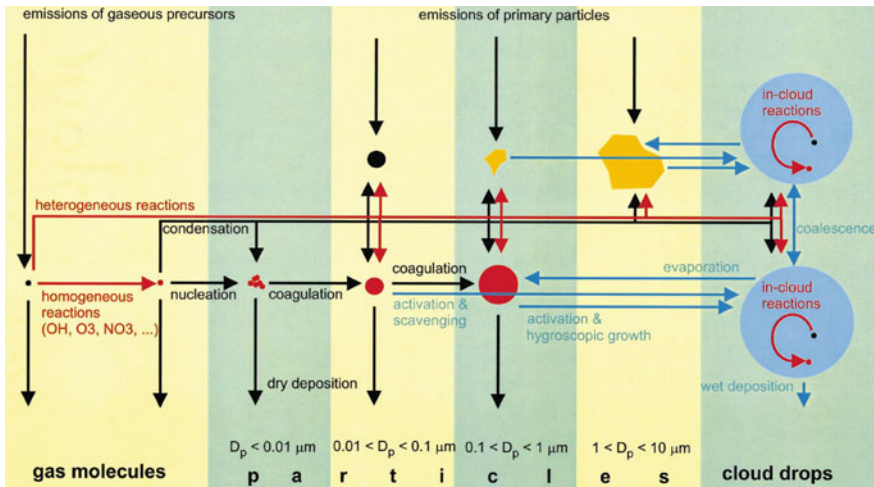
In Chap. 6 daily varying mineral dust emissions for the year 2000 are included following Dentener et al. (2006). Mineral dust is emitted into the insoluble accumulation and insoluble coarse modes.

## 2.6 Aerosol Microphysical Processes

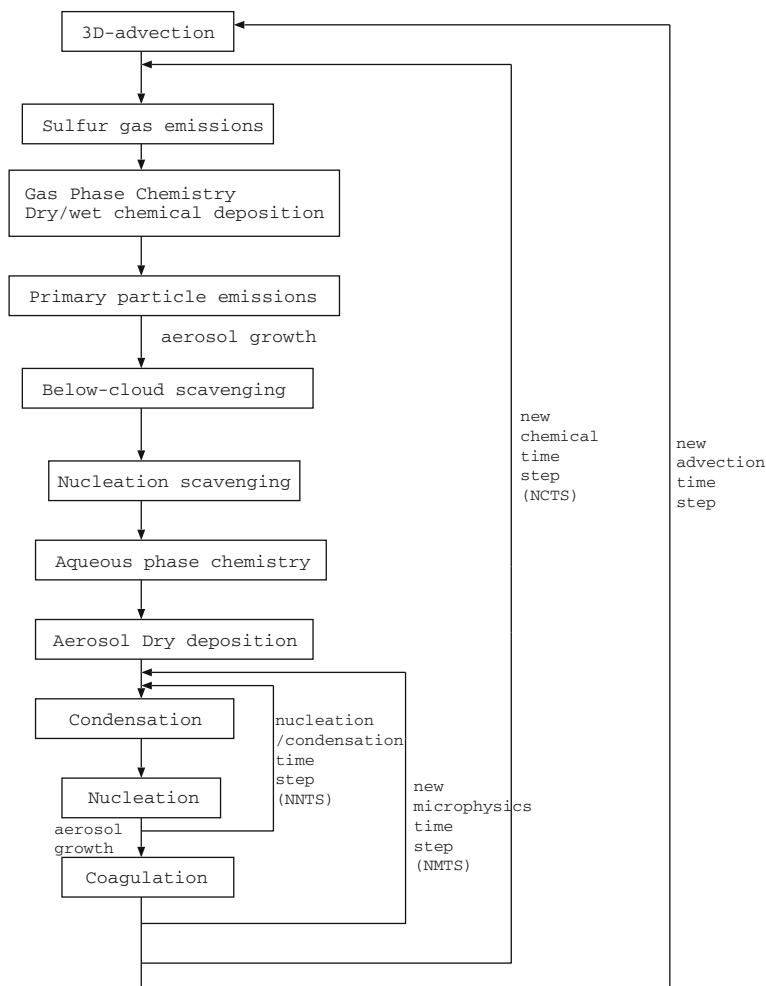
Figure 2.1 depicts the aerosol microphysical processes that shape the aerosol size distribution and influence the lifetime and chemical composition of the aerosol. The following section provides a brief overview of the microphysical processes accounted for in GLOMAP-mode. An in-depth description of the mathematical and numerical representation of the aerosol processes in GLOMAP-mode has been provided by Mann et al. (2010).

### Numerical Treatment

Figure 2.2 shows a flow chart of the order in which GLOMAP-mode treats advective, chemical and microphysical processes which follows the approach by Spracklen et al. (2005a). The differential equations determining particle number and mass concentrations are calculated by means of operator splitting. The version of GLOMAP-mode used here uses an advection time step of 1800 s, which is split into two chemical time steps (NCTS) during which chemistry and emissions are solved. Following that, a further split into two microphysical time steps (NMTS) solving the microphysics is utilised. In order to account for the competition between nucleation and condensation for available  $\text{H}_2\text{SO}_4$  vapour, a further split of NMTS into five timesteps (NNTS) is utilised.



**Fig. 2.1** The aerosol microphysical processes that shape the aerosol size distribution and influence the lifetime and chemical composition of atmospheric aerosol. Figure from Raes et al. (2000)



**Fig. 2.2** Flow chart of the order in which GLOMAP-mode treats advective, chemical and microphysical processes with NCTS = chemical time step, NMTS = microphysical time step, and NNTS = nucleation/condensation time step. Figure from Spracklen et al. (2005a)

## Nucleation

GLOMAP-mode simulates new particle formation in the free troposphere by using the binary  $\text{H}_2\text{SO}_4\text{--H}_2\text{O}$  nucleation scheme from Kulmala et al. (1998) with temperature, humidity and  $\text{H}_2\text{SO}_4$  concentration being non-linearly related to the nucleation rate (and thus the change in nucleation mode number concentration). Favourable conditions for binary nucleation are a low temperature, a low particle surface area and a high relative humidity, thus the upper troposphere provides an ideal environment for new particle formation to take place (Spracklen et al. 2005a,b). Overall, the precise

mechanism inducing the formation of new particles remains poorly understood, and other mechanisms such as ternary ( $\text{H}_2\text{SO}_4\text{--NH}_3\text{--H}_2\text{O}$ ) nucleation (e.g., Korhonen et al. 1999), and boundary layer nucleation (e.g., Kulmala et al. 2004) have been suggested.

### Coagulation

Once new particles are formed, they are subject to coagulation and condensation which are important processes for the growth of newly formed sub-micron particles. GLOMAP-mode accounts for both intra-modal coagulation (i.e. collision of particles within the same mode) and inter-modal coagulation (i.e. collision of particles in different modes) using a simplified mass conserving scheme following Seinfeld and Pandis (1998) as described in Spracklen et al. (2005a) and Mann et al. (2010). Coagulation is an important growth mechanism for freshly nucleated particles as these small and often numerous particles move rapidly by Brownian motion leading to a high particle collision rate.

### Condensation

Condensational growth of  $\text{H}_2\text{SO}_4$  vapour (and secondary organics) onto existing aerosol follows Fuchs and Sutugin (1971) with condensation rates depending on the existing particle surface areas, the concentration of  $\text{H}_2\text{SO}_4$  vapour and a mode-specific condensation coefficient. Sulphuric acid is a low volatility species, thus condenses easily onto existing aerosol.

### Mode-Merging

In order to avoid growth of particles to sizes outside the specified mode-ranges (see Table 2.2) the geometric mean diameter is used for each mode, and particle number and mass are transferred between the modes if the upper limit of the geometric mean diameter is exceeded.

### Removal Processes

Aerosol can be removed from the atmosphere by either dry deposition or wet deposition. In GLOMAP-mode, the parameterisation of dry deposition of aerosol follows the scheme of Zhang et al. (2001) accounting for gravitational settling, Brownian diffusion, impaction interception and particle rebound. In the model, the dry deposition rate depends on the size of particles, the land use category (e.g. forest, ocean) and the surface wind speed. Generally, the dry removal process is least efficient for particles with diameters of  $1\text{ }\mu\text{m}$  and increases in efficiency for particles larger than  $5\text{ }\mu\text{m}$  (as these gravitationally settle to the surface) and particles smaller than  $0.05\text{ }\mu\text{m}$  (as these diffuse to the surface). Overall, dry deposition leads to the formation of the accumulation mode of an aerosol size distribution.

GLOMAP-mode treats nucleation scavenging (i.e. formation of a droplet around an aerosol nucleus) of in-cloud activated aerosol in grid boxes that are, according to ECMWF reanalyses, precipitating. In GLOMAP-mode large-scale rain removes 99.9% of the aerosol number in the soluble accumulation and coarse modes at a constant removal rate over the course of 6 h. In contrast, convective rain is a sub-grid event for which the cloud-to-rainwater conversion rate is given by Tiedtke (1989) and removal of aerosol is assumed to occur in 30% of the grid box.



GLOMAP-mode accounts for removal of aerosol via impaction scavenging (i.e. below-cloud scavenging or collection of aerosol particles by falling raindrops) using a size-dependent raindrop-aerosol collection efficiency look-up table which is calculated from the aerosol mean dry radius and an estimate of the rainfall size distribution (Sekhon and Srivastava 1971). In the model, impact scavenging is calculated for both dynamic and convective rain.

### Cloud Processing

Here, the term “cloud processing” denotes the growth of already existing aerosol particles (as water droplets in non-precipitating clouds) by uptake and aqueous phase oxidation of  $\text{SO}_2$ . Cloud processing creates, after the evaporation of the drop, a minimum in the particle size distribution (referred to as Hoppel gap, e.g., Hoppel et al. 1994) which defines the Aitken and the accumulation modes. Cloud processing of aerosol in GLOMAP-mode follows Spracklen et al. (2005a) using a globally uniform activation dry radius that defines the smallest particles that are activated to cloud droplets. Throughout this thesis, an activation dry radius of 37.5 nm (corresponding to a cloud supersaturation of 0.2 % for sulphuric acid, which is typical for stratocumulus clouds) is assumed.

#### 2.6.1 Aerosol Activation

Only a subset of the entire aerosol population acts as CCN. Throughout this thesis, CCN were counted as soluble particles with a dry radius larger than 35 nm, which is equivalent to the particles that would activate into cloud droplets at 0.22 % supersaturation (i.e. when considering  $\text{H}_2\text{SO}_4$ ). Several microphysical processes lead to an increase in CCN concentrations:

- (a) nucleation of  $\text{H}_2\text{SO}_4$  vapour to  $\text{SO}_4$  aerosol and its subsequent growth to CCN sizes by coagulation and condensation;
- (b) growth to CCN sizes by condensation of  $\text{H}_2\text{SO}_4$  onto existing particles; and
- (c) cloud processing of Aitken mode-sized particles.

Once CCN are present in the atmosphere, water vapour can condense onto these particles and grow these particles to cloud drop size which is a hundred times larger when compared to the initial CCN-particle size. In this thesis, cloud drop number concentrations (CDNC) were calculated using a physically-based aerosol activation scheme, which is identical to Nenes and Seinfeld (2003) and evaluated for GLOMAP in Pringle et al. (2009). The Nenes and Seinfeld (2003) scheme predicts CDNC by means of calculating the maximum supersaturation for a given updraught velocity and aerosol distribution. Once the maximum supersaturation is obtained, the number of activated cloud drops is equal to the number of particles with a critical supersaturation less than the maximum supersaturation with the respective activation diameter being calculated using Köhler theory. The scheme has been shown to compare well with parcel model simulations (Nenes and Seinfeld 2003; Fountoukis and Nenes 2005)

and empirical schemes (Pringle et al. 2009). CDNC were calculated as a post-processing step using GLOMAP-mode output, thus there is no feedback with aerosol microphysical processes. CDNC were calculated in every grid box regardless of the presence of clouds, however when assessing the cloud-radiative effect a cloud mask was used. In this thesis, CDNC were calculated at the base of stratus clouds (approximately 1 km altitude or 920 hPa) using a globally uniform updraught velocity of  $0.2 \text{ ms}^{-1}$ . Updraught velocities of  $0.1\text{--}0.3 \text{ ms}^{-1}$  are most commonly observed in stratus clouds (e.g., Gultepe and Isaac 1999; Peng et al. 2005). Fountoukis et al. (2007) showed that using an average updraught velocity provides a good approximation of mean CDNC. As with many other aerosol activation schemes (e.g., Chen and Penner 2005; Roelofs et al. 2006), the employed scheme does not account for droplet collision-coalescence (i.e. no droplet loss rate), thus the CDNC shown is representative of the cloud base.

## 2.7 Conclusions

GLOMAP-mode is a comprehensive global aerosol microphysics model embedded in the TOMCAT CTM. GLOMAP-mode uses a two-moment scheme in order to simulate the aerosol size distribution by carrying both aerosol mass and aerosol number. GLOMAP-mode accounts for microphysical processes such as nucleation, coagulation, condensation, cloud processing, and dry and wet deposition—all of which determine the evolution of the aerosol size distribution. GLOMAP-mode can be run using any combination of sulphate, sea-salt, black carbon, organic carbon and mineral dust components. Mann et al. (2010) provided a comprehensive overview of the processes treated in GLOMAP-mode, and of the models performance when compared to observational data sets and other numerical models. For the purpose of the work presented here, the coupled chemistry version of GLOMAP-mode has been employed.

## References

- Allen AG, Oppenheimer C, Ferm M, Baxter PJ, Horrocks LA, Galle B, McGonigle AJS, Duffell HJ (2002) Primary sulfate aerosol and associated emissions from Masaya Volcano, Nicaragua. *J Geophys Res* 107(D23):4682. doi:[10.1029/2002JD002120](https://doi.org/10.1029/2002JD002120)
- Andres RJ, Kasgnoc AD (1998) A time-averaged inventory of subaerial volcanic sulfur emissions. *J Geophys Res* 103:25251–25262
- Atkinson R (2000) Atmospheric chemistry of VOCs and NOx. *Atmos Environ* 34:2063–2101
- Bond TC, Streets DG, Yarber KF, Nelson SM, Woo JH, Klimont Z (2004) A technology-based global inventory of black and organic carbon emissions from combustion. *J Geophys Res* 109:D14203. doi:[10.1029/2003JD003697](https://doi.org/10.1029/2003JD003697)
- Breider TJ, Chipperfield MP, Richards NAD, Carslaw KS, Mann GW, Spracklen DV (2010) Impact of BrO on dimethylsulfide in the remote marine boundary layer. *Geophys Res Lett* 37:L02807. doi:[10.1029/2009GL040868](https://doi.org/10.1029/2009GL040868)

- Breider TJ (2010) Coupled halogen-sulfur-aerosol modeling in a 3D chemical transport model. Ph.D. thesis, School of Earth and Environment, University of Leeds
- Brock CA, Washenfelder RA, Trainer M, Ryerson TB, Wilson JC, Reeves JM, Huey LG, Holloway JS, Parrish DD, Hübler G, Fehsenfeld FC (2002) Particle growth in the plumes of coal-fired power plants. *J Geophys Res* 107:4155. doi:[10.1029/2001JD001062](https://doi.org/10.1029/2001JD001062)
- Chen Y, Penner JE (2005) Uncertainty analysis for estimates of the first indirect aerosol effect. *Atmos Chem Phys* 5:2935–2948
- Chipperfield MP (2006) New version of the TOMCAT/SLIMCAT off-line chemical transport model: intercomparison of stratospheric tracer experiments. *Q J R Meteorol Soc* 132:1179–1203
- Cofala J, Amann M, Klimont Z, Schopp W (2005) Scenarios of world anthropogenic emissions of SO<sub>2</sub>, NO<sub>x</sub> and CO up to 2030. In: Internal report of the transboundary air pollution programme, International Institute of Applied Systems Analysis, Laxenburg, 2005
- Dentener F, Kinne S, Bond T, Boucher O, Cofala J, Generoso S, Ginoux P, Gong S, Hoelzemann JJ, Ito A, Marelli L, Penner JE, Putaud JP, Textor C, Schulz M, van der Werf GR, Wilson J (2006) Emissions of primary aerosol and precursor gases in the years 2000 and 1750 prescribed data-sets for AeroCom. *Atmos Chem Phys* 6:4321–4344
- Fountoukis C, Nenes A (2005) Continued development of a cloud droplet formation parameterization for global climate models. *J Geophys Res* 110:D11212
- Fountoukis C, Nenes A, Meskhidze N, Bahreini R, Conant WC, Jonsson H, Murphy S, Sorooshian A, Varutbangkul V, Brechtel F, Flagan RC, Seinfeld JH (2007) Aerosol-cloud drop concentration closure for clouds sampled during the international consortium for atmospheric research on transport and transformation 2004 campaign. *J Geophys Res* 112:D10S30
- Fuchs NA, Sutugin AG (1971) Topics in current aerosol research, International reviews in aerosol physics and chemistry, vol 2–3. Pergamon Press, New York
- Gong SL (2003) A parameterization of sea-salt aerosol source function for sub- and super-micron particles. *Glob Biogeochem Cycles* 17(4):1097
- Graf HF, Langmann B, Feichter J (1998) The contribution of earth degassing to the atmospheric sulfur budget. *Chem Geol* 147:131–145
- Guenther A, Hewitt CN, Erickson D, Fall R, Geron C, Graedel T, Harley P, Klinger L, Lerdau M, McKay WA, Pierce T, Scholes B, Steinbrecher R, Tallamraju R, Taylor J, Zimmerman P (1995) A global model of natural volatile organic compound emissions. *J Geophys Res* 100:8873–8892
- Gultepe I, Isaac GA (1999) Scale effects on averaging of cloud droplet and aerosol number concentrations: observations and models. *J Clim* 12:1268–1279
- Halmer M, Schmincke H, Graf HF (2002) The annual volcanic gas input into the atmosphere, in particular into the stratosphere, a global data-set for the past 100 years. *J Volcanol Geotherm Res* 115:511–528
- Hoppel WA, Frick GM, Fitzgerald JW, Larson RE (1994) Marine boundary layer measurements of new particle formation and the effects nonprecipitating clouds have on aerosol size distribution. *J Geophys Res* 99:14443–14459
- Kettle AJ, Andreae MO (2000) Flux of dimethylsulfide from the oceans: a comparison of updated data sets and flux models. *J Geophys Res* 105:26793–26808
- Korhonen P, Kulmala M, Laaksonen A, Viisanen Y, McGraw R, Seinfeld JH (1999) Ternary nucleation of H<sub>2</sub>SO<sub>4</sub>, NH<sub>3</sub>, and H<sub>2</sub>O in the atmosphere. *J Geophys Res* 104:26349–26353
- Kulmala M, Laaksonen A, Pirjola L (1998) Parameterization for sulfuric acid/water nucleation rates. *J Geophys Res* 103:8301–8307
- Kulmala M, Vehkamäki H, Petäjä T, Dal Maso M, Lauri A, Kerminen VM, Birmili W, McMurry PH (2004) Formation and growth rates of ultrafine atmospheric particles: a review of observations. *J Aerosol Sci* 35:143–176
- Loevblad G, Tarrason L, Torseth K, Dutchak S (2004) Sulphur. In: Loevblad G, Tarrason L, Torseth K, Dutchak S (eds) EMEP assessment, part I: European perspective, Norwegian Meteorological Institute, pp 15–46
- Malm WC, Schichtel BA, Ames RB, Gebhart KA (2002) A 10-year spatial and temporal trend of sulfate across the United States. *J Geophys Res* 107:4627

- Manktelow PT, Mann GW, Carslaw KS, Spracklen DV, Chipperfield MP (2007) Regional and global trends in sulfate aerosol since the 1980s. *Geophys Res Lett* 34:L14803
- Mann GW, Carslaw KS, Spracklen DV, Ridley DA, Manktelow PT, Chipperfield MP, Pickering SJ, Johnson CE (2010) Description and evaluation of GLOMAP-mode: a modal global aerosol microphysics model for the UKCA composition-climate model. *Geosci Model Dev* 3:519–551
- Merikanto J, Spracklen DV, Mann GW, Pickering SJ, Carslaw KS (2009) Impact of nucleation on global CCN. *Atmos Chem Phys* 9:8601–8616
- Nenes, A, Seinfeld JH (2003) Parameterization of cloud droplet formation in global climate models. *J Geophys Res* 108(D14):4415. doi:[10.1029/2002JD002911](https://doi.org/10.1029/2002JD002911)
- Nightingale PD, Malin G, Law CS, Watson AJ, Liss PS, Liddicoat MI, Boutin J, Upstill-Goddard RC (2000) In situ evaluation of air-sea gas exchange—parameterizations using novel conservative and volatile tracers. *Global Biogeochem Cycles* 14(1):373–387
- Peng Y, Lohmann U, Leaitch R (2005) Importance of vertical velocity variations in the cloud droplet nucleation process of marine stratus clouds. *J Geophys Res* 110:D21213
- Pham M, Mueller JF, Brasseur GP, Granier C, Megie G (1995) A three-dimensional study of the tropospheric sulfur cycle. *J Geophys Res* 100(D12):26061–26092
- Prather MJ (1986) Numerical advection by conservation of second-order moments. *J Geophys Res* 91:6671–6681
- Pringle KJ, Carslaw KS, Spracklen DV, Mann GM, Chipperfield MP (2009) The relationship between aerosol and cloud drop number concentrations in a global aerosol microphysics model. *Atmos Chem Phys* 9:4131–4144
- Raes F, Dingenen RV, Vignati E, Wilson J, Putaud JP, Seinfeld JH, Adams P (2000) Formation and cycling of aerosols in the global troposphere. *Atmos Environ* 34:4215–4240
- Roelofs GJ, Stier P, Feichter J, Vignati E, Wilson J (2006) Aerosol activation and cloud processing in the global aerosol-climate model ECHAM5-HAM. *Atmos Chem Phys* 6:2389–2399
- Seinfeld J, Pandis S (1998) *Atmospheric chemistry and physics: from air pollution to climate change*. Wiley, New York, p 1326
- Sekhon RS, Srivastava RC (1971) Doppler radar observations of drop-size distributions in a thunderstorm. *J Atmos Sci* 28:983–994
- Spracklen DV, Bonn B, Carslaw KS (2008b) Boreal forests, aerosols and the impacts on clouds and climate. *Phil Trans R Soc A Math phys Eng Sci* 366:4613–4626
- Spracklen DV, Carslaw KS, Kulmala M, Kerminen VM, Sihto SL, Riipinen I, Merikanto J, Mann GW, Chipperfield MP, Wiedensohler A, Birmili W, Lihavainen H (2008a) Contribution of particle formation to global cloud condensation nuclei concentrations. *Geophys Res Lett* 35:L06808
- Spracklen DV, Pringle KJ, Carslaw KS, Chipperfield MP, Mann GM (2005b) A global off-line model of size resolved aerosol processes: II. Identification of key uncertainties. *Atmos Chem Phys* 5:3437–3489
- Spracklen DV, Pringle KJ, Carslaw KS, Chipperfield MP, Mann GW (2005a) A global off-line model of size-resolved aerosol microphysics: I. Model development and prediction of aerosol properties. *Atmos Chem Phys* 5:179–215
- Stier P, Feichter J, Kinne S, Kloster S, Vignati E, Wilson J, Ganzeveld L, Tegen I, Werner M, Balkanski Y, Schulz M, Boucher O, Minikin A, Petzold A (2005) The aerosol-climate model ECHAM5-HAM. *Atmos Chem Phys* 5:1125–1156
- Stockwell DZ, Chipperfield MP (1999) A tropospheric chemical-transport model: development and validation of the model transport schemes. *Q J R Meteorol Soc* 125:1747–1783
- Textor C, Graf C, Timmreck HF, Robock A (2004) Emissions from volcanoes. In: Granier C, Artaxo P, Reeves C (eds) *Emissions of chemical compounds and aerosols in the atmosphere*. Kluwer, Dordrecht, pp 269–303
- Textor C, Schulz M, Guibert S, Kinne S, Balkanski Y, Bauer S, Bernsten T, Berglen T, Boucher O, Chin M, Dentener F, Diehl T, Easter R, Feichter H, Fillmore D, Ghan S, Ginoux P, Gong S, Grini A, Hendricks J, Horowitz L, Huang P, Isaksen I, Iversen I, Kloster S, Koch D, Kirkevåg A, Kristjansson JE, Krol M, Lauer A, Lamarque JF, Liu X, Montanaro V, Myhre G, Penner J,

- Pitari G, Reddy S, Seland Ø, Stier P, Takemura T, Tie X (2006) Analysis and quantification of the diversities of aerosol life cycles within AeroCom. *Atmos Chem Phys* 6:1777–1813
- Tiedtke M (1989) A comprehensive mass flux scheme for cumulus parameterization in large-scale models. *Mon Weather Rev* 117:1779–1800
- Van der Werf GR, Randerson JT, Collatz GJ, Giglio L (2003) Carbon emissions from fires in tropical and subtropical ecosystems. *Glob Chang Biol* 9:547–562
- Vignati E, Wilson J, Stier P (2004) M7: An efficient size-resolved aerosol microphysics module for large-scale aerosol transport models. *J Geophys Res* 109:D22202
- Whitby ER, McMurry PH (1997) Modal aerosol dynamics modeling. *Aerosol Sci Technol* 27:673–688
- Woodhouse MT, Carslaw KS, Mann GW, Vallina SM, Vogt M, Halloran PR, Boucher O (2010) Low sensitivity of cloud condensation nuclei to changes in the sea-air flux of dimethyl-sulphide. *Atmos Chem Phys* 10:7545–7559
- Woodhouse MT, Mann GW, Carslaw KS, Boucher O (2008) New directions: the impact of oceanic iron fertilisation on cloud condensation nuclei. *Atmos Environ* 42:5728–5730
- Zhang L, Gong S, Padro J, Barrie L (2001) A size-segregated particle dry deposition scheme for an atmospheric aerosol module. *Atmos Environ* 35:549–560

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