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MATCH-SALSA – Multi-scale Atmospheric Transport and CHEmistry model coupled to the SALSA aerosol microphysics model – Part 1: Model description and evaluation

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Abstract. We have implemented the sectional aerosol dynamics model SALSA (Sectional Aerosol module for Large Scale Applications) in the European-scale chemistry-transport model MATCH (Multi-scale Atmospheric Transport and Chemistry). The new model is called MATCH-SALSA. It includes aerosol microphysics, with several formulations for nucleation, wet scavenging and condensation. The model reproduces observed higher particle number concentration (PNC) in central Europe and lower concentrations in remote regions. The modeled PNC size distribution peak occurs at the same or smaller particle size as the observed peak at four measurement sites spread across Europe. Total PNC is underestimated at northern and central European sites and accumulation-mode PNC is underestimated at all investigated sites. The low nucleation rate coefficient used in this study is an important reason for the underestimation. On the other hand, the model performs well for particle mass (including secondary inorganic aerosol components), while elemental and organic carbon concentrations are underestimated at many of the sites.

Further development is needed, primarily for treatment of secondary organic aerosol, in terms of biogenic emissions and chemical transformation. Updating the biogenic secondary organic aerosol (SOA) scheme will likely have a large impact on modeled PM2.5 and also affect the model performance for PNC through impacts on nucleation and condensation.

1 Introduction

Most aerosol properties relevant to climate are both size and chemical composition dependent. Thus, there is a need to resolve the size distributions of particle mass, number and chemical composition in climate models (e.g., Chen and Penner, 2005; Roesler and Penner, 2010). Aerosol particles also have adverse effects on human health (e.g., Pope and Dockery, 2006), which depend on particle size and chemical composition (WHO, 2013). In particular, ultrafine particles (with diameters of less than 100 nm) may be important for their potential impacts on human health (e.g., Oberdörster et al., 1995; Peters et al., 1997; Knol et al., 2009), but there is still limited epidemiological evidence of their health effects (WHO, 2013). The ultrafine particles do not contribute significantly to the particle mass concentration (PM), but they constitute a large proportion of the particle number concentration (PNC). Aerosol microphysical processes need to be considered in greater detail in order to describe PNC and size distributions accurately (e.g., Adams and Seinfeld, 2002). This has led to an increased need for realistic treatment of aerosols in atmospheric models.

A number of chemical transport models (CTMs), which are used operationally for simulating atmospheric chemistry in Europe, were recently reviewed by Kukkonen et al. (2012). The aerosol descriptions in such types of models can be classified into three main categories: bulk schemes, modal schemes (Whitby and McMurry, 1997) and sectional...
schemes (Gelbard et al., 1980). In bulk schemes, typically the total mass concentration of particles, or the mass in a certain size interval, is modeled. LOTOS-EUROS (LOng Term Ozone Simulation – EUropean Operational Smog; Schaap et al., 2008), DEHM (Dutch Eulerian Hemispheric Model; e.g. Frohn et al., 2002) and the EMEP MSC-W model (European Monitoring and Evaluation Programme Meteorological Synthesizing Centre – West; Simpson et al., 2012) are examples of bulk-type models.

In modal schemes, the aerosol size distribution is represented by a small number of modes, typically logarithmic normal size distribution for the modes. The description of new particle formation is limited in modal schemes. Modal schemes are computationally more expensive than simple bulk schemes, but less than the sectional approach, which is why they are commonly used in regional and global CTMs and climate models, e.g., the Regional Particulate Model (Binkowski and Shankar, 1995), CMAQ (Byun and Schere, 2006), CAM5-MAM3 (Liu et al., 2012), TM5 (Aan de Brugh et al., 2011), GLOMAP-mode (Mann et al., 2012), EMAC (Pringle et al., 2010), ECHAM5-HAM2 (Zhang et al., 2012), GISS-MATRIX (Bauer et al., 2008).

The sectional scheme, in which the size distribution is represented by a large number of discrete bins, is the most flexible and accurate choice – but computationally the most expensive. Many modern CTMs and global climate models (GCMs) include the sectional approach, e.g., PM-CAMx (Fountoukis et al., 2011), GLOMAP-bin (Hann et al., 2011), ECHAM5-SALSA (Bergman et al., 2012) and GISS-TOMAS (Lee and Adams, 2010). PM-CAMx and GLOMAP-bin make the assumption of internally mixed particles, in GLOMAP described by 20 size bins, whereas GISS-TOMAS includes externally mixed particles described by 30 size bins. Such a high size bin resolution is computationally demanding. GLOMAP uses prescribed monthly mean oxidant fields. Mann et al. (2014) compared the performance of 12 global aerosol microphysics models using modal and sectional approaches.

The standard version of the MATCH (Multi-scale Atmospheric Transport and Chemistry) model (Robertson et al., 1999; Andersson et al., 2007) uses a simple bulk scheme for treating aerosols, with four size bins for primary particles, without any aerosol dynamics treatment (except hygroscopic growth in some model versions), but with dry and wet deposition of primary particles dependent on particle size. The particle species considered in previous applications (e.g., Andersson et al., 2007, 2009) were primary anthropogenic elemental carbon (EC), organic carbon (OC) and non-carbonaceous particles, as well as secondary inorganic aerosol (sulfate, nitrate, ammonium) and sea-salt particles. Secondary organic aerosol was not included in the model. PNC formation and growth was not described. MATCH was adapted to assess anthropogenic ultratfine particles in an urban environment in a previous study (Gidhagen et al., 2005); seven monodisperse sizes were used and the aerosol dynamics considered water uptake, coagulation and dry deposition, but without inclusion of nucleation or condensation processes.

The MATCH model includes photo-chemistry for calculating oxidant fields that can be used for online coupling to oxidation of organics and sulfur compounds, resulting in a coupled photo-chemistry and aerosol dynamics description. Further, MATCH contains a number of advanced features, including data assimilation (Kahnert, 2008) and inverse modeling of aerosol optics of both surface observations and satellite data (Kahnert, 2009). These assimilation techniques are uncommon in models that include advanced aerosol dynamics.

We have implemented the sectional aerosol dynamics model SALSA (Sectional Aerosol module for Large Scale Applications; Kokkola et al., 2008) in the European-scale CTM MATCH (Robertson et al., 1999; Andersson et al., 2007). SALSA was chosen since it was developed to describe the PNC well; it includes several nucleation mechanisms and the sectional approach used in SALSA is an advantage for simulating new particle formation (e.g., Korhola et al., 2014). The coupling of SALSA to MATCH introduces a description of particle microphysics and aging in the model. New features include particle nucleation, condensation, coagulation and activation, leading to a description of the temporal evolution of the particle number size distribution in a number of bins, through the sectional approach. The model also describes the mixing state of the particles. The physical treatment of aerosol microphysics and the particle size distribution is described in Sect. 2.2; further details about the specific setup used in this study are given in Sect. 3. We discuss the performance of MATCH-SALSA in relation to other models in Sect. 4.

This paper presents the resulting new aerosol dynamics version of the MATCH model; the new model is called MATCH-SALSA. The model was detailed in a report from SMHI (Andersson et al., 2013), which is included as a supplement to this paper (Supplement A). In this paper, we highlight the main new features and present the results from evaluation tests. In a second paper (Andersson et al., 2015) results from various sensitivity tests will be presented. The aim of MATCH-SALSA is to describe particle mass and number concentrations, and particle size distribution on the European scale. The new model features – inclusion of sectional descriptions of aerosol microphysics and particle number size distributions – are developed with the aim to couple the MATCH-SALSA model to climate models and radiative transfer calculations; the new model can also be utilized for the estimation of human exposure to particles of different sizes.

2 Description of MATCH-SALSA

The layout of MATCH-SALSA is illustrated in Fig. 1. After initializations are completed, the model integrates starting
Figure 1. Model layout and time stepping in MATCH-SALSA.

with reading or interpolation of weather data, reading emissions and setting lateral and top boundary concentrations of the chemical species. After this, the emissions are injected and model transport fluxes are calculated with the internal sub-stepping time steps. Subsequently, the model gas-phase and wet-phase chemistry, aerosol microphysics and cloud droplet number concentrations are calculated. Meteorological data are read at regular intervals, typically every 3 or 6 hours. Boundary conditions may be updated at compound-dependent time intervals.

Natural and anthropogenic emissions are included in the model. Sea salt and isoprene emissions are calculated online, whereas anthropogenic and other emissions (volcanic sulfur, marine dimethyl sulfide (DMS) and biogenic monoterpenes) are given as input data to the model in the setup used in the present study. All primary particle components are emitted both as mass and number. Sea-salt emissions are modeled as described by Foltescu et al. (2005) but modified to allow arbitrary size bins. For the smallest bins (diameters of ≤ 1 µm), the description by Mårtensson et al. (2003) was used; for larger sizes the sea-salt generation function was taken from Monahan et al. (1986). Biogenic emissions of isoprene are calculated using the E-94 isoprene emission methodology proposed by Simpson et al. (1995). Emissions from wildfires and agricultural burning are not included in the present version of the model.

The transport model includes advective and turbulent transport. Particle number and mass are transported independently in MATCH-SALSA. The transport scheme is described in detail in Robertson et al. (1999).

2.1 Chemistry

The original MATCH photochemistry scheme (Langner et al., 1998) was, to a large extent, based on the EMEP MSC-W (European Monitoring and Evaluation Programme Meteorological Synthesizing Centre – West) scheme (Simpson, 1992; Simpson et al., 1993), but with an alternative treatment of isoprene chemistry, using an adapted version of the Carter one-product mechanism (Carter, 1996; Langner et al., 1998). A simplified mixture of a dozen representative compounds (“lumped molecules”) was used to model all organic molecules emitted to the atmosphere (e.g., o-xylene represents all emitted aromatic species).

The gas-phase chemistry scheme in MATCH has remained mostly the same since 1998, but a number of reaction rates have been updated, taking into account new recommendations from the International Union of Pure and Applied Chemistry (IUPAC; Atkinson et al., 2006) and the Master Chemical Mechanism, MCM v3 (Jenkin et al., 1997; Saunders et al., 2003, via http://mcm.leeds.ac.uk/MCM); a few new gas-phase components have also been added to the scheme. The revision of the MATCH chemistry scheme was based closely on the updates done in the EMEP MSC-W model, during 2008–2009, as documented by Simpson et al. (2012); the updated gas-phase reaction scheme in MATCH is nearly identical to the EMEP MSC-W Em-
using a relatively coarse particle size resolution, and includ-
reached by keeping the number of tracer variables low, by
The SALSA model was designed to obtain a balance between
reaction rates, see Supplement B.
2.2 Aerosol microphysics
The SALSA model was designed to obtain a balance between
cost of MATCH-SALSA some further modifications related to particle formation have been made
and the scheme used in the present work consists of ca. 140
thermal, wet and photolysis reactions, including ca. 60 dif-

different chemical species.
The chemistry code includes a simple scheme for sec-
ondary organic aerosol (SOA) formation from biogenic
monoterpene emissions; α-pinene is used as a surrogate for
all monoterpenes. In the present study, we assume rapid for-
mation of condensable SOA after gas-phase oxidation of α-
pinene (by O₃, OH or NO₃; oxidation rates are based on
MCM v3.2, http://mcm.leeds.ac.uk/MCM); we assumed that
all oxidation paths for α-pinene produce low-volatility SOA-
forming compounds, with 10% (mass-based) yield. These
compounds are included in the condensation scheme for or-
ganic compounds in SALSA. The SOA yield used here for
α-pinene is relatively high compared to some reported SOA
yields for this monoterpene in smog chamber experiments
(e.g., Mentel et al. (2009) found about 5% yield). However,
recent findings by Ehn et al. (2014) regarding formation of
extremely low-volatility organic compounds from ozonol-
ysis of α-pinene indicate that SOA yields from this process
may be higher than 10% above forest canopies. We also note
that there are recent studies that indicate that SOA yields
based on smog chamber studies may be underestimated by
up to a factor of 4, due to wall losses of gas-phase semi-
volatile organic (Kokkola et al., 2014; Zhang et al., 2014).
Note that the simplified biogenic secondary organic aerosol
(BSOA) “scheme” used in the present study is included to
test the organic-aerosol parts of MATCH-SALSA, with mi-
imal changes to the standard photochemistry scheme; it is not
expected to model BSOA formation in a very realistic way
compared to real-world conditions, but, given the high un-
certainties in monoterpene emissions and the neglect of other
BSOA-forming emissions, it was considered a reasonable ap-
proach for the development phase of MATCH-SALSA.

The chemical equations are solved prior to SALSA.
There is no internal sub-stepping between the chemistry and
SALSA (cf. Fig. 1). For a detailed description of the MATCH
chemistry scheme, including a full list of the reactions and
reaction rates, see Supplement B.

In addition, the chemical compounds that are treated in each
size range are chosen depending on the compounds that are
relevant to that size of particles in the atmosphere (for details,
see Kokkola et al., 2008):

1. The first size range (nucleation and Aitken modes) in-
cludes sulfate (SO₄²⁻) and OC.
2. The second (accumulation-mode) size range includes
SO₄²⁻, EC, OC, sea salt (NaCl) and mineral dust in two
externally mixed parallel size bins for each size section.
3. The third (coarse-mode) size range also includes SO₄²⁻,
EC, OC, sea salt (NaCl) and mineral dust in three exter-

Note that EC is not included in the Aitken mode, which is a
shortcoming of MATCH-SALSA. The reason for this choice
in SALSA was to reduce the CPU burden.
The hygroscopicity of the aerosol is calculated using the
Zdanovskii–Stokes–Robinson method (Jacobson, 2002). At
the end of each microphysical time step, the size distribution
is updated to take into account the growth of particles due to
dynamic and chemical transformation processes.
Nitrate in coarse-mode particles is treated separately as a simple tracer compound. Other particulate nitrogen species are described by a simplified chemistry scheme (see Supplement B), currently handled outside SALSA i.e., ammonium salts (e.g., ammonium nitrate) are not taken into account in the modeling of the aerosol microphysical processes. After the aerosol microphysical processes have taken place, ammonium bound to sulfate is distributed according to the size distribution of particulate sulfate and ammonium nitrate is distributed according to the available aerosol surface area. However, this condensation of ammonium and nitrate does not affect the particle radius in the model, and thus they do not influence the shape of the size distribution. A possible consequence of the simplified treatment can be underestimation of condensational growth, which may cause overestimation of nucleation, due to a too-small condensational sink for the nucleation-mode particles. The lack of ammonium nitrate condensation in the aerosol microphysics could cause underestimation of cloud droplet number concentration (CDNC).

In this study nucleation is simulated through an activation-type nucleation formulation (Kulmala et al., 2006; Riipinen et al., 2007) and the formation rate of 3 nm particles (J3) is calculated according to Lehtinen et al. (2007). Nucleation is solved concurrently with condensation, using the methodology of Jacobson (2002): this takes into account the competition of nucleation and condensation in the mass transfer of volatile species between gas and particle phase. The MATCH-SALSA model also includes other nucleation schemes, for example binary nucleation (Vehkamäki et al., 2002), ternary nucleation (Napari et al., 2002a, b) and activation of both H2SO4 and organic vapors (Paasonen et al., 2010; Supplement C). Tests of these alternative nucleation schemes will be presented in the companion paper (Andersson et al., 2015).

The scheme used for gas-to-particle transformation is the analytical predictor of condensation scheme, with saturation vapor pressure set to zero (Jacobson, 1997). The scheme solves condensation and evaporation of semi-volatile compounds over a discrete time step. It is very well suited for large-scale atmospheric models, such as MATCH, since it requires no iteration, it is mass conserving, and it has been shown to be accurate over a time step length of 7200 s when condensation is the only active process (Jacobson, 2005).

Coagulation is described using a semi-implicit scheme (Jacobson, 1994). Similarly to the condensation scheme, a semi-implicit coagulation scheme does not require iteration and it is mass conserving. Since coagulation is the (computationally) most time-consuming microphysical process, it is neglected between aerosol pairs for which the coagulation efficiency is low. The detailed list of selected collision pairs accounted for in the coagulation routine is given in Kokkola et al. (2008).

Further details of the SALSA model are given by Kokkola et al. (2008) and Bergman et al. (2012).

2.3 Deposition

Dry deposition of trace gases is calculated with a simple resistance approach (Chamberlain and Chadwick, 1965), which depends on land use and season. Wet scavenging of most gaseous species is proportional to the precipitation intensity. For ozone, hydrogen peroxide and sulfur dioxide, in-cloud scavenging is calculated assuming Henry’s law equilibrium; sub-cloud scavenging is neglected for these species. For ozone, sub-cloud scavenging is likely to be negligible; O3 has a very low solubility in water and wet deposition is not an important sink process for this species. For SO2, the omission of sub-cloud scavenging likely leads to a slight underestimation of the wet-deposition losses; however, SO2 also has a relatively low solubility and a modeling study of wet scavenging of sulfur (Berge, 1993) found that sub-cloud scavenging by precipitation was small (only about 1 % of the total S-deposition was due to sub-cloud scavenging). The absence of sub-cloud scavenging for H2O2 probably leads to a substantial underestimation of wet deposition for this compound. In recent MATCH model simulations that included sub-cloud scavenging of H2O2, it was found that sub-cloud scavenging contributed about 20–40 % to the total wet deposition of H2O2. Wet and dry deposition of gases in the MATCH model is described in detail by Andersson et al. (2007).

Particle dry deposition (including the effects of hygroscopic growth) is calculated using a scheme based on Zhang et al. (2001), adapted to a smaller set of land use classes (water, forest, low vegetation and vegetation-free land areas). More details regarding the dry deposition of particle species are given in Supplement A.

Particles are wet deposited through in-cloud and sub-cloud scavenging. The in-cloud scavenging depends on the fraction of cloud water (or ice) that is precipitated in each grid box, the fraction of the box that is cloudy, the concentration of particles and the fraction of particles in each particle size bin that are inside the cloud droplets. MATCH-SALSA includes a simplified scheme, based on Seinfeld and Pandis (1997), to estimate the fraction of particles that are activated as cloud droplets (and thus are located inside the droplets) – in-cloud particles larger than 80 nm in diameter are considered activated as cloud droplets. This simplified description is used in the present study.

A more advanced (and CPU-time-consuming) formulation for cloud activation is also implemented in MATCH-SALSA. The model can be run coupled to an online cloud activation model that computes CDNC based on the prognostic parameterization scheme of Abdul-Razzak and Ghan (2002). The number of activated particles in each size bin is determined by the particle size distribution, their number concentration and chemical composition, as well as the updraft velocity and the maximum supersaturation of the air parcel. Running the model with particle activation is optional. Optionally, the resulting activated particle fraction in each size bin can be used.
for the calculation of in-cloud scavenging of particles. In this formulation, the activated fraction of each particle class is calculated in each time step for each grid point. The online cloud-activation scheme was not used in the present study, but in Supplement A it is compared to the simplified scheme used here.

The sub-cloud scavenging in the model is treated in a similar way as by Dana and Hales (1976). In MATCH-SALSA, a simplified approach is used where a monodisperse washout coefficient is calculated for each particle bin, and a standard rain drop spectrum\(^1\) is assumed for all precipitation. The washout coefficient (i.e., the fraction of a species that is removed by precipitation below clouds) depends on precipitation amount and takes into account particle collection by Brownian diffusion, inertial impaction and interception. The total wet deposition is the sum of the in-cloud and sub-cloud scavenging.

Further details on the wet scavenging of particles are given in Supplement A and in the companion paper Andersson et al. (2015).

### 3 Model setup

In this section we describe the setup of the simulation used to evaluate MATCH-SALSA in Sect. 4.

Meteorological data are input at regular time intervals; here we used 3-hourly fields from the HIRLAM (Hi-Resolution Limited-Area Model; Undén et al., 2002) weather forecast model. The meteorological data are interpolated to hourly resolution. The model domain covers Europe with a spatial resolution of ca. 44 km. The lowest model level is ca. 60 m thick, and, in total, 22 vertical levels are used; the top level is at about 5 km height. The vertical structure of MATCH-SALSA is the same as in the meteorological model, in this case hybrid \((\eta)\) coordinates, with shallow terrain following layers close to the ground and thicker pressure levels higher up.

For the aerosol size distribution, the following settings were used (see Fig. 2): the first subrange covered the diameter interval 3–50 nm, with three lognormally distributed size bins; the second subrange covered the diameter interval 50–700 nm, with four bins each for soluble and insoluble particle types; the third subrange covered the diameter size range of 700 nm–10 µm, with three size bins for each of the following three particle types: sea salt, soluble particles and insoluble particles.

The top and lateral boundary concentrations of gaseous and particle species, including seasonal variation for some species, were set as described in Andersson et al. (2007). However, boundary concentrations of particulate organic matter (OM) on the southern, western and northern bound-

\(^1\)A representative frontal rain spectrum is used, \(R_g = 0.02\) cm, \(\Sigma_g = 1.86\) (Dana and Hales, 1976).
Figure 3. Calculated annual mean (2007) particle number concentration (PNC) in Europe. Total PNC (sum of all sizes) (a), PNC in size bins PNC$_{3<d<7}$ nm (b), PNC$_{7<d<20}$ nm (c), PNC$_{20<d<50}$ nm (d), PNC$_{50<d<98}$ nm (e), PNC$_{98<d<192}$ nm (f), PNC$_{192<d<360}$ nm (g), PNC$_{360<d<700}$ nm (h). Observed annual mean PNC (filled circles) at the observation sites, Hyytiälä (Finland), Aspvreten (Sweden), Melpitz (Germany) and K-puszta (Hungary), when observed numbers exist in the indicated interval. Units in cm$^{-3}$.

The anthropogenic emissions of gases and primary aerosols are taken from the TNO-MACC (Monitoring the Atmospheric Composition and Climate) emission inventory (Kuenen et al., 2011; Pouliot et al., 2012; see also the MACC project web page http://www.gmes-atmosphere.eu/). The TNO-MACC emissions are given as annual totals. Seasonal, weekday and diurnal variations of the emissions are based on results from the GENEMIS project (http://genemis.ier.uni-stuttgart.de/; Friedrich and Reis, 2004).

The particle emissions of EC and OM$^2$ are distributed over different particle sizes according to sector resolved mass size distributions described by Visschedijk et al. (2009). Details about the size distributions are given in Supplement A (Table S4, p. 16). Emissions from most Selected Nomenclature for Sources of Air Pollution (SNAP) sectors are described by unimodal distributions, while emission from two sectors (international shipping and SNAP sector 4: production processes) are described by bimodal distributions.

$^2$OM emissions are assumed to be distributed over different particle sizes in the same way as OC.

The emissions of oxidized sulfur (SO$_x$) were split into 99 % SO$_2$ and 1 % H$_2$SO$_4$. The split is intended to account for sub-grid-scale processes of gas-phase transformation and gas-to-particle partitioning. The distribution of SO$_x$ emissions between SO$_2$ and more oxidized compounds is discussed in Spracklen et al. (2005b) – the fraction of SO$_2$ increases with grid resolution and it is typically set to between 95 and 100 % in European-scale models. The assumed fractions have large uncertainties and it is not clear from the literature how to optimally partition SO$_x$ emissions between SO$_2$(g), H$_2$SO$_4$(g) and particulate sulfate in modeling studies. The best distribution depends on model resolution (Spracklen et al., 2005b). Lee et al. (2013) have shown that the uncertainties in the sub-grid production of sulfate particles in plumes are more important for cloud condensation nuclei (CCN) uncertainty than the uncertainties in the total anthropogenic SO$_2$ emissions. Since we expect that the choice of distribution of SO$_x$ emissions has a large impact on the model results, we investigate this further in a companion paper (Andersson et al., 2015). The size distribution of the emitted sulfate is the same as for OM. NO$_x$ and non-
methane volatile organic compound emissions were handled in the same way as in Andersson et al. (2007).

4 Evaluation of MATCH-SALSA

In this section we compare our model results to observations at a number of measurement sites throughout Europe. The evaluated model results are extracted from the lowest model level. The statistical measures used are defined in Supplement A. We evaluate the PNC, both in terms of total number concentration, accumulation-mode number concentration and temporal and spatial distribution. We also evaluate the particle mass, including speciation of secondary inorganic aerosol, EC and OC.

4.1 Measurement data

Most measurement data were extracted from EBAS (http://ebas.nilu.no). Details of the stations used in the evaluation of particle number size distribution, PM$_1$, PM$_{2.5}$, EC and OC are given in Supplement A (Table 5). The secondary inorganic aerosol (SIA) components (nitrate, sulfate and ammonium) were evaluated against available measurements in the EMEP network for 2007 (http://www.emep.int).

For evaluation of PNC, four stations from EBAS were chosen to represent different parts of Europe, all classified as rural background sites. Two of the measurement sites, Melpitz (in eastern Germany) and K-puszta (in central Hungary), are relatively close to regions with large emissions. Hyytiälä (inland southern Finland) and Aspvreten (ca. 70 km southwest of Stockholm, in southeastern Sweden) were chosen as regional background stations occasionally impacted by aged particles due to transport from large emission sources in Europe.

4.2 Model evaluation of PNC

Figure 3 shows the modeled annual mean PNC in Europe; both total PNC (Fig. 3a) and the PNC in the different model size bins up to 700 nm are shown (Fig. 3b–g). Corresponding measured annual mean PNC values at the four measurement sites are also displayed in circles for particle sizes where measurements are available.

The largest modeled total PNC values (Fig. 3a) are found in areas with high SO$_x$ emissions (e.g., areas around large point sources in Spain, Poland, southeastern Europe, Ukraine, Russia and the area around Mount Etna; as well as along shipping routes around the Iberian Peninsula and the Gibraltar Strait). These results are in line with other model studies (e.g., Yu and Luo, 2009; Spracklen et al., 2010; Ahlm et al., 2013).

Most of the total PNC in the model resides in the Aitken-mode bins (particle diameters of 7–20 and 20–50 nm; Fig. 3c and d). The highest PNC values in the smallest bin (Fig. 3b), indicating recent nucleation, are found in Russia and Ukraine. Increased values in this bin are also seen along shipping lanes; the modeled high nucleation in marine areas is not in agreement with observations (Heintzenberg et al., 2004). Metzger et al. (2010) found similar nucleation over oceanic regions with large sulfur emissions when traditional activation-type nucleation mechanisms were used; their results, with a new organic activation mechanism, captured the observed lack of nucleation in marine areas, indicating that organic molecules may have a critical role in the nucleation.

The Aitken-mode PNC pattern (Fig. 3c and d) is similar to the total PNC distribution (Fig. 3a). The highest concentrations are found in areas in Spain, Turkey, the Balkan Peninsula, and northwestern Russia, and around the volcano Etna. The highest accumulation mode (50–700 nm) PNC values (Fig. 3e–h) are found in southern Europe. This is partly due to relatively large emissions of primary fine particles and gaseous SO$_x$ and partly due to less precipitation in southern Europe, compared to the north and west, allowing accumulation-mode particles to reside longer in the atmosphere.

We evaluate the model performance (see Figs. 4–6) in terms of total and accumulation-mode particle number concentration (PNC and PNC$_a$, respectively) against observations at the four European surface sites. Due to seasonal differences in emissions and atmospheric processes, we separate performance during summer half-years (April–September) from winter half-years (January–March, October–December). For example, residential biomass burning emissions are much higher during winter than during summer, while biogenic VOC emissions are largest during summer. Both these sources are associated with large uncertainties regarding the emissions and modeling. It should be noted that the size ranges for PNC and PNC$_a$ vary between the stations depending on the measurement interval.

4.2.1 Spatial distribution

Modeled total PNC shows moderate to poor agreement with the observations (Fig. 4a). At most sites, the deviation between observed and modeled mean is large both in summer and winter, and the correlation coefficients for daily mean PNC values are low ($r$ values range from 0.05 to 0.66).

The model captures the general observed features of lower total and accumulation-mode PNC in the northern and northwestern parts of Europe (Fig. 3). Aspvreten and Hyytiälä have the lowest modeled and observed PNC values (Fig. 4a). However, looking in more detail at the stations (Fig. 4) there are some discrepancies. Melpitz clearly has the highest observed total PNC (during both winter and summer; Fig. 4a); the model severely underestimates the PNC at Melpitz and predicts much higher total PNC at K-puszta than at Melpitz. The highest observed accumulation-mode PNC values are found at K-puszta and Melpitz (the PNC values are at similar levels for both seasons and both sites; Fig. 4b); just as for to-
Figure 4. Mean particle number concentration (PNC) during winter (January–March, October–December) and summer (April–September) half-years at four sites in Europe. Top panel (a): mean observed and modeled total PNC. Bottom panel (b): mean observed and modeled PNC in the accumulation mode. The interval above the site name indicates the particle size interval (nm). The number above the season shows the (Pearson) correlation coefficient (r) of daily mean PNC. Note that the size intervals differ between the stations: the same size interval is used for both modeled and observed values at each site. Units in cm$^{-3}$.

Figure 5. Modeled and measured winter (January–March, October–December) and summer (April–September) half-year mean particle number concentration size distribution at four measurement sites in Europe during 2007. Units in cm$^{-3}$.

Spracklen et al. (2010) investigated the impact of different nucleation mechanisms, including the impact of using different nucleation rate coefficients in the activation mechanism. They chose to investigate three rate coefficients, $A = 2 \times 10^{-7}$ s$^{-1}$, $2 \times 10^{-6}$ s$^{-1}$ and $2 \times 10^{-5}$ s$^{-1}$, for which they evaluated the bias to global observations in the free troposphere, and the marine and continental boundary layers. In the continental boundary layer the two lowest nucleation rate coefficients resulted in mean underestimations of $-48$ and $-29$ % respectively, whereas the highest rate resulted in a slight overestimation, on average, of 12 %. The nucleation rate coefficient used in MATCH-SALSA in the present study is near the lower end of the interval ($A = 7.3 \times 10^{-7}$ s$^{-1}$), which may explain our underestimation of nucleation at the central and northern sites. In fact, the nucleation rate coefficient in the activation scheme should be site and time dependent in the European boundary layer (e.g., Sihto et al., 2006; Riipinen et al., 2007): observations of this coefficient vary by $\sim 4$–$5$ orders of magnitude for different European measurement sites, ranging from $3.3 \times 10^{-8}$ to $3.5 \times 10^{-4}$ s$^{-1}$ (Riipinen et al., 2007). Thus, a more advanced description of the nucleation, e.g., time-varying and space-varying rate coefficients, should be included in MATCH-SALSA.

Organic nucleation is not included as a nucleation process in the evaluated base case simulation, resulting in a possible underestimation of nucleation in areas with high BVOC concentrations and possibly overestimated nucleation in regions with low concentrations of organic aerosol precursors (similar to the overestimated nucleation in the model in oceanic high-SO$_4$ regions, discussed above). This may also be an explanation for the overestimated nucleation at K-puszta. Sensitivity tests including organic nucleation will be discussed.
4.2.2 Size distribution

The modeled and observed size distributions at all four stations are shown in Fig. 5. A common feature for the PNC size distribution is that PNC values are underestimated, or on the same level as the measurements, except at K-puszta, where the PNC values of the smallest particles are overestimated both during winter and summer (Fig. 5c). At K-puszta, the mean total PNC is overestimated but the PNC in the accumulation mode is underestimated (Fig. 4). At all stations, the shape of the size distribution is captured relatively well, but during winter at K-puszta (Fig. 5c) and during summer at Aspvreten (Fig. 5a) and Hyytiälä (Fig. 5b), the modeled size distribution peaks at smaller sizes than in the observations. The reason for the maximum occurring at too-small sizes, in combination with underestimated accumulation-mode PNC, may be too-weak condensation onto nucleating particles in the model. Bergman et al. (2012) also evaluated the modeled particle number size distribution at measurement sites, including Aspvreten, Melpitz and Hyytiälä, and found that the model ECHAM5-HAM underestimated the number concentrations at all three measurement sites for sizes larger than about 20 nm, both when using the aerosol dynamics modules of M7 and SALSA. SALSA performed better than M7 for PNC above 100 nm at the dirtier measurement sites (e.g., Aspvreten and Melpitz) while M7 performed better at cleaner sites (e.g., Hyytiälä), but the differences between the two models were not large. Bergman et al. (2012) concluded that the growth in SALSA probably was too slow.

4.2.3 Temporal evolution

Figure 6 shows the modeled and observed temporal variation of the daily mean PNC at the four sites. New particle formation in the model is seen in the form of peak concentrations of the smallest particle sizes. These peaks coincide with the observed maximum total PNC on some occasions; sometimes there is a time shift of a few days between the modeled and observed peaks. Many of the observed nucleation peaks at Hyytiälä (Fig. 6a), Aspvreten (Fig. 6b) and Melpitz (Fig. 6d) are not seen in the model results. Reddington et al. (2011) simulated hourly PNC with diameters larger than 15 nm using the GLomap model and evaluated these against measurements over 1 month (May 2008). Depending on the nucleation parameterization, the correlations ($R^2$)
Figure 7. Modeled annual mean concentrations (for 2007) of PM$_{10}$ (a; peak at 37 µg m$^{-3}$ in Moscow) and its particle components: elemental carbon (b), organic matter (c), anthropogenic primary inorganic aerosol (d), sulfate (e), nitrate (f), ammonium (g) and sea salt (h). Units in µg m$^{-3}$.

between model and measured PNC were less than 0.03 at Aspvreten, Hyytiälä and Melpitz, and less than 0.10 at K-puszta. For PNC with larger sizes (> 100 nm), the correlations were less than 0.01 at K-puszta and higher at the other sites (< 0.13 at Aspvreten, < 0.20 at Melpitz and < 0.45 at Hyytiälä). Spracklen et al. (2006), on the other hand, captured the nucleation at Hyytiälä very well with GLOMAP; however, they only studied a short period (22 days) in May with clear sky conditions. With MATCH-SALSA, the hourly correlations ($R^2$), for single months of 2007, for PNC with a diameter larger than 50 nm are in the range of 0–0.17 for Hyytiälä (for May: 0), < 0–0.20 for Aspvreten (May: < 0), < 0–0.20 for K-puszta (May: 0.01) and < 0–0.41 for Melpitz (May: 0.41). These low correlations illustrate that nucleation events are difficult to capture by models when running over long time periods for a large region. One reason for this is the coarse scale of the model – each grid cell is representative of a large area (for MATCH-SALSA, ca. 44 × 44 km$^2$ and for GLOMAP 2.8° × 2.8°). Another reason is that the simple activation-type nucleation scheme needs a site- and time-varying nucleation parameter to work well (Riipinen et al., 2007). Furthermore, the wintertime nucleation peaks in the observations that are absent in the model may also be explained by a temperature dependence in the nucleation that is not accounted for in the model (Dal Maso et al., 2005); or the observed peaks could be of local origin that can not be captured by a regional-scale CTM.

The best correlation between modeled and observed daily mean PNC is found at Melpitz ($r = 0.70$; Fig. 6d) but the model underestimates PNC most of the time; the observed PNC is almost always high at this site. The model grossly overestimates the total PNC at K-puszta (Fig. 6c) during summer, but the temporal variation for particles sizes > 20 nm follows the measurements fairly well ($r = 0.32$); during winter the model PNC is in better agreement with the observations. At Hyytiälä (Fig. 6a), a lot of nucleation is observed; this is not captured by the model, possibly due to the lack of organic nucleation in this simulation; this will be discussed in detail in the companion paper (Andersson et al., 2015).

Spracklen et al. (2010) calculated the correlations ($R^2$) between monthly mean modeled and observed PNC for sites where the monthly means varied by more than a factor of 2 during the year 2000 (Aspvreten was excluded due to too-small of a variation). K-puszta was not included in the assessment. Their results were $R^2 = 0.39$ and 0.28 for the sites
Hyytiälä and Melpitz, respectively. With MATCH-SALSA, we obtained \( R^2 = 0.67 \) and 0.08, respectively, for the same sites (for PNC with diameter > 50 nm). Using kinetic nucleation description Spracklen et al. (2010) achieved higher monthly correlations than with activation-type nucleation at most evaluated sites, including Hyytiälä and Melpitz.

4.3 Model evaluation of particle mass and composition

Simulated annual average total PM\(_{10}\) and the chemical components that constitute PM\(_{10}\) are displayed in Fig. 7. The largest concentrations of total PM\(_{10}\) (Fig. 7a) are found in anthropogenic emission hotspots (e.g., northern Italy, Moscow and eastern Ukraine) and over the Atlantic Ocean and parts of the Mediterranean Sea. The highest modeled concentrations over land are due to large anthropogenic emissions of primary anthropogenic inorganic aerosol (Fig. 7d), except in northern Italy, where there is a large contribution from ammonium nitrate (Fig. 7f–g), and in southeastern Europe and some sulfur emission hotspots, where sulfate (Fig. 7e) dominates PM\(_{10}\). Over the oceans (and in large parts of western and northern Europe), the largest contribution to PM\(_{10}\) is from sea-salt particles (Fig. 7h); important sulfate contributions are also seen, especially around Mount Etna and the eastern Mediterranean Sea. OM (Fig. 7c) gives the largest modeled non-sea-salt contributions to PM\(_{10}\) in northern Europe and also in some parts of southern and western Europe. In the following subsections we present evaluation statistics for the different particle components.

4.3.1 Secondary inorganic aerosol (SIA)

Statistics from the evaluation for SIA components (particulate sulfate, SO\(_4^{2-}\); nitrate, NO\(_3^-\); ammonium, NH\(_4^+\)) are shown in Table 1 and in Supplement A (Tables A15–A19 and Figs. A32–A36). In order to avoid biases due to possible incorrect separation of gas-phase and particle-phase nitrogen in the measurements, we also include evaluation results for total nitrate (TNO\(_3^-\): HNO\(_3\)(g) + NO\(_3^-\) (p)) and total reduced nitrogen (TNH\(_4^+\): NH\(_3\)(g) + NH\(_4^+\)(p)).

Sulfate has a low mean bias (4%) whereas the root mean square error normalized to the observed mean (CV(RMSE)) is around 50%. The average (Pearson) correlation coefficient (average \( r \) at the different sites, based on daily means) is 0.52 and the spatial correlation coefficient (“spatial” \( r \) for the annual mean concentration at all the stations) is 0.57. The model performance for the nitrogen compounds (NO\(_3^-\), HNO\(_3\)+NO\(_3^-\), NH\(_4^+\) and NH\(_3\)) at individual stations is of similar quality as that for sulfate. The model underestimates the concentration of the nitrogen components by about 10–20%, while the CV(RMSE) values are a bit lower than for sulfate (ranging from 36 to 49% for the four N components). The average \( r \) at the measurement sites vary between 0.44 and 0.59 for the N components, whereas the spatial correlation coefficients are higher (between 0.79 and 0.87).

![Figure 8. Evaluation of elemental carbon (EC) for 2007](image)

Figure 8. Evaluation of elemental carbon (EC) for 2007 (a: April–September mean; b: January–March, October–December mean). Observed and modeled mean concentrations (µg m\(^{-3}\)), and correlation coefficients of daily mean concentrations are indicated below the bars. The number of daily mean values is indicated by the numbers in the parentheses. Correlation coefficients were calculated for measurement sites with more than 10 daily observations. Site codes as defined by EMEP, see Supplement A, Table 5.

4.3.2 Elemental and organic carbon

The organic aerosol measurements used for model evaluation in this study are organic carbon (OC) measurements. The model describes organic matter (OM). In the evaluation we assume an OM:OC ratio of 1.4. The actual ratio varies with location and season (e.g., Simon et al., 2011) and is usually between 1.25 and 2.5, with a greater ratio for more aged OM (Turpin et al., 2000; Kupiainen and Klimont, 2007; Aiken et al., 2008). The choice of a fixed OM:OC ratio for the evaluation will lead to model under- or overestimation, depending on the measurement site and time of year. Figures 8 and 9 show the annual observed and modeled mean concentrations of EC (Fig. 8a–b) and OC (Fig. 9a–b) at individual measurement sites, as well as the associated correlation coefficients, based on daily data; detailed results are given in Table 2.

Both EC and OC are underestimated at many of the sites. The underestimation is especially large at the Italian sites and Payerne (Switzerland) during winter, for both EC (Fig. 8b) and OC (Fig. 9b), and for EC at Melpitz (Fig. 8a–b). Correlation coefficients are higher for EC than OC; OC is more complicated to model than EC, since it is a combination of primary and secondary components, many of them semi-volatile. The reasons for the model–measurement differences are likely to vary between seasons and locations; e.g., winter-time emissions from residential combustion are often underestimated (e.g., Simpson et al., 2007; Gilardoni et al., 2011;
Table 1. Comparison of modeled secondary inorganic aerosol (SIA) components to daily observed concentrations. Average results covering available measurements for the year 2007 (results for individual stations are given in Tables A15–A19 in Supplement A). In addition to the SIA components, the total nitrate (\(\text{TN}O_3 = \text{HNO}_3 + \text{NO}_3\)) and total reduced nitrogen (\(\text{TNH}_x = \text{NH}_3 + \text{NH}_2\)) are also evaluated. 

| Measure | Global/temporal | Spatial |
|---------|----------------|---------|
| NO\textsubscript{2}\textsuperscript{−} | Mean obsd \(\mu g\text{g}^{-1}\text{N m}^{-3}\) | Mean model \(\mu g\text{g}^{-1}\text{N m}^{-3}\) | Rel. bias | mean\textsuperscript{*} | mean\textsuperscript{*} CV(RMSE) | No. obs | Rel. bias | r | CV(RMSE) | No. stns |
| SO\textsubscript{4}\textsuperscript{2−} | 0.63 | 0.65 | 4 | 0.52 | 16 033 | −6 | 0.57 | 53 | 52 |
| NO\textsubscript{3} | 0.40 | 0.32 | −21 | 0.44 | 72 49 | −22 | 0.83 | 48 | 23 |
| TNO\textsubscript{3} | 0.49 | 0.40 | −19 | 0.59 | 11 039 | −21 | 0.85 | 41 | 35 |
| NH\textsubscript{4}\textsuperscript{+} | 0.72 | 0.64 | −12 | 0.57 | 97 28 | −11 | 0.79 | 37 | 31 |
| TNH\textsubscript{4} | 1.27 | 1.01 | −21 | 0.53 | 10 137 | −20 | 0.87 | 38 | 32 |

* Weighted average of correlation coefficients and CV(RMSE) values at individual stations.

Table 2. Statistics of the comparison of MATCH-SALSA results to daily observed concentrations of elemental carbon (EC) and organic carbon (OC) in PM\textsubscript{1}, PM\textsubscript{2.5} and PM\textsubscript{10} for the year 2007. Obs stands for measured concentration, Mod for modeled concentration, MAE for mean absolute error, \(r\) = Pearson correlation coefficient (only calculated for sites with more than 10 measurements). Relative bias and MAE are given as percentage of the observed average. For further information about the measurement stations, see Table A5 in Supplement A.

Bergström et al., 2012), and during the summer half-year biogenic VOC emissions and wildfires may be more important sources of carbonaceous particles.

At Ispra (IT04) in northern Italy, the model performs fairly well for carbonaceous aerosol during summer but greatly underestimates both EC and OC during wintertime (Figs. 8 and 9 and Fig. A15 in Supplement A). One reason may be the underestimation of residential wood combustion emissions (e.g., Bergström et al., 2012). The model also underestimates NO\textsubscript{2} (by 43 % in summer and 51 % in winter). Both the observations and the model results show a clear seasonal cycle with higher concentrations during winter for NO\textsubscript{2} as well as for EC and OC. However, for EC and OC the model underestimation during winter is much larger (−74 % and −87 %).
Figure 9. As Fig. 8 but for organic carbon (OC).

respectively) than during summer (−20 and −37 %, respectively) (Supplement A, Fig. A15). The poor model performance for EC and OC during winter is likely due to a lack of emissions from one or more emission sectors, with greater emissions of EC and OC during winter, but relatively small contribution to NO$_2$. This work therefore supports the results of previous studies (e.g., Gilardoni et al., 2011) that have concluded that residential wood combustion emissions are likely underestimated in current emission inventories, at least in the area around Ispra.

For the German site Melpitz, the model grossly underestimates EC throughout the year (Supplement A, Fig. A37). OC is generally captured fairly well at the station, with underestimation of OC in PM$_{2.5}$ and PM$_{10}$ (but not PM$_1$) during winter and overestimation for OC in PM$_{2.5}$ and underestimation (−25 %) in PM$_{10}$ during summer (Supplement A, Fig. A38). Part of the reason for the relatively high EC measurements at Melpitz is that the measurement technique used at this site to separate OC from EC has no charring correction and is expected to lead to too-high EC values and to underestimate OC (see Genberg et al., 2013, and references therein). There are large peaks during spring and late autumn of OC (and EC) in PM$_{2.5}$ and PM$_{10}$, which are clearly underestimated. The peak in the beginning of April coincides with a vegetation fire episode (Genberg et al., 2013); the earlier peaks and the late autumn peaks are perhaps more likely due to residential combustion or other missing/underestimated sources, possibly, also due to fires in eastern Europe (Jönsson et al., 2013). Stern et al. (2008) suggested that the large underestimations of EC may be an indication that emissions in the central European region were underestimated during these episodes.

4.3.3 Total particulate matter (PM$_1$ and PM$_{2.5}$)

Evaluation of PM$_1$ and PM$_{2.5}$ at 28 measurement sites is presented in Fig. 10 and in Supplement A (Table A21 and Fig. A39); detailed time series plots are given in Supplement A, Figs. A17, A40–A41. For PM$_1$ the annual means at the sites with the lowest observed concentration (three Nordic sites: NO01, FI17, DK41) are overestimated by the model. On the other hand, at the central European sites the PM$_1$ concentrations are much better captured. The model underestimates PM$_{2.5}$ by 14 % (spatial average) and the spatial correlation coefficient is 0.64. Six of the 35 evaluated annual means (PM$_1$ and PM$_{2.5}$) deviate by more than 50 % from the measured concentrations. The largest underestimations of PM$_{2.5}$ are seen at the sites with the highest observed annual mean. The underestimation of PM$_{2.5}$ can be due to a
number of reasons, including underestimated emissions, too-
short aerosol lifetime or too-small secondary aerosol produc-
tion. There is probably too little EC and OC in the model, at
least at some of the sites, which can be explained by under-
estimated emissions.

The treatment of sea spray needs to be further evaluated
and the model scheme for sea-salt particles may need to be
updated. For PM$_1$ the annual means at the sites with the
lowest concentrations are overestimated by the model. This
seems to be partly due to overestimation of sea salt. Eval-
uation scores for modeled PM$_1$ and PM$_{2.5}$ excluding sea-salt
aerosol in the total PM mass (see Supplement A, Table A21,
Figs. A18 and A39) give higher correlation coefficients for
daily mean PM$_{2.5}$ or PM$_1$ at 22 of the 28 sites (and lower at
only one site) than when sea salt is included. This is an in-
dication of too much sea salt at the wrong time. It may be due
to too-strong sea-salt emissions and/or too-weak sink processes
for the sea salt, since substantial improvements in correlation
are seen also at some far inland sites.

5 Conclusions

We have implemented the sectional aerosol dynamics model
SALSA (Kokkola et al., 2008) in the European-scale CTM
MATCH (Multi-scale Atmospheric Transport and Chem-
istry; Robertson et al., 1999). The new model is called
MATCH-SALSA. It includes aerosol microphysics with sev-
eral options for nucleation, wet scavenging and condensa-
tion.

In general, the model reproduces the observed lower parti-
cle number concentration (PNC) in northern and northwestern
Europe and remote regions than in central Europe. The
model peak in the particle number size distribution occurs at
the same or smaller particle size as the observed peak. To-
tal PNC is underestimated at northern and central European
sites. The low nucleation rate coefficient used in this study is
probably one important factor for the underestimation, al-
though other reasons may also contribute; e.g., organic nu-
cleation is not included and EC is not emitted in the Aitken
mode. The model performs well for particle mass, includ-
ing secondary inorganic aerosol components. Particulate ele-
mental and organic carbon concentrations are underestimated
at many of the sites.

Before using the model for simulating total PM$_{2.5}$, the
SOA formulation needs further improvements. MATCH-
SALSA is computationally costlier than MATCH, which also
puts restrictions on when the model can be used.

The development of the MATCH-SALSA model is con-
tinuing and in the near future focus will be on the following
areas:

- An updated biogenic emission module is needed for re-
  alistic treatment of BSOA formation. Updating the bio-
  genic SOA scheme will likely have a large impact on
  modeled PM$_{2.5}$ and also affect the model performance
  for total PNC through impacts on nucleation and cond-
 ensation.
- Updating the nucleation rate coefficients possibly with
  time- and space-varying rate coefficients.
- Nitrogen gas–particle partitioning should be coupled to
  the microphysics. This may increase condensational
  growth, which is underestimated in the present version
  of the model.
- Emissions from open fires (wildfires and agricultural
  burning activities) will be added to the model.
- Dust emissions from road traffic, agricultural activities
  and non-vegetated soils including desert areas should be
  included in the model.
- Processes affecting sea salt need further work and eval-
  uation. This study has shown large modeled sea-salt
  peaks that are not seen in the measurements. Both emis-
  sions and deposition of sea-salt particles should be in-
  vestigated.
- Emission inventories need to be improved, especially
  for EC and OC emissions.

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