

Sectional aerosol
module for large
scale applications

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SALSA – a Sectional Aerosol module for Large Scale Applications

H. Kokkola¹, H. Korhonen², K. E. J. Lehtinen^{1,3}, R. Makkonen⁴, A. Asmi²,
S. Järvenoja^{2,†}, T. Anttila², A.-I. Partanen¹, M. Kulmala⁴, H. Järvinen²,
A. Laaksonen^{2,3}, and V.-M. Kerminen²

¹Finnish Meteorological Institute, Kuopio Unit, P.O. Box 1627, 70211 Kuopio, Finland

²Finnish Meteorological Institute, P.O. Box 503, 00101 Helsinki, Finland

³Department of Physics, Univ. of Kuopio, P.O. Box 1672, 70211 Kuopio, Finland

⁴Department of Physical Sciences, P.O. Box 64, 00014 Univ. of Helsinki, Finland

[†]deceased

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Correspondence to: H. Kokkola (harri.kokkola@fmi.fi)

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Abstract

The sectional aerosol module SALSA is introduced. The model has been designed to be implemented in large scale climate models, which require both accuracy and computational efficiency. We have used multiple methods to reduce the computational burden of different aerosol processes to optimize the model performance without losing physical features relevant to problematics of climate importance. The optimizations include limiting the chemical compounds and physical processes available in different size sections of aerosol particles; division of the size distribution into size sections using size sections of variable width depending on the sensitivity of microphysical processing to the particles sizes; the total amount of size sections to describe the size distribution is kept to the minimum; furthermore, only the relevant microphysical processes affecting each size section are calculated. The ability of the module to describe different microphysical processes was evaluated against explicit microphysical models and several microphysical models used in air quality models. The results from the current module show good consistency when compared to more explicit models. Also, the module was used to simulate a new particle formation event typical in highly polluted conditions with comparable results to a more explicit model setup.

1 Introduction

Aerosols are currently an inherent part of many types of large-scale atmospheric models, including regional and global climate models, air quality models and chemical transport models. There are two things that need to be kept in mind when designing the description of aerosols in these models. First, the climatic and other effects of atmospheric aerosols have been demonstrated to be sensitive to aerosol size distribution, size-resolved chemical composition and the mixing state of the particles (Myhre et al., 2004; Chen and Penner, 2005). Second, understanding the linkage between the emissions of aerosols or their precursors and various impacts by these aerosols is not

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possible without consideration of the atmospheric processes that modify the different aerosol properties (Adams and Seinfeld, 2002; Spracklen et al., 2005).

5 Treating aerosols in a large-scale modeling framework is always a compromise between the detail of description and computational efficiency. One approach in this regard is to approximate the aerosol population as a sum of a small number of log-normal modes. The modal approach has been used in regional air quality models for more than a decade (Binkowski and Shankar, 1995; Ackermann et al., 1998; Binkowski and Roselle, 2003), and more recently it has also been applied to various global modeling frameworks (Ghan et al., 2001; Wilson et al., 2001; Stier et al., 2005). The drawback of the modal approach is that many of the processes relevant to indirect climatic aerosol effects such as aerosol formation and growth, cloud processing, and aerosol ageing in terms of its cloud nucleating properties, are difficult to simulate properly. Also, the calculation of the effective radii of the particles can be inaccurate when using modal approach (Weisenstein et al., 2007).

15 A more general way of treating aerosols in large-scale models is to describe the aerosol size distribution with a limited number of moments (McGraw, 1997). All aerosol processes in this approach are tied into the moments and thus getting information about their physical values at any time requires a separate retrieval. The moment approach is computationally very effective but has basically the same drawbacks as the modal approach. Additionally, the moment approach is not intuitive for use, as the modal values themselves are not observed parameters beyond the first few integer values.

25 In principle, sectional models can be used to avoid the problems associated with modal and moment approaches. However, the calculation of microphysical processes affecting the aerosol size distribution using sectional approach is usually more computationally demanding than modal or moment method approaches. On the other hand, a sectional model can describe different microphysical processes more accurately than modal and moment methods. Because of the computational demands, sectional approaches have often neglected one or more major aerosol types and assumed an

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internal mixture between the simulated components (e.g. [Jacobson, 2001](#); [Rodriguez and Dabdub, 2004](#); [Spracklen et al., 2005](#)). Another way of tackling this problem is to apply the sectional approach for some of the aerosol sizes and to treat the rest of the aerosol sizes using either modal or bulk mass based approach ([Liao and Seinfeld, 2005](#); [Liu et al., 2005](#); [Reddy et al., 2005](#)).

There are several ways to improve the efficiency of a sectional aerosol model. Most importantly, the model development should concentrate on the main application of the model (e.g. climate effects in this case) and try to minimize less important processes on different aerosol sizes and compositions. Especially, reduction of number of computational tracers is of major importance, as they affect the model efficiency severely. We have developed a new aerosol model, which tries to approach the simulations of aerosol dynamics from climate effect point of view using a highly flexible sectional model approach, with computational efficiency as a major design criteria.

2 Model description

There are several ways by which the computational efficiency of multi-component sectional aerosol models could be improved to make them more suitable for large scale applications. We postulate the major design choices of our improvements:

1. The width of the particle size bins need not be fixed as usually assumed. Instead, lower size resolution could be used in regions influenced less by microphysical processes, or in regions that are less important for the problem under investigation;
2. Since different aerosol types are concentrated over different parts of the particle size spectrum, the set of chemical components included in simulations could vary between different size regions;
3. The relative importance of different microphysical processes varies strongly with particle size. This makes it possible to simplify the treatment of individual aerosol

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processes, or to neglect them altogether, in some particle sizes. The model developed in this study is designed to use the simplification mentioned above to minimize the amount of calculations and the variables passed on to the host climate model to maximize the computational efficiency of the model without compromising too much the accuracy of the aerosol model.

Using these basic postulates as our design criteria, we have created a highly flexible modelling framework SALSA. In the following we present the description of the model and the suggested base configuration for climate studies. The model also introduces organic compounds which have, until recently, been usually neglected in sectional models used in large scale climate applications.

2.1 Representation of the aerosol size distribution

In the SALSA model, the particle size spectrum is divided into three subranges based on the following criteria:

1. What aerosol sources and sinks are important?
2. What atmospheric processes are relevant?
3. What chemical components need to be included?
4. How the aerosol mixing state should be described?
5. Which aerosol quantities need to be modelled accurately to obtain relevant climate or air quality effects?

After this, the number of size bins, chemical components, externally mixed groups of particles, and simulated processes can be optimized for each sub-range of the distribution.

The degree of internal mixing of the particles is important for determining the optical properties of the particles, their ability to act as Cloud Condensation Nuclei (CCN),

and their lifetime under cloudy conditions. Due to the diverse aerosol sources of accumulation and coarse mode particles, and the relatively slow ageing of large particles due to microphysical processes, the degree of internal mixing usually decreases with increasing particle size.

5 The optimization of the position of the limits between the subranges in the base configuration was done by calculating the number of activating droplets for three different size distributions using updraft velocities of an adiabatically rising air parcel between 0.1–1.0 m/s. The positions of the limits were varied and the results were compared to those calculated using a detailed model setup for 300 size bins in size space. The
10 chosen positions for the limits were chosen which gave the smallest least square error. The method for the calculation of CDNC is presented in more detail later in the manuscript in Sect. 2.6.

The three size distributions used in the optimization were urban, rural, and marine size distributions reported by Jaenicke (1993). In the optimization procedure, the particles were assumed to be internally mixed consisting purely of sulfate.

15 Based on this optimization, we have selected the subranges of particles and their chemical composition as follows: subrange 1: particles with diameter $3 \text{ nm} < D_p < 51 \text{ nm}$, subrange 2: $51 \text{ nm} < D_p < 730 \text{ nm}$ and subrange 3: $730 \text{ nm} < D_p < 10 \mu\text{m}$. A schematic of the division into sections of the size distribution is shown in Fig. 1. The figure shows
20 the sections in different subranges, the chemical compounds treated in each subrange and in externally mixed size bins.

The division of size distributions in the size bins presented here is the default model setup and designed for purposes of large scale models. Nevertheless, the number of size bins is adjustable and can be chosen differently for different types of simulations
25 or model configurations.

Subrange 1:

Particles with diameter less than 51 nm are estimated to mainly originate from nucleation processes rather than from primary sources. The composition and size of these particles are strongly affected by aerosol dynamics, coagulation and condensation with

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principal compounds being low-volatile material. Particles of this size range can be assumed to be approximately internally mixed. Particles in this size range coagulate effectively with large particles, but for example cloud processing should have no direct effect on the number concentration of these particles. In the SALSA model, there are three internally mixed size sections for particles in subrange 1. The compounds treated in subrange 1 are sulfate and low-volatile secondary organics.

Subrange 2:

Particles in this range are assumed to evolve from subrange 1 by aerosol processing, but this subrange also includes particles from primary emissions. The main chemical components for this subrange are sulfate, primary organics, black carbon, low-volatile secondary organics and sea salt. In the aerosol model, the subrange is divided into four size sections. Since in the atmosphere there is some degree of external mixing for particles of these sizes, and especially as the composition of these external mixtures can play a significant role in cloud activation, the model uses two externally mixed parallel size bins for each size section (2a and b; see Fig. 1). The sizes of particles in subrange 2 are fairly insensitive to processing by coagulation and dry deposition. The particles in this subrange are important in cloud activation as they are the dominant source of cloud droplets. Also, wet deposition is a significant sink for these particles. For these reasons, subrange is described using a higher resolution of size sections.

Subrange 3:

Particles larger than 730 nm contain primary particles from sea salt emissions, dust emissions and primary biogenic emissions also including all the chemical compounds found in smaller particles. Coarse particles have a large impact on the number concentration of the particle population since they effectively collect smaller particles by coagulation and are deposited by both dry and wet deposition. Still, the sizes of coarse particles are not affected significantly by aerosol dynamics.

In the model, subrange 3 is divided into three size sections. The degree of external mixing is high for particles in this size range, so the sections include three parallel size bins (referred to as 3a, b, and c; see Fig. 1).

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2.2 Treatment of microphysical processes

The microphysical processes included in the model are nucleation, coagulation, condensation, and hydration. The processes are calculated for each fixed time step independently, except for nucleation and condensation which are calculated simultaneously to represent the competition between these processes more accurately. The redistribution of particles which grow or shrink out of the boundaries of their initial size bin are redistributed to the appropriate new size bins at the end of each time step, when all microphysical processes have been already taken into account.

For sparse size bins, the moving center scheme has proven to be an efficient way to describe the time dependent development of particle size distribution (Jacobson, 2005) and this method is also used in the current aerosol model for particles in the subranges 1 and 2. Since aerosol dynamics do not affect particles larger than 730 nm significantly, size bins in subrange 3 are treated in a fixed sectional grid to decrease the calculation time and the number of tracers in the model.

The division between insoluble and soluble bins in subrange 2 requires further explanation. As insoluble particles grow in subrange 2 they collect soluble compounds from condensation and thus their water uptake increases. We have selected the particles that are able to form cloud droplets in 0.5% supersaturation to be “soluble”, rest “insoluble”. This way we have tried to get more climatologically relevant choice of solubility, which can provide the most appropriate measure of particle ability to act as a CCN.

2.3 Nucleation and condensation

Nucleation is taken into account by calculating the rate of formation of 3 nm sized particles resulting from nucleation processes. This is calculated using the expression for the parameterized formation rate of detectable 3 nm particles given by Kerminen and Kulmala (2002). The parameterization calculates the formation rate J_{app} of 3 nm particles

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using the “real” nucleation rate $J(t)$ using equation

$$J_{\text{app}}(d_p, t') = J(t) \exp\left(\frac{\eta}{d_p} - \frac{\eta}{d_{\text{nuc,ini}}}\right), \quad (1)$$

where $t' - t = (d_p - d_{\text{nuc,ini}})/\text{GR}$ is the time difference between formation of nuclei of critical diameter $d_{\text{nuc,ini}}$ and their growth to diameter d_p , GR (m s^{-1}) denoting the nuclei growth rate calculated according to [Kerminen and Kulmala \(2002\)](#), where the details of calculating parameters in Eq. (1) are given. The reason for this shift in size-space is the choice of minimum size of the particles in SALSA.

The “real” nucleation rate in Eq. (1) can be calculated in the default model setup using description for binary nucleation ([Vehkamäki et al., 2002](#)), ternary nucleation ([Napari et al., 2002a,b](#)), kinetic nucleation ([Sihto et al., 2006](#); [Riipinen et al., 2007](#)), or activation type nucleation ([Kulmala et al., 2006](#); [Riipinen et al., 2007](#)). However, other methods of nucleation mechanisms can be used, if the host model together with SALSA can provide the necessary inputs for parameterizations.

Mass transfer of gas molecules to particles is calculated using the condensation equation summed over all size bins ([Lehtinen et al., 2004](#))

$$\frac{dC_i}{dt} = - \sum_{j=1}^n \beta_{i,j} C_i, \quad (2)$$

where C_i is the gas phase concentration of the condensing gas phase compound i in units m^{-3} , and $\beta_{i,j}$ is the collision rate between the gas molecules and pre-existing particles for size bin j . $\beta_{i,j}$ can be expressed as

$$\beta_{i,j} = \frac{2(d_i + d_j)(D_i + D_j)(Kn + 1)}{0.377Kn + 1 + \frac{4}{3\alpha}(Kn^2 + Kn)}, \quad (3)$$

where d_i and d_j are the diameters and D_i and D_j the diffusion coefficients of the condensing molecule and particle in bin j , respectively. The Knudsen number Kn

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is defined as

$$Kn = \frac{2\lambda}{d_i + d_j}, \quad (4)$$

where λ is the mean free path of the condensation process

$$\lambda = \frac{3(D_i + D_j)}{\sqrt{\bar{c}_i + \bar{c}_j}}. \quad (5)$$

5 Here \bar{c}_i and \bar{c}_j are the thermal speeds of the molecule and particle, respectively.

Similarly, the change of particle phase concentration $c_{i,j}$ of compound i in each size bin j is

$$\frac{dc_{i,j}}{dt} = \beta_{i,j} C_i. \quad (6)$$

For a finite time step Δt Eq. (6) can be written as

$$10 \quad c_{i,j}(t) = c_{i,j}(t - \Delta t) + \Delta t \beta_{i,j} C_i(t). \quad (7)$$

Since mass is conserved,

$$C_i(t) + \sum_{j=1}^n c_{i,j}(t) = C_i(t - \Delta t) + \sum_{j=1}^n c_{i,j}(t - \Delta t). \quad (8)$$

Substituting (7) into (8) and solving gas phase concentration, we get

$$C_i(t) = \frac{C_i(t - \Delta t)}{1 + \Delta t \sum_{j=1}^n \beta_{i,j}}. \quad (9)$$

15 Using this equation, the particle phase concentration can be calculated from

$$c_i(t) = c_i(t - \Delta t) + \frac{\beta_{i,j}}{\sum_{j=1}^n \beta_{i,j}} (C_i(t - \Delta t) - C_i(t)) \quad (10)$$

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When also nucleation is taken into account, an extra term describing nucleation is added to Eq. (3) for the smallest size bin 1. Including this term in Eq. (3), the collision rate for H₂SO₄ in size bin 1 is defined as

$$\beta_{\text{H}_2\text{SO}_4,1} = 2 \frac{(d_{\text{H}_2\text{SO}_4} + d_1)(D_{\text{H}_2\text{SO}_4} + D_1)(Kn + 1)}{0.377Kn + 1 + \frac{4}{3\alpha}(Kn^2 + Kn)} + \frac{J_{\text{app}}n_{3\text{nm}}}{C_{\text{H}_2\text{SO}_4}(t - \Delta t)}. \quad (11)$$

Here, $n_{3\text{nm}}$ denotes the number for molecules in 3 nm particles.

The number concentration of particles in size bin 1 is then updated according to equation

$$n(t) = n(t - \Delta t) + \left(\frac{J_{\text{app}}(c_{\text{H}_2\text{SO}_4}(t) - c_{\text{H}_2\text{SO}_4}(t - \Delta t))}{(J_{\text{app}} + \beta_{\text{H}_2\text{SO}_4,1} C_{\text{H}_2\text{SO}_4}) n_{3\text{nm}}} \right) \quad (12)$$

2.4 Hydration

The equilibrium sizes of particles in different size bins j are calculated using the ZSR method (Stokes and Robinson, 1996). The water content c_w (kg m⁻³) in particles is given by equation

$$c_w = \sum_{j=1}^n \frac{c_j}{m_{j,a}}. \quad (13)$$

In the equation, c_j is the molar concentration of species j , and $m_{j,a}$ is the molality of solute species alone in the solution. Summation in (13) is done over all solute species in each size bin. To decrease the amount of calculation, hydration is not calculated for insoluble size bins in subranges 3b and 3c.

The binary molalities for inorganic salts are calculated using parameterizations given by Jacobson (2005). For organic compounds, binary molality is calculated using equation

$$m_{j,a} = 1/(a_w v_w) - 1/v_w, \quad (14)$$

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where v_w is the molar volume of water and a_w is the water activity. Equation (14) assumes ideal behavior for the organics.

2.5 Coagulation

In a typical large scale aerosol code, coagulation calculations are one of the most computationally expensive among the different microphysical processes. To decrease the calculation time for this process, exact coagulation coefficients are calculated off-line for each pressure level at the beginning of the simulation. During the model simulation, coagulation coefficients are scaled according to current particle wet size using linear scaling with respect to diameter. Also, the coagulation is not calculated for all collision pairs. In subranges 1 and 2a, the coagulation in each size bin is calculated with larger particles. In subrange 2b, the coagulation for each size bin is calculated only with larger particles, and particles in the parallel size bin in subrange 2a. The collisions between particles in subrange 3 are not taken into account. The changes in particle number and volume concentrations due to coagulation are then calculated using the semi-implicit coagulation scheme which requires no iteration (Jacobson, 1994).

2.6 Cloud droplet activation

The number of activating cloud droplets is calculated using parameterization by Abdul-Razzak et al. (1998); Abdul-Razzak and Ghan (2002). Coarse sectional resolution (i.e. wide size bins) can cause problems with cloud activation when each bin covers a fairly wide range of critical supersaturations. One possibility around this problem is to activate only fraction of the particles in the critical size bin. The easiest way to do this is to assume a number concentration profile inside the critical size bin. This can be done in several ways. Korhonen et al. (2005) have presented a method to describe the number concentration using linear dependency between the centers of two adjacent size bins. In this model, the method by Korhonen et al. (2005) was further improved by assuming the number concentration dependence between two bins to be of 2nd

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order polynomial form. Figure 2 illustrates the calculated number of activated cloud droplets (CDNC) for urban conditions using three different approaches to describe the size distribution in a bin. The results are compared with an accurate solution obtained using 300 size bins in size space (thick black curve). The first approach was to assume constant number concentration within one bin (dash-dot curve). Second, the method by Korhonen et al. (2005) was used (dashed curve). Third, a modified method of Korhonen et al. (2005) was used: the number concentration inside the bin was described with a 2nd order polynomial function dependent on the particle concentration in the surrounding size bins.

From Fig. 2 it can be seen that the accuracy in the CDNC is significantly increased when a 2nd order polynomial function is used to describe the number size distribution within a size bin.

2.7 Sulfate production in cloud droplets

Salsa calculates the sulfate production calculating the oxidation of SO_2 by H_2O_2 and O_3 in cloud droplets. The aqueous phase concentration of SO_2 is calculated according to Henry's law, accounting for dissolution effects (see e.g. Seinfeld and Pandis, 1998). The liquid water available for liquid phase chemistry is calculated from the liquid water content dividing the liquid water equally among activating cloud droplets.

2.8 Other processes

The aerosol model SALSA itself calculates only processes described above. Other processes have to be calculated in the host large scale model, which SALSA is coupled with. The treatment of other processes, such as particle emissions, dry and wet deposition, and radiative effects of the aerosols are dependent on the host model.

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3 Model evaluation

3.1 Coagulation and condensation

The performance of the aerosol model was tested by calculating different microphysical processes and comparing the results against previous aerosol models used in large scale models. Coagulation and condensation were tested comparing the model results and a model comparison for air quality models by Zhang et al. (1999). This comparison also included accurate numerical solutions for condensation and coagulation that have been calculated using an explicit microphysical models CONFEMM and COAGUL for condensation and coagulation, respectively.

Size distributions used in the model verification for condensation and coagulation are given in Table 1. The given size distributions represent three different types of conditions, clear, hazy and urban conditions. In the evaluation, particles were assumed to be dry.

Figure 3 illustrates results for a case when only coagulation is taken into account. The dotted red curve represents the initial size distribution. The initial size distribution used here was the urban case given in Table 1. The solid red line is the size distribution after 12 h calculated using COAGUL. The dashed curve is calculated using a modal approach used in Models-3, and the blue solid curve is calculated using SALSA using 15 min time step.

Figure 3 shows that when using the sectional approach, the shape of the accurate size distribution is reproduced clearly better than when using modal approach. The final total number concentration calculated using SALSA is approximately 5.5% smaller than the total number concentration calculated using COAGUL.

Coagulation was also tested running SALSA using different time steps. With time steps less than 1 h, SALSA reproduced the shape of the accurate results and when the time step was less than 100 s, the accuracy was no longer improved by shortening the time step.

Figure 4 shows results for simulations where condensation was the only microphysi-

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cal process taken into account. The dotted red curve represents the initial volume size distribution, and the red solid line the final volume size distribution given by CONDEFF. The figure also shows results for the final size distribution calculated using different aerosol models. Lines with markers represent results from sectional aerosol models, and the dashed green curve shows results from Models-3 using modal approach. The solid blue curve is the final size distribution calculated using SALSA. The Fig. 4 illustrates that SALSA reproduces the shape of the accurate result of CONDEFF. The total volume concentration calculated using SALSA was approximately 1.4% smaller than the total volume concentration calculated using CONDEFF.

3.2 Hygroscopic growth of the particles

The ability of SALSA to describe the effect of composition on the wet aerosol size distribution was tested comparing SALSA against EQUISOLV II (Jacobson, 1999) which is an explicit thermodynamical equilibrium model calculating the thermodynamical equilibrium between gases, liquids, ions and solids. The compositions used in the subranges are shown in Table 2

Figure 5 shows the equilibrium sizes for particles in different size bins as a function of the water saturation ratio S . In the figure, subranges are denoted as 1, 2a, 2b, and 3a together with the index of the size bin beside the axes. The blue lines are the results from SALSA and the red lines are the results from EQUISOLV II.

The largest differences between SALSA and EQUISOLV II appear in the smallest subrange 1, and the smallest bins in subrange 2a at high water saturation ratios. This is probably due to different treatment of organics between the two models. EQUISOLV II also calculates explicitly the dissociation of sulfate into HSO_4^- and SO_4^{2-} . The smallest droplets are more highly concentrated and more sensitive to changes in the Kelvin effect, thus leading to large differences in the equilibrium droplet size.

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3.3 Insoluble vs. soluble size bins

Many combustion sources produce originally rather insoluble soot particles which, after spending some time in the atmosphere, become hygroscopic enough to act as cloud condensation nuclei. (e.g. [Riemer et al., 2004](#); [Zuberi et al., 2005](#); [Moteki et al., 2004](#)).

5 The same aging process concerns originally insoluble dust particles. A large-scale atmospheric model should be able to capture the hygroscopic aging of particles correctly. In practice this means that particles from the insoluble size bins should be transferred to the soluble size bins at the right moment. Too early transfer of insoluble particles would artificially reduce the hygroscopic properties of already soluble particles, whereas too
10 late transfer would cause underestimation of the number concentration of soluble particles prior to the transfer. In both cases, incorrect number of cloud droplets during the cloud formation would be predicted.

Testing how well a model performs in treating the insoluble-to-soluble transition is notoriously difficult. Ideally, such a test should be made in a three-dimensional framework with multiple sources of particles of different solubility and using a model that has
15 a large number of size bins with multiple solubility classes in each size bin. Here we take a simpler approach by testing how sensitive the predicted cloud droplet number concentrations are to the transfer of particles from insoluble to soluble size bins.

To start with, we made a large number of simulations using the detailed cloud model
20 with 300 size bins mentioned in Sect. 2.6. In these simulations, we varied the air updraft velocity (range 0.1–1 m/s), particle number size distribution (marine, rural and urban distributions in Table 1), soluble volume fraction of “insoluble” particles (range 0.0001–0.1 for the volume fraction of sulfate in subrange 2b), and soluble volume fraction of “soluble” particles (volume ratios of 50/50, 75/50 and 100/0 between sulfate and black
25 carbon in subrange 2a). An equal number of insoluble and soluble particles were assumed. Each simulation was made in two different ways: with or without moving particles from the insoluble size bins into the corresponding soluble size bins.

Figure 6 illustrates the relative differences in calculated cloud droplet number con-

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centrations between the cases where all insoluble particles were moved into soluble size bins and cases where they were not. The urban particle number size distribution was chosen because it led to the largest differences. We may see the relative differences increased with increasing soluble fraction of “insoluble” particles but were quite low (<0.01) over the whole variable range considered. This suggests that the cloud droplet nucleating properties of externally-mixed particle populations are not very sensitive to the exact treatment (timing) of the insoluble-to-soluble particle transfer, provided that this transfer is made before “insoluble” particles become very hygroscopic.

Next, we performed the same set of simulations with the difference that the model runs with insoluble-to-soluble transfer and subsequent cloud droplet activation were made using SALSA. The results are shown in Fig. 7. We may see that due to the course resolution (10 size bins) of SALSA compared with the cloud model with 300 size bins, the relative differences in predicted cloud droplet number concentrations are clearly larger than those in Fig. 6. However, the differences can still be considered fairly low considering that this is kind of a “worse case scenario”. Under conditions more typical for the global atmosphere, the number concentration of insoluble particles is expected to much lower than that of soluble particles. We conclude that our method of moving particles from insoluble to soluble size bins can be used with a relatively good confidence.

3.4 All microphysical processes combined

Finally, the model was evaluated simulating a nucleation event typical in a highly polluted area of Po Valley, Italy. The ambient size distribution and the values 280.6 K for temperature and 83% for relative humidity were taken from measurements (Hamed et al., 2007). The gas phase concentration for sulfuric acid was derived from measured OH concentrations and can be seen in Fig. 8a. Since there was no measurement data available for organic gases, the gas phase concentrations were assumed to be equal to those of sulfuric acid. The event was compared against an accurate model simulation using SALSA with 300 size bins in size space. The initial conditions were approximated

using measured conditions at 08:00 am before the observed nucleation event. Initial ambient size distribution was chosen as tri-modal with geometric median diameters of 5 nm, 20 nm, and 100 nm, number concentrations of 4000 cm^{-3} , 8000 cm^{-3} , and 2000 cm^{-3} for each mode, respectively. The geometric standard deviation for each mode was set to be 1.5.

Figure 8b illustrates the contour plot of the particle number concentrations during a simulated nucleation event calculated using 300 size bins. In Fig. 8c, the same event was simulated using the default bin setup of 3 size bins in subrange 1, 4 size bins in subrange 2, and 3 size bins in subrange 3. In Fig. 8d, the total number concentrations for these two simulations are presented.

From Fig. 8 it can be seen that, overall, the default model setup reproduces the number concentrations well compared to the accurate model setup. At the beginning of the simulation, the total number concentrations are underestimated, and also after the actual nucleation burst, the processing of the aerosol size distribution by condensation and coagulation shows surprisingly good agreement when the accurate and coarse simulations are compared.

3.5 Technical details

The model is written in Fortran 90 in modular form to make it easy to include it in existing large-scale models. The selection of number and location of size sections, processes active on different sections as many key parameters are easily changeable from model to another.

Preliminary simulations using SALSA coupled with ECHAM5 general circulation model (Stier et al., 2005) show less than 30% increase in calculation time for one month simulations replacing the modal aerosol model HAM. We can therefore conclude that the model SALSA is computationally efficient enough to be used in large scale circulation models.

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We have developed an aerosol dynamics model package suitable for addition climate modelling. Sectional approach provides a much more flexible way to simulate the aerosol size distribution, as it does not assume any explicit shape of the population.

Our approach, which tries to only simulate the climatologically important aerosol properties without oversimplifying the processes, is in our opinion a good way to prepare a complete view of aerosol dynamic effects to the climate. However, realities of model development inherently forces the developers to make simplifying assumptions or model design choices.

One such issue is the choice of number of sections. We have selected the number of sections and their properties in the default case to best produce in our opinion the most important aerosol process – aerosol-cloud interactions – as well as possible given the calculation resource restrains. A more detailed size distribution handling is of course possible in the model framework, but as the model evaluation in this paper shows, the current model does already very good effort in capturing the processes. An another question altogether is can the number of sections be reduced. The advantages of such approach are clear, as the number of sections is the most critical issue for computational efficiency of the model. Based on the work here, we can predict that at least a radical reduction of number of sections will lead to significant drop in model performance. However, such changes should be thoroughly tested for microphysical accuracy.

The default model configuration takes into account both particle size and composition in account in cloud activation processes. The particle size resolution is increased in the sizes where the cloud activation will most likely to be sensitive to the particle diameter. Additionally, the particle activation is also handled in sub-section parameterization. For particle composition effect, we have taken into account external mixing of soluble and insoluble particles in the most sensitive aerosol fraction. It is an ongoing debate in the cloud activation literature on which of the effects are more important (Dusek et al.,

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2006; McFiggans et al., 2006), but on our opinion relevant microphysical handling of both is necessary in the default model configuration.

Even though the default model takes into account only one kind of organic species, this is not a major limiting factor for the model. In the default configuration, the role of the model organic is just to act as a surrogate for most organic species. This is also obvious on the quite simplistic way of calculating the binary molality of the model organic. Addition of more detailed organic package is beyond the intended scope of this work, but the model framework is well suited for such more detailed studies of organic aerosol effects.

The default model framework does assume that the parent model can provide many necessary boundary conditions and concentrations for SALSA model. The choice of input parameters is selected to best fit as an aerosol model for ECHAM5 climate model (Stier et al., 2005). Especially some aerosol related input functions, such as emission modelling is not included and should be separately investigated for each model to best fit the intended resolution and model configuration.

5 Conclusions

A new sectional aerosol microphysical model designed for large scale modelling purposes has been developed and tested against existing aerosol models.

The aerosol size distribution in the model is divided into three subranges according to dominant processes and chemical components in different sized particles. Only the relevant processes and components are calculated for each subrange. The densest size section spacing is reserved for particles from 51 nm to 730 nm in order to capture the changes in this climatically most important size range.

The model uses the moving-centre scheme by Jacobson (2005) to describe the aerosol size distribution of particle smaller than 730 nm, i.e. below this threshold size average number as well mass concentrations of chemical components in each section are carried as tracers. As particle growth for particles in the coarse mode is very slow,

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particles larger than 730 nm are represented with fixed size sections.

Comparison against explicit solutions of coagulation and condensation equations show that the model captures the changes in the aerosol size distribution well despite its relatively coarse size resolution of 10 sections from 3 nm–10 μm .

5 It is particularly noteworthy that this fairly coarse sectional representation tracks the evolution of the particle size distribution much more accurately than a multimodal representation. Especially, the concentration of accumulation mode particles, the most important size range concerning direct and indirect climate effects of aerosol particles, was reproduced much better with the new model owing largely to the size distribution description in subrange 2. The denser size sectioning in this subrange led to a more accurate description of cloud droplet activation and sulphate production in cloud droplets. Although the currently often used modal descriptions of atmospheric particulate matter are capable of reproducing observed aerosol mass, it is the size together with the composition of particles that determines their radiative and human health effects.

15 *Acknowledgements.* This work has been partly funded by European Commission project EU-CAARI (N₀036833-2(EUCAARI)).

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Table 1. Parameters for the size distributions used in coagulation and condensation tests, taken from Zhang et al. (1999). Subscripts n , a , and c refer to nuclei, accumulation, and coarse modes, respectively.

Parameter	Clear	Hazy	Urban
Mean diameter (μm)			
d_n	0.03	0.044	0.038
d_a	0.2	0.24	0.32
d_c	6.0	6.0	5.7
Standard deviation			
σ_n	1.8	1.2	1.8
σ_a	1.6	1.8	2.16
σ_c	2.2	2.2	2.21
Total volume ($\mu\text{m}^3 \text{cm}^{-3}$)			
V_n	0.03	0.09	0.63
V_a	1.0	5.8	38.4
V_c	5.0	25.9	30.8

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Table 2. Chemical composition in subranges used in equilibrium size calculations.

subrange	$(\text{NH}_4)_2\text{SO}_4$	OC	BC	Sea salt
1	0.2	0.8		
2a	0.6	0.2	0.1	0.1
2b	5×10^{-3}	0.01	0.985	
3a				1

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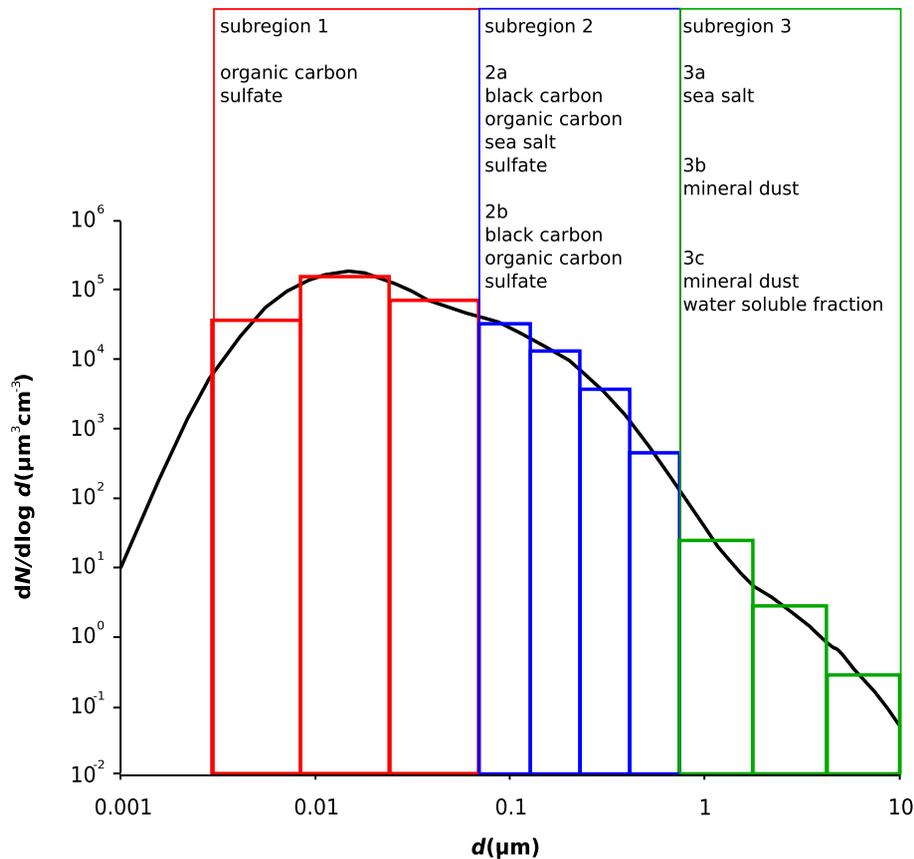


Fig. 1. Schematic of the division of size distribution into subranges and sections in the aerosol model SALSA.

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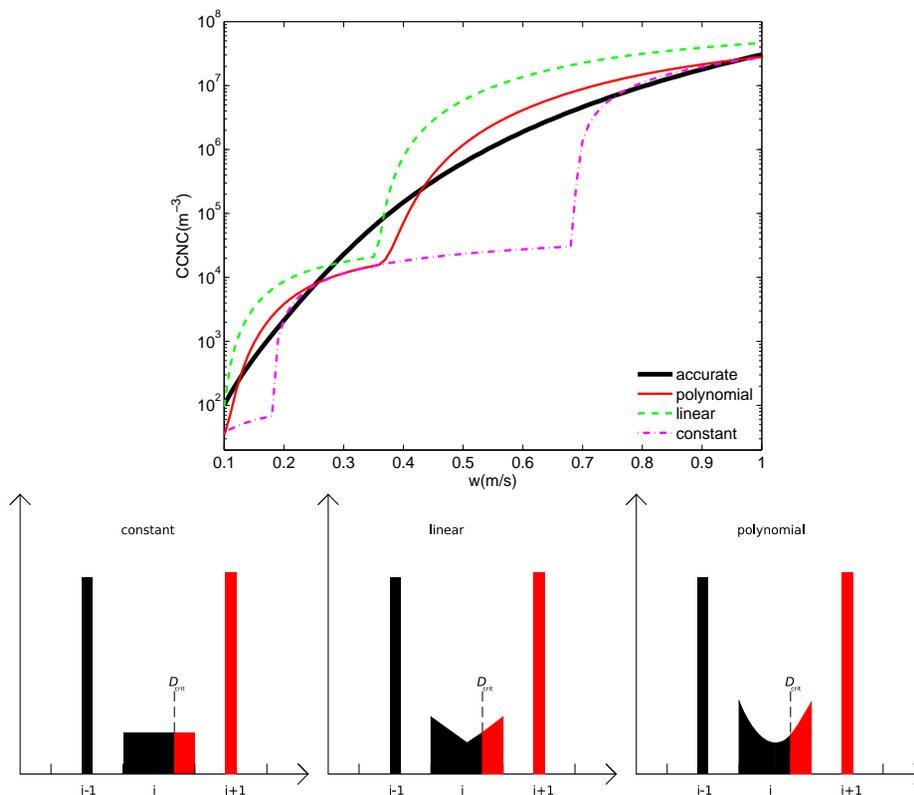


Fig. 2. Cloud droplet number concentration (CDNC) as a function of updraft velocity w for four different approaches. Lower panels illustrate three different approaches for coarse grid setup.

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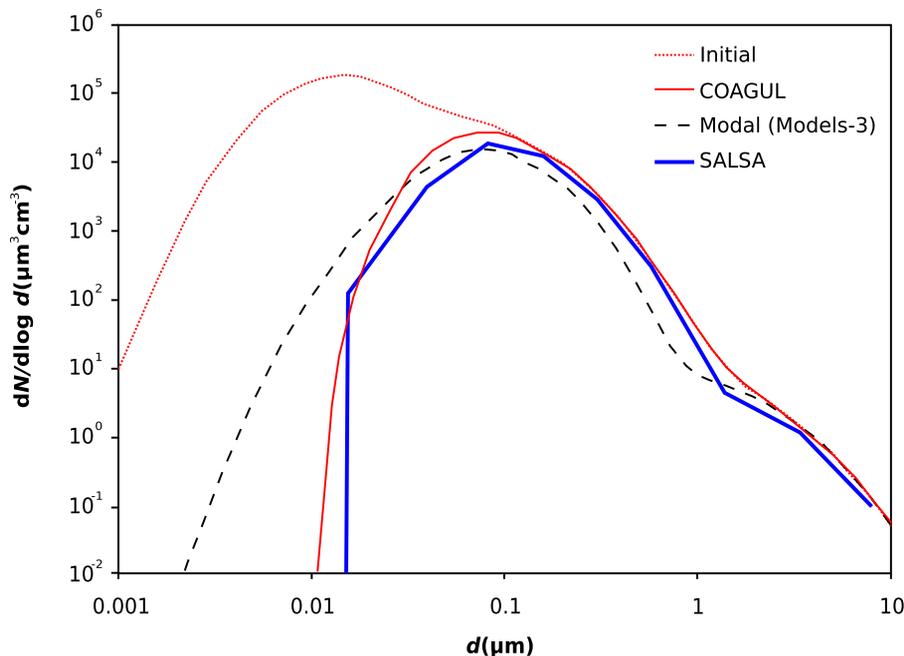


Fig. 3. Initial and final size distributions coagulation simulation. The dotted red curve is the initial size distribution. The red curve is the final size distribution using an explicit coagulation model COAGUL, The black dashed curve represents the final size distribution using a modal aerosol model at the thick blue line is the final size distribution calculated using SALSA. Results for COAGUL and modal aerosol model were adapted from Zhang et al. (1999).

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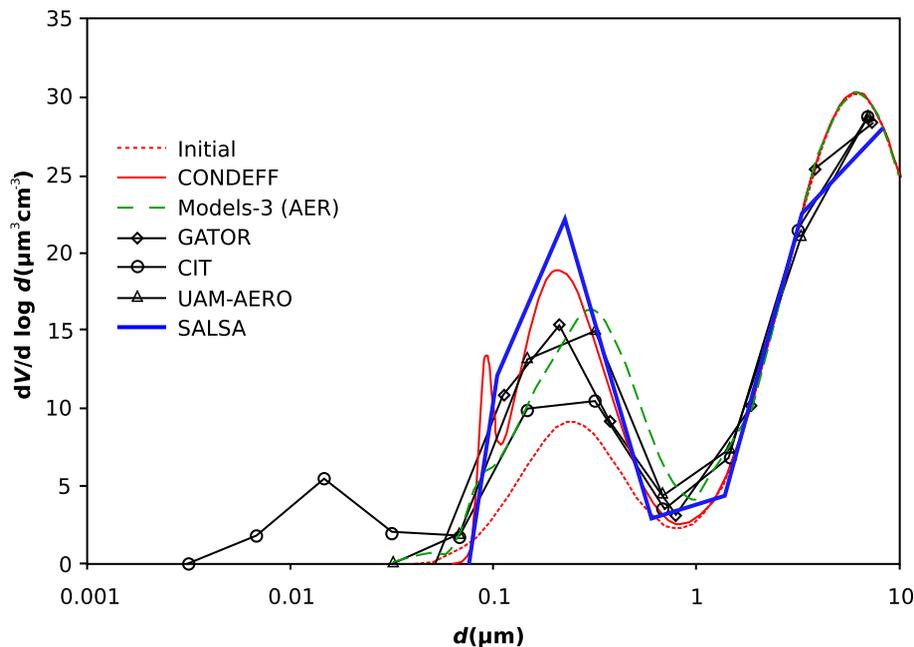


Fig. 4. Initial and final particle number concentration, for 12 h simulations for urban conditions. The dotted red curve is the initial size distribution and the solid red curve is the final size distribution from CONDEFF. The dashed green curve is the final size distribution calculated using a modal model, the black curves accompanied with markers are the final size distribution calculated using different sectional aerosol models, and the thick blue curve is the final size distribution calculated using SALSA. Results for models in comparison were adapted from Zhang et al. (1999).

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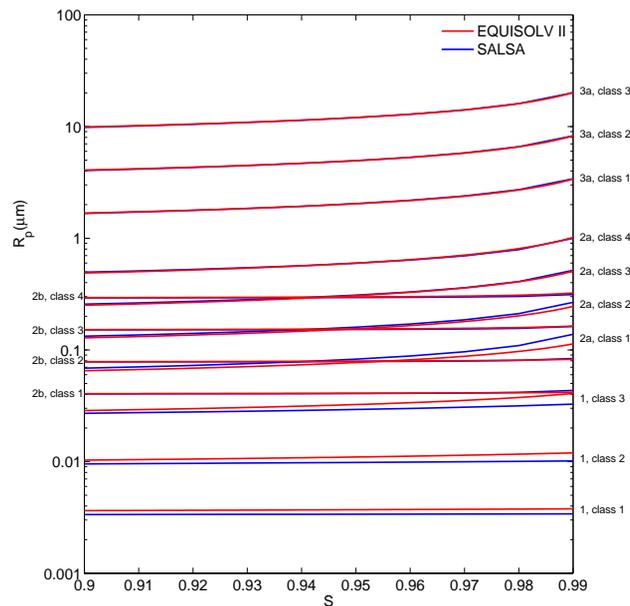


Fig. 5. Wet diameter of different size bins in subranges 1, 2a, and 3a as a function of water saturation ratio calculated using EQUISOLV II (red curves) and SALSA (blue curves).

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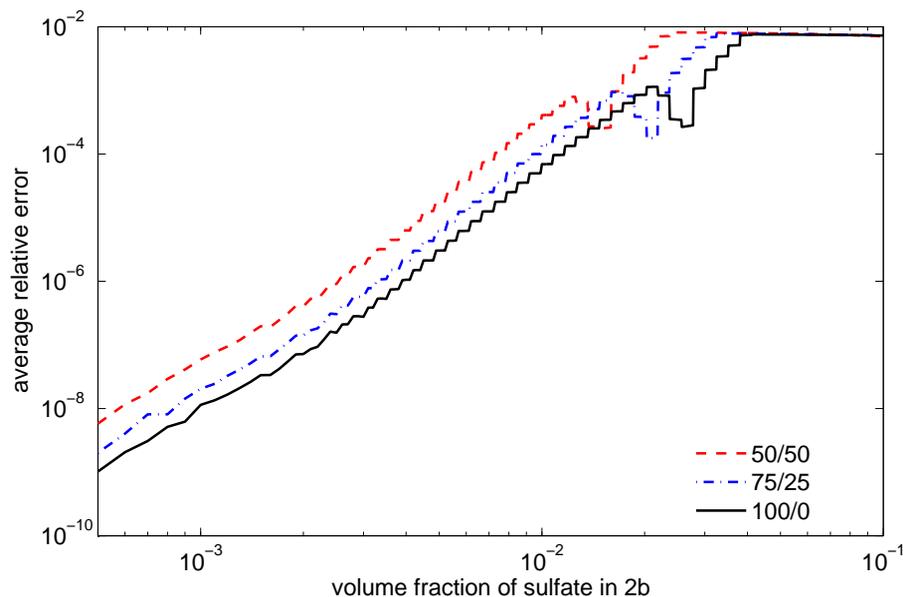


Fig. 6. Average relative difference in the number activating cloud droplets when particles are moved from insoluble size bins to soluble size bins compared to a case where particles remained in their original size bins. Relative error is given as a function of volume fraction of sulfate for different volume ratios of sulfate and black carbon in subrange 2a. Calculations were done using 300 size bins in size space.

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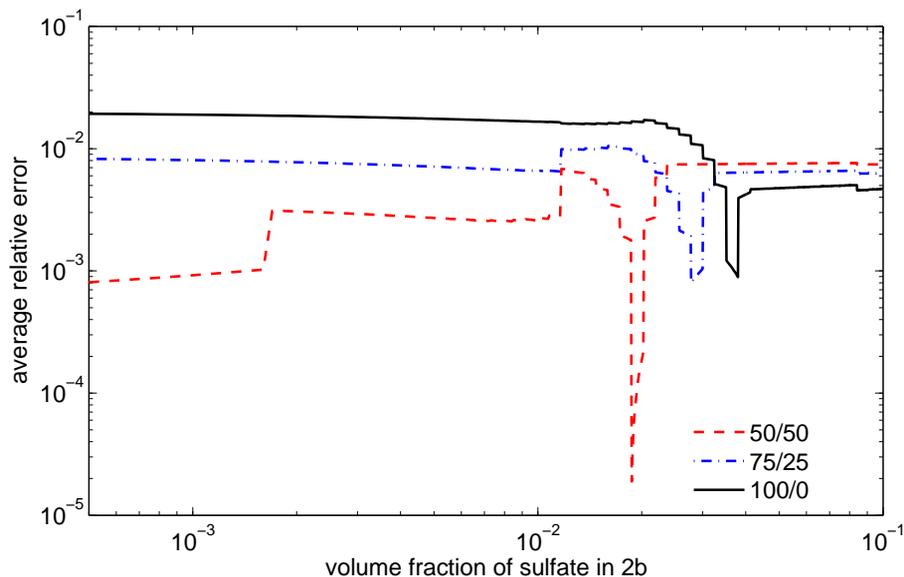


Fig. 7. Average relative error in the number activating cloud droplets when particles are moved from insoluble size bins to soluble size bins using 10 size bins in size space, compared to a case calculated using model setup of 300 size bins in size space where particles remained in their original size bins. Relative error is given as a function of vertical velocity for different volume ratios of sulfate and black carbon.

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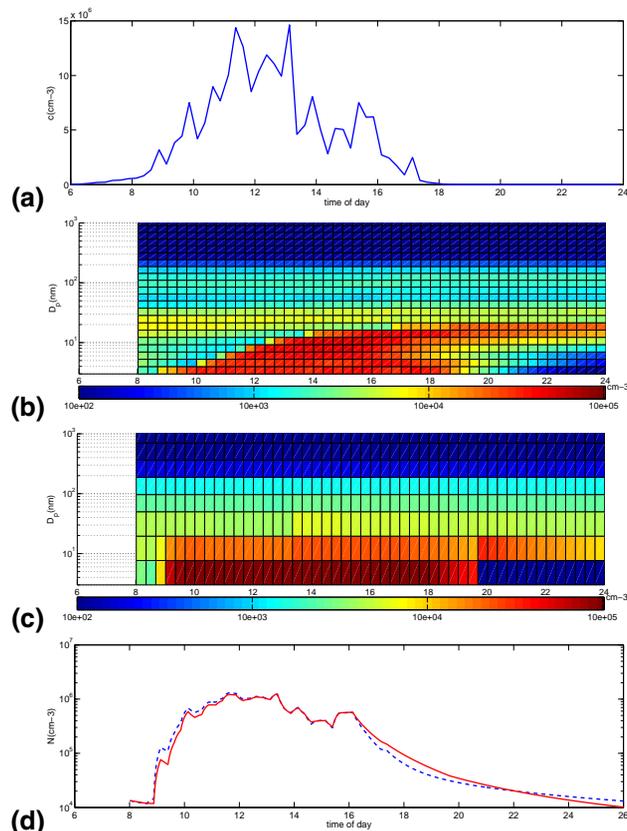


Fig. 8. (a) Gas phase concentration of sulfuric acid and condensable organic vapor as a function of time. (b) Number concentrations of particles as a function of time and diameter calculated using 300 size bins in size space. (c) Number concentrations of particles as a function of time and diameter calculated using 3, 4, and 3 size bins in subranges 1, 2, and 3, respectively. (d) Total number concentrations for accurate (blue dashed line) and coarse model setup (red solid line).

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