Response to reviewer #1

ACP-2014-201: "Development of an aerosol microphysical module: Aerosol Two-dimensional bin module for foRmation and Aging Simulation (ATRAS)" by H. Matsui et al.

We thank the reviewer very much for reading our paper carefully and giving us valuable comments. We have revised our paper by taking into account the reviewer's suggestions. Detailed responses to individual comments are given below.

1) Needed accuracy of aerosol microphysical simulations

Reviewer's comment:

The statement given in lines 16-18 on page 10661 is very strong. While a more detailed description of aerosol microphysics is likely to increase the accuracy of a simulation, it is still unclear how detailed description is necessary in various atmospheric applications. As a compromise between bulk approaches and very detailed approaches, such as the model framework introduced here, many large-scale models use currently a modal approach to represent the aerosol size distribution and allow a simplified treatment of aerosol mixing state.

Response:

We slightly toned down the description because it depends on cases whether a detailed aerosol model is necessary or not. When we need detailed information of size distribution and mixing state (e.g., for comparisons with measurements), simulations with a detailed scheme (such as ATRAS) is necessary. On the other hand, for global and/or long-term simulations, more computationally efficient (modal or bulk) scheme may be reasonable for the balance between accuracy and computational cost.

2) Model evaluation

Reviewer's comment:

Model evaluation presented in this paper was very crude, which is understandable. However, I would like the authors to provide some recommendations on how to evaluate detailed aerosol microphysical models in large-scale applications. What quantities should be looked at? How can one demonstrate, by comparing with measurements, that this kind of a very detailed model is essentially better than e.g. a modal model?

Response:

For comparisons of the performance between a detailed model and a simple model, CCN (at several constant supersaturations) and cloud droplet number concentrations may be good parameters to evaluate the number concentration and size distribution of aerosols, though these parameters are influenced by the treatment of emissions, transformation, and deposition processes. Mass absorption cross section is probably a good parameter to evaluate BC mixing state (lens effect) in large-scale applications.

We can show the advantage of a detailed model especially when detailed aerosol parameters are compared between measurements and model simulations. For example, number median diameter is a good parameter to evaluate new particle formation events (Matsui et al., 2011). Shell-to-core diameter ratio is also a useful parameter to validate BC mixing state (Matsui et al., 2013a). Our previous studies clearly showed the improvement of model performance for these parameters by using a detailed aerosol model. With the development of more advanced observational techniques, we need more detailed and sophisticated aerosol representation in models for comparisons.

In many cases, it is not easy to show that a detailed bin aerosol model has better agreement with measurements than a simple modal or bulk model because of the uncertainties in emissions and processes. But a detailed bin model is conceptually better (e.g., Korhola et al., GMD, 2014) and uses less assumption than a simple modal model.

3) Areas requiring further development in large-scale modeling

Reviewer's comment:

Based on our current scientific knowledge, how should one develop aerosol microphysics models further? Do this and other existing large-scale models miss some major processes altogether, or should some processes treated in more detail and how? As one example, do the recent findings about extremely low-volatile organic vapours have any implication on how atmospheric new-particle formation and SOA formation should be treated in large scale models. There are probably many other examples like this.

Response:

As the reviewer suggested, low-volatile organic vapors will be important in our future study (and in many other aerosol models). They may promote the occurrence of new particle formation and enhance SOA concentrations. A recent study (Metzger et al., PNAS, 2010) developed and used a nucleation parameterization considering the contribution of organic vapors to nucleation. The parameterization may be useful for further improvement of our model. Few 3-D models consider brown carbon (Feng et al., ACP, 2013) and OA formation in aerosol phase (e.g., Liu et al., JGR, 2012). They are also key processes for the improvement of OA formation and its radiative effect.

Including IN formation from aerosols is another important step for studies on aerosol-cloud interactions. To extend BC mixing state treatment to dust particles for resolving pure dust, mixed dust, and dust-free particles may be a key issue for more realistic simulations of IN concentrations and their formation pathways.

4) Minor/technical issues:

Reviewer's comment:

The present tense in section 4 sound a bit strange. Perhaps past tense would be more appropriate.

Response:

We have revised section 4 as the reviewer suggested.

Response to reviewer #2

ACP-2014-201: "Development of an aerosol microphysical module: Aerosol Two-dimensional bin module for foRmation and Aging Simulation (ATRAS)" by H. Matsui et al.

We thank the reviewer very much for reading our paper carefully and giving us valuable comments. We have revised our paper by taking into account the reviewer's suggestions. Detailed responses to individual comments are given below.

1)

Reviewer's comment:

Even though the individual model components are described in previous publication are short description of the NPF (e.g. whether The H2SO4-Water system or a more complex scheme (neutral and ion induced nucleation, amines, etc.) is used) and the SOA scheme (number of volatility classes, hygroscopicity, ageing included or not) should be included in the manuscript.

Response:

We have added several sentences to the manuscript to describe the NPF and SOA schemes used in this study.

The revised description of NPF scheme is as follows. "The particle formation (nucleation) rate at 1 nm is estimated by activation-type (e.g., Kulmala et al., 2006) or kinetic nucleation (e.g., Kuang et al., 2008) in the boundary layer and by a binary (H_2SO_4-water) homogeneous nucleation (Wexler et al., 1994) in the free troposphere, as described by Matsui et al. (2011). In this study, we use the activation-type nucleation (nucleation rate at 1 nm is proportional to H_2SO_4 concentrations) with a constant rate coefficient of 2×10^{-7} s^{-1}, which value was adopted in our previous studies (Matsui et al., 2011, 2013c)."

The revised description of SOA scheme is as follows. "SOA processes are calculated by the volatility basis-set scheme with photochemical multigenerational oxidation of organic vapors by OH radicals (Matsui et al., 2014), which is similar in many respects to Shrivastava et al. (2011). This scheme uses 9 volatility classes to represent semi-volatile and intermediate volatility organic compounds (S/IVOCs). We consider the formation of first-generation oxidized VOCs (OVOCs) from 9 lumped VOCs; alkanes (ALK4 and ALK5), olefins (OLE1 and OLE2), aromatics (ARO1 and ARO2), isoprene (ISOP), monoterpene (TERP), and sesquiterpene (SESQ). The mass yield of OVOCs from each lumped VOC is calculated with a NO_x-dependent 4-product basis fit (Tsimpidi et al., 2010). S/IVOCs and OVOCs are oxidized to the volatility class with an order of magnitude lower effective saturation concentrations by OH radical with a rate constant of 1×10^{-11} cm^3 $molecule^{-1}$ s^{-1}. Size-resolved OA condensation and evaporation are calculated using the method of Koo et al. (2003) by assuming gas-particle equilibrium partitioning (Schell et al., 2001). A hygroscopicity value (κ) of 0.14 is assumed for all the OA species used in the volatility basis-set scheme (Matsui et al., 2014)."

2)

Reviewer's comment:

The authors mention an increase in computational cost; however, it would be useful for readers to get a reference number (for e.g. the M10_SN simulation) of the CPU hours per simulation day required including information on the processor machine type. This would provide information on the real computational cost compared to other "cheaper" aerosol modules.

Response:

The CPU time required for the M10_SN simulation is about 36 hours per a simulation day in our application (SGI ICE X (Intel Xeon E5-2670 2.6GHz, SUSE Linux Enterprise Server 11SP1, Intel Composer XE 12)). Compared with the original 8-bin MOSAIC simulation, the computational costs are a factor of 14, 3.5, and 1.7 greater in the M10_SN, M01_SN, and M01 simulations, respectively. We have added these description to the manuscript.

3)

Reviewer's comment:

The evaluation is relatively simple; a more detailed evaluation of the full composite of the model components as given by ATRAS could be provided in an electronic supplement.

Response:

The ATRAS simulations were compared with surface and aircraft measurements with more detailed information (e.g., temporal and vertical variability). The results were added to the manuscript (supplement). The following parameters were compared between measurements and model simulations: the temporal variations of BC, sulfate, and OA mass concentrations at Fukue and Hedo (Fig. S1), the vertical profiles of BC mass and LSP (light scattering particles) volume concentrations during A-FORCE (Fig. S2), the temporal variation of mean shell-to-core diameter ratio at a BC core diameter of 200 nm along the flight tracks during A-FORCE (Fig. S2), and the temporal variation of aerosol size distribution at Anmyeon, Korea (Fig. S3, for NPF evaluation). Details of these measurements and the methods for comparisons are described in our previous studies (Matsui et al., 2011, 2013a, 2013c, 2014). Compared with our previous studies, similar or better model performance was obtained for individual parameters by using ATRAS.

4)

Reviewer's comment:

The authors state, that there is no consideration of coarse mode emissions. Why is PM2.5 and not PM1 used for the analysis, as this should be more representative for the simulated fine mode aerosol. Is there a significant amount of larger particles in the simulations and where do they come from? They are not visible in Fig. 6d.

Response:

We use $PM_{2.5}$ because it is calculated and outputted in WRF-chem as a default parameter. Since we do not consider dust and sea salt emissions, there are no significant sources of larger particles in our simulation. $PM_{2.5}$ and PM_1 concentrations are 15.1 and

14.7 µg m^{-3} (period and domain average at 1 km), showing that most of $PM_{2.5}$ is PM_1 in our simulation.

5)
Reviewer's comment:

Does cloud processing and release from evaporated hydrometeors contribute to NPF? If so, how important is this process?

Response:

We made three sensitivity simulations, M04_S, M04_SN_aqoff, and M04_S_aqoff. M04_S is the simulation with SOA processes (without NPF processes) with 4 mixing state bins. M04_SN_aqoff and M04_S_aqoff are similar to M04_SN and M04_S, respectively, but these simulations were conducted without aqueous-phase aerosol formation processes (AQCHEM). The statistics of these simulations are summarized below (period and domain average at 1 km). We can estimate the importance of NPF from the difference between M04_SN and M04_S for both the simulations with and without AQCHEM. These results are also shown below.

Simulations	$PM_{2.5}$ µg m^{-3}	$CCN_{1.0}$ cm^{-3}	N (bin1) cm^{-3}	N (bin 5) cm^{-3}	N (bin 8) cm^{-3}
M04_SN	15.1	2443	12580	834	162
M04_S	15.0	2035	0	0	6.9
M04_SN_aqoff	14.5	2642	15510	1040	212
M04_S_aqoff	14.4	2059	0	0	3.1
M04_SN – M04_S	0.04	408	12580	834	155
M04_SN_aqoff – M04_S_aqoff	0.09	584	15510	1040	209

These results show that AQCHEM reduces the contribution of NPF to $CCN_{1.0}$ concentrations by 30% and to number concentrations from 1 to 40 nm (bin 1 - 8) by 20-25%. AQCHEM suppresses NPF likely because it reduces SO_2 and H_2SO_4 concentrations and increases the concentrations of larger preexisting particles.

6)
Reviewer's comment:

Using the M08_SN, M06_SN and M04_SN simulations the authors state that they can explain a certain percentage of the total effect. How is this determined? Is this simply the difference between the simulations compared to the total spread given by M10_SN compared to M01_SN or are more sophisticated statistical methods, e.g. EOF analysis or other forms of data compression used to explain the variability.

Response:

The percentages are calculated from the difference between the simulations, not more sophisticated methods.

7)
Reviewer's comment:

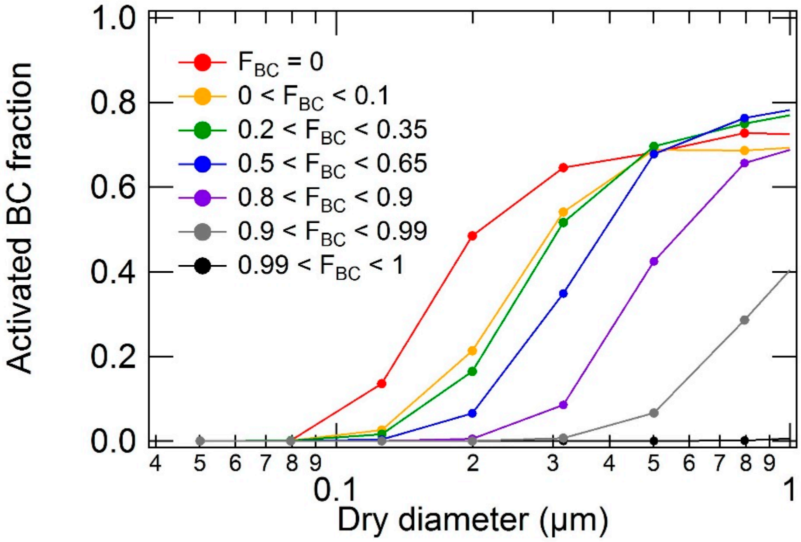
Can coated BC particles contribute to CCN? If so, how does the coating change the activation? I would guess that a coating by inorganics should allow the BC to act as CCN. Do you consider aerosol activation of BC coated particles as internally mixed particles with a total hygroscopicity based on the coating and the BC or as hydrophilic particles with a solid core? The latter one, would likely result in more CCN as the hygroscopicity of those particles is larger. Could you analyse from your data which material is in the activated particles and compare the chemical composition of activated particles with those of the original aerosol distribution, i.e. the fraction of activation for each of the bins?

Response:

Coated BC particles can contribute to CCN in our model. Volume-averaged hygroscopicity is calculated using all chemical species including BC.

We calculated the fraction of activated BC mass to total (both activated and unactivated) BC mass for each size and mixing state bin in the M10_SN simulation. We selected grid points having grid-resolved cloud at 1 km (defined as cloud droplet number concentrations greater than 1 cm^{-3} in this analysis), and the period- and domain-averaged values were calculated. The results are shown by the figure below. This figure shows that the fraction of activated BC largely depends on BC mixing state: the activated fraction decreases with increasing BC mass fraction in a particle at the same diameter bin.

For the grid points selected above, we calculated mean mass concentrations of each chemical species for both activated (cloud-phase) and unactivated (aerosol-phase) particles. Inorganic species (sulfate, ammonium, and nitrate) had higher mass fraction in cloud-phase than carbonaceous species (OA and BC), as shown below.



	Aerosol-phase µg m^{-3}	Cloud-phase µg m^{-3}	Fraction in cloud %
Sulfate	1.25	1.90	60.4
Ammonium	0.54	0.92	63.1
Nitrate	0.37	0.85	69.9
Organic aerosol	1.43	0.76	34.6
Black carbon	0.31	0.083	21.1

8)

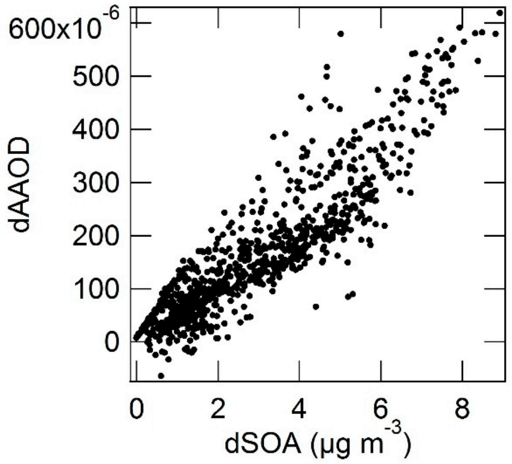
Reviewer’s comment:

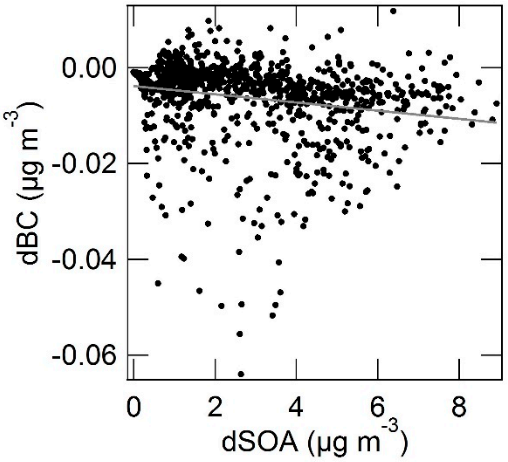
When comparing the compensating effects of absorption and budget changes of BC, and the effects of SOA can you state whether these effects are linear? Is SOA only scattering or do you consider some "brown" carbon absorbing as well?

Response:

SOA is treated as scattering only in this study. To consider brown carbon is a future study. We added this information to the revised manuscript.

We examined the relationship between absorption and budget changes of BC and SOA. The difference between the M10_SN and M10_N simulations was calculated for BC and OA mass concentrations and AAOD. These differences are due to the effect of SOA processes and shown as dBC, dSOA, and dAAOD. The figures below show the correlation between these parameters. Each data point shows period-averaged value at each grid cell at 1 km. As shown by these figures, the relationship between dAAOD (absorption enhancement by SOA) and dSOA is generally linear. The linearity between dBC and dSOA is not so clear, but they have weak negative correlation (more efficient BC removal by SOA).





9)

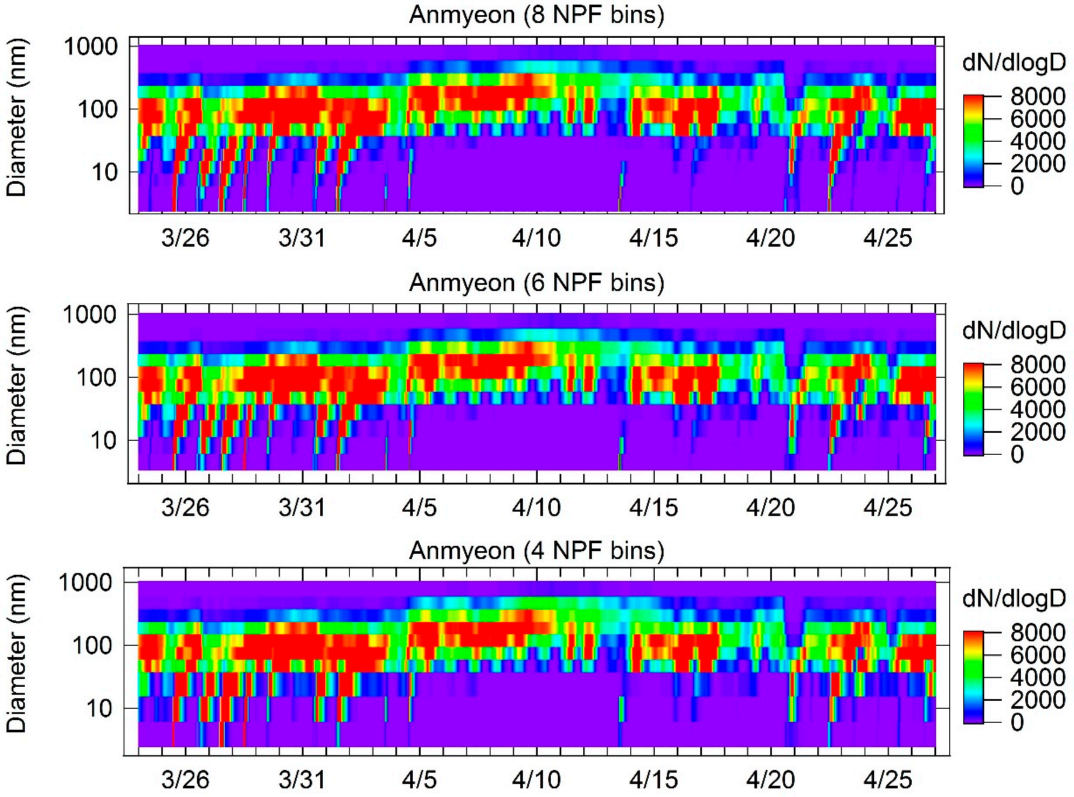
Reviewer’s comment:

To which degree is the high number of bins necessary, especially for the NPF? Of course, coagulation and further condensation of hydrophilic material and SOA are important for the particle size distribution, but as the model operates on a relatively coarse grid, also the time evolution of the aerosol size distribution from a NPF event to an aged aerosol population is unlikely to be resolved.

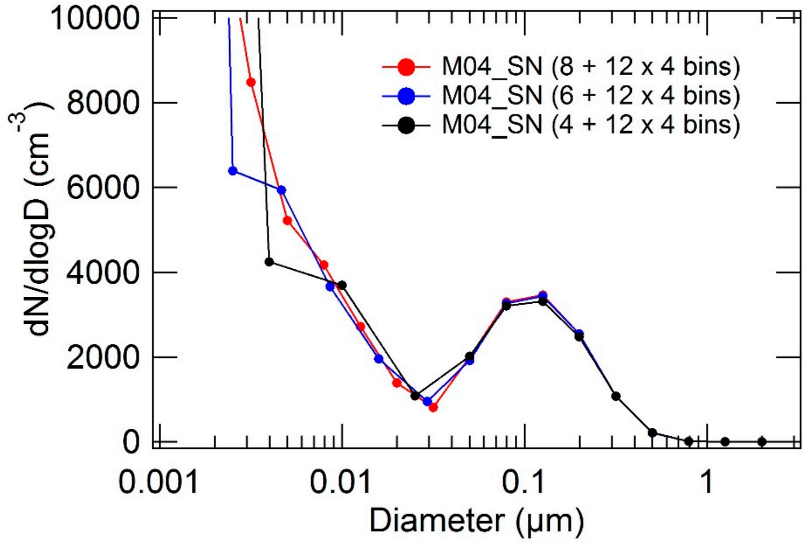
Response:

We made two sensitivity simulations to understand the impact of the number of bins to NPF: one is the simulation with 6 bins between 1 and 40 nm, and the other is the simulation with 4 bins between 1 and 40 nm (12 × 4 bins between 40 nm and 10 µm). These results were compared with the simulation with 8 bins between 1 and 40 nm

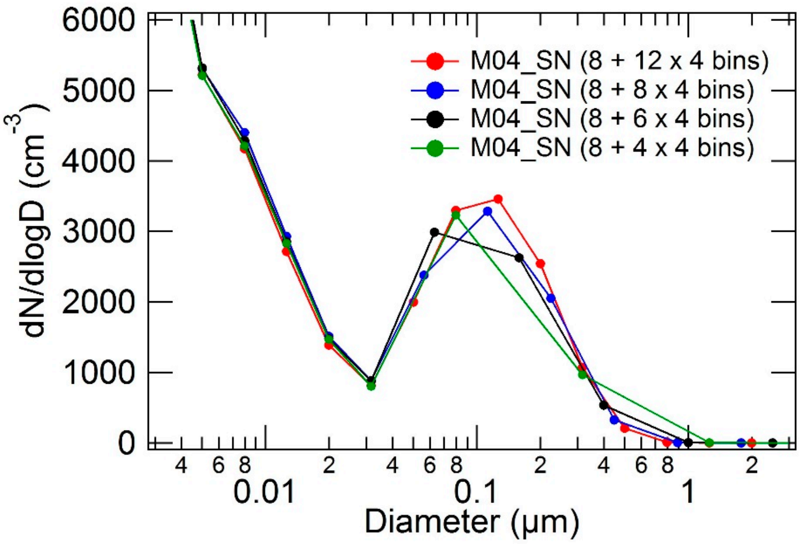
(M04_SN). The results at the Anmyeon site are shown below. These figures show that the simulation with 4 NPF bins can capture important features seen in the simulation with 8 NPF bins in terms of the growth of nucleated particles.



The period- and domain-averaged size distribution at 1 km also shows that the simulations with 4 and 6 NPF bins can capture the absolute number concentrations and their size distribution less than 40 nm in diameter reasonably well. These results suggest that 4 NPF bins may be sufficient for future applications. We have added these description to the text.



We made three additional sensitivity simulations to understand the sensitivity of the number of bins between 40 nm and 10 µm: the simulations with 8, 6, and 4 bins between 40 nm and 10 µm. These results were compared with the simulation with 12 bins between 40 nm and 10 µm (M04_SN). The period- and domain-averaged size distribution at 1 km shows that the simulation with 8 size bins can simulate reasonable number size distributions to some extent, but the simulations with 6 and 4 size bins cannot. Therefore, resolving the size range in Aitken and accumulation modes is important to simulate realistic number size distributions.



10)

Reviewer’s comment:

Comparing the results from MADE-IN (Aquila et al., GMD, 2011) with the findings from ATRAS it seems that the consideration of a mixed BC mode on its own, is already a reasonable improvement for coarse grid models. This manuscript is missing in the list of references.

Response:

We have added the reference to section 1 (Introduction) in the revised manuscript.

11)

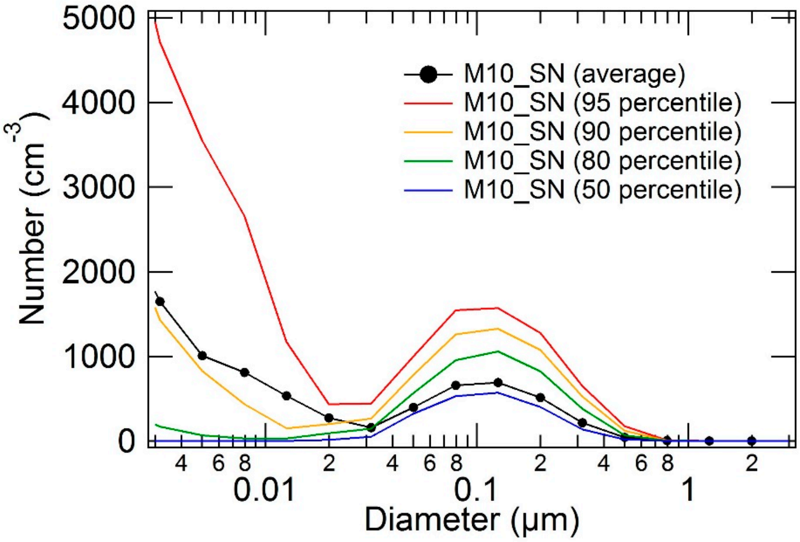
Reviewer’s comment:

Fig. 6d shows a very strong nucleation mode for the period and domain average. However, there are an substantial amount of aitken and some accumulation mode particles available as well. From a typical thermodynamic point of view condensation should be preferred to nucleation which usually takes place in individual events. Is condensation considered as a secondary process in your model, overemphasising NPF? Are the individual nucleation events so strong that they prevail in the spatial and temporal average? How is the variability (spatial and temporal) of this distribution? Could you check how much inorganic material is in the aerosol phase from NPF compared to condensation?

Response:

In the calculation of the nucleation rate in our scheme, we use average H_2SO_4 concentrations before and after the calculation of condensation processes at each time step. So, we do not think nucleation rate is overestimated due to the order of condensation calculation. Condensation is the dominant process in terms of mass transfer from gas to aerosol phase. The mass fraction of sulfate between 1 and 40 nm is only 0.07% of total sulfate mass on period and domain average at 1 km.

The figure below shows 95th, 90th, 80th, and 50th percentiles of number concentrations at each size bin for all spatial and temporal data at 1 km. This figure shows that individual NPF events are strong and that about 10% of all data have large contribution to period- and domain-averaged concentrations in nucleation mode.



12)

Reviewer's comment:

Fig. 8a is in the text referenced as column AAOD, but in the figure caption at a specific altitude. What is correct? The values appear to be relatively large for a single altitude. If the latter is the case what is the thickness of the layer? 8b and 8c are likely given at the specific altitude.

Response:

Column AAOD is used in this study. We revised the caption of Figure 8 to clarify this point.

13)

Reviewer's comment:

Fig. 10 is interesting, but only very shortly described in the manuscript. In my opinion this figure summarises your findings well and its discussion should be extended

- potentially also as part of the conclusions if accompanied with some quantitative numbers.

Response:

We thank the reviewer's positive comment. We think what we want to discuss using Figure 10 is summarized in section 4 (Summary and Conclusions). So, we clarified the findings related to Figure 10 in section 4 (by adding "Fig. 10" to some sentences), rather than adding similar discussions to the end of section 3.4.

Response to reviewer #3

ACP-2014-201: “Development of an aerosol microphysical module: Aerosol Two-dimensional bin module for foRmation and Aging Simulation (ATRAS)” by H. Matsui et al.

We thank the reviewer very much for reading our paper carefully and giving us valuable comments. We have revised our paper by taking into account the reviewer’s suggestions. Detailed responses to individual comments are given below.

1)

Reviewer’s comment:

The discussion of the results is very interesting, but I feel that it lacks some clear and definite conclusion. In particular, after comparing all these simulations with different treatment of BC mixing state, which configuration would the authors suggest? Are 10 bins necessary? I assume that this would be a major limitation for applications to global models.

Response:

We think the number of mixing state bins should be chosen case by case. For example, when we focus on the detailed information of mixing state such as shell-to-core diameter ratios and their frequency distribution (e.g., Matsui et al., 2013), simulations with 10 mixing state bins may be necessary. On the other hand, for global applications focused on total BC mass concentrations and their optical and radiative parameters, simulations with less number of mixing state bins may be reasonable.

This study shows that 4 mixing state bins can simulate BC mixing state effects reasonably well, but since the results are improved (approaching to M10_SN) with increasing the number of mixing state bins (6 or 8 mixing state bins), we cannot conclude that 4 mixing state bins are sufficient. The balance between accuracy and computational cost is important, and it should be decided by each researcher.

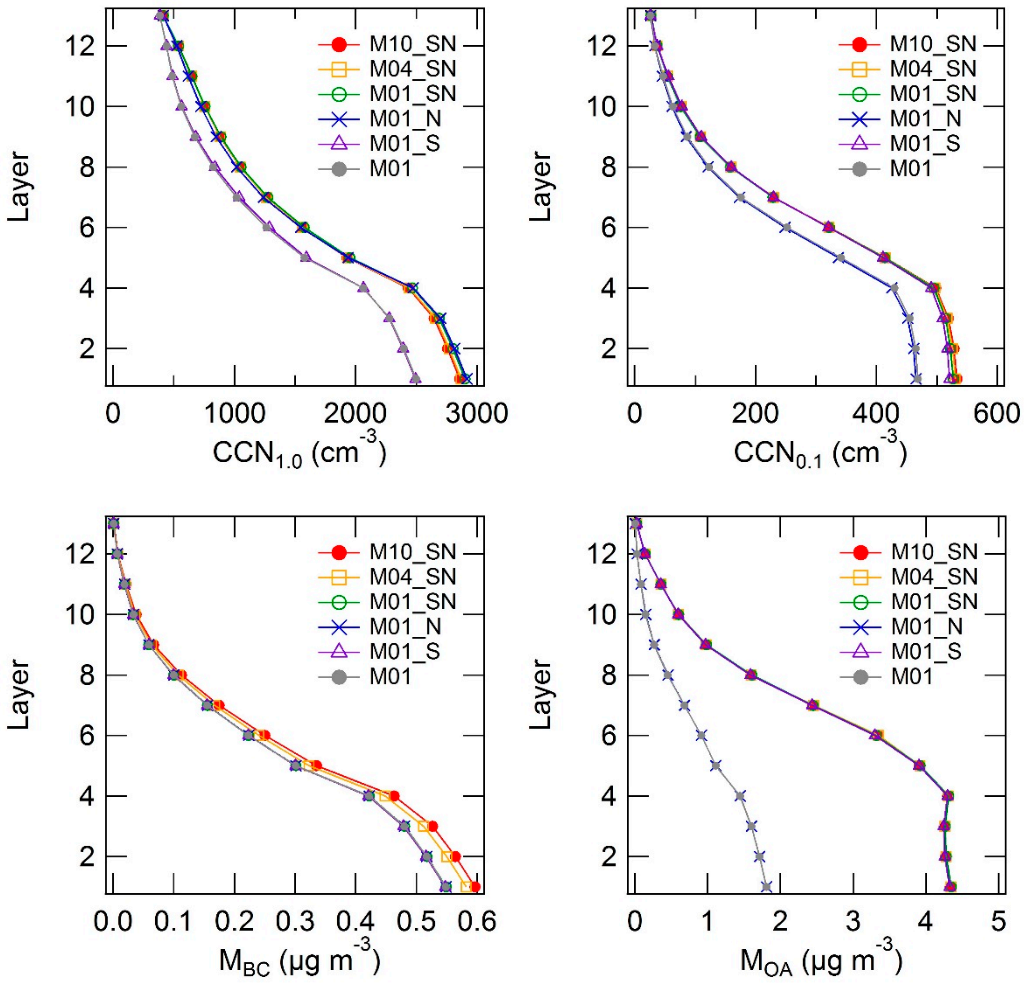
2)

Reviewer’s comment:

All results of 3D variables are shown at 1 km altitude. How do vertical profiles look like?

Response:

The figures below show the vertical profiles of period- and domain-averaged mass and number concentrations in the M10_SN, M04_SN, M01_SN, M01_N, M01_S, and M01 simulations. We have added them to the revised manuscript (Fig. 7). We have also added a paragraph to the end of section 3.2: “The vertical profiles of CCN and mass concentrations show that the features obtained at an altitude of about 1 km (layer 4) are seen at all levels (Fig. 7): OA and $CCN_{0.1}$ concentrations are higher in the simulations with OA formation scheme, BC mass concentrations are higher in the simulations resolving mixing state, and $CCN_{1.0}$ concentrations are higher in the simulations with NPF.”



3)

Reviewer's comment:

I suggest including more description of the SOA processes included, instead of referring only to previous studies. How is SOA transformed into OA? And is SOA a primary emission, or is it calculated from terpene emission? Is organic chemistry included?

Response:

SOA is formed from the oxidation of 9 lumped VOCs (alkanes, olefins, aromatics, isoprene, monoterpene, and sesquiterpene) and the calculation of gas-particle partitioning assuming equilibrium. The oxidation of semi-volatile and intermediate volatility organic compounds by OH radical is considered. Organic chemistry in aerosol phase is not considered in our model.

We have added the description of our OA formation scheme to the manuscript: "This scheme uses 9 volatility classes to represent semi-volatile and intermediate volatility organic compounds (S/IVOCs). We consider the formation of first-generation oxidized VOCs (OVOCs) from 9 lumped VOCs; alkanes (ALK4 and ALK5), olefins (OLE1 and OLE2), aromatics (ARO1 and ARO2), isoprene (ISOP), monoterpene (TERP), and sesquiterpene (SESQ). The mass yield of OVOCs from each lumped VOC is calculated with a NO_x-dependent 4-product basis fit (Tsimpidi et al., 2010). S/IVOCs and OVOCs are oxidized to the volatility class with an order of magnitude lower effective saturation concentrations by OH radical with a rate constant of 1×10^{-11} cm^3 $molecule^{-1}$ s^{-1}."

4)

Reviewer's comment:

Is organic material from SOA considered hydrophilic?

Response:

We assume a hygroscopicity value (κ) of 0.14 for all the OA species used in the volatility basis-set scheme. The value is used for OA in our previous studies (Matsui et al., 2011, 2014). We have added this description to the manuscript.

Specific comments:

5)

Reviewer's comment:

P10664 L2-4: Does this mean that this configuration uses in total 256 bins?

Response:

Yes, in total 256 bins are used to represent aerosol (in both aerosol-phase and cloud-phase) in our model. We clarified this point in the revised manuscript.

6)

Reviewer's comment:

P10664 L5: No BC is emitted as internally mixed. Does this make physical sense? Some aging might be faster than the model time-step (how long is the model timestep?). Would a different choice for emissions make a large difference?

Response:

This assumption (all BC is emitted as pure BC) was adopted to be consistent with the treatment in Matsui et al. (2013a). The uncertainty of this mixing state treatment is described by Matsui et al. (2013a) (section 4.3.2 and Figure 9 of Matsui et al. (2013a)). A sensitivity simulation considering internally-mixed BC emissions showed that BC mixing state in the atmosphere was moderately sensitive to the treatment (the mean SC ratio along flight tracks during A-FORCE increased from 1.26 to 1.35 at a BC core diameter of 200 nm).

The model time step is 6.66 min for inner domain. This time step is probably acceptable to resolve BC aging processes.

7)

Reviewer's comment:

P10666 L26: Does the choice of binning make any difference in the results? Why are the intervals 0.2-0.5 and 0.5-0.8 not as well resolved as the others?

Response:

We used fine resolution between 0.8 and 1.0 to resolve the transition of BC from hydrophobic to hydrophilic. There is no special reason for the choice of partition between 0 and 0.8.

We made a sensitivity simulation (M08_SN_sens) with 8 mixing state bins. In this sensitivity simulation, BC mixing state bins were divided into BC mass fractions of 0, 0-0.2, 0.2-0.4, 0.4-0.6, 0.6-0.8, 0.8-0.9, 0.9-0.99, and 0.99-1. As shown below, the results of mass and number concentrations and optical and radiative parameters were almost identical between the two simulations (period- and domain-averaged values at 1 km).

Parameters	Unit	M08_SN	M08_SN_sens
$PM_{2.5}$	$\mu g\ m^{-3}$	15.1	15.1
BC	$\mu g\ m^{-3}$	0.463	0.464
OA	$\mu g\ m^{-3}$	4.31	4.32
$CCN_{1.0}$	cm^{-3}	2438	2438
$CCN_{0.2}$	cm^{-3}	1081	1083
$CCN_{0.1}$	cm^{-3}	498	500
AOD	---	0.310	0.311
AAOD	---	0.0152	0.0152
SSA	---	0.930	0.931
Heating rate	$K\ day^{-1}$	0.425	0.423
Downward flux	$W\ m^{-2}$	-35.0	-35.0

8)

Reviewer's comment:

P10669 L5: In the simulations without SOA processes, is OA from primary emissions?

Response:

Yes, OA is from primary emissions only in the simulations without SOA processes. We clarified this point in section 3.1 in the revised manuscript.

9)

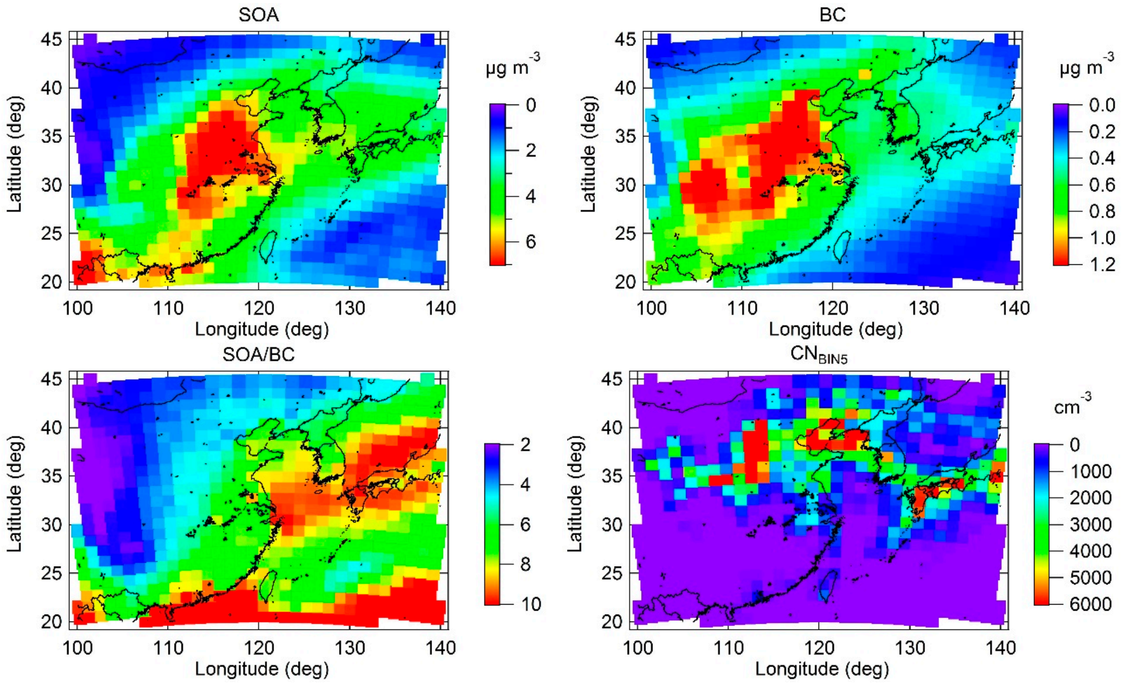
Reviewer's comment:

P10670 L5: It would be useful to see maps of BC, SOA and NPF. SOA is more important in the SE Asia for CCN concentrations: is it because SOA emissions are higher, or because the processes are somehow more efficient? And, in the latter case, what would be the reason?

Response:

The figures below show period-averaged distribution of BC mass, SOA mass, and CN_{BIN5} concentrations and SOA/BC mass ratio at 1 km. CN_{BIN5} is the number concentrations in bin 5 (6.3 – 10 nm), which was used as an indicator of NPF events in our previous studies (Matsui et al., 2011, 2013c). We have added these figures to the manuscript (supplement).

SOA is more important in the SE Asia for $CCN_{0.1}$ concentrations because SOA concentrations and their ratios to preexisting aerosols (see Figure 4 and SOA/BC ratio shown below) are high over the region. High SOA concentrations over the SE Asia are because emissions of SOA precursors (S/IVOCs from biomass burning and biogenic VOCs) are large over the region (Figure 3 of Matsui et al. (2014)).



10)

Reviewer's comment:

P10670 L12: I am confused by figure 6b. M01 and M01_S are overlapping, M01_N has more OA than M01_S. Is it correct? I don't understand how that can be.

Response:

The legend in the submitted version was not correct. We revised the legend of Figure 6b.

11)

Reviewer's comment:

P10671 L2: Fig. 7 does not show local changes, only the regional and time average, hence it does not show what the authors say in this sentence.

Response:

Concentration ratios are calculated for individual horizontal grids (39 × 24 grids) and times (34 days at noon) (39 × 24 × 34 = 31824 data in total), and their average (squares) and 10th – 90th percentiles (vertical bars) are shown in Figure 7. We revised the figure caption of this figure and moved it to supplement in the revised manuscript.

12)

Reviewer's comment:

P10671 L16: when the authors say "overestimation" and "underestimation" they mean with respect to M10_SN. I think it is worth to repeat it, or it seems that they are compared to the real world, i.e. observations. Is there a way to compare these numbers to data?

Response:

We have revised the sentence as follows. “The difference in absorption between the two simulations is due to two effects: the M01 simulation has (1) higher absorption by coating materials (lens effect) and (2) lower BC mass concentrations by efficient wet removal processes (which decreased absorption) than the M10_SN simulation because the M01 simulation assumes internally mixing for all BC particles.”

The validation of absorption enhancement is a future work. If we can measure the ratio of absorption to BC mass concentrations (such as mass absorption cross section), it would be possible to examine the validity of simulated absorption enhancement.

13)

Reviewer’s comment:

P10672 L11: does the lens effect saturate after a certain coating thickness?

Response:

We examined absorption enhancement by SOA (e.g., AAOD increase normalized by SOA increase, from the comparison between the M10_SN and M10_N simulations) but found no clear evidence of its latitudinal dependency. So, we have deleted the sentence from the manuscript.

Technical comments:

Reviewer’s comment:

P10665 L14: eliminate “during”

P10676 L5: change “complicate” into “complicated”

Response:

We have revised the text as the reviewer suggested.