Response to interactive comment on "Size-resolved measurement of the mixing state of soot in the megacity Beijing, China: diurnal cycle, aging and parameterization" by Referee #2

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Major Comments:

This paper presents VTDMA measurements of the size and mixing state of soot containing particles during the CAREBeijing campaign. They use the data to derive a time constant for the conversion of externally mixed soot to internally mixed soot in Beijing. In addition they use observed correlations between the internally mixed soot fraction and other photochemical age indicators (such as the NOz/NOy ratio) to parameterize a calculation of the internally mixed fraction of soot particles using more commonly measured chemical tracers in the absence of VTDMA measurements.

The results presented here are an interesting addition to the current literature on the aging of soot particles and it should be published in ACP after the authors address the following concerns.

Response:

We gratefully appreciate the referee's comments and suggestions. Please find our point-by-point responses in the following text. The suggestions/comments from the reviewers are in italic, our responses are in plain text, and the revision texts in the manuscript are marked in blue.

Comment 1:

*This is a very equation-heavy paper and I don't think that all of the equations presented increase the clarity of the manuscript. For example, it seems to me that equations 2-7 could be combined into 1 or 2 equations. Equations 3 and 7 are already the same. For example you could have Eq 1 as d(nex)/dt = (condensation term ex → in) + (coagulation term ex → in) + emissions + transport + deposition and highlight that that includes both population transfer from ex→in as well as total particle number changes due to physical processes. Then state that, to first order, you are going to ignore everything other than condensation such that the total particle number is assumed to be constant and go right to a combination of 6 and 7 (- dnex/dt = dnin/dt = kex→in*nex for example). Similarly combine 9+10 and, in general, I recommend including only those equations that will help the reader follow the logic rather than converting into equations assumptions that can easily be stated in the text.*

Response:

Very good suggestions and we do appreciate it. Section 2.3 has been reformulated accordingly.

Text:

The turnover rate, $k_{ex\to in}$, can be used to describe the conversion/aging rate of the externally mixed soot to internally mixed particles. It is defined as the rate of percentage change of externally mixed soot particles due to conversion/aging processes (Eq. 2).

$$(\frac{\partial n_{ex}}{\partial t})_{ex\to in} = -k_{ex\to in}n_{ex}$$

(2)

where $(\frac{\partial n_{ex}}{\partial t})_{ex\to in}$ is the rate of change of n_{ex} due to conversion/aging processes.

In case the condensation dominates the conversion, n_{tot}, the number concentration of total (ex+in) soot particles can be considered as a constant. Dividing Eq. (2) by n_{tot}, we have

$$(\frac{\partial F_{in}}{\partial t})_{ex\to in} = k_{ex\to in}(1 - F_{in})$$

(3)

The $k_{ex\to in}$, however, can't be directly solved by substituting n_{ex} or F_{in} obtained from field measurements into Eq. (2) or (3). This is because the observed changes of n_{ex} and F_{in} are subject to multiple processes in the atmosphere, including horizontal/vertical transport (subscribed by "Tran"), emissions (subscribed by "Emis"), deposition (subscribed by "Depo"), condensation (subscribed by "Cond") and the coagulation (subscribed by "Coag") as in Eq. (4)

$$\frac{\partial F_{in}}{\partial t} = \overbrace{(\frac{\partial F_{in}}{\partial t})_{Cond} + (\frac{\partial F_{in}}{\partial t})_{Coag}}^{\text{aging (ex}\to\text{in)}} + (\frac{\partial F_{in}}{\partial t})_{Tran} + (\frac{\partial F_{in}}{\partial t})_{Emis} + (\frac{\partial F_{in}}{\partial t})_{Depo}$$

(4)

in which F_{in} could also be replaced by n_{ex} or n_{in}.

During daytime (the focus period of this study), the condensation dominates the aging processes, especially for relatively large particles (Jacobson, 1997; Riemer et al., 2004). The impact of transport and deposition on the fraction F_{in} is also not prominent because both the internally and externally mixed soot particles undergo similar transport and deposition processes and their ratios are likely conserved (Su et al., 2008; Wiedensohler et al., 2009). Therefore, we assume that $\partial F_{in}/\partial t$ is only controlled by the condensational aging process ("Cond") and emissions ("Emis") while all the other processes are not considered in this study.

When the emission term in Eq. (4) is also neglected, an "apparent" turnover rate can be determined by $k_{ex\to in} = (\Delta F_{in}/\Delta t)/(1-F_{in})$, which attributed all variations in F_{in} to an 'apparent' conversion process. In previous studies, "apparent" $k_{ex\to in}$ of 1.3–5.8% h^{-1} has been reported, corresponding to $(\Delta F_{in}/\Delta t)$ of 1.0–2.3% h^{-1} and F_{in} of 0.2–0.6 (Moteki et al., 2007; Shiraiwa et al., 2007).

Comment 2:

It is hard for me to visualize how the particles might be transitioning between non-BC, internally-mixed BC and externally mixed BC. I wonder, for example if, instead of calling 45%-82% D_{p300}/D_p internally mixed you called D_{p300}/D_p of 30-70% internally mixed, would that change the absolute values of F_{in} but leave the trends (diurnal and as a function of particle size) unchanged? It might be helpful to include a figure of histograms of observed D_{p300}/D_p for at least a subset of the particle sizes/times you are talking about. Maybe 100nm and 200nm at 6:00 and 13:00.

Response:

Yes, we tested several threshold values of $D_{p300}{}^{o}C/D_p$, which defines the soot mixing state. The absolute values of F_{in} are different but the trends are similar. For example, Fig. R1 shows good correlations between hourly F_{in} values calculated by two definitions for 100 nm and 200 nm particles. One (F_{in}, y-axis) is calculated by the same definition as in the paper (45 %< $D_{p,300°C}/D_p$ < 82 % were considered as internally mixed soot particles). The other one ($F_{in}{}^{*}$, x-axis) is calculated by considering all particles with $D_{p,300°C}/D_p$ < 82 % as internally mixed soot particles. Figure R1 has been included in the supplement of the revised manuscript as Fig. S3.
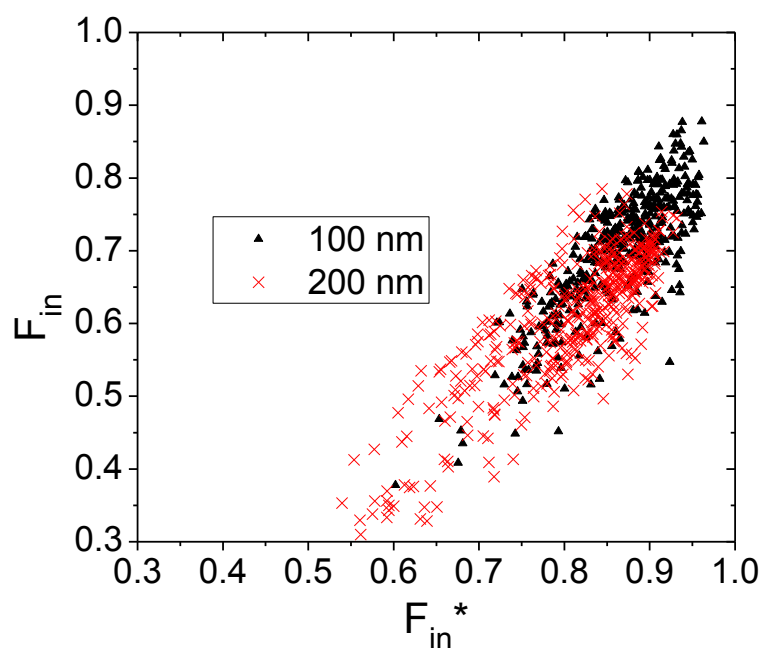

Figure R1: Comparison of F_{in} under different definitions. F_{in} (y-axis) is calculated the same as in the paper (45 %< $D_{p,300°C}/D_p$ < 82 % were considered as internally mixed soot particles). $F_{in}{}^{*}$ (x-axis) is calculated by considering all particles with $D_{p,300°C}/D_p$ < 82 % as internally mixed soot particles.

For a more precise and meaningful description of the mixing state of soot particles, the distribution concept, e.g. the distribution of the relative coating thickness (D_{core}/D_p), would be favorable. However, the problem is that in order to better use and interpret such a distribution concept, supports from an advanced observation-based

aerosol model are desirable. In the present study, we will stick to the definition commonly used in previous VTDMA measurements. We are willing to collaborate with modelers and pursue the distribution concept in future studies.

As suggested by the reviewer, new figures (Fig. R2) of size distributions of nonvolatile residuals with initial diameter D_p of 100 nm and 200 nm, as well as the related texts are included in the revised manuscript to give a hint on the histograms of observed $D_{p,300^oC}$. They have been arranged as Fig. 10 and Sect. 3.7. in the revised manuscript.
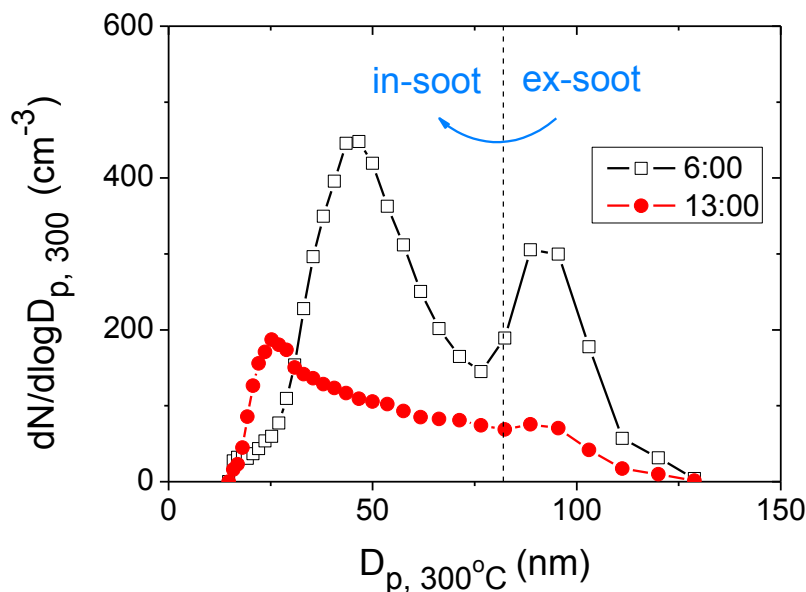
(a)
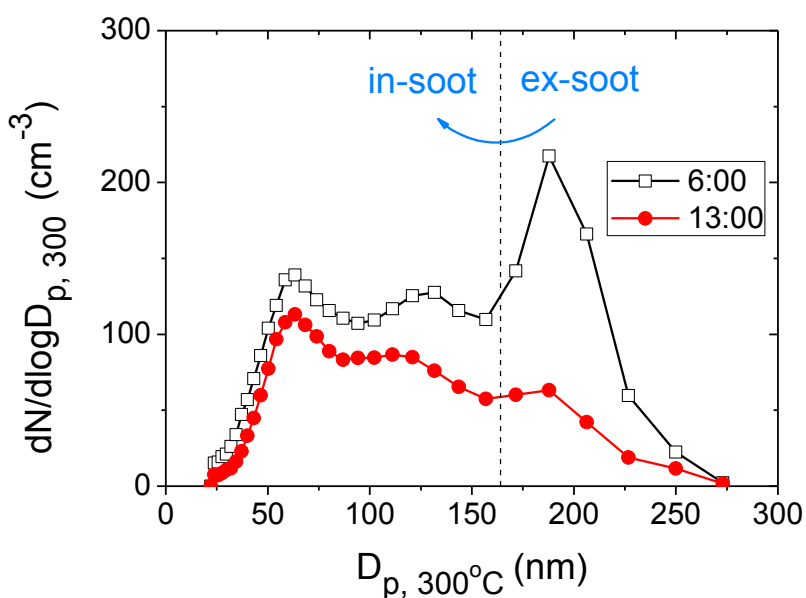
(b)

Figure R2: Size distributions of nonvolatile residuals with initial diameter D_p of (a) 100 nm and (b) 200 nm. $D_{p,300^oC}$ is the diameter of the nonvolatile residuals, i.e.,

particles after being heated at 300°C. The presence of particles with $D_{p,300^\circ C} > D_p$ is due to the transfer function of the DMA used for initial particle selections.

Text:

3.7 Coating thickness distribution

In literature and the present study, the soot particles were classified into two distinct groups (internally and externally mixed soot particles) by certain threshold core/shell ratios (e.g., $D_{p,300^\circ C}/D_p$). However, the transition between the two groups in the atmosphere can be quite smooth, especially in aged air mass (Fig. 10, results at 13:00), and the threshold core/shell ratio should be considered as an arbitrary definition. Such "arbitrary" ratios (e.g., core/shell ratio = 2) have also been used to distinguish different mixing states of soot particles in SP2 studies (e.g., Shiraiwa et al., 2007).

In our case, the use of different threshold ratios will change the absolute values of F_{in} but leave the same/similar trends (see Fig. S3 for example). For a better description of the mixing state of soot particles, we suggest to consider the distribution concept, e.g. the distribution of the relative coating thickness ($D_{p,300^\circ C}/D_p$), in future studies.

Comment 3:

The discussion of the hygroscopicity is rather disconnected from the rest of the discussion. As it now reads the only utilization of the CCNC data is to look at the variation of the width of the kappa distributions as a function of the internally mixed soot fraction measured with the VTDMA and that information is presented simply as a confirmation of measurement reliability. It seems that more could be said about this data. While I realize that, since soot particles likely contribute a very small fraction of the total aerosol mass, it is difficult to relate kappa to soot particle properties but it would be interesting to know if there is any trend in kappa as a function of Fin. Even if there isn't, it would be reasonable to mention that fact and to explain why it's not surprising to see the effect of soot particles manifesting solely in sigma and not in kappa itself. Also, this discussion should probably at least be referenced in the conclusions.

Response:

As suggested by the reviewer, we have included a new figure (Fig. R3) and discussions about the correlation between kappa ($\overline{\kappa_g}$) and F_{in} in the revised manuscript.

Text:

During the CAREBeijing campaign, positive correlation between hourly $\overline{\kappa_g}$ and F_{in} was also found (Fig. 2b) as a result of their concurrent increase during the aging process. The increased $\overline{\kappa_g}$ can be attributed to the increase of IM fraction of aerosol particles. The IM is in general more hygroscopic and CCN-active than the rest of the aerosol chemical species (Pöschl, 2011; Rose et al., 2011). The increase of IM fraction

leads to a corresponding increase of the overall aerosol hygroscopicity and hence $\overline{\kappa}_g$, which represents an averaged hygroscopicity of aerosols (Su et al., 2010).
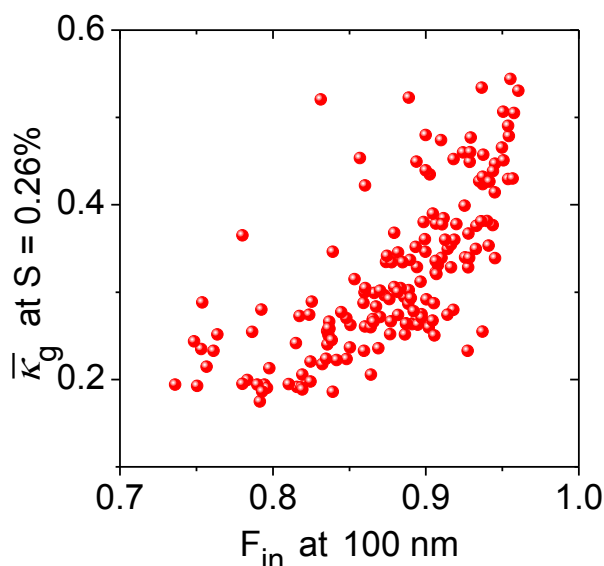

Figure R3. Comparison of aerosol hygroscopicity and soot (NVP) mixing state during the CAREBeijing-2006 campaign. The parameter $\overline{\kappa}_g$ and $\sigma_{\kappa,g}$ were calculated from the aerosol hygroscopicity (i.e., κ) distribution derived at supersaturation $S = 0.26$ % (Su et al., 2010); and F_{in} was determined for particles of diameters at 100 nm by the VTDMA measurements. Note that the mean activation diameter observed at $S = 0.26$ % is 85 nm (Gunthe et al., 2011).

Comment 4:

I have significant difficulties with your discussion of the estimation of the soot emission rate and its incorporation into the calculation of the actual turnover rate of soot. Mainly, even after multiple readings, I don't totally understand what you did or how sensitive your reported turnover rates are to uncertainties in this calculation. To break my questions down into manageable chunks: a.) If I understand your discussion of figure 5 correctly, the "emission rate" trace is simply a plot of normalized CO. If that's the case, just call it that and say that you assume BC emissions have a similar pattern. If not, you should better describe how you got that trace. b.) While I see why it is important to minimize the impacts of vertical and horizontal transport to calculate emissions, it seems that your choice of acceptable time window entirely determines your calculated rate. You say that you wound up doing the calculation at 20:00 because that was when Emis/m was highest but that's not when it reached its peak. It had peaked earlier in the day and that was merely when it was at a maximum for the times you selected (somewhat arbitrarily it seems). If, instead, you had chosen to look at 19:00-7:00 your "maximum" would have been at 19:00 and you would calculate a different emission rate, wouldn't you? c.) Once you have this 13%/hr

emission rate, I don't see how you can assume that that stays constant over the whole day. If that were true (that concentrations continue to go up by 13% /hr over the day) it would be impossible to replicate the observed diurnal profile of EC unless there is significant lateral transport. If lateral transport is that important at certain times of day shouldn't you exclude them from the overall analysis? Later you say that you calculate Emis tot by multiplying a "generic diurnal cycle of emissions" by the 13%/hr so perhaps I have entirely misunderstood what number you are trying to extract but I don't understand what you did or why it makes sense. I would recommend starting this calculation with Eq 16 to illustrate how emissions can impact Fin and to justify the need to calculate Emistot as a function of time. Then you need to clarify your discussion of the calculation of Emistot and how you get from an observation-based calculation of emissions at a particular time of day to an inferred diurnal profile of emissions.

<u>Response:</u>

a) The emission rate of EC is not taken from the CO concentration but from the emission rate of CO. This has been clarified in the manuscript (see revised text below).

b) As mentioned on Page 32176, line 16, we did a sensitivity test by varying the emission rates by a factor of two. The absolute values of the turnover rate are changed but its diurnal profile still holds (Fig. R4). Figure R4 has been included in the revised manuscript as Fig. S2.
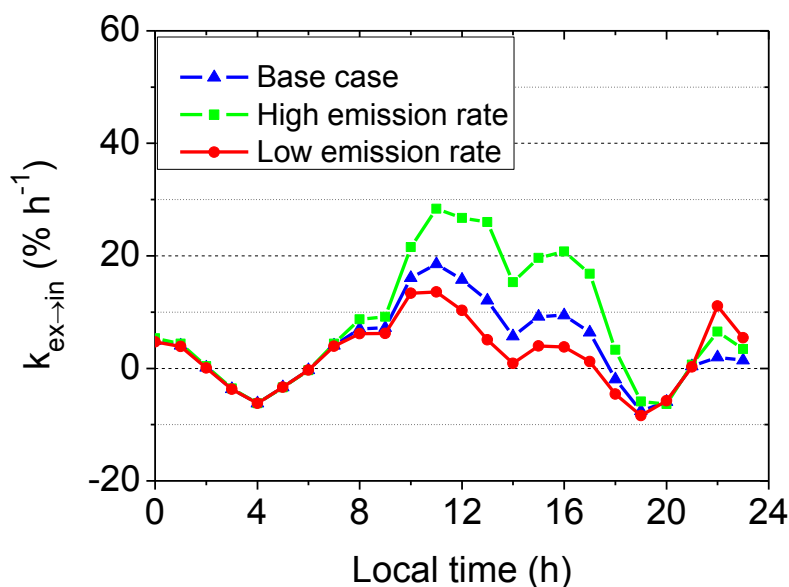
Figure R4. Sensitivity of the actual turnover rate of soot (NVP), $k_{ex \to in}$, to the emission rate ('Emis' in the old version; $(\partial[EC]/\partial t)_{Emis}$ in the revised version). The base case is the same as the case of $\beta = 0.6$ in Fig. 6. In high/low emission cases, the emission rate has been increased/reduced by a factor of two, respectively.

c) The emission rate ('Emis' in the old version; $(\partial[EC]/\partial t)_{Emis}$ in the revised

version) is not assumed constant but with a diurnal variation (as 'Emission' in Fig. 5 in the manuscript). Accordingly to the suggestions of the reviewer, we have reformulated the section for clarity.

Text:
Page 32174 line 1 to Page 32175 line 4

The emission rate can be divided into a diurnal profile describing its relative variation and the absolute value of any point on this profile. In this study, we adopt the diurnal profile of CO (carbon monoxide) emissions from traffic systems in Beijing areas (Zhou et al., 2010) and assume that soot emissions have a similar diurnal pattern. Then measurement data of EC are used to estimate the absolute emission rate at certain period during night-time. With these two kinds of information, the whole emission profile can be quantified.

Note that the emission rate is often expressed as a flux in mass per area per time while its impact on EC concentrations, $(\partial[EC]/\partial t)_{Emis}$, has a unit of mass per volume per time. From the aspect of Eulerian grid models, the diurnal profile (relative variation) of the emission flux and $(\partial[EC]/\partial t)_{Emis}$ is the same while their absolute values differ by a factor of the grid height.

Figure 5 shows the diurnal profile of the emission rate $(\partial[EC]/\partial t)_{Emis}$ used in this study. To estimate its absolute values, we tried to find a time period when the concentration variation is dominated by emissions, i.e., $\partial[EC]/\partial t \approx (\partial[EC]/\partial t)_{Emis}$. The EC concentration is also shown in Fig. 5. It is clear that [EC] is not always increasing though $(\partial[EC]/\partial t)_{Emis}$ is always positive. This is because transport plays an important role on the diurnal variation of [EC].

To minimize the impact of transport and obtain an optimal estimate on absolute values of $(\partial[EC]/\partial t)_{Emis}$, we adopted the following criteria, eliminating days with average wind speed $>2m\ s^{-1}$ (20, 22 August, 3, 4, 5, 6 and 8 September), excluding the time periods with strong vertical mixing (from 8:00 to 19:00 LT), and choosing periods with largest ratio of normalized $(\partial[EC]/\partial t)_{Emis}$ to [EC] (at 20:00 LT). The reason of choosing low wind speeds and night-time periods is to minimize the impact of horizontal and vertical transport processes on [EC]. Large ratios of $(\partial[EC]/\partial t)_{Emis}/[EC]$ ensures that the emission term could dominate the variation of [EC].

Finally, $\Delta[EC]/\Delta t$ at 20:00 (~0.89 $\mu g\ m^{-3}\ h^{-1}$) was taken as an optimal estimation of $(\partial[EC]/\partial t)_{Emis}$ at 20:00. It was then used to calculate $(\partial[EC]/\partial t)_{Emis}$ for the rest time of a day by applying the diurnal profile in Fig. 5. Table 3 summarized the diurnal variation of the measured [EC] and the calculated $(\partial[EC]/\partial t)_{Emis}$. The mean emission rate is ~13% of the mean soot concentration per hour (13% h^{-1}).

Table R1. Mean diurnal variation of [EC] (EC concentrations) and $(\partial[EC]/\partial t)_{\text{Emis}}$ (emission rates)

Time	[EC]	$(\dfrac{\partial[EC]}{\partial t})_{\text{Emis}}$	$\dfrac{(\partial[EC]/\partial t)_{\text{Emis}}}{[EC]}$	Time	[EC]	$(\dfrac{\partial[EC]}{\partial t})_{\text{Emis}}$	$\dfrac{(\partial[EC]/\partial t)_{\text{Emis}}}{[EC]}$
	$\mu g\ m^{-3}$	$\mu g\ m^{-3}\ h^{-1}$	% h^{-1}		$\mu g\ m^{-3}$	$\mu g\ m^{-3}\ h^{-1}$	% h^{-1}
0	6.1	0.191	3.1	12	3.9	0.954	24.5
1	5.8	0.120	2.1	13	3.1	1.092	35.2
2	6.3	0.088	1.4	14	2.7	0.952	35.3
3	6.7	0.074	1.1	15	2.7	0.856	31.7
4	7.2	0.058	0.8	16	2.6	1.114	42.8
5	7.7	0.108	1.4	17	2.8	1.168	41.7
6	8.3	0.407	4.9	18	2.7	0.986	36.5
7	8.0	0.832	10.4	19	3.3	1.052	31.9
8	7.5	0.934	12.5	20	4.4	0.894	20.3
9	7.8	1.022	13.1	21	5.1	0.564	11.1
10	5.1	1.020	20.0	22	5.7	0.495	8.7
11	4.4	1.052	23.9	23	6.1	0.232	3.8

Page 32175 line 9 to Page 32176 line 11

Two processes are considered in the model: the condensational aging process that converts equal amounts of externally mixed particles into internally mixed particles and the emissions. We also assume that all (or most) particles stay in the same size bin after the 1-h aging process. This assumption, to a large extent, simplifies the following analysis, and could at least be valid for a broader size bracket, e.g., [100 nm, 350 nm]. The transport and dry deposition processes are supposed not to significantly change F_{in} (assuming n_{in} and n_{ex} have the same gradient and dry deposition velocity) and are therefore ignored in this analysis.

Then the variation of F_{in} in the time interval Δt can be expressed by Eq. (5):

$$\Delta F_{\text{in}} = \frac{n_{\text{in}} + (\frac{\partial n_{\text{in}}}{\partial t})_{\text{ex}\rightarrow\text{in}} + (\frac{\partial n_{\text{in}}}{\partial t})_{\text{Emis}}}{n_{\text{tot}} + (\frac{\partial n_{\text{tot}}}{\partial t})_{\text{Emis}}} - F_{\text{in}} = \frac{n_{\text{in}} + k_{\text{ex}\rightarrow\text{in}} n_{\text{ex}} + \beta(\frac{\partial n_{\text{tot}}}{\partial t})_{\text{Emis}}}{n_{\text{tot}} + (\frac{\partial n_{\text{tot}}}{\partial t})_{\text{Emis}}} - F_{\text{in}} \tag{5}$$

in which $\beta = (\partial n_{\text{in}}/\partial n_{\text{tot}})_{\text{Emis}}$ is the fraction of internally mixed soot in fresh emitted soot particles, which is assumed to be a constant. The minimum $F_{\text{in}} \sim 0.6$ can be considered as the upper limit of β, and we also tested other β values (0.2 and 0.4) for sensitivity studies.

Dividing the numerator and fraction of the first term on the right-hand side of Eq. (5) by n_{tot}, we have

$$\Delta F_{in} = \frac{F_{in} + k_{ex \to in}(1 - F_{in}) + \beta[\frac{1}{n_{tot}}(\frac{\partial n_{tot}}{\partial t})_{Emis}]}{1 + [\frac{1}{n_{tot}}(\frac{\partial n_{tot}}{\partial t})_{Emis}]} - F_{in}$$

(6)

Assuming $\frac{1}{n_{tot}}(\frac{\partial n_{tot}}{\partial t})_{Emis} = \frac{1}{[EC]}(\frac{\partial [EC]}{\partial t})_{Emis}$ (see Table 3), $k_{ex \to in}$ becomes the

only unknown parameter and can be calculated by solving Eq. (6).

Comments 5:

Similar to the above, I am unclear on what you are doing in Section 3.5. It would help if you gave units for Kshift. The units given in the figure are nm^-2 but there must also be a t^-1?

Also, the equations from Seinfeld and Pandis gave you changes in size distributions as a function of time but I don't see how you can talk about only internally or externally mixed BC using the same equations because you are looking at not only growth for the whole population but also a conversion from externally mixed to internally mixed particles. You probably even have some particles that are falling off into the non-BC categorization at the same time. (a 30nm particle that has a 15nm soot fraction, once it grew to the 50nm size range would no longer be classified as soot containing).

Are you trying to quantify the rate at which internally mixed particles in the aitkin mode transition to the accumulation mode? If all you are trying to say is that condensational growth will tend to move particles from the smaller size bins into the accumulation mode, you could do that using equations 18-20 and figure 7. It is much more complicated to try to tease out the combined effects of morphology changes, conversions between externally mixed to internally mixed and the overall condensational growth of the whole population. If you want to do that, I think this treatment is poorly explained and probably overly simplistic. Also, I don't think it adds substantially to the conclusions of the paper. The final paragraph of the section is the only place where you relate the equations presented to the observed behavior and it is both qualitative and confusing. I don't understand how condensational growth could ever lead to an increase in externally mixed particles for example. Also, I don't understand how emissions "overwhelm condensation" to increase Fin for 30nm particles late in the day when the coated fraction of the emissions should be lower than the observed Fin. In summary, I would recommend omitting the second half of this section. If the discussion must be kept it needs a substantial rewrite.

Response:

We have double checked the unit of K_{shift} and found nm^{-2} correct. The unit of t was counteracted by t^{-1} in the parameter A in Eq. (20).

Our main purpose is to demonstrate the different impact of changes in size distribution for Aitken mode and accumulation mode particles, rather than to predict the temporal evolution of size distributions or mixing states. Therefore we prefer to decouple it rather than to mix it with conversions and emissions.

We agree with the reviewer and have deleted the second half of this section in the revised manuscript.

Comment 6:

In section 3.6 I think you need to make very clear up front that this parameterization is specific to Beijing. That point should already be apparent to the reader and it is mentioned in the conclusions but it needs to be highlighted in the discussion as the emission ratios of the different observed components could be wildly different in other locations with different sources (and with different OH concentrations). I don't understand where equation 27 comes from and I therefore can't assess how specific this parameterization is to Beijing but the authors should comment on potential sources of variation for these numbers.

Response:

We have added the following sentence in the end of section 3.6.
Text:
This parameterization method requires caveats because considerable variability can be expected between different sites or even at a given site, like the scattered data shown in Figs. X8 and 9. To validate the parameterization methods, more measurements should be carried out in other environments.

Equation 27 is a function we found that could well describe the observed size dependence. We have clarified this fact in the revised manuscript.

Text:
Secondly, we find that the size-resolved F_{in} in the accumulation mode can be calculated by the following equation from F_{in} at 150 nm:

Minor Comment 1:

What is the lower limit for the size of particles detected by the VTDMA?

Response:

In the present study, the lower detection limit of the second DMA in the VTDMA system was ~ 12 nm (Wehner et al., 2009). To save the scanning time, the lower limits in each diameter scans were set as in Table R2.

Table R2. Lower limits of particle diameters scanned at 300℃ in VTDMA measurements

	Initial diameter D_p (nm)						
	30	50	100	150	200	260	320
Smallest $D_{p,300^oC}$ (nm)	10	12	15	21	22	27	32

Minor Comment 2:

   You should have some references to previous work using the NOx/NOy and (IM+OM)/EC ratios as indicators of photochemical age.

Response:

We have added several references in which the same/similar indicators were used/discussed: (Parrish et al., 1992; O'Brien et al., 1997; Cheng et al., 2006).

Minor Comment 3:

   In the conclusions "time courses" → "behavior"

Response:

We have implemented the suggestion in the revised manuscript.

Minor Comment 4:

   Figure 9 - It would be interesting to see this figure color-coded by the size of the particles.

Response:

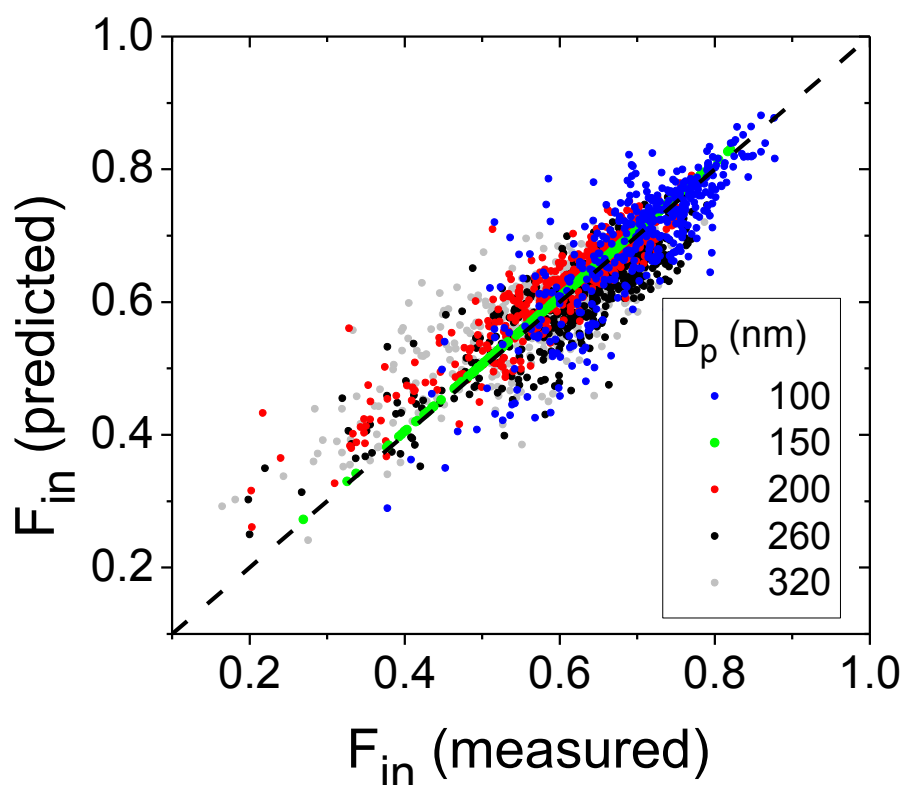   We have replotted the figure in the revised manuscript (Fig. R5).

Figure R5: Measured and predicted F_{in} over 100 nm to 320 nm (2145 data points). The dashed line represents the 1:1 line.

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