Response to Referee#1

We would like to thank Referee#1 for the review and the constructive suggestions for the improvement of our manuscript, which will be implemented upon revision. Detailed responses to the individual comments are given below.

Referee comment:

Page 2: "apparent elemental and organic carbon". I would remove the word "apparent" since it is not clear what you mean. You can say "observed elemental and organic carbon inferred from thermo-optical measurements" if that makes you feel more comfortable than simply removing the word "apparent". I would also remove the subscript "a" for the variable "EC_a" (which I assume stands for "apparent"). There is uncertainty in the EC measurement, and perhaps there are conditions where the measurement does not accurately describe the elemental carbon content of the aerosol, but every measurement is subject to uncertainties and caveats. For instance, particle size inferred from light scattering measurements may depend on the particle refractive index (which depends on composition) and particle shape. These unknowns add uncertainty to the sizing measurement - but measuring particle size with light scattering is still a valid technique. Putting the word "apparent" here makes it seem that the EC measurement technique is not valid. It would be better to quantify the uncertainty in the measurement, rather than to cast a vague doubt about the measurement technique.

Authors' response:

Thanks for the suggestion; we understand the referee's concern. Note that EC is just operationally defined and difficult to measure and hence should be regarded as apparent elemental carbon (Andreae and Gelencsér, 2006). For simplicity and to avoid confusion, we will delete the subscript "a".

Referee comment:

Page 2: The equation assumes that EC is not contributing much to the CCN-active aerosol mass. This is a bit confusing when you say later in the abstract that EC mass fraction is _30% in the fresh pollution outflows. I would expect that the EC mass present in the fresh outflows can become internally mixed as the aerosol population ages, and therefore should be included in the kappa parameterization. I understand that EC does not contribute to soluble mass, but it can contribute to total mass (the denominator of your parameters "f_org" and "f_inorg") depending on how these mass fractions are defined. Please clarify this point, and be consistent and clear in your terminology. It is confusing that you apparently define mass fraction one way (relative to AMS organic + inorganic) and then in the next paragraph define it a different way (relative to soot + AMS organic + inorganic)

Authors' response:

We thank the referee for raising this point and will clarify the terminology in the revised manuscript. Please note that in the abstract we correctly mention that κ_p is "parameterized as a function of AMS based organic and inorganic mass fractions", and does not include EC.

Nevertheless, EC may contribute to the overall particle hygroscopicity. Including the measured EC mass in the κ parameterization, however, is not possible in the present case since the overlap in EC and AMS data is not continuous during the measurements, and more importantly, unlike the AMS measurements EC measurements were not size-resolved.

Referee comment:
Page 4: replace "air particulate matter" with "atmospheric particulate matter".
Authors' response:
We will change "air particulate matter" to "atmospheric particulate matter".

Referee comment:
Also, is there another more permanent source you could reference for the population of Beijing? http://en.wikipedia.org/wiki/Beijing would be a better source (and it says 19.6 million, not 22 million).
Authors' response:
In the revised manuscript, we will reference the web page of the National Bureau of Statistics of China and change the sentence to "Beijing, the capital of China, is a megacity with a population of about 20 million people (http://www.stats.gov.cn/english/newsandcomingevents/t20110429_402722516.htm).

Referee comment:
You write that in addition to the "domestic, industrial, and traffic emissions" there is "regional pollution originating from the highly industrialized areas to the south" – are agriculture/biomass burning and dust other important sources that you should mention here?
Authors' response:
We do not think agriculture/biomass burning and similar other sources contribute substantially in Beijing. If so, then it may be episodic, which we did not see during our measurement period. In the revised manuscript we will adjust this paragraph accordingly.

Referee comment:
Page 7: the calibration line you report for S assumes that temperature is constant. You should at least mention this, and also mention how much a 10oC temperature change will effect supersaturation (10oC appears to be approx. how much the temperature changes over any given day, from Fig. 3). From Rose et al. 2008 (Fig. 8a), a temperature change of 10oC can change supersaturation by more than 15%. I'm guessing that pressure and CCNc flow rate do not change much, and therefore the temperature variability is your biggest source of uncertainty - but that is worth looking into as well. It is possible that a temperature increase of 10oC results in a >20% decrease in supersaturation (depending on the thermal efficiency of the instrument). Therefore your reported uncertainty of < 10% seems a bit low to me, even

though a precise solute activity model was used, since the operating parameters were not factored in to the estimated supersaturation (except for deltaT).

Authors' response:

We understand the referee's concern. However, kindly note that the 10°C temperature variability on any given day as shown in Fig. 3 is for ambient temperature measured on the roof top close to the inlet, and not for the environment of the CCNC. The temperature within the room where the CCNC was installed was fairly constant over the entire period of measurements as it was modulated by means of an air conditioner. Hence, we are confident that the reported uncertainty of <10% for the estimated supersaturation, especially the lowest supersaturation, seems to be reasonable.

Referee comment:

Also, at high CCN concentrations the supersaturation in the CCNc can be significantly depressed, which should be at least mentioned (Lathem and Nenes, Water Vapor Depletion in the DMT Continuous-Flow CCN Chamber: Effects on Supersaturation and Droplet Growth, Aerosol Science and Technology, 45, 5, 2011) - How high were CCN concentrations at any given time in the instrument (a given size cut and supersaturation)?

Authors' response:

During our measurements the maximum CCN number count in the CCNC column did not exceed more than ~400 cm^{-3}, which is well below the CCN concentration above which the supersaturation is known to decrease significantly (~5000 cm-3; Lathem and Nenes, 2011). In the revised manuscript we will include this statement.

Referee comment:

Page 8: for the correction factor, you should say what units D is supposed to be in (presumably nm), or rather the units of x2. Also, report the Rˆ2 for this fit.

Authors' response:

We thank the referee for pointing this out. We will include R^2 of the fit and the unit of D in the revised manuscript.

Referee comment:

For the multiply charged correction, do you just assume that all of the multiply charged particles you calculate from the aerosol size distribution and charge distribution act as CCN? (since they are larger and likely more hygroscopic, according to Fig. 6, than the singly charged particles). What kind of aerosol charger did you use upstream of the DMA?

Authors' response:

For the charge correction we do not assume that all of the multiply charged particles act as CCN. The number concentration of the activated multiply charged particles is rather calculated from the CN number concentration and the activated fraction (as can be obtained from the CCN efficiency spectrum) at the respective larger sizes. For more details we refer to Frank et al. (2006). The bipolar charger we used upstream of the DMA was a Ni-63 (555 MBq).

Referee comment:
Also, why doesn't the cumulative Gaussian distribution function seem to fit very well for particle sizes just above the inflection point for your dataset?
Authors' response:
The Gaussian fit is just an approximation. It will depend on the actual hygroscopicity, which cannot be perfectly fitted and expected to be a single Gaussian fit.

Referee comment:
Page 9: It is not correct to say that not accounting for the externally-mixed CCN-inactive particles in calculation of kappa is the same as obtaining the "effective overall properties" of the aerosol population. kappa and MAF can vary independently from each other, and they have different effects on the shape of the CCN spectrum. The 2- parameter fit will only have predictive ability when the externally-mixed fraction is low. When MAF is < 1, the 2-parameter fit will always overestimate D_a. Just because this bias acts in the same direction as a lower MAF (reducing predicted CCN concentrations for a given S), this doesn't mean that the two biases (overestimate of MAF and overestimate of D_a) cancel each other out. Assuming MAF = 1 also does not help with understanding or modeling the physical processing that are occuring. Therefore, the 2-parameter fit is inferior, but it seems that you are stating the opposite. I don't know why the 2-parameter fit is used at all.
Authors' response:
Ambient aerosols generally have a κ-distribution rather than a single κ. The results based on 2-parameter and 3-parameter fits both gave mode κ-values rather than a whole distribution. From this aspect, the κ-values from both fits represent the "effective overall properties (i.e. hygroscopicity distribution)" of a certain aerosol population.
The particles will all be activated when their sizes (supersaturation) become large (high) enough, and for the CCN spectra (κ-distributions) MAF of the complete distribution must always be 1. This is not an assumption but a fact, and it is the base of the 2-parameter fit (MAF). The 2-parameter fit can be considered as a 3-parameter fit to the whole distribution, while in a 3-parameter fit only part of the CCN spectra (κ-distributions) were taken into account. Therefore, we made the following statement: "the 3-parameter fit results represent the average properties of the CCN-active aerosol particle fraction, whereas the 2-parameter fit results are proxies for the effective overall properties".
The reason of using the 2-parameter fit is because it has shown good predictive ability in previous studies. We have demonstrated that κ_t is suited for effectively predicting the CCN number concentration under a wide range of CCN number concentrations and mixing states (e.g. Gunthe et al. 2009; Rose et al. 2010).

Referee comment:

Although you are basically copying the text from Rose et al, 2010, it is not correct to say "The difference between unity and maximum observed CCN efficiency... represents the fraction of externally mixed CCN-inactive particles at D_max or averaged over the diameter range of D_a to the largest diameter of the measured spectrum (D_max)". Mixing state is defined for a given (constant) particle size. For a given size, if you increase the supersaturation, you activate more particles, until the point when all CCN-active particles have activated - at that point, you can say that what remains are the "externally-mixed CCN-inactive particles". In Fig. 2a, MAF(0.07%S)=0.81 - but when you increase the supersaturation, MAF(0.26%S)=0.98. Just because you measure MAF(0.07%S)=0.81, that doesn't mean 19% of the _300nm particles are CCN-inactive - apparently, they just need a slightly higher supersaturation in order to activate. Therefore, your dataset shows that the aerosol composition for these large particles is a continuum, rather than two discrete populations.

Authors' response:

In our view, the mixing state is not only defined for a given particle size but is defined in a 2-D space of chemical composition vs. size (diameter). A group of sulfate particles around 50 nm diameter and soot particles around 200 nm would still be called externally mixed.

As shown by Su et al. (2010), a single mode (smooth continuum) of aerosol composition and hygroscopicity will result in CCN spectra with the appearance of a single-mode CDF (cumulative distribution function). In this case, the activated fraction CCN/CN reaches unity after a single sigmoid increase as shown by the green lines in Fig. R1. This is not only the case for a given particle size but also for CCN scans at a given supersaturation and varying particle size as illustrated in Figs. R1 and R2. Discrete modes of particle composition and hygroscopicity will result in CCN spectra with the appearance of a bimodal or multimodal CDF, in which two or more steps are needed for the CCN/CN fraction to reach unity, i.e. a stepwise or sigmoid increase followed by a flat plateau, then another stepwise or sigmoid increase and a flat plateau, etc. Thus, a plateau level of CCN/CN <1 indicates the existence of at least two discrete modes in the κ-distribution and thus at least two particle populations with different chemical composition.

Our measurement data shown in Fig. 2a resemble a bi-modal CDF (red lines in Fig. R1), rather than a single-mode CDF. Similar to Rose et al., 2011 (Fig. 5), the non-activated fraction ($1-\mathrm{MAF}_f$) plotted against $D_a+3\sigma_a$ (the smallest particle size at which CCN spectrum reaches its maximum), follows a similar trend as the number fraction of low volatile particles obtained from parallel VTDMA measurements. This finding supports the interpretation that $1-\mathrm{MAF}_f$ represents the fraction of externally mixed weakly CCN-active particles with $\kappa < \kappa_c(D_{max},S)$ at D_{max} (Su et al., 2010). Detailed analyses of hygroscopicity and volatility distributions, however, go beyond the scope of the present study, which is focused on the contrast of CCN properties in air masses flowing into and out of Beijing. Follow-up studies using VTDMA data will address the topic of aerosol mixing state in more detail.

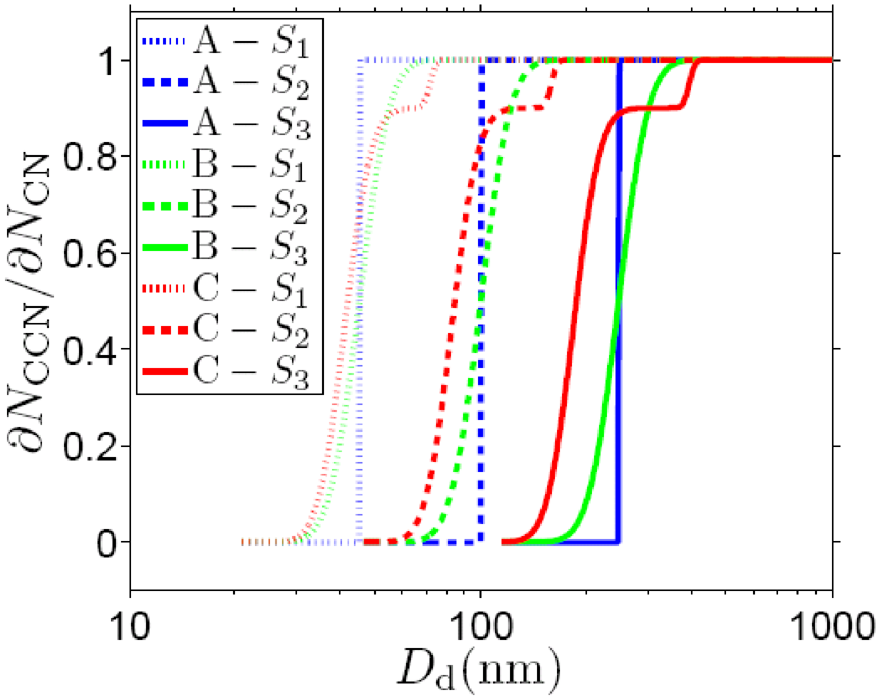

Fig. R1: CCN efficiency spectra (activation curves) of hypothetical model aerosols. Each line represents the spectrum of one type of mode aerosol (Case A, B, or C as specified in Table 1) obtained by D_d scans at a fixed supersaturation S. The supersaturation levels are S_1=0.86%, S_2=0.26%, or S_3=0.067%. This figure is taken from Su et al. (2010, Fig. 3).

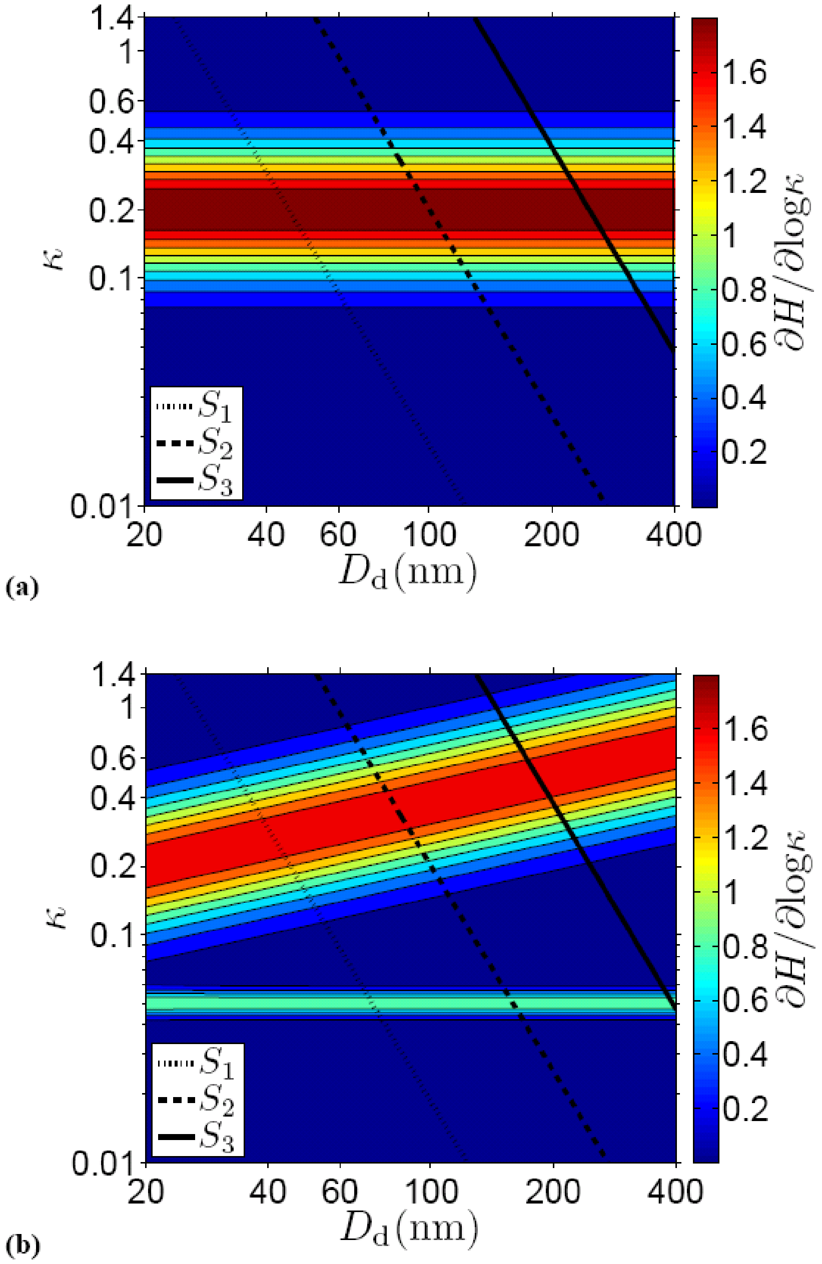

Fig. R2: Logarithmic probability distributions of particle hygroscopicity, $h_{10}(\kappa, D_d) = \partial H(\kappa, D_d)/\partial \log\kappa$, plotted over effective hygroscopicity parameter (κ) and dry particle diameter (D_d) for model aerosols of Case B (a) and of Case C (b). The tilted black lines indicate D_d scans at fixed supersaturations (S_1=0.86%, S_2=0.26%, S_3=0.067%). This figure is taken from Su et al. (2010, Fig. 4).

Referee comment:

By the way, Table 1 and Fig. 2a do not match up. For the entire campaign, MAF_m(0.26%S) appears from Fig. 2a to be 0.98, not 0.91 as reported in Table 1. and MAF_f(0.26%S) > 0.9 (from Fig. 2a) while Table 1 reports MAF_f(0.26%S) = 0.90. I did not check all values in the table.

Authors' response:

As specified in the manuscript, the values given in Table 1 are arithmetic mean values while median values are presented in Fig. 2a. A table of median values is given in the online supplement (Table S1).

Referee comment:
In fact, I think it is incorrect to use the terminology maximum activated fraction (MAF) in this way, first of all because the fraction of activated particles for a given size maybe greater than what you measure at some arbitrary supersaturation (as explained above) and also because you cannot assume that the activated fraction will always increase with particle size (the activated fraction of large particles can actually be lower than smaller particles, due to differences in their composition and mixing state – therefore the activation spectrum as a function of particle size is not necessarily a cumulative distribution).
Authors' response:
As stated in the manuscript, our definition of MAF refers specifically to our measurement data and fit results obtained for a given set of supersaturations and range of diameters. We see no point in defining MAF differently, because the maximum activated fraction will always reach unity if the supersaturation and diameter range is unrestricted. As explained below, the CCN spectra observed at constant supersaturation are similar to those that would be observed at constant particle size when the κ-distribution exhibits a smooth size-dependence.

Referee comment:
To have a cumulative distribution, you would have to see how the activated fraction changes as a function of supersaturation, not particle size (See Fig. 3, Cerully et al, Aerosol hygroscopicity and CCN activation kinetics in a boreal forest environment during the 2007 EUCAARI campaign, ACPD, 11, 15029-15074, 2011).
Authors' response:
Yes, the referee is right that the cumulative distribution is a function of supersaturation (*S*). However, as shown in the work of Su et al. 2010, the shape of the cumulative distribution function (CDF) at constant D would be similar to that at constant *S* for smooth variations of CDF against particle sizes. For example, Fig. R3 shows similar CCN spectra at constant D and at constant *S* for a κ-distribution with $\kappa = 0.2(D/20nm)^{0.4}$ (similar to the CAREBeijing study). Considering other potential uncertainties related to measurement techniques and conditions, the mathematical deviations between partial and total derivative in such cases appear relatively minor.

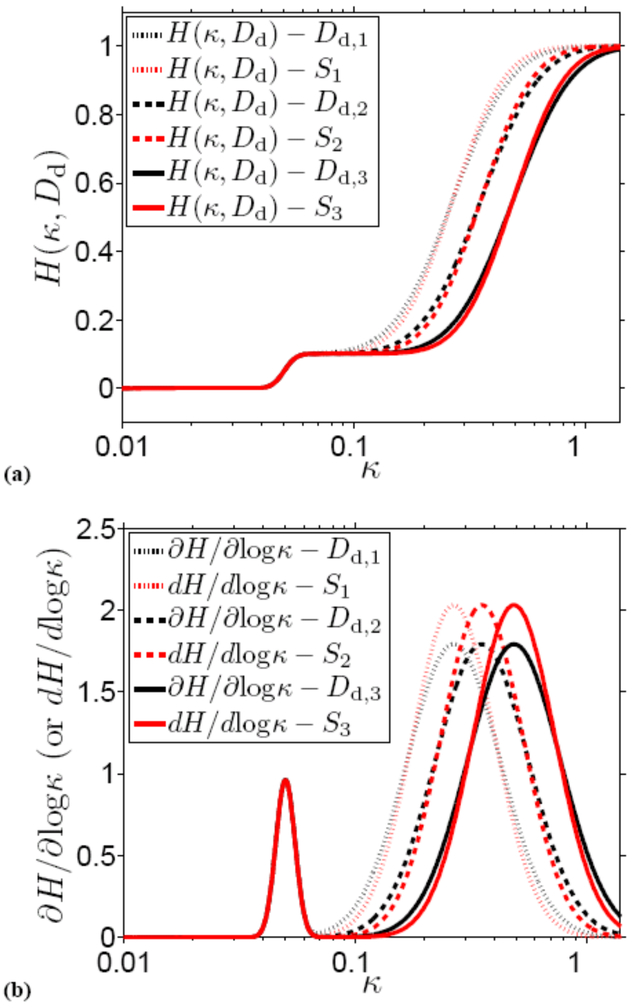

Fig. R3. Cumulative particle hygroscopicity distributions, $H(\kappa,D_d)$ (a), and corresponding logarithmic probability distributions, $\partial H(\kappa,D_d)/\partial \log \kappa$ or $dH(\kappa,D_d)/d\log \kappa$ (b), for the model aerosol of Case C obtained by method I ("S scan" at $D_{d,1}$, $D_{d,2}$ or $D_{d,3}$) and method II ("D_d scan" at S1=0.86%, S2=0.26% or S3=0.067%). This figure is taken from Su et al. (2010, Fig. 6).

Referee comment:

I don't believe you are capable of determining the externally-mixed fraction of CCNinactive particles for sizes < _80nm with this dataset, since you have only 3 datapoints (supersaturations) for any given particle size. You can say that at least _80% of 60 nm particles activate. and you can say that at least _60% of 45 nm particles activate. but you do not know if the supersaturation were increased further slightly (> 0.86%) if more of the 45 nm particles would activate or not. Therefore, you cannot say how the externally-mixed fraction changes as a function of particle size. You can only say that particles > _80nm do not have a significant externally-mixed CCN-inactive fraction (for the average).

Authors' response:

The referee is right. Scans at higher supersaturation (S) are needed to determine the fraction of CCN-inactive particles for smaller particles. For example, S>1% are needed to determine the fraction of particles with κ <0.1 at 45 nm (marked by blue dashed line in Fig. R4).

We did not mean to say that MAF (or the externally-mixed fraction) of smaller particles (e.g., 45 nm) could be determined from our measurements. Therefore, we changed the following sentence “The difference between unity and the maximum observed CCN efficiency ($1-\text{MAF}_m$ or $1-\text{MAF}_f$) represents the fraction of externally mixed CCN-inactive particles at D_{max} or averaged over the diameter range of Da to the largest diameter of the measured spectrum (D_{max})” to “The difference between unity and the maximum observed CCN efficiency ($1-\text{MAF}_m$ or $1-\text{MAF}_f$) represents the fraction of externally mixed CCN-inactive particles measured at D_{max} or fitted over the whole diameter range of each scan. Though derived from a fit to the whole scan, MAF_f is only valid at D_{max} and the nearby diameter range with a plateau of activated particle fraction N_{CCN}/N_{CN}”.

Indeed, for D=45 nm, we do not derive a full κ-distribution from our data. The coverage of κ-distributions by CCN scans is illustrated in Fig. R2 (Su et al. 2010).

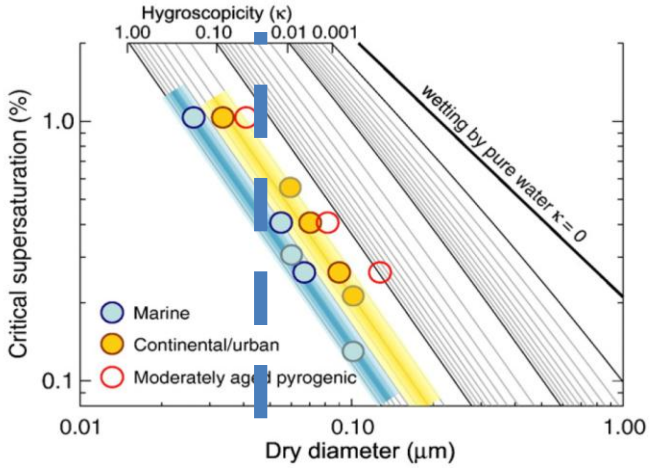

Fig. R4: Average relationships between aerosol dry diameter and critical supersaturation. The lines of constant hygroscopicity, κ, are from Petters and Kreidenweis (2007). This figure is adopted from Andreae and Rosenfeld (2008, Fig. 5).

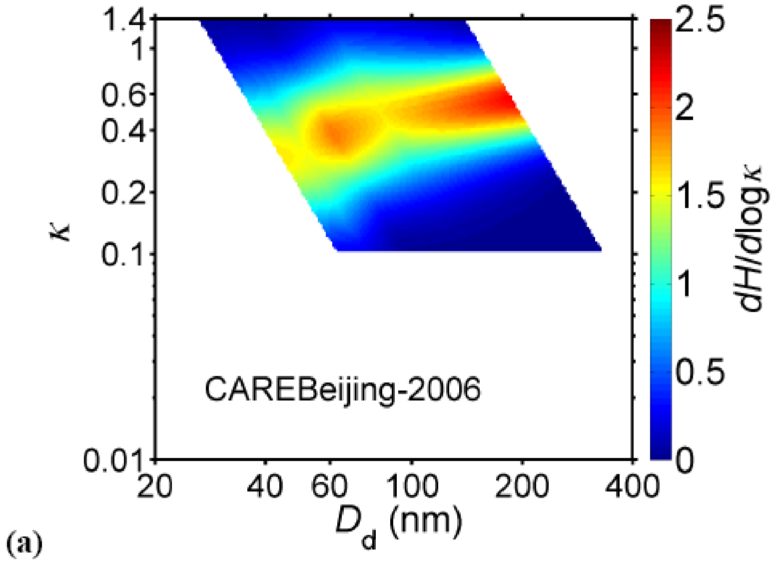

Fig. R5: Average logarithmic probability distributions of particle hygroscopicity, dH(κ,Dd)/dlog κ, plotted over effective hygroscopicity parameter (κ) and dry particle diameter (D_d) for CAREBeijing-2006 (a). This figure is taken from Su et al. (2010, Fig. 9).

Referee comment:

Page 10: again, even though it has been previously published in Rose et al, 2010, and Gunthe et al, 2009, I don't agree with the statement: "kt is better suited for the calculation of CCN number concentrations..." this statement assumes that MAF is not known. If the CCN activation spectra are obtained, then there is no reason why MAF would not be known, or why the 2-parameter fit would ever be used. Even if CCN activation spectra are not obtained, mixing state can be derived from single particle composition measurements or humidified tandem DMA measurements.

Authors' response:

When talking about a single parameter for the prediction of the CCN number concentration, κ_t is better suited than κ_a. The emphasis on a single parameter will be clarified in the revised version of the manuscript (p. 9968, l. 20): "… for single-parameter prediction of CCN number concentrations …" If we allow for two parameters, the combination of κ_a and MAF is indeed also well suited for CCN prediction and effectively equivalent to κ_t as demonstrated by Rose et al. (2011, Sect. 3.2.2). As discussed above, however, MAF is only operationally defined and depends on the investigated particle diameter range and supersaturation level. It is not clear if a campaign average value of MAF averaged over all investigated supersaturation levels would be meaningful and yield reasonable results in CCN prediction. This question goes beyond the present study but will be investigated in follow up studies. For campaign average values of κ_t, however, we have already shown in several earlier studies that it enables efficient prediction/approximation of CCN concentrations. Thus, we maintain our (clarified) statement. Beyond that we agree that mixing state information can be derived from single particle composition measurements or humidified tandem DMA measurements, and that such information is very useful for full elucidation of aerosol hygroscopicity distributions (Su et al. 2010). However, such measurements are not always available, and additional mixing state information does not invalidate the well documented fact that CCN predictions using κ_t alone work better than CCN predictions using κ_a without MAF.

Referee comment:
Page 13: Is it possible that the externally-mixed CCN-inactive particles at 250 nm are dust and not primary soot particles? From studies in other locations (e.g. Mexico City, Houston) I would expect the mean of the primary soot number mode to be much smaller (e.g. dp = _60nm mobility diameter). Also, I expect that there can be a significant amount of dust in Beijing. Dust can also be an absorbing aerosol, which might register as ECa? I understand that you will focus on the "abundance, properties and effects of the externally mixed, weakly CCN-active particles" in later studies, but I would like a short discussion on this here, since you say that these large (> 250 nm) particles are likely soot, but don't rule out the possibility (or even mention) that they could be dust (which seems more likely to me). If you believe that they are not likely dust, then I'd like you to give a short explanation why you think this.
Authors' response:
We understand the referee's concern, and we will address the issue in the revised manuscript. Nevertheless, we think that soot particles are much more likely than dust to account for the externally mixed weakly CCN-active particles observed during our measurement campaign. Dust particles are usually most concentrated in the coarse mode (> 1µm) of the particle number size distribution (e.g., Wu et al., ACP, 2009). They are not likely to represent a significant fraction in the size range we investigated in our study (< 300 nm), except for dust storm episodes. Remote sensing data, however, do not show any evidence for dust storm episode during our measurement campaign. We will add this information in the supplementary material of the revised manuscript.

Referee comment:
In general, more topic sentences would be nice. Especially for the paragraph introducing Fig. 2c on Page 14. This is an important paragraph - it would be nice to know that up-front.
Authors' response:
We are not sure which kind of topic/introductory sentences the referee would like to see at this or other positions in the manuscript. The main text of the manuscript follows the numbering and appearance of the figures, and we try to emphasize the most important messages at the end of each subsection as well as in the abstract and conclusions.

Referee comment:
I wish you had used different variable names, because I have to keep referring back to page 8 for their definitions. They are not obvious. I think it would be a lot easier to keep track if the following names were used:
....
3 parameter fit:

MAF (instead of MAF_f, what does the subscript f stand for anyway? fit? it seems like the same subscript should be used as the other fit parameters, or no subscript at all)
Da_3 (instead of Da_a)
sigma_3 (instead of sigma_a)
....
2 parameter fit:
Da_2 (instead of Da_t)
sigma_2 (intsead of sigma_t)

Authors' response:

We understand the referee's concern, but we are not sure if the suggested symbols would indeed be more self-explanatory. For compatibility with our previous studies, we prefer not to introduce new symbols.

Referee comment:

I don't really understand why MAFm is useful. Since MAFf is fit to the activation spectrum, then it should correspond fairly closely to MAFm. Why does the variability of this one point (the activated fraction at Dmax) matter more than for the other points? you could have simply used MAFf = MAF, since MAF for the 2-parameter fit is always 1, and since MAFm isn't really useful. Also, some of the variables (e.g. NCN, tot and NCN,30) are defined in table captions, when they should be defined in the text. they should also be defined before they are used (Table 2, where NCN,30 is defined, is introduced after it is used, for example). What does the subscript "p" stand for in "kappa_p"? why not use "kappa_AMS" to be more straightforward?

Authors' response:

The point mentioned by the referee is well taken. Depending upon the fitting practice to CCN spectrum there can be a slight difference in the maximum value generated by a fit and the maximum value measured as identified by the last point. Since we regard it good scientific practice to report both fitting values and directly measured values, we have chosen to mention MAF_f and MAF_m. However, as the referee has pointed out and mentioned in the manuscript the CCN efficiency measured at the largest diameter of each spectrum was generally in good agreement with MAF_f as derived from the 3-parameter fit. To improve the distinction among the meaning of the different variables, we will add a table in the revised manuscript listing the short description of all CCN parameters and other symbols used in the present study. Regarding the subscript "p" for predicted parameter values, we prefer to remain compatible with earlier studies. Moreover, we point out that different approaches of prediction may include more than just AMS data (see Rose et al., 2011).

Referee comment:

Page 17: "...the mass concentrations determined by AMS were enhanced by factors of 1.5-2..." although I know what you mean, you should say explicitly, enhanced relative to what?

Authors' response:
Thanks for pointing that out. We have modified the sentence accordingly in the revised version.

Referee comment:
Page 19: "An apparent overprediction of _a in the Aitken size range during the focus period of fresh city pollution (< 100 nm, Fig. 6c) is likely due to enhanced mass fractions of soot and elemental carbon (Tab. 3), which are not captured by the AMS data." You need to rethink this statement. For kappa_p to overestimate kappa_a, a compound must be measured by the AMS, but not actually contribute soluble material. Presumably, soot is not measured by the AMS. Instead, perhaps some of the organic compounds measured by the AMS during this focus period are not actually soluble? It seems reasonable to suggest that kappa_org is lower (< 0.1) for these fresh organics than on average. In that case, not only the fraction of organics matters, but it can also matter (to a lesser extent) whether the organics are fresh or aged...
Authors' response:
Thank you for pointing this out. We will change the sentence into "… due to enhanced mass fractions of internally mixed soot and elemental carbon, which are not captured by the AMS data." Moreover we will add a statement that changes in κ_{org} may also play a role.

Referee comment:
Page 21: "Thus, the CCN concentrations were generally larger (_(0.8-13)×103 cm-3 vs. _(0.4-8)×103 cm-3), although the total CN number concentrations were smaller (_1.8×104 cm-3 vs. _2.2×104 cm-3)." you should generally avoid words like "larger" and "smaller" when talking about particle concentrations (and instead used "greater" and "lesser", "higher" and "lower", or "more" and "fewer") to avoid confusion with statements about particle size.
Authors' response:
Thanks for the suggestion; we will change the sentences accordingly.

Referee comment:
Page 22: again I don't believe this statement is correct: "An apparent overprediction of _a in the Aitken size range during the focus period of fresh city pollution was likely due to enhanced mass fractions of soot and elemental carbon, which are not captured by the AMS"
Authors' response:
Please see the response to the above comment.

Referee comment:
Page 23: "...suggests that the fresh outflow from Chinese megacity centers may generally contain..." remove the word "may" or "suggests". both words together in the same sentence (along with "generally") makes it sound like you are not confident in your results.
Authors' response:

Thanks for pointing that out; we will change it accordingly in the revised version of the manuscript.

Referee comment:
Figure 2: It seems that your fits of the CCN activation spectra fit better to the smaller particles than to particle sizes just above the inflection point. Can you explain this? Does it have to do with the variable composition (or variable MAF) as a function of size? or multiply charged particles? What does it mean?
Authors' response:
As mentioned above it is related to the fitting procedure and technique. We do not think that it is related to the multiply charge particles. However, we will address these issues in more detail in follow up studies where we will show the effect of multiply charge correction and transfer function correction on the fits and derivation of the activation diameter and other parameters. Nevertheless, our preliminary analysis shows that the difference in activation diameter and other related parameters derived from the CDF fit are <5%.

Referee comment:
Figure 3 has a lot of information in it - it's very difficult to convey that much information in one figure. It would be nice to see a diurnal plot (data as a function of time of day, instead of one long timeseries) or a series of such plots that are colored by whatever variable you are interested in (e.g. temperature, organic mass fraction, humidity or wind direction). This would more directly illustrate the correlation(s) that you want to show. If you do decide to keep Figure 3, you should at least label all of the plots (a through i), in correspondence with the caption. Please discuss why the relative humidity (presumably after the drier) is so high (> 50% at times) during the green shaded periods. were you sampling in fog during those times? on page 16 you said there was a "frontal passage" during this period. If you were sampling in cloud, perhaps you were sampling interstitial aerosol, which appears to be consistent with your observations (e.g. lower CCN, lower kappa, lower MAF...)
Authors' response:
We thank the referee for the suggestion and accordingly we will label the panels (a to i) in Fig. 3. The diurnal variation are planned to be discussed in more detail in a follow up study. Please note that the relative humidity shown in Fig. 3 is the ambient relative humidity measured close to the inlet on the roof top and not the relative humidity of the aerosol in the sampling line. To avoid confusion we will add the word "ambient" in front of "relative humidity" in the figure caption.

Referee comment:
Figure 5: Why are there so few data points at S = 0.86%? It might be nice to show the different focus periods on this same plot (using different marker symbols?)
Authors' response:

As mentioned in the manuscript the mass fraction used in Fig. 5 were derived by integrating the AMS mass over the size interval of D_a-σ_a to D_a+σ_a. Furthermore the data points for which the mass concentration in the integration interval was less than 1 µg m^{-3} were excluded. For S=0.86% the average D_a and σ_a are ~45 nm and ~5 nm respectively implying the average integration range of 40 to 50 nm. Hence, within such a small interval of integration there were lots of data points with AMS mass <1 µg m^{-3}, which were to be excluded leaving very few data points for the correlation at S=0.86%.
As suggested by the referee we will separate the different focus periods in the plots using different symbols.

Referee comment:
Figure 6: This figure is very nice. It would have been nice to see how the externally mixed fraction varies as a function of particle size too. but I don't think you are capable of determining this from your dataset. However, if I am wrong about that, please include it here. you could at least show a lower limit for the externally-mixed raction...
Authors' response:
We thank the referee for the suggestion. We could include 1-MAF$_f$ as an indicator of the externally mixed fraction as shown by Rose et al., 2011. However, during the CAREBeijing campaign MAF was significantly <1 only at lowest S. We are planning to address these and related issues in a follow up study.

Referee comment:
In generaly, you should order citations by date (oldest first, to give credit to the person originally doing the work).
Authors' response:
We thank the referee for pointing that out and we will correctly reorder the references in the revised manuscript.

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