We thank the reviewer for the detailed and thorough comments. We implemented a lot of the suggestions (especially the kappa-representation throughout the manuscript) and now discuss all possible explanations of the observed results in more detail. However we kept the derivation of kappa (linear interpolation of CCN spectra in kappa space) as it was, for reasons detailed below.

In light of these big revisions, many changes have been made. The theory section has been rewritten. All Figures and Tables and all sensitivity studies have been updated and partially redone at RH=85%. Please note that the extra data in Figure 5 for 150 nm particles have been omitted. In the original version there was an error in the symbols corresponding to different fuel types. This has been corrected.

Page 29859, line 16 (and the whole manuscript)

Answer: We now use the kappa representation throughout the manuscript. This resolves the issue

Section 3.3: You define your "significant" supersaturation from the CCN activation curve where 50% of the particles are activated. In order to be sure that this method is correct you have to be sure, that you got successfully rid of any multiple charged particles and you don't have any background counts in the CCNC, so that your activation curve starts from 0. And also that at full activation the CCN/CN ratio is 1 (which is often not the case because of the independent measurements of the CN and CCN concentrations and/or not sufficient counting statistics at low concentrations). Did you measure both plateaus for the activation curves (for the example scan which is shown in Figure 2, neither the full activation nor the non-activation is captured)?

Answer: The full activation plateaus were to a good approximation at 1 (at least within uncertainties) for larger particles of 150 and 200 nm. The 200 nm particles were fully activated at most supersaturations and therefore not used in this study, but served as a good measure of the plateaus.

With the resolution of our spectra we cannot prove that our correction method eliminated all the doubly charged particles, but due to the low resolution we also have no way of estimating a possible amount of doubly charged particles left. The 50nm particles really reach zero at the lowest 3 supersaturations measured, but at this size the doubly charged particles are low, so this is probably not a solid 'proof' of our correction method.

Using another approach to define a "significant" supersaturation helps to get rid of this problem: fitting a sigmoid or cumulative Gaussian function on the data, where the base and maximum values are allowed to vary, is a solution. Of course it is only possible if you have sufficient measurement points. Which can be critical if measurements were performed only at 5 different supersaturations.

Answer: Indeed, a well-constrained fit of a sigmoid function is not possible for our data, especially not for the 50 and 150 nm data. Therefore we have to resort to an interpolation method.

The difference between the derived supersaturations using the two methods (CCN/CN vs. supersaturation or epsilon) originates from the low resolution of your measurements as well (as you have mentioned). Searching for the 50% activation point in the different spaces (CCN/CN vs. S or CCN/CN vs. epsilon space) is equivalent to different methods how you connect the two closest measurement points that surround the 50% activation: assuming a linear relationship (linear interpolation in the CCN/CN vs. S space) or a specific non-linear relationship (the epsilon vs. S relationship multiplied with a linear relationship which is equivalent to the linear interpolation in the CCN/CN vs. epsilon space). The observed difference between epsilon_CCN and epsilon_G is simply due to the different interpolation. With higher supersaturation resolution this difference would decrease, and in ideal case with arbitrary high resolution it would even disappear. So actually this is a measurement error caused by your low resolution which is independent from the uncertainty of the single measurement points. Therefore the two errors has to be summed to get the true error, and the "interpolation" error cannot be neglected just because it is smaller than the error originating from the uncertainty of the individual measurement points.

However, probably it is still better to use S_c^m as "significant" supersaturation (linear interpolation in the CCN/CN vs. S space) and not the roughly 20% higher value. If the aerosol is not highly externally mixed (which is suggested by figure 1) then the CCN/CN spectrum is roughly anti-symmetrical around the 50% activation point. So a linear interpolation between the measurement points around the 50% activation is a better idea.

Answer: It is true that the problem arises due to poor resolution of our CCN spectra. This poor resolution results from the fact that a static thermal diffusion chamber was used for the CCN measurements, which are not very fast. In order to assure stability of the aerosol during one CCN activation curve, only few data points could be measured. However, it is still possible to estimate which interpolation method is the best approximation.

Generally, the point can be made that due to the roughly linear relationship between growth factor and effective soluble volume fraction (and kappa) the growth factor measurements are made in 'epsilon space' and therefore CCN measurements should be evaluated in epsilon space as well. This can be substantiated by a simple calculation, illustrated in Figure 1, in the attached file. From the growth factor measurements it is clear that the effective soluble volume fraction (and kappa) in our experiments are roughly normally distributed. If we assume an aerosol with a nominal diameter of 100 nm and soluble volume fractions (model substance: levoglucosan) normally distributed around 0.35, we can estimate the cumulative kappa distribution (blue line, panel a). From this we can estimate a CCN activation

curve (blue line, panel b). The red line in panel b shows the linear interpolation between the supersaturations at which the CCN activation was measured in our experiments. The red line in panel (a) shows the interpolation in epsilon space. (Note that for conversion to kappa you would have to multiply the x-axis in panel (a) with 0.2). The red line in panel b shows the interpolation in S space.

It is clear that in the case of a normal distribution of soluble volume fractions, the linear interpolation method in kappa space is superior. Therefore we choose this method and treat it as a systematic correction, instead of an additional uncertainty. This is made clearer now in the revised manuscript. We realize that deriving kappa from the 50% activation point is the more common practice and it is in fact very useful and should be the method of choice to parameterize CCN spectra. However in this work, we have to derive the kappa value that is most comparable to the HTDMA measurements in order not to introduce errors, which leads to the use of a different method.
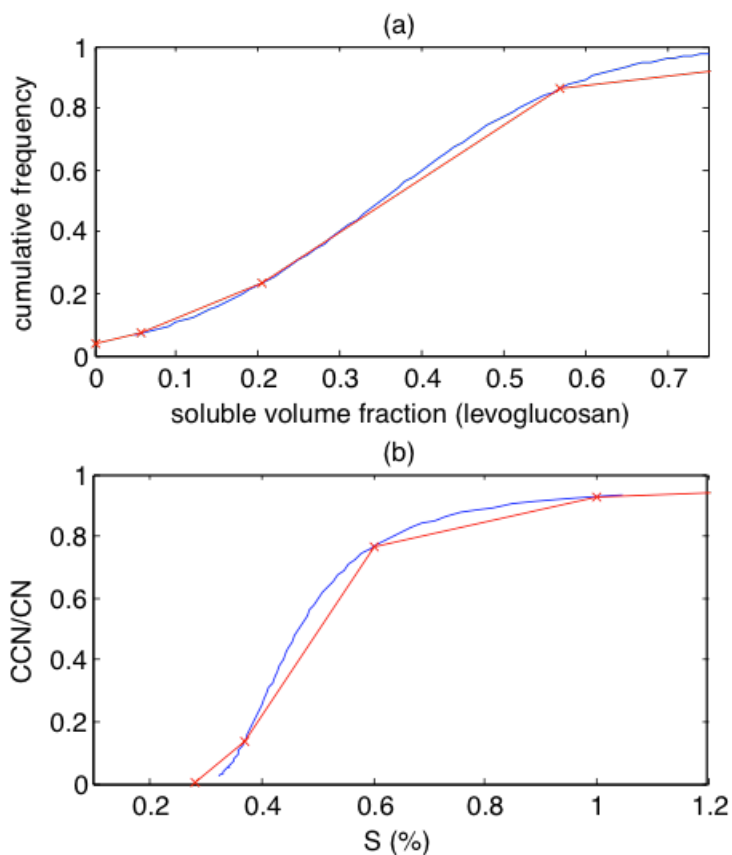
One should also note that this example only applies for a roughly symmetrical distribution of soluble volumes. If the distribution is clearly skewed (e.g. lognormal), then no matter how precise the measurement and how good the interpolation, it will always be inappropriate to compare the 50% CCN activation point to the mode of the growth factor distribution.

Sections 5 and 6: During the hygroscopicity closure mainly the mean soluble fractions from the HTDMA and CCNC are compared. However, for some individual experiments, "behavior", which is clearly different from the average "behavior" can be observed (e.g. the experiment where the 150nm particles seem to have at least a factor 2 higher epsilon_G than epsilon_CCN). I would suggest to discuss those extreme cases as well.

The error bars on the data represent 1 sigma uncertainty, one would therefore expect roughly 30% of that data to not include the 1:1 line within the respective error bars and about 5 % of the data to be outliers of more than 2 standard deviations. Some or all of the outliers could be therefore due to statistics. In the experiments in question, we could not find anything that would explain strong deviations. In the case of 150 nm the poor correlation (definitely more than 30% of that data points do not include the 1:1 line with the error bars) is already discussed in the text.

Page 29855, line 4-5: Did you really burn and measure peat and grass? You show results only on different wood types. It would be actually nice to add a summarizing table with all experiments and fuel types to the manuscript.

Yes, but this is part of a forthcoming paper about the EFEU experiment in general. We removed the reference to that in the abstract.

Page 29856, line 1-2: The insoluble part plays a role as well, since the absolute amount of insoluble and soluble material determines the particle's size, which plays a big role in hygroscopic growth and CCN activation.

Done

Page 29856, line 19-20: This is a strong statement. Can you really objectively say that?

From the overview of the relevant literature, I believe this to be the case. Although I weakened the statement slightly, since I did not do an exact meta analysis on this.

Page 29858, line 10-12: If I am correct, for the multiple charge correction the size distribution data is needed. Did you have this information? Then please include the description of the instrument in the experimental part as well.

Done

Page 29858, line 2-12: Please report on the length of one CCNC scan. How stable was the sampled aerosol during this period? How many CCN scans could you do during the 1 hour burns? Please answer the same question for the HTDMA measurements as well.

Done

Page 29858, line 21-29: At which RH was the HTDMA measuring?

This information is included in the revised manuscript

Page 29860, equation 3: This is not the definition for the ideal solutions, for ideal solutions i=1, this is a general equation for non-ideal solutions as well.

This is true, but not applicable in the revised manuscript, the same goes for the next 2 comments

Page 29863, line 25 to page 29864, line 1: The RH measurement has an uncertainty of +-2%. This causes roughly +-3.5% uncertainty in the GF for sodium chloride measurements. You argue that since the measured biomass burning aerosol is much less hygroscopic, the uncertainty originating from the not precise RH measurement will be less than +-2%. From my point of view this argument is not correct. The propagation of the relative error on the RH is solely dependent on the first derivative of the GF(RH) function and has nothing to do with the GF(RH) value itself. So it is not a correct argument that the biomass burning aerosol is much less hygroscopic. If you have information on the first derivative of the GF(RH) function at the RH where the measurements were done (85%?) for both sodium chloride and the biomass burning aerosol then you can determine the propagated error on the GF. Otherwise, if the needed data is not available, I suggest to calculate epsilon_G for your individual measurement points assuming 2% lower and 2% higher RH as well to get the correct uncertainty of epsilon.

We do not have any information about the first derivative of the Gf(RH) function. Unfortunately, we also don't have precise information on Gf at 2% higher and lower RH. We therefore include a rough theoretical estimate of the uncertainty assuming an idealized biomass burning particle (30% levoglucosan, 70% insoluble, ideal solution), as suggested by reviewer 2. Assuming a smooth activation curve, this should be a good approximation of the actual uncertainties. If there are any 'steps' due to deliquescence events or phase transitions, this would not hold. However, we don't have any data on this, so it is better to stick with a very rough Gf uncertainty of 1-2%.

Page 29864, line 14 and 15: the CCN/CN term in the formulas after the delta sign has to be in brackets.
Done

Page 29864, section 3.5: This section belongs more to the sensitivity studies, since as you show, the non-sphericity of the particles can lead to significant differences between epsilon_G and epsilon_CCN. I would move this section under section 4.

Done

Page 29866, line 8: Change "We can deduce from his example" to "We can deduce from

this example"
done

Page 29867, line 1-2: How "roughly" can levoglucosan be approximated to be an ideal solution? Is the non-ideality of levoglucosan neglectable when one compares it to the non-ideality of ammonium sulfate?

This was not very well formulated in the original manuscript, since we did not want to approximate a 'real' levoglucosan particle, but actually need a particle with ideal behavior in order to isolate the desired effects. In the revised manuscript this is reformulated to:

"For the purpose of these sensitivity studies we consider an idealized biomass burning particle consisting of 70% insoluble material and 30% levoglucosan that behaves as an ideal solution with a van't Hoff factor of 1. This means we ignore the weak solution nonidealities (Mochida and Kawamura, 2004; Svenningsson et al., 2006) and the weak effect of levoglucosan on surface tension at high solute concentrations (e.g., Svenningsson et al., 2006). In this way we can study the desired effect without interference from non-ideal solution effects. "

Page 29867, equations 11 and 12: Repetition of the equations. Could be easily referenced if a good theory section existed.

Not applicable in the revised manuscript

Page 29867, line 12: Why did you solve it for RH=90%? It was not clearly stated but it seems to me that the GF measurements were performed at 85%. Please do the calculations at the RH where the measurements were done.

In this case the 90% RH was a typo, but we rechecked everything and made sure that all the calculations are done at 85% RH

Page 29867, line 18-21: Here, you only test the effect of the non-ideality of your model salt. The effect of the non-ideality of the biomass burning aerosol has to be added to this. Can you be sure, that that is not much higher than for ammonium sulfate (e.g. if there are, give references where biomass burning aerosol seems to behave ideally, or close to that)?

The discussion if the effects of non-ideality has changed due to switching to the kappa formulation. Now we cite Petters et al., 2007 for this information.

Please emphasize here as well, that epsilon_G and epsilon_CCN are calculated using the surface tension of pure water

Done

When you used levoglucosan as a model substance, did you assume again its ideal behavior?

Yes. It is now mentioned with equation 6 that ideal solutions are assumed for all sensitivity studies.

Page 29868, line 17: "Epsilon_G (or kappa_G) is not influenced by a change in surface tension" Please reformulate this sentence, they are influenced but only slightly.

Done

Page 29868, line 17-23: Please state at which RH did you do the calculations

Done

Page 29868, line 24-27: Why is it a problem if the hygroscopicity parameter exceeds 1? It simply means that you have something, which is more hygroscopic than pure levoglucosan, or has lower surface tension. If you prefer ammonium sulfate as a model substance, just use it (it is your free choice, section 4.1 shows that ammonium sulfate is a usable model salt).

Does not apply to the revised manuscript

Page 29869, line 7: at which temperature does succinic acid has this solubility?

At 25°C

Page 29869, 2nd paragraph: Please mention already before the equations that succinic acid is treated as an ideal solution.

Done

Page 29869, line 17-18: if the solubility is used in g/l, then the density of water has to be also in g/l in equation 13. Please confirm that you used the correct units.

Yes. Also we omitted the units in the revised manuscript to avoid confusion.

Page 29870, line 4: Again, why 90%?
Changed to 85%

Page 29870, line 18: Was something different during those pre-experiments?

Does not apply to the revised manuscript. I realized it was somewhat awkward to add extra data for only one diameter, so I omitted them in the revised version.

Page 29870, line 27: Janhäll et al., 2010 is not in the reference list! Do you mean this paper: Biomass burning aerosol emissions from vegetation fires: particle number and mass emission factors and size distributions in ACP? In that paper I did not find anything about the hygroscopicity.

As stated in the text, this is a manuscript in preparation

Page 29871, 10-18: Can you give an estimate to what extent should the slightly soluble material be enriched?

Now explained in the text. To explain the increase in kappa_CCN, the soluble fraction must increase by 50%. Assuming that since the kappa values agree at 100 nm, there are only small amounts of slightly soluble material present, the fraction of slightly soluble organics would have to increase from roughly 0% to roughly 30% of all water soluble organics.

Page 29871, 18-25: I don't see, why the lowered surface tension is the most likely explanation. You have shown in section 3.5 that a relative small deviation from the sphericity could cause the observed phenomenon as well. Why is this less likely?
Smaller parcticles -> less condensed organics and therefore higher soot fraction -> less spherical particles. For me this explanation seems as reasonable as the lowered surface tension.

This is now discussed in the text in more detail. The problem is that not only the difference in kappa at 50 nm, but also the increase in kappa_CCN with decreasing particle size must be explained. For both effects combined, the surface tension effect seems most likely, otherwise a combination of different effects have to be invoked that accidentally cancel out at large particle sizes, which is not very satisfactory.

Page 29871, 18-25: And could you also give here an estimate what surface tension would be needed to explain the observed discrepancy between the GF and CCN measurements.

This is done in Table 6 (revised manuscript)

Page 29872, 22-26: As you state: both effects reduce epsilon_G compared to epsilon_ CCN. But the epsilon_G values are symmetrically distributed around the mean epsilon_CCN values. So if this was the real reason for the scatter, something else should be responsible for the higher epsilon_G average. What could that be?

This is indeed a problem, and now mentioned in the text. It actually seems reasonable that the particles were pretty spherical, given that Iinuma et al., estimate

the fraction of elemental carbon in this size range to only be roughly 30%. It might also be that we underestimate the uncertainty in growth factor measurements, since we only estimate the shape of the activation curve using an ideal solution.

Table 2: Last column should read epsilon_G. Please mention in the caption that you used the levoglucosan as model substance as well. The growth factor has a symbol of G here, in the other tables it is GF. Please be consistent.

Not applicable in the revised manuscript

All the other comments on Figures and Tables have been implemented.