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 and the study of shape effects has been moved to section 4. All Figures and Tables have been updated. Also 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 for 150 nm particles. This has been corrected.

Answers to the comments:

Abstract, lines 22ff: Does this last sentence relate to the 50nm particles, or to biomass burning particles in general? If it is a general value, how does k=0.15-0.2 compare to k=0.07 you give in line 10 in the abstract?

K=0.07 is for the whole biomass burning particles, including insoluble fraction, whereas k = 0.2 is derived for the water soluble organic fraction. This is now explained better in the abstract.

page 29856, lines 27-28: I suggest you explain this in the light of the deliquescence process (i.e. dilution at the RH where particles just form a saturated solution).

This comment is not completely clear to me, since slightly soluble substances mixed with hygroscopic substances do not really deliquesce but gradually dissolve over a wide range of RH. I think I did not sufficiently understand this comment. However, I tried to extend the discussion round this topic.

page 29857, lines 1-3: The effect you mention (i.e. that SA substances barely affect water uptake at subsaturated conditions, but can have a large effect on the critical supersaturation) is shown in detail in a paper you've cited earlier, anyway (Wex et al. 2008), so you should mention that here, too.

Done

page 29857, line 13: Also Chang et al. (2007) did not achieve closure between N(CCN) as measured directly with those derived from AMS measurements during times when the organic content of the aerosol was high. They also relate this to solubility or surface tension effects. Please add this citation here.

Done

page 29858, line 4: Is this the CCN counter as described in Roberts and Nenes (2005)? If yes, please say so.

No, it is a static thermal diffusion chamber, described in detail in the papers cited in the text.

page 29859, line 2: The sentence needs to be reformulated; it sounds strange that the discrepancies should be based on model calculations.

Done

page 29862, lines 1-2: Please, for clarity, add to the text in the parenthesis, that you, however, omit a concentration dependent behavior of sigma.

Not applicable to the revised manuscript

page 29862, lines 14ff: I found it confusing to have this paragraph first, and then, in the next paragraph, being introduced to the example of very smooth humidified size distributions with only one mode (in Figure 1). Reverse the order of these two paragraphs, and make it clear if the problems you describe for the biomass burning aerosol relate to the broadness of the measured peaks or to multiple peaks that you observed, or to both.

We strongly prefer to keep the order of the paragraphs, but we reformulated then to make them less confusing.

page 29862, line 21: The curves you show in Figure 1 are very smooth – were they really measured and then smoothed, or are these theoretical values? Please explain in the text.

They are measured and smoothed; this is now explained in the text.

page 29863, line 9ff: You introduce an artificial error to your data by your choice of using epsilon(CCN) instead of epsilon(S). In the past, this kind of data (as in Figure 2a) has been evaluated by fitting an error-function to the data (e.g. Rose et al. (2008)). This would have been better instead of using an interpolation to get Sm, as you did. It gets worse when you derive epsilon for all your measured data and interpolate then. The deviation between epsilon(CCN) and epsilon(Sm) will depend strongly on the number of measurement points you have in the interval around CCN/CN=0.5. You can still show the error that is introduced by the second method you showed, but for the evaluation of your own data you use later on, I suggest following what has been established as a feasible way of data evaluation, i.e. the error function fit, or at least use epsilon(Sm).

As the reviewer notes, the problem arises mainly 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 is not very fast. In order to assure stability of the aerosol during one CCN activation curve, only few data points could be measured. However, even though there are only few data points available 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 or kappa space' and therefore CCN measurements should be evaluated in epsilon or kappa 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 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 salt: levoglucosan) normally distributed around 0.35, we can estimate the cumulative epsilon distribution (blue line, panel a). From this we can estimate a 'true' 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.
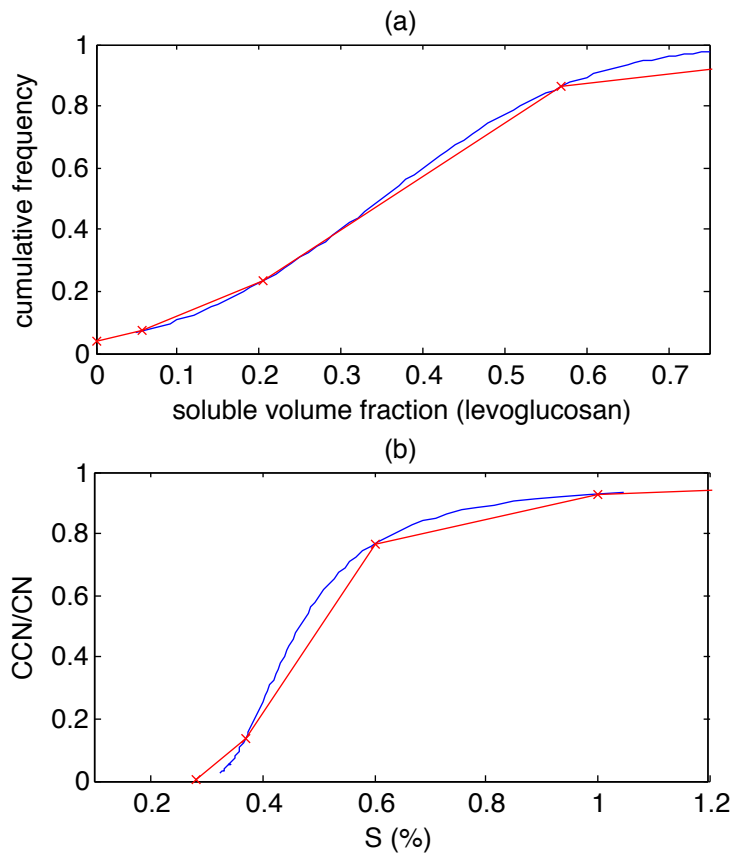

Figure (1)

It is clear that in the case of a normal distribution of soluble volume fractions, the linear interpolation method in kappa space is superior. Since we are forced to interpolate, 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. 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 lead us to using 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 significantly 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, since the mode, median and mean of the kappa distribution are not the same any more.

page 29863, line 23: Please add to the text, if you used a shape factor for NaCl, and if yes, which one?

Sodium chloride particles were atomized during the experiment on regular intervals at the same RH, where laboratory measurements of the fuel samples were carried out (RH= 85%). The growth of the sodium chloride particles from this study was compared to a former laboratory measurement, where the hygroscopic growth of sodium chloride was derived under extremely stable conditions with the same H-TDMA system verifying that measurements were carried out within +/- 1.5 % RH during the acquisition of this "calibration curve". No shape factor was applied to the calibration data from this study and to the former acquired "calibration curve". The comparison revealed that the RH during the measurements of this study was within 85 +/- 2%. Nowadays we use ammonium sulfate calibration measurements and compare them to theory, which might give a little bit more exact results.

page 29864, line 1ff: Where do the <2% come from? Do you simply relate the 2% uncertainty in RH to 2% uncertainty in the growth factor? This cannot be done so easily, because at lower RH (i.e. where growth factor curves are less steep) an uncertainty in RH translates into a much smaller uncertainty in the growth factor than at higher RH (where the growth factor curve is steep). Anyway: This whole paragraph goes back and forth a bit. I suggest you include a Figure showing the theoretical growth factor curve for an aerosol with a GF of 1.1 at 85% RH (and maybe a further one for a GF of 1.4) (this corresponds to kappa of about 0.06 to 0.09). From this curve clearly will follow the uncertainty in the growth factors for the 2% uncertainty in RH. This saves you a lot of guessing and you can write this paragraph more straight forward.

We did not include a Figure, but otherwise followed the suggestion to calculate the uncertainty of the growth factor theoretically for a representative particle with a representative kappa. This is of course very uncertain, since we do not know the shape of the activation curve of the actual biomass burning particles and assume an ideal particle. Therefore we refrained from putting in a Figure, because this could lead to confusion and give the impression that we actually know shape of the activation curve.

page 29865, lines 1-2: What do you mean by "The critical supersaturations : : : do not depend on the equivalent particle diameter"? You measure a pair, consisting of "supersaturation" vs. "critical dry diameter", so there is a dependency. I guess I do not

understand what you want to say -> please clarify.

This is reformulated in the revised manuscript. The sentence reads now: "The critical supersaturations derived from CCN measurements are an intrinsic property of the particles studied. However, the dry particle diameter is subsequently ... "

page 29865, lines 6-7: This is only true if the dry particle is non-spherical – please clarify in the text!

Done

page 29866, line 4: Please mention that Eqs. 11-12 will follow in 4.1.

Does not apply to the revised manuscript

page 29866, lines 4-5: Add / replace text, starting at the beginning of the sentence in the middle of line 4: "To derive epsilon(g) we then use the calculated d(wet) and d(m) of 100nm, as it were done to evaluated the data if the shape factor were not known. [The measured hygroscopic growth factor : : : strongly with increasing X], i.e. the derived epsilon(g) will be too small, and increasingly so with increasing shape factor. Also, the critical supersaturations were determined as described in 4.1. [Since the : : :]"

Done with a slight change in formulation

page 29867, lines 1ff: As you say, water uptake of levoglucosan can be approximated by that of an ideal solution with a van't Hof factor of 1. This touches one of my main concerns with your way of evaluating the data: by the use of ammonium sulfate as the proxy, from which you derive epsilon, you will get an artificial error for any substance that has a different concentration dependency in its non-ideal behavior than ammonium sulfate. If one wants to be able to disentangle these kinds of effects at all, it would be of advantage to first assume as little as needed when deriving epsilon (or kappa, for that means), and go on from there. You show correctly that assuming a non-ideal behavior that might not be there introduces an error. If you examined a substance that has a non-ideal behavior different than that of ammonium sulfate, these two different non-idealities start to be entangled so much that information is really lost. Therefore, as mentioned above, it might be advantageous to use kappa throughout your work. If, however, you decide to continue using epsilon, please add to the text how large the error introduced here is, compared to the errors introduced by the shape factor, SA and SS.
Kappa is now used throughout the manuscript

page 29868, 4.2: Start the chapter with a description of how you obtained the values in Table 3, e.g.: Values that would be measured for Sc if the surface tension was reduced were calculated, and from these values epsilon was derived while using the surface tension of water

done

page 29868, lines 5-6: Please explain how you know that the increase in epsilon(CCN) is independent of particle diameter.

It is now mentioned that this can be seen from Table 2 (new manuscript)

page 29868, lines 10-11: It is exactly this sentence that made me suggest do change your way of data evaluation, because this is absolutely correct!

Done

page 29868, lines 24ff: Table 3 does not show that levoglucosan is not the model substance of choice. The surface tension reduction you assume here is pretty large, already, compared to what is generally observed (a lowering of 30% might be as low as it was observed for atmospherically relevant substances). So you do not really observe that epsilon exceeds 1. Also, if epsilon exceeds 1, this only means that the hygroscopicity of the particle is larger than that of levoglucosan. These large epsilon values >1 could, however, still be used. (Similarly it could be argued that the water uptake of e.g. NaCl particles cannot be described by using epsilon and ammonium sulfate as a proxy, and with this discard also ammonium sulfate as a suitable proxy.)

Does not apply to the new version of the manuscript

page 29870, lines 5ff: You showed earlier, that at least the shape effect can introduce a similar error, and the use of ammonium sulfate as a proxy acts similar. These effects cannot be disentangled from the effect of SS, therefore your conclusion here is too strong.

It is now mentioned in the manuscript that this would only be the case, if the other effects were not present.

page 29872, line 1: Why do you show these additional data, if they were done for different burning conditions? Show at least which of your data originated from that earlier campaign, e.g. through the use of different symbols.

They are not shown anymore.

Figure 3: Add error bars to the epsilon values.

Done.