

Interactive comment on “Measured and modelled cloud condensation nuclei concentration at the high alpine site Jungfraujoch” by Z. Jurányi et al.

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We would like to thank Anonymous Referee #2 for his/her valuable comments, it helped us improving our manuscript and making it more understandable.

Comment:

Technically, and grammatically, the paper is very good. I do have some general concerns with regards to inconsistencies with other publications. There are multiple sections within the text used to infer the role of surface tension. It is clear from the measurements, assuming the correct kappa values are derived, that you can assume the value of that of pure water. However on page 8863 you make the statement ‘Lance et al. ... 0.015N/m lower and concluded CCN predictions became much worse. The

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same is true for most hygroscopicity-CCN closure studies’. Is this true? This goes against the findings and conclusions presented in the following papers, to name but a few: ...

Response:

It is true that several laboratory studies on organic aerosols indicate surface tension suppression effects. However, most ambient studies report overprediction of CCN concentration, which cannot be explained by lowered surface tension. These facts have been acknowledged in the revised Sect. 4.5: “Several laboratory studies on organic aerosols indicate that surface tension suppression effects may occur (e.g. Shilling et al., 2007; King et al., 2009). In contrast the results of our closure study give no indication for a suppressed surface tension of the aerosol at the point of CCN activation. Aerosols with suppressed surface tension activate at lower SS_{crit} because the Kelvin effect term in Eq. (1) is decreased. Assuming a 10% reduction of surface tension at the point of CCN activation relative to pure water increases the predicted CCN concentration. Consequently the overprediction becomes even higher and reaches a factor of 1.12 averaged over the whole data set compared to a factor of 1.04 with assuming surface tension of pure water. Kammermann et al. (2010a) compiled an overview of the performance of existing hygroscopicity-CCN closure studies, revealing that the CCN concentration was underpredicted in only one out of 10 cases, always assuming surface tension of pure water. This is in line with our result that assuming reduced surface tension impairs the performance of CCN predictions for ambient aerosols. However, Good et al. (2010) also reported underprediction of CCN concentration, which might be an indication for suppressed surface tension in their case. One has to note that closure with assuming surface tension of pure water might be untruly achieved by compensating errors if both kappa hygroscopicity parameter and surface tension are in reality smaller than the values used in the calculations (see also Sect. 4.7). Furthermore, it is also not possible to distinguish between a true absence of surface tension depression and the presence of surface active species compensated by bulk/surface partitioning

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effects (Sorjamaa et al., 2004) such that one can still assume surface tension of pure water at the point of CCN activation.”

Comment:

There is also inconsistency between statements made in this paper and those made in a similar articles I have encountered in ACP, including one written by same authors which need to be addressed. I have read the following paper with much interest: ‘Widening the gap between measurement and modelling of secondary organic aerosol properties? N. Good, . . . Atmos. Chem. Phys., 10, 2577-2593, 2010. However, I found no reference to this paper within any portion of the current manuscript. However, from the abstract of said paper we have the following statement: ‘..the ability of the simpler single parameter model to predict cloud activation behaviour was dependent on the instrument used to measure sub-saturated hygroscopicity and the relative humidity used to provide the model input. ... The difference in HTDMA data from validated and extensively used instruments means that it cannot be stated with certainty the detail required to predict the CCN activity from sub-saturated hygroscopicity. In order to narrow the gap between measurements of hygroscopic growth and CCN activity the processes involved must be understood and the instrumentation extensively quality assured. It is impossible to say from the results presented here due to the differences in HTDMA data whether: i) Surface tension suppression occurs ii) Bulk to surface partitioning is important iii) The water activity coefficient changes significantly as a function of the solute concentration. Derivation of Kappa values has to come from the HTDMA during ambient campaigns, but the variability and concluding remarks published by Good et al (2010) is worrying. I would like the authors to comment on how results from that paper impact on this study. At least, this should be commented on in light of the sensitivity studies presented here. For example, would any propagated errors change the conclusions regarding closure with measured CCN concentrations if the averaged kappa values change? Again, I appreciate the need to investigate the applicability of these models in field campaigns using the excellent sensitivity studies you have conducted,

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but evaluation surely depends on instrumentation errors. If this is supposed to be implied within the kappa sensitivity section, then this should be stated explicitly within the main body of text, referring to the recent publications.

Response:

This is a valid objection raise by the referee. It is now discussed in the last paragraph of Sect. 4.7: “CCN predictions are complicated by several factors including the potential occurrence of surface tension suppression, bulk to surface partitioning effects of surface active species, or strong dependence of the water activity coefficient on the concentration of the solution droplet. Good et al. (2010b) reported that experimental differences between HTDMA instruments operated in parallel made it impossible to determine whether or not the above effects occur. Furthermore, CCN properties predicted from the hygroscopic growth measured at subsaturated RH were substantially different when using data from different HTDMA instruments. What does this mean for the interpretation of our closure results? Good et al. (2010b) investigated pure organic and organic dominated aerosols (organic fraction >80% for the most part) produced in a smogchamber by photo-oxidation of alpha-pinene. Duplissy et al. (2009) reported for this particular aerosol type discrepancies between different HTDMA instruments that compared well for inorganic aerosol during the same intercomparison study. Whether or not such discrepancies between different HTDMA instruments also occur for ambient aerosols has not yet been shown to our knowledge. The organic aerosol investigated Good et al. (2010b) has a rather low hygroscopicity ($\kappa \sim 0.1-0.15$), which makes predicted CCN properties much more sensitive to experimental uncertainties (see Fig. 8), compared to the aerosol at the Jungfraujoch. Furthermore, the study by Good et al. (2010) investigates the properties of CCN rather than CCN number concentration. Comparison of CCN properties emphasizes small discrepancies, whereas CCN number concentrations of a polydisperse aerosol are much less sensitive to uncertainties in kappa. This fact is illustrated by Figs. 4, 8 and 9 of this study: substantial differences in kappa translate only into small differences of corre-

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sponding CCN number concentrations. As a consequence it is not possible to attribute the differences kappa values reported here to experimental uncertainties or one of the above-mentioned effects with certainty, but we can constrain the kappa hygroscopicity parameter from either AMS/MAAP or HTDMA measurements well enough to predict the CCN number concentration within a few percent.”

Minor comments:

Comment:

In the abstract you make the statement ‘a sensitivity study showed that the temporal variability of the chemical composition at the Jungfraujoch can be neglected for a reliable CCN prediction’. Reading through the manuscript this seems to be simply because, as you state on page 8874, the ‘aerosol with a relatively constant chemical composition’ was studied. Thus, the chemical composition does not vary, and therefore can be ignored. Reading the abstract however it appears that there might be some variability but ignoring this makes no difference.

Response:

The statement in the abstract is correct as it is. The temporal variability of aerosol chemical composition at the JFJ is indeed slower and less pronounced than at sites near sources. Nevertheless substantial changes are also observed. More detailed statements about the variability of the chemical composition and its impact on CCN concentration are already made in the ACPD manuscript: “The volume fraction of the inorganic compounds ranged from 20% to 80% with the 10th percentile of 37%, median of 45% and 90th percentile of 66% during the one-month observation period. Even though substantial variations in chemical composition were observed, significant differences between the reference prediction and the time averaged kappa case cannot be seen. The model performance gets only slightly worse (larger chisquare values at most of the SS, not shown here) if the time variance of the chemical composition is ignored. Based on this analysis, the temporal variability of the chemical information could be

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skipped for the calculation, still yielding a reliable CCN prediction at the JFJ.”

Comment:

‘Our observations give no indication that the surface tension of the aerosol was suppressed’. This actually shows that the behaviour of material at the bulk/surface interface may be such that the surface tension of pure water can be used in this case. Or, it also may mean that any postulated bulk/surface partitioning, which arises from a suppressed surface tension effect, is such that one can assume the surface tension of water in this case. . .provided we can trust the HTDMA to infer these processes?

Response:

This has been clarified with the following paragraph added to subsection 4.5: “. . .Furthermore, it is also not possible to distinguish between true absence of surface tension depression and presence of surface active species compensated by bulk/surface partitioning effects (Sorjamaa et al., 2004) such that one can assume surface tension of pure water at the point of CCN activation.”

Comment:

Conclusions. Again, you make the statement ‘no substantial surface tension reduction occurred’. Again, I think this should be placed in context of the fact that this may indicate potential processes which we, apparently, cannot decipher using current instrumentation at relative humidities less than say 98%.

Response:

The statement in the conclusions reads now: “Significant underprediction was not experienced, indicating that one can assume surface tension of pure water at the point of CCN activation.”

Comment:

‘The HTDMA data can be used as a proxy for the chemical composition, instead of the

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AMS and MAAP'. I believe this statement is too broad if not taken within the appropriate context. Yes, the HTDMA can be used to infer mixing state, but to prescribe broad chemical proxies alone is not correct. The similar statement made in the abstract is less broad, suggesting that if this is the only option then it might be ok to use.

Response:

This has been clarified: "Here we show that HTDMA data can also be used to determine the kappa hygroscopicity parameter required for CCN prediction instead of the chemical composition data."

Interactive comment on Atmos. Chem. Phys. Discuss., 10, 8859, 2010.