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Atmospheric Chemistry and Physics Discussions

Interactive comment on "On aerosol hygroscopicity, cloud condensation nuclei (CCN) spectra and critical supersaturation measured at two remote islands of Korea between 2006 and 2009" *by* J. H. Kim et al.

Anonymous Referee #1

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The authors present hygroscopicity and CCN data from for field campaigns near the Korean peninsula from 2006-2009. Different episodes (polluted/unpolluted) are identified and growth factor measurements and CCN activity for these periods are compared throughout all four data sets. Four different methods are applied in order to perform CCN closure studies and it is concluded that the assumption of a constant kappa (0.3) for all particles is not appropriate as it leads to an overprediction of the CCN activity of smaller particles (< 100 nm). Instead it is suggested that size-resolved measurements should be used to predict CCN numbers. Aerosol-cloud interactions are among the most uncertain factors in assessing the anthropogenic impact on radiative forcing. Thus, evaluating the ability of aerosol particles to act as CCN in different regions of the world is certainly an important and valuable task. Thus, the current study warrants publications in ACP. However, the current study (and many prior ones, too) analyze data in much detail and often come to the conclusion that 'only detailed (e.g., size-resolved) information gives the 'best CCN closure'. While this might be true, such information can never be implemented into climate models and thus a broader assessment is needed that states 'how wrong are the predictions of we make simpler assumptions'. I think the current study has the potential to provide such useful guidance as I will detail below in my comments. In addition, I have several minor comments that should be addressed in order to improve the readability and clarity of the manuscript.

General comments

1) The authors compare four different methods for CCN closure and discuss that Method-2GF leads to the best CCN closure whereas the other methods that either ignore size-resolved information show larger deviations. While the data in Table 5 show the deviations for each method from 'truth' (i.e. measurements) a comparison would be useful that shows how much the individual methods differ among each other. I.e., given that aerosol/cloud modelers look for simplicity, how 'bad' would the CCN closure be if e.g. constant hygroscopicity vs size/temporal resolved one is assumed. I recognize that this information could be derived from the data given here, but an additional figure would make it clearer that shows (CCN-predicted by Method 1, 2 or 3) versus CCN(predicted using Method 4). The results of this comparison should be briefly discussed in light of discussions that compare CCN effects on cloud droplets e.g., (Rissman et al., 2004; Cubison et al., 2008).

2) The suggestion by Andreae and Rosenfeld (AR08) to use kappa = 0.3 for continental aerosol is definitely a useful one and gives 'reasonable' results. As the authors show it could be improved since obviously smaller particles are less soluble. Since it is not feasible to take into account (due to lack of data) or implement size-resolved hygro-

scopicity data on a global scale into models, could you come up with a refinement of the AR08 recommendation? Does a fixed hygroscopicity (kappa < 0.3) for particles < 100 nm give robust and good results?

Specific comments

p. 19684, l. 12: Are these the minimum/maximum values or ranges of standard deviation?

p. 19684, ,l. 19 (and throughout the text): Use 'CCN activation' instead of 'cloud activation' since (i) activation occurs for an individual particles (CCN) and (ii) cloud formation would include many additional processes that are not taken into account by CCN studies.

p. 19685, l. 8-10: This statement is somewhat misleading. It is true that the aerosol indirect effect has been identified the largest uncertainty in understanding radiative forcing. However, the role of CCN is only one of many issues that is encompassed by the term 'aerosol/cloud interactions'. So, your sentence should be reworded.

p. 19685, l. 25: Vapor is continuously condensing on particles; however, in equilibrium it also evaporates at the same rate. To be correct, you should say that above Sc, the former rate is much greater than the latter leading to continuous and efficient growth.

p. 19686, l. 3: I don't think that Sc has ever been measured in clouds. The typical supersaturation in clouds is < 1% with significant temporal and spatial variability. Thus, measuring it in 'real' clouds seems impossible.

p. 19690, l. 19: Be more specific here and replace 'some important statistics of the aerosol physical properties' by something like 'Average number concentrations and standard deviations of the aerosol populations. . .'

p. 19691, l. 15: A brief summary of the method by Swietlicki would be helpful.

p. 19692, l. 7: Can you really say based on a single GF that particles were internally

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mixed? Without any further composition information, I think, the only statement you can make is that they had very similar hygroscopicity.

p. 19692, l. 4: what does ‘which is located at the dominant upwind region’ refer to?

p. 19693, l. 7: Why could the curve not be constructed?

p. 19693, l. 24/25: Be more specific: What is it in the studies by Mocida that supports your results ?

p. 19694, l. 13-16: I don’t understand why the Ddry-scan and the S-scan should give different results in terms of CCN activity as both are based Kohler equation. Are you saying that the S-scan is often done for the whole aerosol population and thus is biased if hygroscopicity varies with size?

p. 19695, l. 15/16: How relevant is $S > 0.2\%$ for atmospheric clouds? Stratocumulus clouds, the most abundant cloud type, might have S smaller than that. It has been shown that CCN predictions for much lower S is associated with greater uncertainties (e.g., Kammermann et al., 2010 and references therein). Some words on these issues should be added.

p. 19696, l. 15 ff: This is not conclusive: On the one hand you say that the solubility of organics cannot fully explain the differences in kappa derived from HTDMA and CCN measurements (l. 17/18); on the other hand you are saying the high organic fraction of the Gosan and BCMO aerosol could have caused this effect.

p. 19607, l. 22: Is this sentence basically just repeating that you assume an internal mixture?

p. 19697, l. 25 ff: (i) You show in Table 3 that kappa derived from CCN and HTDMA differ quite a bit but nonetheless you use the HTDMA-derived value for your CCN closure (due to the lack of data from CCN-derived kappas) and add a fair amount of discussion on the importance of differences in kappa for CCN closure. This seems to be somewhat inconsistent and the uncertainties as introduced by using a ‘wrong’ (too small)

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kappa and should be discussed in light of the resulting differences in CCN closure for the different methods assumed here. (ii) I got confused about the number of the different methods. I count five different ones (1-GF, 2-GF, 2-Sc, 3-GF, and ‘4’) whereas 3-GF was only applicable to part of the data. In order to avoid the confusion, I suggest renaming Method 4 to something like ‘Method constant-kappa’. In addition, I suggest adding a table (or information to Table 5) summarizing briefly the characteristics of each method.

p. 19700, l. 6: Are there any data sets that suggest that the hygroscopicity varies over seasons and years?

Table 5: Add information of different Methods here or refer to (an additional) table or to the respective text section.

Technical comments

p. 19684, l. 6: add ‘s’ to measurement

p. 19687, l. 27: ‘replace in each day’ by ‘of each day’

p. 19689, l. 21: remove ‘the’ (. . .lower PM2.5) p. 19690, l. 1: replace ‘the’ by ‘a’ (a 13-year. . .)

p. 19690, l. 4: replace ‘their’ by ‘the’

p. 19691, l. 11: remove ‘fresh’ (seems redundant)

p. 19691, l. 20: remove ‘done’

p. 19692, l. 9: remove ‘within the sample’ (not clear what is meant by it)

p. 19693, l. 19: replace ‘can act as good CCN as sulfate’ by ‘is as hygroscopic as sulfate’

p. 19695, l. 5: remove ‘is’ (Wiedensohler. . . presumed)

p. 19701, l. 28: replace ‘its’ by ‘their’

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p. 19707. l. 33: Weingartner misspelled Figure 6: Quality should be improved by increasing figure size and/or line weight and symbol size.

Figure 7 and 9: Hard to read, use larger font.

Figure 11: Define k(GF) and k(Sc) in figure caption

Figure 12: (i) the 50% lines are not visible (ii) increase font and symbol size for better readability (iii) Refer in Figure caption to Text section and/or table where Methods are explained.

Additional references

Cubison, M., Ervens, B., Feingold, G., Docherty, K., Ulbrich, I., Denkenberger, K. A., Shields, L., Prather, K., Snyder, D., Schauer, J. J., Hering, S., and Jimenez, J. L.: The influence of chemical composition and mixing state of Los Angeles urban aerosol on CCN number and cloud properties, Atmos. Chem. Phys., 8,5649-5667, 2008. Rissman, T., A. Nenes, and Seinfeld, J. H.: Chemical Amplification (or Dampening) of the Twomey effect: Conditions derived from Droplet Activation Theory, J. Atmos. Sci., 61, 919-930, 2004.

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