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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Effects of aerosol organics on cloud condensation nucleus (CCN) concentration and first indirect aerosol effect" *by* J. Wang et al.

Anonymous Referee #1

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Review

The study by Wang et al. addresses the analysis of cloud condensation nuclei (CCN) data acquired during the MASE campaign. The authors have identified three different air masses (boundary layer (BL), free troposphere (FT), and above clouds (AC)) that differ in their origin, and, thus, in aerosol chemical composition and size distributions. They show that for BL and FT aerosols the assumption of an internal mixture of inorganic and insoluble organics is sufficient to obtain good closure between measured and predicted CCN number whereas they suggest that for AC aerosols more information on organics' hygroscopicity have to be available. Based on the variation in

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organic properties they discuss in the CCN closure study the authors have performed an estimate of the extent to which uncertainty in organic aerosol properties translate into uncertainties in radiative forcing. They show that composition effects are greatly reduced in clouds and they conclude that the error is only significant if particles are composed of $>$ 90% organics; in all other cases the properties of the inorganic fraction dominates the hygroscopic properties of the population and the influence of the organics on cloud drop number concentration over a wide range of hygroscopicity is minor. This study represents a nice combination of a detailed analysis of CCN data and a discussion of the significance of measurement and/or model uncertainties for the broader picture of radiative forcing. The topic is certainly well suited for Atmospheric Chemistry and Physics and I recommend its publication after my comments below have been addressed.

General comments

1) The authors conclude that because of the different shape of the AC aerosols size distributions as compared to BL and FT aerosols, the organics in the AC aerosols are externally mixed. Since this assumption has a significant impact on their following results, more detail should be given to their linear combination approach to fit separate modes to the measured size distributions (p. 9794, l. 4 ff.). The large underestimate of CCN in Figure 6 is due to the fact that about 90% of all particles are excluded as CCN, and, thus, it is not so much the organic properties but the mixing state of the organic (insoluble) particles that leads to the significant deviation in Figure 6. The assumption certainly represents an extreme case and the authors should emphasize this fact more clearly in their discussion: How likely is it that 'above cloud' particles are externally mixed with all other aerosol compounds? Can any information be given about the origin, and, thus, age of these particles? In Figure 4, it seems that the smaller particles ($<\tilde{}$ 200 nm) have even higher organic fractions that the larger ones. Is it possible that different organic compounds are present and only newly formed small organic particles are externally mixed whereas other organics are internally mixed in the larger mode?

ACPD

8, S4711–S4715, 2008

2) As a consequence of the mixing state assumption for AC particles, the authors show that AC particles have to be hygroscopic (kappa = 0.12) whereas best closure for BL and FT masses is obtained for the assumption of insoluble organics. If the extreme mixing state is indeed reality, how can the difference in organic properties be explained for the different scenarios? It has been shown that the kappa of organics can span several orders of magnitude (Petters and Kreidenweis, 2006). For the sensitivity analysis of N(CCN), N(drops) and radiative forcing, it might be more useful to investigate a wider range of kappa covering a wider range (on a logarithmic scale).

Specific comments

p. 9786, l. 10: What 'chemical processes during cloud nucleation' are you referring to?

p. 9788, l. 20: Replace 'as possible' by 'if possible'

p. 9788, l. 22: What is meant by 'state parameters'? 'Mixing state'?

p. 9789, l. 19 ff.: Was the supersaturation corrected for gradients in the CCN counter ?

p. 9792, l. 12: The van't Hoff factor is the product of the number of ions and the osmotic coefficient. Was the factor for the inorganic salts corrected for non-idealities or was the maximum number of ions assumed?

p. 9796, l. 10/11: It has been shown in CCN studies that water-soluble organics do not necessarily exhibit a kappa value greater than 0 (cf. e.g., Ervens et al., 2007). The reason for this apparent contradiction is that the analytical method that is used t extract water-soluble organics from particulates (Particle-Into-Liquid-Sampler) operates at very high dilution whereas the water/solute mass ratio at particle activation (S ˜ 1) might not be sufficient to dissolve the organics.

p. 9796, l. 22: Rephrase and use something like 'the number of solute moles was determined by the inorganics' since - according to Koehler theory - it is the number of moles not the ion concentration (note: most organic do not form ions) that determines hygroscopicity

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8, S4711–S4715, 2008

Interactive Comment

p. 9798, l. 1ff: Clarify that the drop-CCN relationship is valid for internally mixed aerosols that have bulk composition (i.e. no change in composition with size). Was the study by Sotiropulou also performed for stratus/stratocumulus clouds?

p. 9798, l. 25: Repeat here that the large variation in radiative forcing for the AC aerosols is due to the assumption of a different mixing state.

p. 9804, l. 27: This statement of the accurate prediction using an average kappa seems very strong. Since one would predict probably similarly good results if kappa is different within a factor of 2, I suggest to rephrase: 'can be predicted sufficiently accurate'.

p. 9805, l. 12: The statement that 'an average kappa(org) may be sufficient for many aerosol types' seems to be a contradiction to your CCN analyses where you showed that for two out of three cases kappa can be assumed to be zero. Here again, you should clarify that successful CCN closure could be only obtained under the assumption of external mixing of hygroscopic organics.

Figure 2c: Can you comment on the size distributions of the organics? Are the peaks around 90 nm and 200 nm different modes or artifacts due to the resolution of the measurements?

Fig. 8: (i) The figure caption should refer to Eq.-17 in order to make it easier to understand the connection between the two y-axes. (ii) Add to the figure caption that this estimate refers to an internal mixture without any size-dependent composition and a reference value of kappa = 0.12.

Technical comments

p. 9789, l. 21; p. 9790, l. 5: quadrupole

p. 9789, l. 25: 'particle time of flight' (not fight)

p. 9792, l. 22/23: Reword: Either 'the concentration [...] is small' or 'the solution is [...]

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8, S4711–S4715, 2008

dilute’

p. 9795, l. 26: add ’with’ (associated with)

p. 9796, l. 10: Replace organics by organic

p. 9804, l. 23: Replace ’archived’ by ’achieved’

p. 9806, l. 25: Worsnop

p. 9806, l. 23: physical

Figure 5: Choose different colors. The cyan and blue lines are hard to distinguish.

Interactive comment on Atmos. Chem. Phys. Discuss., 8, 9783, 2008.