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Atmospheric
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Discussions

Interactive comment on "Aerosol mixing-state, hygroscopic growth and cloud activation efficiency during MIRAGE 2006" *by* S. Lance et al.

Anonymous Referee #2

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This manuscript presents results from measurements outside Mexico City over a 16 day period. The measurements focus on the water uptake properties of the ambient aerosols. Several state of the art instruments were used including an aerosol mass spectrometer for measurements of aerosol chemical composition, a continuous flow CCN counter for determination of critical supersaturations and a HTDMA system for measurements of hygroscopic growth.

The experimental data are analyzed in the framework of kappa-Köhler theory. Kappa values derived from the different techniques are compared. Kappa_CCNc decreases as function of size during all time periods while the ccn active fraction increases with particle size. Kappa derived from AMS measurements agrees reasonable well (with some deviations) with the kappa values derived from CCN measurements.

The authors find that there is a correlation between new particle formation events and increased kappa values. Particles are externally mixed during morning rush hours and internally mixed at mid-day. The authors point out that it is important to account for aerosol mixing state, otherwise one can overestimate CCN concentrations by 50-100%.

The manuscript is well written, the figures are very nice and interesting data are presented. I have some comments as outlined below which the authors should address before publication can be recommended.

General comments In several places it is unclear what the averaging period is. For example: Fig 2: "Average diurnal profile…" – average over which period – the whole campaign? Fig 5: is this average over NPF days? This should be clearly stated.

Specific comments

Page 15713 The authors provide nominal and real dry particle diameters – how were the "real" values determined?

P. 15714 Why are the points at 40 and 60 nm at the lowest set points excluded? Please explain. How was the CCNC calibrated?

P.15715 Since the HTDMA system as I understand was employed here for the first time it could be described in a little more detail. For example: what is the residence time in the nafion humidifier tube. With respect to the calibration – it says that the expected growth factor for NaCl is 1.3 at 90% RH based on Petters and Kreidenweis and that the RH was calibrated accordingly. The growth factor for NaCl at this RH is higher, see for example (Brechtel and Kreidenweis, 2000) or (Carrico et al., 2008). Was a kappa value used? If so why? Would it not seem more direct to use a growth factor at the specific relative humidity? Was a shape factor used/assumed for the NaCl particles?

P. 15616 To help the reader I suggest to show an activation spectrum (non-averaged) or at least refer the reader to figure 7 to get an idea of what it looks like. How was double charged particles accounted for – looking at fig 7 it seems like they were somehow removed before fitting?

P. 15719: Is there a reference for how multiple charging was accounted for that could be given? As discussed by the authors later in the manuscript it complicates the fitting when the ccn-spectrum does not reach a stable activated fraction (for the smallest particles) – it could be described in 2.2.1 how that was handled.

Page 15716-15717 For practical reasons the sheath to aerosol ratio in the DMA in front of the CCN counter is very low (∼2.7). It would be nice to show an example demonstrating how much of the slope can be attributed to the DMA transfer function and how much to other effects (chemical heterogeneity, slightly soluble material, surface tension). Other groups have also investigated such effects and the authors could consider citing some of those: e.g. (Rose et al., 2008) (Svenningsson and Bilde, 2008).

Page 15719: "The measured and simulated CCN concentrations are inverted to determine nCCN(dp,S)=. . .." It is not clear how this was done and when these inverted number concentrations where used. This should be explained in more detail. What is shown in Fig 1 a and Fig9 – is it CCN concentration measured by the CCN counter or has it been somehow inverted? I find the use of the * a bit confusing. Why not call it kappa_CCNc, kappa_AMS, kappa_HTDMA for the experimentally determined values and then in eq (5) use kappa as also done in Petters and Kreidenweis eq 10?

Page 15728 The authors write in the conclusion that a linear fit is obtained with lower slope than observed in several other studies (I assume this refers to Fig. 6) but when discussing fig 6 in the main text it was concluded that the results fall within results from several other studies. This seems inconsistent.

P. 15723: The use of the term "often" in the discussion of five days is maybe too strong – instead it should be specified on how many days out of the five considered this was the case.

Page 15725: the authors state that the increase in both kappa_CCNC and kappa_HTDMA during NPF events is contrary to the observations of Dusek et al. 2010 – what could be the reason for this? The authors could consider also to compare/contrast with ccn activity and NPF events in other environments e.g. (Sihto et al., 2011).

Page 15723: Please provide uncertainties on the determined kappa_inorg and kappa_org from the intercepts in fig. 6. Figures: Fig 3: a: critical supersaturation had distinct values – I would suggest to indicate those with separate colors rather than let it look like a continuum (right hand scale). Fig 2: "Average diurnal. . ." the time of the average should be given. Fig3: red and grey lines should be explained in the caption Fig 4: It would be nice to see error bars on the "all other days" data to see if there is a significant difference between NPF days and "all other days". Fig 6: What were the particle sizes used in the literature studies shown? It should be stated in the figure caption which days are included in the figure- Fig 7: is the morning rush hour a campaign average?

Minor comments: in the introduction it is explained what T1 is – it should also be explained what T0 is.

On page 15712 it says that the campaign was in the period 16-31 March – in Figure 1 it looks like it is 15-30 March.

Was there air-condition in the container to keep the cold end of the CCN counter at a constant temperature? This could be stated.

P. 15721: Line 23: it should say in the text that this is for 1% supersaturation.

Page 15718: it says that measured values of kappa* are referred to as kappa_CCNc , I suggest to rephrase (kappa is not measured, it is derived from measurements.)

References Brechtel, F. J., and Kreidenweis, S. M.: Predicting particle critical supersaturation from hygroscopic growth measurements in the humidified TDMA. Part II:

Laboratory and ambient studies, Journal of the Atmospheric Sciences, 57, 1872-1887, 10.1175/1520-0469 2000.

Carrico, C. M., Petters, M. D., Kreidenweis, S. M., Collett, J. L., Engling, G., and Malm, W. C.: Aerosol hygroscopicity and cloud droplet activation of extracts of filters from biomass burning experiments, Journal of Geophysical Research-Atmospheres, 113, 10.1029/2007jd009274, 2008.

Rose, D., Gunthe, S. S., Mikhailov, E., Frank, G. P., Dusek, U., Andreae, M. O., and Poschl, U.: Calibration and measurement uncertainties of a continuous-flow cloud condensation nuclei counter (DMT-CCNC): CCN activation of ammonium sulfate and sodium chloride aerosol particles in theory and experiment, Atmos. Chem. Phys., 8, 1153-1179, 2008.

Svenningsson, B., and Bilde, M.: Relaxed step functions for evaluation of CCN counter data on size-separated aerosol particles, J. Aerosol Sci., 39, 592-608, 10.1016/j.jaerosci.2008.03.004, 2008.

Sihto, S. L., Mikkila, J., Vanhanen, J., Ehn, M., Liao, L., Lehtipalo, K., Aalto, P. P., Duplissy, J., Petaja, T., Kerminen, V. M., Boy, M., and Kulmala, M.: Seasonal variation of CCN concentrations and aerosol activation properties in boreal forest, Atmos. Chem. Phys., 11, 13269-13285, 10.5194/acp-11-13269-2011, 2011.

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