

This paper presents aerosol hygroscopicity growth and CCN activation spectra of submicron aerosol particles observed at T1 site outside of the Mexico City during MIRAGE 2006 study. During new particle formation (NPF) events, increased particle characteristic hygroscopicity (κ^*) and CCN activation fraction were observed, the increases were more significant for smaller particles. It was found that fresh traffic emissions resulted in large fraction of externally mixed particles in early morning, whereas at midday particles could be described as internally mixed. The authors also report that the size dependence of κ^* cannot be predicted using size-resolved AM measurement alone. The discrepancy is attributed to the lack of information on mixing state and refractory material, which cannot be provided by AMS. Hygroscopicity derived from composition measured by AMS (κ_{AMS}) was able to describe CCN concentration reasonably well, especially at highest CCN concentrations, partly because the highest CCN concentration was observed during the daytime when aerosol was more internally mixed and the organic fraction was relatively low. However, falling to account for aerosol mixing state often resulted in significant overestimation of CCN concentration during early morning rush hours. These findings are consistent with earlier studies. The topic is well suited for Atmospheric Chemistry and Physics. I'd like to compliment the authors on this interesting study. Following are my comments and suggestions.

Page 15712, line 1. After reading the section 3.4, I realized this is based on the comparison between CCN concentrations calculated using κ_{AMS} , E , σ_{κ} and the measured N_{CCN} . Therefore I'd suggest change the sentence to “ κ_{AMS} is able to describe CCN concentrations reasonably well, provided mixing state information is available, ...”

Page 15712, line 3-5 (abstract). “is partly due to the fact... organic fraction is relatively low”. The discussion on this point is missing from the main text. Please include relevant discussions in the manuscript.

Page 15714, line 1- 12: When CCNC is operated at low supersaturations (i.e. $< \sim 0.1\%$), the driving force for droplet growth is low. Therefore a reduced CCN flow is required such that particles could grow to sizes larger than the OPC detection threshold. This also ensures that interstitial aerosol particles under the low supersaturation could be distinguished from activated CCN (Roberts and Nenes 2005, Lance et al., 2006). The CCN counter was operated at a relatively high flow rate of 0.75. How did the high flow rate impact the measurements at lower supersaturations?

Page 15715, line 5-7. Given the high RH, did RHs measured at different locations of the 2nd DMA agree?

Page 15715, line 12: I think the growth factor of NaCl at 90.8% RH is much larger than 1.3. Is this the value of κ instead?

Page 15716, Line 21: As described later, the function is influenced by both the chemical characteristics of the activated particles and the shape of DMA transfer function.

Page 15719, line 9, “the fraction of particles with $Sc < S$ is”, is this fraction the normalized activated fraction for size selected particles (i.e., $Ra^*(s)$)? Please clarify.

Page 15719, line 11, “The measured and simulated CCN concentrations...” Is the measured CCN concentration total CCN concentration or size resolved? This paragraph appears to be quite confusing. Please rewrite and clarify.

Page 15720, line 13-14: Please rephrase this sentence. I assume that your method (Eqn. 11) does not include externally mixed non-CCN-active particles. However, the sentence appears to suggest that method reported by Su et al. (2010) does not include externally mixed non-CCN-active particles.

Page 15721, line 9-10. These assumptions are not always appropriate, especially during early morning traffic hours. Please include some discussions on the appropriateness of these assumptions.

Page 15721, Line 14-15. I understand that soot often has fractal geometry, but how does it contribute to measured increase in organic mass fraction at smaller particle sizes? Are you suggesting that particles with higher organic mass fraction were more fractal, such that their d_{va} was smaller? Please clarify.

Page 15723, line 15. Based on Figure 2b, f_{org} should be lower at 40 nm than that at 100 nm (f_{org} of activated CCN).

Page 15723, line 16. Please clarify what the “aerosol dilution” means.

Page 15723, line 25-26. $\kappa_{org} = 0.04$ and 0.21 for 100 and 40 nm, respectively. Do AMS data show higher POA at 100 nm?

Page 15726 and 15727. Among the main results of the paper are (1) externally mixed primary particles emitted during the morning rush hours rapid grew into larger particles that were internally mixed; and (2) mixing state information is important for describing CCN concentration during the morning rush hour, and calculated N_{ccn} often significantly overestimates measured CCN concentration. In addition, the calculated CCN concentration agree well with the measured during daytime when aerosols are more internally mixed.

I would suggest discuss these results in relation to earlier studies pointing in the same direction, especially those carried out in the Mexico City during the same field campaign:

Moffet, R. C., and Prather, K. A.: In-situ measurements of the mixing state and optical properties

of soot with implications for radiative forcing estimates, Proc Natl Acad Sci USA, 106, 11872-11877, 2009.

Wang, J., Cubison, M. J., Aiken, A. C., Jimenez, J. L., and Collins, D. R.: The importance of aerosol mixing state and size-resolved composition on CCN concentration and the variation of the importance with atmospheric aging of aerosols, Atmos. Chem. Phys., 10, 7267-7283, 2010.

Page 15727, section 3.4. Does Figure 9 show the comparison of total CCN concentration (as a function of S) or size resolved CCN concentration (a function of S and D_p)? It appears that the comparison is for total CCN concentration. But I couldn't find any description on total CCN concentration measurements in the manuscript. In addition, if the comparison is for predicted and measured total CCN concentrations, then the data point would be colored by supersaturations instead of particle size. Please clarify and include more details on what quantities are compared in the figure.

Page 15727, section 3.4. I'd suggest include the comparison for CCN predicted using κ_{AMS} , $E=1$, $\sigma_k=0$. This represents the calculations when only AMS data are available, and particles are assumed as internally mixed.