Atmos. Chem. Phys. Discuss., 7, S6679–S6683, 2007
www.atmos-chem-phys-discuss.net/7/S6679/2007/

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Influence of particle size and chemistry on the cloud nucleating properties of aerosols" *by* P. K. Quinn et al.

Anonymous Referee #1

Received and published: 8 November 2007

General comments:

The paper presents measurements of particle size distribution, particle composition and CCN concentrations made on board a research vessel in the Gulf of Mexico during the Texas Air Quality - Gulf of Mexico Atmospheric Composition and Climate Study in 2006. The authors show that the chemical composition (the fraction of insoluble organic material) becomes important in cloud activation efficiency for small particles and small supersaturations. The observed dependence of cloud activation efficiency on particle chemistry from the field data is compared with model calculations and with other field studies. The error induced by neglecting chemical composition in predicting cloud nucleating efficiency is calculated as a function of size, insoluble mass fraction and supersaturation.

Generally the paper is well written and understandable. It is a valuable contribution to aerosol-cloud research and fits well into the scope of ACP. I encourage publication after the major comments (see below) have been addressed.

I have four major comments:

1) The CCN measurements are not size resolved. Therefore, the method to infer the critical diameter as is explained in section 3.2.1 relies on the assumption that the CCN/CN ratio (the activation efficiency) is an ideal step function that jumps from 0 to unity at the critical diameter. In reality this is usually not the case: Firstly, the activation curve is broadened around D_{crit}, and secondly, the activation curve sometimes lies below unity for larger particles (due to measurement uncertainties or due to incomplete activation), sometimes also above unity (due to measurement uncertainties). Size resolved CCN measurements usually define the critical diameter as D_{50}, i.e., the point where the activation curves reaches 50% of the maximum value. What is the uncertainty for D_{crit} induced by the method presented here?

2) The method used to infer the geometric mean diameter D_{gn} needs to be discussed in more detail. The authors fit lognormal distribution to the Aitken mode and to the accumulation mode und take D_{gn} of the accumulation mode, if an accumulation mode is present. In cases without an accumulation mode, they use D_{gn} of the Aitken mode. To my understanding this leads to two problems: 1) an overrepresentation of small accumulation modes. 2) A "jump" of D_{gn} from the accumulation mode to the Aitken mode at the moment when the accumulation mode disappears. Thus, I expect that the geometric diameter in Figure 3 is biased in favor of larger diameters. Wouldn't it be more useful to "force" a monomodal distribution through both Aitken and accumulation mode?

3) The comparison with the Dusek 2006 data is not correct: Dusek 2006 did not differentiate between HOA and OOA. Assuming all POM in the Dusek 2006 study to be HOA leads to a HOA fraction that is too high. If one assumes the same HOA/OOA ratio

ACPD
7, S6679–S6683, 2007

as the present Quinn manuscript (HOA/OOA ca 2:1 for sub-200), the HOA fraction is overestimated by approximately 33%. This would shift the Dusek box to 40 - 60%, into a range where the CCN concentrations are higher and the errors are lower. In this range "size matters more than chemistry". Furthermore, how was the range of D_{gn} of Dusek estimated? The simulations in Dusek 2006 were made for the whole campaign, not only for the 4 case studies plotted in Fig 2 of Dusek 2006. The range of D_{gn} used for the simulations is not given in Dusek 2006.

4) Please give the time period when the measurements were performed. Only in Figure 2 the reader can learn that two measurements were made on 8/5 and 9/11 (which I assume to be month/day). The times of the measurements should be noted in the introduction (e.g., on page 14174), and also indicated in Figure 1. Are the presented data (in terms of meteorological conditions and air mass trajectories) representative only for summer/fall or also for the rest of the year?

Minor Comments:

Abstract:

p. 14172, lines 2-5: Move first two sentences to introduction p. 1763, lines 5-9: These sentences are almost exactly repeated in the introduction, they can be omitted here.

Introduction:

p. 14173, line 18: Replace "cloud parcel" with "air parcel"

p. 14175, line 19: It should be noted here that a vacuum aerodynamic diameter of 200 nm can correspond to roughly 300 nm mobility diameter (assuming density 1.5, shape factor 1)

Methods:

p. 14177, line 20: Are the DMA columns custom built? In not, give type and manufacturer or reference.

ACPD
7, S6679–S6683, 2007

EGU

p. 14177, line 25: "Stratmann" (also on page 14196, line 33)

p. 14180, line 14: Why was 1000 mBq/m^3 used as a threshold? Are there any references?

Section 3.1.2

p. 1483, lines 22-25: These statement is a repetition of page 14178, lines 4-8.

Section 3.2.2

p. 14186, lines 17-25, also Table 2: The results and the methods of the PCA needs more explanation. Is this a more qualitative analysis or does a number like "0.76" in Table 2 mean that 76% of the variation in critical diameter can be explained by the sub-200nm mass fraction? A reference for the method is needed. What is "factor 2"? What is "CCN activation factor"? In the text it is only stated that "Table 2 shows the factor with the highest loading for the critical diameter. "

p. 14186, line 24: Why "negatively" instead of "negative"?

p. 14187, line 6: Equation 2 is usually referred to as "Kohler Equation" or "Köhler Equation" (e.g. Equ. 2 in McFiggans 2006).

Replace in Equ. 2 the second "=" by "≈" since this is an approximation (see McFiggans 2006).

Figures and Tables:

Figure 1:

I would suggest to plot the insert in the same size below the main figure

Figure 2:

- Make plot sizes of c + d uniform

- Indicate error bars in AMS size distributions

ACPD

7, S6679–S6683, 2007

Interactive Comment

- Legend: sub -> d < 1 μm, sub 200 -> d < 200 nm

- Insert space between the three bars "sub" and the three bars "sub200"

Table 2: replace "subum" by "submicron"

Table 3 and 4: The sentences "calculated using a growth factor of 1.3" that are marked with an asterisk should be in the footnote.

Interactive comment on Atmos. Chem. Phys. Discuss., 7, 14171, 2007.