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Interactive comment on “Technical note: Characterization of a static thermal-gradient CCN counter” *by* G. P. Frank et al.

Anonymous Referee #1

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General Comments

This note presents calibration data for a static diffusion chamber (SDC) CCN counter developed at the University of Mainz. As indicated in the abstract, the paper describes in detail methods and results for calibrations of the CCN counter with respect to both supersaturation and concentration. Also considered is the effect of water vapor depletion on the instrument. These experiments lead the authors to three main conclusions: 1) experimental calibrations of SDC CCN counters with respect to supersaturation are important; 2) the number concentration calibration should be performed as a function of the instrument supersaturation; and 3) there was no evidence of water vapor depletion affecting the supersaturation in the instrument.

A well-developed paper presenting the information described above would be suitable for publication as a technical note. Such suitability would rest entirely on the potential strength of the final two conclusions; the methods described in the paper are not particularly unique, and the first conclusion has been raised previously by several other papers. Unfortunately, the evidence presented is insufficient to support the second and third conclusions. As such, this paper should not be accepted for publication in Atmospheric Chemistry and Physics without extensive modification. Specific issues that the authors should address further are discussed below.

Specific Comments

Page and line number are as in the print version of the paper available on the ACPD website.

Page 2154, Lines 7-26: These two paragraphs make clear that the two calibration methodologies presented in the paper have been developed and used several times previously. Specifically, Delene and Deschler (2000) developed the technique wherein the number concentration measured by the CCN counter is compared to parallel measurements with a CN counter. Similarly, it is now a common technique to experimentally determine the instrument supersaturation by stepping through a range of particle diameters and maintaining a constant supersaturation (or equivalently, by keeping the particle size constant while stepping through supersaturations). Indeed, the authors themselves note numerous papers in which these techniques are used. Furthermore, the two techniques are often performed in parallel; in addition to the papers cited in the text, the authors are encouraged to note the calibration data presented by Raymond and Pandis (2002), VanReken et al. (2003), and Roberts and Nenes (2005). Given the degree to which these calibration techniques are currently employed, describing their use is insufficient to merit publication in itself.

Page 2155, Lines 1-2: This statement is somewhat misleading. The first paper cited, that of Jennings et al. (1998) is not particularly recent; their efforts to validate their

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instrument are not notably less advanced than other studies published around that time (e.g., Delene et al., 1998; de Oliveira and Vali, 1995). The more recent paper (Roberts et al., 2003) indicates that calibrations were performed, and that these data are available from Dr. Roberts' thesis. The authors have cited this thesis elsewhere in the paper, and are presumably using the same instrument in the current work.

Page 2155, Lines 3-6: The objectives of the paper as stated here are not sufficient for a stand-alone publication. Almost all of the CCN instrument calibrations cited by the authors, as well as those cited in the above comments, were included as sections in larger papers describing either an instrument's development or its use in a scientific study. The methods described here are not novel, and calibration data for a specific instrument are not by themselves interesting enough to merit publication. As noted in the general comments, this paper is only interesting to the degree that it increases our understanding of the performance of SDC CCN counters. The objectives should be changed to emphasize the portion of the paper that attempts to do this: the dependence of the number calibration on supersaturation and the effect of water vapor depletion on instrument performance.

Page 2156, Lines 8-11: The authors themselves demonstrate here that one of their chief conclusions (that experimental CCN calibrations are important) is already widely realized by the CCN measurement community, and that the techniques for performing such calibrations are already well-established.

Page 2157, Lines 9-11: The authors should make more clear why the calibration technique was different at the lowest supersaturations. As I understand it, it was faster/easier to vary supersaturations than particle size so this was the preferred technique. However, at the lowest supersaturations (and the highest?), it was not possible to scan the entire supersaturation range, so particle size was varied instead.

Page 2157, Line 9- Page 2158, Line 27: This description of the analysis is difficult to follow, primarily because of the chosen subscripts for the various supersaturations involved. Snom is fine, but Sexp and Sc lead to confusion the way they're used here. Sc is a thermodynamic value for a single particle, but is used here to denote the actual supersaturation in the diffusion chamber; perhaps Sact could be used instead? Sexp is normally used to express an experimental supersaturation, but is used here in a somewhat different sense. Perhaps it could also be replaced with a different symbol? Labels in Figure 3 contribute to this general confusion.

Page 2158, Lines 14-18: This is indeed a large difference between the nominal and the actual supersaturation. How much different would T have to be from the nominal value to explain such a large change in supersaturation? Also, has any attempt been made to model the temperature discrepancy? Giebl et al. (2002) attempted this with some success, and Lance et al. (2006) recently considered the issue in some detail (albeit for a different CCN instrument design).

Page 2159, Lines 15-16: The authors note here that the measuring volume has been determined in a previous calibration. How was this calibration performed? Given that the volume can be changed, how confident are the authors in the stability of this number?

Page 2159, Lines 15-28: A couple questions on this discussion. First, the authors seem to indicate that the offset at higher concentrations is entirely due to coincidence errors. Why was water vapor depletion eliminated as a potential cause? The analysis in the next section looks for this effect at concentrations less than 4000 cm-3, but here you indicate that the data don't become unreliable until concentrations exceeding 5000 cm-3 are reached. Also, how did you settle on 5000 cm-3 as the upper limit for reliability?

Page 2160, Lines 3-18: These are good points presented in these paragraphs (though the overestimation is a bit hard to see in Figure 4b). However, because the bias should always be positive as a result of the analysis procedure, it seems that the uncertainty in line 17 should not be expressed as plus or minus a constant (i.e., the positive and

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negative uncertainties should be different).

Page 2160, Line 19- Page 2161, Line 10: As has been noted above, this is potentially the most important point in the entire paper. If the counting efficiency is indeed a strong function of supersaturation across the entire range and even at low concentrations, then that finding should be widely known. However, the evidence must be presented more convincingly, and the argument developed more fully. For whatever reason, this experiment is the only one for which individual data points were not made available; the figure would be more persuasive if they data were included. It strikes me also that if droplet growth were the cause of the variability in counting efficiency, then composition would also affect it. In that case, how could ambient aerosol (with unknown composition) be analyzed reliably? The ramifications of this finding should be explored in much more detail.

Page 2161, Lines 12-23: Like the preceding point, this examination of the effect of water vapor depletion is potentially important. The limited evidence that is presented does support the authors' claim, but more extensive exploration of the effect would be beneficial. Specifically, what would happen at lower or higher supersaturations? What about at higher particle concentrations? The effect in the latter case would likely be difficult to determine, given the difficulty in separating the effect of droplet coincidence from that of water vapor depletion.

References

References not included below are cited by the authors in the original text.

Lance, S., Medina, J., Smith, J.N., and Nenes, A.: Mapping the operation of the DMT continuous flow CCN counter, Aerosol Sci. Technol., 40(4), 242-254, 2006.

Raymond, T.M., and Pandis, S.N.: Cloud activation properties of single-component organic aerosol particles, J. Geophys. Res., 107(D24), 4787, doi:10.1029/2002JD002159, 2002.

Roberts, G.C., and Nenes, A.: A continuous-flow streamwise thermal-gradient CCN chamber for atmospheric measurements, Aerosol Sci. Technol., 39(3), 206-221, 2005.

VanReken, T.M., Rissman, T.A., Roberts, G.C., Varutbangkul, V. Jonsson, H.H., Flagan, R.C., and Seinfeld, J.H.: Toward aerosol/cloud condensation nuclei (CCN) closure during CRYSTAL-FACE, J. Geophys. Res., 108(D20), 4633, doi:10.1029/2003JD003582, 2003.

Interactive comment on Atmos. Chem. Phys. Discuss., 6, 2151, 2006.