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Interactive comment on "Closure between measured and modeled cloud condensation nuclei (CCN) using size-resolved aerosol compositions in downtown Toronto" *by* K. Broekhuizen et al.

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In response to the important points raised by Dr. McFiggans, we have expanded our closure calculations. Although we had included in the original manuscript a very simple treatment of external aerosol mixing, we have now replaced that with a more realistic approach motivated by Dr. McFiggans' comments. The nucleation and accumulation mode were decoupled from one another on the two days, September 15 and 16, where there were obviously two distinct particle modes. (On the other days, the modes were not sufficiently decoupled in the mass distributions to make this possible, nor was there a significant accumulation mode.) The contributions of these two modes to CCN concentrations were calculated independently, as described in the revised manuscript. The

removal of the accumulation mode soluble material, such as sulfate and ammonium, from the nucleation mode did result in a higher activation onset diameter for that mode. This decrease in the calculated number of CCN was partially offset by the additional contribution of relatively small (50-60 nm) accumulation mode particles to the total CCN number. Closure within experimental uncertainty was still achieved with this new analysis assuming external mixing. Sensitivity studies were also performed with regard to chamber supersaturation, soluble organic carbon, and droplet surface tension.

While it is true that all closure analyses are underconstrained without explicit knowledge of the mixing state of the aerosol, we nevertheless feel that this work does represent a significant contribution to the field. Previous closure analyses were performed with considerably less knowledge of the size-resolved particle composition in the CCN size range. However, we fully agree with Dr. McFiggans that all assessments of closure rely upon a number of critical assumptions. For that reason, we have explicitly stated that closure has been achieved under the assumptions set forth in the paper, most notably full insolubility for the organics and a droplet surface tension equal to that of water. The finding of closure could be later invalidated as our knowledge of organic solubility, surface tension or mixing states of the particles improves. Regardless, previous closure studies have had difficulty under conditions of high anthropogenic emissions and we have shown in this paper that size-resolved composition measurements can help resolve some of these issues, under a set of reasonable assumptions.

In response to the other points:

We have included sensitivity analysis with regard to solubility and surface tension. As described in the text, the overall findings are that a 10% decrease in the growing droplet surface tension or a 10% by mass water soluble organic content predict closure just within our estimated uncertainties. In general, the uncertainties associated with these assumptions and other aspects of the CCN calculations and measurements are significantly larger than the precision uncertainties in our closure plots.

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Concerning the AMS collection efficiencies, the 50% collection efficiency was only applicable to the two days in August mentioned in the original manuscript, not to the full study and, in particular, not for days for which we present CCN data. For that reason, we have clarified the manuscript to stress that the 100% AMS collection efficiency used in this work is largely based on the comparison to PILS data, but we also refer the reader to the Buset et al. (2005) paper to give more information on the comparison to the TEOM data over the whole sampling period. Full description of the TEOM comparison is beyond the scope and not central to the focus of the paper.

Interactive comment on Atmos. Chem. Phys. Discuss., 5, 6263, 2005.