

Interactive comment on “A closure study of cloud condensation nuclei in the North China Plain using droplet kinetic condensational growth model” by F. Yang et al.

A. Laaksonen (Referee)

ari.laaksonen@uef.fi

Received and published: 10 January 2012

This paper presents a closure study of CCN concentrations in an polluted environment in North China. CCN concentrations were measured at different supersaturations using a continuous flow CCN counter. Measured particle size distributions were used as a basis for the closure study, in which the particles were assumed to be either internal or external mixtures of ammonium sulfate (AS) and insoluble matter. CCN concentrations were calculated using a (nonequilibrium) condensational growth model, and alternatively using the Köhler theory. It was concluded that the best agreement with theoretical and measured CCN concentrations is achieved using an external mixture

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with a 60-70% mass fraction of AS. Discrepancies between the predictions from the condensational growth model and Köhler theory were discussed in light of kinetic effects taking place inside the CCN flow tube.

I think that this paper could ultimately be published in ACP. I especially like the discussion of the kinetic effects inside the CCN counter. However, I find the closure study oversimplified, and to some extent contradictory to the authors' previous work, and therefore suggest major revisions.

In the previous paper (Deng et al., 2011) dealing with the same data as the current paper, it was stated, based on measurements of size-resolved activation ratios, that "...the aerosols in the North China Plain consist of highly soluble material. Soluble fractions of more than 83% calculated based on the Köhler theory with assumed aerosol composition of ammonium sulfate and insoluble core, are needed to activate these particles". This seems contradictory to the information given in Fig. 2, which suggests that even an AS mass fraction of 60% yields too high modelled CCN concentrations. The reason for this apparent discrepancy should be clarified. Furthermore, it would be interesting to know how low AS mass fraction (in internal mixture) is needed for achieving reasonable agreement with the measurement.

Secondly, the external mixture of AS and insoluble matter with a size-independent AS mass fraction appears oversimplified and also contradictory to the data in Deng et al. (2011). With such an external mixture, the insoluble particles should not be able to activate at all, so that the activated fraction will simply decrease linearly with decreasing AS mass fraction, and a reasonable agreement with the measured CCN will be achieved just by decreasing the AS mass fraction enough. However, the size resolved activation fractions in Deng et al. (2011) indicate that above a dry diameter of 100 nm (150 nm), more than 90% of the aerosol activates at a supersaturation of 0.72% (0.36%), and therefore the 60-70% external mixture employed in the present paper appears contradictory to the measurements.

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In general, I think that when polluted regions are being considered, the use of ammonium sulfate and insoluble matter with a constant mass fraction throughout the whole aerosol size range is too simple regardless of the mixing state assumed. In any case, there are so many unknowns (including the accommodation coefficient) that there are multiple ways of achieving an apparently reasonable closure, and the question then is, what is the value of such closures? Unfortunately, no chemical composition data or growth factors appear to be available to the authors. Nevertheless, I believe that a better use can be made of the size distributions, together with the size resolved activation ratios.

I suggest that the authors select a few cases, based on Fig. 1, for a closer scrutiny. For example, take a couple of cases in which the datapoints are close to the 1:1 -line, a couple cases in which the calculated CCN is much above the 1:1 -line, and a case from the few datapoints in which the calculated CCN is below the 1:1 -line, and examine the measured size distributions. How do the size distributions differ? Can they be described as superpositions of two or more lognormal modes? If so, could some well reasoned compositions be assigned to the different modes, so that the aerosol is both internally and externally mixed? In this way, perhaps a closure with measured CCN can be obtained in such a way that the modeled data is also in accord with the measured activation fractions. This would make the study much more convincing.

Interactive comment on Atmos. Chem. Phys. Discuss., 11, 28969, 2011.

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