

### **General comments**

*The authors have conducted CCN measurements in China, nearby Beijing, during three consecutive weeks in 2009 and 2010. The authors have used a CCN counter with two columns to measure total CCN concentrations, total CCN activation efficiency and size-resolved CCN efficiency. A closure study comparing the results derived from the two columns was performed along with sensitivity studies showing that the size distribution is more important in predicting the CCN concentrations compared to the size-resolved activation efficiency (which reflects the chemistry and mixing state of the aerosol). The study provides new information on CCN characteristics in a relative polluted region, and meets the criteria required for publication in Atmospheric Chemistry and Physics. Before publication, the following relatively minor comments need to be considered.*

**Thanks for your comments.**

### **Major issues**

1. *Presenting some additional results would shed more light into the activation behaviour of the sampled aerosol. First, please provide time series for the bulk (absolute) CCN and CN concentrations for the measurement period. Second, it would be interesting to see a comparison between  $D_m$  and the so-called  $D_{50}$  diameter, i.e. the diameter at which the activation efficiency is 50%. Finally, using the inferred  $D_{50}$  diameters and modeling the activation behaviour with a step function, the CCN concentrations could be recalculated and compared to calculations based on Eq. 2.*

**The time series of wind, CN and CCN concentrations were shown in the revised manuscript.**

**The relationship between  $D_m$  and  $D_{50}$  is discussed below in following special comments.  $D_m$  is calculated based on the measured aerosol number size distribution and CCN number concentration. The aerosol number concentration for particles larger than  $D_m$  equals the measured CCN number concentration. This inferred cut-off diameter are in the ranges of 190-280, 160-260, 95-180, 65-120 and 50-100 nm for supersaturations of 0.056, 0.083, 0.17, 0.35 and 0.7%, with their mean values 230.1, 198.4, 128.4, 86.4 and 69.2 nm, respectively.  $D_{50}$  is the diameter where the activation ratio is 50% for the fitted activation ratio curve using the measurements at the 6 diameters. The fitting results for  $D_{50}$  depend on the choice of particle diameters, the number of selected diameters, and other uncertainties. The activation curve is not ideally sigmoidal. This paper did not give the fitted  $D_{50}$ , but provided an estimation of  $D_{50}$ . More than 50% of the particles larger than 200, 170, 90, 70, and 45 nm are activated at supersaturations of 0.058, 0.085, 0.18, 0.36 and 0.72%, respectively. The  $D_{50}$  is generally smaller.**

**And the result of calculated CCN concentration with mean  $D_m$  is discussed in the text.**

2. *The usage of English in the manuscript. The manuscript contains several grammatically incorrect and unclear expressions. Some of the language issues are pointed out below but, if possible, a native English speaker should proof-read the manuscript to improve the language and overall flow of the text.*

**We have improved the English usage accordingly in the revised version.**

3. *The description of methodology for calculation CCN concentrations in Section 2 needs some revisions. Related to this, the usage of the symbol  $N_{CCN}$  is not consistent in the manuscript: please use e.g. different subscripts to differentiate between  $N_{CCN}$  calculated under different*

assumptions. See specific comments below.

**We have re-defined the symbols of CCN in the text:**

**$N_{CCN}$ : CCN Number Concentration**

**$N_{CCN-obs}$  : Observed CCN Number Concentration**

**$N_{CCN-int}$ : Calculated CCN Number Concentration assuming internal mixture**

**$N_{CCN-ext}$ : Calculated CCN Number Concentration assuming partially external mixture**

**$N_{CCN-meas}$ : CCN Number Concentration at the supersaturations of size-resolved measurements obtained by fitting the  $N_{CCN-obs} \sim S^{\alpha}$**

**$N_{CCN-calc}$ : Calculated CCN Number Concentration using equation (3)**

**$N_{CCN-calc-realtme}$ : Calculated CCN Number Concentration using equation (3) with the measured data**

**$N_{CCN-calc-ConstAR}$ : Calculated CCN Number Concentration using equation (3) with the size-resolved activation ratio averaged throughout the campaign**

**$N_{CCN-calc-ConstNcn}$ : Calculated CCN Number Concentration using equation (3) with the particle number concentration averaged throughout the campaign**

**$N_{CCN-calc-ConstNASD}$ : Calculated CCN Number Concentration using equation (3) with the normalized aerosol size distribution averaged throughout the campaign**

### ***Specific comments***

1. Page 1, lines 26-28. Please report representative values (median or mean, for example) for the inferred cut-off diameters instead of making a qualitative statement only.

**This sentence was changed into ‘This inferred cut-off diameter are in the ranges of 190-280, 160-260, 95-180, 65-120 and 50-100 nm for supersaturations of 0.056, 0.083, 0.17, 0.35 and 0.7%, with their mean values 230.1, 198.4, 128.4, 86.4 and 69.2 nm, respectively. ’**

2. Page 2, lines 6-8. The last sentence of the paragraph should be removed because it refers solely to the results of another study that has not been yet published.

**This sentence was removed.**

3. Page 2, lines 14-15. Please clarify the last sentence of the paragraph, for example as follows: “The NCCN can be reliably estimated using time-averaged, size-resolved activation efficiencies without accounting for the temporal variations.”

**This sentence was revised as suggested.**

4. Page 2, lines 21-23. The sentence starting with “Comparison of the...” needs rewording, for example as follows: “Closure studies where measured and predicted CCN concentrations are compared provide a test for quantitative understanding of CCN activation properties, and to parameterization schemes used in large-scale models in particular.”

**This sentence was revised as suggested.**

5. Page 2, lines 27-29, second and third sentences of the paragraph. These sentences need also rewording. If I understood right, the authors want to express the fact that measurements of aerosol size distribution and chemical composition can be used to predict CCN concentrations reliably. There is no need to use vague and too general expression "...are the two most important aerosol properties that can be measured...". Please re-word the sentences so that the underlying thought becomes clear.

**The sentence was revised as 'The measurements of aerosol size distribution and chemical composition are often utilized to predict the  $N_{CCN}$ '**

6. Page 2, lines 30-31. Please explain the concepts PM1 and PM10. Also, please do not use expression "such" here because it is not clear to which word it refers to in this sentence.

**This sentence was revised as 'Bulk chemical composition of aerosol particles with diameter smaller than 10 or 1  $\mu\text{m}$  from filter sampling is often used in closure studies (Bougiatioti et al., 2009),...'**

7. Page 3, lines 5-6, first sentence of the paragraph. I disagree with the notion that measurements of chemical composition would be necessarily time-consuming (take AMS studies, for example) but detailed chemical analysis of compounds present in the aerosol phase certainly is. Please explain in more detailed fashion why direct measurements of CCN activation properties are needed. Also, in its current form, the sentence structure should be changed so that the sentence starts with the clause "Direct and detailed measurement techniques...".

**We removed this vague sentence in the text.**

8. Page 3, lines 14-21. The contents of this paragraph overlap partially with those of the second paragraph of Introduction. Please check and correct this.

**We rewrote the paragraphs as suggested. Thanks.**

9. Page 3, lines 20-21. Please explain the concept of mixing state.

**This sentence was revised as "Mixing state indicating the heterogeneity of the chemical composition of a particle distribution also needs to be accounted for (Anttila, 2010; Medina et al., 2007)."**

10. Pages 3-4, lines 30-32 and 1-4, respectively. The authors should consider of omitting this paragraph because, in my opinion, it does not contain any useful information that is relevant for this study. After all, the authors investigate CCN activation properties of ambient aerosol, not cloud formation or cloud microphysics.

**This paragraph was deleted from the text.**

11. Section 2, pages 4 and 5. I recommend slight re-organization and some re-writing to make the section more accessible. First, the second paragraph is not needed as equations (1) and (2) can

*be used without the Koehler theory. Also, the critical supersaturation is not (necessarily) determined by Koehler theory and not even by size and composition alone, because e.g. particle morphology may play a role as well. Second, I'd split the section into two sub-sections so that the cases with internal (fourth paragraph) and external (fifth and sixth paragraphs) are in separate sections. Third, using the critical dry diameter of NaCl particles to illustrate the minimum diameter needed for activation is problematic because, in principle, it is possible that the atmosphere contain substances which are even more hygroscopic than NaCl and/or depress the droplet surface tension so that the corresponding particles are even more CCN active than NaCl particles. Instead, the authors could introduce, for example, an operationally defined diameter  $D_{min}$  which is the minimum diameter needed for activation (within the experimental uncertainties).*

**We revised the paragraph as suggested. We used  $D_{HSP}$  to represent the critical dry diameter for highly soluble particles.**

12. Section 2, pages 4 and 5. Please use the  $N_{CCN, Cal}$  consistently when presenting the calculations based on equation (2) in the manuscript. Also,  $N_{CCN}$  is now defined twice: through equation (1) and eq. (5). Please correct this.

**We corrected these equations. Different subscription for  $N_{CCN}$  are added in the text.**

13. Page 5, lines 29-30. Would "has been undergoing" be the right expression instead of "underwent"? Also, "has been causing", not "caused"?

**These two sentences were revised as suggested.**

14. Page 5, line 30. "Pollution episodes", not "pollutions"? Are there any studies or documents to support the statement? Please refer, if available.

**"Pollution" was changed into "pollution episodes". A citation was added.**

15. Page 6, lines 4 and 5. Should be: "The field study... WAS carried out...". Also, "...focusing on aerosol CCN and optical properties...", for example.

**This sentence was revised as "The field study, focusing on aerosol, CCN activation properties and aerosol optical properties, was carried out in January 2010."**

16. Section 3.1, pages 5 and 6. Please provide a more detailed description of the immediate surroundings of the measurement site and on the local meteorological conditions.

**More information was added in the revised manuscript. A detailed description of the measurement site and the meteorological conditions was available in the paper of Xu et al. (2011) in this issue.**

17. Section 3.2, page 7, lines 11-12. Please provide a reference for the applied form of the Koehler equation.

**The citation (Pruppacher and Klett, 1997) was added in the manuscript.**

18. Section 4.1, page 8. Please insert a figure showing the time series of CN and bulk CCN concentrations.

**The figure was provided in the revised manuscript as Fig. 4**

19. Section 4.1, page 8, lines 12-13. The authors claim that the NCCN depended on the meteorology during the measurement without giving any evidence. Do the authors have any evidence to substantiate this claim? I'd recommend that the authors elaborate further the connection between NCCN and meteorology.

**The dependence of CN and CCN concentrations on wind was shown in the new added figure (Fig. 4).**

20. Section 4.1, page 8, lines 28-30. Please provide explicit calculations for the error introduced in NCCN when using a fixed  $D_m$  at various supersaturations.

**We rewrote the sentence as "The predicted  $N_{CCN}$  with a fixed critical dry diameter inferred from the measurement result in an average relative deviation of 69%, and the bias of -26% (see the definition in caption of Fig. 9)."**

21. Section 4.2, page 9, line 3. "shows" instead of "is".

**This sentence was revised as suggested.**

22. Section 4.2, page 9, lines 4-9. The two sentences are irrelevant in this context and the topic is also covered in Section 2. Please remove the overlap.

**These sentences were removed in the text as suggested.**

23. Section 4.2, page 9, lines 13 and 14. In this sentence, three diameters are given but only two supersaturations are mentioned. Please correct.

**This sentence was revised as "Particles larger than 75 nm are mostly activated at supersaturations above 0.36%."**

24. Section 4.2, page 9, third and fourth paragraphs. The discussion regarding the results presented in Figure 6 could be extended by reporting the diameter at which the activation efficiency is 50%,  $D_{50}$ , for different supersaturations. Also, it would be interesting to see how well  $D_{50}$  compares with  $D_m$ .

**$D_m$  is calculated based on the measured aerosol number size distribution and CCN number concentration. The aerosol number concentration for particles larger than  $D_m$  equals the measured CCN number concentration. This inferred cut-off diameter are in the ranges of 190-280, 160-260, 95-180, 65-120 and 50-100 nm for supersaturations of 0.056, 0.083, 0.17, 0.35 and 0.7%, with their mean values 230.1, 198.4, 128.4, 86.4 and 69.2 nm, respectively.**

**$D_{50}$  is the diameter where the activation ratio is 50% for the fitted activation ratio curve**

using the measurements at the 6 diameters. The fitting results for D50 depend on the choice of particle diameters, the number of selected diameters, and other uncertainties. The activation curve is not ideally sigmoidal. This paper did not give the fitted D50, but provided an estimation of D50. More than 50% of the particles larger than 200, 170, 90, 70, and 45 nm are activated at supersaturations of 0.058, 0.085, 0.18, 0.36 and 0.72%, respectively. The D50 is generally smaller.

25. Section 4.2, page 9, line 17. "shows" instead of "is".

**This sentence was revised as suggested.**

26. Section 4.2, page 9, line 22. "...activate at all measured supersaturations.", instead of "...are activated at all of the measured..."

**This sentence was revised as suggested.**

27. Section 4.2, page 9, lines 24-26. Please insert ", respectively" to the end of the sentence.

**This sentence was revised as suggested.**

28. Section 4.3, I'd split the section into two subsections so that the last five paragraphs would form a subsection that deals with the sensitivity studies.

**This section was split into two subsections.**

29. Section 4.3, page 10, line 18. "showed", not "shows".

**This sentence was revised as suggested.**

30. Section 4.3, page 10, third and fourth paragraphs. Please describe the fitting it in a more detailed fashion. The sentence starting with "If an activation curve is measured..." is vague, please clarify.

**This sentence was revised as " $N_{CCN}$  at the effective supersaturations of the size-resolved measurements are achieved by fitting the CCN spectra with Eq. (3)"**

31. Page 10. Only two equations are presented before Eq. 5 so it should be Eq. 3.

**This was corrected in the manuscript.**

32. Page 11, line 2. "measurements", not "measurement".

**This sentence was revised.**

33. Page 11, lines 22-28. The description of the sensitivity cases needs to be clarified. At least, provide explicit equations showing how the CCN concentrations are calculated in the three

*sensitivity studies described in this paragraph.*

**We added six equations to describe how we calculated the CCN number concentrations.**

34. *Page 12, lines 7 and 8. Please clarify the sentence starting with “The calculated NCCN for each supersaturation,...”.*

**We removed this vague sentence.**

35. *Page 12, line 12. Please insert “...,respectively” to the end of the sentence.*

**This sentence was revised as suggested.**

36. *Page 12, line 28. “Provide”, not “gain”.*

**This sentence was revised as suggested.**

37. *Page 13, lines 8-23. This paragraph does not summarize the results of the study but could be classified under the title “Discussion”. Therefore, re-organize the section and add a separate section for discussion.*

**We changed the title of section 5 into “Summary and Discussion”.**

38. *Page 13, lines 9 and 10. The sentence “The cloud property...” contains vague expressions such as “The cloud property”, “thermodynamic processes”, and “microphysical processes”. Please clarify.*

**This sentence was removed in the text.**

39. *Page 13, lines 29-30. How the soluble fraction was calculated? Please describe the method briefly.*

**This sentence was revised as “Soluble fractions of more than 83%, calculated based on the Köhler Theory with assumed aerosol composition of ammonium sulfate and an insoluble core, are needed to activate these particles.”**

40. *Page 13, lines 31-32. Please provide evidence for this claim.*

**We added two citations for this sentence.**

41. *Page 14, lines 13-21. Please express the finding that the variation in the size-resolved activation ratio did not have large impact on NCCN in a more condensed manner. In particular, the sentence starting with “The activation ability of aerosol...” is not relevant in this context.*

**This part was revised accordingly.**

42. Page 14, lines 18-19. The sentence starting with “This average activation property...” contains vague expressions (“average activation property” and “well predicted aerosol size distribution”). Please revise.

**The sentence was revised as “This average size-resolved activation ratios during this campaign and measured aerosol size distribution can be used to predict  $N_{CCN}$  successfully.”**

43. Table 1, text. Add word “particles” to the end of the sentence.

**This was revised as suggested.**