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Interactive comment on “Aerosol hygroscopicity and CCN activity obtained from a combination analysis based on size-resolved CCN and aerosol chemical composition observations during the AC³Exp13 campaign” by F. Zhang et al.

F. Zhang et al.

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General comments: The paper “Aerosol hygroscopicity and CCN activity obtained from a combination analysis based on size-resolved CCN and aerosol chemical composition observations during the AC3Exp13 campaign” by F. Zhang, et al. 2014 provide a new data set in the southeast area of Beijing, China. The authors present useful observations on chemical composition, size distribution or mixing state of aerosol,

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and the results are consistent with the previous studies. However, the data analysis provides only qualitative explanation, which is known by previous studies. The paper titled “a combination analysis based on size-resolved CCN and aerosol chemical composition observations”, however authors did not clearly demonstrate the advantage of using size-resolved measurements. Authors may consider providing more analysis/discussion which separates aerosol size effect with its chemical composition effect by size-resolved measurements.

Re: The reviewer made a good point. By using size-resolved CCN data, CCN activity can be investigated for aerosol particles with different size range. However, we cannot separate aerosol size effect with its chemical composition effect only based on size-resolved CCN measurements. To do this, size-resolved chemical composition data (measured by high resolution AMS) are required at least. We feel a little pity for that we are not able to do this in this paper due to the lack of such measurements and dataset (size-resolved chemical composition data) during the campaign. That's just why we provide more qualitative explanations on impacts of chemical composition on CCN activity. However, based on the limited data in the region, we have shown some interesting results and highlighted the importance of aerosols chemical composition on determining the activation properties of aerosol particles, which was not taken into account in previous/early literatures conducted in the North China Plain. Recently, we are planning size-resolved chemical composition measurements combined with size-resolved CCN measurements in the region (probably in Beijing). Thus, some more quantitative analysis and results would be expected in our future papers.

In addition, the closure study shows that NPO test has a similar or may even better agreement between CCN_estimation and CCN_measurement than PO closure test. That is odd to reviewer.

Re: Thanks a lot for reviewer pointing this and thus let us to check and revise the data again, including multiple charge correction and transfer function which are applied for each CN size distribution spectrum as well as CCN efficiency spectrum in the revi-

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sion. Then, the CCN efficiency spectrum fitted by CDF method as well as the three parameters (MAF, D_a and σ) is revised. Finally, we used the revised activation ratio curve and calibrated CCN number concentrations to do the CCN closure test. Fig 5 and Fig.6 show the revised results of PO and NPO closure tests. If we just look at the closure results at $SS > 0.2\%$, although the slopes for both PO and NPO closure tests don't show significant differences, the correlations (R^2) for NPO test is much worse than PO test at a given SS. The lower correlation in NPO test just indicated that about 30%-40% uncertainties in NCCN prediction are associated with the changes of particle composition.

At lower SS, particles will active at larger size and it is supposed their chemical composition will be closer to ACSM measurement. Why lower correlation exhibited at lower SS?

Re: One reason for the slight worse closure between measured and estimated CCN number concentrations at lower SS lies in that size-resolved activation ratios exhibit a larger variability at low SS than that at higher SS. Also, compared to the activated CCN number concentrations at high SS, the NCCN is with a less amounts at low SS, which would lead to larger uncertainties in CCN prediction.

Specific comments: Page 6, Line 3-6: the field campaign was mainly conducted in June and July, 2013. However, the results are presents in different periods without clearly stating the criteria for selection. For example, figure 1 is for 07/07-07/21/2013, figure 3 is for 07/07- 07/19/2013, and figure 9 is for 06/19-06/24/2013. Please clarify.

Re: In the campaign, aerosol particle size distribution (10-700 nm) and bulk CCN activation were measured from 1 June to 25 June 2013. Size-resolved CCN was measured from 7 July to 21 July 2013. Aerosol chemical composition was measured from 31 May to 30 June 2013. Therefore, the CCN efficiency spectra (Fig. 1) are derived from the size-resolved CCN observation from 7 July to 21 July 2013. The aerosol particle size distribution data measured from 1 June to 25 June 2013 combing with the derived CCN

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efficiency spectra (Fig. 1) is used for NPO CCN closure test. We have clarified this in the revised version (see Page 7, lines 5-17). In Fig. 9, an example (or case) for temporal variations of bulk AR at SS of 0.5%, derived κ_{chem} and NCN from a few days (19-24 June) during the observed periods is given.

Page 6, Line 17: what is the inlet RH for CCN?

Re: the inlet RH for CCN is <30%, which has been included (see Page 7, line 21).

Page 7, Line 10: Need more information on BC analysis, because BC probably is the main source for externally mixed inactive CCN.

Re: The campaign averaged mass concentrations of BC are $\sim 4.2 \mu\text{g m}^{-3}$, and the averaged mass fractions are about 6%, with maximum of 18% and minimum of 2%. However, after doing the revision and analyzing the data more carefully, we have derived more confident explanations and suppose on the composition and mixing state of aerosol particles during the campaign. The revised results suggest the aerosols are with fewer portions of externally mixed particles than that previously derived.

Page 7, Line 20: the criteria of clean day and pollution day is little weak. May consider include gas phase measurement and meteorology data, such as wind directions.

Re: In the revision, we have examined the synchronal winds to classify the data as background and pollution conditions. Basically, the polluted conditions are with $\text{NCN} > 15000 \text{ cm}^{-3}$ when the airflow came from southeast/east and the background cases are with $\text{NCN} < 15000 \text{ cm}^{-3}$ always associated with the winds from west or north-west.

Page 8, line 2, page 12, section 4.1.1, and Table 1: It is expected that D_{cut} is larger than D_a . But at $\text{SS} = 0.079\%$ in table 1, D_{a_POL} is larger than $D_{\text{cut_POL}}$. Why?

Re: thanks a lot for the reviewer's careful check. We just checked and revised the data again. In the revised version, the parameters have been recalculated in Table 1, and also the standard deviations of some parameters, e.g. D_{a_POL} , $D_{\text{cut_POL}}$, K_{a_POL} ,

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Ka_BG, etc., have also been added in Table 1.

Page 11, line 11-14: it is not true. With MAF only, you can not characterize aerosols with homogeneous composition and a core-shell structure aerosols may has MAF = 1.

Re: Yes, the reviewer is right. We just corrected this point in the revised version (see Page 15, line 23; and Page 16, lines 1-2).

Page 12, Line 1-3: With increasing SS, the aerosol particle critical activation size changes too. With ACSM measurement, authors can only get bulk chemical composition and chemical composition varies with aerosol size. The conclusion maybe true, but cannot draw from the differences of AR. In addition, Kuwata et al. paper was published in 2007.

Re: In a new version of the paper, we have revised this.

Page 12, section 4.1.2: the discussion here is not consistent with figure 1. In figure 1, the average AR at 0.109% is around 0.6 for POL and 0.8 for BK cases. The substantial portion of externally mixed CCN-inactive particles is around 20-40%, not 14-22%. The significant increase of MAF at POL case suggests that there is a large variability in mixing state at different size particles. What do authors mean by “a higher sensitivity of particle MAF dependence on SS”?

Re: “a higher sensitivity of particle MAF dependence on SS” just means that MAF would be very different for different SS. However, the point has been revised in the paper.

Page 14: Line 23: for particles at size range of 30-60 nm, kappa for polluted particles is high than clean aerosols. Why? Is clean case supposed to have more aged aerosols? Or does that mean the fresh biomass burning aerosol has higher kappa?

Re: the reviewer makes a good point. Although we cannot figure out accurately and definitely why the kappa for polluted particles is higher than clean aerosols, it's really interesting. For background days, larger particles were on average more hygroscopic

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than smaller particles. This result is consistent with most of field observed results. However, κa at polluted cases exhibited relatively flat trend. It didn't exhibited significant increase but with larger error bars around a given D_p , suggesting the complex and diversities of particle compositions and their mixing state at polluted cases. In this case, larger particles are even less hygroscopic than the smaller particles. We think one of the possible reason for the changes of κa at polluted cases may be due to the high organics freshly emitted from biomass burning, which would coat on those larger particles and then lead the particles with less hygroscopic. Overall, κ for polluted aerosols are about 20% lower than that of clean aerosols for particles in accumulation size range; but for these particles in nucleation or Aitken size range, κa for polluted particles is slightly higher than that at background cases. Based on laboratory experiment, Petters et al., (2009) examined the hygroscopic properties of particles freshly emitted from biomass burning. They found that κ was a function of particle size, with 250 nm particles being generally weakly hygroscopic and sub-100 nm particles being more hygroscopic. During the campaign at Xianghe, the biomass burning aerosols are the lead particles for the selected polluted cases. The laboratory results, to some extent, can thus explain our field measurements. But further investigations including laboratory experiments and field measurements of size-resolved chemical composition are needed to confirm and clarify this.

Figure 2, Is the data presented here are average of whole campaign? Please add error bar or uncertainty estimation.

Re: Yes, the data presented in Figure 2 are average of whole campaign during background and polluted conditions. The error bars (with one standard deviation) have been added in Figure 2.

Interactive comment on Atmos. Chem. Phys. Discuss., 14, 14889, 2014.

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