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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 AC<sup>3</sup>Exp13 campaign” by F. Zhang et al.***

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Anonymous Referee #2 Received and published: 31 July 2014

This manuscript describes a CCN activity in highly polluted location as observed at the Xianghe Atmospheric Observatory, China, and presents the analysis of the aerosol size and chemical composition effects on the CCN activity. While the data set is interesting and could provide new information, especially considering the location, the analysis and discussion is lacking clarity and thoroughness.

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General comments The fact that MAF is always significantly below 1 raises some questions, it well may be due to externally mixed hydrophobic particles as explained by the authors; however, this has to be proven. First of all, it should be shown or at least checked if MAF is equal to 1 for the calibration particles (e.g. ammonium sulphate) at high number concentrations ( $\sim 25000$ ). This would rule out effects of water depletion and system setup. Once this is proven, an evidence or indication that externally mixed fresh black carbon particles were present during these observations should be provided. Actually, black carbon is not even mentioned here, some vague explanation of "externally mixed particle composition" ((page 14901, L 19) is presented, which, to my opinion, is not enough to prove such a significant effect.

Re: the reviewer made a good point regarding our paper's revision. The comments from the reviewer just 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 revision. Then, the CDF fit curve as well as the three parameters (MAF,  $D_a$  and  $\sigma$ ) is revised (see Fig. 1 and Table 1). Because the effect of water depletion is so small that it can be neglected. And also MAF is actually equal to 1 for pure ammonium sulphate particles (0.05mol L<sup>-1</sup>). Furthermore, the observations have indicated that particles can be able to activate to CCN completely when  $D_p > 300$  nm even at  $SS = 0.08\%$ . Therefore, a smaller portion ( $1 - \text{MAF}$ , 2-6%) (in Fig 1) is just caused by an error of CDF fit method which will lead to lower MAF than it should be.

Also, the paragraph 4.2 on  $\kappa$  dependency on  $D_p$  requires more details. What is  $D_p$  in this case? How the dependency is derived? I assume it is the same  $D_p$  as in Figure 4. If data presented in figure 2 are averages of the data shown in Figure 4, then I see some inconsistency, which has to be verified:  $\kappa_{\text{cut\_POL}}$  at  $D_p = 163\text{nm}$  would average out to  $\sim 0.46$ , which contradicts the number presented in figure 2 ( $< 0.4$ ), but would be in line with other kappa's at different sizes and there would be no more reduction in kappa with  $D_p$ . . . I may be wrong, but this should be checked. In any case, one should be

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cautious by basing the conclusion on one point, which seems to be outlier. However, if you trust this point and think that it is outside a kappa uncertainty range, then it should be shown and proven in this paragraph.

Re: The  $D_p$  in the previous version of Figure 2 is  $D_a$  and  $D_{cut}$ , which we used to derive the  $\kappa_a$  and  $\kappa_{cut}$ . Thus, in Figure 2, that's just  $\kappa_a$  dependency on  $D_a$ , and  $\kappa_{cut}$  dependency on  $D_{cut}$ . We have clarified this in the revised paper. In the revised paper, Figure 4 showed the PDF of  $\kappa_a$ . Therefore, the PDF of  $\kappa_a$  in Figure 4 should be consistent with the  $\kappa_a$  showed in Figure 2. I

The potential of information on chemical particle composition has not been fully used here.  $\kappa_{chem}$  differs from other kappa's derived in this paper, which is attributed to incorrect kappa used for the organics, then why not to try different and correct kappa's? Why not to link organic kappa with f44, especially that authors indicate AF dependency on f44 in polluted cases.

Re: In the revised paper, we used a linear function between  $\kappa_{org}$  and f44 derived by Mei et al., (2013) to estimate the  $\kappa_{org}$  in our study. The formula is written as  $\kappa_{Org} = 2.10 \times f44 - 0.11$  (Mei et al., ACP, 2013). By applying the function, calculated  $\kappa_{org}$  is  $0.115 \pm 0.019$  during the observed period. Then we used the estimated  $\kappa_{org}$  to calculate  $\kappa_{chem}$ .

Figure 5 indicates two modes, at least for the background  $\kappa$ , thus two values at  $\sim 0.35$  and  $0.45$  would be consistent with figure 2, which shows different kappa at different sizes. Could you check if this would be consistent with particle size distribution: if lower  $\kappa_{chem}$  occurs for the events dominated by smaller particle mode and vice versa – larger  $\kappa_{chem}$  for particle distributions dominated by larger particles (size distributions could be obtained from concurrent smps measurements).

Re: the reviewer's suppose is right. However, we just looked the data and plotted the time series of  $\kappa_{chem}$  and NCN for Nuc., Ait and ACC modes particles (see Fig 1s). No obvious correlation between  $\kappa_{chem}$  and particles size was observed. We think that

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two reasons can well explain this: Firstly, it can be seen from the Figure 2,  $K_{chem}$  increased with the  $D_p$  at background conditions, while they didn't change much along with the particles size under polluted events (with high NCN). In other words, larger  $K_{chem}$  is not always dominated by larger particles in such cases. Second,  $K_{chem}$  estimated in this paper are a bulk hygroscopicity parameters, each  $K_{chem}$  derived at every observed time indicated overall averaged hygroscopicity of the particles with particle size  $<1000$  nm (PM<sub>1.0</sub>). Thus, the relationship between  $K_{chem}$  and particle size is complex. Size-resolved chemical composition data can well address this issue. As we stated earlier, 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.

The whole paragraph on chemical composition (4.6) is very qualitative, it requires more quantitative approach: I suggest the authors provide scatter plots with proper statistics, or at least quantify "high correlation", "well correlated", "moderate correlation". Time trends are not enough for proper analysis.

Re: In the revised version, we have included a scatter plot (Fig. 8) to address the correlation of  $K_{chem}$ ,  $f_{44}$  and AR under background and polluted conditions.

Specific comments: P14895 L3 and L13: RH range should be specified.

Re: revised (see Page 8, lines 11-12).

P14896 L1: More details on ACSM operation should be provided, e.g. collection efficiency applied in this study; information on the inlet: particles were or were not dried; calibrations.

Re: Some more details regarding the ACSM measurements have been included. (see Page 8, lines 19-25; Page 9, lines 1-15).

P14896 L11: Specify  $N_{cn}$ , what were the lowest and highest size limits?

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Re: in this study, we used a SMPS to measure aerosol particle size distribution with size range of 10-700 nm. Thus, the NCN is aerosol number concentrations with particle size range of 10-700 nm.

P14899 L14: Why sea salt and dust are expected to be negligible for the size range of < 500 nm? Give some more information, discussion, references.

Re: revised. (see pages 13, lines 11-15)

P14899 L15: Define size (mobility diameter, vacuum aerodynamic?) why 10-500nm if ACSM measures 50-1000 nm, I guess it is in  $D_m$ , but it should be specified.

Re: revised. Here the 10-500 nm is just a written mistake, it has been corrected. (see page 13, line 14).

P14903 L17-18: This sentence is confusing – reduction due to good activity?

Re: revised.

P14905 L17: To me the range is 0.15-0.52 as shown in Figure 5. Figure 5: Include a description of the red line at 0.3 and fittings into the caption.

Re: Yes, the reviewer is right. But,  $K_{chem}$  estimated in this paper are a bulk hygroscopicity parameters, each  $K_{chem}$  derived at every observed time indicated overall averaged hygroscopicity of the particles with particle size <1000 nm (PM<sub>1.0</sub>). But  $K_a$  and  $K_{cut}$  derived from size-resolved CCN measurement refer to the particle hygroscopicity parameter at a given  $D_p$ . The comparison between  $K_{chem}$  and  $K_{CCN}$  ( $K_a$  and  $K_{cut}$ ), to some extent, is thus not such reasonable. One probably can compare the campaign mean  $K_a$  averaged by all the  $K_a$  at different size diameter with the mean  $K_{chem}$ . In the revised paper, we removed the PDF of  $K_{chem}$  in section 4.4. In section 4.6, we will focus on examining the relationship between bulk AR,  $f_{44}$  and  $K_{chem}$ , trying to understand the impact of chemical composition and particle size on aerosol hygroscopicity and CCN activity.

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P14908 L8, L11, L25, L24 and others: define goodness with R or R2.

Re: revised. (see Pages 21-22, Fig. 8)

P14910 L22-23: this sentence is confusing- smaller particles were more aged and coated than larger particles? It requires better explanation.

Re: revised. (see Pages 18-19). For these particles in nucleation or Aitken size range,  $\kappa_a$  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  $\kappa$  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.

P14910 L13: Provide a reference for the typical atmospheric SS of 0.4 %.

Re: in the revised version, such saying has been removed.

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

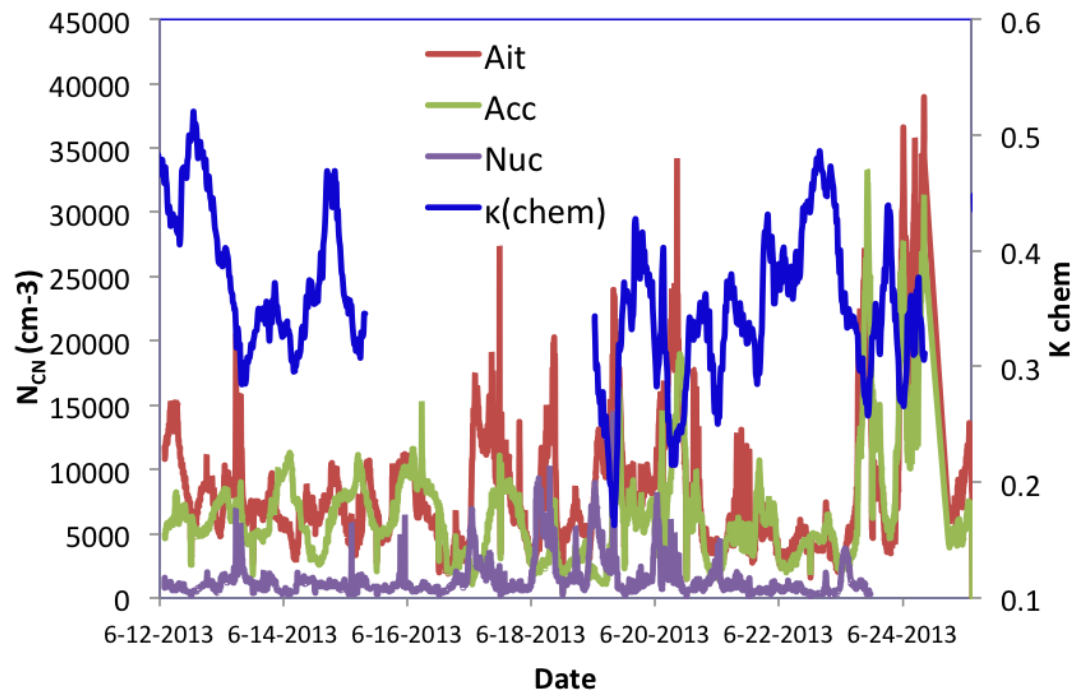
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**Fig. 1.** Fig 1s Time series of Kchem and NCN for Nuc., Ait and ACC modes particles

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