

Anonymous Referee #1

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General comments:

The paper “Challenges of parameterizing CCN due to changes in particle physicochemical properties: implications from observations at a suburban site in China” by F. Zhang, et al. 2015 provide more surface measurement dataset in China. The authors present useful observations on CCN activation properties and the results are consistent with the previous studies. However, the paper is lack of the discussion on what affects CCN properties, such as distinguishing the effect of chemical composition from size distribution or mixing state of aerosol. Thus, the results and conclusion are not new and expected from previous studies. Authors may consider providing more analysis/discussion which separates aerosol size effect with its chemical composition effect.

Re: The reviewer made a good point. In the previous version of this paper, we mainly focus on examining the influence of extremely high volume fraction of organics on CCN prediction in polluted area of China, to our knowledge, which have not been examined in the region. However, in the revision, the sensitivity of both the volume fraction organics (x_{org}) as well as oxidation level (using f_{44} , the fraction of m/z 44 in total organics, as an indicator) of organics on estimating N_{CCN} is examined (see Section 4.4).

But as proposed by the reviewer, to separate aerosol size effect with its chemical composition effect is the key point to study the relative importance of size and composition impact on CCN activity. We are now drafting another paper and mainly concern the impacts of chemical composition and mixing state on CCN activity by using size-resolved chemical composition data measured by AMS as well as size-resolved CCN data measured by SMPS-CCNc. In the paper, we will distinguish the effect of chemical composition from size distribution or mixing state of aerosol.

In addition, the figure 1 shows the maximum activation fraction is around 0.9 for both sites, but in the paper by Zhang et al. 2014, the table 1 shows that MAF is larger than 0.94%. Please explain the inconsistency.

Re: there are two reasons can explain the inconsistency: firstly, the CCN spectra here are plotted by the observed campaign averaged values, but in the paper published previously, we plotted the fitted curve by using CDF method at both background and polluted cases; secondly, a further calibration and correction method was applied to the data, which forced the AR values to 1 if it is higher than 1 when the $D_p > 300$ nm, and thus leading to slight lower MAF. This treatment we think is more reasonable for that the large particles should be activated but the AR would never be larger than 1.

Specific comments:

P16143, Line 10-15, P16154, section 4.3.2: When author mentioned PSD effect was examined, how does author exclude the chemical effect/mixing state effects? If you cannot separate those effects, it is impossible to examine the influence of the PSD on Nccn estimation.

Re: This section is with the aim to examine the impact of variation of PSD on the CCN number concentration. Thus, the campaign averaged PSD (not changed along with the time) is used for calculating CCN number concentrations by multiplying the time dependent CCN spectra (changing of CCN spectra can indicate the variation of chemical composition and mixing state).

P16146, section 2, line 14: what do you mean “relatively little”? Is it occasionally local interference? If so, was the data screened? What percentage of the data is screened? Do they happen in the same pattern? Such as all in the morning?

Re: here it means that the site is with very little influence from the local vehicles and industries. The data points, which are probably due to the local vehicle or industrial emissions, account for <1% during the campaign. Here, we focus on the chemical and physical impacts on the CCN activity but the variations of CCN and aerosol particles, thus we didn't distinguish the data as like local influenced or regional background. However, the invalid data, which are due to the instrumental problems,

were removed. We do observed diurnal cycles of N_{CN} during the observed periods showing high level of N_{CN} usually presenting at about 11:00-12:00, which we think was closely related to the new particle formation events. We are drafting another paper mainly concerning the impacts of new particle formation on CCN activity.

P16148, line 7: what the mass concentration from ACSM? Is the BC concentration significant comparing the rest of chemical compositions?

Re: the campaign averaged mass concentration of PM1 by ACSM is $31.6 \mu\text{g m}^{-3}$. The mass concentration of BC, which was measured by a seven-wavelength aethalometer (Model AE31), was $\sim 2.5 \mu\text{g m}^{-3}$ during the campaign. Thus, compared with the other chemical composition measured by ACSM, BC mass concentration is much lower.

P16148, line 16-20, What is the percentage of valid data? Is there a time pattern for the invalid data appearance?

Re: the valid data account for about 80% during the whole campaign. We didn't observe an apparent time pattern for the invalid data.

P16149-150, section 3, it is almost identical with the paper published in Zhang et al. 2014. Please consider remove it and refer to the paper.

Re: thanks for the comments, some corrections have been made in the section and the paper Zhang et al., 2014 is referred to.

P16151, line 7, because the maximum activation fraction is around 90%, should the cut-off diameter at AR=50% represent the critical activation

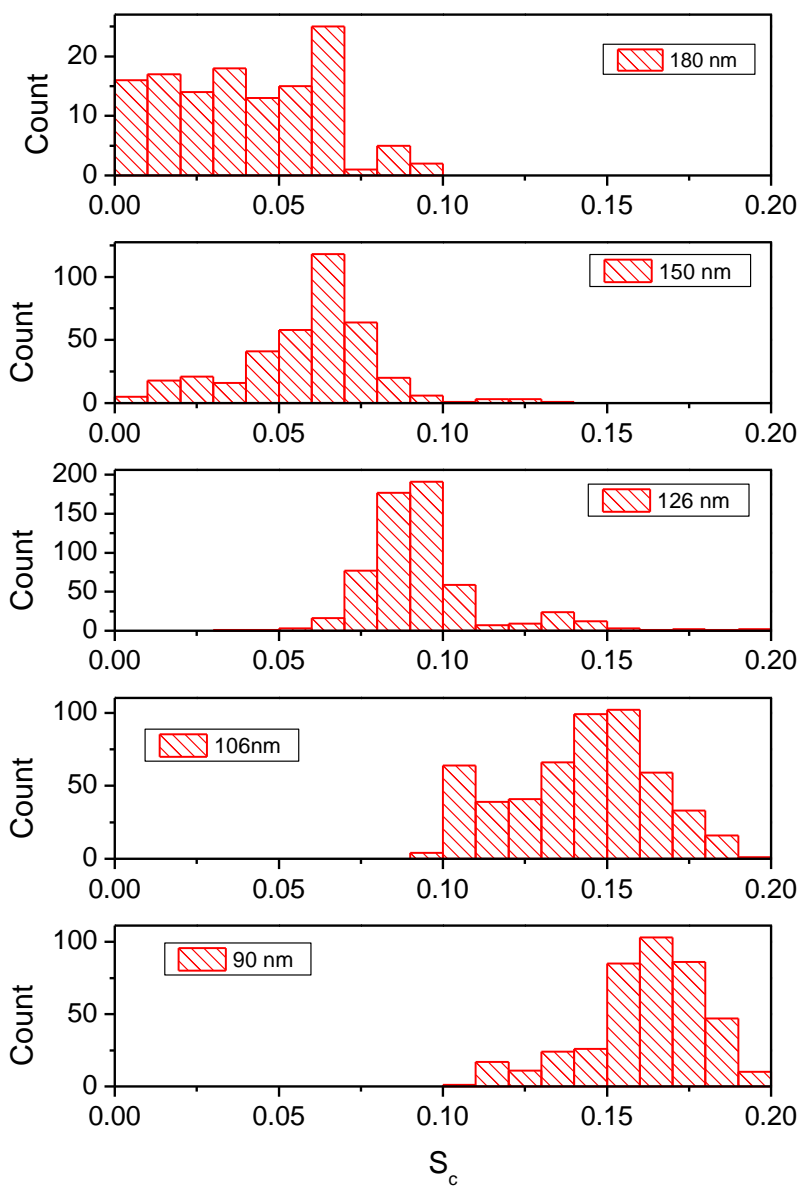
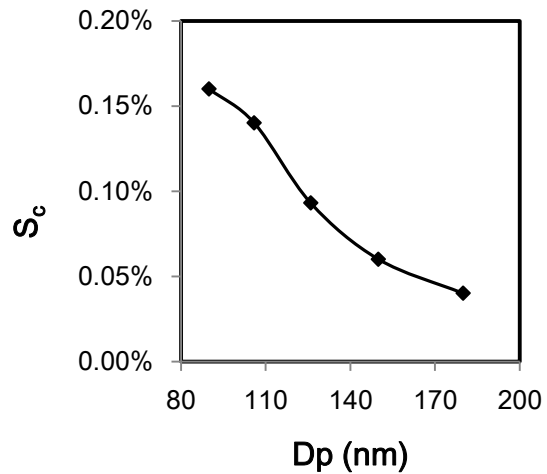
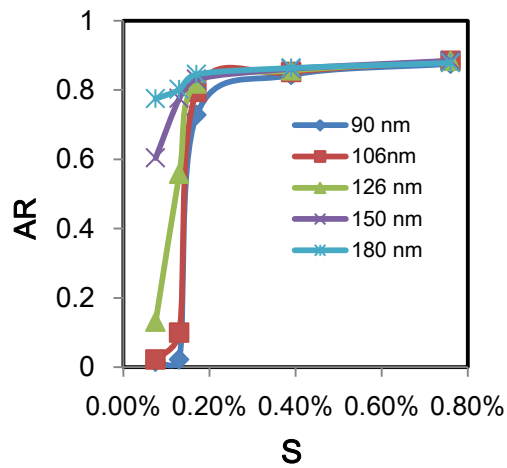
size?

Re: here, we just refer to an ideal case, when all CCN-active particles have the same composition and size, a steep change in AR from 0 to 1 would be observed as D_p reached D_{cut} when AR=50%. However, just as proposed by the reviewer, the actual observed maximum activation fraction (MAF) is around 90%. At the real cases, the D_{cut} is usually defined as when AR=MAF/2.

P16151, line 10-15, To discuss the heterogeneous of hygroscopicity of aerosol, it is better to analyze data using supersaturation vs activation fraction. Here is an ex-ample of such discussion:

<http://www.atmos-chem-phys.net/13/12155/2013/acp-13-12155-2013-supplement.pdf>

Re: thank you very much for the suggestion and giving the example. We just plot data using supersaturation vs activation according to the method in the pdf file. The probability distribution functions (PDFs) of the S_c for the particles with D_p of 90 nm, 106 nm, 126 nm, 150 nm and 180 nm are showed in the following figure. PDFs for each D_p have a wider distribution, suggesting heterogeneous of hygroscopicity of aerosol. But considering the major concern of this paper, we decide not to address this in this paper.



P16152, line 4-5, what is the height of the back-trajectory running at?

Re: the height is 10 m from the ground.

P16154, section 4.3.2, it is well know that the size effect of aerosol on CCN concentration. If author wants to discuss that, please add more qualitative analysis.

Re: according to the comments from the reviewers, this section was removed in the revised manuscript.