

Dear anonymous reviewers,

Thank you very much for your comments on our manuscript [acp-2014-269]. Your comments and suggestions are valuable and very helpful for improving our manuscript. Based on your comments and suggestions, we carefully revised the manuscript, and the point-to-point responses to your comments and suggestions are listed below.

Thank you once again for your time and consideration.

Responds to the comments:

Note: All the revisions are based on the previously submitted manuscript in word format which corresponds to the ACPD. For the modified portions, we provide the page and line numbers of the ACPD format before modification.

Anonymous Referee #1

There is short of the impacts of heave foggy event on CCN, and more studies about this issue are needed in the subsequent works of the authors.

We thank you for your kind comments and good suggestions. In this study, no heave foggy case was observed and this disables us to explore its impact on CCN. However, a foggy-hazy case was observed which could partly act as references. We will pay more attention to the heave foggy

case and its effect on CCN in the future.

It seems that it is not quite often to use CN larger than a certain size (e.g. CN larger than 80 nm and 100 nm in this paper) to get the activated fraction.

We thank you for your comments. Aerosol CCN activated fraction is usually calculated by using the whole CN size and we have presented this method in our submitted manuscript. However, CN larger than a certain size (e.g. CN larger than 80 nm and 100 nm in this paper) can still be used as references for further analysis, because smaller aerosol particles (e.g. particles smaller than 80 nm) are not activated at 0.2% SS. It will help to reduce the uncertainties from the small particle size which contributes little to CCN formation, and help to gain more information about the CCN-active aerosols.

This paper should give more explanation to MPL, such as calibration, data processing.

We thank you for your comments and suggestions. We have added more explanation to MPL in our revised manuscript. Detailed changes are as following:

Page 17003, line 16: add “The description of the retrieval of aerosol parameters by the MPL will be only briefly summarized here as it has

been given by He et al (2006a). The vertical profile of the aerosol extinction coefficient is determined by a near end approach in solving the lidar equation (Fernald, 1984). The PBL height is determined by the MPL lidar at the altitude where a sudden decrease of scattering coefficient occurs (Boers and Eloranta, 1986). The overlap problem must be solved because it can lead to an underestimation of aerosol backscatter and extinction coefficients in the lowest altitudes having the majority of aerosols (He et al., 2006). Outlined by Campbell et al (2002), overlap is typically solved experimentally. The system is set to point horizontally to an averaged data sample with no obscuration, such as the late afternoon, when the atmosphere is well mixed and the aerosol loading is low. The backscattering over the target layer is roughly assumed constant. The similar calibration has been performed before this study.” behind “...Brooks, 2003).”

A lidar ratio plot would help to gain more information about the aerosol
We thank you for your comments and valuable suggestions. We agree with you that a lidar ratio plot would help to gain more information, but the MPL software we are using now is an old version and this disables the derivation of lidar ratio. We will explore the lidar ratio as soon as the software is updated in the future.

The paper should add more address clearly on time-scale of averaging for meteorological factors, CCN, BC and other data.

We thank you for your comments and suggestions. The PBL and vertical extinction coefficient are processed into 1-min averages. The other factors are processed into 1-hour averages.

Figure 1 needs some modification for time labels.

We accept your valuable suggestions and have modified the time labels for Figure 1 as Figure R1

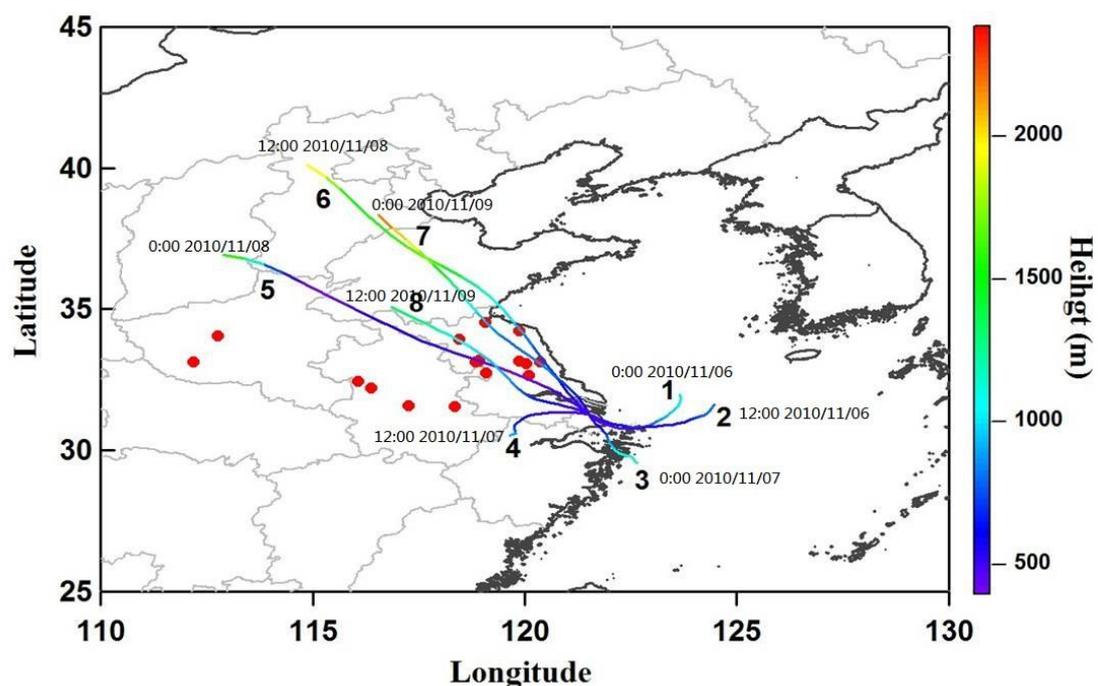


Figure R1 Agricultural fire scattering areas and air mass transport pathways across these regions. All red spots represent biomass burning sites on 7 November measured from MODIS satellite. Starting time (LT) is labeled in the figure.

The authors should give more explanation for the equation 2. More related references should be added and the references style should be adjusted correctly.

We thank you for your comments and suggestions. More explanation and the derivation process for the equation 2 have been introduced in detail by Petters and Kreidenweis (2007) in ACP. Therefore, there is only brief summarization here.

The saturation (i.e. S) over an aqueous droplet can be calculated by:

$$S = a_{\omega} \exp\left(\frac{4\sigma_{s/a} M_{\omega}}{RT\rho_{\omega} D}\right)$$

According to the effect on the water activity of the solution, the hygroscopicity κ is defined as:

$$\frac{1}{a_{\omega}} = 1 + \kappa \frac{V_S}{V_{\omega}}$$

where V_S is the volume of the dry particulate matter and V_{ω} is the volume of the water.

For the equilibrium of a multicomponent system, the total volume of the water is the sum of the water contents due to the individual components, i.e. $V_{\omega} = \sum V_{\omega i}$ by using the ZSR (Zdanovskii, Stokes and Robinson) assumption. The individual $V_{\omega i}$ can be derived for $a_{\omega i} = a_{\omega}$.

$$V_{\omega} = \frac{a_{\omega}}{1 - a_{\omega}} \sum_i \kappa_i V_{s i}$$

The total volume of the system (water+solute) is

$$V_T = \sum_i V_{si} + \sum_i V_{oi} = V_s + V_w$$

The individual dry component volume fractions are defined as $\varepsilon_i = V_{si}/V_s$, then

$$V_T - V_s = \frac{a_w}{1 - a_w} V_s \sum_i \kappa_i \varepsilon_i$$

Defining D_d as the dry diameter, $D_d^3 = 6V_s/\pi$, also $D^3 = 6V_T/\pi$

Then we can derive the equation 2 in ACPD, namely “ κ -Köhle

Anonymous Referee #2

Specific comments:

-The language could be improved by a native English speaker in many parts.

We thank you for your kind comments and good suggestions. We have read the paper very carefully to avoid any grammar errors and mistakes.

-The presented data period is too short to give any general statements of the effects of fog/haze. Please, check and modify this throughout the text.

We thank you for your comments. This study is a case analysis concerning the effect of the fog/haze cases on CCN. We have checked and made a statement of this throughout the text in our revised manuscript.

-Due to the short data period it is bit misleading to talk about “hazy days”, “clear days” etc. I strongly recommend to replace all “day(s)”-words with “case” throughout the paper and to strengthen this fact in all analysis.

We thank you for your valuable comments and good suggestions. We agree with you and have replaced all “day(s)”- words with “case” throughout the paper and strengthened this fact in all analysis in our revised manuscript.

-Related to the short period, the authors could also speculate what effect the diurnal variation has on their results (e.g. foggy-haze case at night/in the morning other during the day).

We thank you for your valuable comments and good suggestions. In our previous studies (Leng et al., 2013), the averaged diurnal CCN number concentrations showed a uni-modal pattern in autumn with a sole peak at 14:00 LT around noon. The foggy-hazy case occurring at night and in the morning could be influenced by the diurnal variations and this could partly explain the low CCN number concentrations and aerosol activity during the foggy-hazy case. However, the effect of diurnal variation could be ignored for the clear and hazy cases because both the cases spanned roughly over one whole day. In addition, the result keeps the roughly the same when we consider the relative diurnal variations (i.e. diurnal variations divided by their autumn averages).

Methods

-Section 2.1: Please specify the time period instruments were on the site and also the time period used in this paper, if different.

We thank you for your valuable comments. The instruments have been on

the site since October 2010 and the time period used in this paper was from 6 to 9 Nov. 2010. We have added this statement in Section 2.1 in our revised manuscript as following:

Page 17002, line 3: replace “were” with “have been”

Page 17002, line 5: add “since October 2010” behind “(...121°29'E)”

-Section 2.2: The authors could add a sentence to justify the selection of SS 0.2% for further analysis.

We thank you for your valuable comments and suggestions. We have added a sentence to justify the selection of SS 0.2% for further analysis in Section 2.2 in our revised manuscript as following:

Page 17002, line 21: add “In real atmosphere, SS varies from slightly less than 0.1% in polluted conditions to over 1.0% in clean-air stratus cloud (Hudson and Noble, 2014). The selection of SS 0.2% in the present study would benefit to the measurements in the urban environment for further analysis.” behind “...within 0.2-1.0%.”

-Section 2.2: The instrumentation needs more specific introduction: WPS (operating principle, number of size bins, time resolution, flows, calibration), AE-31 (flow, calibration), MARGA (time resolution, flow, calibration), PM2.5&Hydromet (time resolution).

We thank you for your valuable comments and good suggestions. We have added more specific introductions for the instrumentations in Section 2.2 in our revised manuscript as following:

Page 17003, line 2: add “The principles of the instrument, which have been introduced in detail by Gao et al (2009), combine the Laser Light Scattering (LPS), Condensation Particle Counting (CPC) and Differential Mobility Analysis (DMA). The DMA and CPC can effectively measure aerosol particles distributed in the size range of 10-500 nm in up to 96 channels. The LPS scan the size range of 350-10,000 nm in 24 additional channels. In the present study 60 channels in DMA and 24 channels in LPS for the sample mode were chosen and 3 minutes were needed to scan the entire size range completely, as it took 2 seconds for scanning each channel. DMA was calibrated with NIST SRM 1691 and SRM 1963 PSL spheres (mean diameter of 0.269 and 0.1007 μm , respectively) to maintain DMA transfer function properly and accurate particle sizing traceable to NIST. Four NIST traceable sizes of PSL (i.e. 0.701, 1.36, 1.6 and 4.0 μm) were used to calculate LPS.” behind “...10 nm-10 μm .”

Page 17003, line 8: add “The instrument was operated at an airflow rate of 5 l/min.” behind “...5 min time resolution.”

Page 17004, line 1: add “at 1-hour time resolution. An air pump controlled by a Mass Flow Controller (MFC) draws ambient air with airflow of 1 m^3 /hour into the Sample Box. An internal calibration method by using bromide for the anion chromatograph and lithium for the cation chromatograph was operated over the entire measurement period to ensure this instrument to identify and measure ion species successfully.”

behind “particles.”

Page 17004, line 6: add “at 5-min time resolution” behind “monitor”

-Section 2.2: Lidar methods: what is the overlap, time resolution and range of the lidar? How is the PBL defined? How is the extinction retrieved? What is the effect of the overlap on the comparison to ground-based measurements, if any?

We thank you for your comments and suggestions.

Page 17003, line 15: add “(30 s)” between “resolution” and “information”.

Page 17003, line 16: add “The range of lidar is roughly 30 km at night and 10 km during the daytime. The description of the retrieval of aerosol parameters by the MPL will be only briefly summarized here as it has been given by He et al (2006). The vertical profile of the aerosol extinction coefficient is determined by a near end approach in solving the lidar equation (Fernald, 1984). The PBL height is determined by the MPL lidar at the altitude where a sudden decrease of scattering coefficient occurs (Boers and Eloranta, 1986). The overlap problem must be solved because it can lead to an underestimation of aerosol backscatter and extinction coefficients in the lowest altitudes having the majority of aerosols (He et al., 2006). Outlined by Campbell et al (2002), overlap is typically solved experimentally. The system is set to point horizontally to an averaged data sample with no obscuration, such as the late afternoon,

when the atmosphere is well mixed and the aerosol loading is low. The backscattering over the target layer is roughly assumed constant. The similar calibration has been performed before this study.” behind “(...Brooks, 2003).”

-Section 2.3: Trajectories are calculated in 12h-interval and 24h hours backward. Why these selections, why not more frequent and longer? Later in line 21 you mention that air mass changed at 8 am on Nov 7, how is this defined?

We thank you for your comments. The foggy-hazy case started around 0:00 (23:00 LT on 6 Nov. 2010) and ended near 12:00 (10:00 LT on 7 Nov. 2010) and the hazy case lasted until one hour after 12:00 (13:00 LT on 8 Nov. 2010). The calculations for trajectories in 12h-intervals are corresponding to the case transition time. The air mass pathways in 24h backward have scanned almost all the biomass burning areas in China. The air mass changed its direction around 12:00 on Nov 7, and we have corrected this in our revised manuscript. In figure 1, one can see clearly that the air changed its pathways from southeast to southwest and then northwest after 12:00 on Nov. 7.

-Section 2.3: Are trajectories calculated at 500m AGL or ASL? Why at 500m? Sometimes the PBL is said to be lower than 500m (see page 17006, line 10), does this cause any potential uncertainties?

We thank you for your comments and kind remind. The trajectories are

calculated at 500 m AGL because the PBL was mostly around 500 m, especially during the hazy case. The PBL height seldom reached below 500 m minimized the potential uncertainties.

Results

-Page 17005, line 21: The PBL is not presented from 6 to 9 Nov. Why?

We thank you for your comments. We have presented the PBL data for the whole period in Figure R3 as following:

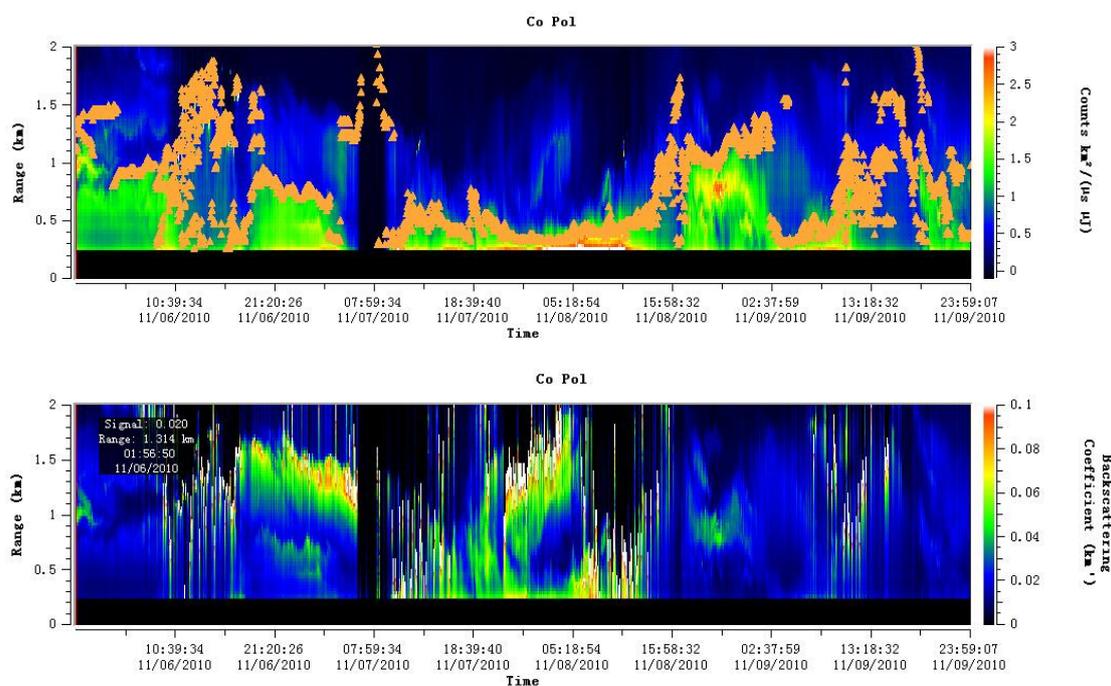


Figure R3 Temporal variations of PBL and vertical extinction coefficient measured by MPL lidar.

-Section 3.2: For explaining the differences in the aerosol physical properties between the cases, I strongly recommend to plot average size distributions for all the three cases (hazy, fog/hazy, clear). This would

make it very visible to reader what is the difference between these.

We thank you for your valuable comments. This is a good suggestion. A plot of average size distributions for all the three cases would make it quite visible to reader what is the difference between these. We have added a new Figure R5 and corresponding analysis in our revised manuscript as following:

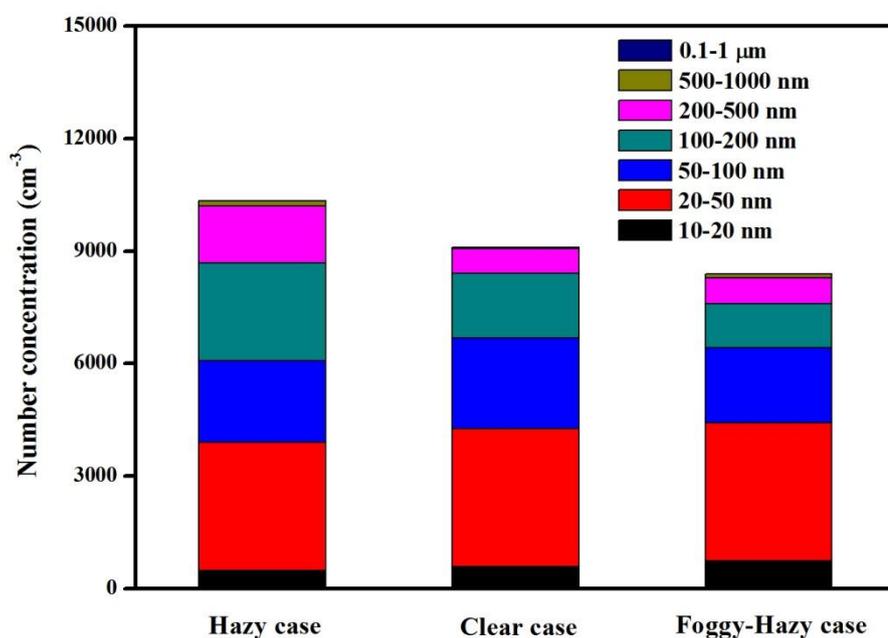


Figure R5 Average size distributions for all the hazy, foggy-hazy, and clear cases.

Page 17007, line 3: add “In addition, Figure 5 shows the average size distributions for all the three cases. It is very visible that it contains relatively more large-sized (e.g. 100 nm) aerosol particles in the air during the hazy case than that during the clear and foggy-hazy cases. Especially aerosol particles larger than 200 nm (a typical CCN size at SS

0.2%) were significantly enhanced.” behind “...particles and N_{CN} .”

-Page 17007, line 3-5: Could you explain and justify more the use of surface and length distribution and retrieving the morphology. What is the benefit here?

We thank you for your comments. We read the paper very carefully and consider the use of surface and length distribution needless in this study. We have deleted this part and relevant figure in our revised manuscript as following:

Page 17007, line 3-8: delete “Figure shows the size distributions...enhance particle aging.”

-Page 17010, line 16-18: More BC particles arrive but the extinction coefficient shows reduction. I would generally expect the opposite.

We thank you for your good comments and kind remind. More BC particles arrived during the time period of 16:00 on 7 to 10:00 on 8 Nov. 2010, when the extinction coefficient shows enhancement within the time period from 22:00 on 7 to 10:00 on 8 Nov. We have corrected this in our revised manuscript as following:

Page 17010, line 18: replace “reduction” with “enhancement”

-Page 17013, line 6-7: Where are the closure calculations?

We thank you for your comments. We add Figure R10 to show the closure calculations in our revised manuscript as following:

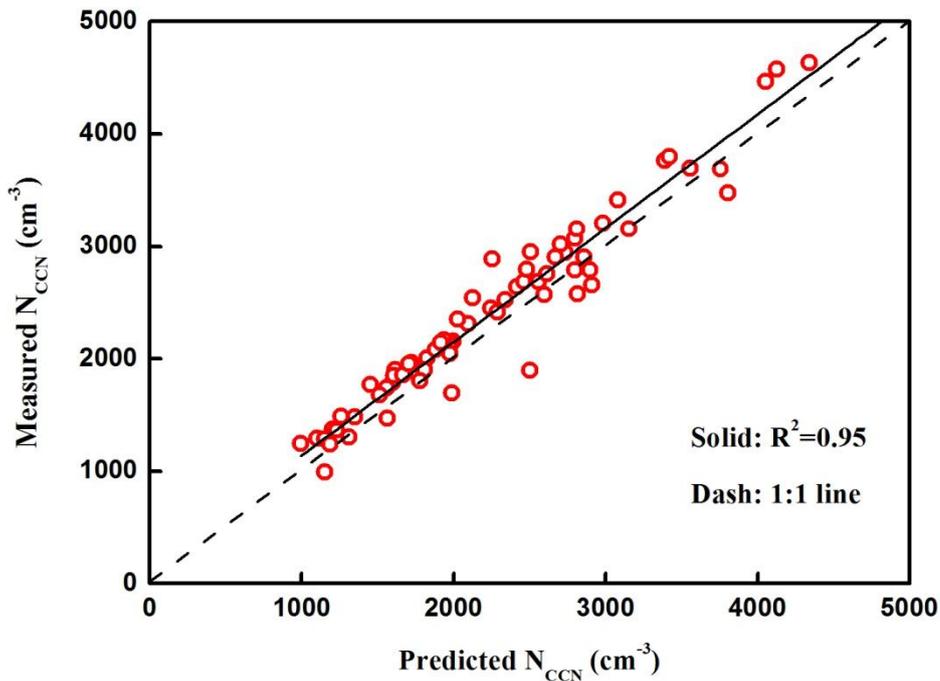


Figure R10 Scatterplot of the simplified closure analysis at SS 0.2%.

Page 17013, line 3: add “In this calculation, the predicted N_{CCN} of hourly-averaged were compared with the measured ones correspondingly.” behind “...the upper boundary.”

Page 17013, line 3: add “The results of this closure analysis are shown in scatterplot in Figure 10 and 11.” before “The prediction for CCN...”

Page 17013, line 3: add “throughout the entire data set. The linear regression between predicted and measured N_{CCN} produces a slope of 1.012 and an intercept of 128.3 cm^{-3} ($R^2=0.95$), and the average ratio of predicted versus measured N_{CCN} is 0.94 (Figure 10). The results indicate some moderate underestimate (about 6% on average) but the agreement is still excellent. The achieved closure calculation suggested that water

soluble inorganic ions played a major role in contributing the κ value. In fact, 83.8% of the κ was expressed by $\text{SO}_4^{2-} + \text{NO}_3^- + \text{NH}_4^+$ in total (in another study by our group, not published yet), with their individual contribution to be 39.8%, 31.7% and 12.3%, respectively. In addition,” behind “...generally success.”

Page 17013, line 3: replace “It” with “it”

Page 17013, line 10: replace “On the other hand” with “In all cases”

Page 17013, line 6-10: delete “The achieved closure...12.3%, respectively.”

Conclusions

-Page 17013, line 15-17: Is the great influence of ws, wd and temperature shown somewhere in the result section?

We thank you for your valuable comments. We have added the influence of ws, wd and temperature in Section 3.1 in our revised manuscript as following:

Page 17013, line 18: replace “CN” with “PM_{2.5}”

Page 17013, line 18: delete “(500 m)”

Page 17013, line 19: add “For example, PM_{2.5} and BC accumulated in mass concentrations and reached their maximums when these meteorological parameters remained at low level (e.g. wind speed at 2 m s⁻¹, PBL height around 0.5 km). The later disappearance of the pollutants at the end of the hazy case was mostly attributed to the wind speed

increasing to 6 m s^{-1} , and the PBL height rising to 1.4 km (Figure 2).”
behind “...BC loadings (Figure 3).”

-Page 17014, line 12: Was the CCN closure presented in the result section?

We thank you for your comments. We have presented the CCN closure in the result section. Please refer to the reply to “-Page 17013, line 6-7: Where are the closure calculations?” in detail.

Tables

-Table 1: What is the origin of the different PBL heights for the cases? Could it be that the cases happen in different time of the day? Or does the PBL detection suffer from the fog?

We thank you for your comments. The effect of diurnal variations on PBL heights for the clear and hazy cases could be ignored since both the cases spanned over one whole day. The PBL height for the foggy-hazy case occurring at night and in the morning could be partly attributed to the diurnal variation. The main origin of the different PBL heights for the cases is the pollutant loadings. PBL detection probably suffered from the fog during the period 5:00-9:00 am on 7 Nov. 2010 since the RH reached its maximum during this time. This is further confirmed by another MPL measurement performed in Shanghai Meteorological Bureau, roughly 12 km southwest of our site.

-Table 3 would benefit of some references.

We thank you for your valuable comments and good suggestions. The reference “Yue, D. L., Hu, M., Zhang, R. J., Wu, Z. J., Su, H., Wang, Z. B., Peng, J. F., He, L. Y., Huang, X. F., Gong, Y. G., and Wiedensohler, A.: Potential contribution of new particle formation to cloud condensation nuclei in Beijing, *Atmos. Environ.*, 45, 6070-6077, 2011” has been added for Table 3 in our revised manuscript.

Figures

-Please mention the meaning of the red and black boxes of the plots in the Figure captions.

We thank you for your good suggestions. We have mentioned the meaning of the red and black boxes of the plots in our revised manuscript.

-Figure 3: Why is the PBL not for the whole period? The values near the end of the “red-box period” are unreal and the PBL values are not true. One can see that there is something below the plotted 300m line that blocks the beam (the fog probably) and there is no signal from above. How about the other PBL values, could it be that some other of the low values are due to instrumental limitations.

We thank you for your comments. We have presented the PBL for the whole period as a new Figure 3. PBL detection probably suffered from the fog during the period 5:00-9:00 am on 7 Nov. 2010 since the RH reached its maximum during this period. This was further confirmed by another MPL measurement performed in Shanghai Meteorological

Bureau, roughly 12 km southwest of our site.

-Figure 5: Is this figure needed, what is the information?

We thank you for your valuable comments and good suggestions. We read the paper carefully and consider Figure 5 is not needed. We have deleted this figure in our revised manuscript.

Technical corrections

-Page 17010, line 24: “ R^2 are listed”, maybe “presented” fits better.

We thank you for your good suggestions. We have replaced “listed” with “presented” here.

Page 17010, line 24: replace “listed” with “presented”

-Page 17012, Equation 2: Define D_d .

We thank you for your comments and suggestions.

Page 17012, line 6: add “ D_d is the dry diameter,” behind “T is temperature,”

-Figure 9: Define which subplot is a), b),...

We thank you for your valuable comments and good suggestions. We have defined and labeled the subplots a), b), c), d) in Figure R9.

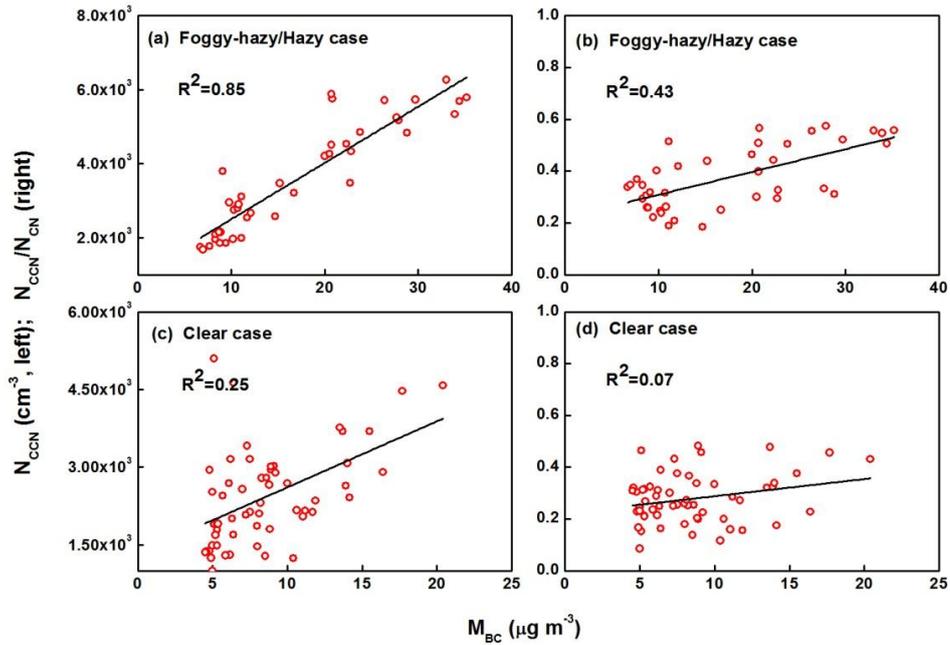


Figure R9 Correlations of BC mass concentration (M_{BC}) to N_{CCN} and N_{CCN}/N_{CN} (0.2% SS) during the foggy-hazy/hazy (a, b) and clear (c, d) cases.

-Figure 10: 1:1 line would be informative.

We thank you for your valuable comments and good suggestions.

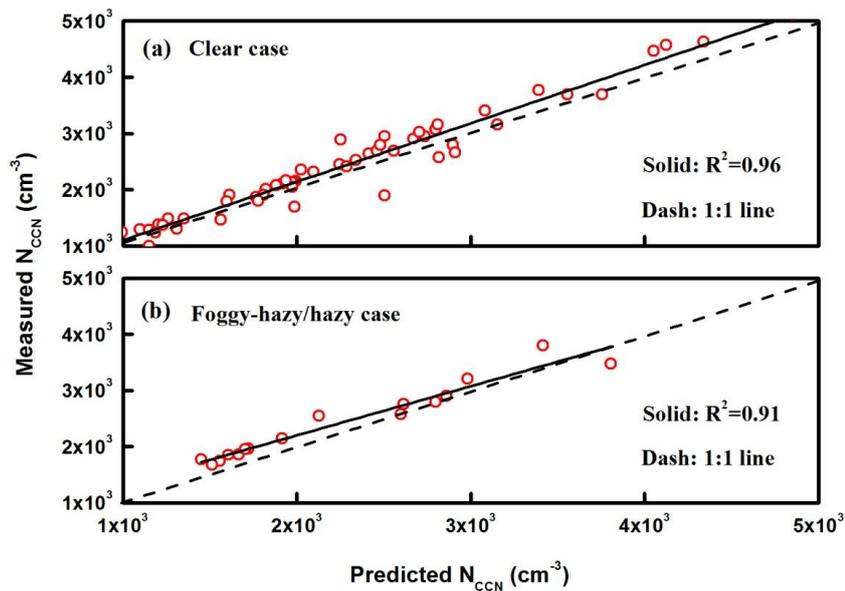


Figure R11 Correlations of observed and predicted N_{CCN} (0.2% SS) in the clear (a) and foggy-hazy/hazy (b) cases.