

Reviewer #2:

*General comments:*

*The present paper focuses on improving our understanding of the aerosol mixing state in a background site of the North China Plain in China. This is achieved by combining various techniques, including HTDMA, CCN counter, VTDMA, and SP2. The study provides a first-time intercomparison of the four aerosol mixing state parameters from the instruments above and offers insights into the interlink among these parameters and potential sources. I find this research to be important and interesting for aerosol mixing state studies. The manuscript is well-structured and scientifically engaging for the aerosol society. However, in terms of writing, it would be beneficial for non-expert readers if certain sentences were shortened and explanations were provided before reaching conclusions. Please see the detailed comments below. I suggest publishing the manuscript after a minor revision.*

**Response: Thanks for your comments. Suggestions and comments are addressed point-by-point and corresponding responses are listed below.**

*Specific comments:*

1) Line 70. “..lead to substantial overestimation”. Could you provide more details about the magnitude of the substantial overestimation?

**Response: Thanks for your suggestion. There can be overestimation of NCCN from 10% to 30%, and we have revised this sentence as:**

**“Using simple internally mixing state assumptions for aerosol chemical compositions in estimating CCN number concentrations can lead to substantial overestimations (up to 30%, Deng et al., 2013; Farmer et al., 2015; Ren et al., 2018; Ching et al., 2017, 2019; Tao et al., 2021).”**

2) Line 94. “highly correlated to those of a VTDMA at high temperature”. Which temperature do you refer to and why?

**Response: In general, in order to remove most non-refractory materials in aerosol, 300 °C is used in VTDMA measurement (Philippin et al., 2004; Wehner et al., 2009; Zhang et al., 2014; Hong et al., 2017; Wang et al., 2022). But this temperature can be lower to 200 °C depending on the aerosol chemical compositions. We have revised this sentence as:**

**“Thus, measurements of an SP2 are highly correlated to those of a VTDMA at high temperatures (higher than 200 °C and up to 300 °C), with their differences reflecting**

variations in aerosol density, shape or volatility (Philippin et al., 2004; Wehner et al., 2009; Adachi et al., 2018, 2019; Wang et al., 2022).”

3) Line 127-129. Please summarize the key messages of the meteorology influences on the aerosol mixing state.

**Response:** Thanks for your suggestion. As reported by Kuang et al., 2020, the secondary aerosol formations under low RH conditions, mainly taken place in gaseous phase, would change to that mainly taken place in aqueous phase under high RH conditions. As secondary aerosol formed through different mechanism have different chemical compositions and add mass to different particle groups, secondary aerosol formations under different meteorological conditions can affect the aerosol mixing states (Tao et al., 2021). We have revised this sentence as:

“Meteorological conditions can greatly impact the secondary aerosol formation in the NCP, which can be significantly exacerbated during severe pollution events. The secondary aerosol formations under low RH conditions, mainly taken place in gaseous phase, would change to that mainly taken place in aqueous phase under high RH conditions (Kuang et al., 2020). As secondary aerosols formed through different mechanisms have different chemical compositions and add mass to different particle groups, secondary aerosol formations under different meteorological conditions can affect the aerosol mixing states (Tao et al., 2021).”

4) Line 168 BBOA, line 173 FFOA..., please explain the full name when introducing a new term and check out the remaining of manuscript.

**Response:** Thanks for your suggestion. The full names of BBOA and FFOA are Biomass Burning Organic Aerosol and Fossil Fuel Organic Aerosol, respectively. We have added a table listing the definition and description of the abbreviations as follow:

**Table 1. Definition and description of the abbreviations.**

| Abbreviations | Full name and/or Definition   |
|---------------|---|
| BBOA          | Biomass Burning Organic Aerosol<br>In this study, characterized by obvious m/z 60 (mainly C <sub>2</sub> H <sub>4</sub> O <sub>2</sub> <sup>+</sup> ) and 73 (mainly C <sub>3</sub> H <sub>5</sub> O <sub>2</sub> <sup>+</sup> ), which are two indicators of biomass burning |
| FFOA          | Fossil Fuel Organic Aerosol   |

|                    |  |
|--------------------|--|
|                    | A mixed factor in this study that comprises traffic emissions and coal combustions, which was characterized by typical hydrocarbon ion series                |
| OOA                | Oxidized Organic Aerosol   |
| OOA1 and OOA2      | Two SOA factors derived from the PMF analysis  |
| SOA                | Secondary Organic Aerosols<br>Summation of BBOA and FFOA   |
| POA                | Primary Organic Aerosols<br>Summation of OOA1 and OOA2   |
| PM <sub>2.5</sub>  | Particulate Matter with aerodynamic diameter less than 2.5 $\mu\text{m}$   |
| PM <sub>1</sub>    | Particulate Matter with aerodynamic diameter less than 1 $\mu\text{m}$   |
| NR-PM <sub>1</sub> | Non-refractory PM <sub>1</sub>   |
| MF                 | Mass Fraction  |
| D <sub>p</sub>     | Particle diameter after humidification or heating  |
| D <sub>d</sub>     | Particle diameter under dry conditions without humidification or heating   |
| K                  | Hygroscopicity parameter   |
| SS                 | Super-saturation   |
| SPAR               | Size-resolved Particle Activation Ratio<br>Size-dependent CCN activity under a specific SS   |
| MAF                | Maximum Activation Fraction<br>An asymptote of the measured SPAR curve at large particle sizes and represents the number fraction of CCNs to total particles |

|   |   |
|---|---|
| $D_a$   | Midpoint activation diameter<br>Linked to the hygroscopicity of CCNs  |
| GF  | Growth factor<br>The ratio between particle with and without humidification, and is linked to aerosol hygroscopicity  |
| SF  | Shrink Factor<br>The ratio between particle with and without heating, and is linked to aerosol volatility   |
| PDF   | Probability Distribution Function   |
| $NF_H$  | Number Fraction of Hydrophilic aerosol whose hygroscopicity parameter is higher than $\sim 0.07$ .  |
| $NF_V$  | Number Fraction of Volatile aerosol whose Shrink Factor at $200\text{ }^\circ\text{C}$ is lower than 0.85.  |
| $NF_{noBC}$   | Number Fraction of BC-free particles  |
| $NF_{CBC}$  | Number Fraction of thickly coated BC particles  |
| $R_{exBC}$  | Ratio of the number concentration between externally BC particles and BC-containing particles.<br>Externally BC particles are defined as BC-containing particles in this study. |
| $NF_A - NF_B$<br>( $NF_{noBC} - NF_H, NF_V - NF_H,$<br>$NF_{noBC} - NF_V, NF_V - MAF,$<br>$NF_{noBC} - MAF$ ) | The difference between the number fraction of A and B.  |

**In addition, we have also added the definition and description of the abbreviations when first introduced in each section and also in the caption of the figures for clarification.**

5) Line 177, what do you mean by “different chemical process” and could you give more details?

**Response: Secondary organic aerosol formations originated from volatile organic compounds precursors could be formed in differ formation pathways such as aqueous phase reactions, heterogeneous reactions or gas phase reactions and also might be oxidized under different conditions, for example oxidized under different nitrogen oxide conditions with different oxidation capacity and oxidants. The following sentences is added in the revised manuscript.**

**“Secondary organic aerosol formations originated from volatile organic compounds precursors could be formed in differ formation pathways such as aqueous phase reactions, heterogeneous reactions or gas phase reactions and also might be oxidized under different conditions, for example, oxidized under different nitrogen oxide conditions with different oxidation capacity and oxidants. Two resolved SOA factors were found to display different spectral patterns, correlations with tracers and diurnal variations, suggesting that they resulted from different chemical processing, however, their formation mechanisms remain to be explored in our future studies. In general, the OOA factor 1 (Oxidized Organic Aerosol, OOA1) had higher CO<sub>2</sub>+C<sub>2</sub>H<sub>3</sub>O+ (3.9) and O/C (0.91) ratios compared to the OOA factor 2 (OOA2, 2.1 and 0.78).”**

*6) Line 187, why do you choose these three supersaturations for CCN measurements?*

**Response: As particle size is the most important parameter in determining CCN activity (Duesk et al., 2006), measurement of CCN activity can indicate particle hygroscopicity in different particle size ranges. In general, the three supersaturations indicate the particle hygroscopicity in particle size range from 100 nm to 200 nm.**

**In order to perform intercomparisons among instruments, three supersaturations (SSs) of 0.08%, 0.14% and 0.22% were applied in a single cycle of about 15 minutes. CCN measurement under these three SSs reveals mainly CCN activity of aerosols reside in accumulation mode aerosol with diameter range of about 100-200 nm, which are close to diameters of HV-TDMA measurements, and higher SSs would reveal CCN activity of smaller aerosol particles (<100 nm) where DMA-SP2 measurement is not available:**

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7) Line 211, the maximum temperature you chose is 200 degree Celsius, why do you choose this threshold?

**Response:** The HV-TDMA were scanning at different temperatures and diameters for the HV-TDMA system, to ensure the time duration of one full cycle is about 3 h, we limited the number of temperatures and diameters. Most importantly, results of previous studies in the North China Plain have shown that 200 degree Celsius is enough for removing most non-refractory aerosols (>80%) (Xu et al., 2019).

8) Line 225-229, regarding the chosen size for SP2, which system was conducted for this study, with or without thermodenuder-bypass? Since you are expected to compare with HTDMA and VTDMA, why not choose the same sizes to measure for the three instruments?

**Response:** The DMA-SP2 system was conducted both with and without thermodenuder-bypass depends on time, and detailed periods are added in the revised manuscript. Compared to HTDMA and VTDMA, more particle sizes are selected in the measurement DMA-SP2 system for obtaining more information of BC mass concentration and mixing states at different particle diameters for other scientific purposes. Because the time needed for a single particle size measurement of DMA-SP2 system is much shorter than that of HTDMA and VTDMA, and one full cycle for H/VTDMA lasts 3 hours. We have added corresponding description into the manuscript as:

“The DMA-SP2 setup was able to measure the mixing states of aerosols at diameters of 100 nm, 120 nm, 160 nm, 200 nm, 235 nm, 270 nm, 300 nm, 335 nm, 370 nm, 400 nm, 435 nm, 470 nm, 500 nm, 535 nm, 570 nm, 600 nm, 635 nm, 670 nm, 700 nm within 20 minutes, when it wasn't placed after a thermodenuder-bypass switch system (13<sup>th</sup>-24<sup>th</sup> October, 09:00 am of 5<sup>th</sup> November to 09:00 am of 8<sup>th</sup> November). However, it only measured mixing states at diameters of 120 nm, 160 nm, 200 nm, 250 nm, 300 nm, 400 nm, and 500 nm when it was placed after a thermodenuder-bypass switch system (11:00 am 24<sup>th</sup> October to 08:00 am 5<sup>th</sup> November, and 09:00 am of 8<sup>th</sup> November to 06:00 pm of 17<sup>th</sup> November). Because the measurements of HTDMA and VTDMA are conducted solely by a single H/VTDMA system working in different mode, the time needed for a single particle size measurement of HTDMA and VTDMA is much longer than that of DMA-SP2 system. Thus, more particle sizes are selected in the measurement DMA-SP2 system for acquiring BC mass concentration and mixing state at more diameters, compared to those of HTDMA and VTDMA”

9) Line 235, does the flow rate influence the measurements and by how much?

Response: This change satisfied the flowrate requirements of this instrument (0.03 to 0.18 L/min), and 0.12 L/min was typically used. The flow rate change does not affect the measurements when aerosol number concentration is not small. Actually, at the very beginning, 0.1 L/min (less than the typical one 0.12L/min) was usually used because the NCP is generally polluted, and higher flow rate would produce larger data storage, however, does not affect the statistical results. We change to 0.12 L/min is because that we realized that we scan up to 700 nm using the DMA-SP2 which is different with previous studies where aerosol number concentration is much smaller and a larger sample flow rate should be better.

10) Section 2.3.1, the MAF is a fitting parameter from eq.7, what is the physical meaning of this parameter? Is it the maximum activation fraction?

Response: Yes, it's the maximum activation fraction and we have revised the corresponding description as:

“.. MAF (Maximum Activation Fraction) is an asymptote of the measured SPAR curve at large particle sizes as shown in Fig. S4, and it represents the number fraction of CCNs to total particles. ...”

To be noted, a schematic of the SPAR parameterization scheme and the corresponding fitting parameters is added into the supplement for clarification as:

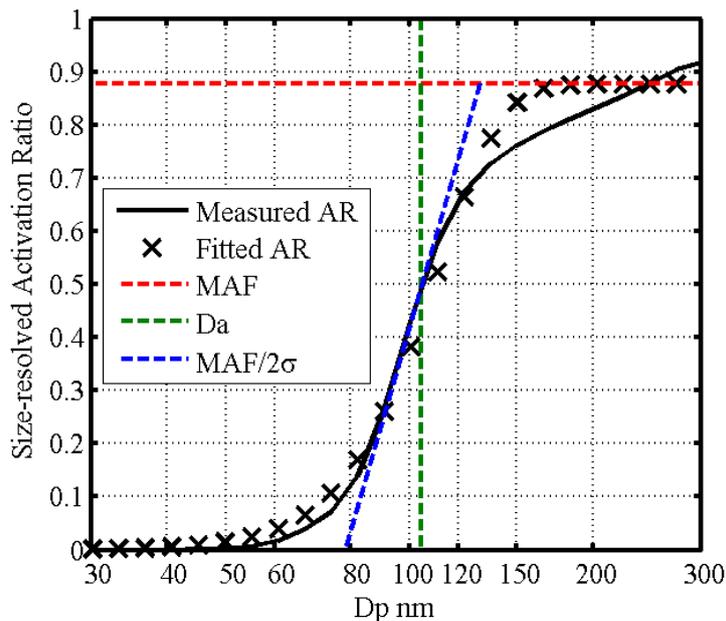


Fig. S4. Schematic of the parameterization scheme of SPAR curves. The black solid

curve and the black crossing are the measured SPAR and fitted SPAR with the parameterization scheme. The red, green and blue dashed lines indicate the fitting parameters of Maximum Activation Fraction (MAF), the midpoint activation diameter (Da) and s, respectively.

11) Line 267, add sizes for the GF “The GFC for the four measured particle sizes were 1.1, 1.15, 1.175 and 1.2”.

**Response:** The GFC for particle size of 50, 100, 150 and 200 nm are 1.1, 1.15, 1.175 and 1.2, respectively. We have revised it as:

“The  $GF_c$  for the four measured particle sizes of 50 nm, 100 nm, 150 nm and 200 nm were 1.1, 1.15, 1.175 and 1.2, respectively”

12) Section 2.3.3. Here you use the lag time between the peak of the scattering signal and the incandescence signal to classify the bare and coated BC. Is it related to the BC-coating mass ratio? The mass ratio is more commonly used and intuitive to understand.

**Response:** Thanks for your suggestion. It is related to the coating thickness of the BC-containing aerosols. The BC-coating mass ratio cannot be directly obtained in SP2 measurement, due to the lack in the accurate density and shape of the BC core. In addition, the lag time is positively correlated to the coating thickness, but their relation cannot be directly quantified and also calibrated. Thus, a critical value of lag time rather than coating thickness or coating mass ratio is used to classify the bare and coated BC. We have revised this sentence as:

“In this study, a two-mode distribution of the lag time ( $\Delta t$ ) was observed. As the lag time is positively correlated to the coating thickness, a critical lag time (0.8  $\mu s$ ) was used to classify the BC-containing particles into thinly coated (or bare) BC ( $\Delta t < 0.8 \mu s$ ) and thickly coated BC ( $\Delta t \geq 0.8 \mu s$ ), respectively.”

13) Line 297-299, please give exact values of PM mass for the heavily polluted and clean period.

**Response:** Thanks for your suggestion. Non-refractory  $PM_{10}$  mass for the heavily polluted and clean period are  $49.5 \pm 22.5$  and  $5.1 \pm 3.3 \mu g/m^3$ , respectively. We have revised this sentence as:

“The mass concentrations of different aerosol compositions increased significantly from October 23rd to November 6th (heavily polluted period with average non-

refractory PM<sub>1</sub> mass concentration of 49.5±22.5 µg/m<sup>3</sup>) and decreased to much lower levels after November 6th(clean period with non-refractory PM<sub>1</sub> mass concentration of 5.1±3.3 µg/m<sup>3</sup>).”

14) Line 315-316. “At lower SSs, the rapid increases in SPAR curves occur at larger particle sizes and the maximum AR of SPAR curves becomes smaller”. Please briefly explain why.

**Response:** Thanks for your suggestion. For lower SSs, particle size need for CCN activation is larger, thus SPAR curve start to increase from 0 at larger particle size. Because only SPAR in particle size lower than 300 nm is presented and there was less particle to be CCN active under low SSs, the maximum AR of SPAR curves becomes smaller under low SSs. We have revised this sentence as:

“At lower SSs, the rapid increases in SPAR curves occur at larger particle sizes, since particle size need for CCN activation is larger. In addition, as SPAR in particle size lower than 300 nm is presented, the maximum AR of SPAR curves becomes smaller as there was less particle to be CCN active under low SSs.”

15) Line 318, add SS for the “increases in SPAR curves, are approximately 90 nm, 120 nm and 180 nm”

**Response:** Thanks for your suggestion. We have revised it as:

“... increases in SPAR curves, are approximately 90 nm, 120 nm and 180 nm for the three SSs of 0.08%, 0.14% and 0.22%, respectively.”

16) Fig 2. Are bars representing the standard deviation of the campaign average?

**Response:** Yes, and we have added corresponding description in the end of the caption of Figure 2 as:

“The shaded areas indicate the standard deviations.”

17) Line 331-333, “In general, the size dependence of MAF, NFH, NFV and NFnoBC were similar to one another, suggesting they were dominated by the same particle group, namely BC-free particles”. I think this statement is not well supported, I would suggest weakening it or proving it with more evidence. For example, thickly coated BC particles can be very CCN-activate, hydrophilic and volatile, if mostly contain SIA.

**Response:** Thanks for your suggestion and we fully agree. We have revised this sentence as:

“In general, the size dependence of MAF,  $N_{FH}$ ,  $N_{FV}$  and  $N_{noBC}$  were similar to one another, suggesting they were likely dominated by the same particle group, namely BC-free particles. ”

18) Line 335, please provide exact values of the fraction of BC-containing particles and the applied diameter range, because the terms “higher” or “larger” are not accurate. Check out similar issues for the remaining manuscript too.

**Response:** Thanks for your suggestion. We have revised this sentence as:

“This suggests that primary emissions tend to have higher fractions of BC-containing particles in larger diameter ranges, for example, the fraction of BC-containing particles increases from ~0.1 to ~0.4 as particle size enlarges from 200 nm to 500 nm during the clean period.”

**We have also checked the manuscript and revised the following:**

**L333:** “This particle group had the highest fraction (higher than 0.7) during the heavily polluted period and the lowest fraction (down to 0.5) during the clean period, ...”

**L347:** “... when the nitrate fraction was the highest (~30%) and the SOA fraction was the lowest (~7%) among all three periods, ...”

**L354-355:** “but were larger than  $N_{FH}$  (by ~0.2) during the moderately and heavily polluted periods when the POA/SOA fractions were higher (~40% vs ~35%).”

19) Line 342, what do you mean by “the more efficient secondary aerosol formation”, increase by secondary aerosol mass or particle size?

**Response:** Here we are referring to that the formation rate of secondary aerosol mass is more efficient on larger particle, and we have revised this sentence as:

“... while the decrease of  $Rex_{BC}$  with increasing particle diameter size in the polluted period confirms secondary aerosol formation to be more efficient on particles with larger diameter.”

20) Line 356-357, what is the kappa value for hydrophobic mode aerosol?

**Response:** The kappa value for hydrophobic mode aerosol is less than 0.07 and we have revised this sentence as:

“The critical  $\kappa$  of hydrophilic mode aerosols was 0.07, suggesting that a higher fraction of aerosols had  $\kappa$  below 0.07 (i.e. hydrophobic mode aerosol in this study) during the

moderately polluted period.”

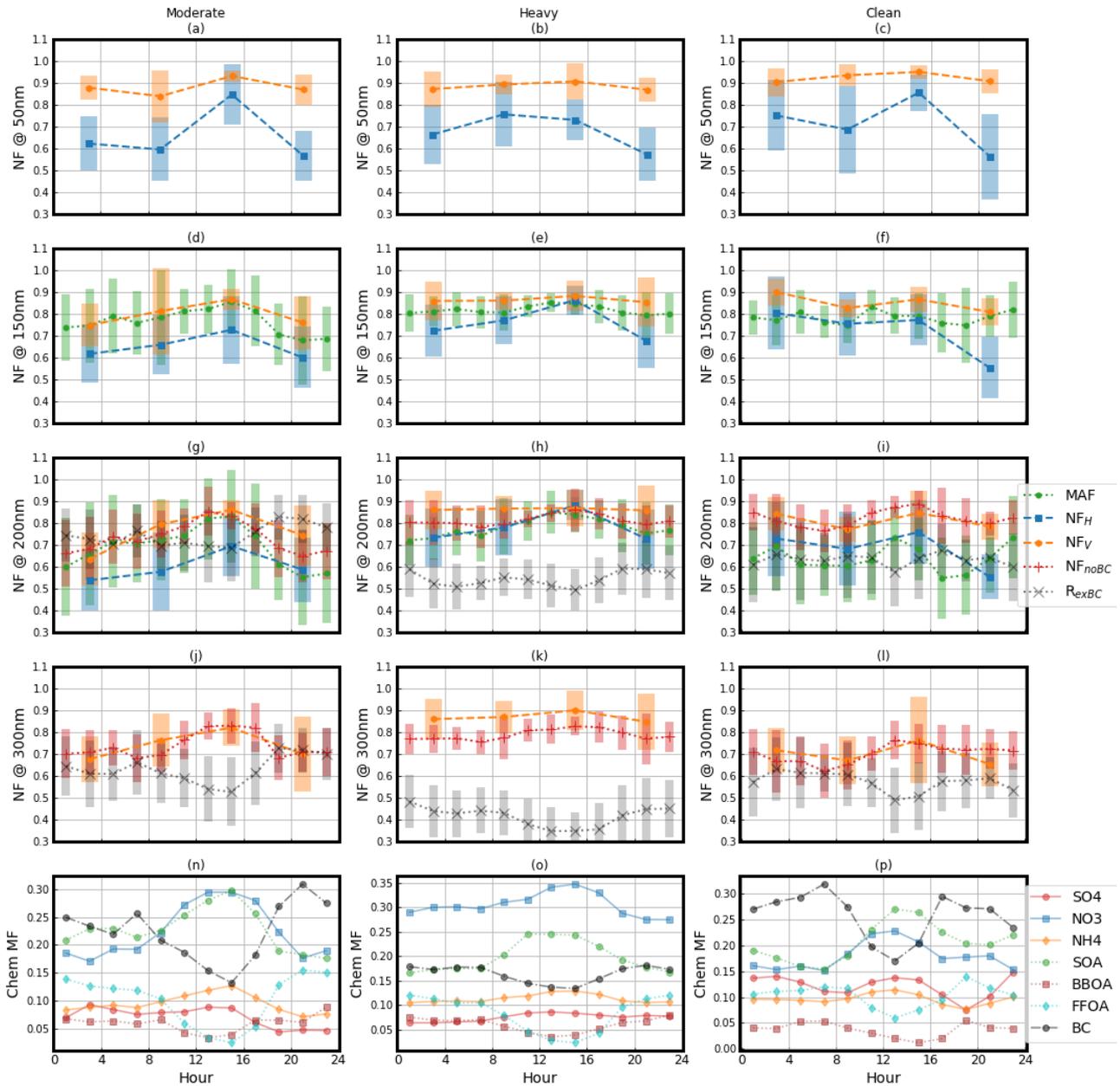
21) Line 361, how do you get this statement with “lower than 0.07 but still CCN active”? please explain in detail.

**Response:** In this part we are referring to that the difference among MAF,  $N_{F_v}$ ,  $N_{F_H}$  and  $N_{F_{noBC}}$  and we found that  $N_{F_H}$  is significantly smaller than the other three parameters. This may indicate that a portion of particles to be CCN active but not hydrophilic, i.e. with  $\kappa$  lower than 0.07. We have revised this sentence as:

“The  $N_{F_H}$  was consistently lower than  $N_{F_v}$  and  $N_{F_{noBC}}$  (the average difference between  $N_{F_H}$  and  $N_{F_{noBC}}$  was about 0.2), especially during the moderately polluted period. As mentioned above that  $N_{F_H}$  was also lower than MAF during the moderately polluted periods, there may be a significant fraction of volatile BC-free aerosols had hygroscopicity lower than critical  $\kappa$  of 0.07 but were still CCN-active and therefore not fully hydrophobic.”

22) Fig4, I would suggest simplifying the plot and keeping the sizes with most concurrent measurements, e.g. 150, 200 and 300 nm. Put other sizes to the supplement.

**Response:** Thanks for your suggestion. We have revised Figure 4 and its caption as:



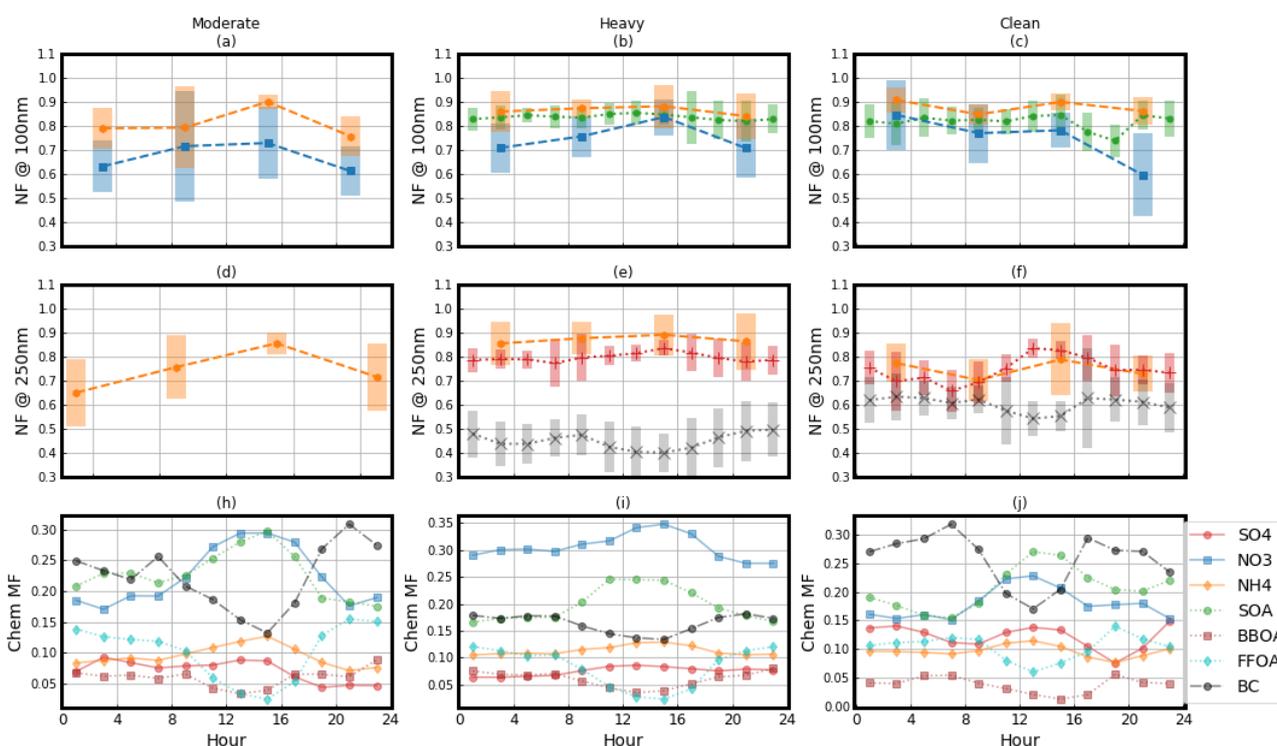
“Figure 4. (a-l) Diurnal variations of aerosol mixing state parameters (identified by color and marker) at different particle sizes (50, 150, 200 and 300 nm) during the three periods. The shaded areas indicate the standard deviations. MAF (Maximum Activation Fraction): An asymptote of the measured SPAR curve at large particle. NF<sub>H</sub>: Number Fraction of Hydrophilic aerosol whose hygroscopicity parameter is higher than ~0.07. NF<sub>V</sub>: Number Fraction of Volatile aerosol whose Shrink Factor at 200 °C is lower than 0.85. NF<sub>noBC</sub>: Number Fraction of BC-free particles. R<sub>exBC</sub>: Number fraction of externally BC particles in total BC-containing particles. (m-o) Diurnal variations of mass fractions of aerosol chemical compositions including secondary organic aerosols (SOA), biomass burning organic aerosol (BBOA), fossil fuel organic aerosols (FFOA), and

inorganic ions including sulfate (SO<sub>4</sub>), nitrate (NO<sub>3</sub>) and ammonium (NH<sub>4</sub>) (identified by color and marker) during the three periods.”

We have revised the corresponding description of Figure 4 as:

“For particle sizes larger than 100 nm (shown in both Fig. 4 and Fig. S5), there were maxima in the afternoon for MAF, NF<sub>H</sub>, NF<sub>V</sub> and NF<sub>noBC</sub>, indicative of a peak during this time due to the increase in secondary aerosol compositions like nitrate and SOA, and the decrease of POA and BC.”

We have also added Figure S4 into the supplement as:



“Fig. S5. (a-l) Diurnal variations of aerosol mixing state parameters (identified by color and marker) at different particle sizes (50, 150, 200 and 300 nm) during the three periods. The shaded areas indicate the standard deviations. (m-o) Diurnal variations of mass fractions of aerosol chemical compositions (identified by color and marker) during the three periods.”

Line 362- 366, the diurnal variations should be described more explicitly as the pattern of RexBC is clearly different from the other three mixing state parameters and explain why.

Response: Thanks for your suggestion. We have added more discussion in the end of this paragraph as:

“R<sub>exBC</sub> tended to be lower during the daytime and its diurnal variation was more

significant in larger particle sizes. In general, these diurnal variations for  $R_{\text{exBC}}$  are opposite to those of  $NF_{\text{noBC}}$  and secondary aerosol mass fractions, and agree better with those of the primary aerosol mass fractions. This is because BC particles originate from primary emissions and are mainly externally mixed. After experiencing aging process in the atmosphere, BC particles can be coated by secondary aerosol formed on, resulting in more coated BC particles and less externally mixed BC particles. As the secondary aerosol tends to form on larger particles, the diurnal variations of secondary aerosol formations may affect more significantly on those of mixing state of BC particles and thus  $R_{\text{exBC}}$  in larger particle sizes.”

23) Line 384, table S1 is quite interesting for readers thus I suggest putting it or making a correlation plot into the main context.

**Response:** Thanks for your suggestion. We agree that useful information is contained in this table, we also struggled before we decided to put it in the supplement. We want readers focus more on key parts of those intercomparison results, however, it was also available in the supplement in case that readers want to know all scenarios.

24) Line 385, why do you choose these three sizes? The critical size for the setting SS?

**Response:** As shown in Figure 2, the particle size where the rapid increases in SPAR curves are approximately 90 nm, 120 nm and 180 nm for the three SSs of 0.22%, 0.14% and 0.08%, respectively. And the diameter range of rapid increases in SPAR curves are determined by aerosol hygroscopicity in this particle size ranges. Thus, the three particle sizes of 100 nm, 150 nm and 200 nm are chosen in comparison to the MAF at the three SSs of 0.22%, 0.14% and 0.08%, respectively. We have revised this sentence as:

“Note that MAF at SSs of 0.08%, 0.14% and 0.22% was used for comparison at particle sizes of 200 nm, 150 nm and 100 nm. This is because that the diameter range of rapid increases in SPAR curves are determined by aerosol hygroscopicity in this particle size range, and the midpoint of rapid increase diameter ranges of SPAR curves at SSs of 0.08%, 0.14% and 0.22% are approximately 180 nm, 120 nm and 90 nm (as shown in Fig. 2). ”

25) Line 386. A classification of the correlation should be clarified, such as the  $r$  range for the weak, moderate, and strong correlation.

**Response:** Thanks for your suggestion. The value range of correlation coefficient for

weak, moderate and strong was generally less than 0.3, from 0.3 to 0.5 and larger than 0.5. We have added detailed value of correlation coefficient into the manuscript including:

L386: "... moderate correlations ( $r \sim 0.5$ ) .."

L392: "... the correlation became weaker ( $r \sim 0.4$ ), ..."

L421: "..., and weak correlations ( $r < 0.3$ ) ..."

L440: "... a strong positive correlation with  $\text{MF}_{\text{SO}_4}$  ( $r > 0.5$ ). ..."

L445: "... the weaker correlations with SOA ( $r \sim 0.3$ ) seen in Fig. 8."

L454: "..., the strong positive correlations between  $\text{NF}_v$  and secondary aerosol formations ( $r \sim 0.6$ ) ..."

L457: "... strong positive correlations ( $r \sim 0.5$ ) ..."

L465: "... a strong negative correlation with  $\text{MF}_{\text{NH}_4}$  and  $\text{MF}_{\text{NO}_3}$  (mainly -0.6) ..."

26) Line 392, what do you mean by saying "...while the degree was the least for the correlation.."?

**Response:** We are referring to that the degree of the reduction of correlation was the least for the correlation between MAF and  $\text{NF}_v$ , and we have revised this sentence as: "At smaller particle size, the correlation became weaker ( $r \sim 0.4$ ), while the degree of the reduction was the least for the correlation between MAF and  $\text{NF}_v$ ."

27) Fig. 5, what is the  $r$  in the plot? It would be more intuitive to use the same marker to represent different periods in the plot.

**Response:** The variable  $r$  represent the correlation coefficient and we have added corresponding description into the caption as "with  $r$  representing the correlation coefficient." At the request of the Copernicus Publications, the marker used to present different periods are set to be different in order to making this figure friendly to readers with color vision deficiencies.

28) A summary table (or correlation matrix plot) of  $r$  in Fig5-7 will be helpful for readers to better understand the interlink between mixing state parameters and chemical composition.

**Response:** Thanks for your suggestion and the correlation between mixing state parameters and aerosol chemical composition as well as detailed correlation during

different pollution periods were summarized in Figures S6 and S8 (Figures S5 and S6 in old version). We have added introduction of these figures into the manuscript as:

In the end of Section 3.3: “In addition, the correlation between mixing state parameters and primary aerosol composition during the campaign and different pollution periods were summarized in Fig. S7.”

In the end of last second paragraph of Section 3.4: “Besides, the correlation between mixing state parameters and secondary aerosol composition during the campaign and different pollution periods were summarized in Fig. S9.”

29) Line 400, please give values to the sentence “correlation with MFFFOA was much weaker compared to MFBBOA”.

**Response:** Thanks for your suggestion. We have revised this sentence as:

“However, the correlation with MFFFOA (-0.45~-0.74) was much weaker compared to MFBBOA (-0.10~-0.45).”

30) Fig.7. Which size of data do you use?

**Response:** The size is 200 nm and we have added corresponding description into the caption of Figure 7 as “The correlation between the difference among the four aerosol mixing state parameters at particle size of 200 nm and MF of primary organic aerosol components during different periods.”

31) Line 428, please introduce what the difference (NF<sub>noBC</sub>-NF<sub>H</sub> and NF<sub>V</sub>-NF<sub>H</sub>) represents first before jumping to the results.

**Response:** Thanks for your suggestion. We have added the definition of these abbreviations as:

**L427-434:** “The difference between NF<sub>noBC</sub> and NF<sub>H</sub> (NF<sub>noBC</sub>-NF<sub>H</sub>) had significant positive correlations with MF<sub>FFOA</sub> and MF<sub>BBOA</sub> ( $r > 0.5$ ), suggesting that a substantial proportion of POA resided in BC-free aerosols and was volatile but contributed substantially to nearly hydrophobic aerosols. So did the difference between NF<sub>V</sub> and NF<sub>H</sub> (NF<sub>V</sub>-NF<sub>H</sub>). The mass fractions of BBOA and FFOA were poorly linked with the difference between MAF and NF<sub>V</sub> (MAF-NF<sub>V</sub>), or MAF and NF<sub>noBC</sub> (MAF-NF<sub>noBC</sub>), or NF<sub>V</sub> and NF<sub>noBC</sub> (NF<sub>V</sub>-NF<sub>noBC</sub>) (Fig. S7). The difference between MAF and NF<sub>H</sub> (MAF-NF<sub>H</sub>) had a positive correlation with MF<sub>BBOA</sub>, further suggesting BBOA contributed to nearly hydrophobic aerosols under subsaturated conditions, however, their hygroscopicity

was enhanced and became CCN-active at supersaturated conditions.”

**L464:** “Difference between  $NF_{noBC}$  and  $NF_H$  ( $NF_{noBC}-NF_H$ ) showed a strong negative correlation with  $MF_{NH4}$  and  $MF_{NO3}$ . So did the Difference between  $NF_V$  and  $NF_H$  ( $NF_V-NF_H$ ). So did the difference between  $NF_V$  and  $NF_H$  ( $NF_V-NF_H$ ).”

**L483:** “The difference between  $NF_{noBC}$  and  $NF_V$  ( $NF_{noBC}-NF_V$ ) is negatively correlated with  $MF_{NO3}$ , which is consistent with the semi-volatile nature of nitrate.”

**L555:** “... the two resolved SOA factors exhibited different impacts on the difference between  $NF_V$  and  $NF_H$  ( $NF_V-NF_H$ ), ...”

**Figure 10:** “.....(d and e) The variations of the difference between  $NF_V$  and  $NF_{noBC}$  ( $NF_V-NF_{noBC}$ , blue large circle) and the difference between  $NF_V$  and  $NF_{noBC}+NF_{CBC}$  ( $NF_V-(NF_{noBC}+NF_{CBC})$ , yellow small circle) with the mass concentration of SA at particle size of 200nm (d) and 300nm (e). MAF (Maximum Activation Fraction): An asymptote of the measured SPAR curve at large particle.  $NF_H$ : Number Fraction of Hydrophilic aerosol whose hygroscopicity parameter is higher than  $\sim 0.07$ .  $NF_V$ : Number Fraction of Volatile aerosol whose Shrink Factor at 200 °C is lower than 0.85.  $NF_{CBC}$ : Number Fraction of thickly coated BC particles.”

32) *Line 438, why do you choose 200nm?*

**Response:** This is mainly because we focus on the comparison of the four aerosol mixing state as well as their relationship with aerosol chemical compositions, but only in 200 nm were all the four aerosol mixing state parameters measured. We have added corresponding description into the manuscript as:

“To be noted, in order to compare the four aerosol mixing state as well as their relationships with aerosol chemical compositions at the same time, the analysis is conducted in only 200 nm where all the four aerosol mixing state parameters were measured.”

33) *Line 459-462, out of curiosity, does the transport of ageing aerosols play a role in the increasing fraction of non-BC particles?*

**Response:** The reviewer raised a very interesting topic. Indeed, the transport of aging aerosols could play a role in variations in fraction of non-BC particles, for example, during the clean period. However, for periods of the moderately to heavily polluted, the wind speed generally lower than 2 m/s, with strong local emissions (represented quick increase of rBC and POA in the afternoon) of secondary aerosols formations (represented by quick nitrate and SOA formations), the transport of aging aerosols

should play a negligible role.

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