

Editor:

As also noted by the referees the paper has improved substantially, however, there is still more to be done (hence major revision with additional review by the referees)

Response: Thanks for your comments. Thank you for your feedback and for taking the time to review our manuscript, we really appreciate your insightful comments

Suggestions and comments are addressed point-by-point and corresponding responses are listed below.

Referee 2 gives some hints, please consider them with the exception mentioned below; further comments are given in the following:

- I agree that the paper lists observations on similarities and differences in the various fractions, however, more needs to be said what is new, what is expected, and what is in contrast to expectations. You should also give recommendations for future research with such a setup, and list the advantages and disadvantages.

Response: Thanks for your comments. We have added some discussions in sections 3.3 and 3.4 to better claim what is new and what is expected as follow:

L397: We have added a sentence as:

“This may be due to the high relative humidity during the pollution period, and the formation of SA occurs mainly in the aqueous phase, which contributes to the formation of particles with larger diameters (accumulation-mode and droplet-mode particles, Kuang et al., 2020).”

L503: We have added a paragraph as:

In general, both field and laboratory studies have shown that primary organic aerosols from the combustion of biomass and fossil fuels are less hygroscopic. In laboratory experiment, it is found that organic aerosols produced by fossil fuels have very low hygroscopicity, significantly less than 0.1 (Vu et al., 2015, 2017; Fofie et al., 2018; Zhang et al., 2018; Mukherjee et al., 2021). Observations have also found that the organic aerosols associated with fossil fuel combustion have low hygroscopicity, which may be due to the low soluble components in FFOA (Qiu et al., 2019; Li et al., 2021). The aerosol composition produced by biomass burning is complex, with a large number of organic aerosols (BBOA) and inorganic components being produced at the initial stage, making important contributions to CCN (Spracklen et al., 2011; Bougiatioti et al., 2016; Po’hlker et al., 2018). These primary organic aerosols (i.e.

BBOA) is generally semi-volatile (May et al. 2013) and less hygroscopic (Engelhart et al., 2012; Hennigan et al., 2012), which has a negative contribution to overall hygroscopicity (Bougiatioti et al., 2016; Kuang et al., 2020b, 2021, Cai et al., 2022), resulting in weaker overall aerosol hygroscopicity in the initial stage of the biomass burning (Engelhart et al., 2012, Hennigan et al., 2012, Po'hlker et al., 2018). However, laboratory experiments found that BBOA may contain organic substances with different hygroscopicity under different saturation ratios (Malek et al., 2022), lead to increased hygroscopicity and enhanced CCN activity of BBOA under supersaturation conditions (Hersey et al., 2013). Our results generally agree with previous studies and provide evidences of the enhanced CCN activity of BBOA under supersaturation conditions in field campaigns. Furthermore, the different impacts of aerosols emitted from biomass burning and fossil fuel combustion on CCN is directly observed in this campaign used newly developed advanced aerosol-cloud sampling system, which show that biomass burning aerosols are efficient CCN even under low supersaturations (<0.05%), however, aerosols from fossil fuel combustions can only activate at higher supersaturations (~>0.14%). These results suggest simultaneous measurements of aerosol GF distributions, SPAR curves and BC mixing states and their comparisons could shed novel insights into different synergistic hygroscopic, volatile and activation properties of aerosols from different sources in the atmosphere.

L530: We have added sentences as:

“Recent studies reported that catalyst or photochemical reactions on BC particles can contribute the formation of secondary aerosols (Zhang et al., 2020; Zhang et al., 2021). Our results may indicate SA formation on BC particles might not be a significant pathway that contribute substantially to haze formation, and the underlying mechanisms need to be further resolved.”

L563: We have added a sentence as:

“Our results on OOA agree with previous studies, that OOA are reported to be volatile (Kim et al., 2020; Cai et al., 2022) but can have a positive or negative impact on hygroscopicity depending on its oxidation level (Kim et al., 2020; Kuang et al., 2021; Cai et al., 2022).”

Also, the conclusions are modified to better deliver new findings of this study in the context of existing literatures, and recommendations are also concluded in this part as follow:

“Fossil fuel combustion and biomass burning emissions represent two major primary sources of global aerosol burden and are dominant primary aerosol sources in this campaign. It is known that the chemical compositions of both these primary sources are dominated by organics and BC. However, the intercomparison results among instruments revealed significant differences in the physical and chemical properties of aerosols emitted from these two sources. The combination of HTDMA, DMA-SP2, as well as aerosol source apportionment confirmed that substantial portions of BC-free aerosols from both biomass burning and fossil fuel combustion are nearly hydrophobic under sub-saturated conditions. Additionally, BC from fossil fuel combustion tends to be more externally mixed with other aerosol compositions than those from biomass burning. However, additional insights from DMA-CCN measurements revealed that substantial portions of BC-free aerosols, nearly hydrophobic from biomass burning, could serve as CCN, while a substantial portion of those from fossil fuel combustion could not. Previous studies have confirmed the hygroscopicity difference of aerosols from biomass burning under sub- and supersaturated conditions in laboratory settings; however, such differences have rarely been confirmed in field measurements. Moreover, comparisons between sub- and supersaturated conditions for aerosols from fossil fuel combustion have been rarely undertaken, even in laboratory studies. This finding is quite important because the ability of primary organic aerosols from biomass burning and fossil fuel combustion is often treated as the same in models (Liu et al., 2021; Pöhlker et al., 2023).

Secondary aerosol formation substantially alters aerosol mixing state. The different variations in mixing state parameters can also help reveal mechanisms of secondary aerosol formation. For example, the two resolved SOA factors exhibited different impacts on the differences between N_{F_V} and N_{F_H} ($N_{F_V} - N_{F_H}$), and their correlations with N_{F_V} and N_{F_H} revealed that OOA1 was more hygroscopic but less volatile, suggesting distinct formation mechanisms for these two OOA factors. Further analysis might help link SOA formation mechanisms to aerosol physical

properties, which is important for connecting aerosol chemistry to aerosol climate effects determined by aerosol physicochemical properties. Additionally, variations in size-resolved NF_{noBC} revealed that secondary organic and inorganic aerosol formations led to the migration of BC-free particles towards larger diameters more quickly than that of BC-containing particles. This phenomenon is more likely to occur when aqueous pathways dominate secondary aerosol formation because BC-containing particles generally exhibit weak hygroscopicity and do not favor aqueous processes.

The findings of this study highlight the markedly different effects of primary emissions and SA formation on aerosol mixing states and suggest that comparisons of aerosol mixing states obtained using various techniques are useful for gaining insights into the hygroscopicity, volatility, and CCN activity of different aerosols. Recommendations are listed for future studies based on the findings of this study: (1) When exploring the impact of aerosol emissions and secondary aerosol formations on aerosol hygroscopic under sub- and supersaturated conditions, we recommend employing simultaneous DMA-SP2 measurements to better represent BC characteristics; (2) Simultaneous DMA-CCNC, V-HTDMA, and DMA-SP2 measurements could enhance studies on secondary aerosol formation mechanisms. Conversely, if formation mechanisms and pathways are clear, these measurements could elucidate how secondary aerosol formation impacts aerosol physical properties from different aspects. (3) To be cautious in the application of aerosol mixing state parameters from HV-TDMA to conduct aerosol optical property investigations because the suitability of HV-TDMA-derived mixing state parameters for representing BC mixing states is largely dependent on the composition and mass of the secondary aerosols, and DMA-SP2 measurements are recommended for this purpose.”

- Then, most importantly, there are still a lot of unclear sentences (e.g. L 477-481), or unsupported statements (e.g. L 483), which make it impossible to accept the paper at the current stage.

Response: Thanks for your comments. We have revised these unclear sentences and responded to the corresponding comments later. In addition, we have checked the manuscript and revised the unclear sentences and unsupported statements as follow:

L166: “The flow rate is carefully adjusted in the inlet in order to ensure accurate aerosol particle size cutoff.”

L269: “... when it was placed after a thermodenuder-bypass switch system (during the following time periods: 11:00 am of the 24th of October ...”

L272-273: “Thus, for the same measurement cycle (2h), more particle sizes were selected in the DMA-SP2 system to acquire the BC mass concentration and mixing state at larger diameters than HTDMA and VTDMA.”

- Please check the attached annotated manuscript; these comments are just examples and not meant to be complete.

Response: Thanks for your comments. We have revised the manuscript based on the comments in the attached annotated manuscript.

- In contrast what Referee 2 suggested you are fine with using L as symbol for liter; according to a recent decision by the ACP Editorial board the symbol L will be used for liter in all future publications, to avoid confusion with the capital I.

Response: Thanks for your kind reminding.

- Concerning the number of figures I leave this up to the authors: Please decide what you consider appropriate number of figures, based on the suggestion of Referee 2 in terms of readability of the paper.

Response: Thanks for your comments. After careful consideration, we have decided to retain all the figures in the manuscript. While we acknowledge that some figures may appear similar at first glance, each figure conveys distinct messages and contributes to the comprehensive understanding of our research findings. We believe that retaining all figures is crucial in presenting a thorough analysis and ensuring that all aspects of our study are adequately represented.

1. Abstract: The journal now has new guidelines, see https://www.atmospheric-chemistry-and-physics.net/policies/guidelines_for_authors.html.

Specifically, the abstract should have less than 250 words (but also check the other items of the guidelines)

Response: Thanks for your kind reminder, we have shortened the abstract to less than 250 words as follow:

“This study compares aerosol mixing state parameters obtained via simultaneous measurements using DMA-CCNC, H/V-TDMA, and DMA-SP2, shedding light on the impacts of primary aerosol emissions and secondary aerosol (SA) formation. The analysis reveals significant variations in mixing-state parameters among different techniques, with V-TDMA and DMA-SP2 indicating that non-volatile particles mainly stem from BC-containing aerosols, while a substantial proportion of nearly hydrophobic aerosols originates from fossil fuel combustion and biomass burning emissions. Synthesizing the results, some nearly hydrophobic BC-free particles were found to be CCN-inactive under supersaturated conditions, likely from fossil fuel combustion emissions, while others were CCN-active, linked to biomass burning emissions. Moreover, BC-containing aerosols emitted from fossil fuel combustion exhibit more external mixing with other aerosol components compared to those from biomass burning. Secondary nitrate and organic aerosol formation significantly affect aerosol mixing states, enhancing aerosol hygroscopicity and volatility while reducing heterogeneity among techniques. The study also highlights distinct physical properties of two resolved secondary organic aerosol factors, hinting at formation through different mechanisms. These findings underscore the importance of comparing aerosol mixing states from different techniques as a tool in understanding aerosol physical properties from different sources and their responses to SA formation, as well as aiding in the exploration of SA formation mechanisms.”

2. *English:*

L187: “... by obvious m/z 60 (mainly $C_2H_4O_2^+$) and 73 (mainly $C_3H_5O_2^+$):”

L276: “The BC-containing particles passing through the laser beam **became** incandescent by absorbing radiation.”

L498: *“The difference between MAF-NFH ...”*

Response: Thanks for your comments. We have revised these sentences as:

“by abundant fragments of m/z 60 (mainly C₂H₄O₂⁺) and 73 (mainly C₃H₅O₂⁺)”

“The BC-containing particles passing through the laser beam become incandescent by absorbing radiation.”

“The difference between MAF and NF_H ...”

3. L265: *“... (the 13th to the 24th of October, 09:00 am of the 5th of November to 09:00 am of the 8th of November)”*: Not clear

Response: Thanks for your comments. We are referring to the time periods that DMA-SP2 was not placed after an denuder-bypass switch system and we have revised this sentence as:

“... when it was not placed after an denuder-bypass switch system (during the following time periods: the 13th to the 24th of October, 09:00 am of the 5th of November to 09:00 am of the 8th of November).”

4. L348: *“In contrast, the highest mass concentrations of SOA, POA, and BC reached 10 µg/m³.”* : still not clear: each component beyond 10 or the sum of all three together beyond 10?

Response: Thanks for your comments. It should each component of SOA, POA, and BC reached 10 µg/m³ and we have revised this sentence as:

“the highest mass concentrations of SOA, POA, and BC all reached 10 µg/m³”

5. L390: *“... during the clean period”*: clean period, not cleaning period. Multiple instances

Response: Thanks for your comments. We have revised them accordingly.

6. L393: *“RexBC”*: define at first instance

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

“As for RexBC , which is defined as the number concentration ratio of externally mixed BC particles in total BC-containing particles, the small ...”

7. L407: "... because the SF of this type of volatile BC-containing aerosols has an SF lower than 80/200 ...": 80/200 not defined (it's probably both nm, but this needs to be mentioned)

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

"because this type of volatile BC-containing particles aerosols has an SF lower than 0.4 (=80nm/200nm),"

8. L452-453: "The agreement between MAF and NFV was slightly higher than that between MAF and NFH or between NFH and NFV with similar correlation coefficients (~0.65).": this sentence is not clear, there also seems to be a discrepancy to the sentence before (unless I got it wrong, but then it is indeed not clear)

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

"The agreement between MAF and NF_v was slightly higher than that between MAF and NF_H or between NF_H and NF_v. In detail, compared to the other two, the agreement between MAF and NF_v has a similar correlation coefficients (r~0.65) and a smaller systematic differences (slope and intercept were much closer to 1 and 0, respectively)."

9. L454: "However, smaller systematic differences (slope and intercept) were much closer to 1 and 0, respectively.": not clear

Response: Thanks for your comments. This sentence is wrong and we have revised as mentioned in the response of the comment before.

10. L457: "... whereas the degree of reduction was the lowest ...": what does this mean: still highest correlation between MAF and NFV ?

Response: Yes, the correlation between MAF and NFV is the highest and r is about . We have revised this sentence as:

"whereas the degree of reduction was the lowest for the correlation between MAF and NF_v (r~0.53)."

11. L476-482: "At the same MFFFOA, NFH was obviously lower than NFnoBC (NFH and NFnoBC were larger and smaller than 0.7 when MFFFOA was larger than 0.1), demonstrating that a substantial portion of nearly hydrophobic aerosols was not contributed by BC-containing aerosols (BC-containing aerosols of 200 nm with BC core smaller than 80 nm which is smaller than the detection limit of SP2 likely to be quite aged in the air, thus not

possible to be nearly hydrophobic), but likely by FFOA- or BBOA-dominant aerosols (NF_H also had a negative correlation with MF_{BBOA}).”: sentence not clear. Shorten

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

“At the same MF_{FFOA}, for example, when conditions of MF_{FFOA} >0.1 were met, NF_H (<0.7) demonstrated a noticeable decrease compared to NF_{noBC} (>0.7), and NF_H showed a negative correlation with both MF_{BBOA} and MF_{FFOA}, suggesting that a substantial portion of nearly hydrophobic particles originated from FFOA- or BBOA-dominant rather than BC-containing particles. Additionally, markedly different correlations were observed between MAF and MF_{FFOA} (r=-0.62), and between MAF and MF_{BBOA} (r=-0.2), implying that nearly hydrophobic but CCN-active aerosols likely originated from biomass burning.”

12. L483-484: “... between MAF and MF_{BBOA} (r=-0.2) imply that nearly hydrophobic but CCN-active aerosols were likely contributed by biomass-burning emissions.”: I don't understand this conclusion. You compare MAF and MF_{BBOA} here, nothing about NF_H. If you could still show this with the appropriate data it would need an interpretation as it is quite opposite to the expectations.

Response: Thanks for your comments. We have revised this sentence as shown in the response to the previous comment.

13. L489: “... suggesting that BC-containing aerosols emitted from fossil fuel combustion tended to be more externally mixed with other aerosol components than those emitted from **biomass burning**.”: This is a good example where the finding should be put into the context of previous literature (as requested from a referee). This finding probably relates to the fact (described in the literature) that the BC fraction in fossil fuel combustion emissions is higher than in biomass burning emissions.

Response: Thanks for your comments. In a field campaign in the North China Plain, Zhang et al. (2020) reported that BC-containing particles originated from the source of fossil fuel tends to be more externally mixed than that originated from biomass burning. Thus our finding agree with the previous literature. We have revised this sentence as:

“... BC-containing particles emitted from fossil fuel combustion tended to be more externally mixed with other aerosol components than those emitted from biomass burning, which is consistent with the results of previous studies (Schwarz et al., 2008; Laborde et al., 2013; Liu et al., 2017; Zhang et al., 2020).”

14. L499-501: "... suggesting that BBOA contributed to nearly hydrophobic aerosols under subsaturated conditions; however, their hygroscopicity was enhanced, and they became CCN-active under supersaturated conditions." another finding without an attempt of an explanation. Could this be weakly soluble compounds that are soluble at high LWC, but not at low LWC?

Response: Thanks for your comments. The enhanced hygroscopicity of BBOA under supersaturated conditions may be attributed to:

(1) surface tension lowered by surface-active organic solutes (Hodas et al., 2016; Ruehl et al., 2016);

(2) variations of both particle diameter and surface tension due to liquid–liquid phase separation (Ovadnevaite et al., 2017; Liu et al., 2018);

(3) dissolution of sparingly soluble compounds at higher saturated conditions (Wex et al., 2009; Dusek et al., 2011);

(4) highly viscous organic aerosol which takes up water by surface water adsorption under sub-saturated conditions and by absorption of water under super-saturated conditions (Pajunoja et al., 2015);

We have added a sentence into the manuscript as:

"The enhanced hygroscopicity of BBOA under supersaturated conditions may be attributed to: (1) surface tension lowered by surface-active organic solutes (Hodas et al., 2016; Ruehl et al., 2016); (2) liquid–liquid phase separation (Ovadnevaite et al., 2017; Liu et al., 2018); (3) dissolution of sparingly soluble compounds at higher saturated conditions (Wex et al., 2009; Dusek et al., 2011); (4) highly viscous organic aerosol which takes up water by surface water adsorption under sub-saturated conditions and by absorption of water under super-saturated conditions (Pajunoja et al., 2015)."

15. L512-513: "... the secondary inorganic aerosol components dominated over SA (approximately 50% vs. approximately 70%), ...": Not clear. Do you mean dominated over SOA? And what are these percentages?

Response: We are referring to that SIA rather than SOA dominate SA and the percentage is about 70%. We have revised this sentence as:

"... the secondary inorganic aerosol components dominated SA (the mass ratio between SIA and SA is approximately 70%), ..."

16. L527-530: *“The increase in N_{FnoBC} at 200 nm as a function of the SA MF suggests that SAs migrated to a higher fraction of BC-free aerosols smaller than 200 nm to particle size of 200 nm, highlighting that SAs tended to form more quickly on BC-free aerosols than on BC-containing aerosols.”: What would be the reason for SA condensing more quickly on BC-free aerosols? Partitioning? But this depends also on the chemical composition of the rest of the 200-nm particle; the BC core can be as small as 80 nm. I believe this conclusion is only partly true. A higher mass fraction simply means that the primary particle size (before the condensation) is smaller, hence possibly a smaller BC size, and therefore possibly smaller than the detection limit of the SP2*

Response: Thanks for your comments. We agree with the reviewer that this conclusion is partly true can there can be BC-containing particles with BC cores smaller than the detection limit of the SP2. As shown in Fig.5c of Li et al. (2023), secondary aerosol formations mainly add mass to BC-free particles (particles with BC lower than detection limit is not excluded), and similar results is found in this campaign as shown in Fig. R1. We now are preparing a manuscript to address this phenomenon from the review of chemical mechanisms, the main reason behind this phenomenon is that water uptake abilities of BC-containing particles are generally small while some BC-free particles would uptake a lot of water which provide site for chemical reactions. However, BC-containing particles with BC mass lower than detection limit at 200 nm likely have abundant hygroscopic coating materials which also favor greatly aerosol chemical compositions, therefore we agree with the reviewer that this conclusion is only partly true.

We have revised this part as:

“The increase in N_{FnoBC} at 200 nm as a function of the SA MF suggests that SAs migrated to a higher mass fraction of BC-free particles smaller than 200 nm to particle size of 200 nm, suggesting that SAs tended to form more quickly on BC-free particles than on BC-containing particles with BC higher than SP2 detection limit.”

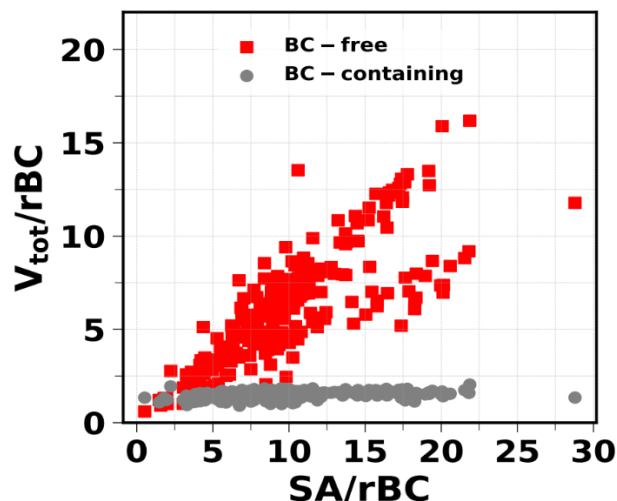


Figure R1: Scatter plots of SA / rBC and the ratio between total volume (V_{tot}) of BC-free and BC-containing aerosols to mass concentrations of rBC.

17. L557-558: “... and is likely formed mainly on BC-free particles.”: see comment above on BC-free particles

Response: Thanks for your comments. Based on the response before, we have revised this sentence as:

“...indicates that the difference is smaller when there is more OOA2, implying that OOA2 is also a semi-volatile compound and is likely formed mainly on BC-free particles (particles with BC mass lower than detection limit are not excluded).”

18. L571-572: “significantly enhanced aerosol mixing state parameters”: not clear. A mixing state parameter cannot be enhanced

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

“this increase in SAs significantly enlarged the value of aerosol mixing state parameters”

19: L598: “However, the differences between these mixing state parameters vary significantly under different conditions.” : use consistent past or present tense

Response: Thanks for your comments. We have revised it accordingly

Reviewer #1:

General comments:

Thanks for the revision. The paper looks much improved, now easy to follow, and readable to non-expert audiences. I agree with the author's argument that the correlation is not causality but this could be the best available way to understand the interlinks among various chemical-physical properties. I have some further questions as below and would suggest accepting with a minor revision.

Response: Thanks for your comments, we really appreciate for your time and careful inspection of our manuscript. Suggestions and comments are addressed point-by-point and corresponding responses are listed below.

1. Regarding the two OOA factors from PMF, are they related to Less-oxygenated and More-oxygenated OOAs?

Response: Terms of Less-oxygenated and More-oxygenated OOAs were used in previous studies when they have distinct O/C ratios. O/C ratios of OOA1 and OOA2 have a difference, however, their difference is small. Therefore, we used terms of OOA1 and OOA2 to avoid misleading.

2. Reviewer2, comment 8. In the last sentence of your paragraph. ...Thus, for the same measurement cycle (2h), more particle sizes were selected in the DMA-SP2 system to acquire the BC concentration...

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

3. Reviewer2, comment 9. Are you adjusting the flow rate to ensure enough aerosol particles in the inlet? I do agree with you that the flow rate doesn't influence mixing state measurements as it is not influenced by total mass. If yes, I suggested to add 1-2 sentences to clarify in the paper.

Response: We adjust the flow rate in the inlet in order to ensure accurate aerosol particle size cutoff. We have added these sentences into the manuscript as:

“The flow rate is carefully adjusted in the inlet in order to ensure accurate aerosol particle size cutoff.”

4. Reviewer2, comment 20. According to Fig1 in Petters and Kreidenweis (2007), to activate a particle with kappa of 0.07, the aerosol particle size should be around 200-300nm. What are the composition and sources of these aerosol particles?

Response: In the NCP, aerosol with hygroscopicity kappa lower than 0.07 are likely to be dominated by BC and primary organic aerosol, from biomass and fuel combustion (Tao et al., 2021; Shi et al., 2022).

Reviewer #2:

General comments:

The manuscript has improved significantly compared to the first version. Thanks a lot for the improvement!

However, there is still some work, which needs to be done, mainly in the discussion of the results in section 3. Here, a lot of text describes too many similar looking figures. It needs to be indicated, what is really new. Other detailed comments are given below.

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

Furthermore, this study claims to use for the first time all these measurements and methods in parallel. But, what is the outcome? Which methods are comparable? Are all of them needed? E.g., if one has to reduce to setup to 2 mixing state parameters, which would you recommend? Any other general conclusions regarding the methodology?

Response: Thanks for your comments. We have added some discussions in sections 3.3 and 3.4 to better claim what is new and what is expected in the context of existing literatures.

Since this is definitely a new approach, it should be discussed and interpreted in the conclusion. Please also indicates in the conclusions which findings are new or you assume them to be new. There are too many 'findings' listed and the reader does not know, what is important.

Response: Thanks for your comments. Conclusions are modified to better deliver new and important findings of this study in the context of existing literatures, and recommendations are also concluded in this part as follow:

“Fossil fuel combustion and biomass burning emissions represent two major primary sources of global aerosol burden and are dominant primary aerosol sources in this campaign. It is known that the chemical compositions of both these primary sources are dominated by organics and BC. However, the intercomparison results among instruments revealed significant differences in the physical and chemical properties of aerosols emitted from these two sources. The combination of HTDMA, DMA-SP2, as well as aerosol source apportionment confirmed that substantial

portions of BC-free aerosols from both biomass burning and fossil fuel combustion are nearly hydrophobic under sub-saturated conditions. Additionally, BC from fossil fuel combustion tends to be more externally mixed with other aerosol compositions than those from biomass burning. However, additional insights from DMA-CCN measurements revealed that substantial portions of BC-free aerosols, nearly hydrophobic from biomass burning, could serve as CCN, while a substantial portion of those from fossil fuel combustion could not. Previous studies have confirmed the hygroscopicity difference of aerosols from biomass burning under sub- and supersaturated conditions in laboratory settings; however, such differences have rarely been confirmed in field measurements. Moreover, comparisons between sub- and supersaturated conditions for aerosols from fossil fuel combustion have been rarely undertaken, even in laboratory studies. This finding is quite important because the ability of primary organic aerosols from biomass burning and fossil fuel combustion is often treated as the same in models (Liu et al., 2021; Pöhlker et al., 2023).

Secondary aerosol formation substantially alters aerosol mixing state. The different variations in mixing state parameters can also help reveal mechanisms of secondary aerosol formation. For example, the two resolved SOA factors exhibited different impacts on the differences between NF_V and NF_H ($NF_V - NF_H$), and their correlations with NF_V and NF_H revealed that OOA1 was more hygroscopic but less volatile, suggesting distinct formation mechanisms for these two OOA factors. Further analysis might help link SOA formation mechanisms to aerosol physical properties, which is important for connecting aerosol chemistry to aerosol climate effects determined by aerosol physicochemical properties. Additionally, variations in size-resolved NF_{noBC} revealed that secondary organic and inorganic aerosol formations led to the migration of BC-free particles towards larger diameters more quickly than that of BC-containing particles. This phenomenon is more likely to occur when aqueous pathways dominate secondary aerosol formation because BC-

containing particles generally exhibit weak hygroscopicity and do not favor aqueous processes.

The findings of this study highlight the markedly different effects of primary emissions and SA formation on aerosol mixing states and suggest that comparisons of aerosol mixing states obtained using various techniques are useful for gaining insights into the hygroscopicity, volatility, and CCN activity of different aerosols. Recommendations are listed for future studies based on the findings of this study: (1) When exploring the impact of aerosol emissions and secondary aerosol formations on aerosol hygroscopic under sub- and supersaturated conditions, we recommend employing simultaneous DMA-SP2 measurements to better represent BC characteristics; (2) Simultaneous DMA-CCNC, V-HTDMA, and DMA-SP2 measurements could enhance studies on secondary aerosol formation mechanisms. Conversely, if formation mechanisms and pathways are clear, these measurements could elucidate how secondary aerosol formation impacts aerosol physical properties from different aspects. (3) To be cautious in the application of aerosol mixing state parameters from HV-TDMA to conduct aerosol optical property investigations because the suitability of HV-TDMA-derived mixing state parameters for representing BC mixing states is largely dependent on the composition and mass of the secondary aerosols, and DMA-SP2 measurements are recommended for this purpose.”

One general formal comment. The unit liter has the abbreviation ‘l’, not the capital ‘L’. This should be corrected through the whole text.

Response: According to a recent decision by the ACP Editorial board, the symbol L will be used for liter in all future publications to avoid confusion with the capital I.

The table with abbreviations also helps a lot. But could you please put it in an alphabetical order? This would be even better! And the term ‘SA’ is missing there. Maybe also indicate which parameters refer to certain diameters?!

Response: Thanks for your suggestions. We have revised this table as:

Table 1. Definition and description of abbreviations.

Abbreviation	Full name and/or Definition
	Biomass Burning Organic Aerosol
BBOA	Characterized by obvious m/z 60 (mainly C₂H₄O₂⁺) and 73 (mainly C₃H₅O₂⁺), which are two indicators of biomass burning
D_a	Midpoint activation diameter
	Linked to the hygroscopicity of CCNs
D_d	Particle diameter under dry conditions without humidification or heating
D_p	Particle diameter after humidification or heating
	Growth factor
GF	The ratio between particles with and without humidification and is linked to aerosol hygroscopicity
κ	Hygroscopicity parameter
MF	Mass Fraction
	Maximum Activation Fraction
MAF	An asymptote of the measured SPAR curve at large particle sizes and represents the number fraction of CCNs to total particles
	Number Fraction of Hydrophilic aerosol whose hygroscopicity parameter is >~0.07 at particle size of 50, 100, 150 and 200 nm
NF_H	
	Number Fraction of Volatile aerosol whose Shrinkage Factor at 200 °C is <0.85 at particle size of 50, 100, 150 and 200 nm
NF_V	
	Number Fraction of black carbon (BC)-free particles at particle size of 200, 250, 300 and 370 nm
NF_{noBC}	
	Number Fraction of thickly coated BC particles at particle size of 200, 250, 300 and 370 nm
NF_{CBC}	
NF_A-NF_B	
(NF_{noBC}-NF_H, NF_V-NF_H, NF_{noBC}-NF_V, NF_V-MAF, NF_{noBC}-MAF)	The difference between the number fraction of A and B at particle size of 200 nm
OOA1 and OOA2	Two OOA factors resolved from the PMF analysis
PDF	Probability Distribution Function
PM_{2.5}	Particulate Matter with an aerodynamic diameter <2.5 μm
PM₁	Particulate Matter with an aerodynamic diameter <1 μm
	Primary Organic Aerosol
POA	Summation of BBOA and FFOA
	The number concentration ratio of externally mixed BC particles in total BC-containing particles
R_{exBC}	

	Externally mixed BC particles are defined as identified bare/thinly coated BC-containing particles at particle size of 200, 250, 300 and 370 nm
SA	Secondary Aerosols, including nitrate, sulfate, ammonium and the two OOA factors Shrinkage Factor
SF	The ratio between particles with and without heating and is linked to aerosol volatility
SIA	Secondary Inorganic Aerosols, including nitrate, sulfate, and ammonium
SOA	Secondary Organic Aerosols, including the two OOA factors
SPAR	Size-resolved Particle Activation Ratio Size-dependent CCN activity under a specific SS

Comments in detail:

Line number in the following mean the corresponding lines in the manuscript with tracked changes.

Line 158 – 160: please keep the old version to recognize the origin of the abbreviation.

Response: Thanks for your suggestion. We have revised it accordingly.

Line 223 ff: Do you think that AMS measurements and PNSD experience similar losses? If you say, that they agree well, this is hypothetical to my impression.

Response: Thanks for your comments. The agreement between AMS measurements and PNSD is confirmed in their comparison as shown in Figure S3:

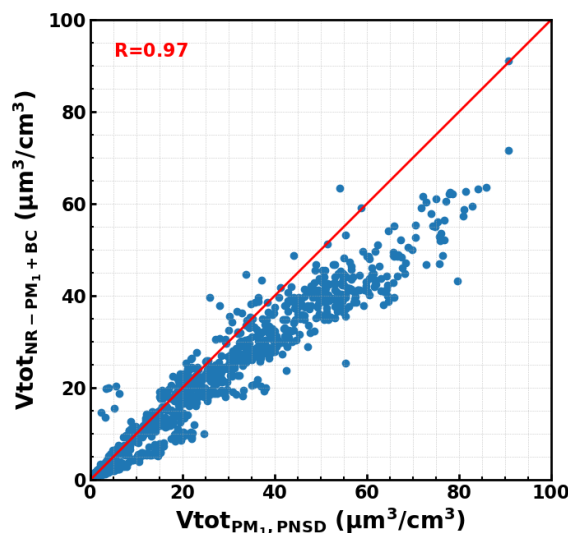


Fig. S3. Comparison between aerosol volume concentration derived from measurements of PNSD and aerosol chemical compositions.

Line 346: How did you choose e.g., the critical GF? Did you plot for each diameter the PDF? Other studies used a common GFc for all diameters, why do you think this is different here? Does this correspond to Figure 2c?

Response: Thanks for your comments. The critical GFs are determined based on the GF PDF for each diameter. The critical GF in this study is different from those in other studies, because there is difference in aerosol micro-physical properties and we want to distinguish between different aerosol groups for comparison with aerosol groups inferred from measurements of other instruments. We have added corresponding description into the manuscript as:

“These values of GFC and SFC divide the probability density functions (PDFs) of SF and GF into two modes as shown in Figure 2c and 2d, consistent with prior NCP studies (Liu et al., 2011; Zhang et al., 2016), and may be different from those GFC and SFC in other studies because of the difference in aerosol micro-physical properties.”

Line 361 and many other times: coating thickness of aerosols: it might be that I am a bit too picky here, but I think a coating thickness is always related to aerosol particles and not to aerosols (mixture of gas and particles), therefore I suggest to check the usage of the word ‘aerosols’. I think in most cases it should be ‘aerosol particles’.

Response: Thanks for your suggestion. We have checked the usage of the word ‘aerosols’ and revised the following:

“BC-containing aerosols” to “BC-containing particles”

“BC-free aerosols” to “BC-free particles”

Line 375, 425 and caption of Figure 1 and 3: The word ‘compositions’ should be ‘components’. I do not see too much sense in ‘mass concentrations of aerosol composition’

Response: Thanks for your comments. We have revised them accordingly.

Figures 4 – 9: Similar parameters are plotted, but I personally think, there are too many figures. For some the particle diameter (200 nm) is given, for others not in the caption. Please add this information in the figure caption. Are all figures really necessary? It would

be better to exclude one or two from the main paper or combine some of the results. The reader feels a bit overloaded with so many scatter dots.

Response: Thanks for your comments. After careful consideration, we have decided to retain all the figures in the manuscript. While we acknowledge that some figures may appear similar at first glance, each figure conveys distinct messages and contributes to the comprehensive understanding of our research findings. We believe that retaining all figures is crucial in presenting a thorough analysis and ensuring that all aspects of our study are adequately represented.

In addition, we have taken your suggestion into account and will ensure that each figure caption includes essential information, such as the particle diameter, for clarity and consistency.

Line 532 – 538: The sentence is too long and not understandable for me. Please avoid such long sentences with too much additional information in brackets.

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

“At the same MF_{FFOA} , for example, when conditions of $MF_{FFOA} > 0.1$ were met, $NF_H (< 0.7)$ demonstrated a noticeable decrease compared to $NF_{noBC} (> 0.7)$, and NF_H showed a negative correlation with both MF_{BBOA} and MF_{FFOA} , suggesting that a substantial portion of nearly hydrophobic particles originated from FFOA- or BBOA-dominant rather than BC-containing particles. Additionally, markedly different correlations were observed between MAF and MF_{FFOA} ($r = -0.62$), and between MAF and MF_{BBOA} ($r = -0.2$), implying that nearly hydrophobic but CCN-active aerosols likely originated from biomass burning.”

Line 569: the word ‘compositions’ should be components, as written in the corresponding caption of Figure 8.

Response: Thanks for your comments. We have revised it accordingly.

Line 633: better ‘components’ instead of ‘compositions’ Line 640: please use ‘components’ instead of ‘compositions’

Response: Thanks for your comments. We have revised them accordingly.

Section 3, in particular 3.3 and 3.4 are too long and not well connected to other studies. What is really new in your study? Many correlations are obvious and well-known, here you

should compare with literature. E.g., that SA increases the hygroscopicity of hydrophobic particles is not new. There are similar examples. I would strongly recommend to remove some of the figures and reduce the text. For the results indicate the well-known facts with references or remove them and highlight those results which are new or opposite to former findings.

Response: Thanks for your comments. We have added discussions about the our results in the context of existing literatures as shown below. In addition, as mentioned in the response to the comment earlier, we have decided to retain all the figures in the manuscript after careful consideration and we have also revised the conclusion to better deliver new and important findings of this study.

L397: We have added a sentence as:

“This may be due to the high relative humidity during the pollution period, and the formation of SA occurs mainly in the aqueous phase, which contributes to the formation of particles with larger diameters (accumulation-mode and coarse-mode particles, Kuang et al., 2020).”

L489: We have revised this sentence as:

“... BC-containing particles emitted from fossil fuel combustion tended to be more externally mixed with other aerosol components than those emitted from biomass burning, which is consistent with the results of previous studies (Schwarz et al., 2008; Laborde et al., 2013; Liu et al., 2017; Zhang et al., 2020).”

L499-501: We have added a sentence as:

“The enhanced hygroscopicity of BBOA under supersaturated conditions may be attributed to: (1) surface tension lowered by surface-active organic solutes (Hodas et al., 2016; Ruehl et al., 2016); (2) liquid–liquid phase separation (Ovadnevaite et al., 2017; Liu et al., 2018); (3) dissolution of sparingly soluble compounds at higher saturated conditions (Wex et al., 2009; Dusek et al., 2011); (4) highly viscous organic aerosol which takes up water by surface water adsorption under sub-saturated conditions and by absorption of water under super-saturated conditions (Pajunoja et al., 2015).”

L503: We have added a paragraph as:

“In general, both field and laboratory studies have shown that primary organic aerosols from the combustion of biomass and fossil fuels are less hygroscopic. In laboratory experiment, it is found that organic aerosols produced by fossil fuels have very low hygroscopicity, significantly less than 0.1 (Vu et al., 2015, 2017; Fofie et al., 2018; Zhang et al., 2018; Mukherjee et al., 2021). Observations have also found that the organic aerosols associated with fossil fuel combustion have low hygroscopicity,

which may be due to the low soluble components in FFOA (Qiu et al., 2019; Li et al., 2021). The aerosol composition produced by biomass burning is complex, with a large number of organic aerosols (BBOA) and inorganic components being produced at the initial stage, making important contributions to CCN (Spracklen et al., 2011; Bougiatioti et al., 2016; Pöhlker et al., 2018). These primary organic aerosols (i.e. BBOA) is generally semi-volatile (May et al. 2013) and less hygroscopic (Engelhart et al., 2012; Hennigan et al., 2012), which has a negative contribution to overall hygroscopicity (Bougiatioti et al., 2016; Kuang et al., 2020b, 2021, Cai et al., 2022), resulting in weaker overall aerosol hygroscopicity in the initial stage of the biomass burning (Engelhart et al., 2012, Hennigan et al., 2012, Pöhlker et al., 2018). However, laboratory experiments found that BBOA may contain organic substances with different hygroscopicity under different saturation ratios (Malek et al., 2022), lead to increased hygroscopicity and enhanced CCN activity of BBOA under supersaturation conditions (Hersey et al., 2013). Our results generally agree with previous studies and provide evidences of the enhanced CCN activity of BBOA under supersaturation conditions in field campaigns. Furthermore, the different impacts of aerosols emitted from biomass burning and fossil fuel combustion on CCN is directly observed in this campaign used newly developed advanced aerosol-cloud sampling system, which show that biomass burning aerosols are efficient CCN even under low supersaturations (<0.05%), however, aerosols from fossil fuel combustions can only activate at higher supersaturations (~>0.14%). These results suggest simultaneous measurements of aerosol GF distributions, SPAR curves and BC mixing states and their comparisons could shed novel insights into different synergistic hygroscopic, volatile and activation properties of aerosols from different sources in the atmosphere.”

L530: We have added sentences as:

“Recent studies reported that catalyst or photochemical reactions on BC particles can contribute the formation of secondary aerosols (Zhang et al., 2020; Zhang et al., 2021). Our results may indicate SA formation on BC particles might not be a significant pathway that contribute substantially to haze formation, and the underlying mechanisms need to be further resolved.”

L563: We have added a sentence as:

“Our results on OOA agree with previous studies, that OOA are reported to be volatile (Kim et al., 2020; Cai et al., 2022) but can have a positive or negative impact on hygroscopicity depending on its oxidation level (Kim et al., 2020; Kuang et al., 2021; Cai et al., 2022).”

Is it really necessary to jump always between NF and MF? This is very confusing for the reader. In between, the word 'fraction' is used and nobody knows, what you mean here, e.g. line 604. Please also state clearly which parameters are related to certain diameters or diameter ranges. This is not clear in the text.

Response: Thanks for your comments and suggestions. This part revised as:

“The difference between NF_{noBC} and NF_H ($NF_{noBC}-NF_H$) showed a strong negative correlation with MF_{NH_4} and MF_{NO_3} (mainly -0.6), as did the differences between NF_v and NF_H (NF_v-NF_H). Ammonium nitrate is a pure-scattering semi-volatile compound with strong hygroscopicity, the increase of its mass fraction can enhance can increase both aerosol volatility and hygroscopicity, therefore resulting in a smaller difference between NF_{noBC} , NF_H , and NF_v .”

▪ **Reference:**

Bougiatioti, A., Bezantakos, S., Stavroulas, I., Kalivitis, N., Kokkalis, P., Biskos, G., Mihalopoulos, N., Papayannis, A., and Nenes, A.: Biomass-burning impact on CCN number, hygroscopicity and cloud formation during summertime in the eastern Mediterranean, *Atmos. Chem. Phys.*, 16, 7389–7409, <https://doi.org/10.5194/acp-16-7389-2016>, 2016.

Cai, M., Huang, S., Liang, B., Sun, Q., Liu, L., Yuan, B., Shao, M., Hu, W., Chen, W., Song, Q., Li, W., Peng, Y., Wang, Z., Chen, D., Tan, H., Xu, H., Li, F., Deng, X., Deng, T., Sun, J., and Zhao, J.: Measurement report: Distinct size dependence and diurnal variation in organic aerosol hygroscopicity, volatility, and cloud condensation nuclei activity at a rural site in the Pearl River Delta (PRD) region, China, *Atmos. Chem. Phys.*, 22, 8117–8136, <https://doi.org/10.5194/acp-22-8117-2022>, 2022.

Dusek, U., Frank, G.P., et al., 2011. Water uptake by biomass burning aerosol at sub- and supersaturated conditions: closure studies and implications for the role of organics. *Atmos. Chem. Phys.* 11 (18), 9519–9532.

Engelhart, G. J., Hennigan, C. J., Miracolo, M. A., Robinson, A. L., and Pandis, S. N.: Cloud condensation nuclei activity of fresh primary and aged biomass burning aerosol, *Atmos. Chem. Phys.*, 12, 7285–7293, <https://doi.org/10.5194/acp-12-7285-2012>, 2012.

Fofie, E. A., Donahue, N. M., and Asa-Awuku, A.: Cloud condensation nuclei activity and droplet formation of primary and secondary organic aerosol mixtures, *Aerosol Science and Technology*, 52, 242–251, <https://doi.org/10.1080/02786826.2017.1392480>, 2018.

Hennigan, C. J., Westervelt, D. M., Riipinen, I., Engelhart, G. J., Lee, T., Collett Jr., J. L., Pandis, S. N., Adams, P. J., and Robinson, A. L.: New particle formation and growth in biomass burning plumes: An important source of cloud condensation nuclei, *Geophysical Research Letters*, 39, <https://doi.org/10.1029/2012GL050930>, 2012.

Hersey, S. P., Craven, J. S., Metcalf, A. R., Lin, J., Lathem, T., Suski, K. J., Cahill, J. F., Duong, H. T., Sorooshian, A., Jonsson, H. H., Shiraiwa, M., Zuend, A., Nenes, A., Prather, K. A., Flagan, R. C., and Seinfeld, J. H.: Composition and hygroscopicity of the Los Angeles Aerosol: CalNex, *Journal of Geophysical Research: Atmospheres*, 118, 3016–3036, <https://doi.org/10.1002/jgrd.50307>, 2013.

Hodas, N., Zuend, A., et al., 2016. Discontinuities in hygroscopic growth below and above water saturation for laboratory surrogates of oligomers in organic atmospheric aerosols. *Atmos. Chem. Phys. Discuss.* 1–34, 2016.

Kuang, Y., He, Y., Xu, W., Yuan, B., Zhang, G., Ma, Z., Wu, C., Wang, C., Wang, S., Zhang, S., Tao, J., Ma, N., Su, H., Cheng, Y., Shao, M., and Sun, Y.: Photochemical

Aqueous-Phase Reactions Induce Rapid Daytime Formation of Oxygenated Organic Aerosol on the North China Plain, *Environmental Science & Technology*, 54, 3849–3860, <https://doi.org/10.1021/acs.est.9b06836>, 2020.

Kuang, Y., Huang, S., Xue, B., Luo, B., Song, Q., Chen, W., Hu, W., Li, W., Zhao, P., Cai, M., Peng, Y., Qi, J., Li, T., Wang, S., Chen, D., Yue, D., Yuan, B., and Shao, M.: Contrasting effects of secondary organic aerosol formations on organic aerosol hygroscopicity, *Atmos. Chem. Phys.*, 21, 10375–10391, <https://doi.org/10.5194/acp-21-10375-2021>, 2021.

Laborde, M.; Crippa, M.; Tritscher, T.; Jurañyi, Z.; Decarlo, P. F.; Temime-Roussel, B.; Marchand, N.; Eckhardt, S.; Stohl, A.; Baltensperger, U.; Prévôt, A. S. H.; Weingartner, E.; Gysel, M. Black carbon physical properties and mixing state in the European megacity Paris. *Atmos. Chem. Phys.* 2013, 13 (11), 5831–5856.

Li, F., Luo, B., Zhai, M., Liu, L., Zhao, G., Xu, H., Deng, T., Deng, X., Tan, H., Kuang, Y., and Zhao, J.: Black carbon content of traffic emissions significantly impacts black carbon mass size distributions and mixing states, *Atmos. Chem. Phys.*, 23, 6545–6558, [10.5194/acp-23-6545-2023](https://doi.org/10.5194/acp-23-6545-2023), 2023.

Liu, D.; Whitehead, J.; Alfarra, M. R.; Reyes-Villegas, E.; Spracklen, D. V.; Reddington, C. L.; Kong, S.; Williams, P. I.; Ting, Y.-C.; Haslett, S.; Taylor, J. W.; Flynn, M. J.; Morgan, W. T.; McFiggans, G.; Coe, H.; Allan, J. D. Black-carbon absorption enhancement in the atmosphere determined by particle mixing state. *Nat. Geosci.* 2017, 10 (3), 184–188.

Liu, P., Song, M., et al., 2018. Resolving the mechanisms of hygroscopic growth and cloud condensation nuclei activity for organic particulate matter. *Nat. Commun.* 9.

Liu, P., Kaplan, J. O., Mickley, L. J., Li, Y., Chellman, N. J., Arienzo, M. M., Kodros, J. K., Pierce, J. R., Sigl, M., Freitag, J., Mulvaney, R., Curran, M. A. J., and McConnell, J. R.: Improved estimates of preindustrial biomass burning reduce the magnitude of aerosol climate forcing in the Southern Hemisphere, *Science Advances*, 7, [10.1126/sciadv.abc1379](https://doi.org/10.1126/sciadv.abc1379), 2021.

Malek, K. A., Gohil, K., Al-Abadleh, H. A., and Asa-Awuku, A. A.: Hygroscopicity of polycatechol and polyguaiacol secondary organic aerosol in sub- and supersaturated water vapor environments††Electronic supplementary information (ESI) available: Detailed experimental procedures, and figures and tables showing data analysis. See DOI: [10.1039/d1ea00063b](https://doi.org/10.1039/d1ea00063b), *Environmental Science: Atmospheres*, 2, 24–33, <https://doi.org/10.1039/d1ea00063b>, 2022.

May, A. A., Levin, E. J. T., Hennigan, C. J., Riipinen, I., Lee, T., Collett Jr., J. L., Jimenez, J. L., Kreidenweis, S. M., and Robinson, A. L.: Gas-particle partitioning of primary organic

aerosol emissions: 3. Biomass burning, *Journal of Geophysical Research: Atmospheres*, 118, 11,327-11,338, <https://doi.org/10.1002/jgrd.50828>, 2013.

Mukherjee, S., Anil Kumar, V., Patil, R. D., Meena, G. S., Buchunde, P., Waghmare, V., Deshmukh, S., Dhavale, V., Ray, A., Panicker, A. S., Sonbawne, S. M., Safai, P. D., and Pandithurai, G.: Investigation of physico-chemical characteristics and associated CCN activation for different combustion sources through Chamber experiment approach, *Atmospheric Environment*, 266, 118726, <https://doi.org/10.1016/j.atmosenv.2021.118726>, 2021.

Ovadnevaite, J., Zuend, A., Laaksonen, A., Sanchez, K.J., Roberts, G., Ceburnis, D., Decesari, S., Rinaldi, M., Hodas, N., Facchini, M.C., Seinfeld, J.H., O' Dowd, C., 2017. Surface tension prevails over solute effect in organic-influenced cloud droplet activation. *Nature* 546, 637–641. <https://doi.org/10.1038/nature22806>.

Pajunoja, A., Lambe, A.T., Hakala, J., Rastak, N., Cummings, M.J., Brogan, J.F., Hao, L., Paramonov, M., Hong, J., Prisle, N.L., Malila, J., Romakkaniemi, S., Lehtinen, K.E.J., Laaksonen, A., Kulmala, M., Massoli, P., Onasch, T.B., Donahue, N.M., Riipinen, I., Davidovits, P., Worsnop, D.R., Petäjä, T., Virtanen, A., 2015. Adsorptive uptake of water by semisolid secondary organic aerosols. *Geophys. Res. Lett.* 42, 3063–3068. <https://doi.org/10.1002/2015GL063142>.

Pöhlker, M. L., Ditas, F., Saturno, J., Klimach, T., Hrabě de Angelis, I., Araùjo, A. C., Brito, J., Carbone, S., Cheng, Y., Chi, X., Ditz, R., Gunthe, S. S., Holanda, B. A., Kandler, K., Kesselmeier, J., Könemann, T., Krüger, O. O., Lavrič, J. V., Martin, S. T., Mikhailov, E., Moran-Zuloaga, D., Rizzo, L. V., Rose, D., Su, H., Thalman, R., Walter, D., Wang, J., Wolff, S., Barbosa, H. M. J., Artaxo, P., Andreae, M. O., Pöschl, U., and Pöhlker, C.: Long-term observations of cloud condensation nuclei over the Amazon rain forest – Part 2: Variability and characteristics of biomass burning, long-range transport, and pristine rain forest aerosols, *Atmos. Chem. Phys.*, 18, 10289–10331, <https://doi.org/10.5194/acp-18-10289-2018>, 2018.

Pöhlker, M. L., Pöhlker, C., Quaas, J., Mülmenstädt, J., Pozzer, A., Andreae, M. O., Artaxo, P., Block, K., Coe, H., Ervens, B., Gallimore, P., Gaston, C. J., Gunthe, S. S., Henning, S., Herrmann, H., Krüger, O. O., McFiggans, G., Poulain, L., Raj, S. S., Reyes-Villegas, E., Royer, H. M., Walter, D., Wang, Y., and Pöschl, U.: Global organic and inorganic aerosol hygroscopicity and its effect on radiative forcing, *Nature communications*, 14, 6139, [10.1038/s41467-023-41695-8](https://doi.org/10.1038/s41467-023-41695-8), 2023

Qiu, Y., Xie, Q., Wang, J., Xu, W., Li, L., Wang, Q., Zhao, J., Chen, Y., Chen, Y., Wu, Y., Du, W., Zhou, W., Lee, J., Zhao, C., Ge, X., Fu, P., Wang, Z., Worsnop, D. R., and Sun, Y.: Vertical Characterization and Source Apportionment of Water-Soluble Organic Aerosol with

High-resolution Aerosol Mass Spectrometry in Beijing, China, *ACS Earth Space Chem.*, 3, 273–284, <https://doi.org/10.1021/acsearthspacechem.8b00155>, 2019.

Ruehl, C.R., Davies, J.F., Wilson, K.R., 2016. An interfacial mechanism for cloud droplet formation on organic aerosols. *Science* 351, 1447–1450, 6280.

Schwarz, J. P.; Gao, R. S.; Spackman, J. R.; Watts, L. A.; Thomson, D. S.; Fahey, D. W.; Ryerson, T. B.; Peischl, J.; Holloway, J. S.; Trainer, M.; Frost, G. J.; Baynard, T.; Lack, D. A.; de Gouw, J. A.; Warneke, C.; Del Negro, L. A. Measurement of the mixing state, mass, and optical size of individual black carbon particles in urban and biomass burning emissions. *Geophys. Res. Lett.* 2008, 35, L13810.

Shi, J., Hong, J., Ma, N., Luo, Q., He, Y., Xu, H., et al. (2022). Measurement report: On the difference in aerosol hygroscopicity between high and low relative humidity conditions in the North China Plain. *Atmos. Chem. Phys.*, 22(7), 4599–4613. <https://doi.org/10.5194/acp-22-4599-2022>

Spracklen, D. V., Carslaw, K. S., Poöschl, U., Rap, A., and Forster, P. M.: Global cloud condensation nuclei influenced by carbonaceous combustion aerosol, *Atmos. Chem. Phys.*, 11, 9067–9087, [doi:10.5194/acp-11-9067-2011](https://doi.org/10.5194/acp-11-9067-2011), 2011.

Tao, J., Kuang, Y., Ma, N., Hong, J., Sun, Y., Xu, W., Zhang, Y., He, Y., Luo, Q., Xie, L., Su, H., and Cheng, Y.: Secondary aerosol formation alters CCN activity in the North China Plain, *Atmos. Chem. Phys.*, 21, 7409–7427, <https://doi.org/10.5194/acp-21-7409-2021>, 2021.

Vu, D., Short, D., Karavalakis, G., Durbin, T. D., and Asa-Awuku, A.: Integrating Cloud Condensation Nuclei Predictions with Fast Time Resolved Aerosol Instrumentation to Determine the Hygroscopic Properties of Emissions Over Transient Drive Cycles, *Aerosol Science and Technology*, 49, 1149–1159, <https://doi.org/10.1080/02786826.2015.1105358>, 2015.

Vu, D., Short, D., Karavalakis, G., Durbin, T. D., and Asa-Awuku, A.: Will Aerosol Hygroscopicity Change with Biodiesel, Renewable Diesel Fuels and Emission Control Technologies?, *Environ. Sci. Technol.*, 51, 1580–1586, <https://doi.org/10.1021/acs.est.6b03908>, 2017.

Wex, H., Petters, M.D., et al., 2009. Towards closing the gap between hygroscopic growth and activation for secondary organic aerosol: Part 1-Evidence from measurements. *Atmos. Chem. Phys.* 9 (12), 3987–3997.

Zhang, Y., Yuan, Q., Huang, D., Kong, S., Zhang, J., Wang, X., Lu, C., Shi, Z., Zhang, X., Sun, Y., Wang, Z., Shao, L., Zhu, J., and Li, W.: Direct Observations of Fine Primary Particles From Residential Coal Burning: Insights Into Their Morphology, Composition, and

Hygroscopicity, *Journal of Geophysical Research: Atmospheres*, 123, 12,964-12,979, <https://doi.org/10.1029/2018JD028988>, 2018.

Zhang, Y., Zhang, Q., Yao, Z., & Li, H. (2020). Particle Size and Mixing State of Freshly Emitted Black Carbon from Different Combustion Sources in China. *Environmental Science & Technology*, 54(13), 7766–7774. <https://doi.org/10.1021/acs.est.9b07373>