Response to Reviewer#1

The authors kindly thank the reviewer for the careful review of the manuscript, and the helpful comments and suggestions, which improve the manuscript a lot. After reading the comments from the reviewer, we have carefully revised our manuscript. In the current version, we added more discussions into the text with one new text (Text S4), one new table (Table S2) and five new figures (Figure 5, Figure S1 and Figure S4-6). We highlighted all the revisions and changes in the manuscript and the supporting information with blue color. Our responses to the comments are itemized as below.

Comment:

Li et al. measured water-soluble organic carbon (WSOC) in both gas and particle phases using online and offline measurements over a one-month period in Spring in Shanghai. The author compared measurements between dust storms (DS), haze events, and clean periods. They claimed that high aerosol pH modulated the gas-to-particle partitioning of WSOC during the DS periods and found that DS led to remarkable increases in light absorption due to high concentrations of nitroaromatics, imidazole and water-solute organic nitrogen at the coarse mode.

The study focuses on a short dataset spanning only one month, and the discussion is based solely on the periods of interest that are 3-5 days long. Particularly, they only captured one episode of a dust storm. I do understand that dust storms do not occur with high frequency within one month. My biggest concern is that the levels of detail and discussion are not convincing enough for a publication on ACP. Therefore, I would suggest that the current version of the manuscript be rejected.

Reply: We fully understand the reviewer' concern about the dataset size. Although the periods of the dust storm, pollution event and clean period in this study are only 3-5 days, the sample numbers of each period are 72, 120 and 144, respectively, because the sampling frequency of our instrument is 1-hour (see the manuscript experiment section). For a field measurement on atmospheric aerosol chemistry, a few days of

observation is very common. For example, Wang et al., (2021) did a field investigation on the aqueous production of SOA in Beijing haze from Dec.16-22, 2016. The measurement duration of the Beijing haze is 6 days with a focus on haze chemistry. Another study on sulfate formation mechanism in China haze also used a short time of 1-2 days to investigate the aerosol chemistry during Beijing haze (Wang et al., 2020). Wu et al., (2020) investigated the formation mechanism of ammonium nitrate in Shanghai during a winter dust event. Their sampling duration for the event is less than 5 days from 10/29-11/04, 2019. Lv et al., (2022a, 2022b) investigated the gas-to-particle phase partitioning kinetics of water-soluble organic compounds (WSOC) in Chinese cities (Lv et al., 2022b; Lv et al., 2022a). They also only captured 1-2 two days of episodes for a haze pollution. In western countries researchers have also used a short time to investigate the atmospheric aerosol chemistry sometimes. For example, Campbell et al., (2024) discussed aerosol aqueous formation in Alaska, USA with a focus on a cold haze event that also lasted for less than 5 days. Those studies used an online observation with a time resolution same with ours. One may see that a field observation based solely on the periods of interest that are 3-5 days long is not an unacceptable strategy to explore atmospheric chemistry of an episode such as dust storm and haze event. Therefore, we think the dataset size of this study on the dust storm and haze periods are enough for obtaining a reasonable and convincing conclusion.

We agree with the reviewer that the levels of detail and discussion were not convincing enough. So, we presented more details about the experiment and additional discussions into the manuscript by adding two new texts (Texts S2, S4), one new table (Table S2) and five new figures (Figure 5, Figure S1 and Figures S4-6). Moreover, we also calculated the contributions of specific BrC compounds (eight imidazoles) to light absorption of water-soluble BrC at the coarse modes in NDS and DS periods for further elucidating the enhancing effect of BrC on particles during the Asian dust storm long-range transport. See the details below.

We thank the reviewer for her/his comments, which are very helpful for improving our paper quality.

We highlighted all the revisions and changes in the manuscript and the supporting information with blue color.

Major Comments:

1. Length of the dataset: The study focuses on a dataset with nearly 1 month and discusses three measurement periods of interest (i.e., dust storm, haze event, and clean period). Each measurement period of interest spans 3 to 5 days. Although some of the analyses appear to be technically sound, I believe the sample size is too small to draw a firm conclusion. The current level of discussion is too simplistic and vague to capture the interest of the atmospheric science community. One example is that there are no details about what compounds contribute to the high MAC during periods impacted by dust storms.

Reply: We think that the sample size in this study is enough for drawing a firm conclusion. As we replied before, our instrument time resolution is 1-hour. In other words, the sample numbers of each period including dust storm, haze and clean period we categorized in this study are 72, 120 and 144, respectively. Such a short duration has also been frequently used by many other researchers to study aerosol chemistry of particle pollution events such as dust storm and haze pollution (Wang et al., 2020; Wu et al., 2020; Wang et al., 2021; Lv et al., 2022b; Lv et al., 2022a; Campbell et al., 2024). Thus, we believe it is acceptable that our study focused on a dataset with nearly 1 month and discussed three measurement periods of interest (i.e., dust storm, haze event, and clean period).

We agree with the reviewer's comments that the details about what compounds contribute to the high MAC during periods impacted by dust storms are important. Thus, in the revised version, we made a deep discussion on this issue. Based on those additional discussions, we found NACs and IMs were enriched in the coarse mode through gas-particle partitioning and heterogeneous reactions in dust storm period. Similarly, we also found abundant $WSON_p$ and high MAC values at the coarse mode in dust storm period, confirming that BrC could be formed through gas-particle

partitioning process and heterogenous reactions during the dust particles long range transport. Nitro-aromatic compounds (NACs) light absorption mainly peak at 350-450 nm (Yuan et al. (2020); Liu et al. (2023)). A seen in Figure 4b, in this study MAC was enhanced in the coarse mode at 250-350 nm during the dust storm period. Thus, we quantified the contributions of IMs light absorption to the total BrC, which characteristically peak at 250-300 nm, at the coarse mode in NDS and DS periods. We added those additional discussions into the manuscript as follows. See page 16, lines 374-388 in the revised manuscript.

"From Figure 4b one may further see that the high $MAC_{250\text{-}600}$ values at the coarse mode in the DS period largely occurred at 250-350 nm. As reported by previous studies (Yuan et al., 2020; Liu et al., 2023), light absorption of NACs mainly peaks at a wavelength range large than 350 nm while IMs absorb light mostly at a wavelength less than 300 nm. Thus, here we quantified the contribution of IMs light absorption in the coarse mode during DS period. Based on its concentration and light absorption properties, the light absorption contribution of each individual chromophore to the total BrC in the wavelength range of 250–400 nm was calculated (Text S2). As seen in Figure 5, IMs show two large absorption peaks at wavelengths of 260 and 290 nm, which are mainly associated with the absorption of 2IC and 4IC. Moreover, it can also be seen that IMs light absorption contribution to light absorption of water-soluble BrC in DS is almost twice that of NDS, indicating a significant contribution of aqueous phase formation to BrC in DS. This result further suggests that an enhanced heterogeneous formation of nitrogen-containing organics such IMs on the dust particle surface are responsible for the increased light absorbing of Asian dust during a long-range transport."

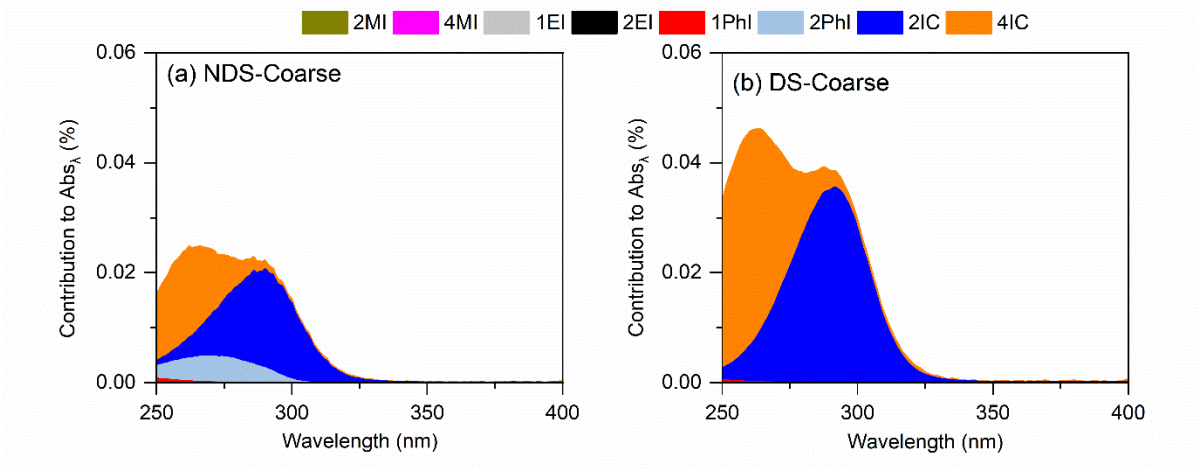

Figure 5. Light absorption contributions of IMs to water-soluble BrC over the wavelength range of 250-400 nm in the coarse mode in NDS and DS periods.

Comments:

2. Table 1: It is good to have Table 1 to summaries the statistics for different measurement periods. When reading the details in Sections 3.1 and 3.2, I found it challenging to keep track of the associated values in Table 1 without searching through the rows one by one. I recommend simplifying Table 1 and presenting only the relevant results in the main text while moving the current version of Table 1 to the supplement.

Reply: Suggestion taken. We simplified Table 1 and moved part of the data to the supplementary materials Table S2. Moreover, we also modified the sequence of the tables. Please see Table 1 in the revised manuscript and Table S2 in the supporting information.

Comments:

3. Sections 3.3 and 3.4: What is the non-dust storm period? Is it equivalent to the haze event plus the clean period? In Sections 3.1 and 3.2, you compare the dust storm with haze event and clean period. However, in sections 3.3 and 3.4, you compare the dust storm periods with non-dust storm periods. Importantly, there is no definition for non-dust storm periods. I am very confused about the consistency between different sections in the manuscript.

Reply: We appreciate the reviewer's suggestion and have added the definition for non-dust storm period in Section 2.1. Please see page 5 lines 108-119.

In section 3.1 and 3.2, we investigated the process of gas-to-particle phase partitioning of WSOC in DS, HE and CP periods from the fine particle (< 2.5 μm) perspective, based on the IGAC-TOC online sampling system (2.5 μm cutoff inlet). We found that the F_p of WSOC was enhanced during DS due to the high pH. In DS, however, the particle is largely dominated by coarse-mode (> 2.1μm). To further elucidate the partitioning processes and formation mechanisms of WSOC, we conducted complementary measurements during DS event (April 11-13) using a 9-stage Anderson impactor (0.4-9.0 μm size range). For comparison, two sets samples were collected during the non-dust storm period (April 7-10). In section 3.3 and 3.4, we investigated WSOC partitioning process, formation mechanism and light absorption of BrC in the fine and coarse modes in the DS period. We found that WSOC partitioning process was enhanced at the coarse mode during the DS period. These results provide crucial mechanistic insights that complement and extend the observations in Sections 3.1-3.2, offering a more comprehensive understanding of WSOC dynamics across different particle size ranges during the dust episodes.

"$PM_{2.5}$ filter samples were collected on a day/night basis by using a high-volume sampler (1.13 m^3 min^{-1}, TISCH Environmental, Inc.) in the spring of 2023 from 27 March to 21 April 2023 (N = 50), while size-segregated aerosols were also collected using an Anderson 9-stage sampler (Thermo Electronic Corporation, USA) at an airflow rate of 28.3 L min^{-1} with cutoff points of 0.43, 0.65, 1.1, 2.1, 3.3, 4.7, 5.8 and 9.0 μm, respectively. In this study four sets of the size-resolved aerosol samples were collected with two sets collected in 11-13 April, 2023, which is a dust storm period (DS) (see discussion later), while other two sets of the size-resolved aerosol samples were collected in other non-dust storm periods (NDS) (7-10 April)."

Comments:

4. F_p: The F_p describes a compound's partitioning between the gas and particle phases. During DS periods, the authors claim the high pH is the main driver when ALWC is

low. By definition, the F_p is governed by the effective saturation vapor pressure and aerosol mass loading (Lutz et al., 2019). When the PM_{10} loading is sufficiently high at the DS periods compared to other measurement periods, how can you be sure that high pH is the only key driver of F_p? Discussions are needed regarding the effect of high aerosol mass loading on F_p during DS periods.

Reply: We thanks the reviewer for the important comment. We added a discussion about effects of aerosol mass loading on the WSOC partitioning process in the revised manuscript. Please see pages 11-12, lines 251-270.

"Apart from the above factors that influence the F_p variations, aerosol mass loading may also affect the partitioning of WSOC (Lutz et al., 2019). According to the Raoult's law, the field gas-particle partitioning coefficient can be described as follows (Pankow, 1994):

$$K_i = \frac{i_{particle}}{i_{gas} M_{org}} \qquad (R1)$$

where K_i is the partitioning constant of i compound, $i_{particle/gas}$ are the particle and gas phase concentrations of compound i, and M_{org} is the concentration of aerosol organic mass ($WSOC_p$ are used here). In this section, our main focus is the F_p of WSOC in $PM_{2.5}$. Thus, i and M_{org} are the concentrations of formic and acetic acids and WSOC in $PM_{2.5}$. By definition, the K_i is governed by the organic aerosol mass loading. However, as shown in Figure S5, the partitioning constants (K_i) of formic and acetic acids did not correlate with $WSOC_p$ in both HE and DS periods, indicating that the effect of aerosol organic mass loading on the uptake of WSOC during the campaign was not significant. As shown in Table 1, the $PM_{2.5}$ and WSOC loadings in DS were comparable to those in HE, although PM_{10} loading was much higher in the dust storm period. Moreover, the pH (4.8 ± 1.5, Table 1) of $PM_{2.5}$ in DS was 1.4 units higher than that (3.4 ± 0.3) in HE, which means that H^+ concentration in DS was one order of magnitude lower than that in HE. According to the equations R2 and R3, the low H^+ concentration in DS was favorable for the organic acid equilibria shifting toward the aerosol aqueous phase. Thus, the impact of pH variation on F_p was much

more significant than organic aerosol mass loading in DS, taking a key role in WSOC partitioning process during the dust storm event.

$$RCOOH(g) \longleftrightarrow RCOOH(aq) \qquad (R2)$$

$$RCOOH(aq) \longleftrightarrow RCOO^- + H^+ \qquad (R3)$$

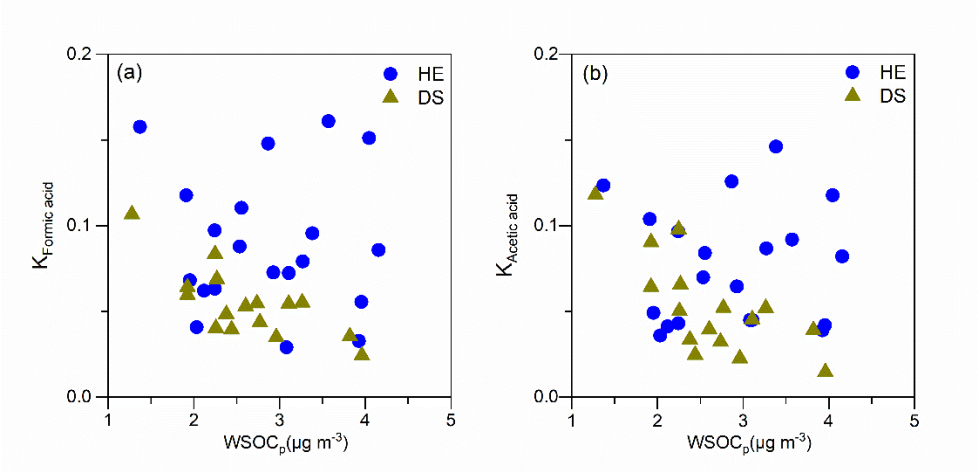

Figure S5. Partitioning coefficients of low molecular organic acids (formic and acetic acids) versus particle phase WSOC during HE and DS periods.

Minor Comments:

1. Lines 52: Specify what the anthropogenic gas pollutants are.

Reply: Suggestion taken. Please see page3 line 51.

"During a long range transport physicochemical properties of dust particles can significantly be changed by a series of chemical reactions especially in the downwind region of East Asia, where anthropogenic gas pollutants such as O_3, NO_x and NH_3 are abundant."

2. Lines 66: What is "such a change in the East Asia atmospheric environment"?

Reply: Thanks for the comment. We have revised the related sentences. See page 3, lines 66-69.

"Such reductions in SO_2 and BC in the East Asia atmospheric environment could also significantly alter the physicochemical evolution process of dust particles during long range transport by adsorption and reactions such as gas-to-particles partitioning of WSOCs and SOA formation on the dust particle surface."

3. Lines 99 and 103: How many $PM_{2.5}$ and PM_{10} filter samples have been collected?

Reply: Thanks for the comment. In this study, we collected 50 $PM_{2.5}$ filter samples in Shanghai, and 3 PM_{10} filter samples in Tengger desert. We have added more detailed information about the samples number in section 2.1. Please see page 5, lines 108-119.

4. Lines 120-127: What are the identified nitroaromatics and eight imidazole compounds?

Reply: Thanks for the comment. In this study, four NACs species were determined, which are 4-nitrophenol (4NP), 2-methoxy-4-nitrophenol (4NGA), 2-methoxy-5-nitrophenol (5NGA), and 5-nitrosalicylicylic acid (5NSA). For imidazoles, a total of eight IMs were determined, which are 2-mechtlimidazole (2MI), 4(5)-methylimidazole (4MI), 1-ethylimidazole (1EI), 2-ethylimidazole (2EI), 1-phenylimidazole (1PhI), 2-phenylimidazole (2PhI), 2-imidazolcarboxaldehyde (2IC) and 4-imidazolcarboxaldehyde (4IC), respectively.

We added the detail description in the revised manuscript in the page 7, lines 144-154.

5. Lines 139–141: Is it your own way of defining a haze episode? I found there is a different definition of haze episodes in Dai et al. (2021). If so, please provide a brief description of how it differs from previous studies.

Reply: Tanks for the comment.

In this study, we defined a haze episode as occurring when the daily $PM_{2.5}$ concentration exceeded 35 μg m^{-3}, which aligns with China's National Air Quality Grade I Standard. However, Dai et al. (2021) adopted a stricter threshold, classifying haze periods based on the National Air Quality Grade II Standard (75 μg m^{-3}). Their study focused on the North China Plain (NCP), one of the world's most heavily polluted regions due to its high population density and numerous emission sources, including traffic and industrial activities. In Dai et al. (2021), $PM_{2.5}$ concentrations

frequently exceeded 75 μg m^{-3}, with peak hourly levels during haze episodes ranging from 198 to 571 μg m^{-3} across their research areas. In contrast, Shanghai exhibits better air quality compared to NCP cities. Our study recorded an average $PM_{2.5}$ concentration of 30 μg m^{-3}, slightly below the Air Quality Grade I Standard of China. Given these lower baseline levels, we maintained the 35 μg m^{-3} threshold to define haze episodes in our analysis.

6. Figure 1: Improvements are needed to enhance the readability of the plot. First, you should provide spaces between each subplot. For some subplots, I am unable to determine whether the y-axis starts at zero. Lines should be thicker, and please avoid using red and green. For Na^+, Mg^{2+}, and Ca^{2+}, I would create a subplot for them. The same applies to NH_4^+ and Cl^-. I cannot identify any trend regarding these species, even with zooming in.

Reply: Suggestion taken. We modified the Figure 1 in accordance to your suggestions. Please see Figure 1 in our revised manuscript.

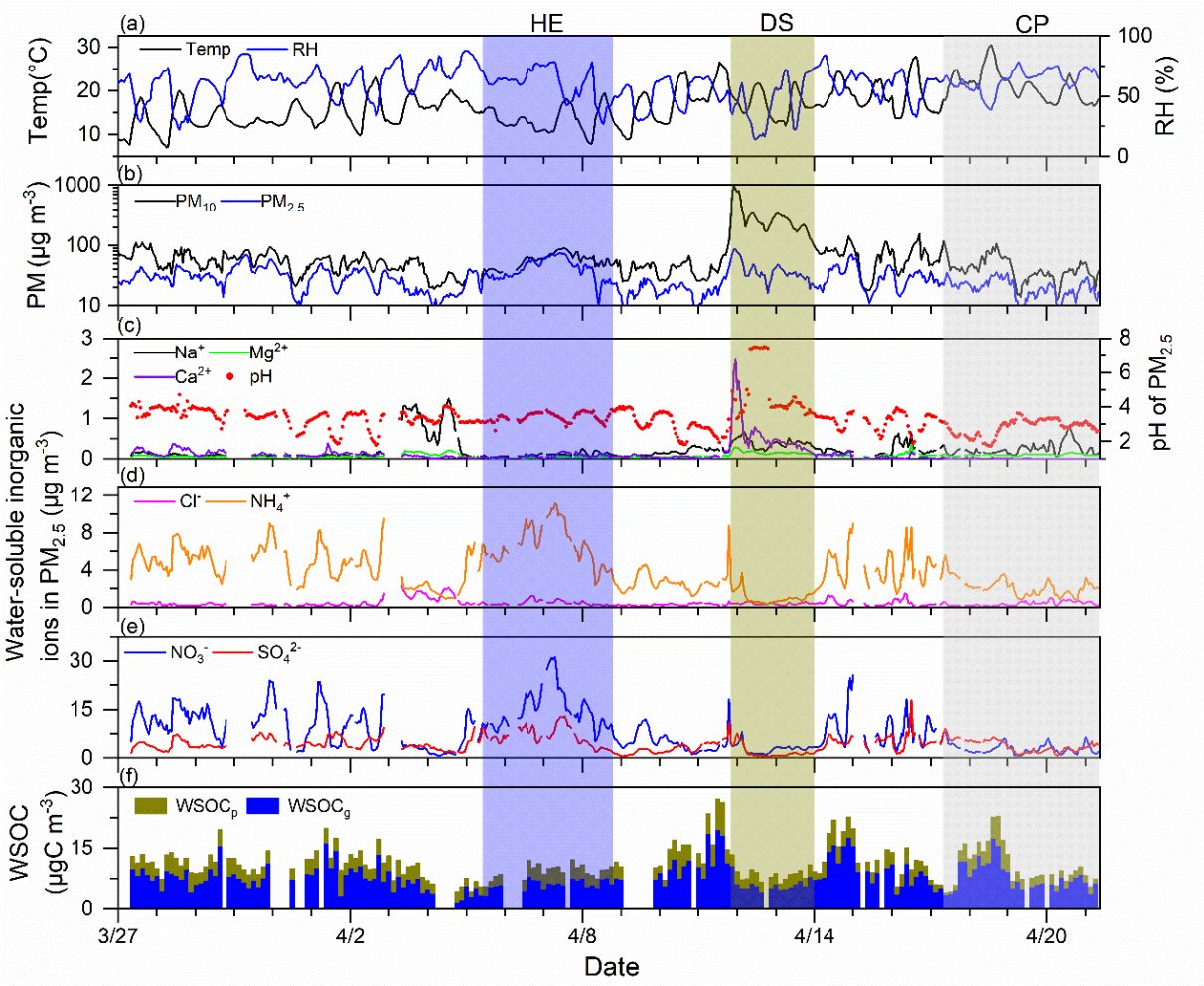

Figure 1. Temporal variations in meteorological parameters and concentrations of major components in $PM_{2.5}$ during the spring of 2023 in Shanghai, China.

7. Table 1: Provide two additional rows for SNA, one for its mass loading and another for the mass contribution to $PM_{2.5}$.

Reply: Suggestion taken. We have added the relative contribution in the Table in the revised manuscript. Please see Table 1 in revised manuscript.

8. Lines 162-163: Please discuss why there is no correlation between $PM_{2.5}$ and $WSOC_p$ in DS.

Reply: Suggestion taken. As seen in Table 1, SNA accounted for 68 ± 13% $PM_{2.5}$ mass in HE but only 16 ± 12% $PM_{2.5}$ mass in DS, because $PM_{2.5}$ was dominated by secondary species in HE but by mineral components in DS, which are primary species emitted from Asian desert region. As we discussed in this work WSOC of $PM_{2.5}$ in

Shanghai during the campaign was mostly derived from secondary formation even in the dust storm period. Thus, $WSOC_p$ correlated with $PM_{2.5}$ in HE there is no correlation between $PM_{2.5}$ and $WSOC_p$ in DS. We added related discussion into the revised manuscript. Please see page 9 line 203-206.

"As seen in Table 1, SNA accounted for 68±13% $PM_{2.5}$ in HE but only 16±12% $PM_{2.5}$ in DS, because $PM_{2.5}$ was dominated by secondary species in HE but by mineral components in DS. Thus, unlike the case in HE, there was no correlation between $PM_{2.5}$ and $WSOC_p$ in DS."

9. Line 205: I cannot see how the uptake of acidic organics is favored by dust. What is it based on?

Reply: Thanks for the comment. In the revised manuscript, we added a detailed discussion on the partitioning of formic and acetic acids as follows.

"To further analyze the factors controlling the WSOC partitioning, we investigated the partitioning process of formic and acetic acids, which are abundant species of WSOCs in the atmosphere (Table S2). As shown in Figure S4, F_p of formic and acetic acids in HE presented strong correlations with ALWC, pH and T. In DS, however, compared with ALWC and T, F_p of formic and acetic acids exhibited a stronger correlation with pH, further indicating that the partitioning of WSOC was more significantly affected by the particle acidity in the presence of dust storm.

Apart from the above factors that influence the F_p variations, aerosol mass loading may also affect the partitioning of WSOC (Lutz et al., 2019). According to the Raoult's law, the field gas-particle partitioning coefficient can be described as follows (Pankow, 1994):

$$K_i = \frac{i_{particle}}{i_{gas} M_{org}} \qquad (R1)$$

where K_i is the partitioning constant of i compound, $i_{particle/gas}$ are the particle and gas phase concentrations of compound i, and M_{org} is the concentration of aerosol organic mass ($WSOC_p$ are used here). In this section, our main focus is the F_p of

WSOC in $PM_{2.5}$. Thus, i and M_{org} are the concentrations of formic and acetic acids and WSOC in $PM_{2.5}$. By definition, the K_i is governed by the organic aerosol mass loading. However, as shown in Figure S5, the partitioning constants (K_i) of formic and acetic acids did not correlate with $WSOC_p$ in both HE and DS periods, indicating that the effect of aerosol organic mass loading on the uptake of WSOC during the campaign was not significant. As shown in Table 1, the $PM_{2.5}$ and WSOC loadings in DS were comparable to those in HE, although PM_{10} loading was much higher in the dust storm period. Moreover, the pH (4.8 ± 1.5, Table 1) of $PM_{2.5}$ in DS was 1.4 units higher than that (3.4 ± 0.3) in HE, which means that H^+ concentration in DS was one order of magnitude lower than that in HE. According to the equations R2 and R3, the low H^+ concentration in DS was favorable for the organic acid equilibria shifting toward the aerosol aqueous phase. Thus, the impact of pH variation on F_p was much more significant than organic aerosol mass loading in DS, taking a key role in WSOC partitioning process during the dust storm event. Previous studies found that diffusivity of organics in particles strongly depends on the phase state of particles (Gkatzelis et al., 2021; Ma et al., 2022). Here we further investigated the impact of particle state on WSOC partitioning during the campaign by calculating the phase state of fine particles (see the detailed method in SI). As shown in Figure S6, fine aerosols were mainly in liquid state during the HE period and mainly in solid or semi-solid phase during the DS period. Thus, F_p linearly and negatively correlated with Tg/T along with an increasing ALWC in HE. However, such a trend was not observed in DS, again suggesting that the key factors controlling the gas-to-particle phase partitioning process of WSOC were different during the two particle pollution events, which was ALWC in HE and pH in DS, respectively.

$$RCOOH(g) \longleftrightarrow RCOOH(aq) \quad (R2)$$

$$RCOOH(aq) \longleftrightarrow RCOO^-(aq) + H^+(aq) \quad (R3)$$

We added the above explanation into the section 3.2 in our revised manuscript. Please see pages 11-12, lines 244-281.

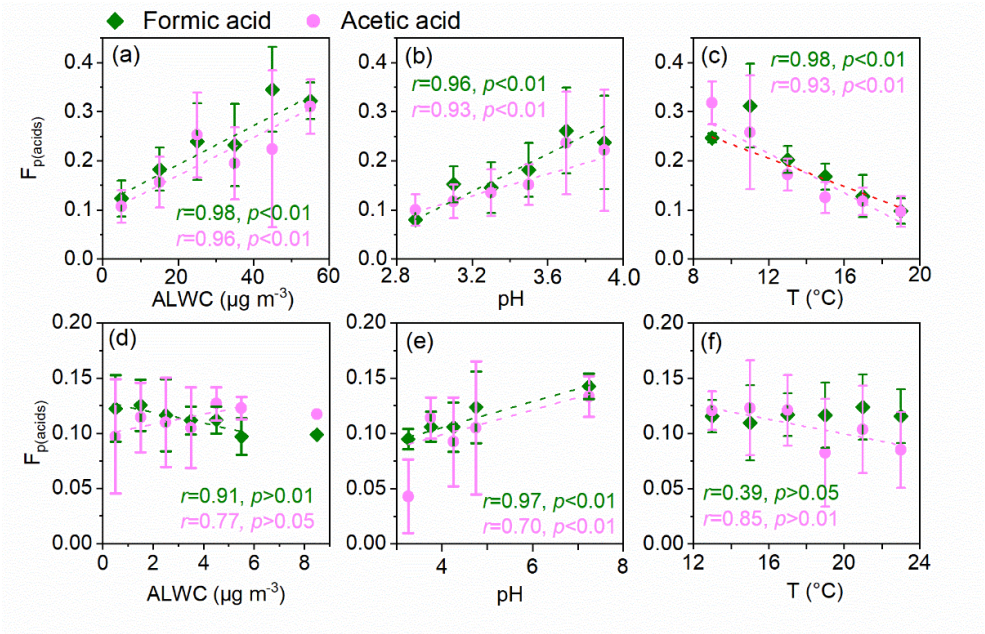

Figure S4. Factors affecting the gas-to-particle partitioning coefficients of formic and acetic acids in (a-c) HE and (d-f) DS periods in spring 2023 in Shanghai.

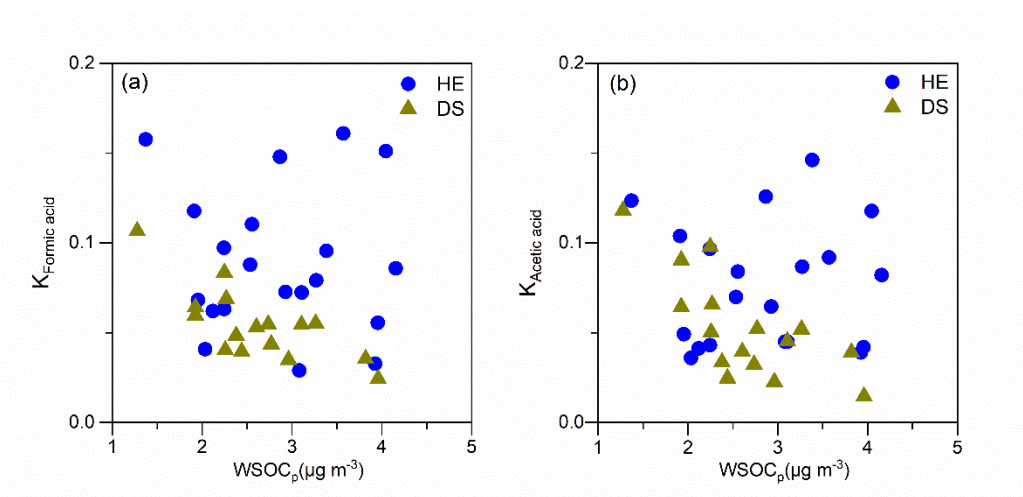

Figure S5. Partitioning coefficients of low molecular organic acids (formic and acetic acids) versus particle phase WSOC during HE and DS periods.

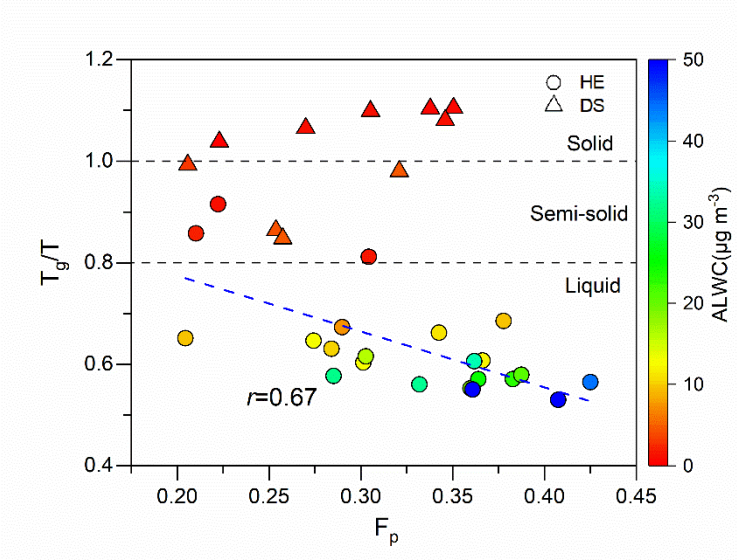

Figure S6. Particle-phase-state transition from solid to liquid particles compared to the partitioning coefficient of WSOC during HE and DS periods.

10. Lines 246 and 249, Figure S5: The discussion is based on the statics of five points for NDS and DS periods. I am concerned with the robustness of the statistics. When examining other figures (e.g., Figures S1 and S2) for statistical analysis, I noticed that more data points are included.

Reply: We fully understand the reviewer's concern about the robustness of the statistics. We made several measures to ensure the robustness of the statistics. For example, for the size-segregate samples, we measured the samples three times to reduce the uncertainty. What's more, the sampling airflow rate for the size-segregated collector was only 28.3 L/min. Each set of samples was continuously for more than 3 days to meet the demand the experimental analysis. To decline the sampling error, we simultaneously collected two sets of samples in each period. In this study four sets of the size-resolved aerosol samples were collected with two sets collected in 11-13 April, 2023, which is a dust storm period (DS), while other two sets of the size-resolved aerosol samples were collected in other non-dust storm periods (NDS). The data exhibited in Figure S5 were the average concentration of pollutants in each size at the coarse mode. In Figures S1 and S2, the analysis was based on the online $PM_{2.5}$ analysis, which has 3-h time resolution. Thus, more data points are included in Figures S1-2, and S5. In the revised version, we added standard variations into the

data points in Figure S9, which is corresponding to Figure S5 in the original version. Please see Figure S9 in the page S10.

11. Line 252: How can you determine that the coarse particles are enriched with $CaCO_3$?

Reply: Thanks for the comment. Coarse particles are derived from soil or dust. These dust-related particles contain carbonates, which are mostly $CaCO_3$ (Laskin et al., 2005; Tobo et al., 2010; Li et al., 2014). In such alkaline conditions, $Ca(NO_3)_2$ are formed from the reactions of $CaCO_3$-containing particles with HNO_3. In fact, Ca-rich particles contain rich nitrate have been detected previously, such as Beijing and Shanghai (Li and Shao, 2009; Wu et al., 2020).

We added the references into our revised manuscript. Please see page 14, line 329.

12. Lines 259 – 263: I do not see the connection between the two sentences. The former is on the discussion about the NDS periods, while the latter is on the discussion on dust surfaces.

Reply: We are sorry for unclear statements. Here we re-wrote the related discussion in the revised manuscript. See page 15, lines 336-339.

"As we discussed in section 3.2, NH_4NO_3 was the biggest contributor to ALWC. Therefore, the formation of hygroscopic species such as NH_4NO_3 can enhance gas-to-particle phase partitioning of $WSOC_g$ and the heterogeneous reactions on the dust surface."

13. Figure 4: Similar to the MAC profile of Tengger Desert PM_{10}, could you please include shaded areas for the MAC profiles of other samples?

Reply: Suggestion taken. Please see Figure 4 in the revised manuscript.

Technical Comment

1. Line 69: Could you use an alternative word instead of "wetted dust"?

Reply: Suggestion taken. We rewrote the sentences. Please see page 3-4, lines 66-69.

"Such reductions in SO_2 and BC in the East Asia atmospheric environment could also significantly alter the physicochemical evolution process of dust particles during long range transport by adsorption and reactions such as gas-to-particles partitioning of WSOCs and SOA formation on the dust particle surface."

2. Lines 110 – 116: The sentence is too long to read. Please split it into two or more.

Reply: Suggestion taken. Please see page 6, lines 125-140 in the revised manuscript.

3. Line 117: Give details about the liquid waveguide capillary UV–Vis spectrometer.

Reply: Done. We gave more detailed description about LWCC spectrometer. Please see page 6, lines 132-137.

"Meanwhile, optical absorptions of WSOC, i.e., water-soluble brown carbon (BrC), in the size-resolved samples were measured using a liquid waveguide capillary cell UV–Vis spectrometer (LWCC, World Precision Instrument, Inc., USA) coupled with a long effective path length. In our application, light from a light source (DH-Mini, Ocean Optics, Inc., USA) is introduced into the LWCC through a fiber optic cable and, after passing through the LWCC, is collected to the detector (Ocean Optics, Inc., USA)."

4. Section 2.3: You should provide the citation from the original papers about ISORROPIA-II, but not three papers in a row from the same group.

Reply: Suggestion taken, we updated the relevant references. See page 7, lines 160-161.

5. Figure 2: Provide details about organic acids in the caption.

Reply: Done. The organic acids were formic, acetic and oxalic acids in Figure 2f. We added the details in the figure caption. Please see Figure 2 in the revised manuscript.

6. Figure 3: What are the dashed lines and filled areas for? Which one is the measured size distribution?

Reply: Thanks for the comment. In Figure 3 and S3, the dashed lines are the measured size distribution and filled areas are the fitting results. We add this description in the caption. Please see Figures 3 and S7 in the revised manuscript.

7. Figure S3: Are these the normalized size distributions?

Reply: Thanks for the comment. They are size distributions normalized by the total concentration in all size stages. We added a note in the figure captions as "C_{total} is the sum of concentration on all the 9-stages".

8. Figure S4: Could you please include the error bars?

Reply: Thank for the comment. Suggestion taken. Please see Figure S8.

9. Line 315: Is it supposed to be NH_4^+?

Reply: We thank for the comment. In the aerosol aqueous phase, carbonyls react with free ammonia (NH_3) rather than ammonium ions to form IMs. Our previous study demonstrates that IMs showed exponential increase with free NH_3 in the humid haze (Liu et al., 2023). Thus, free NH_3 was more suitable than NH_4^+ in herein.

10. Lines 317 – 319: What are the studies from the USA and other developed countries for comparison?

Reply: Thanks for the comment. We add the references in the revised manuscript. Please see page 18, lines 411-412.

11.Table 2: Include the standard deviation for Table 2.

Reply: We thank the reviewer very much for the comment. Suggestion taken.

References:

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