

Dear ACP editor,

After reading the comments from the reviewers, 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.

Anything for our paper, please feel free to contact me via [ghwang@geo.ecnu.edu.cn](mailto:ghwang@geo.ecnu.edu.cn).

All the best,

Gehui Wang

27 May, 2025

**Reviewer #1:**

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-soluble 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's 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  
35 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  
40 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  
45 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  
50 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  
55 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  
60 paper quality.

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

**Major Comments:**

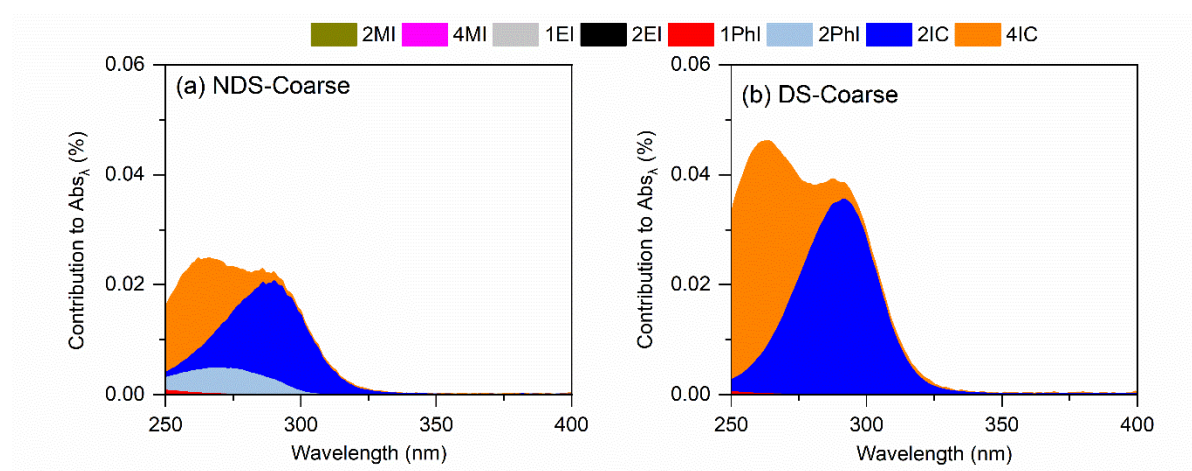
65 1. Length of the dataset: The study focuses on a dataset with nearly 1 month and discusses three measurement periods of interest (i.e., dust storm, haze event, and clean period). Each measurement period of interest spans 3 to 5 days. Although some of the analyses appear to be technically sound, I 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  
70 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  
75 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  
80 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  
85 partitioning and heterogeneous reactions in dust storm period. Similarly, we also found abundant WSON<sub>p</sub> and high MAC values at the coarse mode in dust storm period, confirming that BrC could be formed through gas-particle partitioning process and heterogeneous 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)). As 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  $MAC_{250-600}$  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 as 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:**

115 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.

120 **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:**

125 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.

130 **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 ( $< 2.5 \mu\text{m}$ ) perspective, based on the IGAC-TOC online sampling system ( $2.5 \mu\text{m}$  cutoff inlet). We found that the  $F_p$  of  
135 WSOC was enhanced during DS due to the high pH. In DS, however, the particle is largely dominated by coarse-mode ( $> 2.1 \mu\text{m}$ ). 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 ( $0.4\text{-}9.0 \mu\text{m}$  size range). For comparison, two sets samples were collected during the non-dust storm period (April 7-10).  
140 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<sub>2.5</sub> filter samples were collected on a day/night basis by using a high-volume sampler (1.13 m<sup>3</sup> min<sup>-1</sup>, 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<sup>-1</sup> 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:**

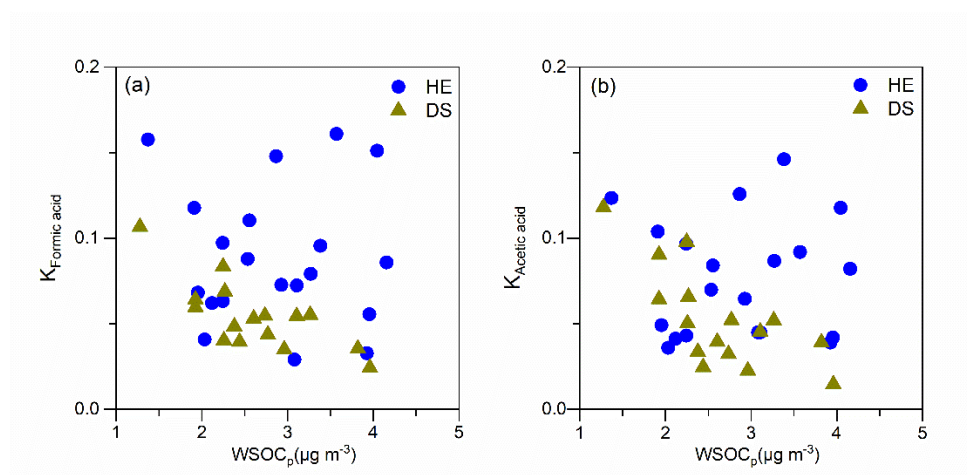
4. F<sub>p</sub>: The F<sub>p</sub> describes a compound's partitioning between the gas and particle phases. During DS periods, the authors claim the high pH is the main driver when ALWC is low. By definition, the F<sub>p</sub> is governed by the effective saturation vapor pressure and aerosol mass loading (Lutz et al., 2019). When the PM<sub>10</sub> 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<sub>p</sub>? Discussions are needed regarding the effect of high aerosol mass loading on F<sub>p</sub> 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<sub>p</sub> 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):

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

where  $K_i$  is the partitioning constant of  $i$  compound,  $i_{\text{particle/gas}}$  are the particle and gas phase concentrations of compound  $i$ , and  $M_{\text{org}}$  is the concentration of aerosol organic mass (WSOC<sub>p</sub> are used here). In this section, our main focus is the  $F_p$  of WSOC in PM<sub>2.5</sub>. Thus,  $i$  and  $M_{\text{org}}$  are the concentrations of formic and acetic acids and WSOC in PM<sub>2.5</sub>. 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<sub>p</sub> 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<sub>2.5</sub> and WSOC loadings in DS were comparable to those in HE, although PM<sub>10</sub> loading was much higher in the dust storm period. Moreover, the pH ( $4.8 \pm 1.5$ , Table 1) of PM<sub>2.5</sub> in DS was 1.4 units higher than that ( $3.4 \pm 0.3$ ) 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.



**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.

195 **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<sub>3</sub>, NO<sub>x</sub>, SO<sub>2</sub> and NH<sub>3</sub>, are abundant.”

200 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<sub>2</sub> 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.”

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3. Lines 99 and 103: How many PM<sub>2.5</sub> and PM<sub>10</sub> filter samples have been collected?

**Reply:** Thanks for the comment. In this study, we collected 50 PM<sub>2.5</sub> filter samples in Shanghai, and 3 PM<sub>10</sub> 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.

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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-nitrosalicylic acid (5NSA). For imidazoles, a total of eight IMs were determined, which are 2-methylimidazole (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.

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



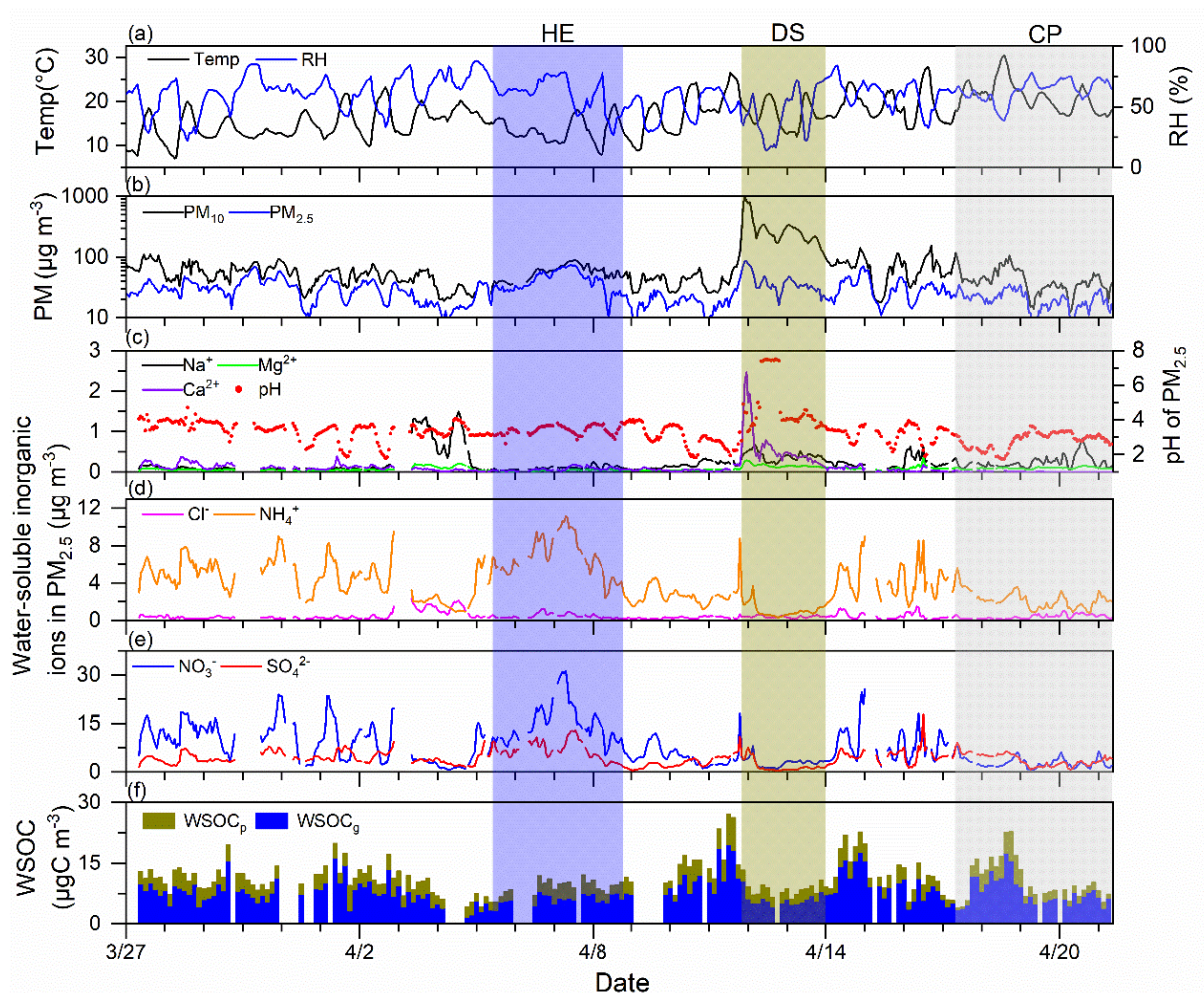
5. Lines 139–141: Is it your own way of defining a haze episode? I found there is a different definition of haze episodes in Dai et al. (2021). If so, please provide a brief description of how it differs from previous studies.

225 **Reply:** Tanks for the comment.

In this study, we defined a haze episode as occurring when the daily  $\text{PM}_{2.5}$  concentration exceeded  $35 \mu\text{g m}^{-3}$ , 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 ( $75 \mu\text{g m}^{-3}$ ). 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),  $\text{PM}_{2.5}$  concentrations frequently exceeded  $75 \mu\text{g m}^{-3}$ , with peak hourly levels during haze episodes ranging from 198 to  $571 \mu\text{g m}^{-3}$  across their research areas. In contrast, Shanghai exhibits better air quality compared to NCP cities. Our study recorded an average  $\text{PM}_{2.5}$  concentration of  $30 \mu\text{g m}^{-3}$ , slightly below the Air Quality Grade I Standard of China. Given these lower baseline levels, we maintained the  $35 \mu\text{g m}^{-3}$  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  $\text{Na}^+$ ,  $\text{Mg}^{2+}$ , and  $\text{Ca}^{2+}$ , I would create a subplot for them. The same applies to  $\text{NH}_4^+$  and  $\text{Cl}^-$ . 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.

250 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.

255 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  $68 \pm 13\%$   $PM_{2.5}$  mass in HE but only  $16 \pm 12\%$   $PM_{2.5}$  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

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campaign was mostly derived from secondary formation even in the dust storm period. Thus, WSOC<sub>p</sub> correlated with PM<sub>2.5</sub> in HE there is no correlation between PM<sub>2.5</sub> and WSOC<sub>p</sub> in DS. We added related discussion into the revised manuscript. Please see page 9 line 203-206.

265 “As seen in Table 1, SNA accounted for 68±13% PM<sub>2.5</sub> in HE but only 16±12% PM<sub>2.5</sub> in DS, because PM<sub>2.5</sub> 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<sub>2.5</sub> and WSOC<sub>p</sub> in DS.”

270 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.

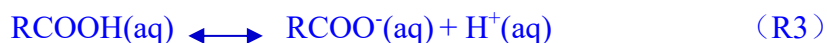
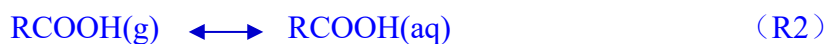
275 “To further analyze the factors controlling the WSOC partitioning, we investigated the partitioning process of formic and acetic acids, which are abundant species of WSOCs in the atmosphere (Table S2). As shown in Figure S4, F<sub>p</sub> of formic and acetic acids in HE presented strong correlations with ALWC, pH and T. In DS, however, compared with ALWC and T, F<sub>p</sub> 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  
280 acidity in the presence of dust storm.

Apart from the above factors that influence the F<sub>p</sub> 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):

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

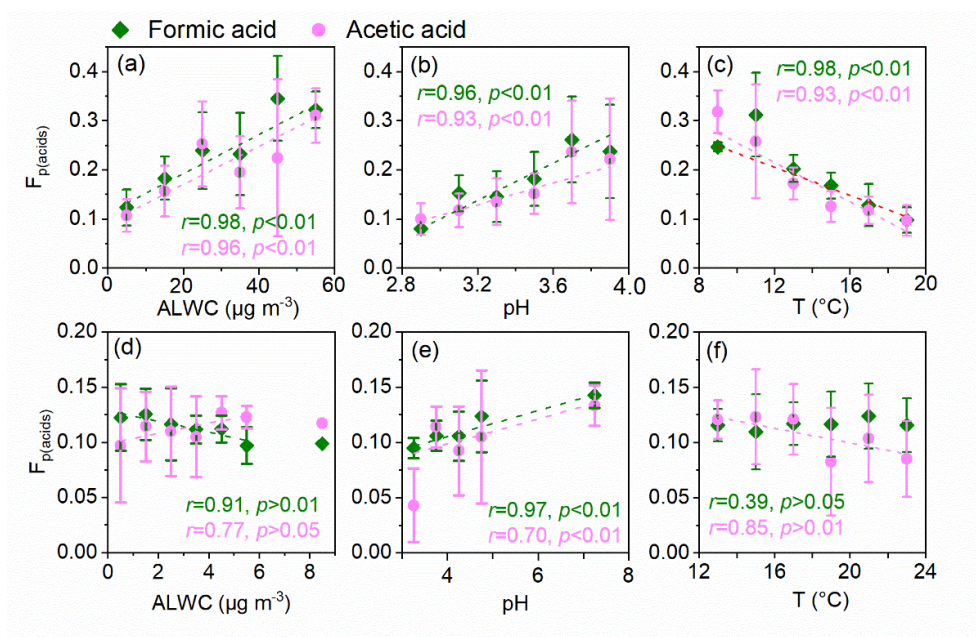
285 where  $K_i$  is the partitioning constant of  $i$  compound,  $i_{\text{particle/gas}}$  are the particle and gas phase concentrations of compound  $i$ , and  $M_{\text{org}}$  is the concentration of aerosol organic mass (WSOC<sub>p</sub> are used here). In this section, our main focus is the F<sub>p</sub> of WSOC in PM<sub>2.5</sub>. Thus,  $i$  and  $M_{\text{org}}$  are the concentrations of formic and acetic acids and WSOC in PM<sub>2.5</sub>. 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<sub>p</sub> 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<sub>2.5</sub> and WSOC loadings in DS were comparable to those in HE, although PM<sub>10</sub> loading was much higher in the dust storm period. Moreover, the pH ( $4.8 \pm 1.5$ , Table 1) of PM<sub>2.5</sub> in DS was 1.4 units higher than that ( $3.4 \pm 0.3$ ) in HE, which means that H<sup>+</sup> concentration in DS was one order of magnitude lower than that in HE. According to the equations R2 and R3, the low H<sup>+</sup> 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.

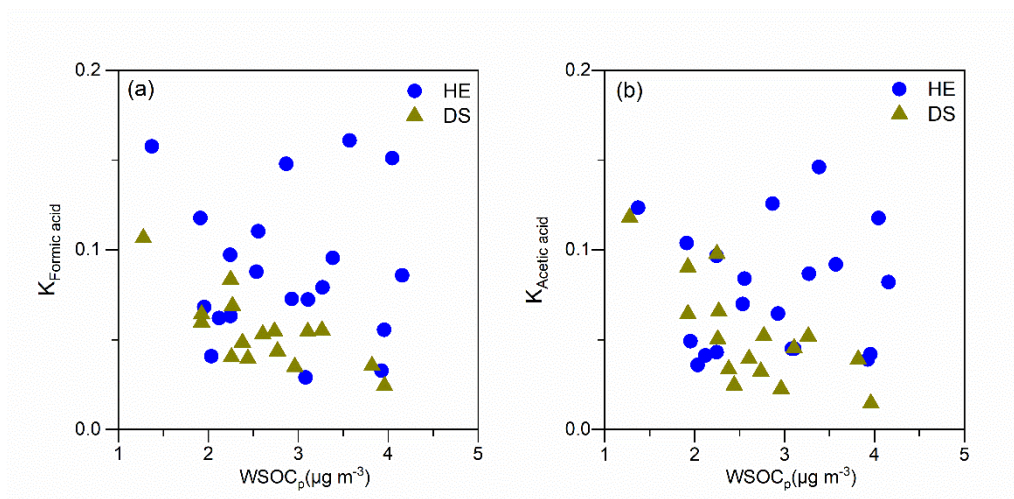


We added the above explanation into the section 3.2 in our revised manuscript. Please see pages 11-12, lines 244-281.





315 **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.

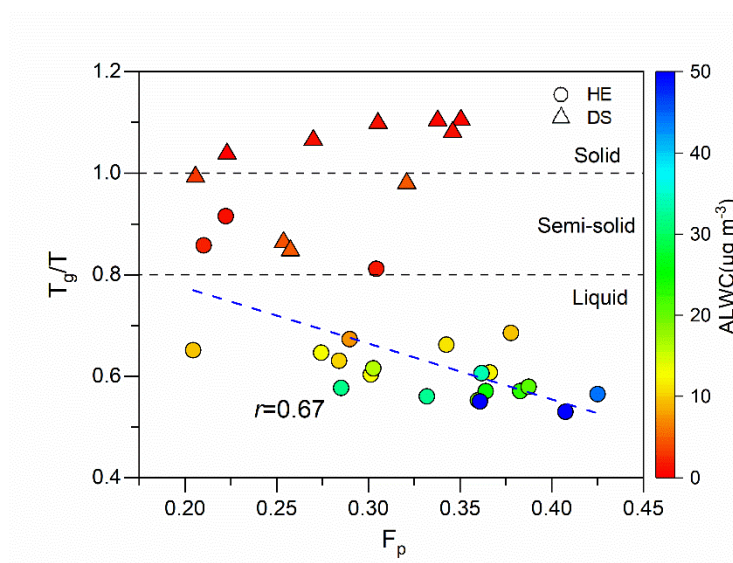


Figure S6. Particle-phase-state transition from solid to liquid particles compared to the partitioning coefficient of WSOC during HE and DS periods.

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<sub>2.5</sub> 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  $\text{CaCO}_3$ ?

345 **Reply:** Thanks for the comment. Coarse particles are derived from soil or dust. These dust-related particles contain carbonates, which are mostly  $\text{CaCO}_3$  (Laskin et al., 2005; Tobo et al., 2010; Li et al., 2014). In such alkaline conditions,  $\text{Ca}(\text{NO}_3)_2$  are formed from the reactions of  $\text{CaCO}_3$ -containing particles with  $\text{HNO}_3$ . In fact, Ca-rich particles contain rich nitrate have been detected previously, such as Beijing and Shanghai (Li and Shao, 2009; Wu  
350 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  
355 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,  $\text{NH}_4\text{NO}_3$  was the biggest contributor to ALWC. Therefore, the formation of hygroscopic species such as  $\text{NH}_4\text{NO}_3$  can enhance gas-to-  
360 particle phase partitioning of  $\text{WSOC}_g$  and the heterogeneous reactions on the dust surface.”

13. Figure 4: Similar to the MAC profile of Tengger Desert  $\text{PM}_{10}$ , 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  $\text{SO}_2$  and BC in the East Asia atmospheric environment could also  
370 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.

375 **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.

380 “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  
385 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.

390

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.

395 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.

400



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 “ $C_{\text{total}}$  is the sum of concentration on all the 9-stages”.

405

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  $\text{NH}_4^+$ ?

410 **Reply:** We thank for the comment. In the aerosol aqueous phase, carbonyls react with free ammonia ( $\text{NH}_3$ ) rather than ammonium ions to form IMs. Our previous study demonstrates that IMs showed exponential increase with free  $\text{NH}_3$  in the humid haze (Liu et al., 2023). Thus, free  $\text{NH}_3$  was more suitable than  $\text{NH}_4^+$  in herein.

415 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.

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

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

425

## **Reviewer #2:**

### **Comment:**

Gas-to-particle partitioning of organics is a key process in secondary organic aerosol (SOA) formation. This manuscript presents field measurements of water-soluble organic carbon (WSOC) in both gas and particle phases, and investigates the characteristics of the particle-phase fraction (Fp), its potential formation processes, optical properties, and size distributions through case analyses. The authors suggest that physicochemical properties such as aerosol pH, aerosol liquid water content (ALWC), and particle composition play dominant roles in governing WSOC formation and Fp dynamics. The manuscript fits well within the scope of Atmospheric Chemistry and Physics (ACP), and the writing is generally clear. However, to support the conclusions more convincingly, the manuscript would benefit from a more thorough and quantitative discussion of the Fp dynamics, detailed analysis of WSOC measurements and chemical composition, more comprehensive characterization of particle size distributions, and improved evaluation of aerosol pH. Therefore, I recommend that the manuscript be reconsidered for publication after substantial revision and inclusion of the necessary details and discussions.

**Reply:** We sincerely appreciate the reviewer's valuable comments. We have carefully addressed all the concerns and thoroughly revised the manuscript accordingly. The key modifications include:

- (1) We have added an in-depth discussion on how organic mass loading and particle phase state influence WSOC partitioning.
- (2) We further analyzed the partitioning behavior of organic acids on a molecular level in both HE and DS to provide a more comprehensive understanding on the key factors controlling WSOC partitioning on dust surface during a long-range transport and its impact on dust particle light absorption.

Detailed responses to each comment are provided below. All the revisions and changes in the manuscript and the supporting information are highlighted in blue color.

455 **Major comments:**

**Comment:**

1. The WSOC data are central to the conclusions of this manuscript. However, the authors only briefly cited previous literature to describe the measurement methods. Detailed descriptions of both the offline and online WSOC measurement techniques, including  
460 sampling protocols, instrument calibration, detection limits, and potential uncertainties, should be clearly provided. In addition, during dust events, the particle mass is primarily dominated by coarse-mode particles. It is unclear how the use of a 2.5  $\mu\text{m}$  cutoff inlet ( $\text{PM}_{2.5}$ ) can adequately represent the WSOC associated with dust particles. The authors should justify the representativeness of the  $\text{PM}_{2.5}$  sampling approach for characterizing  
465 WSOC during dust episodes or provide supporting data to address this limitation.

**Reply:** We appreciate the reviewer's constructive feedback. In response, we have made the following revisions to address the comments:

(1) Methodological clarifications:

As suggested, we have expanded the description in Sections 2.1 and 2.2 to provide further  
470 details on the sampling methods, instrument calibration, detection limits, and potential uncertainties. These additions ensure greater transparency and reproducibility of our methodology. Please see pages 5-7, lines 98-107, 125-140 and 144-154.

(2) Analysis of WSOC partitioning during DS periods:

In Section 3.2, we observed enhanced WSOC partitioning in  $\text{PM}_{2.5}$  during DS episodes.  
475 However, recognizing that atmospheric particles during DS events are predominantly coarse-mode, we acknowledged the limitation of focusing solely on fine particles. To comprehensively evaluate partitioning behavior and validate the Section 3.2 findings, we conducted additional size-segregated sampling and analysis (Section 3.3). The results confirm that WSOC partitioning is indeed amplified during DS periods, particularly in the  
480 coarse mode (see Figure S8).

**Comment:**

2.(1) The pH calculation is based on the ISORROPIA-II, but the basic evaluation of the predicted and observed parameters (such as  $\text{NO}_3^-$ ,  $\text{NH}_3$ , etc) is not given, which is important

for the model performance. (2) In addition, RH is less than 30% in dust haze, which would introduce much uncertainty for pH calculation. It caused the high pH, up to 7.5, and this value is stable. However, during high  $\text{Ca}^{2+}$ , indicator of dust episode, pH is comparable to haze and clean days. The more detailed discussion should be added to clarify this.

**Reply:** (1) We appreciate the reviewer's suggestion and have added the related points to the revised manuscript. Please see the page 7 lines 161-167.

“Figure S1 compares the concentrations of  $\text{NH}_3$  and  $\text{NO}_3^-$  measured by the IGAC instrument with those predicted by ISORROPIA-II model. As shown in Figure S1, the predicted are very close to the measured, suggesting that the metastable mode we used can remarkably reproduce the measured and the model results including ALWC and acidity (pH) are reliable. It should be noted that the samples collected under  $\text{RH} > 95\%$  conditions were excluded in this study, because  $\text{RH} > 95\%$  conditions could increase ALWC and pH estimation uncertainties.”

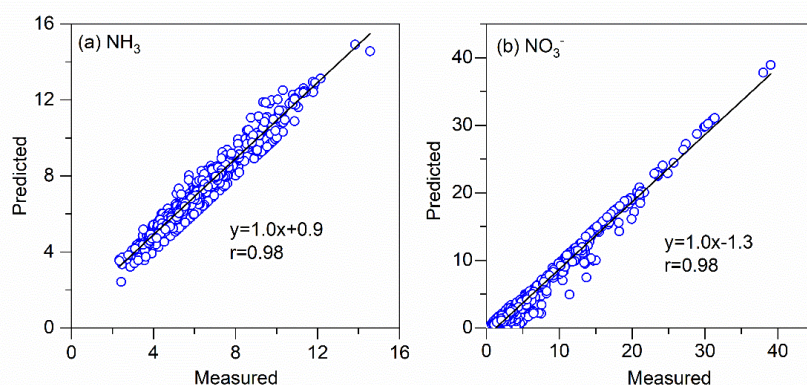
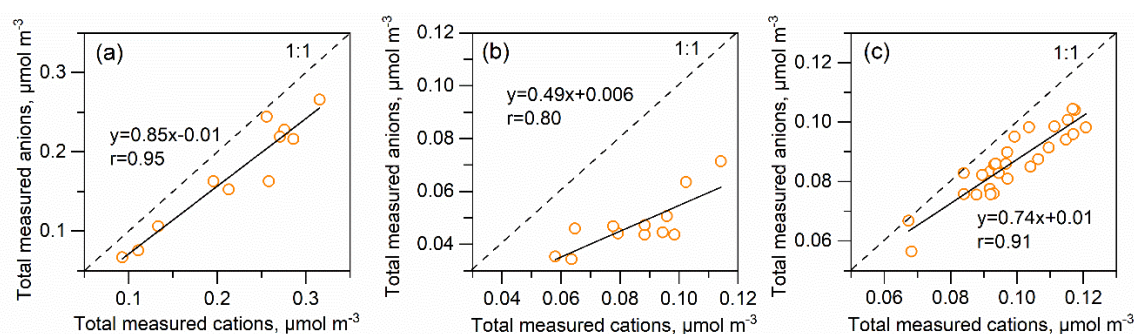


Figure S1. Comparison of measured ammonia and nitrate and predicted by ISORROPIA-II model.

(2) According Fountoukis and Nenes (2007), when the RH ranged between 0.2 and 0.8 at room temperature, the mass fraction of solute as predicted by ISORROPIA-II model agree well with the experimental measurements for a variety of equimolar mixtures of salts. (see section 4 and Fig.3 in Fountoukis and Nenes, 2007). In addition, the high pH value in DS is mainly related to the composition of the particles. As shown in Figure 1c, pH was gradually increased in the begin of DS and up to 7.5 during the stagnant period, then the value of pH at the end of DS gradually decreased and was comparable to those in HE and CP periods. This

phenomenon indicates that the chemical species may be changed during dust particle transport. Figure R1 plots the equivalent ratios of total measured ions in PM<sub>2.5</sub> in DS period.

510 The ratio of total measured anions to cations was close to unity in the dust storm begin period (the first day in DS) (Figure R1a). But the slope of regression line was only 0.49 (Figure R1b) in the dust stagnant period (the second day in DS), then the slope increased to 0.74 (Figure R1c) during the end of dust storm period (the third day in DS), indicating that particles in stagnant period were more basic. When crustal species are in excess compared to  
515 all the anions (such as dust storm samples in this study), ISORROPIA-II model assumes that a presence of excess carbonate in the aerosol phase and the solution is close to neutral (Fountoukis and Nenes, 2007). Thus, it caused the pH of particles was high and stable. The ratio of anions to cations was higher in the end period than in the stagnant period, indicating that more acids replaced CO<sub>3</sub><sup>2-</sup> at the dust particle surface during the dust storm long-range  
520 transport, which could reduce the basicity of dust particles.



**Figure R1.** Correlations for cations and anions in PM<sub>2.5</sub> samples during (a) the begin, (b) the stagnant, and (c) the end period of dust storm. X axis represents the sum of measured Na<sup>+</sup>, NH<sub>4</sub><sup>+</sup>, K<sup>+</sup>, Mg<sup>2+</sup>, Ca<sup>2+</sup>. Y axis represents the sum of total measured Cl<sup>-</sup>, NO<sub>3</sub><sup>-</sup> and SO<sub>4</sub><sup>2-</sup>.

### **Comment:**

3. During the chemical analysis, the organic acids, NACs and IMs are detected. However, the detailed discussion is not given, which is important for WSOC Fp and dynamic characteristics. The Fp and gas-to-particle partitioning (kp) depend on the chemical  
530 properties of organics, aerosol components, particle phase, etc. In this manuscript, the authors just discussed the influences of inorganic components. The driving impacts of organics and particle phase was not considered. It should be added.

**Reply:** Thanks for the comment. We added in-deep discussion on the formic and acetic acids

partitioning in our revised manuscript, which are the most abundant organic acids in  
 535 atmosphere. The  $F_p$  of IMs and NACs calculation requires its gaseous concentration.  
 However, we cannot obtain the gas phase IMs and NACs concentrations due to the  
 instrument limits. In section 3.3, we found that NACs can be formed through gas-to-particle  
 partitioning at the coarse mode in DS and IMs can formed through heterogenous reactions at  
 the coarse mode in DS. More discussion about the driving factors of WSOC  $F_p$  (such as  
 540 organic mass loading and particle phase state) as shown in the latter and revised manuscript.  
 We also analyzed the factors controlling the organic acids partitioning in DS. Please see  
 pages 10-12, lines 244-281:

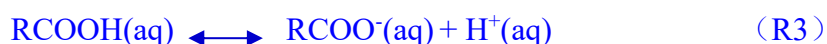
“To further analyze the factors controlling the WSOC partitioning, we investigated the  
 partitioning process of formic and acetic acids, which are abundant species of WSOCs in the  
 545 atmosphere (Table S2). As shown in Figure S4,  $F_p$  of formic and acetic acids in HE  
 presented strong correlations 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.

550 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):

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

where  $K_i$  is the partitioning constant of  $i$  compound,  $i_{\text{particle/gas}}$  are the particle and gas  
 555 phase concentrations of compound  $i$ , and  $M_{\text{org}}$  is the concentration of aerosol organic mass  
 (WSOC<sub>p</sub> are used here). In this section, our main focus is the  $F_p$  of WSOC in PM<sub>2.5</sub>. Thus,  $i$   
 and  $M_{\text{org}}$  are the concentrations of formic and acetic acids and WSOC in PM<sub>2.5</sub>. 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  
 560 WSOC<sub>p</sub> 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<sub>2.5</sub> and WSOC loadings in DS were comparable to those in HE, although PM<sub>10</sub> loading was much higher in the dust storm period. Moreover, the pH ( $4.8 \pm 1.5$ , Table 1) of PM<sub>2.5</sub> in DS was 1.4 units higher than that ( $3.4 \pm 0.3$ ) in HE, which means that H<sup>+</sup> concentration in DS was one order of magnitude lower than that in HE. According to the equations R2 and R3, the low H<sup>+</sup> concentration in DS was favorable for the organic acid equilibria shifting toward the aerosol aqueous phase. Thus, the impact of pH variation on F<sub>p</sub> 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<sub>p</sub> linearly and negatively correlated with T<sub>g</sub>/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.





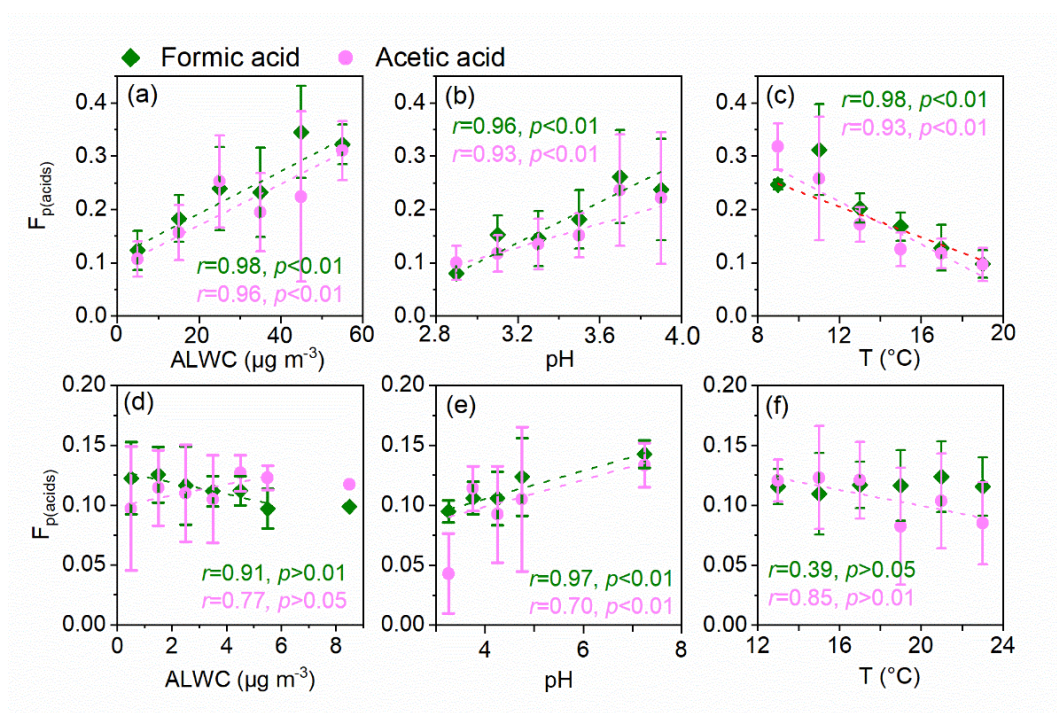


Figure S4. Factors affecting the gas-to-particle partitioning coefficients of formic and acetic acids in (a-c) HE and (d-f) DS periods in spring 2023 in Shanghai.

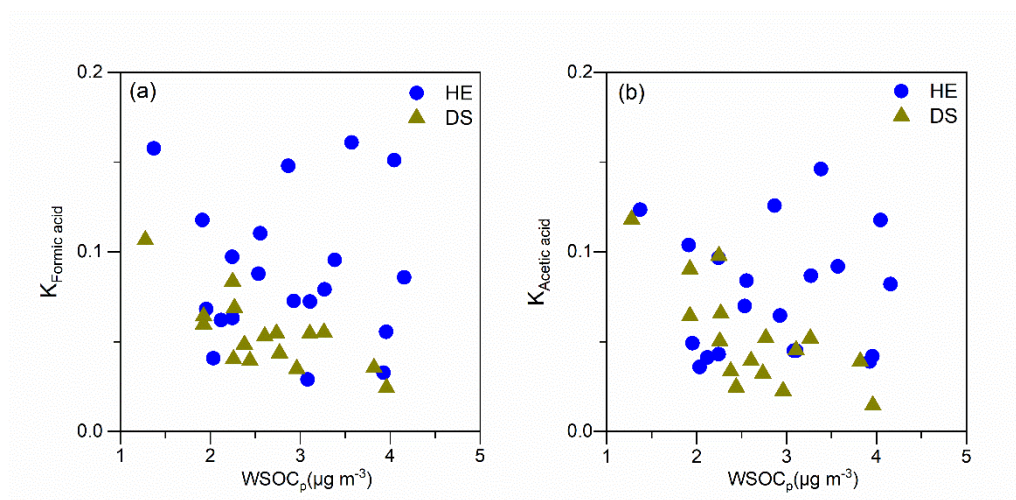


Figure S5. Partitioning coefficients of low molecular organic acids (formic and acetic acids) versus particle phase WSOC during HE and DS periods.



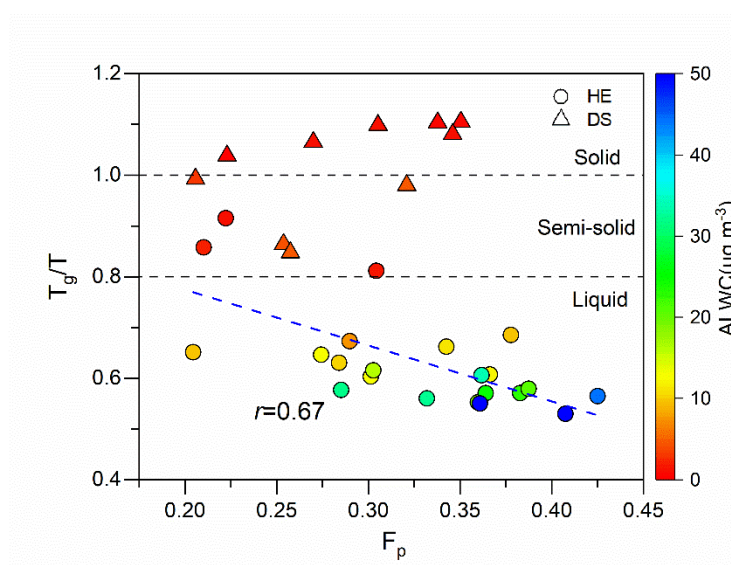


Figure S6. Particle-phase-state transition from solid to liquid particles compared to the partitioning coefficient of WSOC during HE and DS periods.

**Comment:**

4. The size distribution of WSOC, NACs, etc. have been discussed, but the measurement of size distribution was not given. It should be added.

**Reply:** Suggestion taken. In this study, the size-segregated samples were collected using a 9-stage Anderson sampling collector. In section 3.3 and 3.4, the discussion of chemical species was based on the size-segregated samples. We added the sampling methods, sampling instrument and chemical species analysis methods in section 2.1 in the revised manuscript, please see pages 5-7, lines 98-119 and lines 125-154.

**Specific comments:**

1. In the abstract, it mentioned the time resolution of WSOC is 1h, but it is 3h in the method. Please clarify it.

**Reply:** Thanks for the comment. In this study, online  $\text{PM}_{2.5}$  WSOC samples were collected hourly. However, due to the instrument detection limits, three adjacent WSOC samples were merged to provide a large sample volume. Thus, the time resolution of gas and particle phase WSOC measurements were 3h. We revised this sentence in the abstract. Please see lines 25-29.

2. Line 62, the “NO<sub>x</sub>” should be replaced with “NO<sub>x</sub>”. Please checked all the upper and lower superscripts of the article.

**Reply:** Suggestion taken. We have revised the sentences according to the reviewer’s comment and checked all the upper and lower superscripts of the article.

3. Line 153, the author stated a “significant formation of SNA”. Once the term of significance was used, the statistical and significance analyses are necessary. Please add it. In addition, the Ca<sup>2+</sup>, as the indicator of dust, peaked at a short time, indicating minor influence of dust, not lasting three days. Please clarify it.

**Reply:** Thanks for the comment. First, we conducted a significance analysis of the data (t test, p<0.01) and added the result into the revised manuscript. Please see page 8 line 188.

Second, on the third day (13 April) at DS, the ratio of PM<sub>2.5</sub> to PM<sub>10</sub> was  $0.15 \pm 0.03$ , which was 0.18 times that of the HE ( $0.74 \pm 0.17$ ) and 0.29 times that of the CP ( $0.51 \pm 0.14$ ), indicating abundant coarse particles in the Shanghai atmosphere. Moreover, the relative contribution of Ca<sup>2+</sup> to total water-soluble inorganic ions was  $0.09 \pm 0.03$  on the third day, which was slight lower than the whole DS period value ( $0.14 \pm 0.07$ ), but was 18 times of that in HE ( $0.005 \pm 0.003$ ). Thus, based on the above discussion, we think that the impact of the DS on the 13 April in the Shanghai atmosphere still proceeded.

4. In lines157-168, the measured average WSOCp is about  $2 \mu\text{gC m}^{-3}$ , but in Table 1 and 2, the total carbon of oxalic acid, Pyr, NACs and IMs are large than the average WSOCp.

Please checked it.

**Reply:** Thanks for the comment. In the tables, the units of NACs and IMs are  $\text{ng m}^{-3}$ . Other organic acids concentration units are  $\mu\text{g m}^{-3}$ . In this study, the ratio of the sum of organic acids and IMs to WSOC was 6.2%.

5. In section 3.2, the author discussed driving factor of WSOC partitioning. In previous study (Ma et al 2022), it has reported that the particle phase would influence the OVOCs partitioning. For example, when ALWC was over  $15 \mu\text{g m}^{-3}$ , as indicator of liquid phase for particles, it would weaken the gas-to-particle partitioning of OVOCs with  $k_p > 10^{-5}$ . In this

manuscript, the author just discussed the general WSOC partitioning, which was determined  
640 by the chemical components. The more detailed information is needed.

**Reply:** Thank you for the advice. We added the discussion about organic acids partitioning,  
effect of particle phase on the WSOC  $F_p$  in the revised manuscript. Please see pages 11-13,  
lines 244-250, and lines 270-279.

“To further analyze the factors controlling the WSOC partitioning, we investigated the  
645 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  
650 acidity in the presence of dust storm.

Previous study found that the 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  
655 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  
 $T_g/T$  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  
660 in HE and pH in DS, respectively.”

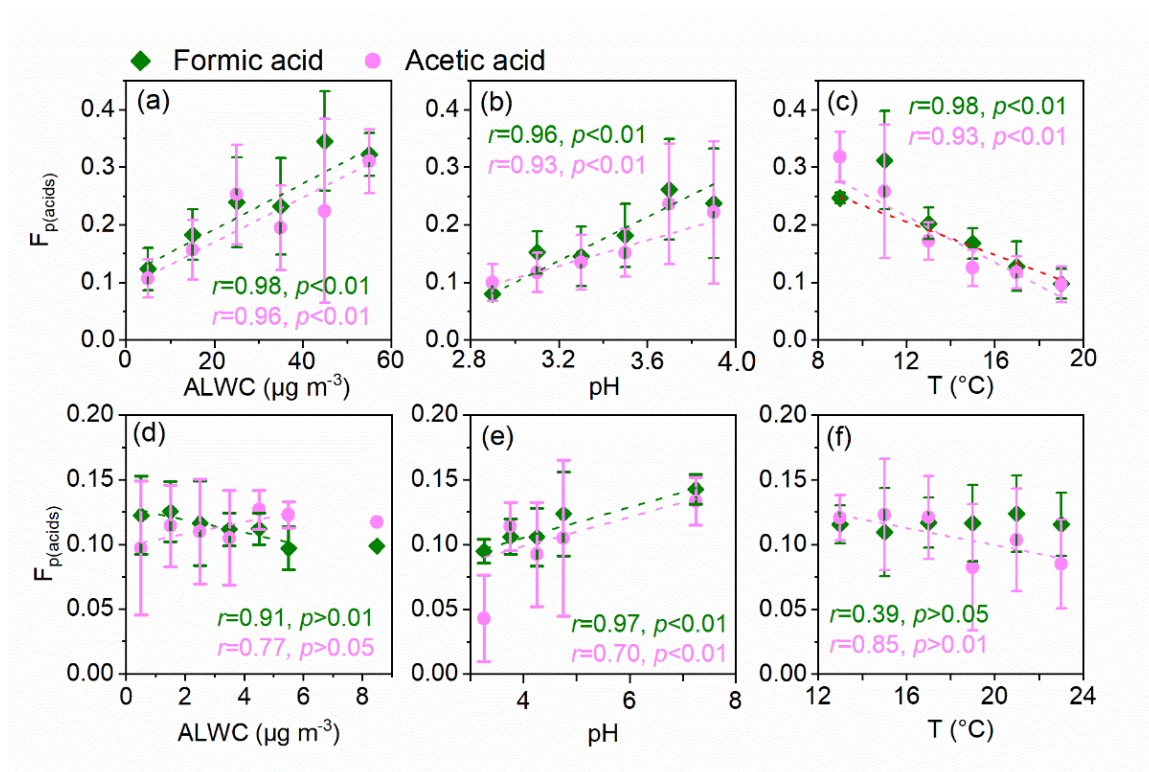


Figure S4. Factors affecting the gas-to-particle partitioning coefficients of formic and acetic acids in (a-c) HE and (d-f) DS periods in spring 2023 in Shanghai.

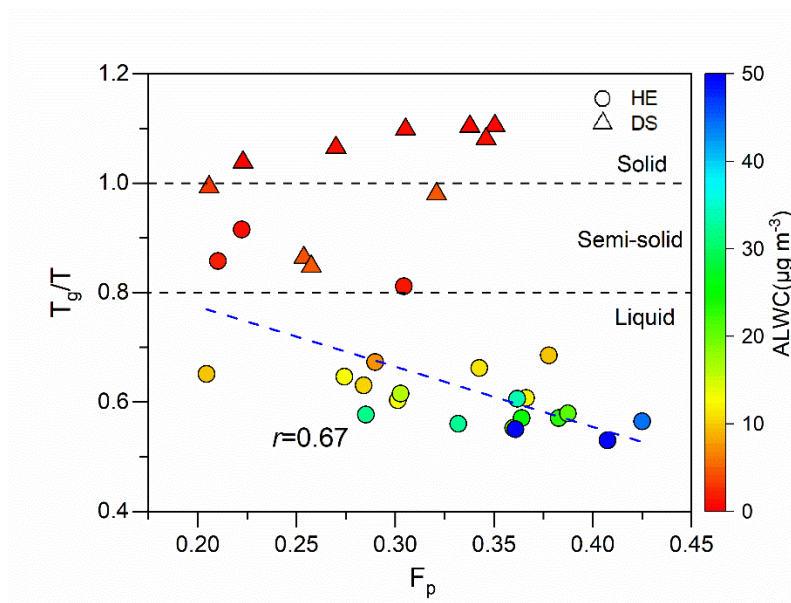


Figure S6. Particle-phase-state transition from solid to liquid particles compared to the partitioning coefficient of WSOC during HE and DS periods.

6. In lines 196–205, the authors suggest that aerosol pH is a key factor influencing WSOC partitioning during the DS period, based on correlation analysis. However, further clarification is needed. During DS,  $\text{Ca}^{2+}$  and  $\text{Mg}^{2+}$  are the dominant cations, whereas during

HE,  $\text{NH}_4^+$  predominates. This shift in ionic composition likely alters aerosol hygroscopicity, thereby affecting aerosol liquid water content (ALWC). The manuscript posited out that in the HE period, higher ALWC dilutes  $\text{H}^+$  concentrations, leading to higher aerosol pH, whereas during DS, lower ALWC results in more concentrated  $\text{H}^+$  and hence lower pH. In fact, the pH in these two cases are the opposite trend. Specifically, the authors should provide detailed data or modeling results to validate the inferred relationship between ALWC, ionic composition, and pH, and to strengthen the mechanistic interpretation of WSOC partitioning behavior across all the measured data.

**Reply:** Thanks for the comment. Indeed, higher ALWC could dilute  $\text{H}^+$  concentrations, leading to higher pH. However, a pH is determined not only by ALWC, but also by the cations and anions. For example, the particles in DS were enriched in  $\text{Ca}^{2+}$ ,  $\text{Na}^+$  and  $\text{Mg}^{2+}$  cations, which significantly reduced the  $\text{H}^+$  concentration and resulted in a higher pH. In fact, in DS the  $\text{H}^+$  concentration was  $9.2 \times 10^{-8} \text{ M}$ , which was one order of magnitude lower than that ( $4.7 \times 10^{-7} \text{ M}$ ) in HE. This is the reason why pH in DS was higher than that in HE, although ALWC was lower during DS. Moreover, for verifying the WSOC partitioning behavior, we also discussed the partitioning process of low molecular organic acids, i.e. formic and acetic acids, in our revised manuscript. We found that those low molecular weight organic acids partitioning behaviors were consistent with that of WSOC. Please see page 11, lines 244-250.

“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.”

7. In Lines 249-253: How can the author determine that the coarse particles are enriched with  $\text{Ca}(\text{NO}_3)_2$  or  $\text{CaCO}_3$ ?

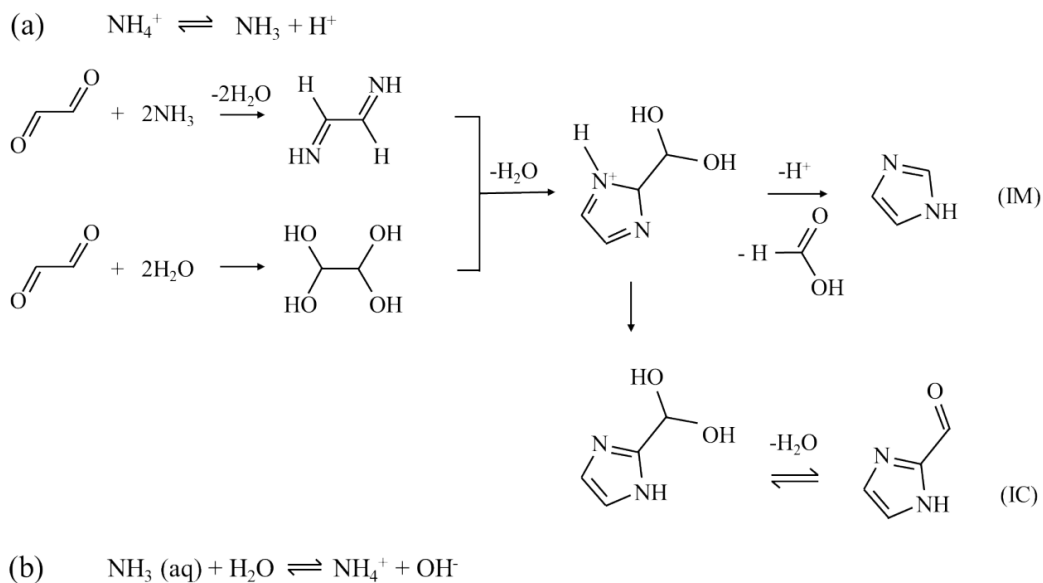
**Reply:** Thanks for the comment. As we replied previously, coarse particles in the planet boundary layer are often derived from soil and dust. Those dust-related particles are enriched in carbonates, which are mostly  $\text{CaCO}_3$  (Laskin et al., 2005; Tobo et al., 2010; Li et al., 2014). In such alkaline conditions,  $\text{Ca}(\text{NO}_3)_2$  are formed from the reactions of  $\text{CaCO}_3$ -containing particles with  $\text{HNO}_3$ . In previous studies, 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 the revised manuscript. Please see page 14, lines 327-330.

8. In 238-246, the role and influence of IMs for  $F_p$  of WSOC was not clear? IMs were mainly formed through the aqueous reaction of  $\text{NH}_3$  and carbonyls. The low ALWC would not favor the formation of IMs. It should give more discussion, such as the phase state.

**Reply:** Thanks for the comment.

Our study confirmed that IMs can be formed through aqueous-phase reactions during DS. Such aqueous-phase reactions can enhance the  $F_p$  of WSOC. Moreover, our previous study confirmed that IMs were mainly formed through the aqueous reactions of free ammonia ( $\text{NH}_3(\text{aq})$ ) with carbonyls (Figure R2a). As shown in Figure R2b, the equilibrium between  $\text{NH}_4^+$  and  $\text{NH}_3(\text{aq})$  depends on the aerosol aqueous phase pH. In DS, the higher pH was favorable for the formation of  $\text{NH}_3(\text{aq})$ , which promoted the reaction of carbonyl with  $\text{NH}_3(\text{aq})$  to produce IMs. In DS, the concentration of  $\text{NH}_3(\text{aq})$  was  $0.76 \pm 0.5$  M, which was more one orders of magnitude higher than that in HE ( $0.017 \pm 0.014$  M). The higher concentration of  $\text{NH}_3(\text{aq})$  in DS was favorable for the formation of IMs. Moreover, we also added in-deep discussion on the influence of the phase state on WSOC partitioning. Please see page 12, lines 270-279.



**Figure R2.** Reactions of free ammonia with carbonyls (cited from Liu et al., (2023)).

725

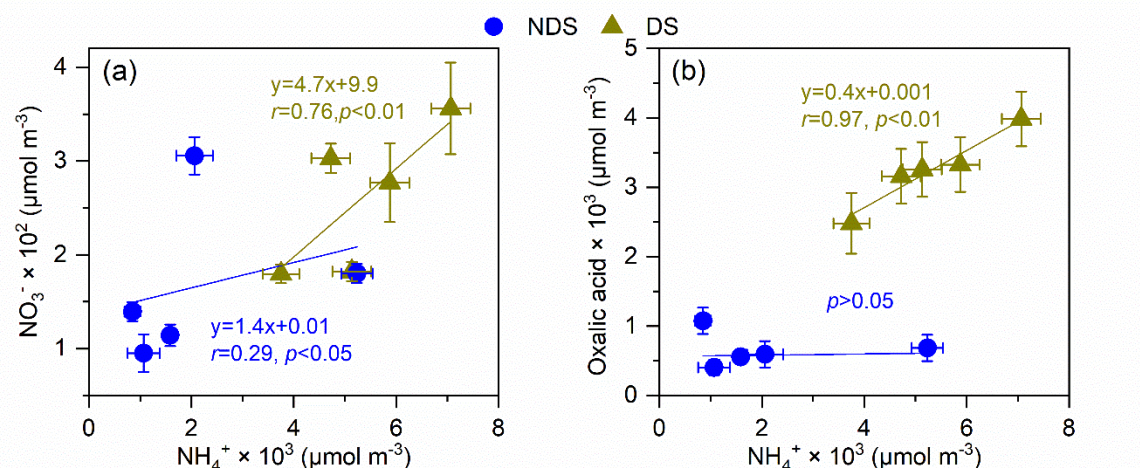
9. In Lines 255-258, the author pointed out that  $\text{N}_2\text{O}_5$  hydrolysis would promote  $\text{NO}_3^-$  formation due to those mineral dust particles and hygroscopic, but in DS, no obvious  $\text{NO}_3^-$  formation occurred compared other observed time. Thus, it did not support a linear correlation between  $\text{NO}_3^-$  and  $\text{NH}_4^+$  in the coarse mode. Please clarify it. In addition, this discussion should apply to the whole campaign, including WSOC,  $F_p$ , pH, size distribution and driving factors.

730

**Reply:** Thanks for the comment. Indeed, there were no obvious  $\text{NO}_3^-$  formation in  $\text{PM}_{2.5}$  during DS (Figure 1). But, as shown in Figure S9, the concentration of  $\text{NO}_3^-$  in the coarse mode in DS was almost twice times higher than in NDS, indicating abundant  $\text{NO}_3^-$  formed in DS, especially in the coarse mode.

735





**Figure S9.** Linear fit regression for the  $\text{NH}_4^+$  with (a)  $\text{NO}_3^-$  and (b) oxalic acid in coarse mode ( $>2.1 \mu\text{m}$ ) of particles in the non-dust storm (NDS) and dust storm (DS) periods in Shanghai during spring of 2023.

10. In Section 3.4, the author should give the detailed information of the measurement of absorption, such as the solvent, the filter mass, etc., to reduce the uncertainty. In addition, the relationship of the concentration of chromophores or the ratio to WSOC with MAC or Abs should be added to differentiate the dust and haze. The filter of dust is yellow, but the haze filter may be black or grey, how to differentiate the influence of inorganic components.

**Reply:** Suggestion taken. The detail information about experiment, including instrument, solvent, filter areas, were added to the Section 2.2. Please see page 5, lines 125-137.

In this study, the filter samples were extracted by ultrapure water. The extract was filtered through a  $0.45 \mu\text{m}$  PTFE syringe filter to remove water insoluble particles (black carbon etc.). The light absorption of water-soluble BrC measured using LWCC (World Precision Instrument, Inc., USA). Inorganic components are light absorbing, but the light absorption of those species is in the wavelength smaller than  $250 \text{ nm}$ . In this study, we investigated the Abs and MAC in the wavelength range of  $\lambda = 250\text{--}500 \text{ nm}$  in Shanghai. Thus, the effects of inorganic components on the light absorption were negligible.

“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.”

11. Check the references (pages, volume, issue, journal (abbreviation or full), etc.) to meet the requirement of ACP.

**Reply:** Done. We have checked the references and revised the incorreced references.

12. The current title of the manuscript is overly long and could be more concise. A shorter, more focused title would improve readability and better reflect the core content of the study.

**Reply:** Suggestion taken. New title was revised as follow:

**Measurement report: Simultaneous measurements on gas- and particle-phase water-soluble organics in Shanghai: Enhanced light absorption of transported Asian dust**

## References

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