

### **Referee#1:**

#### **General Overview:**

Li et al. studied the aerosol scattering hygroscopic growth factor ( $f(RH)$ ) in a Chinese coastal city and compared  $f(RH)$  between NPF and non-NPF days. Based on the ACSM measurement, some analyses and explanations have been provided. The difference in  $f(RH)$  between NPF and non-NPF days is due to the variations in  $SO_4^{2-}$ ,  $NO_3^-$  and the oxidation degree of OA.

The study falls into the scope of ACP. But, compared with existing work, I do not see too much novelty in the current version of the manuscript. The quality of the work has been significantly weakened by but not limited to 1) no detailed composition analysis from the ACSM data (e.g., no PMF analysis), 2) no systematic comparison with literature on  $f(RH)$ , 3) too brief discussion many times, 4) inconsistency throughout the manuscript and 5) the use of language. The manuscript will require major revisions, and the comments below need to be fully addressed before the manuscript can be reconsidered for publication.

*Response:* We sincerely appreciate the critical comments and constructive suggestions. In response to your feedback, we have addressed all of the comments, and particularly adopted the suggestion to polish our results. Our responses for the comments as follow.

#### **Major Comments**

**1. Lines 215 – 219:** It is not straightforward to visualize the comparison in a table. It is recommended that the author use a figure to illustrate the comparison. If necessary, the figure should be considered to be placed in the main text. In addition, the author compares  $f(80\%)$ ,  $f(85\%)$ , and  $f(70\%)$  between studies. This needs to be clarified. What are the reasons behind the differences between  $f(RH)$  from different studies? The author needs to discuss that.

*Response:* Thank you very much for your advice. We have used Figure 1 and placed it in the main text to illustrate the comparison more clearly, and have also clarified the parameters of the comparison in Table S2. In general, the differences in aerosol source, and chemical composition results in the variation of  $f(RH)$  from different studies. For example, marine aerosol has the strongest  $f(RH)$ , followed by urban/continental aerosol, while dust and biomass combustion aerosols are the lowest, and this contents are covered in the introduction. The main revisions are as follows and the Figure 1 is shown in Line 228-243.

“The comparison presented in Figure 1 and Table S2 implied that  $f(RH)$  varied widely across different regions, with consistently higher values observed in Europe compared to China. The differences between  $f(RH)$  in this study and in other regions of China were smaller than those outside of China. In urban China (Lin'an, Beijing, Guangzhou and Xiamen),  $f(RH)$  was generally small (Zhang et al., 2015; Zhao et al., 2019b; Ren et al., 2021; Li et al., 2021); whereas in Raoyang, greatly influenced by anthropogenic polluted aerosols,  $f(RH)$  raised significantly (Wu et al., 2017). Urban area in Europe (Granada, Spain) also displayed similar values of  $f(RH)$  (Titos et al., 2014). Nevertheless, the  $f(RH)$  of continental aerosols in Europe (Jungfraujoch, Swiss)

obviously surpassed what had been observed in China (Zieger et al., 2013). In the Arctic (Ny- Ålesund, Svalbard) and the shore of the ocean (Mace Head),  $f(\text{RH})$  showed high values due to the sea salt (Zieger et al., 2010; Zieger et al., 2013). However, Mace Head, which was also exposed to air pollution from the urban areas, had lower  $f(\text{RH})$  than the undisturbed Arctic. Thus, the large variability of  $f(\text{RH})$  across measurement sites was primarily attributed to the different aerosol sources and chemical composition.”

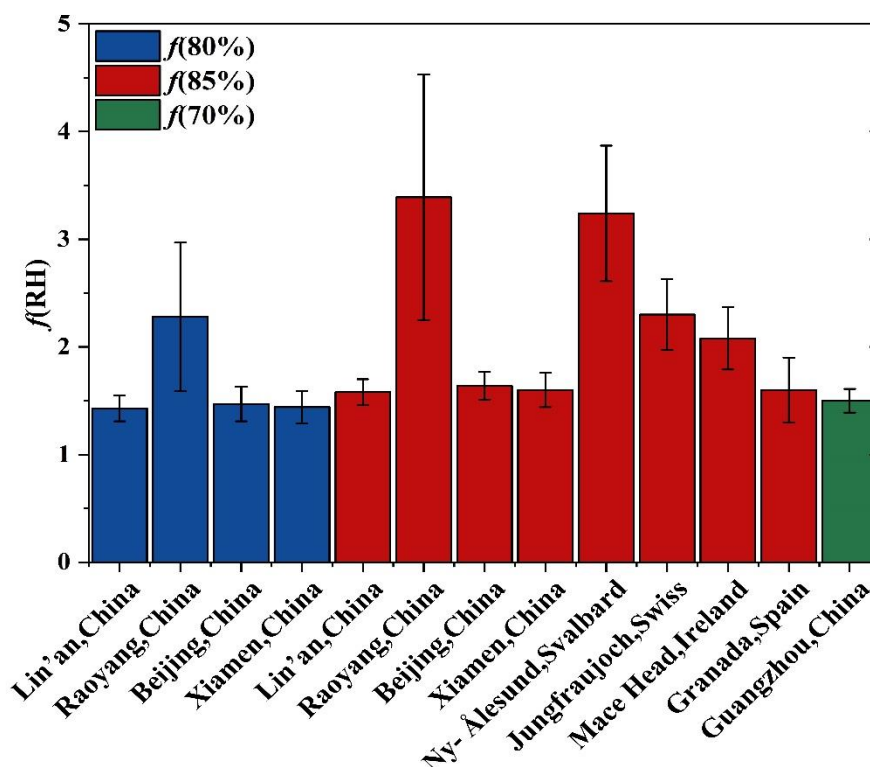


Figure 1. Mean values of  $f(80\%)$ ,  $f(85\%)$  and  $f(70\%)$  values in different observation sites. The error bars represent standard deviation. Blue, green, and red represent  $f(80\%)$ ,  $f(85\%)$  and  $f(70\%)$ , respectively.

#### Reference:

- Li, J. W., Zhang, Z. S., Wu, Y. F., Tao, J., Xia, Y. J., Wang, C. Y., and Zhang, R. J.: Effects of chemical compositions in fine particles and their identified sources on hygroscopic growth factor during dry season in urban Guangzhou of South China, *Science of the Total Environment*, 801, 10.1016/j.scitotenv.2021.149749, 2021.
- Ren, R. M., Li, Z. Q., Yan, P., Wang, Y. Y., Wu, H., Cribb, M., Wang, W., Jin, X. A., Li, Y. A., and Zhang, D. M.: Measurement report: The effect of aerosol chemical composition on light scattering due to the hygroscopic swelling effect, *Atmospheric Chemistry and Physics*, 21, 9977-9994, 10.5194/acp-21-9977-2021, 2021.
- Titos, G., Lyamani, H., Cazorla, A., Sorribas, M., Foyo-Moreno, I., Wiedensohler, A., and Alados-Arboledas, L.: Study of the relative humidity dependence of aerosol light-scattering in southern Spain, *Tellus Series B-Chemical and Physical Meteorology*, 66, 10.3402/tellusb.v66.24536, 2014.
- Wu, Y. F., Wang, X. J., Yan, P., Zhang, L. M., Tao, J., Liu, X. Y., Tian, P., Han, Z. W.,

and Zhang, R. J.: Investigation of hygroscopic growth effect on aerosol scattering coefficient at a rural site in the southern North China Plain, *Science of the Total Environment*, 599, 76-84, 10.1016/j.scitotenv.2017.04.194, 2017.

Zhang, L., Sun, J. Y., Shen, X. J., Zhang, Y. M., Che, H., Ma, Q. L., Zhang, Y. W., Zhang, X. Y., and Ogren, J. A.: Observations of relative humidity effects on aerosol light scattering in the Yangtze River Delta of China, *Atmospheric Chemistry and Physics*, 15, 8439-8454, 10.5194/acp-15-8439-2015, 2015.

Zhao, P. S., Ding, J., Du, X., and Su, J.: High time-resolution measurement of light scattering hygroscopic growth factor in Beijing: A novel method for high relative humidity conditions, *Atmospheric Environment*, 215, 10.1016/j.atmosenv.2019.116912, 2019b.

Zieger, P., Fierz-Schmidhauser, R., Gysel, M., Strom, J., Henne, S., Yttri, K. E., Baltensperger, U., and Weingartner, E.: Effects of relative humidity on aerosol light scattering in the Arctic, *Atmospheric Chemistry and Physics*, 10, 3875-3890, 10.5194/acp-10-3875-2010, 2010.

Zieger, P., Fierz-Schmidhauser, R., Weingartner, E., and Baltensperger, U.: Effects of relative humidity on aerosol light scattering: results from different European sites, *Atmospheric Chemistry and Physics*, 13, 10609-10631, 10.5194/acp-13-10609-2013, 2013.

**2. Lines 316 – 318: Was the wind direction from the major roads near the site? How might the increasing fraction of OM be attributed to the emission from heavy trucks? Did the authors observe any increase in traffic-related marker ions or PMF factors on ACSM?**

*Response:* We thank the referee. There are two major roads with heavy traffic approximately 100 meters to the northwest and northeast of the observation site. According to Figure S1(f) and Figure S6, the wind mainly came from the major roads near the site. Carbon monoxide (CO) and black carbon (BC) are important indicator species for traffic emissions. We analyzed the diurnal trends of CO, BC, and OM on both NPF and non-NPF days (Figure S7). Compared to non-NPF days, OM exhibited homology with BC and CO, showing similar diurnal patterns on NPF days, with high values during the traffic rush hour and nighttime. Therefore, we infer that the increasing fraction of OM on NPF days was related to traffic emissions. This study is mainly focus on the aerosol scattering hygroscopic growth, thereby, we are more concentrated on the variation of  $f(RH)$  related to chemical composition during NPF days. Thank you for your advice; however, source apportionment using ACSM data is not the primary focus of this study, other members of our group are actively working in this area. The modified contents are as follows.

“Based on the wind direction and the homology of OM with BC and CO (Figure S6, S7), the increasing fraction of OM might be attributed to the emissions from heavy truck in the major roads near the observation site, which had a weaker impact on the aerosol hygroscopic growth than the SNA.”

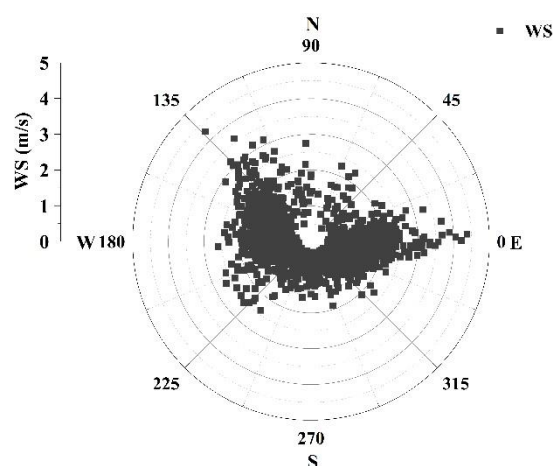


Figure S6. The wind directions and speeds at observation site during the observation period.

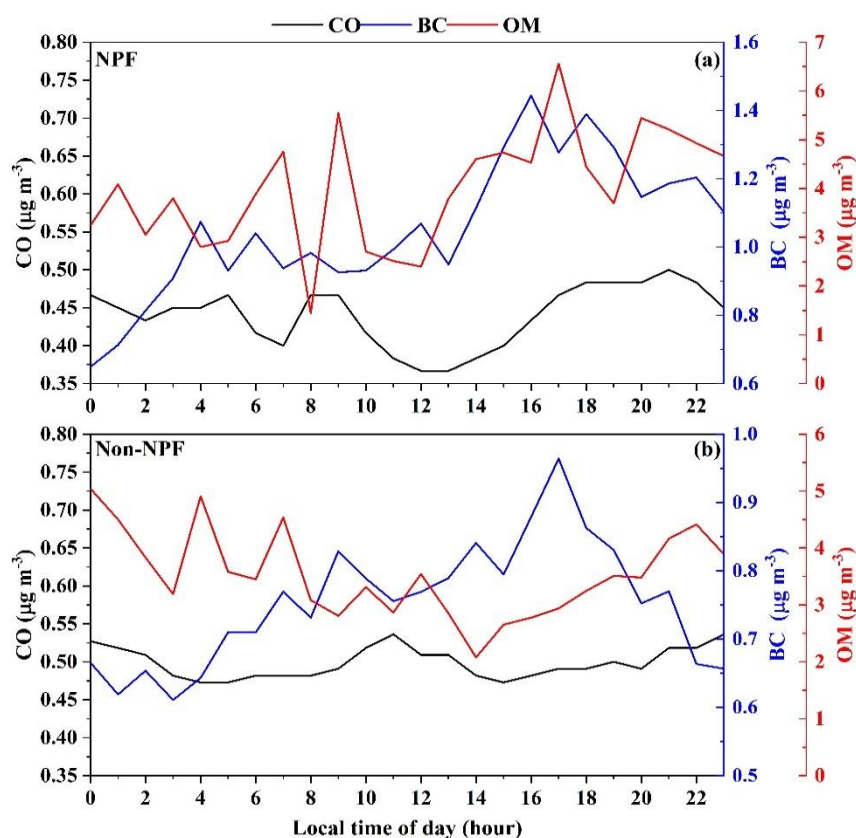


Figure S7. Diurnal variations of CO, BC and OM between NPF (a) and non-NPF (b) days.

3. Lines 320 – 323: The authors claim that the compared to non-NPF days, a higher mass fraction of sulfate on NPF days was possibly due to the nucleation of sulfuric acid. Have the authors considered other possibilities, e.g., the increased sulfate fraction can be attributed to the increase in other species? Have the authors

**observed in any absolute increase in SO<sub>2</sub> on NPF days?**

*Response:* We thank the referee. As suggested, we have analyzed SO<sub>2</sub> concentrations during NPF days and non-NPF days (Figure S8). The SO<sub>2</sub> concentrations on NPF days were clearly higher than those on non-NPF days. Moreover, SO<sub>2</sub> concentrations increased from the morning and peaked around 15:00 on NPF days, which was consistent with the increasing trend of particle number concentration on NPF days. The reaction of SO<sub>2</sub> and OH radicals is considered as the primary pathway of sulfuric acid generation. Condensation mode reactions could occur due to the large amount of sulfuric acid in the atmosphere during NPF events. Recent studies have revealed several mechanisms of sulfate formation, such as manganese-catalyzed oxidation of SO<sub>2</sub> on aerosol surfaces and oxidation of SO<sub>2</sub> by NO<sub>2</sub> and HONO. These mechanisms are reported to occur under the conditions of heavy atmospheric pollution and hazy days. However, during the entire campaign, the atmospheric environment near the observation site was relatively low pollution or even clean conditions, and the concentrations of precursors were low. Therefore, we suggest that condensation mode reactions occurring simultaneously with NPF events were mainly responsible for the higher mass fraction of sulfate during the NPF days observed at our site. Thank you for your insightful comments, we will conduct in-depth analysis of this in future studies. The modified contents are as follows.

“The mass fraction of sulfate was much higher than that of nitrate on NPF days compared to non-NPF days. The SO<sub>2</sub> concentrations on NPF days were clearly higher than that on non-NPF days (Figure S8). Moreover, SO<sub>2</sub> concentrations increased from the morning and peaked around 15:00 on NPF days, which was consistent with the increasing trend of particle number concentration on NPF days. The reaction of SO<sub>2</sub> and OH radicals is considered as the primary pathway of sulfuric acid generation (Sipilä et al., 2010). Condensation mode reactions could occur due to the large amount of sulfuric acid in the atmosphere during NPF events. It is hypothesized that condensation mode reactions occurring simultaneously with NPF events were mainly responsible for the higher mass fraction of sulfate during the NPF days observed at our site (Yue et al., 2010).”

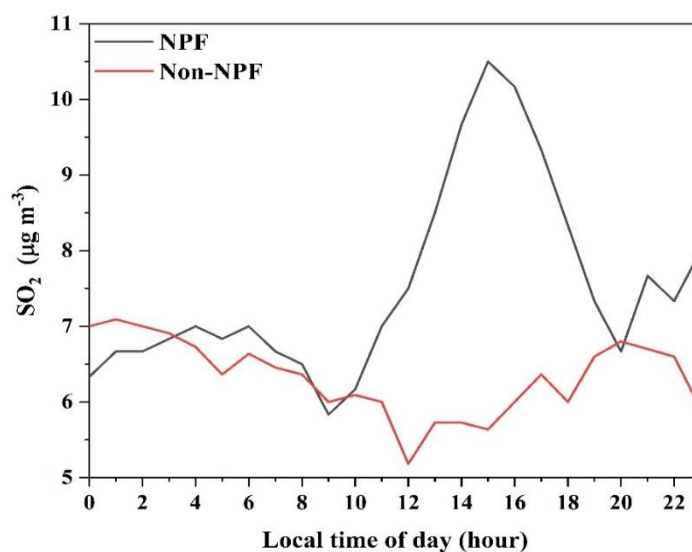


Figure S8. Diurnal variations of SO<sub>2</sub> between NPF and non-NPF days.

Reference:

Sipilä, M., Berndt, T., Petäjä, T., Brus, D., Vanhanen, J., Stratmann, F., Patokoski, J., Mauldin, R. L., Hyvärinen, A. P., Lihavainen, H., and Kulmala, M.: The Role of Sulfuric Acid in Atmospheric Nucleation, *Science*, 327, 1243-1246, 10.1126/science.1180315, 2010.

Yue, D. L., Hu, M., Zhang, R. Y., Wang, Z. B., Zheng, J., Wu, Z. J., Wiedensohler, A., He, L. Y., Huang, X. F., and Zhu, T.: The roles of sulfuric acid in new particle formation and growth in the mega-city of Beijing, *Atmospheric Chemistry and Physics*, 10, 4953-4960, 10.5194/acp-10-4953-2010, 2010.

**4. Section 3.3: As Xiamen is a coastal city, the influence of sea air masses and the contribution of sea salt are expected to be considerable. As far as I know, the ACSM is not good at quantifying the refractory components (e.g., sea salt). When discussing the relationship between  $f(\text{RH})$  and aerosol chemical composition, a thorough discussion on the influence of sea salt must be provided as well.**

*Response:* We thank the advice of referee. To better discuss the influence of aerosol chemical components, including sea salt, on  $f(\text{RH})$ , we have previously used data from Monitor for AeRosol and GaSes (MARGA) and OC/EC Analyzer for analysis. We found that the concentration and mass fraction of sea salt (SS) in the aerosols were very low during the observation period (see figure below), and its influence on  $f(\text{RH})$  was very limited. However, due to the unstable state of MARGA and the large amount of missing data, we ultimately chose to use ACSM data for analysis. Thank you for your insightful comments. We will subsequently focus on the influence of sea salt on  $f(\text{RH})$  in Xiamen and add this to our future study.

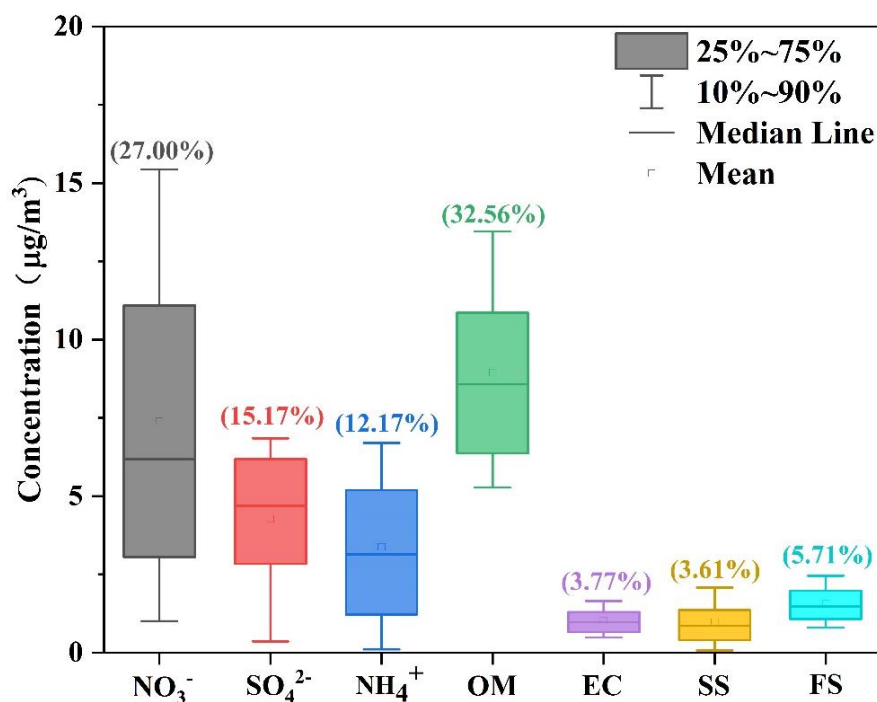


Figure RC1-1. The concentration and mass fraction of each component in the aerosols



**5. Lines 332 – 337: The author speculated on the condensation of large quantities of sulfuric acid and organic vapors on NPF days. Unless there was CIMS measurement, this statement is speculative and can be misleading.**

*Response:* Thank you for your comments. Unfortunately, CIMS was not involved in this observation. In the absence of data on sulfuric acid and organic vapours, we refer to Wu et al. (2016) to explore possible reasons for the low  $f(RH)$  values during NPF days at the observation site. Previous observations and experiments have shown that during particle formation, a large amount of condensable vapors, such as sulfuric acid and secondary organic species, are produced due to strong photochemical activity. These condensable vapours can condense onto pre-existing particles, causing the transformation from an external mixture to an internal mixture. Such a transformation may alter the characteristics of pre-existing particles, including their aerosol optical and chemical properties during new particle formation events. And changes in these properties can affect the aerosol scattering hygroscopic growth. As mentioned above, this is a reasonable assumption based on previous research combined with our observations. In the subsequent work, we are exploring NPF events using CIMS, and will focus on and complement this aspect in our future study. The modified contents are as follows.

“On the basis of aerosol chemical compositions during NPF, it could be speculated that when particle formation occurs in NPF days, the condensation of large quantities of sulfuric acid and organic vapours onto the pre-existing particles, resulting the conversion of mixed state on the surface of particles from external mixture to internal mixture and alteration of the optical and chemical properties of particles, which in turn might change the aerosol scattering hygroscopicity growth (Wu et al., 2016).”

Reference:

Wu, Z. J., Zheng, J., Shang, D. J., Du, Z. F., Wu, Y. S., Zeng, L. M., Wiedensohler, A., and Hu, M.: Particle hygroscopicity and its link to chemical composition in the urban atmosphere of Beijing, China, during summertime, *Atmospheric Chemistry and Physics*, 16, 1123-1138, 10.5194/acp-16-1123-2016, 2016.

**6. Lines 338 -353: Nitrate was assumed to be the driving factor for the aerosol scattering hygroscopic growth. This is supported by a significant decline in  $f(RH)$  when both nitrate content and NOR were low. When NOR and RH were shown in Figure 4, there was no data for nitrate content and  $f(RH)$  in the same figure.**

*Response:* We thank your suggestions. By comparing Figure 4 (a) (c) with Figure 5 (a), the trends for NOR with  $f(RH)$  and nitrate content can be clearly identified. These three graphs show the diurnal patterns of  $f(RH)$ , nitrate content and NOR on NPF days. At around 9:00, NOR began to rise, and  $f(RH)$  and nitrate content increase subsequently. NOR peaked around 15:00 and then declined, while  $f(RH)$  and nitrate content showed the same trend in the following hours. Such a pattern reflected the rapid response of aerosol hygroscopic growth to nitrate, which could be explained by the stronger hygroscopicity of nitrate compared to sulfate. Subsequent correlation analysis in

Section 3.4 also confirmed this point. Your comments made me realize that my statement was not clear enough, the revised contents are as follows.

“We assume that nitrate was essential for aerosol scattering and hygroscopic growth. This assumption was confirmed by comparing Figure 4(a) (c) with Figure 5(a), which show a significant decline in  $f(RH)$  when both nitrate content and NOR were low, especially during NPF days.”

**7. Lines 351 – 353: I understand that the SOR and NOR were higher on non-NPF days compared with NPF days. However, it is unclear to me how the enhanced SOR and NOR were possibly driven by aqueous phase reaction. A detailed analysis needs to be provided to support the statement.**

*Response:* Your suggestions are very helpful to improve the quality of our study. In non-NPF days, RH greater than 60% indicated high humidity condition. When aerosol hygroscopic growth occurs under the high-humidity condition, water vapour condenses on the particles. The increase in aerosol liquid water content (ALWC) on the surface of aerosol particles promotes heterogeneous reactions in the atmosphere. The heterogeneous hydrolysis of  $N_2O_5$  is one of the main ways to produce nitrate, and sulfate can be formed by the oxidation of  $SO_2$  in the aqueous phase. In other words, aqueous-phase reactions contribute to the formation of secondary inorganic aerosols. NOR and SOR are commonly used to characterize the atmospheric secondary transformation of  $NO_2$  and  $SO_2$  to  $NO_3$  and  $SO_4$  in the atmosphere, respectively. Thus, the enhanced SOR and NOR were possibly driven by aqueous phase reaction during non-NPF days. We have added some explanations and literatures in Line 400-409 to support the statement.

“In non-NPF days with high RH, water vapour condensed on the particles and aerosol hygroscopic growth occurred (Martin, 2000). The increase in aerosol liquid water content (ALWC) on the surface of particles is crucial for heterogeneous reactions in the atmosphere (Mogili et al., 2006). The heterogeneous hydrolysis of  $N_2O_5$  (Pathak et al., 2009) and the aqueous-phase oxidation of  $SO_2$  (Seinfeld et al., 1998; Sun et al., 2013) are important pathways for nitrate and sulfate formation, respectively. In other words, aqueous-phase reactions contribute to the production of secondary aerosol (Ge et al., 2012; Xu et al., 2017a). Thus, the elevation of RH during non-NPF days might promote the transformation of  $NO_2$  and  $SO_2$  to nitrate and sulfate via aqueous-phase reactions, manifesting as the enhancement of NOR and SOR.”

Reference:

Ge, X. L., Zhang, Q., Sun, Y. L., Ruehl, C. R., and Setyan, A.: Effect of aqueous-phase processing on aerosol chemistry and size distributions in Fresno, California, during wintertime, *Environmental Chemistry*, 9, 221-235, 10.1071/en11168, 2012.  
Martin, S. T.: Phase Transitions of Aqueous Atmospheric Particles, *Chemical Reviews*, 100, 3403-3454, 10.1021/cr990034t, 2000.  
Mogili, P. K., Kleiber, P. D., Young, M. A., and Grassian, V. H.:  $N_2O_5$  hydrolysis on the components of mineral dust and sea salt aerosol: Comparison study in an environmental aerosol reaction chamber, *Atmospheric Environment*, 40, 7401-7408,



10.1016/j.atmosenv.2006.06.048, 2006.

Pathak, R. K., Wu, W. S., and Wang, T.: Summertime PM<sub>2.5</sub> ionic species in four major cities of China: nitrate formation in an ammonia-deficient atmosphere, *Atmospheric Chemistry and Physics*, 9, 1711-1722, 10.5194/acp-9-1711-2009, 2009.

Seinfeld, J., Pandis, S., and Noone, K.: *Atmospheric Chemistry and Physics: From Air Pollution to Climate Change*, *Physics Today*, 51, 88-90, 1998.

Sun, Y. L., Wang, Z. F., Fu, P. Q., Jiang, Q., Yang, T., Li, J., and Ge, X. L.: The impact of relative humidity on aerosol composition and evolution processes during wintertime in Beijing, China, *Atmospheric Environment*, 77, 927-934, 10.1016/j.atmosenv.2013.06.019, 2013.

Xu, L. L., Duan, F. K., He, K. B., Ma, Y. L., Zhu, L. D., Zheng, Y. X., Huang, T., Kimoto, T., Ma, T., Li, H., Ye, S. Q., Yang, S., Sun, Z. L., and Xu, B. Y.: Characteristics of the secondary water-soluble ions in a typical autumn haze in Beijing, *Environmental Pollution*, 227, 296-305, 10.1016/j.envpol.2017.04.076, 2017a.

**8. Lines 384 – 386: The main drive for aerosol hygroscopicity on NPF days is ambiguous. In lines 345 – 347, nitrate was suggested to be essential for aerosol hygroscopicity on NPF days. Here, the sulfate was suggested to be important in the aerosol hygroscopicity on NPF days. Consistent statements need to be provided through the whole manuscript.**

*Response:* We thank the referee. Through the analyses in Sections 3.3 and 3.4, we emphasized that SNA (SO<sub>4</sub>, NO<sub>3</sub>, and NH<sub>4</sub>) was critical for  $f(\text{RH})$  and aerosol hygroscopic growth, whether on NPF days or non-NPF days, with nitrate having the most pronounced impact. However, during NPF days,  $f(\text{RH})$  and aerosol hygroscopic growth were weaker than those on non-NPF days, due to the higher content of sulfate compared to that of nitrate. Sulfate therefore also had an important effect on  $f(\text{RH})$  during NPF days. We apologize for the misunderstanding caused by the imprecise expression, and the revision is as follows.

“This indicated that sulfate had an important influence on the aerosol hygroscopicity enhancement during NPF period, while nitrate was the primary contributor for non-NPF days.”

**9. Lines 488 – 493: What are the reported k<sub>OA</sub> values in other studies? Could the author use a table or figure to summarize the comparison? How different are the organic aerosol composition between studies?**

*Response:* We thank the referee for this helpful suggestion. We have added Table 1 in Line 567. According to ACSM PMF analysis, we have divided the OA factors into primary organic aerosol (POA) and secondary organic aerosol (SOA) factors. POA is the unoxxygenated component, and SOA is a more oxygenated organic aerosol. The proportion of POA and SOA in OA in this study (Figure S10) showed that higher SOA mass fractions on non-NPF days than on NPF days. Combined with Figure 10(a), it is evident that high SOA mass fractions on non-NPF days corresponded to high  $\kappa_{\text{OA}}$  values. Wu et al. (2016) reported that the POA/OA and SOA/OA ratios in their case were 0.39 and 0.61, respectively, and POA was considered hydrophobic ( $\kappa_{\text{POA}} = 0$ ). Kuang et al.

(2020) found that the rapid formation of oxygenated OA, the secondary factor, and the decrease in POA resulted in the growth of  $\kappa_{OA}$ . In the study by Kuang et al. (2021), the mass fraction of SOA was greater than 70% and had a remarkable effect on  $\kappa_{OA}$ , whereas the hygroscopicity of POA was very low. The study by Chang et al. (2010) showed that the  $\kappa_{OA}$  was also high when the mass fraction of oxygenated organic aerosol was high. These results highlighted that SOA, oxygenated organic aerosol, was very likely the determining factor of  $\kappa_{OA}$ . We have added some explanations in Line 559-571.

“The  $\kappa_{OA}$  values were greater than those of the previous study in China (Wu et al., 2016; Kuang et al., 2020; Kuang et al., 2021), but similar to the findings of Chang et al. (2010) at rural site in Ontario, Canada (Table 1). The proportion of POA and SOA in OA in our study (Figure S10) showed higher SOA mass fractions on non-NPF days than on NPF days. It is evident that high SOA mass fractions on non-NPF days corresponded to high  $\kappa_{OA}$  values. The results from these previous studies (Wu et al., 2016; Kuang et al., 2020; Kuang et al., 2021; Chang et al., 2010) also highlighted that SOA and oxygenated organic aerosols, were likely to be the determinants of  $\kappa_{OA}$ . Furthermore, SOA dominated OA mass both in this study and previous studies; however,  $\kappa_{OA}$  values differed much across studies. Noted that the hygroscopicity of SOA might vary significantly under different emission and atmospheric conditions due to variations in VOC precursors and SOA formation pathways (Kuang et al., 2021).”

Table 1. Comparisons of the average  $\kappa_{OA}$  in different study.

| Study area      | Periods          | Remarks | Mean $\kappa_{OA}$           | Reference           |
|-----------------|------------------|---------|------------------------------|---------------------|
| Beijing, China  | Summer, 2014     | urban   | 0.06                         | Wu et al. (2016)    |
| Dingxing, China | 2018/11/11-12/24 | rural   | $0.08 \pm 0.06$              | Kuang et al. (2020) |
| Heshan, China   | 2018/9/30-11/17  | rural   | $0.085 \pm 0.05$             | Kuang et al. (2021) |
| Ontario, Canada | Spring, 2007     | rural   | $0.22 \pm 0.04$              | Chang et al. (2010) |
| Xiamen, China   | 2022/2-4         | urban   | $0.13 \pm 0.11$              | This study          |
|                 |                  |         | (NPF)                        |                     |
|                 |                  |         | $0.19 \pm 0.21$<br>(non-NPF) |                     |

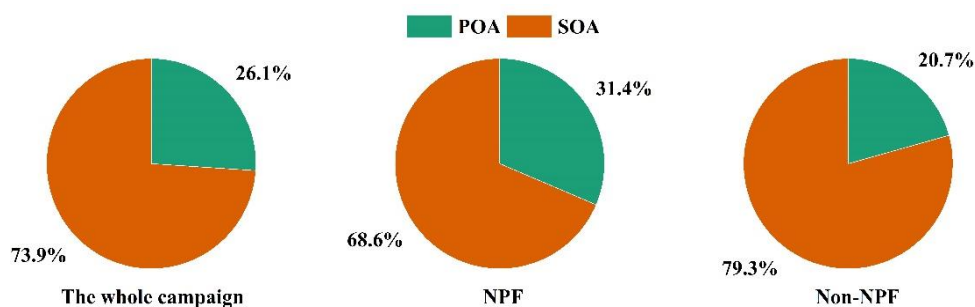


Figure S10. The proportions of POA and SOA in OA.

## Minor Comments

**1. Lines 46-47 and Lines 49 -50 : How does the aerosol hygroscopicity alter particle size and refractive index? How does the aerosol hygroscopicity affect chemical processes and air quality? This is difficult for readers without little background knowledge of hygroscopicity. Could the author give a better explanation of that?**

*Response:* We thank the referee for this useful suggestion. As aerosols absorb water, their sizes, shapes, and refractive indices change, and more particles grow into sizes that are more efficient for light scattering, thus enhancing the scattering of light by aerosol. Due to the aerosol hygroscopicity, ALWC increases as they absorb water in humid environment. The condensed water in aerosols serves as an effective medium for multiphase chemistry, thus promoting the transformation of active gaseous pollutants into particles. The newly formed hygroscopic particle components, such as secondary aerosols, can also alter aerosol hygroscopic behaviors and enhance aerosol extinction efficiency. These processes lead to regional visibility impairment and accelerated formation of heavy haze. As suggested, we have added detailed explanations in Line 49-60.

“Particles absorb water through hygroscopic growth, growing to sizes that are more efficient for light scattering, and their refractive index changes, resulting in enhanced aerosol scattering (Seinfeld et al., 1998).”

“The aerosol liquid water content (ALWC) increases as particles absorb water in humid environment due to aerosol hygroscopicity. The condensed water in aerosols serves as an effective medium for multiphase chemistry, promoting the transformation of active gaseous pollutants into particles. Meanwhile, the newly formed hygroscopic aerosol components, such as secondary aerosols, can also alter aerosol hygroscopic behaviors and enhance aerosol extinction efficiency. These processes lead to regional visibility impairment and accelerated formation of heavy haze. Therefore, aerosol hygroscopicity can profoundly affect atmospheric chemical processes (Wu et al., 2018) and air quality (Liu et al., 2020a).”

## Reference:

Seinfeld, J., Pandis, S., and Noone, K.: Atmospheric Chemistry and Physics: From Air Pollution to Climate Change, Physics Today, 51, 88-90, 1998.

**2. How were the two nephelometers compared when operated under dry conditions?**

*Response:* We thank the referee. The multi-band dual-nephelometer system consisted of two nephelometers. The sample airflow initially entered and passed through Nafion dryers which could reduce the RH of the airflow to less than 30%. After this, the airflow was divided into two routes, one was directed straight into the nephelometer for dry aerosols; while the other was humidified via a humidifier before flowing into the second nephelometer for humidified aerosols. The temperature cycle of the humidifier linked to the second nephelometer was controlled by two water baths, which provided circulating water alternatively for the humidifier. When one water bath was heating up the water for humidifying, the RH of the airflow through the humidifier increased as

the water temperature rose. Simultaneously, another water bath was cooling down the water itself, and no water entered the humidifier. When the airflow had been humidified to a setting maximum RH, the water bath with cool water was switched into the humidifier, causing the RH of the airflow to drop rapidly. As the water bath was heated, the RH of the airflow then rose gradually again. Thus, two nephelometers in the system were not operating under dry conditions simultaneously. This study set the minimum and maximum RH at 40% and 91%, respectively, with a 45-minute cycle for humidification. A separate nephelometer measured the aerosol scattering coefficient under ambient humidity conditions. In Line 140-141, we have corrected the description of circulating water. The detailed principles and operation of the multi-band dual-nephelometer system are shown in Supplement material (Text S1).

“The space between these two tubes contained circulating water, which was heated by two water baths.”

**3. Please give more information about the ACSM. Is it a ToF-ACSM or Q-ACSM? What are the RIEs used for different species?**

*Response:* Thanks. we have added some information about the ACSM in Line 154-161. “The hourly chemical composition of aerosol, including sulfate (SO<sub>4</sub>), nitrate (NO<sub>3</sub>), ammonium (NH<sub>4</sub>), chloride (Cl<sup>-</sup>) and organic matter (OM), was measured by a high-resolution Aerodyne Aerosol Chemical Speciation Monitor (Q-ACSM). The relative ion efficiency (RIE) for SO<sub>4</sub>, NO<sub>3</sub>, NH<sub>4</sub>, Cl<sup>-</sup> and OM was 0.53, 1.1, 5.49, 1.3 and 1.4, respectively. PMF/ME-2 models were performed to identify OA factors resolving primary organic aerosol (POA) and secondary organic aerosol (SOA) in this study. POA is the unoxxygenated component, and SOA is a more oxygenated organic aerosol.”

**4. Lines 177 – 181 should be rearranged into a place where  $\gamma$  and F<sub>0</sub> were in use. I suggest they should be moved closed to the paragraph in lines 403 – 417.**

*Response:* Thank you for this helpful comment. As suggested, we have moved the sentences to Line 467-473.

“The fitting parameter  $\gamma$ , which depends on the aerosol hygroscopicity, is defined as  $\gamma = \ln(RH)/\ln((100/RH_{ref})/(100-RH))$  (Quinn et al., 2005; Zhang et al., 2015). Here  $\gamma$  was based on  $RH_{ref} = 40\%$  and  $RH = 80\%$ .  $\gamma$  can be employed to characterize the relationship between aerosol hygroscopic growth and SNA. The relative quantity of OM and inorganic matters can be expressed as  $F_o = OM/(OM + C_i)$ , where  $C_i$  is the mass concentration of SNA. We chose NO<sub>3</sub>, SO<sub>4</sub> and NO<sub>3</sub> + SO<sub>4</sub> as different SNA constitute in this study, respectively.”

**5. Lines 239-242: Why was hygroscopicity at RH >90% was lower than that at 80%<RH<90% in Zhao et al., (2019)? But why was it the same case in this study? More discussion need to be provided.**

*Response:* We thank the referee. Zhao et al., (2019) reported that the hygroscopicity of PM<sub>2.5</sub> for RH > 90% was lower than that when the RH was between 80 and 90% in Beijing during winter, summer and autumn. We found that when RH was between 80% and 90%,  $f(RH)$  increased more than when RH was below 80% in springtime of Xiamen.

These results indicate that particles exhibited different hygroscopicity behaviors under different RH conditions, which were probably related to the chemical composition, particle size and morphology of aerosols. How aerosol hygroscopicity changes in different humidity ranges, and the specific factors and mechanisms affecting these changes, need to be further studied in the future. The revised contents as follows.

“Moreover, Zhao et al. (2019) found a prominent difference in the aerosol hygroscopicity as RH beyond 90%, with aerosol hygroscopicity in this humidity range being lower than when RH was below 90%. These results indicate that particles exhibited different hygroscopicity behaviors under different RH conditions, which were probably related to the chemical composition, particle size and morphology of aerosols.”

**6. Lines 242 -244: To support the sentence “Secondly, the characteristics of  $f(RH)$ ... were minor”, a statistical analysis (e.g., ANOVA) needs to be conducted.**

*Response:* Thanks. A statistical analysis (ANOVA) has been conducted. The result revealed that  $f(RH)$  was significantly different between NPF days and non-NPF days. The revised contents as follows.

“Secondly, the characteristics of  $f(RH)$  were distinct among different days, especially between NPF days and non-NPF days (Figure S4).”

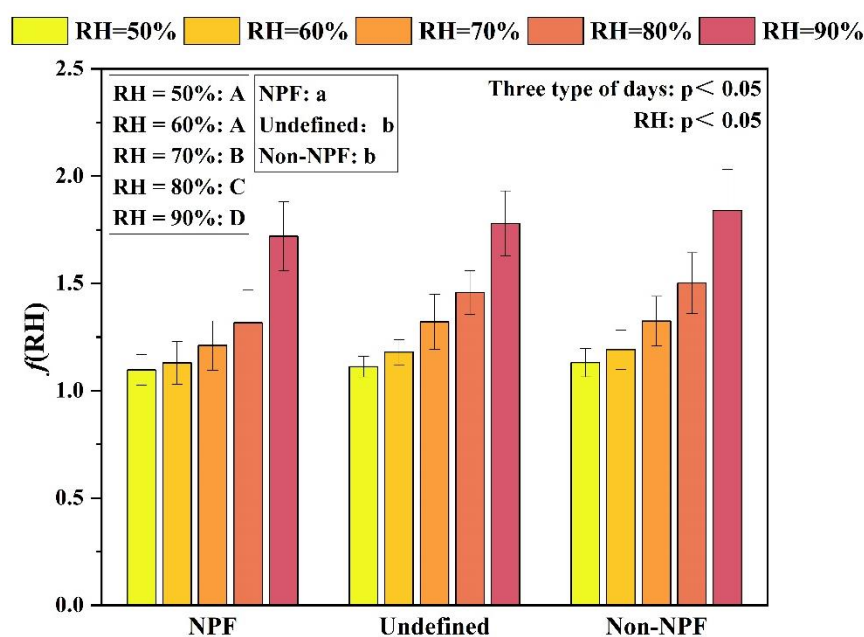


Figure S4. Effects of different days and RH on  $f(RH)$ . Different letters indicate significant differences according to posthoc comparisons. Significance was  $p < 0.05$ . Data are represented as mean  $\pm$  SD ( $n=2$ ).

**7. Lines 249 – 252: Could the authors provide qualitative analysis about the fluctuations of  $f(RH)$  and the dramatic increase in particle number conc., variations in chemical composition?**

*Response:* We thank the referee. The figure below shows the diurnal variation of  $f(80\%)$ , aerosol chemical composition concentrations, and particle number concentrations on

NPF days. It can be seen that the aerosol composition concentrations and particle number concentrations fluctuated in tandem with  $f(80\%)$ . Especially during NPF events, the particle number concentrations increased first, followed by increases in the aerosol composition concentrations and  $f(80\%)$ . The analysis of section 3.3, 3.4 and 3.5 also indicates that the aerosol scattering hygroscopic growth factor is closely related to aerosol chemical composition.

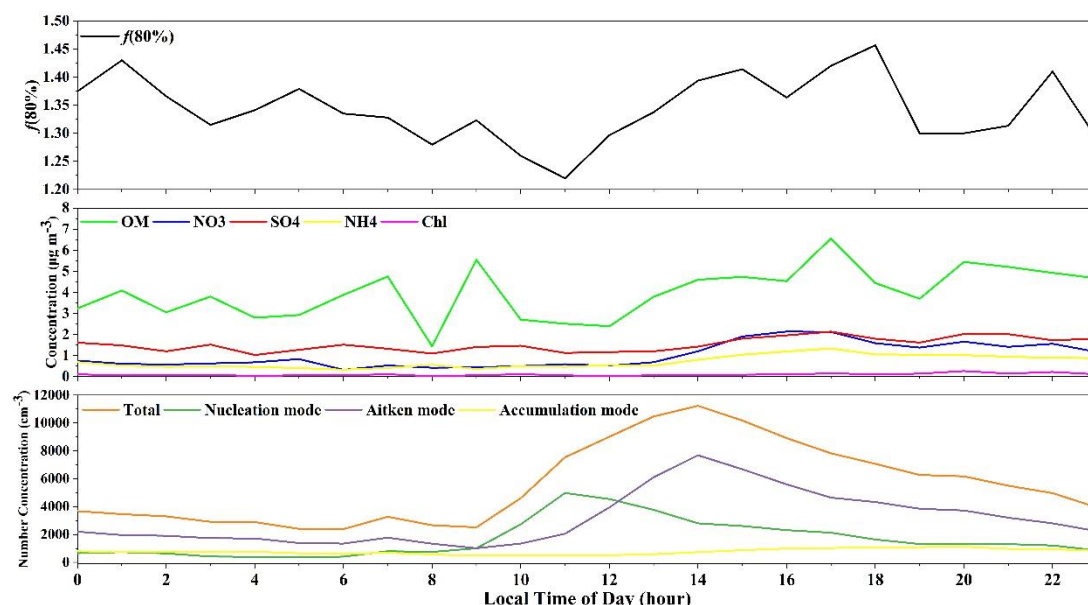


Figure RC1-2. The diurnal variation of  $f(80\%)$ , aerosol chemical composition concentrations, and particle number concentrations on NPF days.

**8. Lines 262 -266: Why is the Eq. (1) the most suitable for the analysis? What is it based on?  $R^2$ ?**

*Response:* We thank the referee. As mentioned in Text S5 and Figure S5 in supplementary materials, the comparison of the fitting curves,  $R^2$  values, simulated and measured values of  $f(80\%)$  for each parameterization scheme was conducted. We found that Eq. (2) (i.e., Eq. (S5) in the supplementary materials) had the best fitting curve, the highest  $R^2$  value, and it also had the smallest difference between the simulated and the measured values of  $f(80\%)$ . Therefore, Eq. (2) was considered to be the most suitable parameterization scheme.

**9. Lines 274 – 275: What are more complex factors? More complex to what? What was compared to?**

*Response:* We thank the referee to point out this. We have modified the contents as follows.

“This reflects the fact that  $f(RH)$  on NPF days was influenced by more complex factors than on non-NPF days, including the source, composition, and morphology of the aerosols.”

**10. Lines 301 – 302: The data showed in Deng et al., 2016 is one-year data but not decade-long data. The sentence needs to be rewritten to avoid misunderstanding.**



*Response:* We thank the referee for careful reading. We had rewritten this sentence to avoid misunderstanding.

“Compared with ten years ago, the concentrations of all SNA in Xiamen have significantly decreased.”

**11. Line 303: What are the control measures?**

*Response:* We thank the referee. In Xiamen, the main measures to control sulfate include the application of desulfurization technology in the flue gas of power plants and coal combustion boilers, as well as the promotion of clean energy for seaships.

**12. Line 304 -306: The statement “the prominent nitrate pollution” is based on a comparison between this study and another study focusing on winter in 2013. In my opinion, the statement completely lacks supporting evidence, unless the cited study focused on the long-term data.**

*Response:* The constructive comments are appreciated. According to the referee’s comment, the revised contents are as follows.

“However, in contrast to the studies in 2011-2013 and 2017 (Wu et al., 2015a; Wu et al., 2020), the ratio of nitrate to sulfate had increased in this study, suggesting that nitrate pollution has become more prominent in recent years.”

Reference:

Wu, S. P., Schwab, J., Yang, B. Y., Zheng, A., and Yuan, C. S.: Two-Years PM<sub>2.5</sub> Observations at Four Urban Sites along the Coast of Southeastern China, *Aerosol and Air Quality Research*, 15, 1799-1812, 10.4209/aaqr.2015.05.0363, 2015a.

Wu, S. P., Cai, M. J., Xu, C., Zhang, N., Zhou, J. B., Yan, J. P., Schwab, J. J., and Yuan, C. S.: Chemical nature of PM<sub>2.5</sub> and PM<sub>10</sub> in the coastal urban Xiamen, China: Insights into the impacts of shipping emissions and health risk, *Atmospheric Environment*, 227, 10.1016/j.atmosenv.2020.117383, 2020.

**13. Lines 328-329: The sentence “This illuminates that fewer hygroscopic particles were born during the NPF event in Xiamen” is not clear and needs revision.**

*Response:* We thank the referee. As suggested, we have revised this sentence.

“This illuminates that aerosols had a lower hygroscopicity during the NPF event in Xiamen.”

**14. Line 331: What were the difference precursors? Please give a brief explanation.**

*Response:* Thanks. According to the study by Liu et al. (2021), the OA on NPF days may primarily form via photooxidation, resulting in more hygroscopic aerosols, while on the non-NPF days, the mechanism of oligomerization is dominant, yields less hygroscopic products. Therefore, Liu et al. (2021) suggested the measured distinct water uptake capacity of OA between NPF and non-NPF events is attributed to the different OA growth processes: nucleation-initiated photochemical oxidation of VOCs to produce water-soluble products (e.g., organic acids) and aqueous oligomerization to

yield less water-soluble products, respectively. The revised contents as follows.

“The study by Liu et al. (2021) also found that the hygroscopicity of 40 nm organic aerosol (OA) was significantly enhanced during NPF days in urban Beijing, which could be derived from different precursors and accounted for the formation of OA during the NPF process.”

**15. Line 376: What is uncertainty range for the linear regression? Please show the range as shaded areas in Figure 5. Same applies for Figure 6.**

*Response:* Thanks, we had corrected them as Figure 6 and Figure 7.

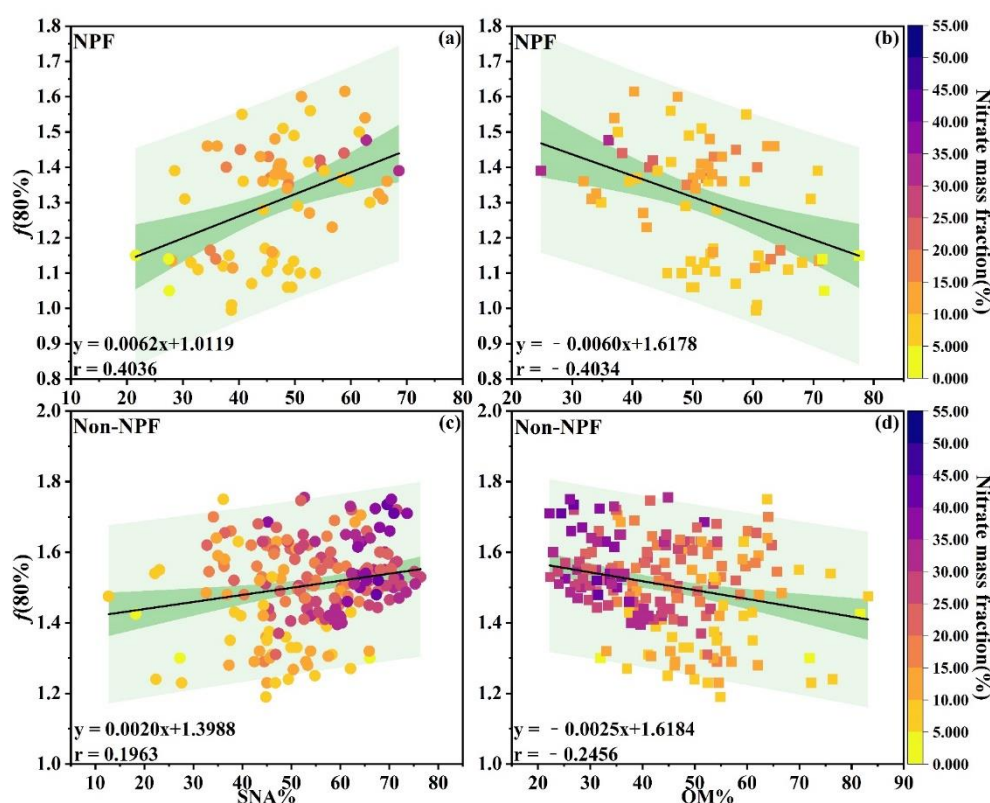


Figure 6. The aerosol scattering hygroscopic growth factor  $f(80\%)$  as a function of SNA and OM mass fraction colored by the nitrate mass fraction. (a) and (b) belong to NPF days; (c) and (d) belong to non-NPF days. The linear regression function and Pearson's correlation coefficient (R) are given in each panel. The dark-color shaded areas denote 95 % confidence levels, and the light-color shaded areas show the 95 % prediction bands for the fits.

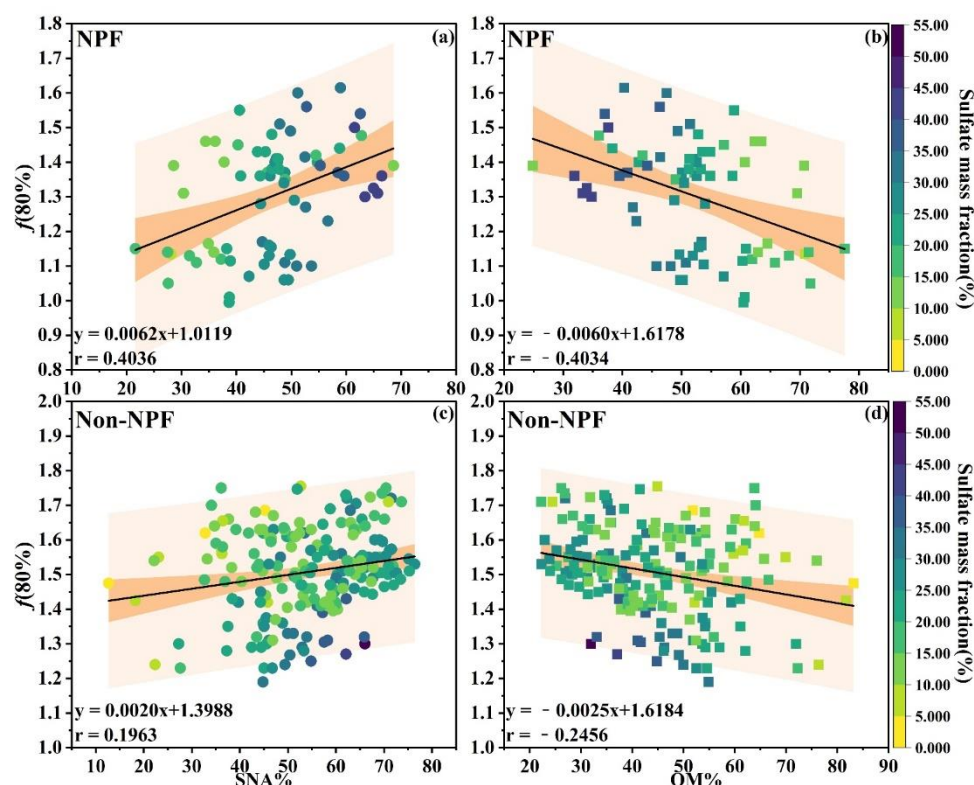


Figure 7. The aerosol scattering hygroscopic growth factor  $f(80\%)$  as a function of SNA and OM mass fraction colored by the sulfate mass fraction. (a) and (b) belong to NPF days; (c) and (d) belong to non-NPF days. The linear regression function and Pearson's correlation coefficient (R) are given in each panel. The dark-color shaded areas denote 95 % confidence levels, and the light-color shaded areas show the 95 % prediction bands for the fits.

16. Lines 415 – 417: What are these studies about? Please provide more background information.

*Response:* Thanks. These studies explored the aerosol chemical composition and aerosol properties, including hygroscopicity and optical properties. They indicated that nitrate played a vital role in aerosol properties and aerosol pollution, which was consistent with the findings in our study.

“This finding also underscored the substantial impact of nitrate on aerosol properties, aligning with recent research conducted in various regions of China.”

17. Line 457: What is the density used for organic when converting the mass fraction into volume fraction?

*Response:* We thank the referee to point out this. The revisions are shown in Text S3.

“The volume concentration of BC was calculated by assuming a density of  $1.7 \text{ g cm}^{-3}$ , and the volume concentration of OA was calculated by assuming that the density of POA is  $1 \text{ g cm}^{-3}$  and density of SOA is  $1.4 \text{ g cm}^{-3}$  (Wu et al., 2016).”

18. Lines 456 – 478: What is the defined RH for  $k_f(\text{RH})$ ?

*Response:* We thank the referee. The hygroscopicity parameter  $\kappa$  retrieved from the

measured aerosol scattering hygroscopic growth factor is usually referred to as  $\kappa_{f(RH)}$ . To reduce the possible influence of observational random errors on the estimated results, all the valid  $f(RH)$  measurements in the period of increasing RH during a complete humidifying cycle are used to derive the overall mean hygroscopicity parameter,  $\kappa$ . Thus,  $\kappa$  calculated from  $f(RH)$  measurements can be understood as an optically weighted  $\kappa$  and represents the overall hygroscopicity of ambient aerosol particles. The RH in  $\kappa_{f(RH)}$  does not have a specific value.

### Technical Comments

**1. Please use “non-NPF days” instead of “Non-NPF days”.**

*Response:* We thank the referee for careful reading. We had corrected “Non-NPF days” to “non-NPF days”.

**2. To increase the readability, it will be good to use a large font size and put labels of “NPF” and “Non-NPF” in figures.**

*Response:* The constructive comments are appreciated. We have improved the figures. And here we did not list the change information but marked in red in the revised manuscript.

**3. Line 52: What RH is classified as high ambient humidity? Could the author define a range?**

*Response:* We thank the referee. In general, a relative humidity of more than 60% is considered high ambient humidity. Xiamen is widely regarded as a warm and humid city. According to our previous research, the average RH in Xiamen was over 60% in spring and summer, and over 50% in autumn and winter (Li et al., 2023). During this observation period, the average RH reached  $63.66 \pm 13.69\%$ , with a maximum value of 87.88%. Furthermore, our analysis shows that the aerosol scattering hygroscopic growth increased significantly when RH was above 60%.

### Reference:

Li, L., Li, M., Zhang, S., Yin, L., Ji, X., Chen, Y., Dong, C., Xu, L., Fan, X., Chen, G., Lin, Z., Hong, Y., Chen, J., and Chen, J.: Seasonal variation of aerosol optical properties in a coastal city of southeast China: Based on one year of measurements, Atmospheric Environment, 305, 119804, <https://doi.org/10.1016/j.atmosenv.2023.119804>, 2023.

**4. Line 62: Lower than what? What is compared with mineral dust and biomass combustion aerosols here?**

*Response:* We thank the referee to point out this. The mineral dust and biomass combustion aerosols are compared here with the marine aerosols and urban/continental aerosols described above. We have made the revisions as follows.

“Mineral dust and freshly emitted biomass combustion aerosols exhibit the lowest  $f(RH)$  values among the aerosol types studied.”

**5. Lines 63 – 66: “Hydrophilic species such...”. Please add references.**

*Response:* Thanks, we have modified the contents as follows.

“Hydrophilic species, such as secondary inorganic components, sea salts, and water-soluble organics in the aerosol are the main contributors to the hygroscopic growth (Li et al., 2021; Fierz-Schmidhauser et al., 2010a), while black carbon and some organic carbons are the major proportion of the hydrophobic species (Liu and Zhang, 2010).”

Reference:

Fierz-Schmidhauser, R., Zieger, P., Vaishya, A., Monahan, C., Bialek, J., O'Dowd, C. D., Jennings, S. G., Baltensperger, U., and Weingartner, E.: Light scattering enhancement factors in the marine boundary layer (Mace Head, Ireland), *Journal of Geophysical Research-Atmospheres*, 115, 10.1029/2009jd013755, 2010.

Li, J. W., Zhang, Z. S., Wu, Y. F., Tao, J., Xia, Y. J., Wang, C. Y., and Zhang, R. J.: Effects of chemical compositions in fine particles and their identified sources on hygroscopic growth factor during dry season in urban Guangzhou of South China, *Science of the Total Environment*, 801, 10.1016/j.scitotenv.2021.149749, 2021.

Liu, X., and Zhang, Y.: Advances in Research on Aerosol Hygroscopic Properties at Home and Abroad, *Climatic and Environmental Research*, 15, 808-816, 2010.

**6. Lines 70 – 71: “For a fixed chemical composition,”. Fierz-Schmidhauser et al., 2010b only measured the  $f(RH)$  of ammonium sulfate and sodium chloride. Please clarify the sentence.**

*Response:* Thanks. For the same substance, the aerosol scattering hygroscopic growth capacity varies with particle size. Generally, this capacity decreases as particle size increases. Zieger et al. (2010) also reported a clear decrease of  $f(85\%)$  with increasing size when assuming the chemical composition to be constant. According to the referee's comment, the revised contents are as follows.

“For a fixed chemical composition,  $f(RH)$  decreases with increasing particle size (Fierz-Schmidhauser et al., 2010b; Zieger et al., 2010; Baynard et al., 2006).

Reference:

Baynard, T., Garland, R. M., Ravishankara, A. R., Tolbert, M. A., and Lovejoy, E. R.: Key factors influencing the relative humidity dependence of aerosol light scattering, *Geophysical Research Letters*, 33, 10.1029/2005gl024898, 2006.

Zieger, P., Fierz-Schmidhauser, R., Gysel, M., Strom, J., Henne, S., Yttri, K. E., Baltensperger, U., and Weingartner, E.: Effects of relative humidity on aerosol light scattering in the Arctic, *Atmospheric Chemistry and Physics*, 10, 3875-3890, 10.5194/acp-10-3875-2010, 2010.

**7. Line 80: What does a relatively low level mean? Relative to what?**

*Response:* We thank the referee. Many researches on  $f(RH)$  were conducted mainly in highly polluted megacities in China, and few studies were conducted in the urban atmosphere with less air pollution. Xiamen is a slightly polluted city compared to megacities, mainly characterized by less particle concentration levels. According to open access data (<https://www.zq12369.com/index.php>), the annual average  $PM_{2.5}$

concentration in Xiamen in 2022 was  $17 \mu\text{g m}^{-3}$ , which was lower than those in many megacities, such as Beijing ( $30 \mu\text{g m}^{-3}$ ), Shanghai ( $25 \mu\text{g m}^{-3}$ ), Guangzhou ( $22 \mu\text{g m}^{-3}$ ), and Xi'an ( $52 \mu\text{g m}^{-3}$ ).

**8. Line 96: I assume these studies point to studies conducted in China. If so, Titos et al., 2014 are miscited here.**

*Response:* We thank the referee to point out this. We had deleted it.

**9. Line 112: What is the meaning of enhanced observation?**

*Response:* We thank the referee. Daily observations at our observation station include measurements of typical atmospheric pollutants (particulate matter, sulphur dioxide, nitrogen oxides, carbon monoxide and ozone), aerosol chemical composition, aerosol optical properties and meteorological parameters. Enhanced observations were carried out to address specific scientific questions, with the aim of this study being to investigate the variation properties of aerosol hygroscopic growth during NPF days. To achieve this, we added a multi-band dual-nephelometer system and a SMPS to our routine measurements to determine  $f(\text{RH})$  and PNSD, respectively. The enhanced observations were conducted continuously from February 1 to April 30, 2022.

**10. Lines 132 -134: Instead of just citing a couple of papers, I suggest the author describe the principle and operation and include them in the supplement.**

*Response:* We thank the referee for the helpful suggestion. The detailed principles and operation of the multi-band dual-nephelometer system have been described in Supplement material (Text S1).

“The multi-band dual-nephelometer system consisted of a nephelometer for aerosol scattering coefficients under dry conditions and another nephelometer for humidified aerosols. The sample airflow initially entered and passed through Nafion dryers which could reduce the RH of the airflow below 30%. After this, the airflow was divided into two routes, one was directed straight into the nephelometer; while the other was humidified via a Gore-Tex tube set in a stainless steel tube before flowing into the other nephelometer. The space between these two tubes contained circulating water. The temperature cycle of the circulating water layer was controlled by two water baths, which provided circulating water alternatively for the humidifier. When one water bath was heating up the water for humidifying, the RH of the airflow through the humidifier increased as the water temperature rose. Simultaneously, another water bath was cooling down the water itself, and no water entered the humidifier. When the airflow had been humidified to a setting maximum RH, the water bath with cool water was switched into the humidifier, causing the RH of the airflow to drop rapidly. As the water bath was heated, the RH of the airflow then rose gradually again. The temperature of the water in the water baths was controlled by an automatic system to ensure the humidifying effect.

In addition, a control software system was used to make sure the RH scans were within a certain RH range. Two combined RH and temperature sensors (Vaisala HMP110; accuracy of  $\pm 0.2^\circ$  and  $\pm 1.7\%$  for RH ranges from 0 to 90 %, respectively, and accuracy



of  $\pm 2.5\%$  for RH ranges from 90 to 100 % according to the manufacturer) were placed at the inlet and outlet of the nephelometer for humidified aerosols, and the measured RHs and temperatures were defined as  $RH_1/T_1$  and  $RH_2/T_2$ , respectively. The dew points at the inlet and outlet of the nephelometer for humidified aerosols were calculated using the measured  $RH_1/T_1$  and  $RH_2/T_2$ , and the average value was considered as the dew point of the sample air. The sample RH can be calculated through the derived dew point and the sample temperature, which is measured by the sensor inside the sample cavity of the nephelometer.”

**11. Line 138: What are the objectives of this study?**

*Response:* Thanks. We do not express the objectives of this study clearly, and we have modified the contents as follows.

“An integrating nephelometer (Aurora-3000, Ecotech, Australia) was used to simultaneously and continuously measure the 5-min average  $\sigma_{sp}$  at the same three wavelengths, and the  $\sigma_{sp}$  at 525 nm was appropriate for characterizing the aerosol scattering coefficient in this study.”

**12. Line 156: Now the sentence reads like “In a 3-month observation, there are 85 days on which NPF can be identified”. This is a very high NPF frequency. In other words, only a few days are classified as non-NPF or unidentified days. Please rewrite the sentence to avoid misunderstanding.**

*Response:* We thank the referee for this valuable suggestion. We apologize for our statement caused you a misunderstanding, and we have made the revisions as follows.

“During the sampling period, a total of 85 days of valid observations were available for PNSD analysis.”

**13. Lines 182 -185: The sentence is too long and difficult to follow. Instead of just citing a paper, the calculation procedure must be detailed in the supplement.**

*Response:* Thanks. As suggested, we have made the revisions as follows and added the detailed calculation procedure in Supplement material (Text S2 and Figure S1).

“The overall hygroscopicity parameter  $\kappa_f(RH)$  can be obtained from the measured  $f(RH)$ . The detailed calculation procedure of this method is shown in Supplement material (Text S2).”

“Brock et al. (2016) proposed a single-parameter representation equation for describing  $f(RH)$ . The equation for  $f(RH)$  is written as:

$$f(RH) = 1 + \kappa_{sca} \frac{RH}{100 - RH} \quad (S1)$$

Where,  $\kappa_{sca}$  is a parameter that fits  $f(RH)$  best.

During processes of measuring  $f(RH)$  with the multi-band dual-nephelometer system, the sample RH in the dry nephelometer condition ( $RH_0$ ) is not zero. Based on Eq. (S1), the measured  $f(RH)_{measured}$  should be fitted using the following equation (Kuang et al., 2017):

$$f(RH)_{measured} = \frac{1 + \kappa_{sca} \frac{RH}{100 - RH}}{1 + \kappa_{sca} \frac{RH_0}{100 - RH_0}} \quad (S2)$$

According to (Kuang et al., 2017), there is a good linear relationship between  $\kappa_{f(RH)}$  and  $\kappa_{sca}$  (Figure S1a). The ratio  $\kappa_{sca} / \kappa_{f(RH)}$  ( $R_\kappa$ ) can be estimated by a look-up table based on the Ångström exponent and  $\kappa_{sca}$  (Figure S1b). With this look-up table,  $R_\kappa$  and  $\kappa_{f(RH)}$  can be directly obtained from measurements of the multi-band dual-nephelometer system. A software for deriving the aerosol hygroscopicity parameter based on measurements from the multi-band dual-nephelometer system and the above principles (BMET, China) was used to obtain  $\kappa_{f(RH)}$  in this study.”

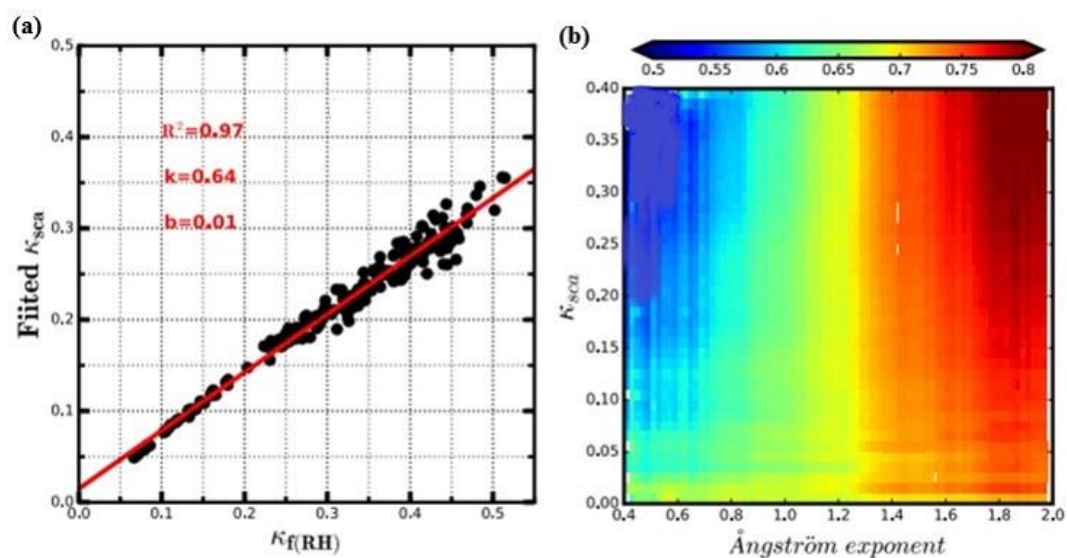


Figure S1. The calculation method of  $\kappa_{f(RH)}$  (Kuang et al., 2017). (a) The good linear relationship between  $\kappa_{f(RH)}$  and  $\kappa_{sca}$ . (b) Colors represent  $R_\kappa$  values and the color bar is shown at the top of this figure. The x axis represents the Ångström exponent and the y axis represents  $\kappa_{sca}$ .

#### Reference:

Brock, C. A., Wagner, N. L., Anderson, B. E., Attwood, A. R., Beyersdorf, A., Campuzano-Jost, P., Carlton, A. G., Day, D. A., Diskin, G. S., Gordon, T. D., Jimenez, J. L., Lack, D. A., Liao, J., Markovic, M. Z., Middlebrook, A. M., Ng, N. L., Perring, A. E., Richardson, M. S., Schwarz, J. P., Washenfelder, R. A., Welti, A., Xu, L., Ziemba, L. D., and Murphy, D. M.: Aerosol optical properties in the southeastern United States in summer - Part 1: Hygroscopic growth, *Atmospheric Chemistry and Physics*, 16, 4987-5007, 10.5194/acp-16-4987-2016, 2016.

Kuang, Y., Zhao, C. S., Tao, J. C., Bian, Y. X., Ma, N., and Zhao, G.: A novel method for deriving the aerosol hygroscopicity parameter based only on measurements from a humidified nephelometer system, *Atmospheric Chemistry and Physics*, 17, 6651-6662, 10.5194/acp-17-6651-2017, 2017.

**14. Line 206: Please clarify the definition of low PM 2.5 pollution in the Chinese AQI standard.**

*Response:* We thank the referee. According to the air quality index (AQI) grading standard of China, a PM<sub>2.5</sub> concentration of 35  $\mu\text{g m}^{-3}$  or less is considered as clean level

(0-50) in the air quality sub-index. Therefore, the average  $PM_{2.5}$  concentration in this study was below  $35 \mu g m^{-3}$ , indicating a relative low  $PM_{2.5}$  pollution level. The revised contents are as follows.

“The mean concentration of  $PM_{2.5}$  was  $24.79 \pm 17.74 \mu g m^{-3}$ , suggested the  $PM_{2.5}$  pollution was relative low in Xiamen referring to the air quality index (AQI) grading standard of China ( $PM_{2.5} \leq 35 \mu g m^{-3}$ ).”

**15. Lines 208 – 209: “standard deviation” is redundant. The format needs to be consistent with other places in the manuscript, e.g., line 205.**

*Response:* Thanks, we have corrected it.

**16. Lines 232- 233: Standard deviation must be provided here for consistency.**

*Response:* Thanks, we have corrected it.

**17. Line 234: Please use “total number concentration”.**

*Response:* Thanks, we have corrected it.

**18. Figure 1: The diverging colour messes up the trend of  $f(RH)$  with increasing RH. Please use sequential colour for better visualization. What are the meanings of whisker and error bars? Can the median be included as well?**

*Response:* We thank the referee for this useful suggestion. The whiskers and error bars represent the mean  $\pm 1.5$  standard deviation. We have improved the figures as Figure 2. The corrected Figure 2 is shown in Line 250.

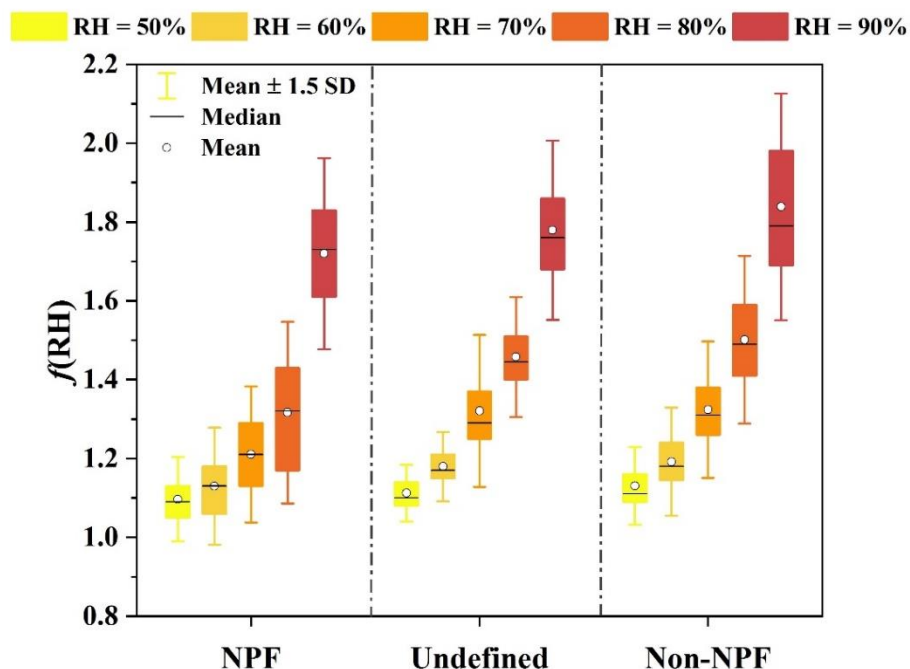


Figure 2. The  $f(RH)$  measured for a given RH in different days. The  $f(RH)$  values at RH=50%, 60%, 70%, 80%, and 90% were counted for the three types of days NPF, Undefined, and non-NPF, respectively. Different colours represent different RH. The error bars represent the mean  $\pm 1.5$  standard deviation.

**19. Lines 238 – 239: Reference(s) is needed for “... the interval of which was the most beneficial to the aerosol scattering hygroscopic growth.”**

*Response:* We thank the referee. The main revisions are as follows.

“Firstly,  $f(\text{RH})$  emerged an approximately exponential rise as RH increasing, with a significant growth when the RH ranged from 80% to 90%, the interval of which was the most beneficial to the aerosol scattering hygroscopic growth (Liu et al., 2013).”

Reference:

Liu, X. G., Gu, J. W., Li, Y. P., Cheng, Y. F., Qu, Y., Han, T. T., Wang, J. L., Tian, H. Z., Chen, J., and Zhang, Y. H.: Increase of aerosol scattering by hygroscopic growth: Observation, modeling, and implications on visibility, *Atmospheric Research*, 132, 91-101, 10.1016/j.atmosres.2013.04.007, 2013.

**20. Line 264: Is Eq (1) in Chen et al., 2014 or in this study?**

*Response:* Thanks, we have corrected it. It is Eq (2) in this study.

**21. Eq (2): Is “a” or “ $\alpha$ ” in the equation? Is the same  $\alpha$  as the one expressed in Eq (1)?**

*Response:* Thanks. It is “a” in the Eq (2). It is not the same  $\alpha$  as expressed in Eq. (1). And “a” is a coefficient that reflects the level of  $f(\text{RH})$  values.

**22. Lines 284 – 286: The sentence “It should be noted that the aerosol scattering... with relatively low levels of particle pollution.” is too hard to follow. What is the meaning of the diminishment of aerosol scattering hygroscopic growth?**

*Response:* We thank the referee. We are sorry for the difficulty of understanding caused by our inappropriate expression. We found that the aerosol scattering hygroscopic growth in lightly polluted areas was not necessarily weaker compared to heavily polluted areas. We have corrected the contents as follows.

“It should be noted that the aerosol scattering hygroscopic growth does not necessarily weaken even in atmospheric environments with light particle pollution.”

**23. Line 296: The sentence “Aerosol chemical compositions play a vital role in the aerosol hygroscopic growth” is redundant here.**

*Response:* Thanks, we have corrected it.

**24. Line 304: Where is the data for winter in 2013?**

*Response:* Thanks. The data for winter in 2013 are from the study by Deng et al. (2016). The contrast here is still with conditions from a decade ago.

**25. Line 312: What is the other event?**

*Response:* We thank the referee. The other event is non-NPF event. Section 3.2 to 3.5 focus on the differences in the aerosol scattering hygroscopic growth and the aerosol hygroscopicity between NPF and non-NPF.

**26. Figure 6: Why is the regression line in d) in red?**

*Response:* We thank the referee to point out this. We have corrected it.

**27. Please add the uncertainty ranges for the linear regression as shaded areas in Figures 2, 5, 6 and 7.**

*Response:* Thanks, we have corrected it. And here we did not list the figures but marked in red in the revised manuscript.

**28. Lines 418 – 420: Please provide references for the sentence “Over the recent decades, the Chinese...”**

*Response:* Thanks, we have modified the contents as follows.

“Over the recent decades, the Chinese government has attached great importance to the air pollution control, and the prominent results have been achieved in reducing SO<sub>2</sub> emissions (Zhang et al., 2019; Zheng et al., 2018).”

Reference:

Zhang, Q., Zheng, Y. X., Tong, D., Shao, M., Wang, S. X., Zhang, Y. H., Xu, X. D., Wang, J. N., He, H., Liu, W. Q., Ding, Y. H., Lei, Y., Li, J. H., Wang, Z. F., Zhang, X. Y., Wang, Y. S., Cheng, J., Liu, Y., Shi, Q. R., Yan, L., Geng, G. N., Hong, C. P., Li, M., Liu, F., Zheng, B., Cao, J. J., Ding, A. J., Gao, J., Fu, Q. Y., Huo, J. T., Liu, B. X., Liu, Z. R., Yang, F. M., He, K. B., and Hao, J. M.: Drivers of improved PM<sub>2.5</sub> air quality in China from 2013 to 2017, *Proceedings of the National Academy of Sciences of the United States of America*, 116, 24463-24469, 10.1073/pnas.1907956116, 2019.

Zheng, B., Tong, D., Li, M., Liu, F., Hong, C. P., Geng, G. N., Li, H. Y., Li, X., Peng, L. Q., Qi, J., Yan, L., Zhang, Y. X., Zhao, H. Y., Zheng, Y. X., He, K. B., and Zhang, Q.: Trends in China's anthropogenic emissions since 2010 as the consequence of clean air actions, *Atmospheric Chemistry and Physics*, 18, 14095-14111, 10.5194/acp-18-14095-2018, 2018.

**29. Line 431: Please rephrase “that of Li’s report”.**

*Response:* Thanks, we have changed the expression of contents as follows.

“However, the correlation between  $\gamma$  and  $F_{O+N+S}$  in this study was lower than that reported by Li et al. (2021).”

**30. Line 468: What does the word “donation” mean?**

*Response:* We thank the referee to point out this. The word “donation” means “contribution” here. The more suitable revision as follows.

“The contribution of AN to aerosol hygroscopicity was predominant during non-NPF days, while the contribution of AS and ABS decreased substantially compared to those during NPF.”

**31. Lines 494 – 497: Please provide references for the sentence about f44.**

*Response:* Thanks. As suggested, we have added literature citations.

“To further investigate the factors affecting  $\kappa_{OA}$ , we compared the effect of OA oxidation level on  $\kappa_{OA}$ , where,  $f_{44}$  was used to represent the level of OA oxidation (Kuang et al., 2020; Chen et al., 2017). The values of  $f_{44}$  in the component mass spectrometry were ratio of  $m/z$  44 to total signals, reflecting the absolute oxidation degree of aerosols (Chen et al., 2022).”

Reference:

Chen, J., Budisulistiorini, S. H., Itoh, M., Lee, W. C., Miyakawa, T., Komazaki, Y., Yang, L. D. Q., and Kuwata, M.: Water uptake by fresh Indonesian peat burning particles is limited by water-soluble organic matter, *Atmospheric Chemistry and Physics*, 17, 11591-11604, 10.5194/acp-17-11591-2017, 2017.

Chen, Y. P., Yang, C., Xu, L. L., Chen, J. S., Zhang, Y. R., Shi, J. Y., Fan, X. L., Zheng, R. H., Hong, Y. W., and Li, M. R.: Chemical composition of NR-PM1 in a coastal city of Southeast China: Temporal variations and formation pathways, *Atmospheric Environment*, 285, 10.1016/j.atmosenv.2022.119243, 2022.

Kuang, Y., He, Y., Xu, W. Y., Zhao, P. S., Cheng, Y. F., Zhao, G., Tao, J. C., Ma, N., Su, H., Zhang, Y. Y., Sun, J. Y., Cheng, P., Yang, W. D., Zhang, S. B., Wu, C., Sun, Y. L., and Zhao, C. S.: Distinct diurnal variation in organic aerosol hygroscopicity and its relationship with oxygenated organic aerosol, *Atmospheric Chemistry and Physics*, 20, 865-880, 10.5194/acp-20-865-2020, 2020.

**32. Line 502: “uplift of oxidation degree of OA” is unclear.**

*Response:* Thanks. The relationship between  $\kappa_{OA}$  and  $f_{44}$  is shown in Figure 10(b). The value of  $f_{44}$  reflects the absolute oxidation degree of organic aerosols (OA). The results showed a positive correlation between  $\kappa_{OA}$  and  $f_{44}$ , indicating that the hygroscopicity of OA increases with the enhanced oxidation of OA. This finding is consistent with many previous studies.



**Referee#2:**

**General comments:**

Aerosol hygroscopicity is a fundamental parameter in atmospheric physics and chemistry, making the results of this study highly relevant to the scope of ACP. While new particle formation (NPF) events are typically associated with active photochemistry and do not directly affect optical hygroscopicity ( $f(RH)$ ), the unique condensation mode chemical reactions occurring during NPF events could have an impact. Therefore, NPF events offer a valuable opportunity to study how the composition of condensation mode aerosols evolves during NPF and how this affects aerosol hygroscopicity. This study presents comprehensive results on the differences in aerosol composition and  $f(RH)$  between NPF and non-NPF days, along with closure studies revealing changes in the hygroscopicity of organic aerosols. Overall, this study provides valuable comparisons between NPF and non-NPF days, such as differences in the RH dependence of  $f(RH)$  and organic aerosol hygroscopicity. As a measurement report, I recommend publication after the following comments are addressed.

*Response:* Thank you very much for your thorough review and constructive comments on our manuscript. As you are concerned, there are several problems that need to be addressed. In response to your valuable feedback, we have carefully considered and made revisions as follow.

**Major comments:**

**1. In the abstract, authors stated that “ NPF occurs frequently and has an obvious effect on  $f(RH)$ ”**

**does NPF could really impacts on  $f(RH)$ ? How was this concluded? NPF means new particle formation, the aerosols with at least size of 150 nm could impact on aerosol scattering thus  $f(RH)$ , and with obvious impacts need aerosol size to be more than 200nm. Authors could estimate how long a newly formed particle could grow to size of more than 200 nm. If authors did the calculation, they would realize that  $f(RH)$  could not be affected by local NPF events, only chemical reactions happened on condensation mode could affect  $f(RH)$ , therefore, if authors find  $f(RH)$  changes during NPF events, it could be condensation mode reactions happened simultaneously with NPF events that changed  $f(RH)$  not the NPF reaction and formation chain.**

*Response:* We sincerely thank the referee for bringing this to our attention. We agree with your point, and your suggestions are very helpful to improve the quality and scientificity of our study. It is highlighted that  $f(RH)$  differed significantly between NPF and Non-NPF days in Xiamen, primarily driven by the aerosol chemical compositions, particularly nitrate and sulfate. Sulfate was a major contributor to SNA on NPF days, distinguishing them from non-NPF days. We have updated the contents as suggested in Line 23-25 and Line 619-622.

**“In the relatively clean atmosphere of Xiamen, NPF events occur frequently, and the variation in chemical composition during the events has a substantial influence on the aerosol scattering hygroscopic growth.”**

“Sulfate was highlighted as the dominance in SNA during NPF days, with weaker  $f(\text{RH})$  compared to non-NPF days. It is likely that the condensation mode reactions occurring simultaneously with NPF events changed the aerosol chemical composition and had an obvious effect on  $f(\text{RH})$ .”

**2. In the abstract, authors state that “main source (30.78%) of the hygroscopicity parameter  $\kappa f(\text{RH})$ ”**

**$\kappa f(\text{RH})$  is not a parameter with absolute amount, therefore, authors cannot say their source, only could state what its variations were driven by what kind of aerosol component. For example,  $\text{NH}_4\text{NO}_3$  played a dominant role in  $\kappa f(\text{RH})$  changes/enhancement. Your statement about influences of organic aerosol is correct.**

*Response:* Thank you very much for kindly reminding us to clarify this point. For the aerosol hygroscopicity during NPF days,  $\text{NH}_4\text{HSO}_4$  (ABS) was the most dominant contributor, as revealed by the aerosol hygroscopicity–chemical composition closure. We agree with the comment and revised the sentence as follows.

“Aerosol hygroscopicity-chemical composition closure demonstrated that  $\text{NH}_4\text{HSO}_4$  was the main driving force (30.78%) of the hygroscopicity parameter  $\kappa_{f(\text{RH})}$  when NPF events happened, while  $\text{NH}_4\text{NO}_3$  played a dominant role in  $\kappa_{f(\text{RH})}$  (up to 35%) for non-NPF days.”

**3. Sect 3.5, OA was not attributed as POA and SOA factors, discussions on OA density should be included.**

*Response:* Thanks for the referee’s valuable suggestion. We have divided the OA factors into primary organic aerosol (POA) and secondary organic aerosol (SOA) factors by the PMF/ME-2 method. The proportion of POA and SOA in OA are displayed in Figure S6. The discussion of POA and SOA with  $\kappa_{\text{OA}}$  is shown in Line 562-571. We apologize for the omission of OA density, and the revisions are shown in Text S3.

“The proportion of POA and SOA in OA in our study (Figure S10) showed higher SOA mass fractions on non-NPF days than on NPF days. It is evident that high SOA mass fractions on non-NPF days corresponded to high  $\kappa_{\text{OA}}$  values. The results from these previous studies (Wu et al., 2016; Kuang et al., 2020; Kuang et al., 2021; Chang et al., 2010) also highlighted that SOA and oxygenated organic aerosols, were likely to be the determinants of  $\kappa_{\text{OA}}$ . Furthermore, SOA dominated OA mass both in this study and previous studies; however,  $\kappa_{\text{OA}}$  values differed much across studies. Noted that the hygroscopicity of SOA might vary significantly under different emission and atmospheric conditions due to variations in VOC precursors and SOA formation pathways (Kuang et al., 2021).”

“The volume concentration of BC was calculated by assuming a density of  $1.7 \text{ g cm}^{-3}$ , and the volume concentration of OA was calculated by assuming that the density of POA is  $1 \text{ g cm}^{-3}$  and density of SOA is  $1.4 \text{ g cm}^{-3}$  (Wu et al., 2016).”

**Specific comments:**

**1. L21, bracket of  $f(\text{RH})$  can be deleted**

*Response:* We thank the referee to point out this. We have deleted it.

**2. L22, describe the aerosol indirectly hygroscopicity**

*Response:* We thank the referee. We have corrected it.

**3. L22, varies greatly due to the influence of aerosol chemical composition and size**

*Response:* Thanks, we have corrected it.

**4. L91-93 Zhao et al. (2019) have summarized this in China**

Zhao, C., Yu, Y., Kuang, Y., Tao, J., and Zhao, G.: Recent Progress of Aerosol Light-scattering Enhancement Factor Studies in China, *Advances in Atmospheric Sciences*, 36, 1015-1026, 10.1007/s00376-019-8248-1, 2019.

*Response:* Thanks. As suggested, we have added this literature citation.

**5. L97-98 “between  $f(RH)$  and NPF events”, as stated in the major comments, NPF itself does not affect, however the unique condensation mode chemical reactions during NPF events could affect.**

*Response:* We thank the referee to point out this. We have modified the contents as follows.

“However, the exploration to the variation of  $f(RH)$  during NPF days is quite few in China.”

**6. L132 Nephelometers**

*Response:* Thanks, we have corrected it.

**7. L136, this is weird, you already have a “dry” nephelometer in the  $fRH$  system**

*Response:* Thanks. This separate nephelometer measured the aerosol scattering coefficient under ambient humidity conditions.

**8. L146, which type of ACSM, Q-ACSM, ToF-ACSM?**

*Response:* We thank the referee. It is a Q-ACSM. We have added this information in Line 154-157.

**9. L147, how BC mass concentrations were corrected, because AE31 is not as AE33 which have auto correction for loading effect?**

*Response:* Thanks. To ensure the data accuracy of the AE-31 aethalometer, we perform a zero-point calibration every two months. During calibration, a filter is connected to the sampling inlet of the instrument and the BC concentration reading is observed on the instrument panel; a concentration of approximately 0 is normal. Zero-point calibration is conducted after a new filter band is changed and typically takes 1-2 hours. In addition, the aethalometer is regularly returned to the manufacturer for system calibration.

**10. L298, SO<sub>4</sub>2- etc, use SO<sub>4</sub> is fine, or sulfate, because the ACSM doe not measure**

**the ion form of sulfate**

*Response:* We thank the referee to point out this. We have corrected it.

**11. L328, fewer hygroscopic particles were born is not correct, because NPF itself does not affect fRH, as you can see from Fig.3 that fRH is lower before NPF event, therefore, it is the background aerosols during NPF events that have lower hygroscopicity**

*Response:* We thank the referee for this useful suggestion, and we have corrected the contents as follows.

“This illuminates that aerosols had a lower hygroscopicity during the NPF event in Xiamen.”

**12. L 332-336, this is speculative, however, authors could state that “On the basis of aerosol chemical compositions during NPF, it could be speculated that .....”**

*Response:* Thank you for this helpful comment. We have made the revisions as follows.

“On the basis of aerosol chemical compositions during NPF, it could be speculated that when particle formation occurs in NPF days, the condensation of large quantities of sulfuric acid and organic vapours onto the pre-existing particles, resulting the conversion of mixed state on the surface of particles from external mixture to internal mixture and alteration of the optical and chemical properties of particles, which in turn might change the aerosol scattering hygroscopicity growth (Wu et al., 2016).”

**13. L356-357, The ACSM measures bulk aerosol compositions, if authors calculated the volume size distribution from PNSD measurements, it could be found that newly formed particle rarely affect bulk ACSM mass, meaning that the different bulk chemical compositions between NPF and non-NPF days could not attributed to formation mechanisms of new particles, it could only be attributed to different background aerosol when NPF occur, and reactions happened on existing condensation mode particles. This sentence could be rewritten as “indicating the remarkably different bulk aerosol compositions and condensation mode aerosol formation mechanisms between NPF and non-NPF days”**

*Response:* Thank you for this constructive comment. We have improved the content as follows.

“Sulfate dominated the SNA during the NPF days, characterized by weaker aerosol hygroscopic growth compared to non-NPF days, indicating the remarkably different bulk aerosol compositions and condensation mode of aerosol formation mechanisms between NPF and non-NPF days.”

**14. L422-425, the difference of organic and inorganic aerosol fraction is the reason, not sulfate. Indeed, kappa of ammonium sulfate is slightly lower than that of ammonium nitrate, however, not the major cause.**

*Response:* We thank the reviewer for this useful suggestion, and we have made the revisions as follows.

“The relative low value of  $f(80\%)$  for NPF days can be explained by the fact that the

organic and inorganic aerosol fractions were distinct, with sulfate being the predominant component of the inorganic aerosol during this period.”