

Response to Reviewer #1

Review of “Measurement report: Dust impact on hygroscopicity and volatility of submicron aerosols: Based on the observation in April of Beijing” by Hu et al. This manuscript investigates the hygroscopicity and volatility of submicron aerosols during a one-month campaign in urban Beijing. The authors present temporal variability in relation to meteorological parameters and $\text{PM}_{2.5}/\text{PM}_{10}$ mass concentrations, provide a statistical overview of the campaign, analyze air-mass back trajectories, and examine the impact of air mass on aerosol properties. Particular attention is given to a short dust event, with a comparison of aerosol hygroscopicity and volatility before, during, and after the episode. The topic is relevant and of potential interest to the community, particularly with respect to understanding the role of dust in influencing aerosol hygroscopic properties. However, I find that the current manuscript has several limitations that, in its present form, raise concerns regarding its suitability for publication in ACP. Below I provide detailed comments and suggestions for improvement:

1. Overall approach and novelty

The manuscript is largely descriptive, particularly in Sections 3.1 and 3.2. The discussion would benefit from a stronger connection to existing literature, as the current version does not sufficiently highlight what is novel compared to earlier studies from the region. I encourage the authors to clarify the new insights gained from this dataset and to better emphasize the scientific significance of their results.

Reply: Thanks for reviewer’s suggestion. To address your concern, we strengthened the discussion in the manuscript, especially in Sections 3.1 and 3.2, and we reorganized the manuscript to clarify the new insights and the scientific significance of the results in the manuscript.

2. Scope and dataset

The dataset, consisting of one month of HV-TDMA measurements without complementary observations of aerosol chemical composition or other physical/optical properties, appears rather limited. As presented, the analysis remains descriptive, which may not meet the standards expected for ACP. The authors may consider expanding the

contextualization of their results within the broader body of work on aerosol hygroscopicity and volatility to strengthen the manuscript.

Reply: Thanks for your comments. In order to improve the manuscript, we have added a discussion on the optical properties of aerosols in the case study. We added a comparative discussion regarding the research results of this study and those of previous studies. We have revised the manuscript carefully according to reviewers' suggestions and comments, hopefully, it is suitable to the scope of the ACP.

3. Title and focus on dust event

The title suggests a comprehensive assessment of dust impacts; however, the analysis relies on a single, short-lived dust event (approximately 6 hours). Drawing broad conclusions on dust impacts from such limited data seems overly ambitious. A more cautious framing of the study in the title and conclusions would be appropriate. Ideally, a longer dataset including multiple dust events would allow for more robust evaluation of dust effects on aerosol properties.

Reply: Thanks for reviewer's suggestion. We agree that a longer dataset including multiple dust events is essential for drawing definitive conclusions on dust impacts. Unfortunately, there was only one dust events during the study period. To address this issue, we have modified the manuscript title, reorganized the content, and explicitly noted the limitation of the short-duration dataset in the conclusion. Besides, we have also discussed the hygroscopicity enhancement factor based on aerosol scattering coefficient measurements to enhance the assessment of dust impacts. **Please see more details in Line 475-497 in the revised manuscript.** We plan to conduct long-term measurements to obtain more data on dust events in the future research to validate and extend the current findings, which will enable a more robust evaluation of dust effects on aerosol properties as recommended. We modified the sentence as:

“This study enhances understanding aerosol mixing state and evolution under different conditions and offer reliable observational constraints for reducing discrepancies between simulation results and observational data. Although only one dust event was captured, the results offer valuable insights into the impact of dust on accumulation mode particles. In the future, more observations including aerosol hygroscopicity, volatility and

chemical composition are needed during dust period to better quantify the influence of dust on aerosol properties.” (Line 592-598 in the revised manuscript)

4. Characterization of the dust event

Additional supporting information would help establish the identification of this episode as a dust event. How exactly is the event defined? While changes in the fine/coarse ratio are a useful indicator, dust events (especially those associated with long-range transport) typically last longer than a few hours. It would strengthen the analysis to incorporate additional observational evidence (e.g., satellite data, lidar, or ground-based measurements) if available. I also note that the link provided to the Beijing Meteorological Observatory does not appear to work and should be updated or replaced.

Reply: Thanks for reviewer’s suggestion. We fully agree that additional supporting evidence is essential to robustly confirm the nature of dust event. To clearly define the haze pollution and dust event, we added a subsection in the Methods section as follow:

“In Beijing, haze pollution (hereinafter referred to as EP) and dust pollution (hereinafter referred to as DS) frequently occur in the spring (Liu et al., 2023). Episodes of EP are typically characterized by a significant increase in PM mass concentration, accompanied by a high $PM_{2.5}/PM_{10}$ ratio, whereas DS events are associated with a sudden rise in PM mass concentration but a low $PM_{2.5}/PM_{10}$ ratio (Wang et al., 2019a; Fu et al., 2010; Wang et al., 2015; Tong et al., 2017). In this study, EP was defined based on the Class II standard values of the Chinese Ambient Air Quality Standards, as well as the $PM_{2.5}/PM_{10}$ ratio. Specifically, an EP event was defined when the $PM_{2.5}$ mass concentration exceeding $75 \mu\text{g}/\text{m}^3$ and the ratio of $PM_{2.5}$ and PM_{10} was greater than 0.3. During the study period, a total of six haze pollution (EP) episodes were identified. For DS events, the identification process involved two main steps. The first step identifying the DS event was to determine the particle mass concentration. We took the PM_{10} mass concentration was larger than $100 \mu\text{g}/\text{m}^3$ and the ratio of $PM_{2.5}$ and PM_{10} was smaller than 0.3 as the threshold to select possible DS events (Wang et al., 2019a; Wang et al., 2015). Second, the Atmospheric Environmental Meteorological Bulletin 2024 was consulted to confirm the occurrence of reported dust events, including their primary affected areas. Based on this combined approach, one DS event that occurred in Beijing in April 2024 was identified.” (Line 223-241 in the revised

Besides, we added the discussion about the variation of aerosol optical properties to explore the dust event.

“To further discuss the changes in aerosol properties during dust period, we analyzed the variations in aerosol optical properties. Figure 9 shows the time series of scattering coefficient, scattering Angstrom exponent, aerosol scattering hygroscopic growth factor for PM_1 and PM_{10} on April 15, 2024. The mean values of σ_{sp} for PM_1 and PM_{10} were 100.9 and 259.8 Mm^{-1} during the dust period. The averages of SAE for PM_1 and PM_{10} decreased sharply from 1.69 and 1.27 before dust period to 0.02 and -0.16 during the dust period, suggested a significant shift in aerosol size distribution toward coarse particle dominance (Hu et al., 2021). The persistently low SAE for PM_1 and PM_{10} values throughout the dust episode suggested the larger particles were dominated. . During the dust period, SAE was comparable to previous results during the dust periods in Beijing and Nanjing (Song et al., 2023; Xia et al., 2019). The mean $f(80\%)$ for PM_1 and PM_{10} decreased from 1.62 and 1.60 before the dust to 1.03 and 1.02 during the dust period, suggested that the submicron and super micron aerosols are almost hydrophobic during the dust period. The $f(80\%)$ during the dust period was similar to the value that observed in Beijing dust period (Xia et al., 2019). Although the scattering coefficient for PM_1 and PM_{10} still remained a low level after the dust period, SAE and $f(80\%)$ gradually rose, implying that the fine particles from anthropogenic sources gradually became dominant.” (Line475-497 in the revised manuscript)

We have re-checked the link to the Beijing Meteorological Observatory thoroughly through multiple browsers (Chrome, Firefox, and Edge) and network environments, and confirmed that it is accessible. The link website screenshot as follow:



Recommendations for improvement

Rather than centering the study on the short dust episode, I recommend the authors consider broadening the scope of their analysis to other features of the campaign dataset. For example, the six identified pollution episodes may provide a stronger basis for discussion and allow for more meaningful conclusions regarding aerosol hygroscopicity and volatility. Additionally, engaging more deeply with the existing literature would improve the scientific context and highlight the added value of this dataset.

Reply: Thank you for your constructive suggestions. We have restructured the manuscript and investigated of aerosol hygroscopicity and volatility under different environment and compared our results with previous studies. Please see more details in section 3.4 (Line 407-453 in the revised manuscript). We also showed as below:

“During the study period, significant differences were observed in the hygroscopicity and volatility of aerosols under different conditions. As shown in Figure 7, with increasing particle size, the dominance of the more hygroscopic mode became more pronounced under haze pollution conditions, implying that the larger size particles exhibited higher hygroscopicity under haze pollution conditions. The number fraction of NF_{MH} at 200 and 300 nm was close to 1, indicating that the particles at 200 and 300 nm were almost internally mixed and highly aged (Wang et al., 2019b). In terms of volatility, the VV mode particles were predominant for 50-300 nm particles, with the number fraction of VV mode particles exceeding 0.9 regardless of particle size.

NF_{vv} for both 50 nm and 300 nm particles was close to 1 under haze pollution conditions, whereas NF_{MH} for 50 nm and 300 nm particles was 0.39 and 0.94, respectively. These distinct hygroscopic and volatile characteristics of 50 nm and 300 nm particles under haze conditions may be associated with differences in their sources and aging processes. Previous study found that the dominant sources of 40 nm particles under polluted conditions were emissions from cooking and traffic (Wang et al., 2019b). Freshly emitted organic aerosols from vehicles are characterized by low-oxidation state, high volatility, and low hygroscopicity (Tiitta et al., 2010; Feng et al., 2023). A study conducted in Beijing reported that cooking organic aerosols (COA) plays a critical role in modifying aerosol hygroscopicity, with higher COA fractions leading to decreased hygroscopicity of organic aerosols (Liu et al., 2021). Additionally, heterogeneous reactions (e.g., aqueous-phase reactions) have been demonstrated to considerably affect the hygroscopicity of accumulation mode particles during haze episodes (Wang et al., 2019b). During haze pollution periods, reduced solar radiation and persistently high relative humidity promote the formation of secondary inorganic components through liquid-phase and heterogeneous reactions, accelerating particle aging in polluted environments (Sun et al., 2016). As a result, internally mixed accumulation mode particles become abundant and exhibit high hygroscopicity and volatility during haze pollution (Wu et al., 2016; Wang et al., 2019b). The HGF increased significantly with particle size under haze conditions, consistent with previous findings (Zhang et al., 2023a), likely due to the higher mass fraction of secondary inorganic aerosols in larger particles (Wang et al., 2018a). VSF did not show a markable size dependency, which is similar to the result under the pollution period observed by Wang et al. (2017) during haze episodes.

During the dust period, NF_{MH} initially increased with particle size but subsequently declined, while NF_{vv} gradually decreased. These suggested that a high proportion of particles with low hygroscopicity and low volatility at 200 and 300 nm. Both HGF and VSF displayed distinct size-dependent behavior during dust events compared to haze pollution conditions. During the dust period, HGF reached a peak at 150nm and then declined as the particle size increased. The VSF for 50nm was the least, and increased with the increase of particle size from 150-300 nm, reaching as high as 0.74 at 300nm during the dust period. More discussion on aerosol hygroscopicity and volatility during the dust period will be given in the following section.”

Besides, we conducted a review of the existing studies on the hygroscopicity and

volatility of aerosols as reviewer's suggestion and modified the introduction as follow:

“Hygroscopicity and volatility are critical physical properties of atmospheric aerosol particles. Hygroscopicity has a significant influence on atmospheric radiative balance and visibility by altering particle size distribution and optical properties. In addition, hygroscopicity indirectly affects the regional and global climate by influencing the lifetime and microphysical properties of clouds (Gunthe et al., 2009; Pöhlker et al., 2023; Rose et al., 2010). Moreover, hygroscopicity plays a key role in particle deposition by changing particle size in the human respiratory tract (Yu et al., 2025a). Volatility plays a crucial role in gas-particle partitioning, the formation and aging process of aerosols (Huffman et al., 2008; Xu et al., 2019). Considering the hygroscopicity and volatility of aerosols in the model is of great significance for reducing discrepancies between simulation results and observational data, and improving the accuracy of model outputs (Gao et al., 2024; Mcfiggans et al., 2006; Pringle et al., 2010; Rissler et al., 2010). Besides, determining the variation of particle size at selected dry diameters under different relative humidity and temperatures can also provide valuable indirect in-situ information regarding the chemical composition, mixing state, and coating properties of aerosols (Chen et al., 2022a; Liu et al., 2025; Massoli et al., 2010).

Hygroscopicity of aerosols have been investigated using several different instruments and techniques. The humidity tandem differential mobility analyzer (H-TDMA), which can provide information on the hygroscopic growth probability distribution of submicron aerosols, is widely used to measure aerosol hygroscopicity worldwide (Coe et al., 2007; Tang et al., 2019). In China, size-resolved aerosol hygroscopicity measurements have been carried out extensively in the North China Plain, Yangtze River Delta and Pearl River Delta. These researches focus on the seasonal variation of aerosol hygroscopicity (Zhang et al., 2023b; Wang et al., 2018b), the characteristics of hygroscopicity under different environment (Chen et al., 2022b; Wang et al., 2017), the impact of aerosol chemical composition and aging processes on aerosol hygroscopicity (Fan et al., 2020; Shi et al., 2022; Zhang et al., 2023a) and the evolution of hygroscopic behavior of atmospheric aerosols during heavy pollution episodes and new particle formation period (Wang et al., 2019b; Wu et al., 2017; Wu et al., 2016). Volatility tandem differential mobility analyzer (V-TDMA) is one of online instruments with high time resolution to measure the aerosol volatility. Multiple studies have been conducted in China using V-TDMA (Wang et al., 2017; Chen et al., 2022a; Wu et al., 2017) and inversed aerosol mixing state based on V-TDMA data (Chen et al.,

2020).

Aerosol hygroscopicity and volatility are correlated with chemical composition of the particles. Simultaneous measurements on aerosol hygroscopicity and volatility can provide new insight about the aging mechanisms of aerosols under different environment and the relationship of hygroscopicity and volatility. The VH-TDMA system was first proposed by Johnson et al. (2004), combining V-TDMA with H-TDMA to obtain both hygroscopicity and volatility simultaneously. Although aerosol hygroscopicity or volatility have been widely investigated worldwide, the simultaneous study of hygroscopicity and volatility in China is still limited (Cai et al., 2017; Kim et al., 2011; Wang et al., 2017; Yu et al., 2025a; Zhang et al., 2016). The results in the rural Pearl River Delta area reported that the photochemically-produced ultrafine particles to consist primarily of non-volatile and hygroscopic (NV-H) particles with a little volatile and non-hygroscopic (V-NH) particles and volatile and hygroscopic (V-H) particles (Kim et al., 2011). Zhang et al. (2016) found that certain fraction of hydrophobic particles is volatile in a rural site of the North China Plain. Wang et al. (2017) demonstrated that a higher number fraction of hydrophobic and volatile particles during the emission control period. The results observed by Yu et al. (2025b) showed that a positive correlation was identified between the number fraction of nearly hydrophobic and non-volatile particles during both the clean and the pollution periods.

Dust particles, suspended in the atmosphere, range from less than 0.1 μm to over 100 μm (Adebiyi et al., 2023). As one of the most important natural aerosols in the atmosphere, dust aerosols significantly affect atmospheric chemistry, human health, climate change, and biogeochemical cycles (Chen et al., 2021; Kurai et al., 2014). Heterogeneous reactions between mineral dust and trace gases can alter the chemical and physical properties of aerosols (Tang et al., 2017; Xu et al., 2020; Kok et al., 2023). Schladitz et al. (2011) demonstrated that the influence of dust particles was observed down to 300 nm during the Saharan Mineral Dust Experiment (SAMUM). Previous studies revealed that the changes of submicron aerosol effective density and optical properties during the dust period (Lu et al., 2024; Xia et al., 2019). Lu et al. (2024) found that the effective densities of 150, 250, 350, 450 nm under dusty conditions were higher than those during non-dusty periods, which reflected the dust influence on accumulation mode particles. Although the climatic and environmental effects of dust are considerable, limited studies focus on the dust effect on aerosol hygroscopicity and volatility simultaneously, especially on submicron aerosols (Kaaden et al., 2009; Kim and Park, 2012; Massling et al., 2007; Schladitz et al., 2011; Schladitz et al., 2009).

The North China Plain (NCP) is a hot spot of anthropogenic emissions, which can lead to severe air pollution. In recent years, the air quality in the NCP has significantly improved due to the strict control measures implemented by the government. However, the air pollution still happens due to unfavorable meteorological conditions, particularly in spring (Hu et al., 2021; Zhong et al., 2021). On the other hand, dust events often occur in the NCP in spring (Gui et al., 2023; Gui et al., 2022), which complicates the characteristics of aerosol properties in the NCP (Pan et al., 2009). Thus, it is necessary to enhance the comprehensive understanding of aerosol hygroscopicity and volatility in spring, particularly under different pollution conditions.

In this study, the hygroscopicity and volatility of aerosols were measured simultaneously using a Volatility-Hygroscopicity Tandem Differential Mobility Analyzer (VH-TDMA) in the spring of 2024. The characteristics of aerosol hygroscopicity and volatility were characterized, and the influence of air mass on aerosol hygroscopicity and volatility was discussed. Moreover, the relationships between hygroscopicity and volatility were explored. Besides, Aerosol hygroscopicity and volatility under different pollution environments were analyzed. Finally, the evolution of aerosol hygroscopicity, volatility and optical properties was investigated through a case study of a dust event. The study aimed to enhance understanding aerosol mixing state and evolution under different conditions and provide reliable observational constraints for reducing discrepancies between simulation results and observational data.” (Line 41-130 in the revised manuscript)

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