Response to Reviewer #3

The manuscript describes an intense measurement period in spring in the North China Plateau (NCP) focusing on sub-micrometre hygroscopicity and volatility measurements. They performed Hysplit back-trajectory analysis and in combination with PM10 and PM2.5 values, determined a special period with most likely dust aerosols from the Gobi desert. The publication is meant as a measurement report rather than a full research article. Generally, characterizing aerosols' water uptake ability and volatility is very important to better constrain model parametrizations. To my mind the article lacks some more discussion regarding what has previously been measured in the NCP and puts very much focus on a single dust event that occurred during the measurement period. I also think, it would be helpful to put the dust period in perspective to other dust events when hygroscopicity and volatility were measured, even outside of the NCP. Furthermore, the article needs a major revision regarding English spelling and structure.

Reply: Thanks for reviewer's constructive suggestions and comments. To strengthen the contextualization of our work within existing literature, we modified Introduction section to synthesize previous measurements of aerosol hygroscopicity and volatility in the China and clarified the importance of this researches. Please see more details in Line 40-130, also shown below:

"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., 2018), 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., 2019; 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 0.1μm to over 100μm (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."*

As reviewer's suggestion, we added the comparison between our findings during the dust period with previous studies as follow:

"*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."* (Line 484-497 in the revised manuscript)

"During the dust period, the proportion of more hygroscopic mode particles decreased rapidly. When the dust arrived, the strong north wind not only brought dust particles but also swept pre-existing fine particulate matter in the atmosphere. The most significant decline in the MH mode number fraction occurred at 200 nm and 300 nm, with minimum values of 0.20 and 0.32, respectively. The mean MH mode number fractions for 200 and 300nm were 0.54 and 0.33, which were much lower than those before the dust period. Previous research also found that the number fractions of MH mode particles for 250 and 350 nm were less than 0.5 during the dust event observed in Tinfou, Morocco (Kaaden et al., 2009). Massling et al. (2007) found that the number fraction of nearly hydrophobic particle from dust particles was approximately 64%, which was consistent with our observation. In terms of volatility, the NV mode of 200

and 300 nm became more prominent during dust period. The mean values of the number fraction of VV mode for 200 and 300 nm particles decreased from 0.93 and 0.94 before the dust period to 0.73 and 0.47 during the dust period, respectively. The minimum NFvv at 300 nm was only 0.18, indicating that approximately 82% of particles with diameter of 300 nm were non-volatile. High number fraction of non-volatile components and nearly hydrophobic particles at 200 and 300 nm during the dust period suggested dust particles can be as small as 200 nm in diameter (Kaaden et al., 2009). The mean HGF for 300 nm particles during the dust period was 1.20, which was close to the observed results of Massling et al. (2007) during the dust period. Besides, low hygroscopicity of aerosol during dust storm was also observed by Shen et al. (2023). VSF increased evidently to the maximum value of 0.89 at 300 nm during the dust period." (Line 511-532 in the revised manuscript)

Besides, we have thoroughly revised the manuscript with attention to grammatical accuracy and reorganized the manuscript thoroughly.

My major concerns are the following:

- The motivation of the study is lacking. The authors' refer to previous publications presenting the aerosols' properties, including hygroscopicity and volatility, in the same area and state that those previous studies related their results to chemical composition. As a major new thing, they state the single dust event. I cannot clearly see how this dataset differs from the previous ones. The authors state that these are the first hygroscopicity and volatility measurements in NCP during a dust event. After a quick literature research, I found several papers discussing dust events in NCP, which are though only partly mentioned in the paper under review. I think it would be very interesting to relate the new data to previous events and for example compare PM values and meteorological conditions and potentially relate the chemical composition measured previously with the hygroscopic growth found in this study. On the other hand, I believe that the dataset also offers other interesting discussion that could be deepened.

Reply: Thanks for reviewer's suggestions. we have revised the introduction thoroughly and added a paragraph in the revised manuscript to explicitly clarify the motivation of study. Aerosol properties are rather complex in the Beijing of Spring because of the mixture of different sources from anthropogenic emissions and dust while previous studies have characterized aerosol hygroscopicity and volatility, limited researches focused on aerosol hygroscopicity and volatility under different conditions simultaneously. Besides, the understanding of dust effect on submicron aerosol remains inadequately understood. We have restructured the manuscript and enhanced the comparison and discussion with the results of previous studies. Moreover, We added the summary of meteorological parameters under different pollution conditions in the supplement (Table. S1) and linked the PM ratio and key meteorological parameters (relative humidity, wind speed, wind direction) from the current study with previous studies as follow:

"*There were six haze pollution events and one dust pollution event during the study period. The haze pollution events were characterized by high $PM_{2.5}$ mass loadings and high $PM_{2.5}$ and PM_{10} ratio (0.46-0.72), which was similar to the results observed in Beijing (Liang et al., 2022). These six haze pollution events were strongly associated with the stagnant meteorological conditions as indicated by the prevailing southerly winds, high RH (>60%), and low wind speed (<1.3m/s) (Table S1). Previous studies in Beijing have shown that aerosol pollution in Beijing possibly contributed by pollutants transported from the south of Beijing (Wang et al., 2013; Yin et al., 2025). Furthermore, low wind speeds favored pollutant accumulation and high relative humidity would further enhance aerosol hygroscopic growth and accelerate liquid-phase and heterogeneous reactions (Zhong et al., 2018). In contrast, during the dust period, the average $PM_{2.5}$ mass concentration was only about half of that observed during haze episodes, whereas the PM_{10} mass loading was approximately twice as high. Besides, low $PM_{2.5}$ and PM_{10} ratio (0.16) was observed during dust period, which was comparable with that observed in Beijing dust period (Lu et al., 2024; Xia et al., 2019; Liang et al., 2022). The dust event was also associated with prevailing northerly winds, low RH (17%), and higher wind speeds (3.5 m/s) (Table S1), consistent with the typical meteorological conditions observed during dust period in previous studies (Lu et al., 2024).*" (Line 254-272 in the revised manuscript)

Table S1. Summary of meteorological parameters under different pollution conditions

	T(℃)	RH (%)	Wind speed (m/s)	$PM_{2.5}$ (μg/m^3)	PM_{10} (μg/m^3)	$PM_{2.5}/PM_{10}$
EP1	14.2±5.2	63±19	1.03±1	92.2±16.5	166.2±37.6	0.56±0.04
EP2	17.5±4.1	66±15	1.26±0.7	128.8±38.4	181.4±60.3	0.72±0.06
DS	18.7±2.1	17±4	3.5±0.6	49.3±19.9	297.1±107	0.16±0.02
EP3	15.4±5.2	62±20	1.15±1.01	86.5±6.6	158.9±8.8	0.54±0.03

EP4	19.5±2.9	67±20	1.16±0.39	81.9±5.5	134.5±6.3	0.61±0.02
EP5	16.1±3.4	76±15	1.06±0.55	81.6±5.1	147.6±8.8	0.55±0.04
EP6	19.6±3.2	80±12	0.85±0.57	86.3±12.2	189.9±28.9	0.46±0.04

Besides, we compared our observational results with previous studies and analyzed the differences in measurements between the two regions based on chemical composition discrepancy in different region. Please see more details in Line 286-295 and 318-327, also shown below:

"During the whole period, the number fraction of more hygroscopic mode particles (NF_{MH}) for 50-300 nm particles was usually larger than 0.50, especially at large size, suggested that more hygroscopic mode particles were dominated. While the results observed in Gucheng, an rural site in the North China Plain, showed that the number fraction of NH mode became more prominent with increasing particle size (Shi et al., 2022). The discrepancy in the size dependency of NF_{MH} in Beijing and Gucheng is likely due to differences in chemical composition. Organics constituted a major fraction of PM_1 at Gucheng (Shi et al., 2022) , whereas secondary inorganic aerosols with high hygroscopicity made the largest contribution to PM_1 in Beijing (Lei et al., 2021)."

"During the sampling period, the mean number fraction of very volatile mode particles (NF_{VV}) for 50, 80, 110, 150, 200 and 300 nm particles was 0.96, 0.95, 0.94, 0.93, 0.91, 0.87, respectively, which were much higher than those measured at Xianghe, a rural site in the North China Plain (Zhang et al., 2016). In other words, the mean number fraction of non-volatile mode particles in Beijing was less than10%, which was lower than the observation results in Guangzhou (Cai et al., 2017) and Shanghai (Jiang et al., 2018). The high NF_{VV} indicated that the majority of particles in Beijing were highly volatile, likely attributable to a lower proportion of black carbon and a higher proportion of secondary inorganic aerosols, especially nitrate (Lei et al., 2021; Sun et al., 2022)."

I believe that the title is misleading as dust is only a very short period during the whole measurement period.

Reply: Thanks for reviewer's suggestion. We have changed the title of this manuscript to "Measurement report: Characteristics of hygroscopicity and volatility of submicron aerosols under different pollution environment in Spring".

- When discussing the differences in measured properties as a function of particle diameter, the authors make a differentiation between organic particles and "secondary particles" that are however, not specified in their composition. From the citation it looks to me like they are referring to "secondary organic aerosols". If this is so, I cannot understand the explanation for the differences between Aitken and accumulation mode particles. The observed differences with size are very interesting but are lacking further investigation.

Reply: We are sorry that we did not express this clearly. We have revised the manuscript as follow:

"During the whole period, the number fraction of more hygroscopic mode particles (NF_{MH}) for 50-300 nm particles was usually larger than 0.50, especially at large size, suggested that more hygroscopic mode particles were dominated. While the results observed in Gucheng, an rural site in the North China Plain, showed that the number fraction of NH mode became more prominent with increasing particle size (Shi et al., 2022). The discrepancy in the size dependency of NF_{MH} in Beijing and Gucheng is likely due to differences in chemical composition. Organics constituted a major fraction of PM_1 at Gucheng (Shi et al., 2022) , whereas secondary inorganic aerosols with high hygroscopicity made the largest contribution to PM_1 in Beijing (Lei et al., 2021)." (Line 286-295 in the revised manuscript)

"*This distinct relationship between hygroscopicity and volatility of accumulation mode particles, compared to Aitken mode particles, is likely associated with differences in aerosol chemical composition. Previous studies in Beijing revealed that the particles below 100nm are mainly organics (Li et al., 2023), while accumulation mode particles are dominated by secondary inorganic aerosols characterized by high hygroscopicity and high volatility (Xu et al., 2015).*" (Line 387-393 in the revised manuscript).

Overall I believe that this manuscript should only be considered for publication in ACP after major revisions.

Reply: Thanks for reviewer's suggestion. We have reorganized the manuscript thoroughly and have done our best to enhance manuscript's quality.

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