Report

Yu et al., describes an interesting study on the hygroscopicity and volatility of aerosols in Beijing urban areas during four weeks' field campaign using a Volatility Hygroscopicity Tandem Differential Mobility Analyzer (VH-TDMA) system, a twin scanning mobility particle sizer, a high-resolution time-of-flight aerosol mass spectrometer and other meteorological instruments. The volatile shrink factor (VSF) and the hygroscopic growth factor (HGF) are reported for aerosols having different sizes: 50 nm, 80nm, 110 nm and 150 nm. Furthermore, the authors try to display the size-resolved probability function on the HGF and VSF determined by two Scanning Mobility Particle sizers (SMPS) implicated in the VH-TDMA system. The bimodal distribution has been shown for both two probability functions. During the field campaign, one clean period and three pollution periods are identified. The authors report also the relation between the hygroscopicity and volatility for submicron aerosols. The back trajectories are also carried out for this observation site. According to the bimodal distribution of size-resolved HGF and VSF, the authors suggest that the presence of submicron aerosols in Beijing urban areas is probably under the external mixture due to different sources.

The manuscript brings a comparatively interesting report but the presentation way, the data analysis methodology and all results related to the multi-charge effect (including HGF-PDFs, VSF-PDFs ,kappa and relation between hygroscopicity and volatility) need to be carefully reviewed before the submission.

1st major comment: the presentation of the measurement campaign is insufficient. A general configuration of all instruments need to be well described, especially how the aerosols are sampled for different instruments. Only a short presentation has been done to indicate the measurement period and the sampling location. All instruments and data should be summarized in this section. However, several fragments could be found in the section of results and discussions, such as meteorological information, back trajectories analysis, etc. Furthermore, the instrumentational information on the three DMAs and two CPCs implicated in the VH-TDMA system is missing, i.e., the model, working conditions... The authors really need to re-organize this section.

2nd major comment: an important part of the conclusions of this manuscript is based on the bimodal distribution of size-resolved HGF and VSF. However, the phenomenon of multicharged effect on the incoming aerosols by the neutralizer is well known. The authors do not provide the size distribution of selected particles by the DMA1 but only suppose that selected particles are monodispersed. The double charged particles potentially correspond to the larger particles which could contribute to the second mode of HGF and VSF probability functions, i.e., it is not clear whether the second mode is contributed by the different hygroscopic or volatile compounds in the aerosols or is contributed to the double charged particles selected by the DMA (generally these double charged particles are larger than what is commanded in the DMA selection). According to the literature, the double charge effect influences importantly the hygroscopicity analysis of SOA (Bouzidi et al., 2022), combustion aerosols (Petters et al., 2009;

Wu et al., 2020), oxygenated aerosols (Petters et al., 2007) at the laboratory and also the measurement during the field campaign (Mochida et al., 2010). Kim et al., 2023 reported a influence of 20% on the k_{CCN}. Several studies suggest how to minimize the double charge effect on the CCN counter (Wang et al., 2015) and HTDMA (Barrett et al., 2012; Duplissy et al., 2009; Oxford et al., 2022). The correction of this multiple charge effect is also provided by Kim et al., 2023 and Petters, 2018. The authors need to take into account this effect in order to better illustrate the conclusions.

Comments in details:

Line 88, what is the reason that authors chose 270 °C for studying the hygroscopicity of non-volatility particles while this temperature is usually set as 300 °C in the literature?

Line 89, in the section "*experimental set-up*", the authors give a description of each instrument used in this study but no information about how aerosols are sampled among different instruments, that is extremely important to validate the results. For example, were these instruments connected to a common sampling inlet or separately? It is suggested to introduce globally the campaign at the very beginning of this section, including the sampling site, the instruments, sampling conditions and the sources of all meteorological data.

Line 97, can authors provide a reference which gives more detailed information on this VH-TDMA (TROPOS, Germany)? Otherwise, authors should provide them in this manuscript. For instance, the model of dryer is missing and at what RH aerosols enter in the neutralizer?

Line 98, what is the model of the DMA1 and in which conditions it works, for example the sheath flow rate? Same questions for the DMA2 and DMA3.

Line 98, what is the model of two CPCs and what size range they measure?

Line 151 and line 156, authors need to declare clearly how the *Dp (Troom, RHdry)* is defined for the data treatment in this manuscript.

Line 161, for having a size-resolved HGF-PDF, how the *Dp (Troom, RHdry)* is defined, the size that the DMA1 selected or a probability density distribution of "monodisperse" aerosols selected by the DMA1? In both case, the multi-charge effect is non-negligible. It is necessaire to provide the size distribution of aerosols selected by the DMA1 or to consider the size distribution of sampled aerosols from site to evaluate this multicharged effect (Duplissy et al., 2009; Kim et al., 2023).

Line 165, the same question as the previous. How the *Dp (Troom, RHdry)* is defined for having a VSF-PDF?

Line 165, the method how to provide the *VHGF-PDF* is missing. Since authors have observed the binormal distribution of the non-volatile core of aerosols after heated, how *Dp(270 ℃ ,RHdry)* is defined for obtaining your results (for example the VHGF-PDF in figure 11), using the first mode size or the second mode size or the probability density distribution?

Line 183, authors really need to take into account the multi-charge effect before calculating the *σHGF-PDF*.

Line 196, could authors provide the size distribution of aerosols measured by the PNSD to show the seven NPF events?

Line 209, authors need to take into account the multi-charge effect before giving the conclusion.

Line 216, in the case of non-consideration of multi-charge effect, the variation (number and mode size) of ambient aerosol size distribution due to clean and/or pollution events could contribute to the variations in the HGF-PDFs and VSF-PDFs. It is suggested that authors provide not only the mean particle size distribution during the clean period as Figure S4, but also that as function as time during the whole measurement campaign.

Line 240, "*narrower*" is not a quantitatively description. Coud authors provide the geometrical standard deviation of two modes for different cases in order to illustrate this conclusion?

Line 265, figure 3 shows the mean HGF-PDF and the mean VSF-PDF of particles having different sizes. In figure 3 a, how to explain the HGF of left part in the first mode (around ¼ visibly) is lower than 100%?

Line 268, the general presentation of the table 1 is missing. In the table 1, authors need to well define the "*Total*". Does "Total" means the whole period from 11 October to 6 November 2023? If yes, it is interesting that the mean k of 50 nm in "*Total*" is slightly higher than that in both "*Clean*" and "*Pollution*". Is it significant large in the period which is not "*Clean*" and "*Pollution*"?

Line 314, "*AMS results show that nitrate is the main inorganic component of PM1 (Fig. 8a), further supporting this viewpoint.*" It is known that nitrate signals in the AMS could represent inorganic nitrate compounds or organic nitrate compounds. Graeffe et al., 2023 reported how to estimate the organic nitrate. Authors should provide more information such as NO+/NO2+ ratio to support this conclusion.

Line 329, "*During the clean period, particle volatility increased dramatically around 10:00 LT (VSFmean decreased) along with the occurrence of NPF events, indicating that the newly formed matter was more volatile.*" Replace "*around*" by "starting from".

Line 348, "*Notably, external mixing was more apparent during the night and early morning, especially for 150 nm particles during the clean period and 350 50 nm particles during the pollution period (Fig. 5d and 5e). This phenomenon could be attributed to the reduced boundary layer height, which leads to the accumulation of nonvolatile particulate matter emissions (e.g. BC, soot aggregates) from cooking or vehicles emissions.*" Authors need to take into account the multi-charge effect which could contribute to the VSF-PDF the same way as the external mixing. It is suggested to combine off-line technics such as Transmission electron microscopy to have a direct prove on the presence of soot particles and/or sea salts mentioned later.

Line 467, it is suggested to present ZSR relation and the method of calculating $HGF_{coating}$ in the section of "*data analysis*".

Line 479, "*As shown in Fig. 8c, variations in HGFcoating basically similar to those hygroscopicity of unheated particles (HGFmean) (Fig.S7) and exhibit significant size dependency*" What is the difference between figure.S7 and figure.2 and figure.S2 on the part of HGF? What is the interest to show the same data by using size independent time series (figure.S7) and using size-resolved probability function time series (figure.2 and figure.S2)? What is the procedure authors calculate HGFcoating, by using HGFmean or HGF-PDF or some other method for the core and for the original aerosols?

Line 501, according to the figure 11, the distribution of core particles (after the heating of 270 °C) is not monodisperse. How authors calculate HGF-core using equation 3?

Line 522, it is the first time to see "VHGF-PDF". The explanation is required.

Line 535, the time scale with multi colors is not explained.

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