Response to Anonymous Referee #2

We thank the reviewer for the constructive suggestions and comments concerning our manuscript entitled "Measurement report: size-resolved particle effective density measured by the AAC-SMPS and implications for chemical composition" (ID: egusphere-2024-3298). Those comments are valuable and very helpful for improving our paper, as well as the important guiding significance to our studies. Below, we provide a point-by-point response to individual comment (Reviewer comments in italics, responses in plain font; page numbers refer to the ACPD version)

The current manuscript presents the characteristics of the ambient aerosol density of one-month online measurements in Hangzhou. The ms is generally written well. I have mainly questions for clarification.

[Comments1] *Do the SHAPs work for particles larger than 350 nm? If not, it deserves to be mentioned in the abstract.*

Responses and Revisions:

Thank you for the advice. According to the SHAP results, the correlation between the effective density and the chemical composition of particles with diameter larger than 350 nm was weak. The Line 17 in abstract has been revised as

"The SHapley additive explanations (SHAPs) revealed good relationships between ρ_{eff} and the bulk composition of particles with diameters smaller than 350 nm, while the relationship of larger particles was weak."

[Comments2] *Lines 33–34: Please specify the size range.*

Responses and Revisions:

Thank you for the advice. This sentence has been revised as

"It collects size-segregated aerosols with different size ranges and subsequently analyses their mass and chemical components, but the temporal resolution of this offline method is relatively low, and the size range is limited (0.056 – 18 μm, depending on size stages of used MOUDI)" (Line 34)

[Comments3] *The current system uses a dryer before the instruments. When RH is high enough, ambient particles may exist as aqueous droplets. Is there any effect of the particle phase state on density measurements?*

Responses and Revisions:

Previous studies have indicated that aerosol particles can absorb and release water when they undergo relative humidity (RH) cycles which can govern the liquid water content, composition, size and phase state (liquid, semisolid, or solid) of aerosol particles (Tan et al., 2024). Liquid water has a much lower density (approximately 1 g/cm^3) compared to solid particles like salts or organic compounds (which can range from 1.5 to 2.5 g/cm^3 depending on the material). The uptake of water can lead to a lower overall density of particles.

In order to avoid the uncertainty of changing RH, we think the dryer before the instrument is necessary. When the particles were introduced into the instrument, it is difficult to keep the RH inside the instrument same as the ambient RH. Different RH can affect the water content of aerosols. Drying the particles standardizes the measurement conditions, making it easier to compare aerosol properties across different sites, times, and studies. Without drying, the measurements could be influenced by varying humidity levels, introducing inconsistencies.

[Comments4] *Please define the sub-density mode.*

Responses and Revisions:

Thank you for the advice. The definition of sub-density has been elaborated in Line 136-140:

"The distribution of ρ_{eff} was fitted to a unimodal Gaussian distribution, whereas some previous studies reported a bimodal distribution, i.e., a mode with higher peak value and larger effective density denoted as main-density mode and another mode with lower peak value and lower effective density denoted as sub-density mode. The sub-density mode was associated with fresh emissions (Qiao et al., 2018; Zhou et al., 2022; Xie et al., 2024)"

[Comments5] *The relationship between density distribution and particle mixing state is unclear. Please elaborate on what the bimodal distribution means.*

Responses and Revisions:

Thank you for the advice. It has been added in Line 131:

"The unimodal distribution denoting an internally mixed aerosol composition and the bimodal distribution with a second, below unity density peak indicating externally mixed BC (Qiao et al., 2018; Ma et al., 2020; Zhou et al., 2022; Xie et al., 2024)."

[Comments6] *The authors conclude no significant diurnal variation in the density values. However, Figure 4a noted a gradual increase in the density for 531 nm particles, which deserves more discussion and comparisons with other studies.*

Responses and Revisions:

Thank you for the advice. The diurnal variation for larger particles has been added in Line 189.

"In addition, compared to small particles, the diurnal variation in effective density of large particles is less pronounced. The decreasing trend during 7:00 to 13:00 becomes less obvious (Fig. S4), primarily because larger particles have longer residence times in the atmosphere and are less influenced by fresh emissions, resulting in a more stable chemical composition (Zhai et al., 2017; Xie et al., 2024). Overall, the effective density of large particles shows a slight increase throughout the day. It would be related to the increasing of RH at night. which facilitates the formation of SIA which has been discussed before. However, compared with smaller particles, larger particles reduce surface reactions and adsorption capacity due to a lower specific surface area (Okuda, 2013). As a result, the ρ_{eff} of larger particles was less sensitive to changes in temperature and RH."

[Comments7] *In this study, only small particles showed a strong correlation between feature values and the prediction. (a) Please specify the feature values. (b) Does the weak correlation for big particles mean that the chemical composition is invariable compared to that of small particles?*

Responses and Revisions:

(a) Feature values estimate the significance of each feature within a model. This sentence has been revise as:

“The cross-validation was used to evaluate the model performance, and the results suggested that RF model was performing well for particles smaller than 350 nm, with the overall R^2 score being greater than 0.55 (Table S1).” (Line 261)

(b) Yes, the weak correlation suggested the relatively stable chemical composition. The inorganic salts dominate in these particles. Previous studies on size-resolved chemical composition of particles have shown that the proportion of inorganic salts increases with particle size (Zhang et al., 2005; Kim et al., 2020; Zhao et al., 2020). The effective density of inorganic salts is approximately 1.77 g/cm^3, causing the effective density of large particles to approach that of inorganic salts.

Minor comments:

[Comments8] ***Line 58:*** *Please rewrite “machine learning (ML) methods, such as ozone pollution and potential aerosol sources.” Ozone pollution and potential aerosol sources are not ML methods.*

Responses and Revisions:

Thank you for the advice. It has been revised as (Lines 58-59):
“Currently, machine learning (ML) methods, such as simulation of ozone concentration and reproduction of aerosol number concentration, are widely used in atmospheric science research”

[Comments9] ***Line 111:*** *Please define “RF.”*

Responses and Revisions:

Thank you for the advice. It has been added in introduction (Line 60):
“Random forest (RF) is a commonly-used machine learning algorithm. Compared to other ML models, such as deep learning models, it maintains a commendable balance between predictive performance and interpretability.”

[Comments10] ***Line 148:*** *Fig.2 -> Fig.3.*

Responses and Revisions:

Thank you for the advice. It has been revised.

Reference

Tan, F., Zhang, H., Xia, K., Jing, B., Li, X., Tong, S., and Ge, M.: Hygroscopic behavior and aerosol chemistry of atmospheric particles containing organic acids and inorganic salts, npj Clim Atmos Sci, 7, 203, https://doi.org/10.1038/s41612-024-00752-9, 2024.