

Dear Editor,

We thank for all the constructive comments and suggestions from referees. We have carefully addressed and provided detailed explanations for their concerns. Point-by-point responses to the suggestions, corresponding updates with the revised manuscript, and the finalized version have been uploaded.

In the following, original suggestions, our response, and updates on the revised manuscript are shown in **bold**, normal, and *italic*, respectively.

Kind Regards,

Jing Chen, Yuhang Hao, and Peizhao Li

Anonymous Referee #2

General comments:

I thank the authors for their efforts and for satisfactorily addressing my earlier comments and suggestions. Although I have indicated the need for minor revisions, the comment below may be considered a major one and must be addressed before the manuscript is accepted.

Response: We appreciate the reviewer for the supportive comments.

Specific comments:

RC1. Regarding Figure R4 in general (and specifically R4a3 and R4b3): Did the authors use the entire time period or only the NPF event time period (8 am –10 pm LT) to create the box-whisker plots? This appears to contradict Figures 4a and 4b, where NF_{accu} and VF_{accu} are clearly higher on non-event days than on NPF event days (for both P1 and P2). Does this suggest that NPF does not contribute to the accumulation mode (both number and volume) during either time period?

Response: We use the entire days' data (24-h) for the box-whisker plots of corresponding concentrations in Figure R4 in our previous response, while Figures 4a-b were the time-averaged diurnal variation results of both number and volume fractions for the accumulation mode particles. To avoid unnecessary misleading, the caption of Figure S7 has been adjusted into “*The number concentrations (left column: a1-a3) and volume concentrations (right column: b1-b3) of different mode particles for the corresponding NPF and non-event days during both P1 and P2 periods.*”.

Accumulation mode particles are suggested to originate predominantly from direct emissions or aging of pre-existing particles (e.g., local sources, regional transport), with NPF contributing marginally to the total number and volume concentrations of the accumulation mode (i.e., without significant increases during the event time periods as shown in Figure S8). Nevertheless, NPF events could significantly modulate variations in $NF_{Acc.}$ and $VF_{Acc.}$ (Figure 4a, b), mainly due to the

shift in the particle number size distribution driven by distinctly enhanced abundances of both nucleation and Aitken mode particles during NPF events. For instance, the discrepancies of $NF_{Acc.}$ and $VF_{Acc.}$ during the non-NPF time window (i.e., unshaded areas in Figures 4a- b) were different from that for the NPF event period in the P1 period. Namely, a higher level of $NF_{Acc.}$ ($VF_{Acc.}$) was observed during the non-NPF time window on the P1 $NPF_{polluted}$ days than on non-event days, yet both $NF_{Acc.}$ and $VF_{Acc.}$ decreased substantially upon NPF event onset. This not only signifies the importance of local accumulation and aging processes on the accumulation mode particles during non-event time window in relatively polluted P1 period, but also highlights the crucial role of NPF in regulating the size distribution pattern thereby the relative abundance of the accumulation mode particles during NPF events. The below Figure R1 shows the mean $NF_{Acc.}$ and $VF_{Acc.}$ results of $NPF_{polluted}$ days (the entire days) and $NPF_{polluted}$ event time periods (08:00-22:00 LT) during the P1 period, as well as that for the corresponding non-event cases. The mean $NF_{Acc.}$ and $VF_{Acc.}$ were quite similar on the whole $NPF_{polluted}$ and non-event days, whereas significantly lower (specifically for $NF_{Acc.}$) during $NPF_{polluted}$ event time periods than that either for the same period of non-event cases or the mean levels for whole days' data (Figure R1). It is noted that the $NF_{Acc.}$ and $VF_{Acc.}$ levels were even lower on the $NPF_{clean, HW}$ days than non-event days during P2 period (Figures 4a-b), possibly due to the rather cleaner environment which amplified the corresponding impacts of NPF on lowering the relative abundances of the accumulation mode.

Updated on the main text:

L499-503: *This is mainly due to the explosive formation of ultrafine particles and subsequent growth on NPF days, significantly altering aerosol size distributions and inducing large fluctuations in the $NF_{Acc.}$ and $VF_{Acc.}$ in comparison to that on non-event days, especially during the P2 period (as shaded in Fig. 4a-b).*

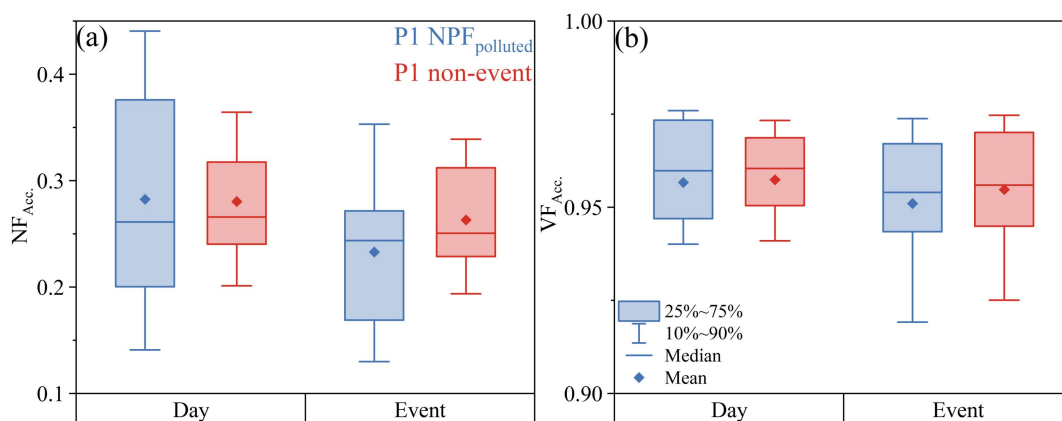


Figure R1. Box plots of $NF_{Acc.}$ (a) and $VF_{Acc.}$ (b) for both the whole days and just during the 08:00-22:00 LT time window in P1 period.

RC2. Additionally, the smallest aerosol effective radius is ~ 80 nm. What fraction of newly formed particles grows to this size and subsequently to the accumulation mode (during both P1 and P2), such that they can be associated with changes in aerosol hygroscopicity resulting from new particle formation?

Response: We're afraid that the newly formed particles could rarely grow into the accumulation mode (e.g., $D_p > 100$ nm) under dry conditions (Kerminen et al., 2018). As shown in Figure S6, the SMPS-determined mean R_{eff} decreased to around 60 nm (50 nm) after the occurrence of the P1 $NPF_{polluted}$ (P2 $NPF_{clean, HW}$) event. Figure R1 has further demonstrated decreases rather than increases in both $NF_{Acc.}$ and $VF_{Acc.}$ during NPF events, although the P1 $NPF_{polluted}$ events exhibited a relatively larger level of R_{eff} . Based on the growth rate (GR) and the duration of NPF events obtained from our observations, the maximum attainable sizes for newly formed particles during both periods can be accordingly estimated. Specifically, the newly formed particles can grow into a maximum dry size of 74.1 ± 17.0 nm during P1 $NPF_{polluted}$ events, while they can grow to 81.5 ± 10.3 nm due to prolonged subsequent growth till the evening during P2, despite of a relatively lower GR. Unless with a pronounced size growth upon aging or hydration (e.g., $D_p > 200$ nm), these maximally grown dry particles would not significantly influence the total light scattering by themselves, as evidenced by the size-resolved light scattering results in Figure S13. Nevertheless,

variations in optical and hygroscopic properties of the aerosol population due to the shift in aerosol size distributions (e.g., altered fractions of ultrafine and accumulation modes induced by NPF events along with the hygroscopic growth and aging processes) could be distinct and nonnegligible, as reflected in Figure S11a-b and Figure S16. In summary, it could be difficult to affect the aerosol optical hygroscopicity solely by the subsequent growth of newly formed particles due to insignificant light scattering efficiencies in the dry condition, yet the hydrated and aged new particles may modulate the aerosol optical and hygroscopic properties largely under specific alterations in the aerosol size distribution patterns.

Anonymous Referee #4

While I do think that this manuscript presents interesting data I also believe that the comments of both referees are well substantiated. My own impression very much conforms to referee 2 which might have major implications on the conclusions drawn.

Response: Thanks for the constructive comments and suggestions.

Specifically the missing nucleation mode particles during the heatwave indicate advection of newly formed particles rather than in-situ formation. This has been addressed in the revised manuscript for horizontal transport but does not take into account potential down mixing during boundary layer evolution. In any case, my concern is that whatever the reason for the lacking newly formed particles is one could equally argue in the other direction that extremely hot weather conditions could suppress NPF at this location. Under the conditions shown, formation and growth rates will simply not be comparable to other periods and will lead to wrong results/interpretation as the sudden appearance is not associated with growth and formation but with changing air mass. I think that the conclusions should be amended accordingly. Maybe also the title should reflect that these data were taken "...during a heat wave..." Furthermore, the text several times mentions "...during weather extremes (e.g., heat waves)..." which does imply also weather extremes beyond heat waves and is not justified in my opinion. I'm therefore ambivalent about this manuscript. I think it is worth publishing but it may still need some optimization in the wording and careful interpretation.

Response: Thanks for the comments. We acknowledge the potential impact of the vertical transport on NPF events (Crumeeyrolle et al., 2010; Lai et al., 2022a; Platis et al., 2016). Actually, the calculated cumulative contributions of transportation (see Figure R1 in our latest response) included both horizontal and vertical transport. As shown in eq. (1-2) and Figure 1 of Cai et al. (2018):

$$\frac{dN_{[i,j]}}{dt} = GR_i n_i - GR_j n_j + CoagSrc_{[i,j]} - CoagSnk_{[i,j]} + TR_{[i,j]} \quad (1)$$

where the subscripts i and j correspond to the specific particle diameters d_i and d_j , respectively; $N_{[i,j]}$ is the number concentration of particles ranging from d_i and d_j ; GR is the condensational growth rate; $CoagSrc_{[i,j]}$ and $CoagSnk_{[i,j]}$ are the formation and loss rates due to coagulation (Cai et al., 2018). $TR_{[i,j]}$ is the transport term:

$$TR_{[i,j]} = \lim_{dt \rightarrow 0} \frac{N_{[i,j]}(t + dt, z2) - N_{[i,j]}(t + dt, z1)}{dt} \quad (2)$$

here, z corresponds to the aerosol populations from different air masses (i.e., either horizontally or vertically) (Cai et al., 2018). A higher mixing layer height (MLH) was indeed observed under the P2 heatwaves, likely favorable for the vertical mixing. Figure R2 displayed the diurnal variations of MLH and the contribution of transport to the nucleation-mode particle number concentration on a NPF_{clean, HW} day. Specifically, MLH gradually developed from 8:00 LT to 17:00 LT and then sharply decreased, with a relatively lower level at night. However, the cumulative contributions of transport significantly increased from 9:00 LT to 11:00 LT and maintained increasing slowly till 21:00 LT, without any observed weakening accompanied with the sharp decline in MLH. In this sense, the contribution of vertical transport (as partly reflected by the MLH evolution) may not be so significant as the horizontal transport to NPF in this study. Considering that our data only represented NPF events during a heatwave of 2022 in southwest China, the findings may not fully capture the impacts of vertical mixing on NPF under different heatwaves. We therefore recommend future investigations to systematically evaluate both horizontal and vertical transport influences on NPF events, especially during heatwaves in diverse regions.

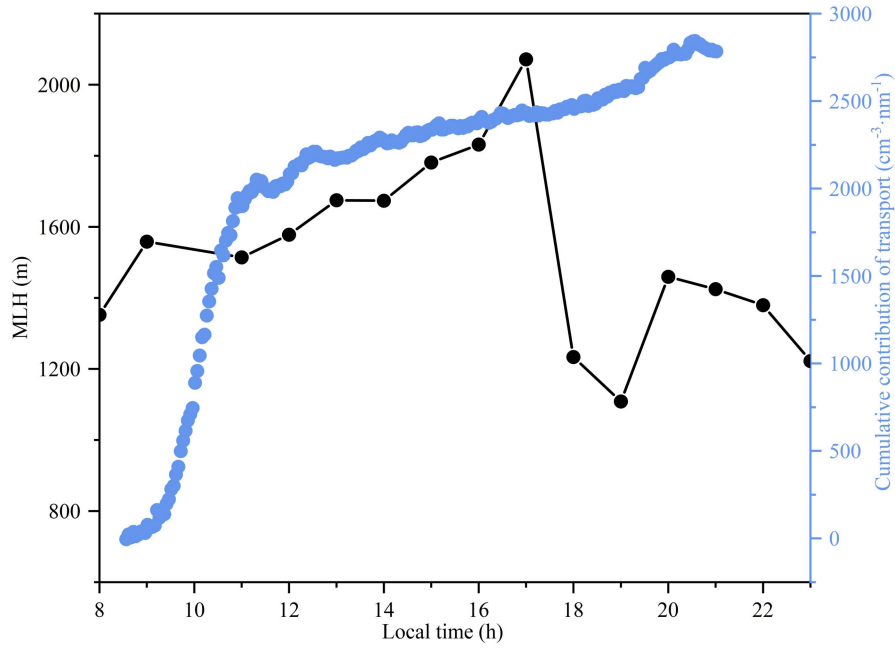


Figure R2. Diurnal evolutions of MLH and the cumulative contribution of transport to the nucleation mode particle number concentration on a $NPF_{clean, HW}$ day (i.e., 9 August 2022 as an example).

To further investigate the formation and subsequent growth of nucleation mode particles under heatwaves, we plotted the geometric mean diameter of fitted ultrafine mode (GMD_{Uf} , see eq.5 in S9 of the supplement) from NPF onset until 19:00 LT using multi-lognormal distribution functions. Unlike the commonly observed discontinuous variations in GMD_{Uf} largely driven by vertical transport during NPF events (Lai et al., 2022; Wu et al., 2024), the nearly uninterrupted GMD_{Uf} results derived from our size distribution data (Figure R3) could clearly demonstrate the local formation of new particles (<25 nm) and the subsequent complete growth process. This highlights that the increase in $N_{Nuc.}$ was not entirely due to the transport, at least not so significantly affected by vertical mixing under heatwaves. The FR and $GR_{<25\text{ nm}}$ remain comparable across different periods of our field observations, particularly when the wind direction and air mass trajectories were similar (Figure 1d, Figure S4). Additionally, previous studies have also compared the FR and GR of different NPF events dominated by local nucleation and transport, or originated from different air

masses (Chandra et al., 2016; Hussein et al., 2009; Komppula et al., 2006; Kulmala et al., 2004; Shang et al., 2023). For example, Chandra et al. (2016) observed different NPF events starting from 5 nm and from ~10 nm due to upstream transport in Fukue Island. Komppula et al. (2006) found a relatively good correlation in GR of two NPF cases in air mass transported between two measurement sites in Northern Finland. Although the “banana-shape” NPF event did not start from the minimum size of SMPS during the P2 period, the evidently continuous GMD_{Uf} can support that the heatwaves did not remarkably suppress NPF events (Figure R3). As both P1 and P2 periods exhibited local formation followed by subsequent growth of NPF, the comparative analysis of FR, GR, and other aerosol physicochemical properties during different NPF events can be regarded as methodologically justified.

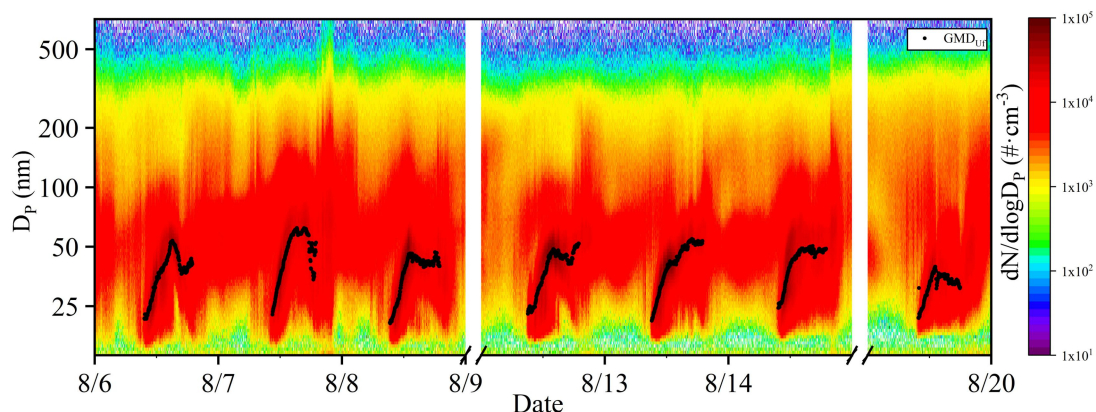


Figure R3. Overview of the measured PNSD and geometrical mean diameter of the fitted ultrafine mode (GMD_{Uf} , black dots) during the P2 $NPF_{clean, HW}$ days.

The comparable NPF occurrence frequencies during P2 (50.0%), P1 (53.8%), and P2²⁰²³ (53.8%; without heatwaves in summer 2023) further indicated that NPF events were not likely suppressed markedly by heatwaves in summer 2022. However, we have to underline that heatwaves did have a notable influence on $NPF_{clean, HW}$ events, as reflected in the potential contributions of transport, the earlier occurrence time, as well as observed reductions in FR, GR, and R_{eff} . Given the more and more frequent heatwave events with the changing climate, we primarily focus on the significant changes observed in both NPF events and aerosol optical and hygroscopic properties against the background of heatwaves.

To avoid controversial conclusions about the impacts of heatwaves on NPF (e.g., either favor or suppress), here we chose to objectively describe the differences in statistical characteristics of NPF events observed under heatwaves, while the detailed formation mechanisms of different NPF events are out of the scope of the current study and will be explored in future work. Although it is difficult to quantify the individual contribution of vertical transport in this study, we have added corresponding discussion on the potential contribution of transport to the main text and amended the Conclusions accordingly. Figure R3 has been added into the revised Supplement (i.e., Figure S15):

L358-367: *Different from that of the P1 NPF_{polluted} cases, the P2 NPF_{clean, HW} event did not start from the minimum size, and the reduced N_{Nuc} during P2 period was likely attributed to the influence of transport on the local nucleation and growth process (Fig. S4; Cai et al., 2023; Lee et al., 2019). Namely, some nucleation mode particles transported from upwind regions or from the mixing layer downwards had undergone atmospheric aging thereby a certain degree of growth upon arrival (Cai et al., 2023; Lai et al., 2022; Platis et al., 2016), resulting in relatively lower concentrations of smaller-sized particles than the case of locally formed. However, the local formation of sub-25 nm particles and the continuous growth process were still distinctly observed under heatwaves (Fig. 1i, Figs. S6, S15).*

Updates in the Conclusions and implications:

L682-685: *Although the air masses and the occurrence frequencies of NPF events were similar during different periods, NPF events exhibited distinct characteristics during the normally hot (P1, relatively polluted) and heatwaves-dominated (P2, quite clean) periods.*

L686-690: *NPF_{clean, HW} events that occurred during the heatwave P2 period were observed with lower CS, CoagS, FR and GR, as well as smaller R_{eff} and D_{mode} , than P1 NPF_{polluted} cases. According to the measured PNSDs, the P1 NPF_{polluted} events were mainly driven by local growth, while NPF_{clean, HW} events may be largely affected by transport under heatwaves.*

L710-713: *This was likely due to the observed lower FR and GR caused by possible evaporation of both unstable clusters and particle coatings under heatwaves (Bousiotis et al., 2021; Cusack et al., 2013; Deng et al., 2020; Garmash et al., 2024),*

L734-739: *This study revealed divergent changes in aerosol optical and hygroscopic properties on different NPF days, thereby modulating the aerosol radiative forcing distinctly during a heatwave in summer 2022. A comprehensive understanding of the formation mechanisms of different NPF events (e.g., local formation versus the horizontal or vertical transport) in diverse environment is crucial in the future.*

Updates in the reference list:

Lai, S., Hai, S., Gao, Y., Wang, Y., Sheng, L., Lupascu, A., Ding, A., Nie, W., Qi, X., Huang, X., Chi, X., Zhao, C., Zhao, B., Shrivastava, M., Fast, J. D., Yao, X., and Gao, H.: *The striking effect of vertical mixing in the planetary boundary layer on new particle formation in the Yangtze River Delta, Sci. Total Environ., 829, 154607, <https://doi.org/10.1016/j.scitotenv.2022.154607>, 2022.*

Platis, A., Altstädter, B., Wehner, B., Wildmann, N., Lampert, A., Hermann, M., Birmili, W., and Bange, J.: *An Observational Case Study on the Influence of Atmospheric Boundary-Layer Dynamics on New Particle Formation, Boundary-Layer Meteorol., 158, 67–92, <https://doi.org/10.1007/s10546-015-0084-y>, 2016.*

We have adjusted the title into “*Divergent changes in aerosol optical hygroscopicity and new particle formation during a heatwave of summer 2022*”, and all the ambiguous statements of ‘weather extremes (e.g., heatwaves)’ have also been revised into ‘heatwaves’.

L43-45: *Further in-depth exploration on molecular-level characterizations and aerosol radiative impacts of both direct and indirect interactions under heatwaves and other weather extremes with the warming climate are recommended.*

L135-136: *specifically under heatwaves with the changing climate.*

L667-668: *This highlights the needs for further in-depth exploration on aerosol radiative impacts under heatwaves with the changing climate,*

L739-744: *The last but not the least, further explorations on detailed molecular-scale characterizations (e.g., molecular structures and compositions of newly and secondary formed particles, as well as particle morphology) and aerosol radiative impacts including the aerosol-cloud interactions of both heatwaves and other weather extremes with the changing climate are highly recommended.*

References

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- Kulmala, M., Vehkamäki, H., Petäjä, T., Dal Maso, M., Lauri, A., Kerminen, V. M., Birmili, W., and McMurry, P. H.: Formation and growth rates of ultrafine atmospheric particles: A review of observations, *J. Aerosol Sci.*, 35, 143–176, <https://doi.org/10.1016/j.jaerosci.2003.10.003>, 2004.
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