

Dear Editor,

We thank for all the constructive comments and suggestions from the referee. We have carefully addressed and provided detailed explanations for all the 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,

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Anonymous Referee #2

RC1. From the title, this study presents divergent changes in aerosol optical hygroscopicity AND new particle formation events, both induced by heatwaves, but conclusions drawn at several places indicate that heatwaves promote NPF, leading to enhanced aerosol hygroscopicity. There is not enough evidence to substantiate this. Note that the aerosol hygroscopicity calculated for total aerosol loading in the atmosphere and NPF's contribution to it is questionable. The manuscript can be accepted, but the authors must tone down statements, including the abstract and conclusion.

Response: Thanks for the comments. To avoid unnecessary misleading, we have modified the statements accordingly throughout the main text.

Updates in the Abstract:

***L25-29:** However, mechanisms regulating aerosol optical hygroscopicity during different NPF days, particularly those under heatwaves due to global warming, remain poorly understood. In the 2022 hot summer in urban Chongqing of southwest China, simultaneous measurements of aerosol optical and hygroscopic properties, PNSD, and bulk chemical compositions were conducted.*

***L35-45:** A generally higher $f(RH)$ was observed on NPF days than non-event cases, partly attributed to distinct changes in PNSD patterns during NPF days. Moreover, heatwave-induced stronger photooxidation may intensify the formation of more hygroscopic secondary components and prolong the atmospheric aging/subsequent growth of both pre-existing and newly formed particles, largely contributing to the enhanced $f(RH)$ especially during NPF_{clean, HW} days. The higher $f(RH)$ and lowered R_{eff} could synergistically elevate the aerosol direct radiative forcing, specifically under persistent heatwave conditions. Further in-depth exploration on molecular-level characterizations and aerosol radiative impacts of both direct and indirect interactions under heatwaves with the warming climate are recommended.*

Updates in the Results and discussion:

***L454-460:** Given that newly formed particles were too small to significantly impact*

the total light scattering (Fig. S11a), this indicates that the atmospheric conditions conducive to the occurrence of NPF may promote further growth (e.g., via intensified/prolonged photooxidation or atmospheric aging processes) of pre-existing particles and newly formed ones, partly contributing to enhanced aerosol optical hygroscopicity as clued from the concurrent variations of ALWC and f_w in urban Chongqing during hot summer (Asmi et al., 2010; Wang et al., 2019; Wu et al., 2016). **L516-517:** , probably attributed to the following two aspects.

L558-562: Specifically, particles could undergo a longer and more intensified photochemical aging process during $NPF_{clean, HW}$ days as influenced by persistent heatwaves, which facilitated the secondary formation of hygroscopic aerosols and jointly contributed to a higher $f(RH)$ after 15:00 LT (Fig. 3b).

L650-652: Our findings suggest that NPF days may possess a relatively higher aerosol optical hygroscopicity in rather hot environments, e.g., the Basin area and tropical regions.

L656-659: Hence, the enhancement of aerosol optical hygroscopicity during the subsequent growth and aging of both pre-existing and newly formed particles possibly exacerbates secondary pollution and even triggers haze events (Hao et al., 2024; Kulmala et al., 2021).

Updates in the Conclusions and implications:

L691-696: In comparison to the P1 $NPF_{polluted}$ events, $NPF_{clean, HW}$ occurred approximately one hour earlier and the subsequent growth was longer during P2, likely intensifying the photochemical oxidation and prolonging atmospheric aging processes under heatwaves, thereby modulating the evolution of aerosol size distributions and chemical characteristics differently.

L704-707: Specifically, aerosol optical hygroscopicity was observed to be higher during the subsequent growth and aging of both pre-existing particles and newly formed ones on P2 $NPF_{clean, HW}$ days than that for P1 $NPF_{polluted}$ days,

L708-709: Compared with non-event cases, the daily mean $f(RH)$ levels were generally higher on NPF days in the 2022 hot summer of urban Chongqing.

L730-738: The above highlights that heatwaves could influence the NPF

characteristics (e.g., the evolution in the aerosol size distribution pattern and chemical composition) and atmospheric processing (although with a decreased aerosol R_{eff} and D_{mode} likely due to evaporation-resulted non-spherical particle morphology under persistently high temperature conditions). Further, variations in the aerosol size distribution and optical hygroscopicity under heatwaves were accompanied with the elevated $HB_{525, RH}/HB_{525}$ ratios, potentially reducing the net solar radiation directly especially in hot summer.

RC2. Authors also stated in their response that "accumulation mode particles are suggested to originate predominantly from direct emissions or aging of pre-existing particles, with NPF contributing marginally to the total number and volume concentrations of the accumulation mode". I do not see a significant enhancement in volume/mass concentrations during NPF events, and also, number concentrations are not very different between NPF and non-events.

Response: Since NPF contributes minimally to the accumulation mode particles, the volume/mass concentrations of the accumulation mode would not enhance significantly during NPF events, instead with only slightly reduced volume fractions of $VF_{Acc.}$ as displayed in the below Figure R1. While in comparison to non-event cases, the explosive formation of new particles commonly leads to a higher number concentration/fraction of both nucleation and Aitken mode particles, coincided with a much lower $NF_{Acc.}$, during NPF event time windows specifically for the heatwave-dominated P2 period with a much cleaner background (i.e., the $NPF_{clean, HW}$ events; Figure R1c, Figure S7). It should be noted that disparities in number concentrations of the accumulation mode are suggested to originate predominantly from variations in pollution levels, emission sources, atmospheric aging degree, etc., rather than the direct contributions of NPF. Hence, the number concentration/fraction of accumulation mode particles was relatively higher on P1 $NPF_{polluted}$ days than that for non-event cases, largely due to the more polluted environment during P1 NPF periods (Figure R1a, Figure S7, Table S2).

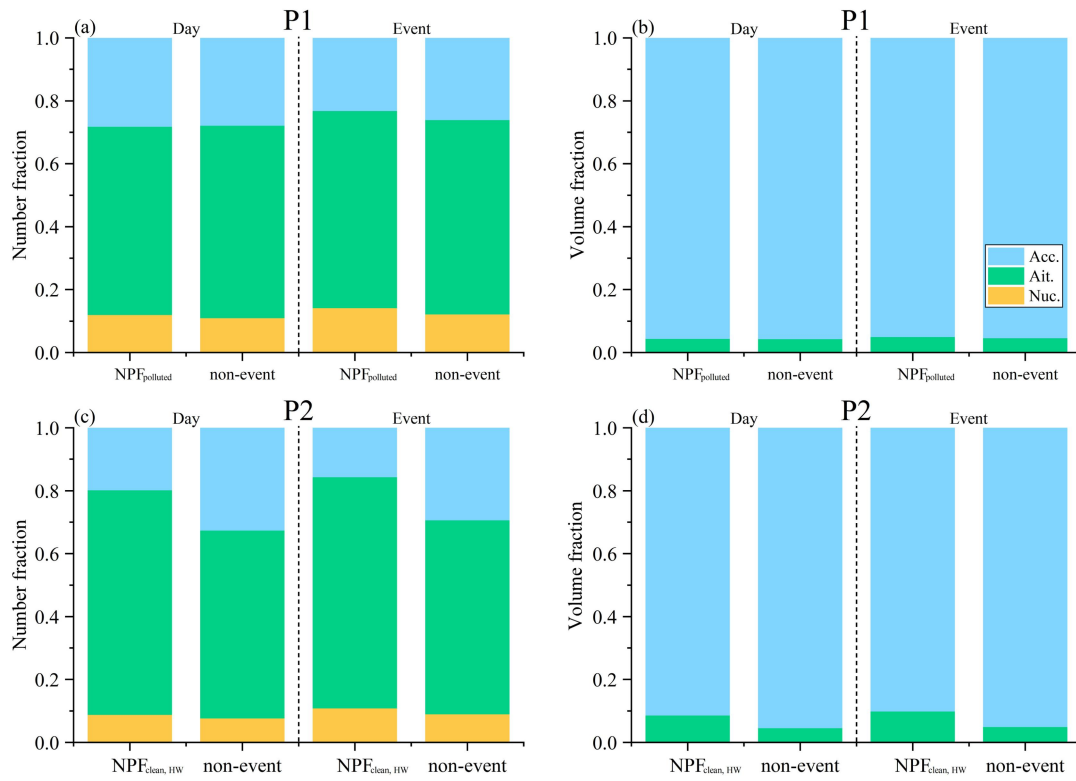


Figure R1. The number and volume fractions of different mode particles for both the whole days (labeled as 'Day') and just during the 08:00-22:00 LT time window (denoted as 'Event') in P1 and P2 period.

RC3. I suggest removing "other weather extremes" throughout the manuscript; it is vague.

Response: We have removed the corresponding statements throughout the manuscript.