



22 **Abstract.** As a crucial climate-forcing driver, the aerosol optical enhancement factor
23 ($f(\text{RH})$) is significantly modulated by the evolution of particle number size
24 distribution (PNSD), e.g., during new particle formation (NPF). The mechanisms
25 regulating aerosol optical hygroscopicity during different NPF events and non-event
26 days, particularly those influenced by heatwaves due to global warming, remain
27 poorly understood. In the extremely hot summer of 2022 in urban Chongqing of
28 southwest China, simultaneous measurements of aerosol optical and hygroscopic
29 properties, PNSD, and bulk chemical compositions were conducted. Two distinct
30 types of NPF were identified: the ones with relatively polluted period (P1) and clean
31 cases during heatwave-dominated period (P2). Heatwaves triggered NPF earlier and
32 prolonged the subsequent growth, resulting in smaller aerosol effective radius (R_{eff})
33 and lower growth rate. This agreed with the concurrently increased aerosol
34 hemispheric backscattering fraction and scattering Ångström exponent. $f(\text{RH})$ was
35 generally higher during NPF events in comparison to that for non-event cases in both
36 periods. Heatwave-induced stronger photooxidation may intensify the formation of
37 more hygroscopic secondary components, as well as the subsequent growth of
38 pre-existing particles and newly formed ultrafine ones, thereby enhancing aerosol
39 optical hygroscopicity especially during heatwave-influenced NPF events. The
40 promoted $f(\text{RH})$ and lowered R_{eff} could synergistically elevate the aerosol direct
41 radiative forcing, specifically under persistent heatwave conditions. Further in-depth
42 exploration on molecular-level characterizations and aerosol radiative impacts of both
43 direct and indirect interactions during weather extremes (e.g., heatwaves) with the
44 warming climate are recommended.

45

46 **1 Introduction**

47 Weather extremes (e.g., heatwaves) have become more and more frequent and
48 intense largely due to the global climate change, and the heatwave-driven
49 environmental, climatic, and health effects have garnered widespread attention



50 (Hauser et al., 2016; Sun et al., 2016). The China Climate Bulletin 2022 confirmed
51 that the national average temperature reached an unprecedented high level since 2012
52 (China Meteorological Administration, 2022), and the risk of heatwaves in China will
53 persist and potentially intensify in the future (Guo et al., 2016; Li et al., 2017).
54 Extreme heatwave events could pose significant threats to human health, the survival
55 of organisms, agriculture, and socio-economic activities (e.g., power supply
56 restrictions) (Anderson and Bell, 2011; Ma et al., 2021; Su, 2021). Moreover,
57 heatwaves can trigger natural disasters such as droughts and wildfires, affecting social
58 stability (Sharma and Mujumdar, 2017).

59 Heatwaves could also affect the atmospheric physical and chemical processes by
60 modulating ambient meteorological conditions. Specifically, extremely high
61 temperature weather is typically characterized by a combination of intensified solar
62 radiation with elevated temperature and low humidity levels. This could significantly
63 affect the formation and evolution of secondary aerosols in the atmosphere (Bousiotis
64 et al., 2021; Hamed et al., 2011; Kurtén et al., 2007), given that the air temperature is
65 crucial for chemical reactions (Xu et al., 2011). New particle formation (NPF) serves
66 as a crucial source of atmospheric particulate matter and plays a significant role in the
67 secondary transformation processes in the atmosphere (Zhu et al., 2021). Generally,
68 NPF involves the initial formation of thermodynamically stable clusters from
69 condensable vapors (e.g., ammonia, sulfuric acid, and organic precursor gases) and
70 subsequent growth of the formed clusters, eventually reaching detectable sizes or even
71 larger dimensions (Kerminen et al., 2018; Kulmala et al., 2003, 2012). Over time,
72 these newly formed particles have the potential to serve as cloud condensation nuclei
73 (CCN), thereby impacting the global climate (Salma et al., 2016). NPF events
74 normally introduce a sharp increase in the number concentration of nucleation mode
75 particles within a short time, altering the particle number size distribution (PNSD).
76 These variations in PNSD likely influence intrinsic physicochemical properties of
77 aerosols, such as the optical hygroscopicity (Chen et al., 2014; Titos et al., 2016; Zhao
78 et al., 2019).



79 Aerosol hygroscopicity plays a critical role in the atmospheric environment and
80 climate change, given the complex interaction between aerosol particles and water
81 vapor (Zhao et al., 2019; Zieger et al., 2011). Water uptake by aerosols not only alters
82 the particle size and composition (e.g., as reflected in the aerosol refractive index) but
83 also impacts aerosol scattering efficiency, which further contributes to the uncertainty
84 in aerosol radiative forcing estimation (Titos et al., 2016, 2021). The aerosol optical
85 hygroscopicity parameter, $f(\text{RH})$, defined as the ratio of the scattering coefficient at a
86 certain RH to that of the dry condition, was widely used to describe the aerosol
87 scattering enhancement through water uptake (Covert et al., 1972; Titos et al., 2016;
88 Zhao et al., 2019). Numerous studies have demonstrated that $f(\text{RH})$ is influenced by
89 the size distribution, in addition to particle chemical composition (Chen et al., 2014;
90 Kuang et al., 2017; Petters and Kreidenweis, 2007; Quinn et al., 2005). NPF could
91 alter the size distribution thereby aerosol optical properties, nonetheless, there is
92 currently limited research on the impact of NPF on aerosol optical hygroscopicity (Ma
93 et al., 2016; Ren et al., 2021). It is suggested that the influence of NPF on aerosol
94 hygroscopicity was likely due to changes in aerosol chemical composition at different
95 stages of NPF events (Cheung et al., 2020), whereas the subsequent particle growth
96 associated with NPF events can significantly affect particle hygroscopicity as well
97 (Wu et al., 2016). Although there have been a great many studies on chemical
98 composition dependences of aerosol hygroscopicity (e.g., the variation in composition
99 of precursor species during NPF events), it is important to acknowledge that the
100 utilized chemical compositions of NPF were either from $\text{PM}_{2.5}$ or PM_1 bulk data,
101 which may differ from the corresponding composition of newly formed ultrafine
102 particles primarily in the nucleation and Aitken modes. This may further introduce
103 bias in exploring the impacts of NPF events on aerosol optical hygroscopicity if solely
104 based on $\text{PM}_{2.5}$ chemical composition, especially in the initial nucleation stage of NPF.
105 Hence, more comprehensive investigations on the influencing mechanisms of aerosol
106 optical hygroscopicity from different perspectives are required, e.g., for the aspects of
107 the evolution of particle size distribution in modulating aerosol optical and
108 hygroscopic properties (Tang et al., 2019; Zhao et al., 2019). Additionally, field



109 observations on $f(\text{RH})$ under extreme weather conditions (e.g., heatwaves) are rather
110 scarce, largely hindering our understanding of how weather extremes (e.g., extremely
111 high temperature) influence the optical hygroscopic properties of aerosols. This
112 knowledge gap further impedes comprehensive understanding of the aerosol water
113 uptake property and resulted effects on air quality and the climate under varied
114 synoptic conditions.

115 During the summer of 2022, a rare heatwave event raged throughout the
116 Sichuan-Chongqing region of southwest China, with the daily maximum temperature
117 exceeding 40 °C lasted for 29 days observed at Beibei meteorological station in
118 Chongqing (Hao et al., 2023). This persistent heatwave not only impacted residents'
119 daily lives significantly, but also affected the aerosol optical and hygroscopic
120 properties likely through NPF and relevant atmospheric processing during the period.
121 In this study, a field observation was conducted by using a combination of a
122 home-built humidified nephelometer system and a scanning mobility particle sizer
123 (SMPS), along with the total suspended particle (TSP) filter sampling. A main goal of
124 this study is to investigate the influence of heatwaves on NPF events and subsequent
125 impacts on aerosol optical and hygroscopic properties. Furthermore, we aimed to
126 explore the mechanisms behind the variability in $f(\text{RH})$ under different meteorological
127 conditions and NPF events. This study will further enrich insights into the potential
128 environmental and climatic impacts due to variations in the aerosol optical
129 hygroscopicity and size distribution, specifically under weather extremes (e.g.,
130 heatwaves) with the changing climate.

131

132 **2 Data and Methods**

133 **2.1 Field observation**

134 A continuous field observation on aerosol optical, hygroscopic and chemical
135 properties was carried out from July 29 to August 19, 2022. The detailed description
136 of the observation site is available in Supporting Information, S1. During the



137 observation period, urban Chongqing suffered a rare heatwave (Fig. S1), which
138 significantly affected the local transportation and industrial activities (Hao et al.,
139 2023). Based on the temperature records and concurrent aerosol light scattering data,
140 the whole study period was categorized into two stages: (1) the normally hot period
141 (with the daily maximum temperature seldomly above 35°C) from 29 July to 6 August
142 (simply labeled as P1); (2) the heatwave-dominated cleaner period (persistent
143 occurrence of the hourly temperature over 40°C, and the hourly total scattering
144 coefficient at 525 nm below 100 Mm⁻¹) during 7-19 August 2022 (marked as P2).

145 2.2 Instrumentation and methods

146 2.2.1 Measurements of aerosol optical hygroscopicity

147 The humidified nephelometer system, consisting of two three-wavelength (i.e.,
148 450, 525, and 635 nm) nephelometers (Model Aurora 3000, Ecotech Inc.) and a
149 humidification unit, was used to determine the aerosol light scattering enhancement
150 factor, $f(\text{RH})$. Briefly, the aerosol scattering ($\sigma_{\text{sca}, \lambda}$) and backscattering coefficients
151 ($\sigma_{\text{bsca}, \lambda}$) were detected in a dry state ($\text{RH} < 30\%$) and at a fixed RH level of $85\% \pm 1\%$,
152 respectively, with the humidification efficiency regulated automatically by a
153 temperature-controlled water bath.

154 Hence, $f(\text{RH})$ could be calculated as the ratio of the aerosol scattering coefficient
155 at a predefined RH ($\sigma_{\text{sca}, \text{RH}}$) to the dry ($\sigma_{\text{sca}, \text{dry}}$) state, i.e., $f(\text{RH}) = \sigma_{\text{sca}, \text{RH}} / \sigma_{\text{sca}, \text{dry}}$
156 (Covert et al., 1972). In this study, the $f(\text{RH})$ discussed is mainly targeted for the 525
157 nm wavelength, unless otherwise specified. More information about the measurement
158 of humidified nephelometer system was illustrated in S2 of the supplement.

159 In additional to $f(\text{RH})$, aerosol optical parameters, such as scattering Ångström
160 exponent (SAE; Schuster et al., 2006) and hemispheric backscattering fraction (HBF;
161 Collaud Coen et al., 2007), were calculated as below:

$$162 \quad \text{SAE}_{\lambda_1/\lambda_2} = \frac{-\ln(\sigma_{\text{sca}, \lambda_1} / \sigma_{\text{sca}, \lambda_2})}{\ln(\lambda_1 / \lambda_2)} \quad (1)$$

$$163 \quad \text{HBF}_{\lambda} = \frac{\sigma_{\text{bsca}, \lambda}}{\sigma_{\text{sca}, \lambda}} \quad (2)$$



164 where $\sigma_{\text{sca}, \lambda}$ and $\sigma_{\text{bsca}, \lambda}$ represent the aerosol scattering and backscattering
165 coefficients at a specific wavelength λ (e.g., λ_1, λ_2), respectively.

166 Both HBF and SAE reflect crucial optical properties of aerosols, e.g., an elevated
167 HBF (or SAE) generally signifies a higher concentration (or a smaller particle size) of
168 fine particles within the aerosol population (Jefferson et al., 2017; Kuang et al., 2017;
169 Luoman et al., 2019). The HBF and SAE discussed in this study are targeted for the
170 dry condition, unless otherwise specified. Based on the measurements with the
171 humidified nephelometer system, the equivalent aerosol liquid water content (ALWC)
172 and the corresponding fraction of ALWC (f_w) can also be obtained (Kuang et al, 2018;
173 see S2 of the supplement).

174 The SMPS-measured concurrent particle number size distributions were further
175 utilized to calculate the aerosol effective radius (R_{eff}) and representative parameters
176 for NPF events, e.g., the growth rate (GR) of new particle, condensation sink (CS)
177 and coagulation sink (CoagS) (Kulmala et al., 2012). More details are provided in the
178 supplement.

179

180 **2.2 Determination of the aerosol direct radiative forcing (ADRF) enhancement** 181 **factor**

182 Given the high sensitivity of aerosol optical properties (e.g., $f(\text{RH})$) to the
183 changes in RH under real atmospheric conditions, the influence of RH, or rather the
184 aerosol hygroscopicity, on ADRF can be quantitatively estimated with the radiative
185 transfer model by the following equation (Chylek and Wong, 1995; Kotchenruther et
186 al., 1999; L. Zhang et al., 2015):

$$187 \quad \Delta F_{\text{R}}(\text{RH}) = -(S_0/4) \times [T_a^2 \times (1 - A_c)] \times [2 \times (1 - R_s)^2 \times \beta(\text{RH}) \times \tau_s - 4 \times R_s \times \tau_a] \quad (3)$$

188 where S_0 is the solar constant, T_a is the atmosphere transmittance, A_c is the
189 fractional cloud amount, R_s is the albedo of the underlying surface, $\beta(\text{RH})$ is the
190 upscattering fraction at a defined RH, τ_s and τ_a are the optical thicknesses of the
191 aerosol layer due to light scattering and light absorption, respectively, which can be
192 expressed as follows (Kotchenruther et al., 1999):



193
$$\tau_s = M \times \alpha_s \times f(\text{RH}), \tau_a = M \times \alpha_a \quad (4)$$

194 where M is the column burden of aerosol (unit: gm^{-2}), α_s is the mass scattering
195 efficiency (MSE), and α_a is the mass absorption efficiency (MAE). The direct
196 radiative forcing is usually calculated with the assumption that the absorption
197 enhancement is negligible, in comparison to the aerosol scattering enhancement (Xia
198 et al., 2023).

199 Hence, the dependence of ADRF on RH (i.e., $f_{\text{RF}}(\text{RH})$) can be estimated by
200 equation (5) (Chylek and Wong, 1995; Kotchenruther et al., 1999; L. Zhang et al.,
201 2015):

202
$$f_{\text{RF}}(\text{RH}) = \frac{\Delta F_{\text{R}}(\text{RH})}{\Delta F_{\text{R}}(\text{dry})} = \frac{(1 - R_s)^2 \times \beta(\text{RH}) \times \alpha_s \times f(\text{RH}) - 2 \times R_s \times \alpha_a}{(1 - R_s)^2 \times \beta(\text{dry}) \times \alpha_s \times f(\text{dry}) - 2 \times R_s \times \alpha_a} \quad (5)$$

203 where the constant parameters used were $R_s = 0.15$, $\alpha_a = 0.3 \text{ m}^2 \cdot \text{g}^{-1}$ (Hand and
204 Malm, 2007; Fierz-Schmidhauser et al., 2010). It should be noted that the assumed
205 constant α_a might introduce some uncertainty in the calculated $f_{\text{RF}}(\text{RH})$, given the fact
206 that the contribution of absorption by brown carbon was unknown, although the mass
207 fraction of BC in TSP remained almost constant (i.e., $4.6\% \pm 1.1\%$, Fig. S2) during
208 the observation period. The parameter α_s was calculated by dividing $\sigma_{\text{sca}, 525}$ in the dry
209 condition by the mass concentration of $\text{PM}_{2.5}$ (i.e., $\alpha_s = \sigma_{\text{sca}, 525} / \text{PM}_{2.5}$). β could be
210 calculated empirically from the measured HBF: $\beta = 0.0817 + 1.8495 \times \text{HBF} - 2.9682$
211 $\times \text{HBF}^2$ (Delene and Ogren, 2002).

212 Results of the offline chemical analysis with TSP filter samples are provided in
213 S3. Given that the particle number and mass size distributions of components such as
214 sulfate and organics from diverse emission sources were primarily concentrated
215 within the submicron size range (An et al., 2024), the bulk chemical compositions of
216 TSP could provide a reasonably good reference for the characterization of NPF and
217 related optical and hygroscopic properties of $\text{PM}_{2.5}$. It should be noted that the
218 corresponding mass fraction of some components (e.g., crustal materials) likely
219 biased for larger particles. The simultaneous meteorological and air quality data can
220 be found in S4.



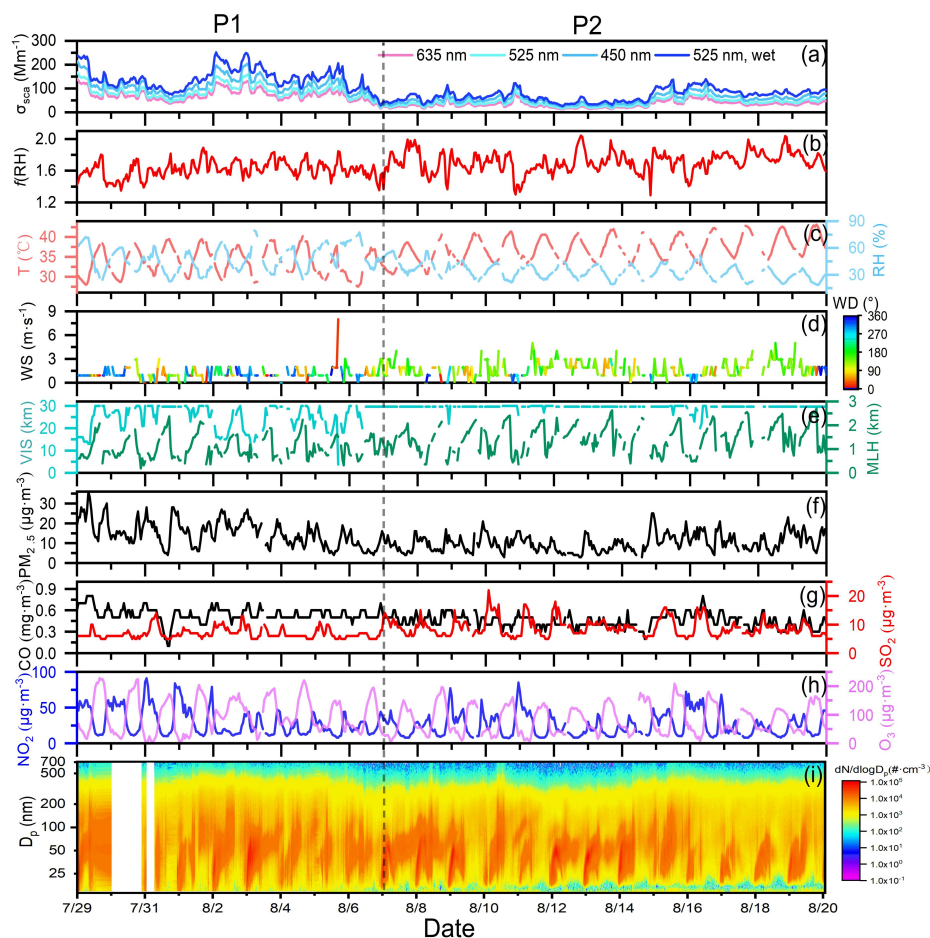
221 **3 Results and discussion**

222 **3.1 Overview of the aerosol optical hygroscopicity and PNSD measurements**

223 Figure. 1 displayed the time series of the measured aerosol scattering coefficients,
224 $f(\text{RH})$, PNSD, and the corresponding meteorological conditions and air pollutants
225 during the study period. A sharp decrease in aerosol scattering coefficients and $\text{PM}_{2.5}$,
226 accompanied with the continuous excellent visibility over 20 km was observed after
227 August 6, indicating a markedly cleaner environment during P2 in comparison to P1
228 in summer 2022 of Chongqing. This could be largely attributed to the reduction in
229 anthropogenic emissions (e.g., NO_2 , CO) from limited outdoor activities influenced
230 by the heatwaves in P2, as well as partly suspended industries and transportation to
231 alleviate the power shortage issue. Notably, the increased wind speed and enhanced
232 mixing layer height (MLH) also enabled a more favorable atmospheric diffusion
233 condition in P2, facilitating the dilution of surface air pollutants (Zhang et al., 2008).
234 However, a higher mass concentration of SO_2 was observed in the P2 period, likely
235 due to a surge in electricity demand and resulted higher emissions from power plants
236 operating almost at full capacity during the heatwave (Su, 2021; Teng et al., 2022).
237 Moreover, significant discrepancies in the aerosol optical and hygroscopic properties
238 were observed under different synoptic conditions (Table S2). Both HBF and SAE
239 were higher during the P2 period, aligning with the smaller R_{eff} (Table S2). The $f(\text{RH})$
240 was found to be larger in heatwave days, with the mean values of 1.6 ± 0.1 and $1.7 \pm$
241 0.2 during the P1 and P2 periods, respectively. Differently, ALWC was more abundant
242 during the normally hot P1 period than the heatwave-dominated P2 period, likely due
243 to that the derivation algorithm of ALWC utilized in this study (Kuang et al., 2018)
244 was partly dependent on (e.g., positively correlated) the aerosol scattering coefficient
245 in the dry condition. The mean $\sigma_{\text{scat}, 525}$ for P2 was about 46.8% of that for the P1
246 period, and the corresponding mean level of ALWC was approximately 55.8% of that
247 for P1. This partly agrees with the stronger aerosol optical hygroscopicity with a
248 marginally higher f_w during the P2 period, highlighting a complex interaction between
249 the optical enhancement and aerosol physicochemical properties.



250 The particle number size distribution data suggested that NPF events appeared in
251 about half the number of observation days (Fig. 1i), with the frequency during the P2
252 period (53.8%) slightly higher than that of P1 (44.4%). This suggests the rather
253 frequent summer NPF events in Chongqing, notably higher than those observed in
254 other regions of the world, e.g., Beijing (16.7%, Deng et al., 2020; ~20%, Wang et al.,
255 2013), Dongguan (4%, Tao et al., 2023), Hyytiälä (<40%, Dada et al., 2017) and
256 LiLLE (<20%, Crumeyrolle et al., 2023). Moreover, the frequent NPF events during
257 heatwaves formed substantially ultrafine particles that are of less contribution to
258 aerosol optical properties in comparison to large particles, partially explaining the
259 significantly lower levels of total scattering coefficients observed during the P2
260 period.



261

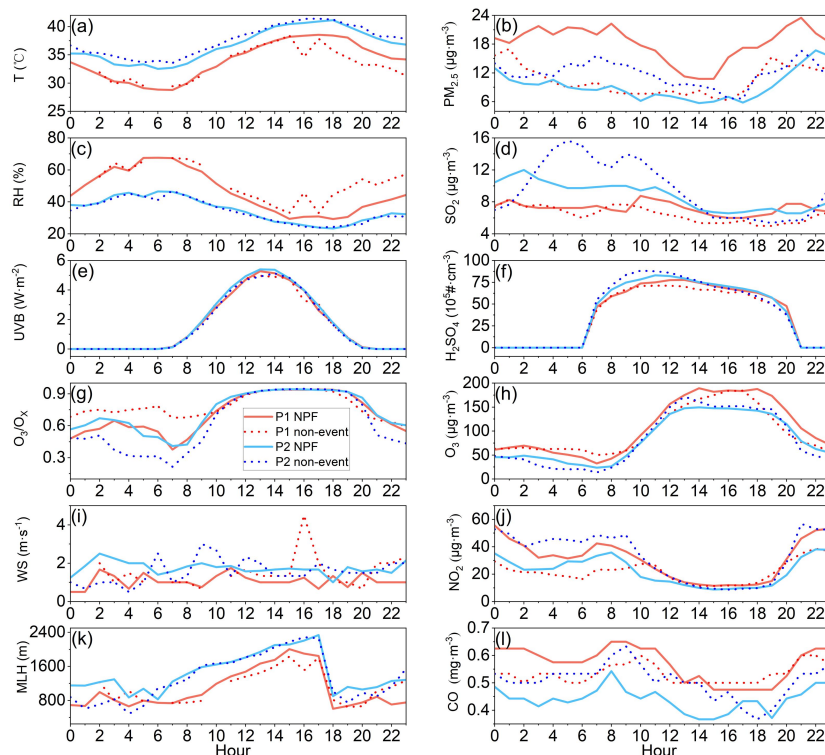
262 **Figure 1.** Time series of the measured aerosol scattering coefficients, $f(\text{RH})$,
263 meteorological conditions, air pollutants, and particle number size distribution during
264 the study period.

265 3.2 Characteristics of NPF events in different periods

266 Aside from gaseous precursors (e.g., SO_2 , volatile organic compounds),
267 meteorological conditions also play a key role in the occurrence of NPF events. In
268 brief, NPF events are more likely to appear under sunny and clean conditions
269 (Bousiotis et al., 2021; Crumeyrolle et al., 2023; Deng et al., 2021; Wang et al., 2017).
270 To further explore the characteristics of NPF events in different periods, the



271 time-averaged diurnal variations of meteorological parameters and air pollutant
272 concentrations during both NPF events and non-event days are presented in Fig. 2.



273
274 **Figure 2.** Diurnal variations of temperature (a), PM_{2.5} mass loading (b), RH (c), SO₂
275 (d), UVB (e), H₂SO₄ (f), O₃/O_x (g), O₃ (h), WS (i), NO₂ (j), MLH (k) and CO (l)
276 during P1 (red) and P2 (blue) NPF events (solid line), as well as the corresponding
277 non-event days (dash line).

278 NPF events during the P1 period tended to occur in relatively polluted
279 environments compared to that of P2 NPF events, as evidenced by the higher $\sigma_{\text{sca}, 525}$,
280 increased air pollutant concentrations and lower visibility levels during P1 (Table S2,
281 Fig. 1). On P2 NPF event days, the overall mean $\sigma_{\text{sca}, 525}$ was $33.2 \pm 11.7 \text{ Mm}^{-1}$,
282 decreased by 68.0% (39.3%) in comparison to that for P1 NPF event days (P2
283 non-event days). In addition, the mean PM_{2.5} concentration was even lower than 10.0
284 $\mu\text{g}\cdot\text{m}^{-3}$, and the corresponding visibility level was almost reaching the upper detection
285 limit of 30 km. All the above implies that the P2 NPF events were generally



286 accompanied with a much cleaner environment. It is notable that the increase in SO₂
287 concentration after 9:00 (Fig. 2d), along with the significant decrease in PM_{2.5} mass
288 loadings thereby lowered CS or CoagS after 8:00 during P1 NPF events (Fig. 2b),
289 likely favored the occurrence of NPF events. The higher gas-phase sulfuric acid (i.e.,
290 H₂SO₄, as estimated with the UVB and SO₂ concentration, Lu et al., 2019, S4) on the
291 same NPF event days (Fig. 2f), further suggesting that sulfuric acid concentration was
292 a critical factor for the occurrence of P1 NPF events.

293 Meanwhile, the diurnal evolutions of meteorological conditions (e.g., T, RH,
294 MLH) for NPF events were distinct between P1 and P2 periods, although relatively
295 insignificant differences were observed for both NPF events and non-event days
296 within a same period (Fig. 2). This might suggest that meteorological factors might
297 not be the predominant determining factor of NPF occurrence, while NPF could be
298 accompanied with quite different meteorological conditions depending on gaseous
299 precursors and preexisting condensation sinks. For instance, the heatwave-influenced
300 NPF events were typically of clean-type NPF, characterized with lower background
301 aerosol loading, higher temperature and favorable atmospheric dispersion capacity
302 with the higher MLH. However, it is reported that excessive heat can increase the
303 evaporation rate of critical acid-base clusters during the nucleation process and reduce
304 the stability of initial molecular clusters (Bousiotis et al., 2021; Kurtén et al., 2007;
305 Zhang et al., 2012). On the other hand, the emission rate of biogenic VOCs (BVOCs,
306 e.g., isoprene, monoterpene) from nearby plants and trees would decrease when
307 temperature exceeded around 40 °C (Guenther et al., 1993; Pierce and Waldruff,
308 1991), despite that BVOCs plays a key role in the nucleation mechanism of NPF
309 (Wang et al., 2017; Zhang et al., 2004). Hence, the even higher temperature (e.g.,
310 T >40 °C) likely hindered the occurrence and subsequent growth of NPF during
311 non-event days of the P2 period, in spite of higher concentrations of SO₂ and H₂SO₄.

312 To further investigate the effect of heatwave on NPF events, the diurnal
313 variations of aerosol number and volume concentrations, as well as R_{eff}, for different
314 modes were illustrated in Fig. S4, and the relationship between temperature and the
315 duration of NPF events was displayed in Fig. S5. The NPF events influenced by



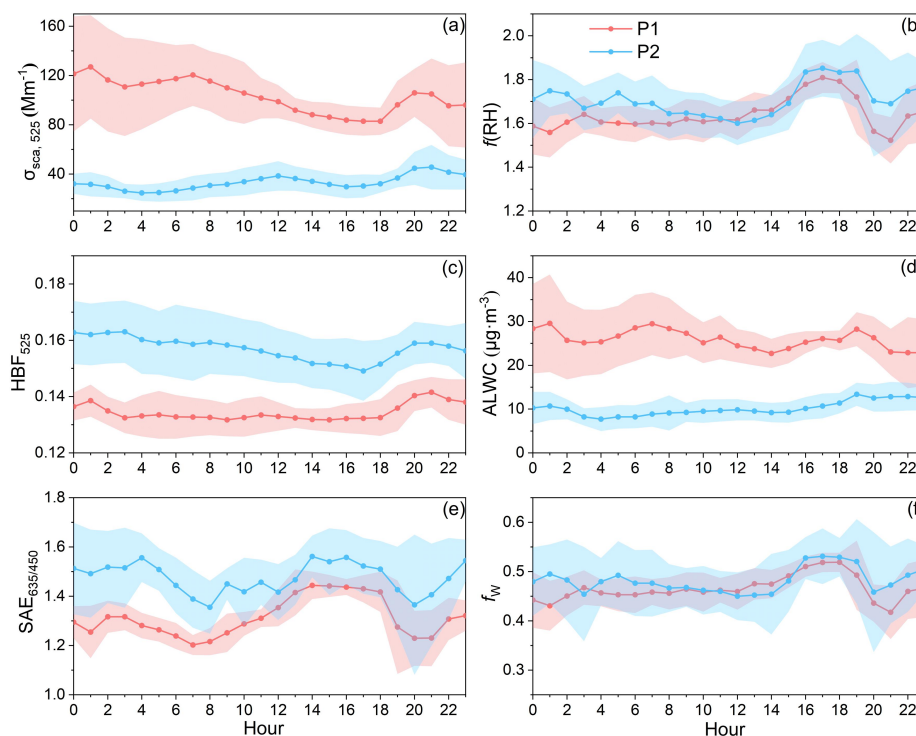
316 heatwaves usually initiated earlier (Fig. S5), with the number concentration of
317 nucleation mode particles ($N_{\text{Nuc.}}$) in P2 NPF cases peaked about an hour earlier (whilst
318 relatively lower) in comparison to P1 event days (Fig. S4a). This implies that
319 heatwaves may accelerate the attainment of the temperature threshold of NPF events,
320 as evidenced by the earlier NPF start time corresponding to higher temperature ranges
321 (Fig. S5). Furthermore, the end time of subsequent particle growth during P2 period
322 was even later (i.e., ~ 21:00 LT) than that of P1 cases (Fig. S5). Given the lower GR
323 during P2 NPF events (Table S2), these explosively formed new particles could
324 persist longer in the warmer atmosphere and probably undergo aging processes with a
325 relatively higher oxidation degree. This is supported by the commonly higher ratios of
326 secondary organic carbon (SOC) to organic carbon (OC) (i.e., $\text{SOC/OC} > 0.5$) during
327 the P2 NPF event days (Fig. S2b). The diurnal patterns of aerosol volume
328 concentrations for different size modes were similar to that of aerosol number
329 concentrations during NPF events (Fig. S4b1-b3). It is worth noting that both the R_{eff}
330 of Aitken mode particles ($R_{\text{Ait.}}$) and accumulation mode particles ($R_{\text{Acc.}}$) were smaller
331 during P2 NPF events than that of P1 NPF events (Fig. S4c2-c3), which may further
332 influence size-dependent aerosol optical and hygroscopic properties (e.g., $\sigma_{\text{sca}, 525}$,
333 HBF, SAE, $f(\text{RH})$). The decrease in $R_{\text{Ait.}}$ and $R_{\text{Acc.}}$ during heatwaves could be
334 attributed to three factors: (1) evaporation of the outer layer of particles due to
335 extremely high temperature (Bousiotis et al., 2021; Cusack et al., 2013; Deng et al.,
336 2020; Li et al., 2019); (2) lower GR of particles under a cleaner environment; (3)
337 reduced emissions of larger primary particles during the P2 period.

338 **3.3 Characteristics of the aerosol optical and hygroscopic properties during NPF** 339 **events**

340 Diurnal variations of the aerosol optical and hygroscopic parameters during NPF
341 events were shown in Fig. 3, and the corresponding results for non-event days can
342 refer to Fig. S6. Generally, $\sigma_{\text{sca}, 525}$ possessed a similar bimodal diurnal pattern to that
343 of the accumulation mode aerosol volume concentration ($V_{\text{Acc.}}$) (Fig. S4b3), as
344 supported by the positive correlation between $\sigma_{\text{sca}, 525}$ and SMPS-measured aerosol



345 volume concentration (Fig. S8). This is also consistent with the Mie theory, with a
346 stronger increase in the scattering efficiency for accumulation mode particles (Titos et
347 al., 2021). The diurnal pattern of $\sigma_{\text{sca}, 525}$ also varied distinctly between different NPF
348 events. Specifically, a minor peak of $\sigma_{\text{sca}, 525}$ around 12:00 (Fig. 3a) was influenced by
349 the newly formed particles during P2 NPF events, which contributed more
350 significantly to the aerosol number and volume concentrations within 100 nm size
351 ranges in pretty clean environments (Fig. S3c, g). Instead of a noontime peak, $\sigma_{\text{sca}, 525}$
352 was observed with an early peak around the morning rush hours and a maximum
353 value similarly occurred at the nighttime on P1 NPF event days.



354 **Figure 3.** Diurnal variations of $\sigma_{\text{sca}, 525}$ (a), $f(\text{RH})$ (b), HBF_{525} (c), ALWC (d),
355 $\text{SAE}_{635/450}$ (e) and f_w (f) on NPF event days during P1 (red line) and P2 (blue line)
356 periods. The shaded areas stand for the corresponding $\pm 1\sigma$ standard deviations.
357

358 Both HBF and SAE during P2 NPF events were significantly higher than that
359 of P1 NPF cases (Fig. 3c, e), largely due to the smaller R_{eff} during P2
360 heatwave-dominated NPF events (Table S2). Moreover, the correlation between HBF



361 (or SAE) and particle size in each mode was relatively weaker on NPF event days
362 than on non-event days, especially for P2 NPF events (Fig. S10). A strongest negative
363 correlation was found between HBF and R_{eff} of the accumulation mode in comparison
364 to other modes, highlighting that HBF is more sensitive to the size distribution of
365 accumulation mode particles (Collaud Coen et al., 2007). Given that NPF would
366 largely enhance the abundance of both nucleation and Aitken mode aerosols, no
367 significant variation in HBF was observed during the daytime due to the weakened
368 correlation between HBF and $R_{\text{Acc.}}$ of NPF events. SAE is commonly used as an
369 indicator of particle size distribution, almost decreasing monotonously with the
370 increase of aerosol size within $1 \mu\text{m}$ (Kuang et al., 2017, 2018; Luoma et al., 2019).
371 Accordingly, SAE decreased over the morning and evening rush hours when coarse
372 particles (e.g., aged particles, road dust, automobile exhaust) generated during
373 anthropogenic activities, accompanied with an increase in CO that is taken as the
374 proxy for primary emissions (Fig. 2l) (Yarragunta et al., 2020). On the contrary, the
375 abundant ultrafine particles formed during NPF events led to a continuous increase in
376 SAE during the day.

377 $f(\text{RH})$ exhibited a similar diurnal pattern on the P1 and P2 NPF event days
378 (Fig. 3b). During the daytime, $f(\text{RH})$ remained relatively stable and gradually
379 increased until peaking around 16:00-18:00, with a generally higher $f(\text{RH})$
380 particularly after 15:00 during P2 NPF events than that of P1 cases. The insignificant
381 fluctuation of relatively lower $f(\text{RH})$ levels before the noon could be attributed to the
382 continuous development of the mixing layer (Fig. 2k), leading to an efficient mixing
383 of particles in the nocturnal residual layer with anthropogenic emissions near the
384 ground. Additionally, photochemical reactions in the afternoon facilitated the
385 formation of more hygroscopic secondary aerosols with a higher oxidation level (Liu
386 et al., 2014; R. Zhang et al., 2015). The diurnal patterns of O_3 and the $\text{O}_3/\text{O}_\text{X}$ ratio (i.e.,
387 an indicator of atmospheric oxidation capacity, where $\text{O}_\text{X} = \text{O}_3 + \text{NO}_2$, Tian et al.,
388 2021) also showed similar trends (Fig. 2g, 2h). The presence of black carbon (BC)
389 mixed with organic compounds (e.g., from traffic emissions and residential cooking
390 activities) explained the rapid decrease in $f(\text{RH})$ during the evening rush hours (Liu et



391 al., 2011). Furthermore, the daily mean $f(\text{RH})$ during NPF events was higher than that
392 of non-event days (Table S2), particularly after the ending of NPF events around
393 12:00. Given that newly formed particles were too small to significantly impact the
394 total light scattering (Fig. S7), this indicates that the atmospheric conditions
395 conducive to the occurrence of NPF may promote further growth (e.g., via
396 photooxidation or atmospheric aging processes) of pre-existing particles and newly
397 formed ones, leading to enhanced aerosol optical hygroscopicity as clued from the
398 concurrent variations of ALWC and f_w in urban Chongqing during hot summer (Asmi
399 et al., 2010; Wang et al., 2019; Wu et al., 2016). The diurnal pattern of ALWC closely
400 mirrored the variation in $\sigma_{\text{sea}, 525}$, while f_w followed the similar evolution of $f(\text{RH})$.
401 This suggests that ALWC was more sensitive to changes in the aerosol volume
402 concentration, as determined by the corresponding retrieval algorithm (Kuang et al.,
403 2018). The relatively higher f_w levels (e.g., even exceeded 50% sometimes) verified
404 the enhancement of aerosol hygroscopicity during NPF events in comparison to that
405 of non-event days.

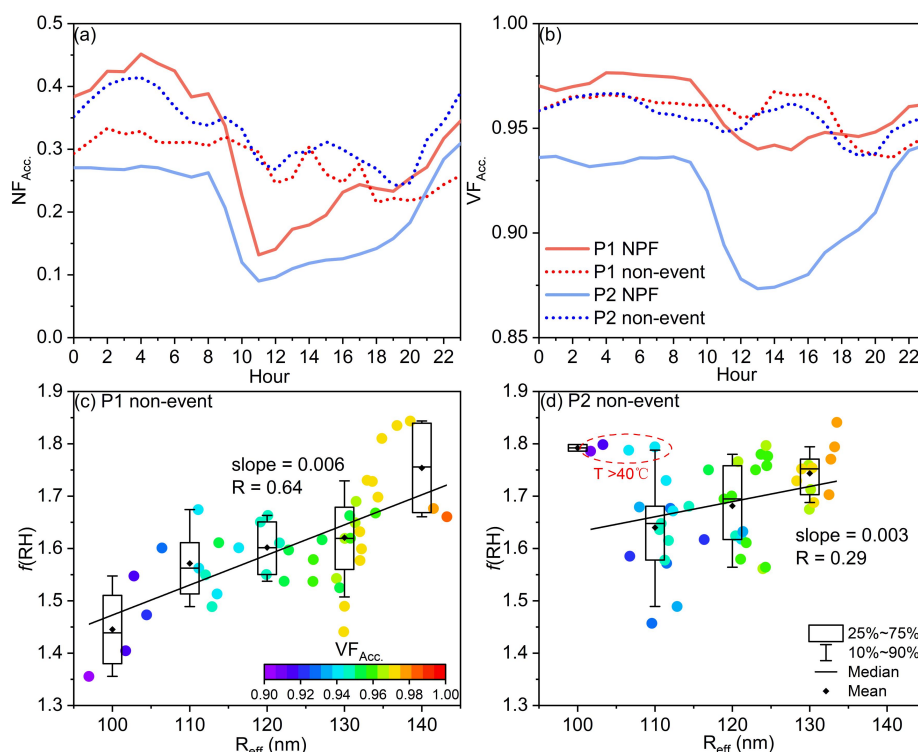
406 **3.4 Heatwave-induced divergent changes in aerosol optical hygroscopicity**

407 To further explore the impacts of heatwaves on $f(\text{RH})$ during diverse NPF events,
408 data mainly within the time window of 08:00-22:00 (i.e., typically covered the
409 complete process of NPF and subsequent growth, while excluded higher RH
410 conditions at night) were utilized for the following discussion.

411 A positive correlation between $f(\text{RH})$, R_{eff} and the volume fraction of
412 accumulation mode particles ($\text{VF}_{\text{Acc.}}$) was found on non-event days (Fig. 4c-d), when
413 the aerosol size distribution was undisturbed by newly formed ultrafine particles and
414 the corresponding $\text{VF}_{\text{Acc.}}$ maintained around a relatively high level of 0.95 (Fig. 4a-b).
415 The notably positive correlation between $f(\text{RH})$ and R_{eff} could be linked to the
416 secondary formation of hygroscopic particles within the accumulation mode,
417 primarily via photochemical reactions and further intensified by heatwaves during the
418 day particularly of the P2 period (Gu et al., 2023; Liu et al., 2014; R. Zhang et al.,
419 2015; Zhang et al., 2024). Consequently, $f(\text{RH})$ at a specific R_{eff} was generally higher



420 during the P2 period in comparison to that of P1 (Fig. 4c-d), also with high $f(\text{RH})$
421 levels observed for smaller size cases of $R_{\text{eff}} < 110$ nm under some extremely high
422 temperature conditions ($T > 40$ °C, as highlighted by the red dashed circle in Fig. 4d).
423 The higher SOC/OC on P2 non-event days further demonstrated the stronger
424 secondary aerosol formation in comparison to P1 non-event days (Fig. S2b).



425
426 **Figure 4.** Diurnal variations of (a) the number fraction (NF_{Acc}) and (b) volume
427 fraction of accumulation mode particles (VF_{Acc}) on P1 (red) and P2 (blue) NPF event
428 days (solid line), as well as non-event days (dash line). The relationship of $f(\text{RH})$ with
429 R_{eff} and VF_{Acc} . (as indicated by the colored dots) on P1 (c) and P2 non-event days (d)
430 during the 08:00-22:00 time window.

431 Nevertheless, $f(\text{RH})$ was almost independent of the two parameters (i.e., R_{eff}
432 and VF_{Acc}) for NPF events (Fig. S11a1-a2). This is mainly due to the explosive
433 formation of ultrafine particles during NPF events, significantly altering aerosol size
434 distributions and inducing large fluctuations in the number and volume fractions of

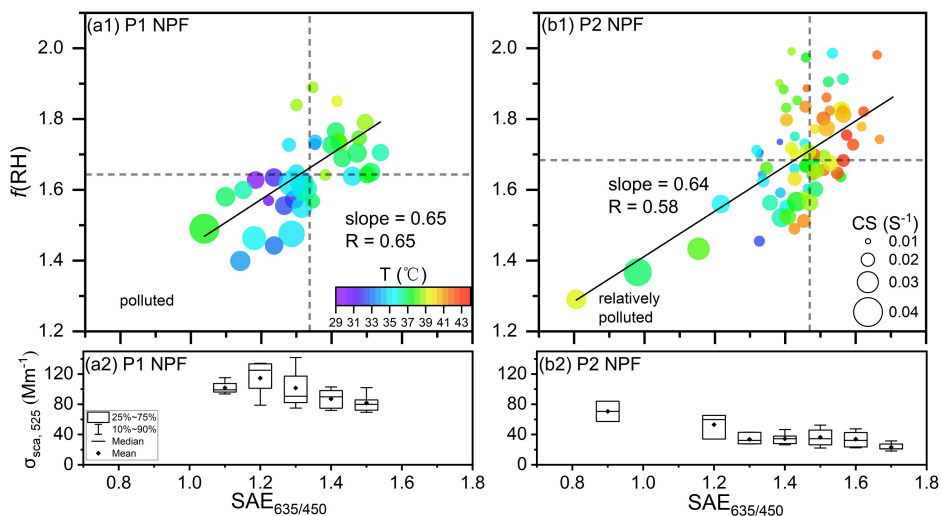


435 accumulation mode particles (Fig. 4a-b). Therefore, characterizing $f(\text{RH})$ with the
436 corresponding R_{eff} of aerosol populations was no longer applicable. Alternatively,
437 SAE was commonly used to estimate or parameterize $f(\text{RH})$ (Titos et al., 2014; Xia et
438 al., 2023; Xue et al., 2022), in line with the similar diurnal patterns of $f(\text{RH})$ and SAE
439 observed in this study. Figure 5 demonstrated a significantly positive correlation
440 between $f(\text{RH})$ and SAE during NPF events, with a similar slope of approximately
441 0.65 suggesting the consistent variation of $f(\text{RH})$ with SAE across both periods. As
442 larger particles contributed higher to the aerosol volume concentrations (Fig. S3), the
443 decrease of SAE also corresponded to an increase in $\sigma_{\text{sca}, 525}$ (Fig. 5a2, b2). In this
444 sense, $f(\text{RH})$ increased with SAE whereas decreased with $\sigma_{\text{sca}, 525}$, or rather the
445 pollution level during NPF events. Meanwhile, the cleaner environment of P2 period
446 generally possessed a lower CS (Table S2, as denoted by the size of circles in Fig. 5),
447 thereby in favor of the occurrence of NPF event. Such a positive (negative)
448 correlation of $f(\text{RH})$ with SAE (CS) was more pronounced in heatwave-induced high
449 temperature days during P2 period. The possible reasons can be attributed to the
450 following two aspects. One is related to the relatively smaller aerosol R_{eff} (with a
451 larger SAE) due to the lower GR, likely influenced by the evaporation of
452 newly-formed unstable clusters and particle coatings under heatwaves (Bousiotis et al.,
453 2021; Cusack et al., 2013; Deng et al., 2020) during the subsequent growth of aerosols.
454 Secondly, the higher temperature was normally associated with stronger
455 photochemical oxidation, which could intensify the formation of secondary aerosol
456 components with a higher hygroscopicity (Asmi et al., 2010; Gu et al., 2023; Liu et al.,
457 2014; Wu et al., 2016; R. Zhang et al., 2015; Zhang et al., 2024). This is further
458 supported by the relatively higher levels of UVB (P1: $2.6 \pm 1.9 \text{ W} \cdot \text{m}^{-2}$ versus P2: 2.7
459 $\pm 2.0 \text{ W} \cdot \text{m}^{-2}$) and O_3/O_x (P1: 0.81 ± 0.17 versus P2: 0.82 ± 0.17) during P2 heatwave
460 days, also in line with a recent study which demonstrated that heatwaves affected
461 secondary organic aerosols (SOA) formation and aging by accelerating
462 photooxidation in Beijing (Zhang et al., 2024).

463 It is worth noting that $f(\text{RH})$ did not show a consistently higher level after the
464 NPF occurrence during P2 period, and it was slightly higher within the first few hours



465 of NPF occurrence during P1 NPF events (Fig. 3b). In fact, aerosol optical
466 hygroscopicity not fully corresponds to the bulk hygroscopicity primarily determined
467 by aerosol chemical components, and the variability in aerosol optical features also
468 plays a key role in $f(\text{RH})$. In this sense, the size-dependency of aerosol optical
469 properties should be considered. The size-resolved $\sigma_{\text{sca}, 525}$ distribution and
470 size-resolved cumulative frequency distribution (CFD) of $\sigma_{\text{sca}, 525}$ over different NPF
471 events were calculated using the Mie theory, with good agreements between the
472 theoretically calculated and measured $\sigma_{\text{sca}, 525}$ values ($R^2 = 0.99$). As shown in Fig. S7
473 and Fig. S9, new particles must grow into the accumulation mode size at least before
474 they can exert a significant influence on the total scattering coefficient. The critical
475 sizes corresponding to the cumulative frequency of 50% in $\sigma_{\text{sca}, 525}$ were 358.7 nm and
476 333.8 nm on P1 and P2 NPF event days, respectively. This indicates that relatively
477 smaller particles contributed a slightly higher portion to $\sigma_{\text{sca}, 525}$ during P2 NPF events,
478 while the $\sigma_{\text{sca}, 525}$ of P1 NPF events was mainly contributed by larger particles.
479 Nevertheless, the Mie theory suggests that these smaller particles generally have a
480 weaker enhancement in total scattering after hygroscopic growth, in comparison to
481 larger size particles (Collaud Coen et al., 2007, Fig. S7). Consequently, the changes in
482 aerosol optical and hygroscopic properties necessitate consideration of both aerosol
483 optical and chemical characteristics during different NPF events. The contribution of
484 newly formed ultrafine particles to aerosol optical properties was insignificant within
485 the first few hours of NPF occurrence, leading to a reduced enhancement in aerosol
486 light scattering as characterized by a smaller R_{eff} during P2 NPF events in comparison
487 to P1 NPF events. In contrast, the growth of pre-existing and newly formed particles
488 into larger sizes would subsequently affect bulk aerosol optical properties, which was
489 evidenced by the enhancement in aerosol extinction coefficient observed after NPF
490 occurrence in a recent study (Sun et al., 2024). Specifically, particles could undergo a
491 longer and more intensified photochemical aging process during P2 NPF events as
492 influenced by persistent heatwaves, which facilitated the secondary formation of
493 hygroscopic aerosols and resulted in a higher $f(\text{RH})$ after 15:00 (Fig. 3b).



494

495 **Figure 5. (a1)** The relationship between $f(\text{RH})$ and $\text{SAE}_{635/450}$, as well as temperature
496 (as indicated by the color of dots) and CS (as denoted by the size of circles), on P1
497 NPF event days during the 08:00-22:00 time window. The vertical (horizontal) dash
498 line represents the median value of $\text{SAE}_{635/450}$ ($f(\text{RH})$). **(a2)** The corresponding $\sigma_{\text{sea}, 525}$
499 under different $\text{SAE}_{635/450}$ levels on P1 NPF event days. **(b1-b2)** The same but for P2
500 NPF event days.



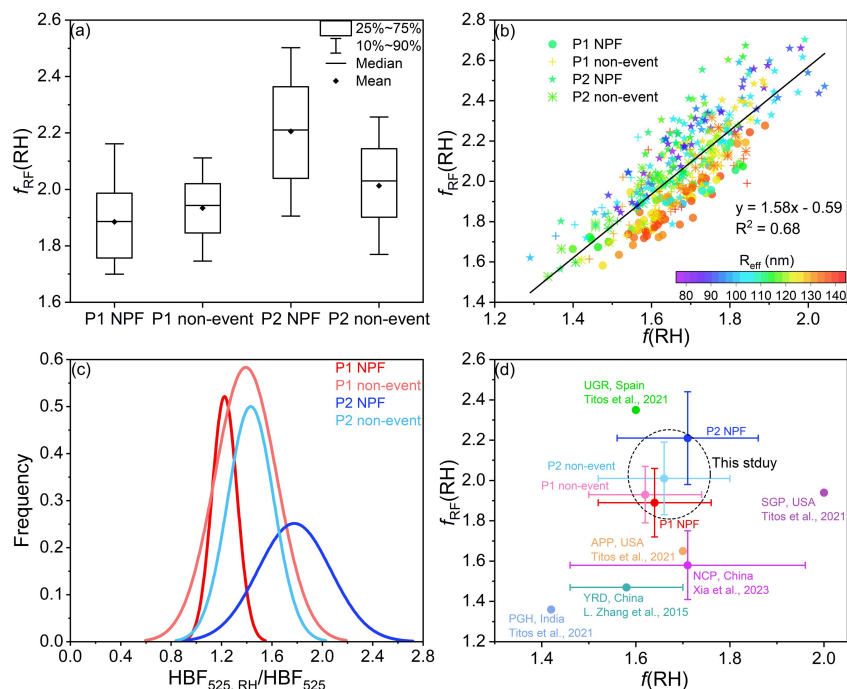
501 **3.5 $f(\text{RH})$ -induced changes in aerosol direct radiative forcing**

502 The changes in $f(\text{RH})$ have significant implications for aerosol direct radiative
503 forcing. Despite considerably lower $\sigma_{\text{sca}, 525}$ results during heatwaves, the
504 corresponding mean $f_{\text{RF}}(\text{RH})$ levels particularly for P2 NPF event days were higher
505 than that of the P1 cases (Fig. 6a). A robust positive correlation ($R^2 = 0.68$) was
506 observed between $f(\text{RH})$ and aerosol radiative forcing enhancement factor, $f_{\text{RF}}(\text{RH})$
507 (Fig. 6b). This is likely attributed to the enhanced $f_{\text{RF}}(\text{RH})$ with the larger forward
508 scattering ratio β , or rather higher HBF for smaller particle sizes, as supported by a
509 generally negative correlation between $f_{\text{RF}}(\text{RH})$ and R_{eff} . Specifically, the highest
510 $f_{\text{RF}}(\text{RH})$ value of 2.2 ± 0.2 was observed on P2 NPF event days, characterized with the
511 highest $f(\text{RH})$ and smallest R_{eff} (i.e., highest HBF) of the entire study period.

512 The definition of $f_{\text{RF}}(\text{RH})$ in Eq.(5) implies the dependences of $f_{\text{RF}}(\text{RH})$ on both
513 $f(\text{RH})$ and HBF-derived $\beta(\text{RH})$ and $\beta(\text{dry})$, or rather the ratio of $\text{HBF}_{525, \text{RH}}/\text{HBF}_{525}$.
514 The mean $\text{HBF}_{525, \text{RH}}$ was generally larger than HBF_{525} in this study, specifically with
515 the $\text{HBF}_{525, \text{RH}}/\text{HBF}_{525}$ ratios centered around 1.8 and even approached 2.5 on P2 NPF
516 event days (Fig. 6c, Table S2). This could be different from the classical Mie theory
517 with the spherical-particle premise, i.e., the observed light backscattering was
518 enhanced after hydration likely resulted from the evolution in particle morphology
519 that significantly influences their optical properties (Mishchenko 2009). The
520 organic-rich particles might remain non-spherical even after water uptake due to the
521 efficient evaporation of organic coatings under extremely hot weather conditions, as
522 evidenced by a recent study that high temperature and RH conditions could accelerate
523 the evaporation rate of SOA (Li et al., 2019). Meanwhile, the backward scattering
524 intensity of non-spherical particles is suggested to be much larger than its spherical
525 counterparts at scattering angles between 90° and 150° (Mishchenko 2009; Yang et al.,
526 2007). Furthermore, ultrafine particles would significantly contribute to both total
527 light scattering and backscattering coefficients (Fig. S7) after hygroscopic growth, if
528 the aerosol population was large enough (e.g., during NPF processes). These
529 combined effects could potentially change particle morphology and optical properties



530 (e.g., elevated the $\text{HBF}_{525, \text{RH}}$) particularly during heatwave-influenced NPF events,
 531 characterized with the smallest aerosol R_{eff} ($102.8 \pm 12.4 \text{ nm}$), lowest number fraction
 532 of accumulation mode particles (0.20 ± 0.10), and a higher SOC/OC ratio. The higher
 533 $\text{HBF}_{525, \text{RH}}/\text{HBF}_{525}$ ratios increased the HBF-derived $\beta(\text{RH})/\beta(\text{dry})$ levels, in
 534 combination of the elevated $f(\text{RH})$, further resulting in the highest $f_{\text{RF}}(\text{RH})$ observed
 535 during P2 NPF events. Given that previously observed $\text{HBF}_{525, \text{RH}}$ was typically lower
 536 than HBF_{525} (Titos et al., 2021; Xia et al., 2023; L. Zhang et al., 2015), the mean
 537 $f_{\text{RF}}(\text{RH})$ results of this study ($f_{\text{RF}}(85\%) = 2.0 \pm 0.2$) were significantly higher than
 538 those observed in the Yangtze River Delta ($f_{\text{RF}}(85\%) = 1.5$, L. Zhang et al., 2015), the
 539 North China Plain ($f_{\text{RF}}(80\%) = 1.6 \pm 0.2$, Xia et al., 2023), and some other regions in
 540 the world (Titos et al., 2021, Fig. 6d). It should be noted that the reported $f_{\text{RF}}(\text{RH})$ for
 541 the UGR site (Spain) was even higher, likely due to the relatively larger HBF in that
 542 area (Titos et al., 2014; 2021).



543
 544 **Figure 6.** (a) The box-plot of $f_{\text{RF}}(\text{RH})$ during P1 or P2 NPF event and non-event days.
 545 (b) The relationship between $f_{\text{RF}}(\text{RH})$ and $f(\text{RH})$, as colored by the corresponding R_{eff} ,
 546 during P1 or P2 NPF event and non-event days (shown in different symbols). (c)



547 Occurrence frequency of the ratio $\text{HBF}_{525, \text{RH}}/\text{HBF}_{525}$ during P1 or P2 NPF event and
548 non-event days. **(d)** The mean $f_{\text{RF}}(\text{RH})$ under different $f(\text{RH})$ levels (the error bars
549 stand for \pm one standard deviations corresponding to $f_{\text{RF}}(\text{RH})$ and $f(\text{RH})$, respectively),
550 along with the reported $f_{\text{RF}}(\text{RH})$ and $f(\text{RH})$ data for other regions in the world.

551 A recent study has indicated that continuous reduction of $\text{PM}_{2.5}$ mass loadings
552 can increase the net solar radiation, thereby promoting NPF events (Zhao et al., 2021).
553 Given the complexity and dynamic evolution of the atmospheric environment, these
554 can further alter the intrinsic properties of aerosol particles (e.g., $f(\text{RH})$, HBF,
555 morphology), potentially feeding back into aerosol-radiation interactions. Our
556 findings suggest that NPF and growth events may elevate aerosol optical
557 hygroscopicity in rather hot environments, e.g., the Basin area and tropical regions.
558 Meanwhile, NPF serves as a crucial secondary transformation process in the
559 atmosphere (Zhu et al., 2021). The favorable atmospheric diffusion capability ensured
560 the mixing of newly formed particles into the upper boundary layer, where is colder
561 and more humid than that near the surface during heatwaves (Jin et al., 2022). Hence,
562 the enhancement of aerosol optical hygroscopicity during the subsequent growth of
563 pre-existing and newly formed particles possibly exacerbates secondary pollution and
564 even triggers haze events (Hao et al., 2024; Kulmala et al., 2021). On the other hand,
565 the new particles of higher hygroscopicity could contribute more to the activation of
566 CCN, thereby modulating the aerosol-cloud interactions and further the global climate
567 (Ren et al., 2021; Sun et al., 2024; Wu et al., 2015). Additionally, the simultaneous
568 decrease in aerosol effective radius and possibly evaporation-induced non-spherical
569 particle morphology further enhance the aerosol direct radiative forcing enhancement
570 factor, potentially amplifying the cooling effect mainly caused by light scattering
571 aerosols. This highlights the needs for further in-depth exploration on aerosol
572 radiative impacts at weather extremes (e.g., heatwaves) with the changing climate,
573 given the continuous reductions of anthropogenic emissions and more intense
574 emissions of biogenic origins with the global warming. Besides, more detailed
575 information on the evolution of particle morphology with the changing environment
576 (e.g., varied temperature and RH) would enrich insights into the aerosol radiative



577 forcing.

578

579 **4 Conclusions and implications**

580 NPF events frequently occurred in urban Chongqing of southwest China in the
581 summer of 2022, accompanied with continuous heatwaves. Concurrent measurements
582 of aerosol optical and hygroscopic properties, PNSD, and bulk chemical compositions
583 were conducted to elucidate the mechanisms behind the variations in aerosol optical
584 hygroscopicity during different NPF event and non-event days.

585 NPF events exhibited distinct characteristics during the normally hot (P1,
586 relatively polluted) and heatwaves-dominated (P2, quite clean) periods. NPF within
587 P1 period was favored by the decrease in background aerosol loading and the higher
588 abundance of H₂SO₄. NPF events that occurred during the heatwave P2 period were
589 characterized with relatively lower CS, CoagS, and GR, as well as a smaller R_{eff}, than
590 P1 NPF cases. In comparison to the P1 NPF events, heatwaves initiated NPF earlier
591 and prolonged the subsequent growth during P2, likely intensifying the photochemical
592 oxidation due to heatwave-induced aging processes and modulating the evolution of
593 aerosol size distributions differently.

594 Heatwaves also significantly influenced the aerosol optical and hygroscopic
595 properties. Distinct diurnal patterns of $\sigma_{\text{sca}, 525}$ were observed for different types of
596 NPF events, with a minor $\sigma_{\text{sca}, 525}$ noontime peak occurred in P2 instead of peaked
597 earlier around the morning rush hours on P1 NPF event days. HBF and SAE were
598 significantly higher on P2 NPF event days, primarily due to the relatively smaller R_{eff}
599 for heatwave-influenced NPF cases. $f(\text{RH})$ remained relatively stable during the
600 daytime of NPF event days and peaked around 16:00-18:00, likely due to the
601 intensive photochemical reactions and accordingly enhanced formation of more
602 hygroscopic secondary aerosols. These secondary components could be more
603 abundant due to heatwave-induced stronger photooxidation, further resulting in a
604 higher $f(\text{RH})$ particularly during the subsequent growth of pre-existing particles and



605 newly formed ultrafine ones during P2 NPF events in comparison to that of P1 NPF
606 cases.

607 Compared with non-event cases, the generally higher levels of daily mean $f(\text{RH})$
608 suggested that the aerosol optical hygroscopicity was enhanced during NPF events in
609 hot summer of urban Chongqing. A significantly positive (negative) correlation
610 between $f(\text{RH})$ and SAE (CS, $\sigma_{\text{sca}, 525}$, or rather the pollution level) was observed for
611 both periods, with a more pronounced correlation during heatwave-influenced NPF
612 events. The aerosol light scattering or volume concentration was mainly contributed
613 by the larger accumulation-mode particles, while more ultrafine particles dominated
614 the size distribution especially for the initial stage of heatwave-influenced NPF events,
615 further leading to a diminished aerosol scattering enhancement capability in
616 comparison to P1 NPF events.

617 Changes in $f(\text{RH})$ have significant implications for the aerosol direct radiative
618 forcing. A robust positive (negative) correlation existed between $f_{\text{RF}}(\text{RH})$ and $f(\text{RH})$
619 (R_{eff}). Despite a lower $\sigma_{\text{sca}, 525}$ during heatwaves, the corresponding mean $f_{\text{RF}}(\text{RH})$ was
620 relatively higher and the maximum value of 2.2 ± 0.2 was observed on P2 NPF event
621 days, associated with the highest $f(\text{RH})$ (1.7 ± 0.2), smallest R_{eff} (102.8 ± 12.4 nm),
622 and highest $\text{HBF}_{525, \text{RH}}/\text{HBF}_{525}$ ratios (1.8 ± 0.3). The above highlights that heatwaves
623 could influence the NPF and atmospheric processing (although with a decreased
624 aerosol effective radius likely due to evaporation-resulted non-spherical particle
625 morphology under persistently high temperature conditions), thereby enhancing
626 aerosol optical hygroscopic growth and potentially reducing the net solar radiation
627 directly especially in hot summer. Further explorations on detailed molecular
628 characterizations and aerosol radiative impacts including the aerosol-cloud
629 interactions of weather extremes (e.g., heatwaves) with the changing climate are
630 highly recommended.

631

632 **Data availability.** Data will be available upon request.

633



634 **Author contributions.** YH and PL: Methodology, Investigation, Data analysis,
635 Formal analysis, Visualization, Validation, Writing – original draft & editing. YG and
636 ZW: Methodology, Investigation, Formal analysis. MT, YC, HX and WH: Data
637 curation, Methodology. FW and YL: Investigation. YK: Methodology, Data analysis,
638 Writing – review & editing. JC: Conceptualization, Methodology, Funding acquisition,
639 Data curation, Writing – review & editing, Supervision.

640

641 **Competing interests.** The authors declare no competing financial interest.

642

643 **Acknowledgement.** This work was supported by the National Natural Science
644 Foundation of China (No. 42105075), Venture and Innovation Support Program
645 for Chongqing Overseas Returnees (No. cx2021021). We thank Biao Xue for the
646 technical support on the utilization and maintenance of the humidified
647 nephelometer system. We also thank Ziqian Wang for the TSP filter sample
648 collection and Jiawei Zhou for the corresponding offline chemical analysis.

649

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