

Synchronization of Source and Sink by Boundary Layer Evolution: A Key to New Particle Formation Under Varying Ozone Pollution

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Abstract. Atmospheric new particle formation (NPF) is a vital source of aerosol and cloud condensation nuclei, regulated by complex meteorological and chemical factors. Utilizing ground aerosol particle size distributions and vertical observations, this study employs a generalized additive model (GAM) and SHapley Additive exPlanations (SHAP) to quantitatively assess the marginal contributions of factors driving NPF under the NPF scenario and three non-NPF scenarios. We found that NPF depends on the synchronization of enhanced source strength and weakened sink intensity, a process controlled by planetary boundary layer evolution. Under the NPF scenario, the breakup of the inversion layer and entrainment of cleaner air aloft promote vertical mixing. This rapidly reduces the condensation sink (CS) while transporting ozone (O₃) to the surface, allowing precursor formation to coincide with a clean background, thus creating a favorable nucleation window. In contrast, nucleation is inhibited in non-NPF scenarios through distinct mechanisms: insufficient oxidation capacity (Non-O₃ scenario), source-sink desynchronization caused by stable stratification suppressing vertical exchange (Low-O₃ scenario), or rapid scavenging by high background particles (High-O₃ scenario). Correlation analysis and SHAP method corroborate the source-sink competition mechanism. Under the NPF scenario, nucleation (Nuc) mode significantly correlates with SO₂, with high temperature and sufficient precursors contributing positively to its predicted value. However, predicted Nuc values are dominated by background particles under the Low-O₃ scenario and negatively influenced by T and SO₂ under the Non-O₃ scenario, while under the High-O₃ scenario, pre-existing aerosols effectively offset precursor contributions.

1 Introduction

Atmospheric new particle formation (NPF) is one of the major sources of atmospheric aerosols (Golden et al., 2017; Jiang et al., 2021) and serves as a critical pathway for the formation of cloud condensation nuclei (CCN) (Matsui et al., 2011; Zhu et al., 2021). **As a result, it** not only significantly impacts air quality (Kulmala et al., 2021; Tang et al., 2021), but also further modulates cloud microphysical processes, radiative balance, and climate effect (Makkonen et al., 2012; Sebastian et al., 2021; Sullivan et al., 2018).

Previous studies generally suggest that the occurrence and development of NPF are jointly regulated by multiple factors. Among them, meteorological elements influence the accumulation or dilution of precursors through their influence on planetary boundary layer (PBL) structure, turbulence, and radiative conditions (Dai et al., 2017; Lai et al., 2022a; Watson et al., 2023). Meanwhile, the concentration and production rate of precursors determine the source strength, representing indispensable prerequisites for NPF (Jin et al., 2025; Wang et al., 2013). Concurrently, the condensation sink (CS), dominated by background aerosol particles, scavenges precursor vapors and newly formed clusters, thereby suppressing nucleation (Du et al., 2022; Liu et al., 2024; Shang et al., 2023).

Ozone (O₃), as a key oxidant for driving the formation of nucleation precursors (Lelieveld et al., 2008; Liao et al., 2009),

exhibits increasingly complex influences on the NPF process (Tiszenkel et al., 2023; Zhang et al., 2024) under the prominent composite air pollution conditions in China (Lyu et al., 2024; Wang et al., 2025; Zong et al., 2021). Although some studies argue that O₃ enhances atmospheric oxidation capacity and accelerates the formation of nucleation precursors (Liu et al., 2022; Tan et al., 2022), thereby promoting NPF, a growing number of observational evidence indicates that NPF does not necessarily occur even under high O₃ pollution conditions (Carnerero et al., 2019; Kanawade et al., 2011; Nie et al., 2014). Therefore, a purely chemical perspective is insufficient to fully explain the behavior of NPF under different O₃ backgrounds.

In fact, as a core driver of atmospheric physical processes, PBL plays an important role in both the variation of ground pollutant concentrations and the NPF process. Lai et al. (2022b) noted that NPF often initiates at the top of the boundary layer or within the residual layer, rather than just at the surface. With the development and lifting of the unstable daytime PBL, particles formed aloft can be entrained into the mixed layer and subsequently transported to the surface (Lampilahti et al., 2021; Nilsson et al., 2001; Wehner et al., 2010), but the intense vertical exchange can also effectively dilute near-surface pollutants (Su et al., 2018; Xia et al., 2022), collectively facilitating NPF. On the contrary, a stable boundary layer structure tends to suppress pollutant diffusion, leading to the accumulation of O₃ and PM_{2.5} near the surface (Ding et al., 2016; Huang et al., 2018; Su et al., 2022; Wang et al., 2018) and a significant enhancement of the background CS (Petäjä et al., 2016), which inhibits nucleation and particle growth. Therefore, the evolution of the boundary layer structure needs to be considered to have a more comprehensive understanding of NPF.

Although previous studies have revealed the influence of physical or chemical processes in the atmosphere on NPF, they are mostly limited to discussions of single processes and qualitative analyses. However, in the real atmosphere, precursors, meteorological elements, background aerosols, and PBL structure are always highly coupled and exhibit significant nonlinear characteristics (Fiore et al., 2015; Jacob and Winner, 2009; Li et al., 2017; Wang et al., 2015). Therefore, traditional correlation analysis is difficult to quantitatively describe the relative importance of different factors (Liu and Shi, 2021; Qi et al., 2024; Zheng et al., 2023), and even more difficult to explain their synergistic control mechanisms, which limits our in-depth understanding of NPF in complex atmospheric environments.

In recent years, interpretable machine learning methods have offered new methods for addressing the above challenges. Among them, the SHapley Additive exPlanations (SHAP) method has been gradually applied in the study of atmospheric and air pollution (Hou et al., 2025; Houdou et al., 2024; Ma et al., 2023; Yenikar et al., 2025), due to its ability to quantitatively analyze the marginal contribution of various input variables to model predictions (Lundberg et al., 2018; Lundberg and Lee, 2017). However, this method is still rarely applied to NPF research, especially regarding the question of whether NPF can occur under different O₃ pollution backgrounds.

To address this gap, this study integrates ground-based aerosol particle size distributions (PSD), vertical observation data, and background particles kinetic parameters to systematically compare the differences between NPF scenario and non-NPF scenarios under three different O₃ pollution backgrounds. By combining the GAM model with the SHAP method, we quantitatively resolve the contributions of different factors to the particle number concentration of the nucleation (Nuc) mode. The results demonstrate that NPF depends on whether the increase in sources and the reduction in sinks occur synchronously. This finding provides a new perspective for explaining the absence of NPF under high O₃ conditions, and is of great significance for understanding aerosol evolution mechanisms under composite pollution.

2 Data and Methods

2.1 Data Collection and Processing

The observation period for this study was from May 30 to June 15, 2018. Hourly surface air quality data (PM_{2.5}, PM₁₀, SO₂, NO₂, CO, and O₃) and meteorological elements (wind speed, wind direction, temperature (T), and relative humidity (RH)) were obtained from the Ministry of Ecology and Environment of China (MEE) (<https://www.mee.gov.cn/>) and the Nanjing

University of Information Science and Technology (NUIST) meteorological station, respectively. In addition, vertical observation data (O_3 , $PM_{2.5}$, and black carbon (BC)) and ground-based aerosol particle size distribution (5-minute resolution, 0.01–10 μm) were respectively collected by an unmanned aerial vehicle (UAV) observation platform and a WPS-1000 instrument at the Intelligent Manufacturing Industrial Park in the northern suburbs of Nanjing (Zhongshan Park; 32.35°N, 118.72°E). Detailed descriptions of the site environment and instrumental configurations can be found in Shi et al. (2022).

To ensure the reliability of the analysis, all observational data were first subjected to quality control. Subsequently, the ground-level meteorological elements and pollutant concentrations used for SHAP analysis were averaged hourly, while the time resolution of the vertical observation data was set to 3 hours to accurately reflect the atmospheric vertical structure during the study period.

2.2 NPF Event Identification and Pollution Scenario Classification

Based on previous studies (Dal Maso et al., 2005; Herrmann et al., 2014; Kerminen et al., 2018; Kulmala et al., 2004; Yue et al., 2011), NPF events in this study were identified according to the following three criteria: (1) A clear new particle mode appears in the particle number concentration size distribution, with an initial particle size within the nucleation (Nuc) mode range (10–20 nm); (2) This mode persists for at least 1 hour during the observation period; (3) The particle size of this mode continuously evolves towards larger sizes over time, accompanied by a continuous increase in the particle number concentration within the mode. Only when all the above conditions are met is it considered an NPF event, and that day is defined as an NPF day, whereas the remaining days were classified as non-NPF days.

Meanwhile, according to the *Ambient Air Quality Standards* (GB 3095–2012) and the *Technical Regulation on Ambient Air Quality Index (AQI)* (HJ 633–2012), the ozone pollution level during the observation period was classified using the maximum daily 8-hour average ozone concentration (MDA8- O_3). Specifically, days were defined as Non- O_3 (MDA8- O_3 < 160 $\mu g/m^3$), Low- O_3 (160 < MDA8- O_3 < 215 $\mu g/m^3$), and High- O_3 (MDA8- O_3 > 215 $\mu g/m^3$).

Finally, by combining the NPF event identification with the ozone pollution classification, the observation period was divided into four scenarios:

- (1) NPF (Low- O_3): 30 May, 3 June, and 11–13 June (hereafter NPF);
- (2) Non-NPF (Non- O_3): 31 May, 5 June, and 9–10 June (hereafter Non- O_3);
- (3) Non-NPF (~~Low- O_3~~) (Low- O_3): 1–2 June, 6–7 June, and 14–15 June (hereafter Low- O_3);
- (4) Non-NPF (~~High- O_3~~) (High- O_3): 4 June and 8 June (hereafter High- O_3).

2.3 Key Kinetic Parameters of NPF

The growth rate (GR, $nm \cdot h^{-1}$) is used to characterize the temporal evolution of the particle diameter of newly formed mode (Dal Maso et al., 2005; Kulmala et al., 2012):

$$GR = \frac{dD_p}{dt} = \frac{\Delta D_p}{\Delta t}$$

where D_p is the geometric mean diameter of the aerosol particles.

The formation rate (FR, $cm^{-3} \cdot s^{-1}$) represents the rate of change of the number concentration of Nuc mode particles, calculated following the method proposed by Kulmala et al. (2004), while neglecting the flux of particles growing beyond the 20 nm upper limit (Wang et al., 2013):

$$FR = \frac{dN_{nuc}}{dt} + Coags_{Nuc} \cdot N_{Nuc}$$

where N_{nuc} is the total number concentration within the Nuc mode, and $Coags_{Nuc}$ denotes the coagulation sink within the Nuc mode. And the coagulation sink (~~Coags- S^+~~) (Coags, s^{-1}) describes the loss rate of target particles due to coagulation

with background particles and is defined as (Kulmala et al., 2001a):

$$Coags(D_p) = \int K(D_p, D'_p) n(D'_p) dD'_p$$

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125 where $K(D_p, D'_p)$ is the coagulation coefficient proposed by Fuchs et al. (1965), $n(D'_p)$ is the number concentration distribution of background particles.

The condensation sink (CS, s^{-1}) is used to characterize the ability of background aerosols to absorb vapor molecules, and was calculated as (Dal Maso et al., 2005; Kulmala et al., 2001b):

$$CS = 2\pi D \sum_i \beta_i d_{p,i} N_i$$

130 where D is the diffusion coefficient of the vapor molecules, β_i is the transition regime correction factor, and $d_{p,i}$ and N_i are the diameter and number concentration of each size bin, respectively.

In addition, based on the empirical linear relationship between GR and sulfuric acid vapor concentration (Kulmala et al., 2001b; Nieminen et al., 2010), the condensable vapor concentration (C , cm^{-3}) was estimated using the following parameterization:

$$C = GR \cdot 1.37 \times 10^7$$

135 Under the steady-state assumption, the condensable vapor production rate (Q , $cm^{-3} \cdot s^{-1}$) can be further expressed as:

$$Q = CS \cdot C$$

2.4 SHAP Method

The SHAP (Shapley Additive exPlanations) method is a game-theory-based method for machine learning model interpretability, used to quantitatively assess the marginal contribution of each input feature to the model prediction (Lundberg and Lee, 2017; Shapley, 1953). Under the SHAP framework, the prediction for an individual sample can be expressed as the sum of a baseline value and the contributions of all features:

$$f(x) = \phi_0 + \sum_{i=1}^M \phi_i$$

145 where $f(x)$ denotes the model prediction for sample x , ϕ_0 is the model baseline value, M is the total number of features, and ϕ_i represents the SHAP value of the i -th feature. The sign of ϕ_i indicates whether the feature promotes or suppresses the prediction, while its absolute magnitude reflects the strength of its contribution.

150 Considering the complex nonlinear characteristics among multiple variables in the atmospheric system, this study utilizes generalized additive models (GAM) and the KernelSHAP algorithm (Aas et al., 2019; Bordt & Luxburg, 2022; Lundberg and Lee, 2017; Murase et al. 2009) to quantitatively assess the relative contributions of meteorological elements, pollutant concentrations, and background particle parameters to the changes in particle number concentrations of Nuc mode. Feature importance plots and SHAP summary plots are used to quantify the relative importance of each factor (Lundberg et al., 2020; Zhao et al., 2025), thereby intuitively identifying the dominant factors under different pollution scenarios. Furthermore, SHAP dependence plots are used to describe the average marginal impact of a single variable on model predictions (Lundberg and Lee, 2017; Wei et al., 2025; Yuan et al., 2025), and nonlinear fitting of the scatter distributions was further performed to elucidate the specific response relationships between the variable and the number concentrations of mode.

3.1 Source–Sink Control Mechanism of New Particle Formation

As shown in Fig. S1, the aerosol number concentration and its size distribution exhibited significant phased evolution characteristics during the observation period. For example, from 6 to 8 June, overall pollution levels were relatively high, with the 20–70 nm size range dominating the particle size distribution and persisting for an extended duration. The total particle number concentration in the 10–100 nm range (N_{10-100}) consistently remained at a high level and displayed a clear increasing trend, with peak values exceeding 10^5 cm^{-3} . Figures 1c–f show that the number concentrations of the four modes of particles also remained high, but there was no obvious enhancement. During this period, the CS level remained persistently high, with an average value of approximately 0.089 s^{-1} (Fig. 1g), and the concentrations of the six environmental factors were generally high but showed no pronounced fluctuations (Fig. S2). These features collectively indicate that aerosol evolution during this stage was predominantly controlled by background particles.

In contrast, during certain periods, the particle size distribution revealed clear growth processes from smaller to larger particle diameters (Fig. S1), lasting approximately 1–2 hours. During these episodes, particle number concentrations in the nucleation (Nuc) and Aitken (Ait) modes decreased sharply (Fig. 1c–d), whereas those in the accumulation (Acc) and coarse (Coa) modes exhibited a decrease followed by a subsequent increase (Fig. 1e–f), reflecting the rapid growth of newly formed particles into larger size modes. Simultaneously, lower CS level (Fig. 1g) and weak wind speeds (Fig. 1a) also provided favorable conditions for the survival and growth of newly formed particles.

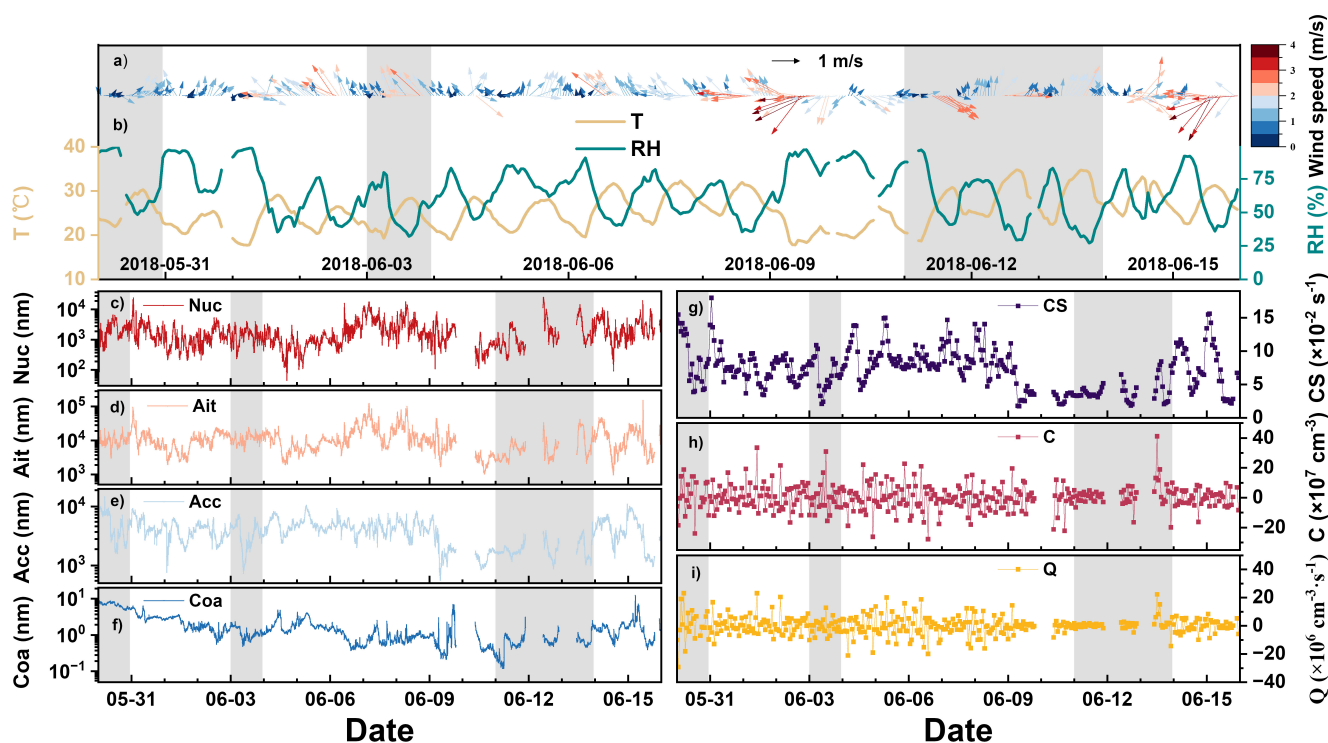


Figure 1. Time series of meteorological parameters, particle number concentrations in different modes, and NPF dynamic parameters from May 30 to June 15, 2018: (a) Wind vector, (b) T and RH, (c) N_{Nuc} , (d) N_{Ait} , (e) N_{Acc} , (f) N_{Coa} , (g) CS, (h) C, and (i) Q. The shaded areas indicate NPF days.

Based on the classification criteria in Sect. 2.2, a total of five NPF days were identified. Following the method of Birmili et al. (2003), this study determined the time range of each NPF process by using the moments when the particle number concentration in the Nuc mode (10–20 nm) rose and fell to $1/e$ of the daily peak value as the onset and termination of the event. Relevant parameters were then calculated based on the formulations described in Sect. 2.3 (Table 1). As shown in Table 1, the intensity and duration of the NPF process are not controlled by a single factor, instead, they are governed by the dynamic balance between precursor strength (source) and background particle removal capacity (sink).

Taking June 13th as an example, although both CS ($6.20 \times 10^{-2} \text{ s}^{-1}$) and Cogas Coags ($18.74 \times 10^{-5} \text{ s}^{-1}$) reached the highest levels among the five events, meaning that newly formed particles and precursors would be strongly removed, which is theoretically unfavorable for nucleation. However, C and Q reached high levels of $7.95 \times 10^7 \text{ cm}^{-3}$ and $3.81 \times 10^6 \text{ cm}^{-3} \cdot \text{s}^{-1}$, respectively, favoring precursor formation. As a result, the FR reached its maximum value of $0.71 \text{ cm}^{-3} \cdot \text{s}^{-1}$, indicating that an extremely strong source can effectively offset the removal effect of a high sink. In contrast, on 3 June, the FR was only $0.03 \text{ cm}^{-3} \cdot \text{s}^{-1}$, suggesting slower nucleation initiation. However, both CS ($3.30 \times 10^{-2} \text{ s}^{-1}$) and CoagS ($6.56 \times 10^{-5} \text{ s}^{-1}$) were the lowest among the five events, the background removal effect was weak, and the final particle growth rate far exceeded that of other events ($\text{GR} = 5.99 \text{ nm} \cdot \text{h}^{-1}$). This comparison clearly demonstrates that the sources and sinks are closely related in the NPF process: a strong source can compensate for the removal effect of a high sink, while a weak sink facilitates sustained particle growth even under conditions of limited precursor supply.

Table 1. Summary of key kinetic parameters for the identified NPF events.

Date	FR ($\text{cm}^{-3} \cdot \text{s}^{-1}$)	GR ($\text{nm} \cdot \text{h}^{-1}$)	C ($\times 10^7 \text{ cm}^{-3}$)	Q ($\times 10^6 \text{ cm}^{-3} \cdot \text{s}^{-1}$)	CS ($\times 10^{-2} \text{ s}^{-1}$)	Cogas Coags ($\times 10^{-5} \text{ s}^{-1}$)
2018.05.30	0.10	1.73	2.37	1.88	5.61	8.90
2018.06.03	0.03	5.99	8.21	2.73	3.30	6.56
2018.06.11	0.05	1.11	1.53	0.63	3.43	6.07
2018.06.12	0.25	0.38	0.52	0.51	4.77	9.80
2018.06.13	0.71	5.80	7.95	3.81	6.20	18.74

However, the analysis above only reveals some of the parametric characteristics of NPF, and the key mechanisms determining whether NPF occurs under different O_3 pollution backgrounds still need to be explored. To further elucidate these differences, we compared the evolution of particle size distributions across different scenarios. And we found that the NPF scenario exhibited a typical new particle formation signal, with Nuc mode particles significantly increasing around 10:00 and continuously growing to larger sizes (Fig. S3a). In contrast, neither the Non- O_3 nor the High- O_3 scenarios showed clear nucleation characteristics (Fig. S3b-c), but the reasons differed substantially between the two cases.

Under the Non- O_3 scenario, the particle size distribution was relatively disordered, and there was no significant growth process of Nuc mode particles (Fig. S3b). Although the relatively low background particle concentrations (Fig. 2) and CS levels (Fig. S5c) would, in principle, favor new particle formation, the high humidity environment created by the daily average RH of 80.1% (Fig. S4b) may also trigger hygroscopic growth effects, thereby exacerbating the scavenging of newly formed clusters by background particles (Molnár et al., 2020; Zang et al., 2019). However, this is unlikely to be the dominant factor for the absence of NPF in this scenario. Instead, the consistently low O_3 concentration (Fig. S4d) and T (Fig. S4a) throughout the day lead to weak photochemical reaction activity. Meanwhile, SO_2 supply was extremely limited: both its mean concentration and temporal increase were the lowest among the four scenarios (Fig. S4h). Under the combined constraints of weak atmospheric oxidation capacity and insufficient precursor accumulation, the source strength was severely limited, as reflected by persistently low values of C (Fig. S5a) and Q (Fig. S5b). Therefore, we infer that the weak source is the fundamental reason for the absence of NPF under the Non- O_3 scenario.

Unlike the Non- O_3 scenario, the High- O_3 scenario exhibited extremely high background particle concentrations, especially from 0:00 to 9:00. However, during the afternoon period with elevated O_3 levels, NPF still did not occur; instead, background particle concentrations even showed a slight decline (Fig. S3d). During the critical nucleation period (08:00-12:00), N_{Acc} remained persistently high (Fig. 2c), resulting in drastic fluctuations in CS with an average value as high as $9.27 \times 10^{-2} \text{ s}^{-1}$, approximately twice that of the NPF scenario ($4.69 \times 10^{-2} \text{ s}^{-1}$) (Fig. S5c). Although elevated O_3 levels in the afternoon could enhance precursor formation (Fig. S4d), as evidenced by strong fluctuations in Q (Fig. S5b), the extremely strong background CS led to rapid uptake of newly generated precursor by pre-existing coarse particles. As a result, C also exhibits unstable fluctuations (Fig. S5a) because it failed to accumulate. Thus, under the High- O_3 scenario, a strong sink can suppress NPF by efficiently removing precursors, despite favorable photochemical conditions.

In contrast, during certain periods, the particle size distribution revealed clear growth processes from smaller to larger particle diameters (Fig. S1), with approximately 1-2 hours.

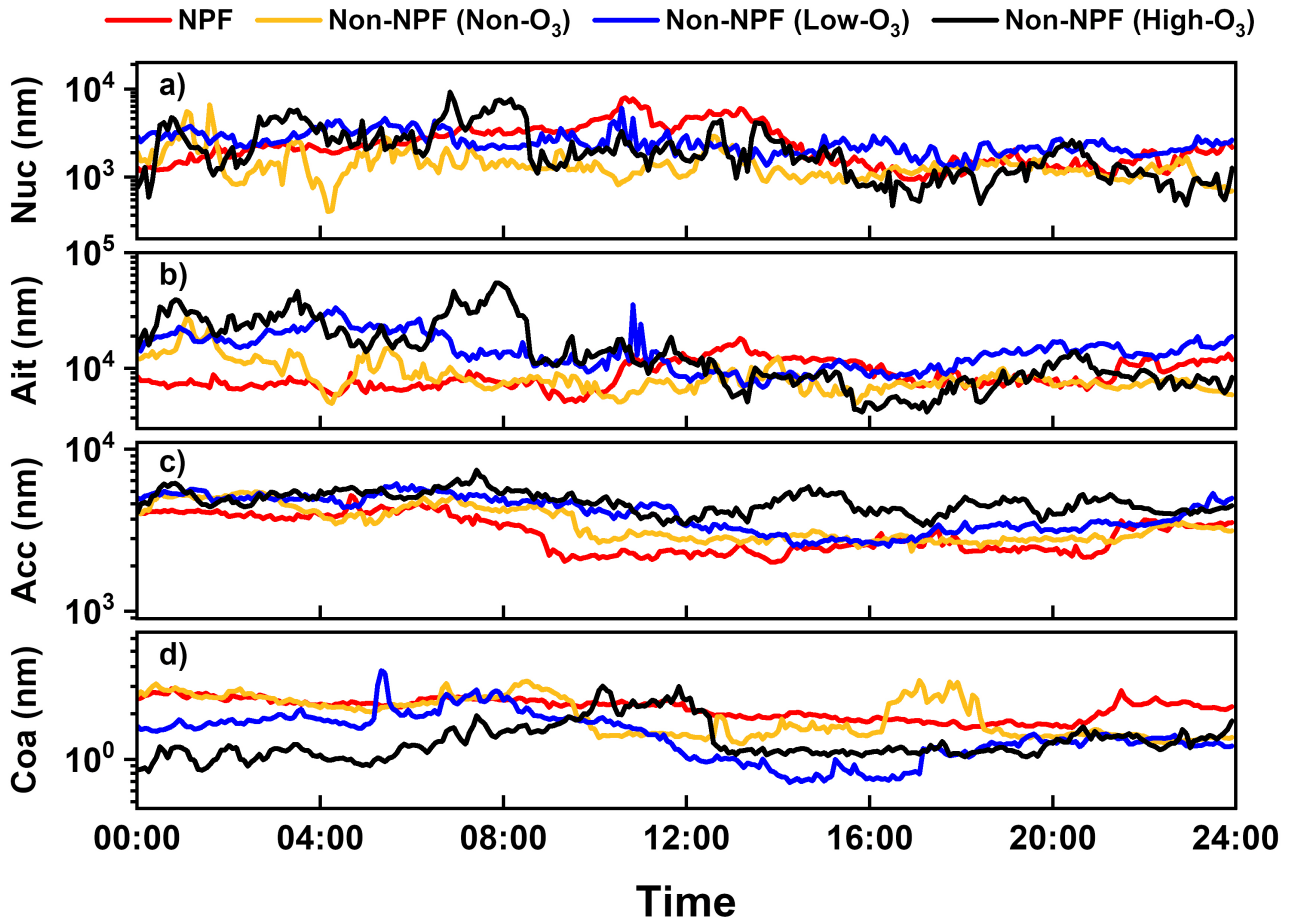


Figure 2. Diurnal variations of particle number concentrations in different modes under four classified scenarios: (a) N_{Nuc} , (b) N_{Ait} , (c) N_{Acc} , (d) N_{Coa} . The red, yellow, blue, and black lines represent the NPF, Non-NPF (Non- O_3), Non-NPF (Low- O_3) and Non-NPF (High- O_3) scenarios, respectively.

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3.2 Regulation of Source–Sink Terms by Boundary Layer Structure under Similar O_3 Backgrounds

According to the classification criteria in Sect. 2.2, both the NPF and Low- O_3 scenarios had similarly low O_3 pollution levels ($160\text{--}215 \mu\text{g}/\text{m}^3$). However, NPF occurred only in the former, whereas the latter exhibited a slight increasing trend in Ait mode particle concentrations (Fig. S3c). To further explain the reasons for this contrast, we compared the spatiotemporal evolution of boundary layer structure under the two scenarios.

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Under the NPF scenario, the near-surface air temperature decreased with height at 05:00, while an inversion layer existed at approximately 900 m (Fig. S6), implying that convective processes could respond rapidly and be initiated after sunrise (Lapworth, 2006). With increasing solar radiation, rapid surface warming (Fig. S4b) promoted the development of turbulence (Kaimal et al., 1976; Li et al., 2026), especially beginning around 08:00, the rapid lifting of the planetary boundary layer established efficient pathways for vertical exchange (Neu et al., 1994; Stull, 2012). On the one hand, cleaner air aloft was entrained into the mixed layer, rapidly diluting aerosols in the lower layers (Nilsson et al., 2001). This was particularly evident below 300 m, where both $\text{PM}_{2.5}$ (Fig. 3) and BC (Fig. 4) concentrations decreased sharply with height. Near the surface, this process manifested as a pronounced decline in N_{Acc} starting around 07:00 (Fig. 2c), leading to a simultaneous decrease in CS (Fig. S5c) (Korhonen et al., 2004; Lehtinen et al., 2003). In addition, the low RH conditions (Fig. S4b) likely suppressed particle hygroscopic growth, further weakening the CS level (Birmili and Wiedensohler, 2000; Hamed et al., 2007). On the other hand, the inversion layer near 900 m was broken (Fig. S6), accompanied by a weakening of the vertical gradient in the O_3 profile within the lower layers (Fig. S9), indicating downward transport of O_3 aloft (Song et al., 2024), which led to

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increased near-surface O₃ concentrations (Fig. S4d) and more favorable conditions for nucleation (Jonsson et al., 2007). Crucially, the process of weakened sinks and enhanced atmospheric oxidation capacity driven by the boundary layer coincided temporally with the increase in SO₂ concentrations between 07:00 and 09:00 (Fig. S4h). Therefore, the precursors were able to survive and accumulate, ultimately triggering nucleation and sustaining growth.

In contrast, the Low-O₃ scenario was controlled by a stable boundary layer before sunrise. At 05:00, a pronounced inversion layer was present within the 100–150 m layer (Fig. S6), accompanied by high relative humidity (Fig. S7) and weak wind conditions (Fig. S8), collectively establishing a highly stable boundary layer structure. This strongly suppressed the development of turbulent mixing after sunrise, and vertical exchange between the lower and upper layers was persistently limited. As a result, near-surface pollutants were not effectively diluted by cleaner air aloft, and PM_{2.5} and BC concentrations remained elevated near the surface (Figs. 3–4), while N_{Acc} showed no significant change (Fig. 2c). Correspondingly, the CS level did not weaken; instead, it increased rapidly around 10:00 (Fig. S5c), further intensifying the sink strength. This enhancement was likely associated with persistently high near-surface RH (Fig. S4b), which promoted hygroscopic growth of background particles and thereby increased CS (~~Ali et al., 2025~~) (Ali et al., 2025). Meanwhile, although SO₂ concentrations showed an increasing trend under this scenario (Fig. S4h), both the magnitude of the increase and the peak values were substantially lower than those observed under the NPF scenario, indicating limited precursor accumulation. More importantly, when SO₂ increased, CS did not show a corresponding decrease but instead remained at a high level throughout the critical period. Therefore, we conclude that the asynchrony between source enhancement and sink weakening effectively suppressed the occurrence of NPF under the Low-O₃ scenario.

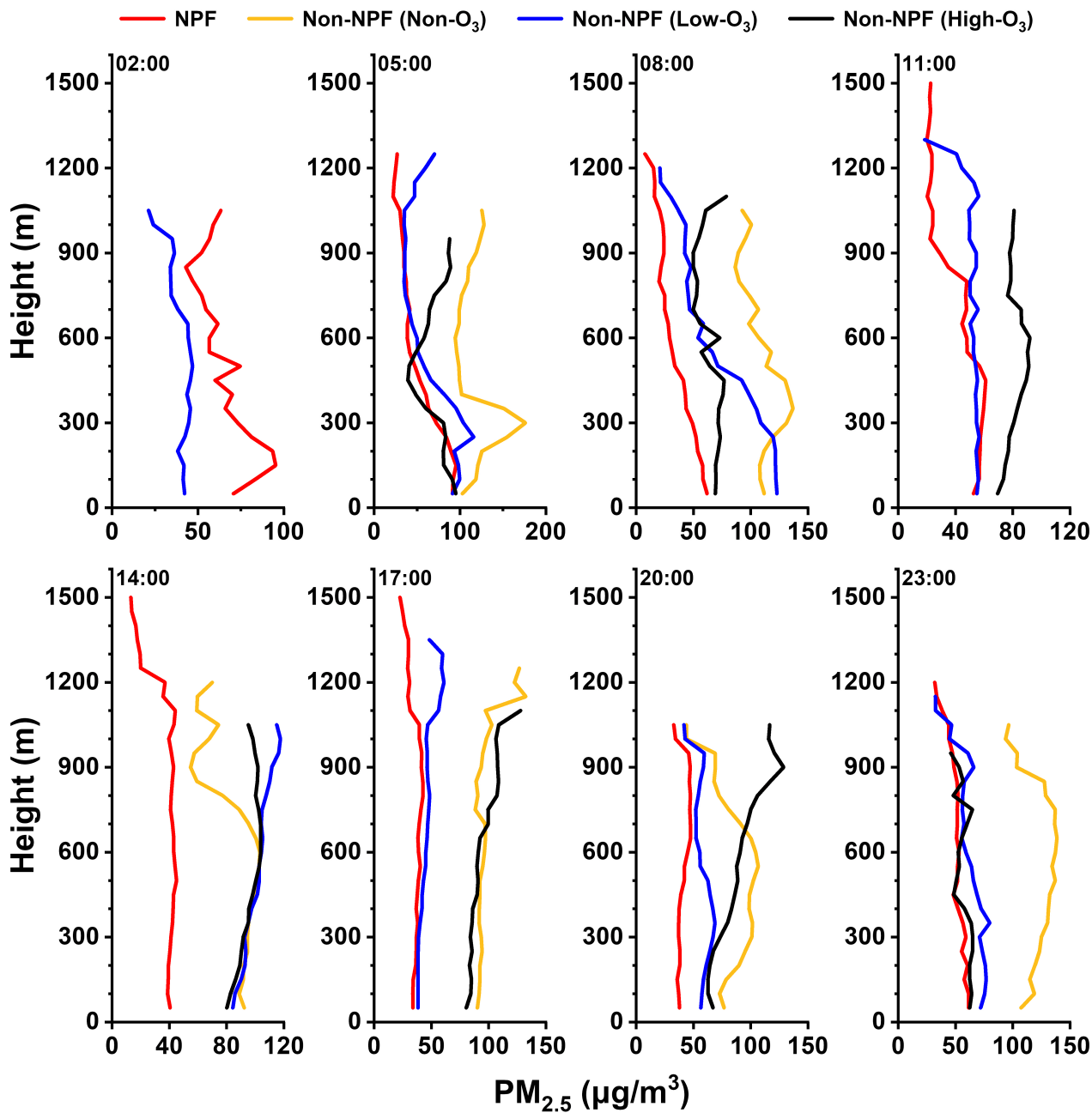
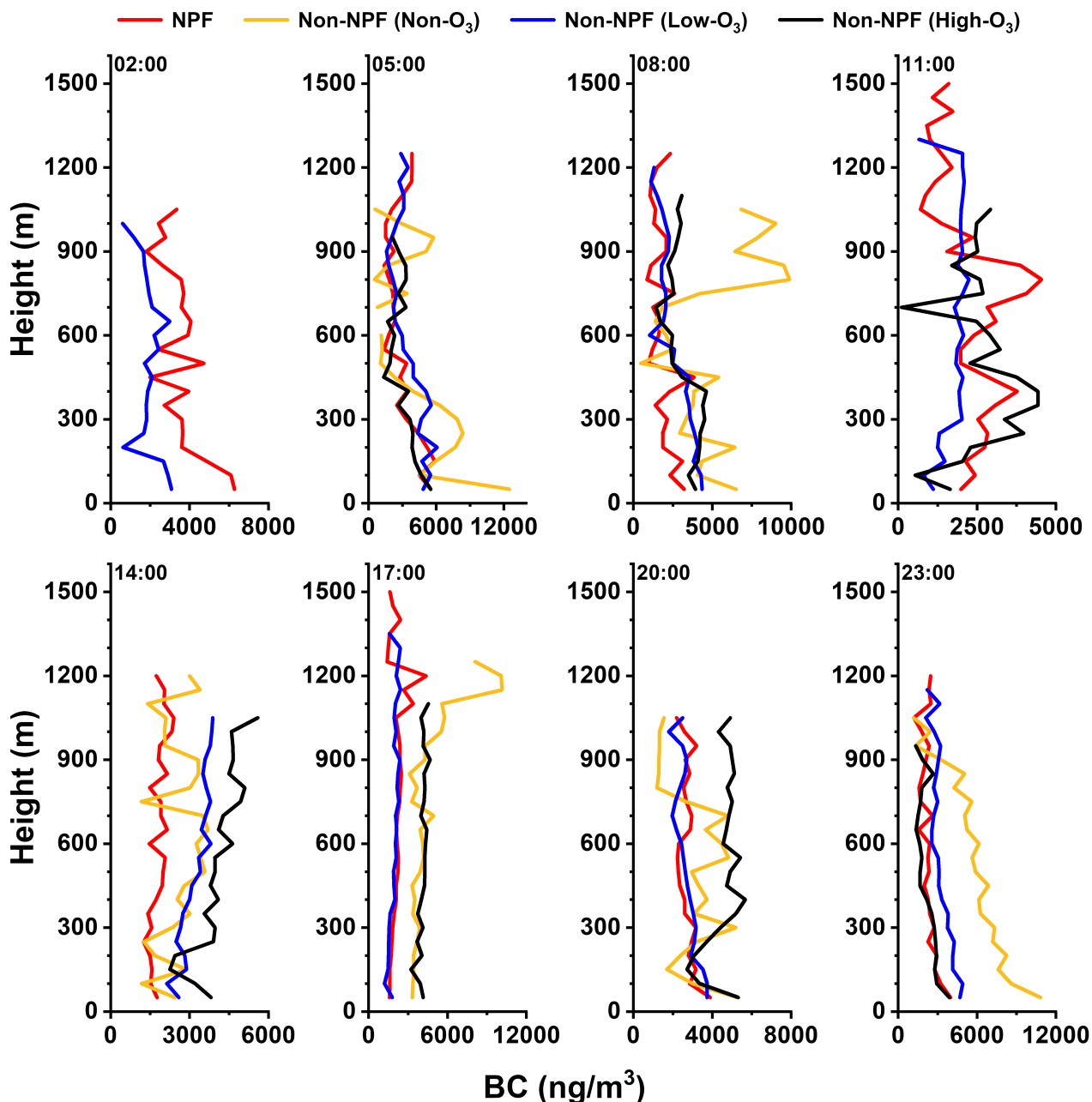


Figure 3. Vertical profiles of $PM_{2.5}$ concentrations at different times under four classified scenarios. The red, yellow, blue, and black lines represent the NPF, Non-NPF (Non- O_3), Non-NPF (Low- O_3) and Non-NPF (High- O_3) scenarios, respectively.



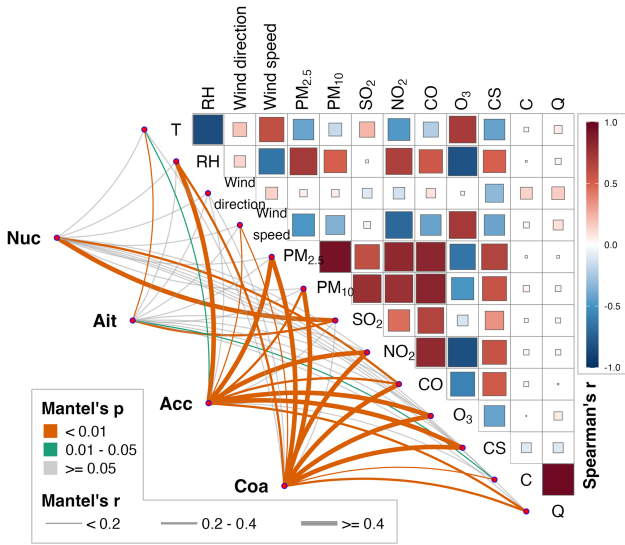
265 **Figure 4.** Vertical profiles of BC concentrations at different times under four classified scenarios. The red, yellow, blue, and black lines represent the NPF, Non-NPF (Non-O₃), Non-NPF (Low-O₃) and Non-NPF (High-O₃) scenarios, respectively.

3.3 Comparison of Nucleation Drivers Based on Correlation Analysis and SHAP Method

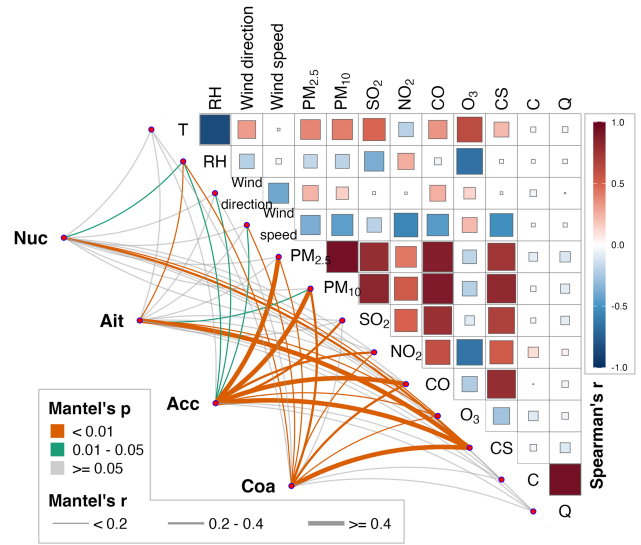
As shown in Fig. 5, the NPF scenario exhibits the most complex and densely connected correlation network (Fig. 5a). Notably, a significant and strong correlation between the Nuc mode particle concentration and SO₂ is observed, which is the
 270 only case among the four scenarios where the Nuc mode is directly coupled with a precursor source, indicating that sufficient precursors are required for new particle formation. Meanwhile, both the Acc and Coa modes show significant correlations with multiple atmospheric pollutants, indicating that particle evolution is jointly influenced by background particles. For the Non-O₃ scenario (Fig. 5b), the correlations are mainly concentrated in the Acc mode, which shows a significant strong correlation with primary pollutants; while under the Low-O₃ scenario (Fig. 5c), the Coa mode is correlated not only with pollutants but
 275 also with meteorological elements. However, unlike the NPF scenario, in these two non-NPF scenarios, the Nuc mode is mainly significantly constrained by CS, and its correlations with source-related factors are markedly weakened. Under the High-O₃ scenario (Fig. 5d), the overall correlation network is further simplified. Only a strong significant correlation between

the Acc mode and CS remains evident, whereas correlations among other particle modes and atmospheric elements are generally weaker.

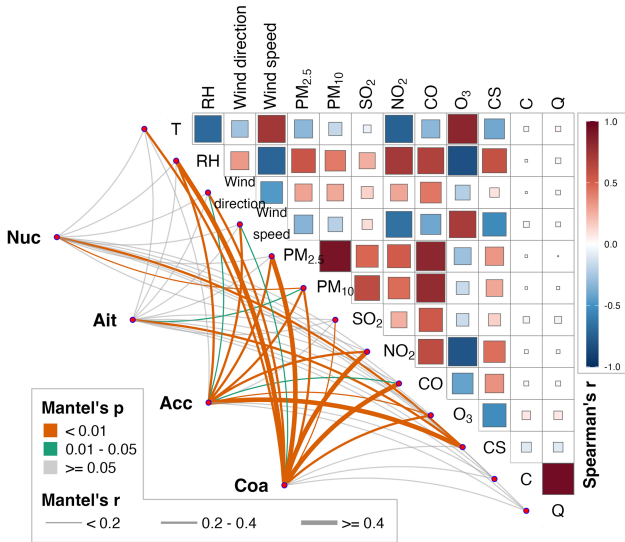
a) NPF



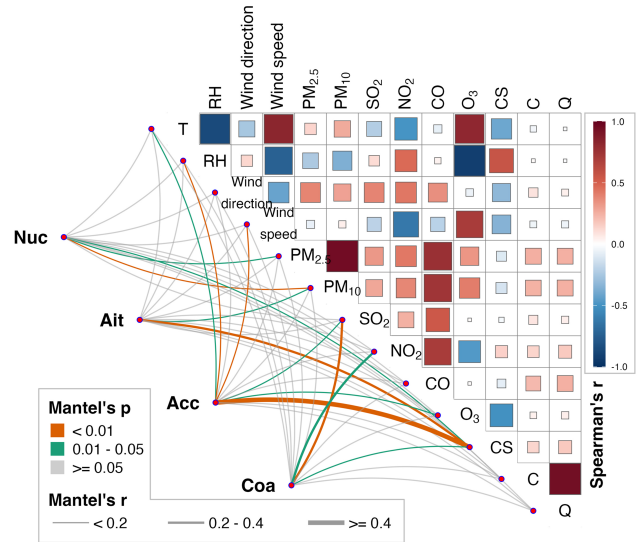
b) Non-NPF (Non-O₃)



c) Non-NPF (Low-O₃)



d) Non-NPF (High-O₃)



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Figure 5. Pairwise correlations of potential drivers and their linkage to particle number concentrations under four classified scenarios: (a) NPF, (b) Non-NPF (Non-O₃), (c) Non-NPF (Low-O₃) and (d) Non-NPF (High-O₃). The upper-right triangle shows Spearman correlations between variables, while the lower-left network illustrates the Mantel test linkages between drivers and particle modes.

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As the initial stage of the NPF process (Kulmala et al., 2004), the Nuc mode is crucial for understanding the underlying mechanisms governing NPF occurrence. However, due to the complex nonlinear interactions among multiple atmospheric variables, conventional correlation analysis alone is insufficient to disentangle these mechanisms. Therefore, this study introduces the generalized additive model (GAM) in combination with the SHAP method (described in Sect. 2.4) to identify the dominant drivers under different scenarios through feature importance and SHAP summary plots (Fig. 6), and to further

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reveal the nonlinear response characteristics of these factors to Nuc mode number concentration through SHAP dependence. Under the NPF scenario, the mean SHAP values of T and SO₂ are 42.9 and 25.2, respectively (Fig. 6a), indicating that both variables generally have a positive impact on the predicted Nuc values. Their importance ranks among the highest across all variables, accounting for 5.5% and 7.3%, respectively (Fig. 6b). Scatter points corresponding to high values (red) are predominantly distributed on the positive SHAP side, whereas low-value samples (blue) mainly correspond to negative SHAP

295 values (Fig. 6b), suggesting a positive association between these two variables and predicted Nuc values. This relationship is further confirmed by the dependence plots, which show a pronounced increase in SHAP values when $T > 28\text{ }^{\circ}\text{C}$ (Fig. 7a) and $\text{SO}_2 > 10\text{ }\mu\text{g}/\text{m}^3$ (Fig. 7f), with the largest amplitudes among all four scenarios. These results indicate that high predicted Nuc values under the NPF scenario are always associated with high temperature and sufficient precursor conditions. In contrast, the CS exerts a clear negative effect on predicted Nuc values, especially when CS is high. Its dependence plot shows that the
300 fitted curve initially increases and then decreases with increasing CS values, turning negative beyond approximately $0.085\text{ m}\cdot\text{s}^{-1}\text{ s}^{-1}$ (Fig. 7m), consistent with a mean SHAP value of -0.9 (Fig. 6a). Under the Low- O_3 scenario, the dominant contributors to predicted Nuc values differ significantly, with the top five variables in terms of importance being NO_2 (15.2%), Coa (13.3%), Ait (10.8%), Acc (7.7%), and CS (7.5%) (Fig. 6d), all closely related to background particles. This indicates that, under Low- O_3 conditions, background particles tend to increase the predicted values.

305 Under the Non- O_3 scenario, the mean SHAP values of T, SO_2 and O_3 are -94.4 , -34.5 , and -26.1 , respectively, and high-value samples are predominantly distributed in the negative SHAP value region (Fig. 6c). ~~This confirms that under conditions of limited oxidation capacity, photochemically related variables generally suppress the predicted Nuc values. This may be attributed to the fact that higher values of these photochemically related variables typically occur in the afternoon, when intensified boundary layer mixing dilutes near-surface particles. Such co-variation explains their generally negative SHAP contributions to the predicted Nuc values.~~ Notably, RH exhibits a high mean SHAP value of 153.5 (Fig. 6a), and its dependence curve increases monotonically from 80% and remained positive thereafter (Fig. 7b), indicating that high humidity conditions are more likely to correspond to higher predicted Nuc values. ~~This is likely because high RH promotes hygroscopic growth of background particles and modulates the small-size end of the particle size distribution, which the model statistically associates with elevated M_{Nuc} .~~ For the High- O_3 scenario, despite elevated O_3 concentrations, the importance of O_3 is only 3.6%, suggesting
315 that O_3 is not the key factor controlling Nuc predictions. Although SO_2 shows a positive mean SHAP value of 81.4 (Fig. 6a), indicating a promoting effect on Nuc predictions, the high-concentration data points of $\text{PM}_{2.5}$ and Coa are almost all fell into the negative SHAP value region (Fig. 6e). Given that these two variables are the most important contributors in this scenario (Fig. 6e), the results demonstrate that background particles significantly inhibit the increase in Nuc mode. This finding is consistent with the previous inference that particles are generated but rapidly scavenged under High- O_3 scenarios.

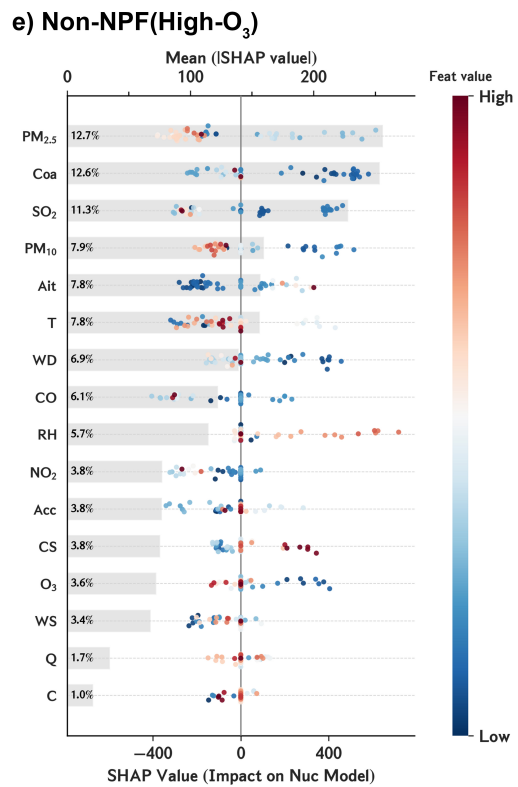
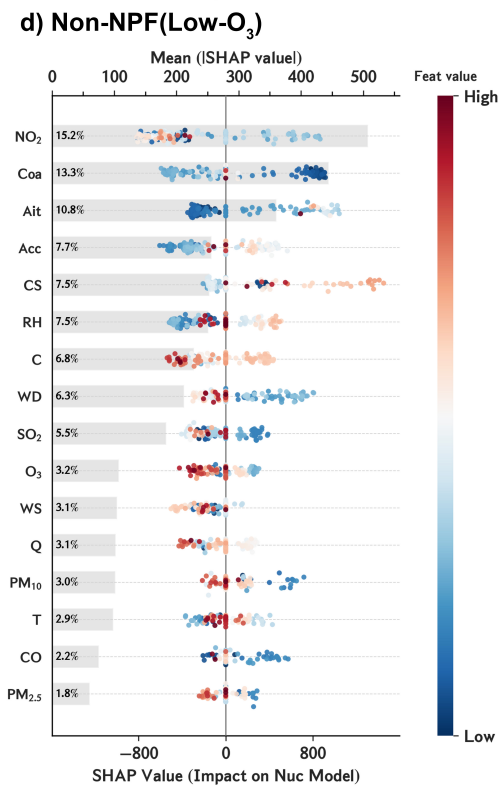
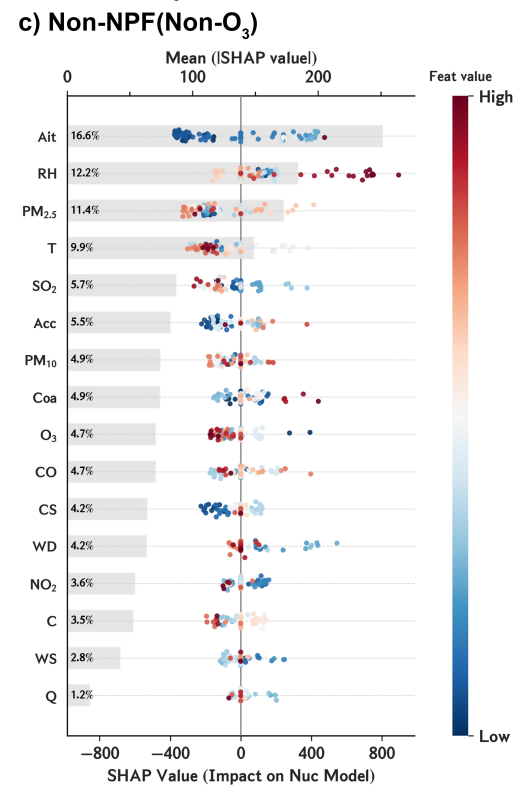
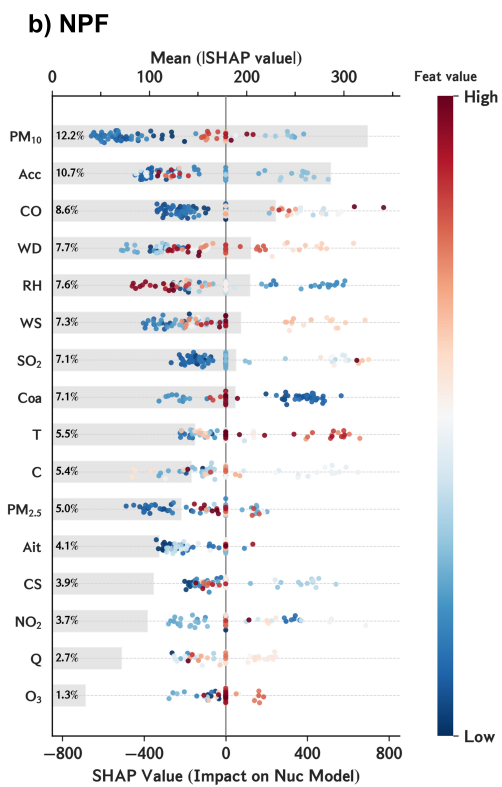
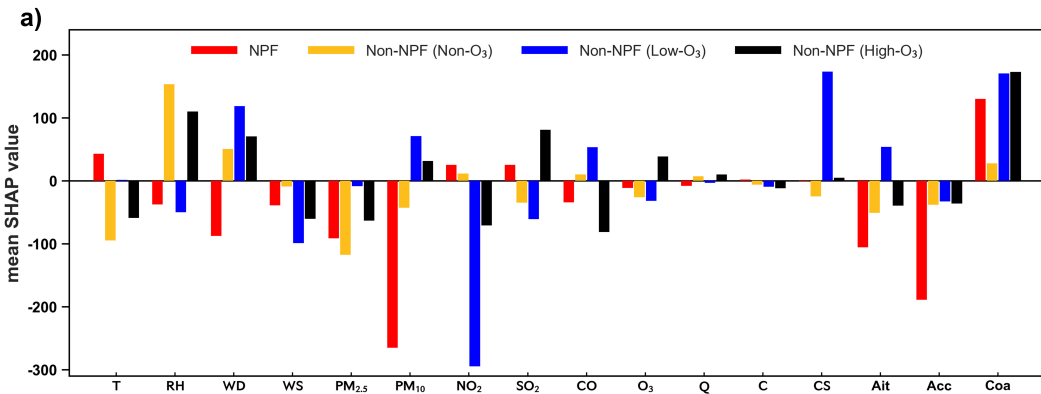


Figure 6. SHAP-based analysis of drivers for particle number concentrations in the Nuc mode. (a) Mean SHAP values across for the four scenarios, with red, yellow, blue, and black bars represent the NPF, Non-NPF (Non-O₃), Non-NPF (Low-O₃) and Non-NPF (High-O₃) scenarios, respectively. (b-e) Feature importance and summary plots for (b) NPF, (c) Non-NPF (Non-O₃), (d) Non-NPF (Low-O₃), (e) Non-NPF (High-O₃) scenario. The bars indicate the mean absolute SHAP values, while the scatter plots show the distribution of SHAP values, with colors representing low (blue) to high (red) feature values. The bar charts indicate the mean absolute SHAP values, while the scatter plots show the distribution of SHAP values, with colors transitioning from blue (low feature values) to red (high feature values).

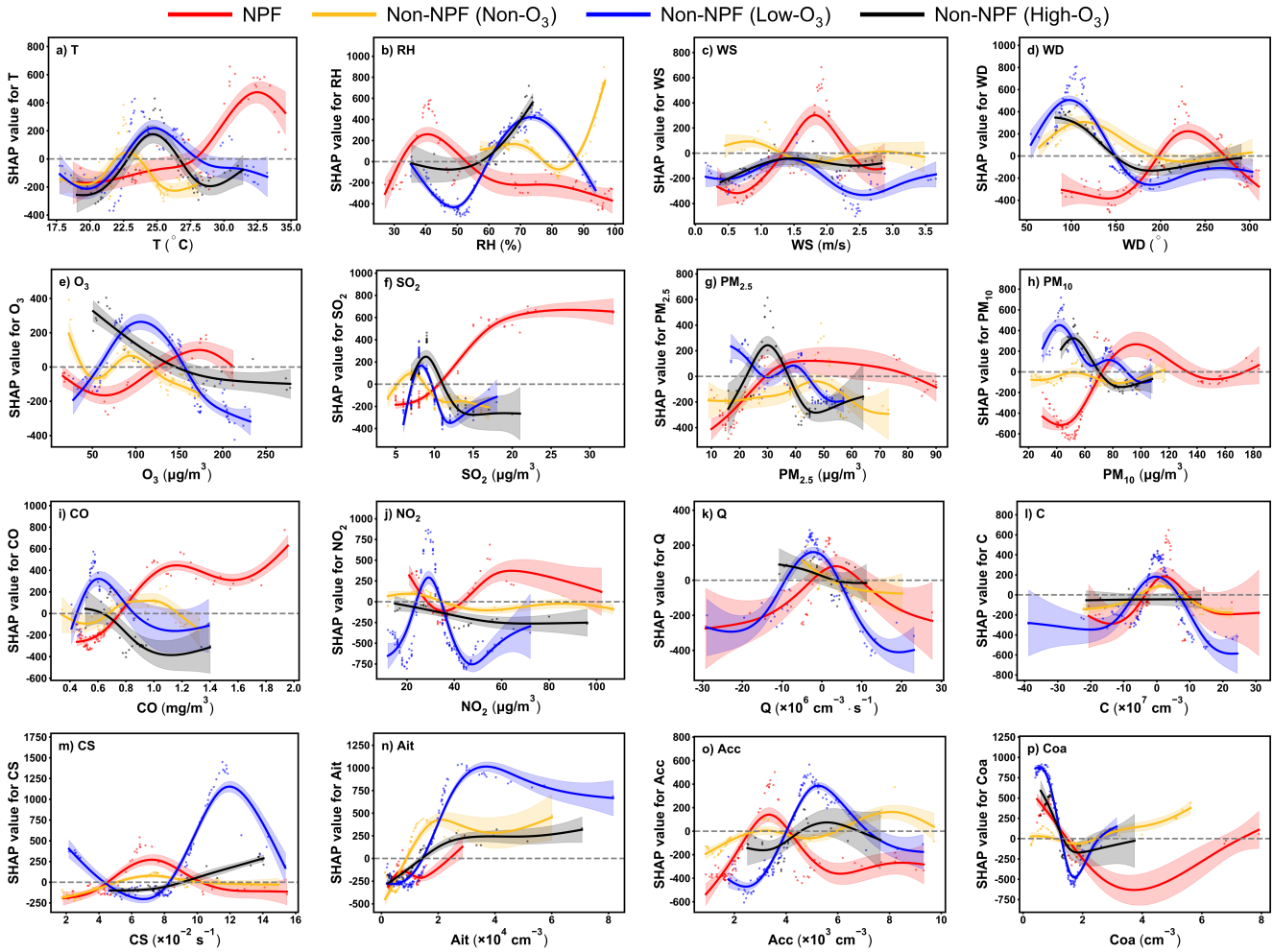


Figure 7. SHAP dependency plots of N_{Nuc} associated with different variables under four classified scenarios: (a) T, (b) RH, (c) Wind Speed, (d) Wind Direction, (e) O₃, (f) SO₂, (g) PM_{2.5}, (h) PM₁₀, (i) CO, (j) NO₂, (k) Q, (l) C, (m) CS, (n) N_{Ait} , (o) N_{Acc} , (p) N_{Coa} . Scatter points depict the SHAP values for individual samples, reflecting the contribution of each feature. The solid curves represent the fitted trends, with shaded areas indicating the 95% confidence intervals. The red, yellow, blue, and black lines represent the NPF, Non-NPF (Non-O₃), Non-NPF (Low-O₃) and Non-NPF (High-O₃) scenarios, respectively.

4 Conclusion

This study systematically analyzed the occurrence conditions and source-sink control mechanisms of NPF under different O₃ pollution backgrounds, based on surface aerosol particle size distribution, pollutant concentration, and vertical observation data. The results demonstrate that NPF is not simply governed by high or low O₃ concentrations; instead, it critically depends on the dynamic balance between precursor formation (source) and background aerosol removal (sink). Under non-O₃ pollution conditions (Non-O₃ scenario), although background aerosol and the CS level are relatively low, insufficient atmospheric oxidation capacity strongly limits precursor formation, making it difficult for the source to be established. Conversely, under high O₃ pollution conditions (High-O₃ scenario), a strongly oxidative environment favors precursor formation; however, the stronger background particle rapidly removes newly formed molecular clusters, thereby suppressing NPF as well. When O₃ concentrations are appropriate, the occurrence of NPF is determined by whether boundary layer can create a favorable window in which source enhancement and sink reduction occur synchronously. Under the NPF scenario, the breakup of the inversion

345 layer after sunrise and the rapid development of the boundary layer plays a decisive role. On the one hand, the entrainment of cleaner air aloft markedly reduces the background CS; on the other hand, enhanced vertical exchange increases near-surface O₃ concentrations. This process is aligned with the rapid conversion of SO₂ into nucleation precursors, allowing the newly formed molecular clusters to survive and grow continuously, ultimately triggering the NPF process. Under the Low-O₃ scenario, the stable boundary layer structure limits vertical mixing, preventing the dilution of near-surface pollutants, and causing the background CS increasing instead of decreasing. Although weak precursor accumulation occurs, its lack of synchronization with sink reduction inhibits the NPF.

In addition, correlation analysis combined with SHAP method provides support for the proposed source–sink competition mechanism. Correlation results show that the NPF scenario is the only case in which the Nuc mode exhibits a strong association with SO₂, which is consistent with the dominant factors identified in the SHAP analysis: high temperature ($T > 28\text{ }^{\circ}\text{C}$) and sufficient precursors ($\text{SO}_2 > 10\text{ }\mu\text{g}/\text{m}^3$) are primary positive contributors for increasing Nuc predicted values under the NPF scenario, whereas CS exerts a consistently negative influence at higher values. In contrast, in the Low-O₃ scenario, background particles such as Coa, Ait, and Acc dominate, indicating that the predicted Nuc values are more constrained by background aerosol levels. For the Non-O₃ scenario, RH is a significant factor in increasing the predicted Nuc values, while T, SO₂, and O₃ show negative contributions. As for the High-O₃ scenario, although the SHAP value of SO₂ is positive, key factors such as PM_{2.5} and Coa exhibit extremely strong negative SHAP contributions in the high-value range, playing a more dominant role in the model predictions. Overall, the physical observations and model results are highly consistent, jointly revealing that the dynamic balance between precursor supply and background removal is the fundamental determinant of NPF occurrence across different O₃ pollution levels.

It should be noted that the data utilized in this study were derived from a 25-day intensive observations conducted at a suburban site within the Yangtze River Delta region. Consequently, the source-sink synchronization framework proposed herein is best regarded as a mechanistic insight at the process level, rather than as a strictly quantitative law. Nevertheless, we believe that this framework can still provide a useful reference for understanding NPF processes in other atmospheric environments, because the temporal synchronization of source enhancement and sink weakening driven by boundary layer development does not depend on the specific emission characteristics or meteorological conditions of this study site. Naturally, the specific forms of the source and sink terms will vary with changing environmental conditions. For instance, in coastal or forested regions, source terms may be dominated by biogenic volatile organic compounds (BVOCs) and their oxidation products, while condensation sinks may be more heavily influenced by sea salt or natural organic aerosols. Conversely, during winter, when photochemical activity is attenuated and stable boundary layers are more prone to persistence, the process of source-sink synchronization becomes more difficult to establish. Furthermore, we hypothesize that this framework holds the greatest explanatory power under conditions of compound O₃-PM_{2.5} pollution; in cleaner atmospheric environments, however, NPF may be more constrained by weak source conditions. Thus, these contextual variations do not negate the applicability of the framework; rather, they indicate that when applying it across different regions or seasons, the specific source and sink terms must be re-identified in accordance with the prevailing atmospheric environment. Therefore, further research is needed to assess the universality of this mechanism based on data from multiple sites and long-term observations, and to quantitatively test its applicability under different atmospheric conditions.

Code and data availability

The observational data used in this study are derived from previously published datasets and are publicly available from Shi et al. (2022) at <https://doi.org/10.1029/2022GL099274>. The SHAP analysis was conducted using publicly available open-source implementations. The SHAP methodology and its reference implementation are available from the official SHAP project website at <https://shap.readthedocs.io/>.

Author contributions

YW carried out the data analysis, conducted the statistical and SHAP analyses, and wrote the original draft of the manuscript. DL contributed to data processing and assisted with the analysis and interpretation of the results. HW and QH designed the study, supervised the research, and revised the manuscript. SS provided the observational data used in this study. ZW and ZL
390 assisted with data analysis and visualization. TZ provided scientific guidance and contributed to the interpretation of the results. LS contributed to data quality control and methodological support. All authors discussed the results, reviewed the manuscript, and approved the final version.

Competing interests

The contact author has declared that none of the authors has any competing interests.

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410 Review statement

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