**We provide detailed responses to all comments below; the responses are shown in blue, and all revisions in the manuscript are highlighted in red, with specific changes bolded.**

**Referee #1**

This manuscript demonstrates that atmospheric new particle formation (NPF) is governed not by ozone ($O_3$) levels alone, but by the temporal synchronization between precursor production and background aerosol removal. The authors show that such synchronization is primarily controlled by the evolution of the planetary boundary layer, with NPF occurring when post-sunrise boundary-layer development simultaneously enhances near-surface oxidation capacity through mixing of $O_3$ from above and reduces the condensation sink. On the other hand, stable PBL conditions can either lead to low $O_3$ levels or high background aerosol concentations leading to low low source rate or high sink of newly formed particles.

In my opinion, methodological strength of this work is the combination of NPF classification with interpretable machine-learning tools. By integrating generalized additive models with SHAP analysis and vertical observational data, the authors quantitatively attribute nucleation-mode particle variability to individual meteorological, chemical, and aerosol parameters under different pollution regimes. This approach represents an advance over traditional correlation analyses and offers a clear framework for diagnosing nonlinear source–sink competition in NPF studies.

In addition to the comments provided by Referee #2, I suggest adding a short discussion on how the authors think the identified source–sink mechanisms would be expected to generalize to other regions, seasons, or pollution regimes. Since this study is based on an intensive observation period of roughly two weeks at a single site, it would help setting this study into broader context and to prevent overinterpretation of the results obtained here.

I recommend the manuscript to be accepted for publication.

**Response:** We sincerely thank the reviewer for the very positive overall assessment of our work and for this constructive and important suggestion. We fully agree that placing the present results in a broader spatial, seasonal, and chemical context is essential, both to underscore the wider relevance of the proposed mechanism and to avoid overinterpretation given the limited duration and single-site nature of our observations.

Following the referee's suggestion, we have added a short discussion at the end of the Conclusion section that explicitly acknowledges the observational constraints of the present dataset and outlines how the identified source–sink synchronization mechanism may be expected to generalize to other regions, seasons, and pollution regimes.

**Modified content (Section 4, Lines 364-380):** 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 $O_3$-$PM_{2.5}$ 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.

**Referee #2**

The manuscript investigates the triggering mechanisms of new particle formation (NPF) under various $O_3$ pollution scenarios, focusing specifically on the dynamic balance between precursor sources and the background condensation sink. The study concludes that NPF is highly dependent on the synchronicity between source enhancement and sink attenuation—a process regulated by the evolution of the planetary boundary layer. Overall, this is a highly interesting and well-structured study that provides valuable quantitative insights through the application of interpretable machine learning methods. The methodology is scientifically sound, and the data robustly support the final conclusions. I recommend publication following minor revisions. The authors are requested to consider and respond to the following specific comments:

**Response:** We sincerely thank the reviewer for the careful reading of our manuscript and for the very encouraging overall assessment of our work. We also greatly appreciate the constructive and detailed comments, which have helped us further improve the clarity, rigor, and overall quality of the manuscript. All comments have been carefully considered, and the manuscript has been revised accordingly. Our point-by-point responses are provided below.

**Major comments**

1. Section 3.2 emphasizes the breakup of the morning inversion layer and the subsequent rapid vertical mixing as the key process leading to the synchronization of source enhancement and sink weakening. Considering that the temporal resolution of the UAV vertical observations is 3 hours, whereas this process may occur on a much shorter timescale, is this resolution sufficient to capture such rapid evolution?

**Response:** We are deeply grateful to the reviewers for raising such detailed and critical methodological questions. We agree that processes such as the breakdown of the morning inversion layer and the rapid development of the boundary layer may indeed occur on time scales shorter than three hours. However, we maintain that this resolution remains sufficient to support the conclusions of the present study, for the following reasons:

First, the objective of utilizing UAV vertical observation data in this paper is not to capture minute-scale turbulent processes, but rather to focus on the structural differences and evolutionary trends of the boundary layer across various stages—such as pre-sunrise, post-sunrise, and mid-morning—under different scenarios; a three-hour resolution is sufficient to characterize these stage-specific differences.

Second, under the NPF scenario, phenomena such as the synchronous decline of CS and $N_{Acc}$ around 07:00 (Figs. 2c and S5c), the accumulation of $SO_2$ between 07:00 and 09:00 (Fig. S4h), and the downward transport of $O_3$ (Fig. S4d) are all finely characterized by high-resolution ground-based data, and they align highly consistently—both temporally and causally—with the evolution of the boundary layer.

2. Section 3.3: In the Non-$O_3$ scenario, RH shows a strong positive SHAP contribution, whereas T, $SO_2$, and $O_3$ show negative contributions. Could the authors provide more scientific interpretation for this?

**Response:** We are very grateful to the reviewer for raising this insightful question. SHAP values reflect the marginal contribution of each variable to the model's prediction of the Nuc mode particle number concentration, rather than strictly representing physical causal effects. The differences in the signs of the SHAP contributions under the Non-$O_3$ scenario essentially reflect the statistical covariation between the individual variables and the Nuc-mode number concentration within a low-ozone background, rather than the NPF nucleation mechanisms themselves.

From a physical perspective, the Non-$O_3$ scenario is characterized by a combination of low $O_3$, low T, and low $SO_2$ levels (Fig. S4)—conditions that fall far short of those required to drive new particle formation. However, high values of these variables tend to occur during the afternoon hours when photochemical activity is relatively high; this coincides with the dilution process of near-surface particulate matter, thereby resulting in a statistically negative correlation with the Nuc mode.

High RH may promote the hygroscopic growth of background particles and influence the distribution of the particle size spectrum in the sub-micron range, leading the model to statistically associate higher RH with higher NNuc values. However, it must be emphasized that this positive contribution does not signify an active NPF process. No distinct nucleation signatures were observed in the Non-$O_3$ scenario (Fig. S3b), indicating that—even though RH shows a positive contribution to the model's predictions—source limitation remains the dominant factor inhibiting the occurrence of NPF.

**Modified content (Section 3.3, Lines 305-314):** 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 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 $N_{Nuc}$.**

**Minor comments**

1. Line 29-33: The first paragraph of the Introduction is rather difficult to read and it is recommended to be reorganized.

**Response:** Thank you for your suggestion. We have rewritten and split the first paragraph of the introduction to make the logic clearer and the sentences more fluent.

**Modified content (Section 1, Lines 29-33):** 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).

2. Lines 107-109: Please delete the space after the hyphen in "Low- $O_3$" and "High- $O_3$".

**Response:** We appreciate you pointing this out and have already made the correction.

**Modified content (Section 2.2, Lines 108-109):** Finally, by combining the NPF event identification with the ozone pollution classification, the observation period was divided into four scenarios:

(1) NPF: 30 May, 3 June, and 11–13 June;

(2) Non-NPF (Non-$O_3$): 31 May, 5 June, and 9–10 June (hereafter Non-$O_3$);

(3) Non-NPF (**Low-$O_3$**): 1–2 June, 6–7 June, and 14–15 June (hereafter Low-$O_3$);

(4) Non-NPF (**High-$O_3$**): 4 June and 8 June (hereafter High-$O_3$).

3. Line 120: "coagulation sink (Coags, $S^{-1}$)" is recommended to be revised to $s^{-1}$.

**Response:** Thank you for pointing out this unit formatting issue.

**Modified content (Section 2.3, Line 120):** And the coagulation sink (Coags, $\mathbf{s^{-1}}$) describes the loss rate of target particles due to coagulation with background particles and is defined as (Kulmala et al., 2001a):

4. Line 122: In the equation, should "Cogas" be corrected to Coags? It seems to be a spelling error.

**Response:** We thank the reviewer's meticulous review. This was indeed a spelling error. We have consistently corrected "Cogas" to "Coags" throughout the formulas.

**Modified content (Section 2.3, Line 123):**

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

5. Section 2.4: Why was GAM chosen instead of random forest or XGBoost?

**Response:** We thank the reviewer for the careful reading of the manuscript. We selected GAM primarily because the objective of this study is not merely to achieve high predictive accuracy, but rather to identify the relative contributions of various influencing factors to the number concentration of Nuc mode particles under different $O_3$ pollution scenarios, as well as their nonlinear characteristics.

RF and XGBoost tend to function more as "black-box" models, which can often result in a loss of physical interpretability. Although they typically demonstrate strong fitting capabilities in predictive tasks, for the purposes of this study, our primary focus lies on the marginal effects of different factors and their underlying physical implications, rather than solely on the performance of the model itself.

In contrast, GAM is more easily associated with actual atmospheric physical and chemical processes. While breaking through the limitations of traditional linear regression, it also avoids the shortcomings of deep machine learning models that lack physical meaning, making it more suitable for our mechanism-oriented requirements.

6. Line 178: "As show in Table 1" should be corrected to "As shown".

**Response:** Thank you for pointing this out. We have made the revisions in the manuscript.

**Modified content (Section 3.1, Lines 179-180): 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).

7. Line 181 & Table1: "Cogas" is also a spelling error here.

**Response:** We thank the reviewer for pointing out this error. All "Cogas" in the manuscript have been uniformly corrected to "Coags" .

**Modified content (Section 3.1):**

**Line 182:** Taking June 13th as an example, although both CS ($6.20\times10^{-2}$ $s^{-1}$) and **Coags** ($18.74\times10^{-5}$ $s^{-1}$) 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.

**Table1:**

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

| Date | FR ($cm^{-3}\cdot s^{-1}$) | GR ($nm\cdot h^{-1}$) | C ($\times10^{7}$ $cm^{-3}$) | Q ($\times10^{6}$ $cm^{-3}\cdot s^{-1}$) | CS ($\times10^{-2}$ $s^{-1}$) | **Coags** ($\times10^{-5}$ $s^{-1}$) |
|---|---|---|---|---|---|---|
| 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 |

8. Lines 219-220: "In contrast…" is repeated from Lines 165–166, please delete it.

**Response:** Thank you for the correction. We have removed the duplicate statement.

9. Lines 226-227: The statement "According to the classification criteria in Sect. 2.2, both the NPF and Low-$O_3$ scenarios had similarly low $O_3$ pollution levels (160-215 $\mu g/m^3$)" does not seem to be mentioned in Section 2.2. Please clarify.

**Response:** Thank you for this insightful comment. The observed $O_3$ concentrations during the NPF days also belong to the Low-$O_3$ range (160 < MDA8-$O_3$ < 215 $\mu g/m^3$). This detail was originally omitted in the text for brevity. We apologize for any confusion this omission may have caused and have now revised the manuscript to clarify this point.

**Modified content (Section 2.2, Lines 106-107):** 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-$O_3$): 1–2 June, 6–7 June, and 14–15 June (hereafter Low-$O_3$);

(4) Non-NPF (High-$O_3$): 4 June and 8 June (hereafter High-$O_3$).

10. Line 254: The citation "Ail et al., 2025" is inconsistent with Ali in the References (Line 392); please revise it.

**Response:** We thank the reviewer for this comment. We have made corrections.

**Modified content (Section 3.2, Line 255):** 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**).

11. Lines 229-300: The phrase "turning negative beyond approximately 0.085 $m \cdot s^{-1}$" is discussing CS, so the authors are advised to check the unit. Should it be $s^{-1}$ instead?

**Response:** We thank the reviewer for the careful examination, and we note that this comment likely refers to Lines 299–300. The unit designation here was indeed incorrect, and we have since corrected it.

**Modified content (Section 3.3, Line 301):** Its dependence plot shows that the fitted curve initially increases and then decreases with increasing CS values, turning negative beyond approximately **0.085 $s^{-1}$** (Fig. 7m), consistent with a mean SHAP value of −0.9 (Fig. 6a).

12. Lines 316–317: Please revise the caption of Figure 6, as "(a) Mean SHAP values across for scenario" is not grammatically smooth.

**Response:** We sincerely appreciate your careful review. The word "for" was indeed a typographical error and should have been "four". We have corrected this spelling mistake and further polished the entire caption of Figure 6 for better clarity.

**Modified content (Section 3.3, Lines 321-327):**

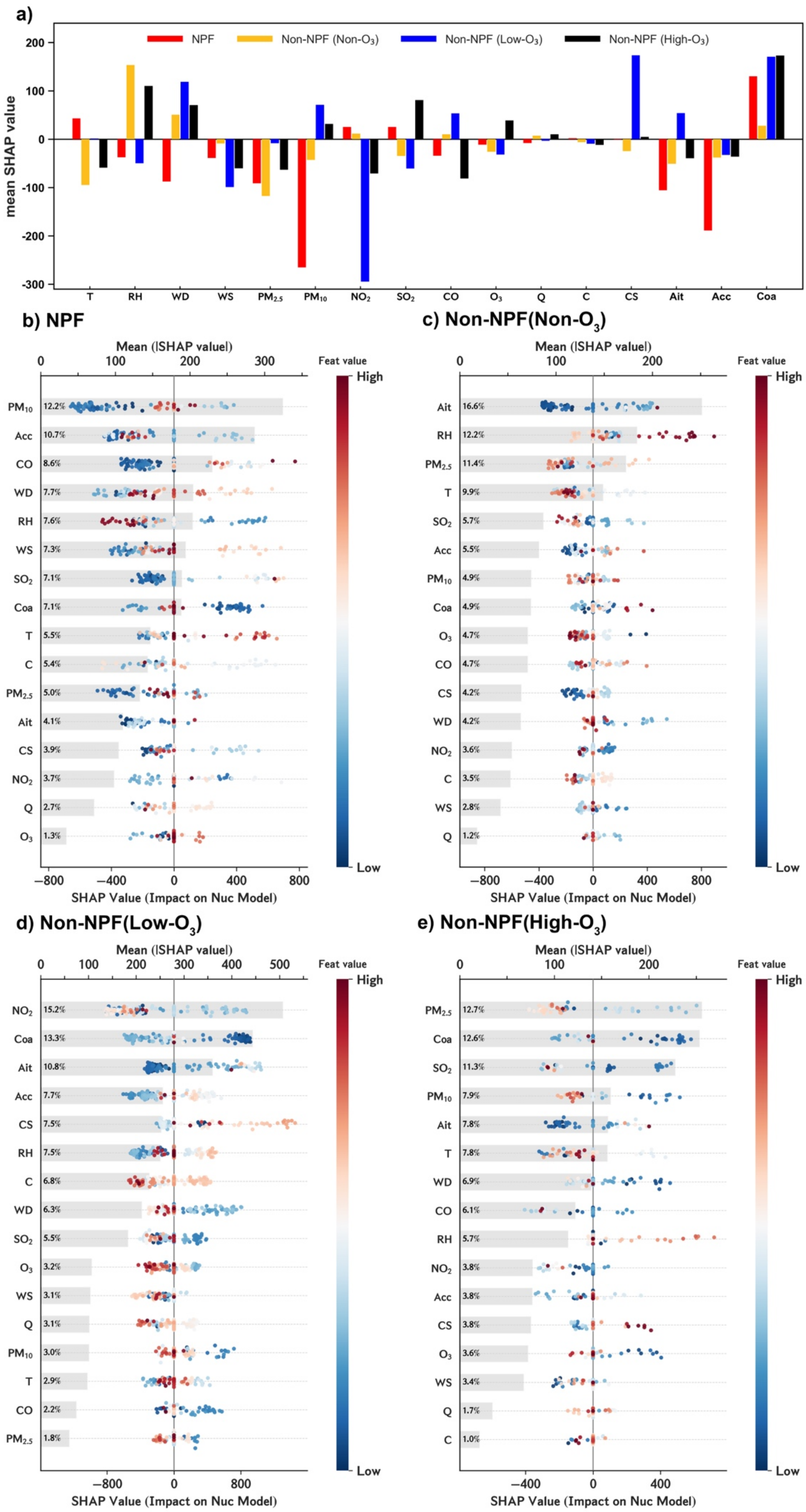
a)
NPF
Non-NPF (Non-O3)
Non-NPF (Low-O3)
Non-NPF (High-O3)
mean SHAP value
200
100
0
-100
-200
-300
T
RH
WD
WS
PM2.5
PM10
NO2
SO2
CO
O3
Q
C
CS
Ait
Acc
Coa
b) NPF
c) Non-NPF(Non-O3)
d) Non-NPF(Low-O3)
e) Non-NPF(High-O3)
Mean (|SHAP value|)
Feat value
High
Low
SHAP Value (Impact on Nuc Model)
PM10 12.2%
Acc 10.7%
CO 8.6%
WD 7.7%
RH 7.6%
WS 7.3%
SO2 7.1%
Coa 7.1%
T 5.5%
C 5.4%
PM2.5 5.0%
Ait 4.1%
CS 3.9%
NO2 3.7%
Q 2.7%
O3 1.3%
Ait 16.6%
RH 12.2%
PM2.5 11.4%
T 9.9%
SO2 5.7%
Acc 5.5%
PM10 4.9%
Coa 4.9%
O3 4.7%
CO 4.7%
CS 4.2%
WD 4.2%
NO2 3.6%
C 3.5%
WS 2.8%
Q 1.2%
NO2 15.2%
Coa 13.3%
Ait 10.8%
Acc 7.7%
CS 7.5%
RH 7.5%
C 6.8%
WD 6.3%
SO2 5.5%
O3 3.2%
WS 3.1%
Q 3.1%
PM10 3.0%
T 2.9%
CO 2.2%
PM2.5 1.8%
PM2.5 12.7%
Coa 12.6%
SO2 11.3%
PM10 7.9%
Ait 7.8%
T 7.8%
WD 6.9%
CO 6.1%
RH 5.7%
NO2 3.8%
Acc 3.8%
CS 3.8%
O3 3.6%
WS 3.4%
Q 1.7%
C 1.0%

Figure 6. SHAP-based analysis of drivers for particle number concentrations in the Nuc mode. (a) Mean SHAP values across the **four** scenarios, with red, yellow, blue, and black bars represent the NPF, Non-NPF (Non-$O_3$), Non-NPF (Low-$O_3$) and Non-NPF (High-$O_3$) scenarios, respectively. (b-e) Feature importance and summary plots for (b) NPF, (c) Non-NPF (Non-$O_3$), (d) Non-NPF (Low-$O_3$), (e) Non-NPF (High-$O_3$) scenario. **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).**