We sincerely thank the reviewers for their careful reading of the manuscript and their constructive comments. We have revised the manuscript and supplementary materials accordingly. Below we respond point-by-point to each comment. Reviewer comments are reproduced in bold, followed by our responses.

Reviewer 1

Although the authors have made certain revisions to the article, further modifications are still required prior to publication consideration. The specific issues are as follows:

1. Validation through numerical simulations is a crucial component for demonstrating the scientific validity of the RIN mechanism in this study. Although the particle number size distribution (PNSD) timeseries (Figure S6) was provided in the previous revision, the three subplots are arranged side by side, resulting in compressed long-term time series data that obscures the daily evolution of the particle size distribution. Since the hourly evolution of the size spectrum is essential for determining the occurrence of new particle formation events, it is recommended to optimize and adjust the presentation. Additionally, further improvement in color scheme is advised. Furthermore, it is suggested to include a discussion on the overall PNSD simulation results in the main text. Otherwise, validating the nearly month-long simulation by selecting only two days for analysis lacks persuasiveness.

Reply:

We appreciate this important suggestion and agree that model performance should be evaluated over the full simulation period. We have strengthened the manuscript accordingly as follows:

1. **Improved presentation of Figure S6:**
   - The original side-by-side layout has been reformatted into a vertically aligned format to improve temporal readability.
   - The horizontal axis has been expanded to better resolve day-to-day variability.
   - The color scheme has been revised to provide clearer and more informative shading.
2. **Expanded discussion of full-month PNSD evolution in the main text** (see reply to Comment #2).
3. **Clarified the role of the two selected days:**
   We now clarify that November 20 and 25 were selected to illustrate diurnal dynamics under relatively stable synoptic conditions, whereas period-wide model performance is evaluated in Table 2 and the new Fig. 8, with

corresponding discussion in the text.

2. In the atmospheric environment, organic compounds play a significant role in particle growth. Based on the comparison of PNSD simulations between the No_RIN and RIN scenarios in this study, the RIN scenario appears to overestimate the particle number concentration within the smaller size range (diameter < 100 nm). Could this be attributed to a lack of consideration for mechanism of particle growth processes in the simulation? Please provide further clarification.

Reply:

Thank you for raising this mechanistic question.

First, we note that the RIN case does not represent a systematic overestimation of total number in the <100 nm range when evaluated over the full month. For example, Table 2 shows that the PNC on average remains lower than observed even in what we regarded as the most physically consistent RIN configuration (Obs: 9354 cm^{-3}; NoRIN: 2287 cm^{-3}; RIN_{Do}: 7307 cm^{-3}). What may have happened is that the simulated Aitken-mode PSD becomes overly broad, so the distribution tails—especially the sub-10 nm number—are exaggerated. We think this bias reflects not only uncertainties in early-growth processes (e.g., condensation and coagulation), but also limitations inherent to the modal representation in CMAQ.

In the current framework, newly formed particles are not represented as a distinct, externally mixed population. Instead, they are immediately assimilated into the pre-existing Aitken mode, which already includes ambient and primary-emitted particles characterized by larger modal diameters and broader geometric standard deviations. This artificial assimilation inevitably broadens the simulated Aitken-mode size distribution and inflates the small-diameter tail. In reality, freshly nucleated particles would initially remain as a separate nucleation-mode population and would be removed efficiently through self-coagulation and scavenging by pre-existing Aitken-mode particles. As a result, far fewer particles would persist in the sub-10 nm range, and the nucleation-mode signature would be less pronounced than implied by the merged-mode treatment.

A related issue is mixing state. Physically, freshly nucleated particles should initially remain largely externally mixed and can exhibit composition-dependent growth rates. In CMAQ, however, they are treated as internally mixed within the host mode, which can alter condensational growth and coagulation dynamics relative to an externally mixed treatment. A rigorous separation of the contributions from (i) growth-process parameterization, (ii) modal merging/broadening, and (iii) mixing-state assumptions would require additional model development and sensitivity experiments that are beyond the scope of this study.

We have incorporated this discussion into Section 4.4 and moved Figure S6 from the Supplement to the main text (as Figure 8).

3. Figure S4 does not include validation for PM2.5, which is inconsistent with the response to the review comments. Given that this study focuses on particulate matter simulation, please supplement the figure with PM2.5 timeseries validation.
Reply: Thanks for pointing this out. In response, we have added $PM_{2.5}$ time-series validation plots to the Supplement and updated the Supplement text to clearly describe the $PM_{2.5}$ model–observation agreement over the study period.

Reviewer 2:
The authors have made efforts to address the previous round of comments, improving the manuscript's clarity and adding supporting information in several areas. However, two critical scientific issues remain inadequately resolved and require further attention before publication can be recommended.
The first major concern relates to the nucleation mechanism framework. While the authors now acknowledge that acid-base and organic nucleation are plausible in Taichung's atmosphere, the model continues to employ only H_2SO_4–H_2O binary nucleation. The justification provided is that binary nucleation is "sufficient" to reproduce observed particle number concentrations and that NH_3 or organics would "not fundamentally alter the qualitative behavior of the system." This assertion, however, lacks empirical support. Without sensitivity tests comparing other nucleation pathways, it remains unclear whether the current framework captures the dominant physics or merely achieves agreement through parameter adjustment. If NH_3 or organics significantly enhance nucleation efficiency—as recent urban nucleation studies increasingly suggest—the CMAQ parameterization developed in Section 4 (which assumes binary-only chemistry) could systematically misestimate PNC under varying NH_3/VOC conditions. The authors' statement that the parcel model serves "qualitative interpretation only" appears inconsistent with subsequently using its mechanistic insights to construct a quantitative CMAQ parameterization. At minimum, sensitivity simulations with at least one alternative nucleation scheme are needed to quantify the uncertainty introduced by this framework choice and to substantiate the claim that mechanism selection does not fundamentally affect the conclusions.
Reply:
We appreciate the reviewer’s emphasis on the nucleation-mechanism choice and the potential implications for generality. We would like to clarify an important point: the parcel-model results were not used to develop or tune the CMAQ parameterization

in Section 4. The CMAQ parameterization in Section 4 was constructed solely from the experimental constraints and observations reported in Section 2, and the parcel model was introduced only as an idealized, qualitative illustration to help interpret how intake-air aerosol loading and incomplete particle vaporization could influence the *tendency* of PNC formation in the exhaust plume. We also removed the sentence that "this most basic pathway alone is able to produce the observed PNC production" to avoid implying it is the correct or dominant nucleation mechanism.

We agree that real-world nucleation and early growth in combustion exhaust can involve multiple pathways and chemical families (e.g., sulfuric acid, ammonia, and organic compounds) and that these mechanisms may be important in ambient urban atmospheres. Our intent, however, was not to claim that H_2SO_4–H_2O binary nucleation is uniquely dominant under all conditions, nor that alternative pathways are irrelevant. Rather, we adopted the H_2SO_4–H_2O binary scheme because it provides a well-established, internally consistent baseline that can demonstrate the *direction* of system response in an idealized framework, without introducing additional poorly constrained parameters under the extreme thermodynamic conditions considered here.

Specifically, although adding ternary H_2SO_4–H_2O–NH_3 (or organic-assisted) nucleation is conceptually straightforward, implementing it in a *physically defensible* manner under high-temperature exhaust-plume conditions is not. Doing so would require thermodynamic and kinetic inputs for multicomponent clusters and freshly formed particles—e.g., activities, surface tension, density, and evaporation/condensation rates—that are poorly constrained at the elevated temperatures and rapid dilution/cooling characteristic of combustion exhaust. Most available ternary/organic parameterizations have been developed and evaluated for atmospheric boundary-layer conditions, and their direct application to this transient, non-equilibrium regime would therefore be uncertain without additional validation. Moreover, explicitly introducing NH_3 and organics would add substantial uncertainty through gas–particle partitioning and evolving composition during cooling, making it difficult to interpret differences as mechanistic insight rather than parameterization artifacts. For these reasons, we do not consider alternative-scheme "sensitivity tests" in this context to be robust, and we wish to avoid presenting results that could be misconstrued as mechanistically definitive.

Nonetheless, we can still discuss sensitivity in a way that does not overstate mechanistic certainty. Figure 6 shows that PNC production is highly responsive to changes in condensable vapor availability—modulated by intake/ambient aerosol loading and the particle vaporization fraction—and therefore to the effective nucleation rate. Importantly, these relationships reflect fundamental physical

controls (competition between vapor generation/supersaturation and removal by condensation/coagulation sinks) that are expected to remain operative regardless of whether nucleation proceeds via binary, ternary, or organic-assisted pathways, even though the *absolute* nucleation efficiency may differ across mechanisms.
To address the reviewer's concern more transparently, we have revised the manuscript to clarify the scope and interpretation in three ways: (1) the parcel model is presented strictly as an idealized tool for qualitative interpretation and is not used to constrain, calibrate, or tune the CMAQ parameterization; (2) the CMAQ parameterization is explicitly framed as being conditioned on an H_2SO_4–H_2O binary baseline and should be interpreted within that assumption set; and (3) we add an discussion noting that NH_3- or organic-enhanced nucleation would most plausibly manifest as an effective scaling (i.e., a multiplicative increase) of nucleation rates, which could amplify PNC under higher NH_3/VOC conditions, while not altering the qualitative direction of the sensitivities and responses reported here.

Reference:
Merikanto, J., Napari, I., Vehkamäki, H., Anttila, T., & Kulmala, M. (2007). New parameterization of sulfuric acid-ammonia-water ternary nucleation rates at tropospheric conditions. Journal of Geophysical Research: Atmospheres, 112(D15).

The second major issue concerns the scope and generalizability of the findings. The revised Introduction and Method appropriately acknowledges that the RIN mechanism operates under specific regional conditions similar to those in Taiwan—heavy pollution, high pre-existing aerosol loading, suppressed NPF events, and a high proportion of motorcycles. However, the title, abstract, and conclusions continue to imply general applicability across urban environments. This creates a misleading impression, as most global cities exhibit substantially different characteristics, and this mechanism may be regionally important in motorcycle-dominated Asian cities but not necessarily representative of urban environments globally. The title, abstract, and conclusions should be revised to explicitly reflect the specific region where this mechanism has been validated.
Reply:
We would like to clarify our intent regarding generalizability. Although the engine platform used in the laboratory was a motorcycle, the proposed RIN mechanism is based on generic processes in combustion exhaust dilution and cooling (e.g., evaporation of condensable material and subsequent nucleation/growth sensitivity to background aerosol loading), which are not inherently unique to motorcycles. Moreover, even within our study region, the majority of urban combustion emissions

arises from passenger cars and other engine types, so our discussion was not meant to frame RIN as a “motorcycle-only” phenomenon. Therefore, the mechanism is applied to all combustion vehicles in our regional simulations. We now stated this in the beginning of Section 4.

To resolve the reviewer’s concern, we will reframe the manuscript language as follows:

1. Evidence statement: emphasize that the mechanism is *demonstrated and evaluated* under Taiwan/West-Pacific urban conditions (in the beginning of section 4).
2. Transferability statement: describe potential applicability to other combustion sources and regions as a hypothesis that requires additional testing, rather than as a conclusion (next to the last paragraph of Section 5.4).

These revisions will ensure the paper accurately distinguishes between what is supported by our data and what remains an extrapolation.