

Reviewer #1 comments

This study attempts to demonstrate that the newly introduced RIN mechanism can significantly improve the simulation of ambient particle number concentrations, thereby highlighting the important role of the RIN process in modeling vehicle-induced particle number concentrations. However, based on the simulation results presented in the current manuscript, the RIN mechanism does not show a clear improvement in simulating particle number concentrations. In particular, the newly added Figure 8 shows that, when compared with observed particle size distributions, the introduction of the RIN mechanism leads to a substantial overestimation of particle number concentrations in the small-size range in most cases. Paradoxically, this overestimation is used as key evidence supporting the claim that the RIN mechanism alleviates the underestimation of particle concentration simulations. The analysis further attributes this overestimation to inherent issues with the model itself. If so, it would be more convincing to first optimize the model before conducting comparative simulations.

Reply:

We thank the reviewer for this important comment regarding the interpretation of the RIN mechanism's impact on particle number concentrations. We agree that the current results do not demonstrate a uniform improvement across all particle size ranges. In particular, as noted by the reviewer, Fig. 8 shows an overestimation of particle number concentrations in the small-size range when the RIN mechanism is included.

Importantly, although an overestimation is evident at the smallest sizes, the Aitken-mode number concentrations remain reasonable and, in some cases, are still slightly underestimated in the monthly averages. This suggests that the discrepancy reflects a redistribution of particle number across the size distribution rather than a systematic overprediction of total particle number. The inclusion of the RIN mechanism reduces the overall low bias in total particle number, although it introduces a size-dependent overestimation at the smallest diameters.

The observed overestimation in the small-size range suggests that, while the RIN mechanism enhances particle production processes, it may also amplify pre-existing model biases. Further analysis indicates that this issue is closely linked to the model's particle size distribution (PSD) representation. Specifically, the geometric standard deviation (σ_g) of the simulated Aitken mode is systematically broader than observed, even with the upper bound imposed by CMAQ. Such behavior is consistent with known limitations of modal aerosol schemes, as discussed in the revised manuscript. This overly broad σ_g is present in simulations without the RIN mechanism and becomes more pronounced when the RIN mechanism is included, leading to an artificial spreading of particle number into the smallest size bins. In addition, uncertainties in the parameterization of RIN-generated particles (e.g., assumptions for μ and σ_g) may further contribute to this size-dependent

discrepancy.

The manuscript has been revised to avoid implying a uniform improvement and instead to emphasize the role of the RIN mechanism in modifying particle number concentrations. A new discussion has been added to highlight the role of PSD representation—particularly σ_g —in contributing to the identified discrepancy. The limitations of the current modeling framework, including the interaction between the RIN mechanism and existing parameterizations, are now more clearly stated. These revisions provide a more balanced and transparent interpretation of the results.

Regarding the reviewer's suggestion to optimize the model prior to comparison, we agree that further refinement of the PSD representation would likely improve model performance. However, the primary objective of this study is to isolate and evaluate the impact of the RIN mechanism on particle number concentrations within the current CMAQ framework. Implementing such refinements would require substantial restructuring of the aerosol representation (e.g., the modal framework and microphysical parameterizations) and is therefore beyond the scope of the present study. This scope has been clarified in the revised manuscript.

The main revisions in the manuscript include the following:

1. The last sentence of the Abstract has been revised as follows:

“The inclusion of RIN reduces the overall low bias in PNC but primarily enhances the smallest size range, leading to overestimation in this regime. Despite this, Aitken-mode number concentrations remain reasonable and are sometimes still slightly underestimated. These results indicate that RIN influences particle production while interacting with existing model limitations.”

2. The following paragraphs are added to the end of Section 5.4:

“An additional limitation is the overestimation of particle number concentrations in the smallest size range when the RIN mechanism is included. Although the Aitken-mode number concentrations remain reasonable—and in some cases still slightly underestimated in the monthly averages—the model overpredicts particle numbers at the lower end of the size distribution (Fig. 8). This reflects not only enhanced particle production by RIN, but also limitations in the PSD representation. Analysis indicates that σ_g of the simulated Aitken mode is systematically broader than observed, even with the upper bound imposed in CMAQ. This bias exists in the baseline simulation and is amplified by RIN, leading to artificial spreading of particle number into the smallest size bins. Consequently, part of the apparent improvement in total particle number arises from redistribution within the PSD rather than an accurate size-resolved representation. In addition, uncertainties in the parameterization of RIN-generated particles (e.g., assumptions for D_g and σ_g) may further contribute to this size-dependent discrepancy.

This σ_g -related bias is consistent with known limitations of modal aerosol schemes, in which a limited number of lognormal modes with prescribed or weakly constrained σ_g can introduce structural biases (Binkowski and Roselle, 2003; Elleman and Covert, 2010). Modal approaches tend to artificially broaden size distributions—particularly in the ultrafine range—due to their inability to explicitly resolve distinct nucleation modes (Mann et al., 2012), and similar PSD-related biases have been reported in regional models (Sartelet et al., 2022). These findings indicate that the impact of RIN is strongly conditioned by the underlying PSD framework. In the current modal structure, newly formed particles are immediately merged into the Aitken mode, precluding representation of a distinct nucleation mode. Addressing this limitation would require improved PSD treatments (e.g., inclusion of an explicit nucleation mode or adoption of sectional approaches) and refinement of early growth parameterizations; however, such developments would involve substantial restructuring of the aerosol representation and are therefore beyond the scope of the present study.”