Response to Reviewer 2

We thank the reviewer for the positive assessment and for identifying important ambiguities in the interpretation of γ and the model documentation. We have revised the central objective, defined the implemented parameter as an effective coefficient for the specific NOCs-forming pathway, clarified the WRF-Chem/AERO5 model and SAPRC-99 configuration, and documented the treatment of primary NOCs. All changes quoted below are marked in red in the revised manuscript.

Major comments

Major comments 1 and 2 (addressed together)

Reviewer comment: *First, the key conclusion from this paper (i.e., $\gamma = 10^{-5}$ is a reasonable upper limit) is not sufficiently justified. It seems the sole reasoning is "NOCs concentrations simulated with the γ of 10^{-5} are the closest to observations" (line 290). However, given the simplifying assumption that all NH3 uptake on SOA leads to NOC formation (line 115), I would expect that the model substantially overestimates NOC production for any given uptake coefficient. In that case, tuning γ downward to match observed NOCs could simply compensate for an overly efficient NOC yield, and the resulting "best-fit" γ may not be physically meaningful.*

Reviewer comment: *More broadly, I encourage the authors to reconsider and clarify the paper's central objective. I do feel the sensitivity experiments are valuable and convincingly show that NH3 uptake on SOA can trigger cascading impacts on NH3, inorganic aerosol, and $PM_{2.5}$, with a sublinear dependence on the assumed uptake coefficient. Several aspects (e.g., induced changes in aerosol–radiation interactions and potential dynamical responses) are interesting and could also be explored further. The current attempt to observationally constrain γ using NOCs, however, does not yet appear robust. If the authors aim to reduce the uncertainty in γ, substantially more work is needed to provide a stronger, more defensible constraint.*

Response: We thank the reviewer for these constructive comments. In the revised manuscript, γ is described as a branch-specific effective reactive uptake coefficient. It represents the net probability that an NH_3 collision with an SOA-containing particle ultimately produces a stable particulate organic-N product. It is not the total physical uptake coefficient of NH_3. The statement in the original manuscript that "all NH_3 uptake on SOA leads to NOC formation" is imprecise and has been revised. What the model assumes is that the NH_3 loss assigned specifically to the implemented NOCs-forming pathway is converted into an aggregate organic-N tracer. Conventional inorganic partitioning and ammonium formation are treated separately by ISORROPIA. The observations include NOCs produced through multiple pathways. We therefore no longer use the observations as a direct mechanistic validation target. Instead, they provide a conservative upper-bound constraint: the contribution from this specific pathway should not substantially exceed the total observed ambient NOCs burden. Accordingly, we have also revised the central objective of the manuscript.

Changes in the revised manuscript: Main text, Abstract, lines 15–21; Introduction, lines 77–81; Sect. 2.1, lines 97–101; Sect. 2.2, lines 116–119; Summary and conclusions, lines 337–339.

> *"In this study, we incorporate a first-order NOCs-forming NH_3–SOA reactive-uptake pathway into WRF-Chem and adopt a literature-based sensitivity range of $\gamma = 10^{-5}$–10^{-3} to evaluate its impacts on air quality and particulate organic nitrogen over the North China Plain during November 2017. Laboratory evidence of declining reaction efficiency and comparison with the measured NOCs burden support considering $\gamma = 10^{-5}$ as a conservative upper limit for sustained operation of this pathway under the conditions examined."*

"In this study, we adopt a literature-based sensitivity range of $\gamma = 10^{-5}$–10^{-3} for a simplified NOCs-forming NH_3–SOA pathway. These values are selected according to laboratory measurements and previous modeling studies. The observations are used to assess the atmospheric plausibility of the contribution of this pathway and the sensitivity simulations are used to evaluate the corresponding air-quality impacts over the NCP."

"The modeled NOCs concentration therefore represents only the product of the implemented NH_3–SOA reactive-uptake pathway and should not be interpreted as total ambient NOCs. Because the measured NOCs burden can include primary emissions and products from other secondary pathways, the model–measurement comparison provides a conservative upper limit for the contribution of this pathway."

"The present parameterization does not resolve these steps separately or represent the complete particulate uptake and fate of NH_3. For consistency with previous laboratory and modeling studies, we retain the notation γ but interpret it here as an effective coefficient for the implemented NOCs-forming pathway."

"Together with laboratory evidence of declining reaction efficiency, we conclude $\gamma = 10^{-5}$ as a conservative upper limit for sustained operation of this pathway under the conditions examined."

Major comment 3a: model architecture

Reviewer comment: *Another concern is the model setup and documentation. Since this is primarily a modeling study, inaccuracies or inconsistencies in the model description are particularly concerning. The manuscript states throughout that WRF-Chem is used, yet the Supporting Information says "aerosols are modeled in CMAQ using a modal approach…". This is confusing because CMAQ and WRF-Chem are two distinct modeling systems. Please clarify whether this is an editing error or whether CMAQ is actually involved, and explicitly state in the main text which aerosol module/configuration is used.*

Response: The simulations use a modified WRF-Chem system; stand-alone CMAQ and an offline WRF–CMAQ system are not used. This architecture is now stated explicitly in both the main text and SI.

Changes in the revised manuscript: Main text, Sect. 2.1, lines 86–89; SI Text S1, lines 13–15.

"WRF-Chem provides the online-coupled meteorology, transport, gas-phase chemistry, photolysis, deposition, and aerosol–radiation framework, whereas aerosol processes are represented by the CMAQ/Models-3 AERO5 modal aerosol module incorporated within WRF-Chem."

"Aerosol processes are represented by the Community Multiscale Air Quality/Models-3 AERO5 modal aerosol module incorporated within WRF-Chem. AERO5 represents coagulation, condensational growth, and new-particle formation using three lognormal modes (Binkowski & Roselle, 2003)."

Major comment 3b: SAPRC-99 and configuration dependence

Reviewer comment: *Also, the manuscript states that a "SAPARC-99" mechanism is used for gas-phase chemistry. Did you mean SAPRC-99? If so, this is an older mechanism, and SAPRC has undergone major updates since then (e.g., SAPRC18). Because the conclusions, especially those involving oxidant levels, sulfate/nitrate formation, and aerosol acidity, can depend strongly on the chemical mechanism and aerosol module, these choices need to be accurately described and clearly documented.*

Response: We have corrected this error throughout the manuscript and Supporting Information. SAPRC-99 is older than recent SAPRC mechanisms, but it is retained because it is part of the internally

consistent WRF-Chem configuration and emission speciation used for these simulations. Replacing it would require emission remapping, re-evaluation of the aerosol treatment, and complete validation of a new base configuration, which is beyond the scope of this pathway-focused sensitivity study.

More importantly, all uptake scenarios use exactly the same gas-phase mechanism, emissions, meteorological configuration, and aerosol treatment. The reported differences therefore isolate the perturbation introduced by the added branch within this model configuration. In addition, the base simulation is evaluated against O_3, NO_2, SO_2, NH_3, sulfate, nitrate, ammonium, and organic aerosol observations.

Nevertheless, we have added the gas-phase mechanism and aerosol-module dependence as a limitation of the present study.

Changes in the revised manuscript: Main text, Sect. 2.1, lines 87–89 and Summary and conclusions, lines 344–347, SI Text S1, lines 17–18.

> *"Gas-phase chemistry is represented by the SAPRC-99 mechanism."*
>
> *"Although SAPRC-99 is older than recent SAPRC mechanisms, it is retained as part of the internally consistent model configuration and emission speciation used here."*
>
> *"Absolute responses may also depend on the gas-phase chemical mechanism and aerosol module. This study uses SAPRC-99 and the incorporated CMAQ/Models-3 AERO5 module; these components are held identical across all sensitivity cases, but comparison with newer mechanisms would require emission remapping and separate model evaluation."*

Major comment 3c: primary NOCs

Reviewer comment: *Relatedly, as the study compares simulated NOC against observations, it is also important to clarify how the primary emissions of NOC are treated in your simulations. All these key model details are unclear in the current manuscript.*

Response: The anthropogenic inventory does not contain an explicit primary-NOC category, and the model has no separate primary-NOC tracer. Some nitrogen-containing organic material may be implicitly included in bulk POA or lumped organic-gas emissions, but it cannot be separately speciated or diagnosed as NOCs. Therefore, the modeled NOCs concentration represents only the product of the implemented NH_3–SOA pathway and is not a simulation of total ambient NOCs.

The Xianghe measurements sum seven quantified NOCs classes and may contain primary material and products from other secondary pathways. The model–measurement comparison is therefore used only as a conservative upper limit for the contribution of this pathway, not as source-resolved closure. This distinction is now stated in the manuscript.

Changes in the revised manuscript: Main text, Sect. 2.1, lines 94–101 and SI Text S2, lines 42–47.

> *"Primary NOC emissions are not explicitly resolved in the anthropogenic emission inventory, and the model does not include a separate primary-NOCs tracer. Some nitrogen-containing organic material may be implicitly included within bulk primary organic aerosol (POA) or lumped organic-gas emissions, but it cannot be separately speciated or diagnosed as NOCs."*
>
> *"The modeled NOCs concentration therefore represents only the product of the implemented NH_3–SOA reactive-uptake pathway and should not be interpreted as total ambient NOCs. Because the measured NOCs burden can include primary emissions and products from other secondary pathways, the model–measurement comparison provides a conservative upper limit for the contribution of this pathway."*

"Nitrogen-containing organic compounds (NOCs) are quantified from high-volume $PM_{2.5}$ filter samples at Xianghe. The measured NOCs burden is the sum of seven quantified classes—free amino acids, amines, amides, nitriles, isocyanates, urea, and cyclic NOCs (Wang et al., 2022)—and may include primary material and products from multiple secondary pathways. It is therefore used as a conservative ceiling for the modeled contribution of this pathway, not as source-resolved closure of the total atmospheric NOCs budget."

Minor comments

Minor comment 1

Reviewer comment: *Line 31: "increased by 75.7 ±6.3%" between what years?*

Response: The period is 2008–2018. We added it and corrected the grammar.

Changes in the revised manuscript: Main text, lines 33–34.

"For instance, ammonia concentrations in East Asia increased by 75.7 ± 6.3% between 2008 and 2018, with an annual growth rate of 5.80 ± 0.61% yr-1 (Van Damme et al., 2021)."

Minor comment 2

Reviewer comment: *Line 126: Define acronym IOA first.*

Response: Corrected. IOA is defined as 'index of agreement' at its first occurrence in the main text and SI.

Changes in the revised manuscript: Main text, lines 17; SI Text S3, lines 74.

"Model performance is quantified using mean bias (MB), root mean square error (RMSE), and the index of agreement (IOA) ..."

Minor comment 3

Reviewer comment: *Lines 198 & 219: "Consistent/inconsistent" reads a bit odd here because you're describing a spatial pattern rather than agreement across datasets or time. Consider wording such as 'the decrease is not spatially uniform' or 'ammonium does not decrease everywhere across the NCP".*

Response: We agree and revised both passages to describe spatial heterogeneity directly.

Changes in the revised manuscript: Main text, lines 189–190 and 209–210.

"In S(-5) (Fig. 4d), the decrease is not spatially uniform, and small positive changes occur in southeastern parts of the domain."

"In S(-4) and S(-5) (Fig. 5c, d), ammonium responses are spatially heterogeneous, with a slight increase north of Beijing."