Response to Reviewer 1

We thank the reviewer for the careful and constructive assessment. In the revised manuscript, we now distinguish the complete particulate fate of ammonia from the simplified NOCs-forming reactive uptake pathway considered in this study. We also clarify that conventional inorganic gas–particle partitioning is already represented in the model, and that the observations are used as an upper-bound constraint on the NOCs budget rather than as a fitting target. All changes quoted below are marked in red in the revised manuscript.

Critical comments

Critical comment 1

Reviewer comment: *As the authors state correctly, the uptake coefficient is considered to be the probability of a molecule to be transferred to a particle after collision. However, not only chemical reactions are considered here, but also solvation processes. Furthermore, the process could be separated into gas-phase diffusion, surface accommodation, solvation and reaction. Of course, implementing a full model framework for the uptake mechanism would be out of the scope of this study, but to account for only the reaction as a sink to the bulk of the total uptake does not seem justified. The process should be discussed following respective studies such as follows: https://doi.org/10.5194/acp-10-9059-2010*

Response: Following Crowley et al. (2010), we now state these steps explicitly and clarify the scope of the parameterization.

We retain the conventional symbol γ for consistency with the cited laboratory and modeling literature, but define it here as an effective net probability for the NOCs-forming pathway. Thus, γ represents only the specific reactive uptake leading to particulate organic-N formation and should not be interpreted as the total physical uptake coefficient of NH_3.

Changes in the revised manuscript: Main text, Sect. 2.2, lines 115–125.

> *"Total heterogeneous uptake of a gaseous species by an aerosol particle may involve gas-phase diffusion, surface accommodation, solvation, condensed-phase diffusion, and chemical reaction (Crowley et al., 2010). 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."*
>
> *"Here, γ is the net probability that an NH_3 collision with an SOA-containing particle ultimately contributes to a particulate organic-N product through the implemented pathway."*

Critical comment 2

Reviewer comment: *If only the reaction sinks are discussed, then at least the acid base reaction leading to ammonium should be part of the sink whenever there is a bit of aerosol liquid water (ALW). Arguably, even in dry aerosol conditions, the reaction with residual sulfuric acid should be a main sink. That would lead to ammonium sulfate. As a result, the shown plots in Fig. 4-8 would significantly change. It is also not discussed, if at such high formation rated of NOC would not be limited by the carbonyl fraction available for the reaction.*

Response: We agree that acid–base partitioning with sulfuric and nitric acids is a major particulate sink of NH_3. This pathway was already active in BASE and every sensitivity simulation through ISORROPIA v1.7, including ammonium-salt formation. The added parameterization neither replaces nor duplicates this inorganic pathway. Figs. 4–8 therefore shows sensitivity-minus-BASE changes caused only by adding the specific pathway while conventional inorganic partitioning remains active in both simulations.

We also agree that sustained NOCs production must be limited by finite carbonyl/dicarbonyl compounds and other reactive organic sites. The finite amount of reactive organic material is, in fact, one of the main motivations for this study. Laboratory experiments show that relatively large initial uptake coefficients decrease substantially with reaction time as reactive sites are consumed or become less accessible because of particle aging and condensed-phase diffusion limitations. Liu et al. (2015), for example, reported a decline from initial values of approximately 10^{-2}–10^{-3} toward 10^{-5} during continued NH_3 exposure.

Changes in the revised manuscript: SI Text S1, lines 18–22; main text, Sect. 3.2, lines 258–259 and 289–292.

> *"Inorganic aerosol thermodynamics are calculated using ISORROPIA version 1.7 (Nenes et al., 1998), which treats partitioning of NH_3 with sulfuric and nitric acids and ammonium-salt formation in BASE and all sensitivity simulations. The added NOCs-forming parameterization does not replace this inorganic pathway but represents an additional simplified pathway to organic nitrogen."*
>
> *"In all sensitivity simulations, the NH_3 loss calculated specifically for the implemented reactive-uptake pathway is converted into a particulate organic nitrogen product while SOA mass is not depleted."*
>
> *"Accordingly, the very high NOCs concentrations in S(-3) and S(-4) should not be interpreted as sustainable atmospheric production. Rather, these cases illustrate the overestimation that can arise when large initial coefficients are applied continuously without explicitly resolving progressive depletion or shielding of reactive organic sites."*

Critical comment 3

Reviewer comment: *Most of the results are discussed in a way that the implemented effect changes the model result away from observation and, as a result, the effect is best represented if the impact is tuned down as far as possible. The discussion if the implemented mechanism represents the current understanding and improves the results at all, are largely missing. The authors should add an additional section to the results, after the model setup and before the variation of the uptake coefficient where the general effect with a conservative estimation of the uptake coefficient is discussed. That way the reader could see the importance of the implementation of such a pathway, before the kinetic constant is tuned.*

Response: We have revised the manuscript to clarify the experimental design and establish the purpose of the sensitivity ensemble before its coefficient-dependent results are interpreted, while avoiding a separate subsection that would duplicate the same analysis.

The three coefficients, $\gamma = 10^{-3}$, 10^{-4}, and 10^{-5}, were specified a priori based on laboratory measurements and previous regional modeling studies (Liu et al., 2015; Zhu et al., 2018; Wu et al., 2021). The larger coefficients represent initial laboratory values and values adopted in previous modeling studies, whereas

10^{-5} represents the lower, late-stage value measured after reaction efficiency decreased because of finite reactive-site accessibility and diffusion limitations. The three simulations were therefore designed as a literature-based sensitivity ensemble. We made some changes accordingly in the main script.

Changes in the revised manuscript: Main text, Abstract, lines 15–17; Introduction, lines 77–81; Sect. 2.2, lines 118–121; Sect. 3.2, lines 299–301; Summary and conclusions, lines 324–327.

> *"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."*

> *"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."*

> *"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. A literature-based range of $\gamma = 10^{-5}$–10^{-3} is selected based on the laboratory results of Liu et al. (2015) and previous modeling studies (Zhu et al., 2018; Wu et al., 2021)."*

> *"Considering both the laboratory evidence for finite and declining SOA reactivity and the ceiling provided by the measured NOCs burden, we interpret $\gamma = 10^{-5}$ as a conservative upper limit for sustained operation of the NOCs-forming pathway under the present conditions."*

> *"We implemented a first-order NOCs-forming NH_3–SOA reactive-uptake pathway in WRF-Chem for a severe $PM_{2.5}$ episode over the NCP in November 2017. Three γ values (10^{-5}–10^{-3}) are selected based on laboratory and modeling literature, and the measured NOCs burden is used as an upper limit for the contribution of this pathway."*

Further comments

Further comment 4

Reviewer comment: *The authors use the term nitrogen-containing compounds (NOCs) and include all kind of NOCs, but the formation mechanism of this compound class is very variable. Ammonia uptake would likely be able to contribute to the formation of imidazoles, if sufficient concentrations of di-carbonyl compounds are available, and to the formation of imines with other carbonyls, but the formation of organonitrates is probably not a direct result of ammonia uptake. Yet, the authors cite the paper of Liu et al. 2015 to compare the behavior of the uptake coefficient. A clear distinction should be made here.*

Response: We have added explanation to further clarify that the modeled NOCs should be interpreted as lumped reduced-nitrogen organic products potentially formed through ammonia reactions with reactive organic components of secondary organic aerosols, such as carbonyl- or dicarbonyl-containing compounds. They should not be interpreted as representing all atmospheric NOCs. Organonitrates formed primarily through VOC oxidation in the presence of NOx are not represented by the implemented tracer.

Changes in the revised manuscript: Main text, Sect. 2.2, lines 127–129.

"In this study, modeled NOCs are a lumped representation of reduced-nitrogen organic products that may form when NH_3 reacts with reactive organic components of SOA, including carbonyl- or dicarbonyl-containing compounds."

Further comment 5

Reviewer comment: *Could the authors investigate the formation of clusters of ammonia and sulfuric acid in the gas-phase and related new-particle formation events and compare that to the uptake and subsequent formation of ammonium sulfate in the particle phase? A high dependency of the local sulfuric acid concentration and ALW would be expected here. This might help to investigate the effect of various uptake coefficients.*

Response: We appreciate this constructive suggestion. We agree that ammonia-sulfuric acid clustering, new-particle formation, and ammonium sulfate formation are important processes affecting the atmospheric fate of ammonia. Their relative importance is expected to depend strongly on gas-phase sulfuric acid concentrations, aerosol liquid water, particle size distribution, and ambient thermodynamic conditions.

In the present model configuration, inorganic aerosol partitioning and ammonium sulfate formation in existing particles are treated using ISORROPIA, whereas molecular NH_3–H_2SO_4 clusters and their subsequent growth are not explicitly resolved. A quantitative comparison would therefore require cluster-resolved nucleation chemistry together with constraints on gas-phase sulfuric acid, aerosol liquid water, and size-resolved newly formed particles, which are not available for the present episode. Moreover, cluster-mediated nucleation is mechanistically distinct from the NOCs-forming NH_3 uptake pathway on existing SOA investigated here. We have therefore explicitly acknowledged this limitation and the associated competition among NH_3 sinks in the revised manuscript.

Changes in the revised manuscript: Main text, Summary and conclusions, lines 347–351.

"In addition, the parameterization does not explicitly resolve molecular NH_3–H_2SO_4 clustering or cluster-mediated new-particle formation. Competition among these processes, uptake by existing particles, and ammonium sulfate formation may depend strongly on gas-phase sulfuric acid and ALW. Resolving these uncertainties requires additional field measurements, laboratory constraints, and model development."

Further comment 6

Reviewer comment: *In line 205 the authors write: 'The increase in NH_3 concentration due to the uptake reaction can also observed…' An uptake is an additional sink for NH_3, as a result the concentration should not increase. This really should be reconsidered and discussed thoroughly*

Response: We now distinguish the direct tendency from the net response of the coupled WRF-Chem system. The small positive differences in a limited number of grid cells can arise from secondary redistribution, aerosol–radiation interactions, meteorological changes, and ISORROPIA gas–particle repartitioning; they do not indicate direct NH_3 production by the uptake reaction. The spatial description was revised accordingly.

Changes in the revised manuscript: Main text, Sect. 3.2, lines 189–191 and 212–226.

"In S(-5), the decrease is not spatially uniform, and small positive changes occur in southeastern parts of the domain. Nevertheless, the domain-average NH_3 concentration decreases by 0.62% …"

"Although reactive uptake itself is strictly a sink for gas-phase NH_3, the difference between the sensitivity and BASE simulations represents the net response of all coupled chemical, thermodynamic, and dynamical processes rather than the uptake tendency alone. A two-way

coupled WRF–CMAQ study of the same NH_3–SOA uptake process have similarly predicted a local increase in NH_3 over Iowa, which is attributed to uptake-induced meteorological feedback that reduces the 10 m wind speed and atmospheric dispersion, thereby promoting NH_3 accumulation in a high-emission region (Zhu et al., 2022). WRF-Chem studies over the NCP have also shown that aerosol–radiation feedbacks can perturb surface temperature, planetary boundary layer development, and wind fields, leading to spatial redistribution of near-surface pollutants (Bei et al., 2017; Wu et al., 2019). In addition, the ISORROPIA thermodynamic module recalculates the equilibrium partitioning between gaseous NH_3 and particulate NH_4^+. Changes in temperature, relative humidity, aerosol liquid water, particle acidity, and nitrate availability may promote NH_4NO_3 dissociation and partially replenish gas-phase NH_3 (Nenes et al., 1998; Pye et al., 2020; Nenes et al., 2020). Therefore, the small positive NH_3 values in a limited number of grid cells do not indicate direct NH_3 production by the uptake reaction, but likely reflect secondary redistribution, aerosol–radiation interactions and gas–particle repartitioning within the coupled modeling system."

Minor comments

Minor comment 1

Reviewer comment: *86 should read: the y of NH_3 on SOA is assumed to be in the range of*

Response: The sentence was replaced by the clearer statement that the literature-based γ range was selected for the NOCs-forming pathway.

Changes in the revised manuscript: Main text, lines 119–121.

"A literature-based range of $\gamma = 10^{-5}$–10^{-3} is selected based on the laboratory results of Liu et al. (2015) and previous modeling studies (Zhu et al., 2018; Wu et al., 2021)."

Minor comment 2

Reviewer comment: *251: should read: much higher*

Response: Corrected. We changed the expression into 'much higher than'.

Changes in the revised manuscript: Main text, lines 261–264.

"S(-3) produces an unrealistically high mean NOCs concentration of 65.5 μg m^{-3}, much higher than the simulated SOA concentration (Fig. 8b)."

Minor comment 3

Reviewer comment: *L.205 should read: can also be observed in FIG.reference*

Response: The ambiguous sentence is removed.

Minor comment 4

Reviewer comment: *Figure references should be unique and clear. E.g. Fig 4b instead of S(-3)*

Response: We agree. Specific spatial results now use unique figure-panel references (e.g., Figs. 4b–d, 5c–d, and 8b–d) rather than scenario labels alone.

Changes in the revised manuscript: Main text, lines 189–191, 203–204 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 the S(-3) (Fig.5b), although the NH_3 uptake reaction decreases NH_3 concentrations by over 30% in the NCP, it only reduces the ammonium concentration by 5.5% or 0.86 μg m^{-3}."

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

Minor comment 5

Reviewer comment: *In the SI, some of the abbreviations are not introduced to the reader*

Response: We checked the SI throughout and defined abbreviations at first occurrence, including WRF-Chem, AERO5, SAPRC-99, SOA, VBS, FTUV, MEGAN, NCP, NOCs, IASI, VCDs, OA, POC, SOC, POA, MB, RMSE, and IOA.