We appreciate the reviewer for the useful comments. In the following, original reviewer comments are shown in black, our point-by-point responses are shown in blue, and updates on the manuscript are shown in blue and highlighted in yellow.

Reviewer Comments to Author:

The manuscript has been improved after the first-round review. I have several comments after reading the revised version and the authors' responses to the previous comments.

1. Manuscript

R3C1: The measurement period is relatively short, as already questioned by both reviewers during the first round. Only one fog event was observed although it last more than one day. In my view, this should be treated more as a case study, and it may not be sufficient to represent typical fog conditions in the studied forest. This limitation weakens the robustness of the conclusions.

One suggestion is to combine previous measurements at the same site.

Response: We thank the reviewer for this constructive suggestion. We agree that a single fog episode, despite its extended duration (~26 hours), is insufficient to draw generalizable conclusions about fog-driven nitrogen transformation in Taiwan mountain forests. Accordingly, we have reframed the study as a case study throughout the revised manuscript, including the title, abstract, introduction, and conclusion sections. We believe this framing more accurately reflects the scope of the current dataset while preserving the scientific value of the detailed process-level observations documented here. Expanded measurements across multiple fog events and seasons will be necessary to establish the broader representativeness of these findings, and we have stated this explicitly in the revised conclusion section. The specific revisions to the manuscript are summarized below:

> (Title) "Size-resolved isotope analysis reveals anthropogenic reactive nitrogen transport and transformation in Taiwan mountain forests: A case study"
>
> (Abstract: Lines 13−32): "Reactive nitrogen (Nr) species such as particulate ammonium ($p\mathrm{NH_4^+}$) and nitrate ($p\mathrm{NO_3^-}$) are critical drivers of air pollution and ecosystem health, yet their transformation processes in mountain forests are not well-characterized. We performed a one-week field campaign in a subtropical mountain forest at Xitou, Taiwan, utilizing size-segregated aerosol sampling coupled with stable isotopic techniques and Bayesian modeling. Functional groups were analyzed by Fourier-transform infrared spectroscopy (FTIR-ATR), and isotopes $\delta^{15}\mathrm{N}$ and $\delta^{18}\mathrm{O}$ were measured using gas chromatography-isotope ratio mass spectrometry (GC-IRMS) to enable quantification of $p\mathrm{NH_4^+}$ source contributions and $p\mathrm{NO_3^-}$ formation pathways. During the sampling week, typical diurnal patterns characterized by higher daytime particle concentrations were disrupted by a stagnant 26-hour fog event, which suppressed the influx of $\delta^{15}\mathrm{N}$-enriched urban plumes and promoted aqueous-phase uptake of isotopically depleted local gas-phase species into the particle phase. Under clear conditions, size-resolved $\delta^{15}\mathrm{N}$-$\mathrm{NH_4^+}$ exhibited a bell-shaped distribution peaking at the accumulation mode, whereas this gradient flattened during the fog event. Size-resolved $\delta^{15}\mathrm{N}$ and $\delta^{18}\mathrm{O}$ signatures of $p\mathrm{NO_3^-}$ revealed two distinct nitrate formation regimes: urban plumes retained $\mathrm{O_3}$-driven

oxidation signatures with higher $\delta^{18}O$, and rural/local regimes dominated by RO_2-involved processes with greater isotopic depletion and/or enhanced biogenic contributions. Bayesian source apportionment analysis constrained by $\delta^{15}N\text{-}NH_4^+$ indicated that 50−83% of NH_3 emissions originated from combustion-related sources. Concurrently, source apportionment analysis of $\delta^{18}O$ revealed that RO_2-initiated oxidation dominated daytime pNO_3^- formation (42−95%) and heterogeneous reactions contributed 6−84% at night. Although based on a short-term campaign capturing a single fog episode, this case study highlights the value of size-resolved isotopic approaches for characterizing reactive nitrogen transport and evolution under contrasting meteorological conditions, providing important mechanistic insights into processing in complex environments."

(Introduction: Lines 94−100): "While previous work at Xitou has characterized winter Nr sources (T.-Y. Chen et al., 2022), the influence of prolonged fog and stagnant atmospheric conditions on size-segregated isotopic distributions remains poorly resolved. In this case study of a one-week field campaign capturing a rare ~ 26-hour fog episode, we examine (1) how prolonged fog modifies the size-dependent isotopic structure of pNH_4^+, and (2) how the combination of $\delta^{15}N\text{-}NO_3^-$ and $\delta^{18}O\text{-}NO_3^-$ with size resolution resolves the partitioning between O_3- and RO_2-initiated nitrate formation pathways. By combining size-segregated sampling with Bayesian source apportionment, we evaluate the relative contributions of NH_3 emission sources and evaluate how fog conditions modulate pNO_3^- formation pathways."

(Conclusion: Lines 495−514) "A one-week campaign at a subtropical mountain forest in Taiwan revealed that daytime upslope transport of urban plumes consistently elevated particle concentrations above nighttime levels. Conversely, a single ~ 26-hour fog event suppressed urban input and drove progressive isotopic depletion in both $\delta^{15}N\text{-}NH_4^+$ and $\delta^{15}N\text{-}NO_3^-$. Simultaneously, size-resolved $\delta^{15}N\text{-}NH_4^+$ signatures shifted from a bell-shaped distribution with a peak at accumulation-mode under clear conditions to a flattened distribution during the fog event, while a marked decline in $\delta^{18}O\text{-}NO_3^-$ indicated a transition from O_3- to RO_2-dominated nitrate formation. Because these distinct signals are easily obscured in bulk measurements, this study underscores the value of size-segregated isotope analysis. Furthermore, Bayesian source apportionment analysis showed that 50–83% of total NH_3 was consistently attributed to combustion-related sources, suggesting that targeting combustion-related NH_3 emissions could represent an effective PM mitigation approach in Taiwan.

Overall, this study demonstrates that size-resolved isotope analysis provides process-level insights into nitrogen transport and transformation within a complex, high-altitude forest ecosystem. Nevertheless, certain analytical limits remain: accurate source apportionment of NH_3 and the precise quantification of discrete pNO_3^- formation pathways remain constrained by simplified equilibrium fractionation assumptions and overlapping isotopic end-members. Future research incorporating direct gas-phase isotope observations and controlled chamber studies of aqueous-phase reactions will help improve the differentiation between urban and biogenic contributions, enabling a more rigorous evaluation of targeted emission reduction frameworks. Finally, as this work constitutes a specialized case study from a short spring campaign capturing a single fog episode, multi-

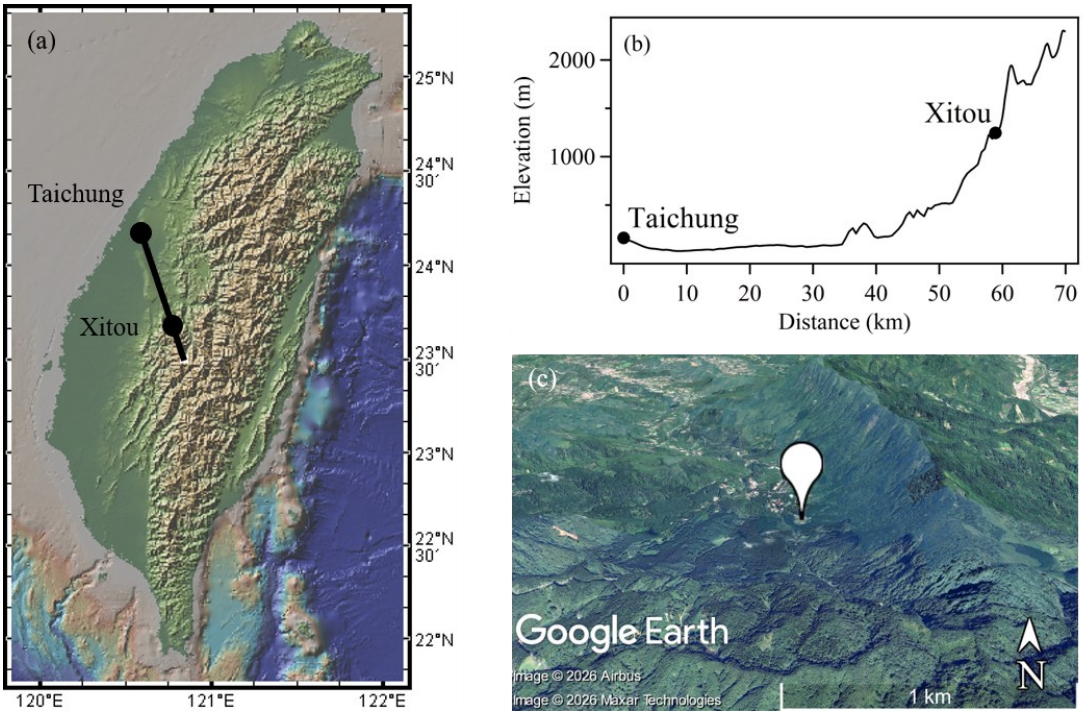
event observations across seasons and contrasting meteorological regimes are needed to establish the broader generality and long-term representativeness of Nr evolution."

R3C2: Regarding the abundance of biogenic VOCs, I agree that forested areas are generally expected to exhibit elevated levels. However, this measurement site is located near a mountain top, where air masses may be substantially diluted. Additional observational evidence or supporting measurements are needed to support this statement.

Response: We thank the reviewer for pointing this out, as it highlights a geographic misunderstanding that we have now rectified. We would like to clarify that the sampling site at Xitou is not located near a mountain summit, but rather situated at 1,179 m a.s.l. within a deeply carved valley-basin terrain. The site is enclosed on three sides (south, east, and west) by substantially higher mountain ridges (up to ~2,000–3,000 m a.s.l.), with a single opening to the northwest facing low-altitude urbanized and agricultural regions (Taichung and surrounding areas). We acknowledge that this topography was not clearly illustrated in the original version of Figure S1, and we have updated the introduction of the main text and the topographic map in the Supplementary Information to explicitly show this basin-like configuration as follows:

(Lines 77−82) "Xitou, a representative cloud forest in central Taiwan, offers a suitable field setting to investigate these gaps. The site is situated at 1,179 m a.s.l. in a valley-basin terrain enclosed on three sides by higher mountain ridges (up to ~ 2,000−3,000 m a.s.l.) to the south, east, and west, with a topographic opening toward the northwest facing urbanized and agricultural lowlands. This specialized geographic configuration drives predictable valley-mountain breeze circulations that transport anthropogenic plumes into a biogenic-rich environment where Nr accounts for 13−23% of PM_{10} mass (Chen et al., 2021). […]"

(Figure S1) "



> **Figure S1.** (a) Horizontal distance between the sampling location (Xitou) and Taichung, plotted using GeoMapApp (ver 3.7.4). (b) Elevation profile along the same transect between the sampling location (Xitou) and Taichung. (c) Three-dimensional terrain visualization of the sampling site (white pin) at Xitou. Imagery from Google Earth Pro."

Regarding direct observational evidence for biogenic VOC abundance at this site, previous field measurements using a PTR-ToF-MS at Xitou during October–November 2015 confirmed isoprene, monoterpenes, and sesquiterpenes along with their first-generation oxidation products (pinonaldehyde, norpinonic acid, pinic acid, and pinonic acid) (Salvador et al., 2020). Critically, the diurnal profiles of isoprene, pinonaldehyde, norpinonic acid, and pinic acid showed strong daytime peaks that tightly correlated with peak UV radiation. This synchronous UV-correlated signal established that these compounds are mainly produced in situ via active local photochemistry within the canopy rather than being diluted by high-altitude clean air masses or transported from long distances. Furthermore, while the campaign by Salvador et al. (2020) was conducted during the fall–winter transition (a period of lower temperatures and reduced biological activity), it still detected substantial biogenic activity. Because BVOC emissions from subtropical forests are strongly temperature- and light-dependent, emission rates during our spring campaign, characterized by warmer temperatures and higher solar radiation, are expected to be comparable to or higher than those reported in the fall–winter study. We have revised the relevant text in the manuscript to integrate this empirical evidence and added a note addressing the lack of concurrent spring BVOC measurements as a campaign limitation.

> (Lines 80−85) "[…] This specialized geographic configuration drives predictable valley-mountain breeze circulations that transport anthropogenic plumes into a biogenic-rich environment where Nr accounts for 13−23% of PM_{10} mass (Chen et al., 2021). The densely vegetated catchment and enclosed terrain promote the accumulation of locally emitted biogenic VOCs, supporting active RO_2 chemistry at the site (Salvador et al., 2020). This mixing vessel is characterized by frequent fog and persistent humidity, with a mean relative humidity (RH) above 80%. […]"

R3C3: L32–33: Please provide references to support this statement.

Response: We thank the reviewer for the reminder. The relevant references have been added to the revised manuscript to support this statement in the introduction section as follows:

> (Lines 35−37) "Anthropogenic activities have significantly altered the global reactive nitrogen (Nr) cycle, with profound implications for climate change (Pinder et al., 2012), biodiversity loss (Humbert et al., 2016), acid deposition (Doney et al., 2007), and regional air quality degradation (Cheng et al., 2016)."

R3C4: L110, L127, L169, and elsewhere throughout the manuscript: Please carefully check the reference formatting.

Response: We thank the reviewer for pointing this out. Inclusion of authors' initials in the text citations was initially intended to disambiguate different references from the same publication year whose first authors shared identical surnames (e.g., W. -C. Huang et al., 2024 vs. P. -C. Huang et al., 2024; T.-Y. Chen et al., 2022 vs. Z.-L. Chen et al., 2022). However, we realize that the original formatting did not adhere strictly to standard guidelines (e.g., the APA style). We

have carefully reviewed and corrected the reference formatting throughout the entire revised manuscript. A few examples are provided as follows:

> (Lines 94–96) "While previous work at Xitou has characterized winter Nr sources (T.-Y. Chen et al., 2022), the influence of prolonged fog and stagnant atmospheric conditions on size-segregated isotopic distributions remains poorly resolved."
>
> (Lines 116–118) "Among the trace gases measured by AQB, carbon monoxide (CO) was selected as the primary tracer for analysis due to its reliable calibration (W.-C. Huang et al., 2024)."
>
> (Lines 135) "Detailed descriptions of isotope measurement procedures can be found in T.-Y. Chen et al. (2022)."
>
> (Lines 178–181) "These sources can be categorized into two groups based on their δ^{15}N-NH_3 signatures: (1) volatilization-related sources (fertilizer and waste) with lower δ^{15}N-NH_3 values, and (2) combustion-related sources (NH_3 slip and fossil fuel emissions) with higher values (Z.-L. Chen et al., 2022)."
>
> (Lines 260–262) "These results are consistent with previous winter observations at this site (−3.7‰ to 21.39‰, with an average of 11.95 ± 2.65‰) (T.-Y. Chen et al., 2022)."
>
> (Lines 352–354) "These findings underscore the need for regional emission control strategies targeting anthropogenic NH_3 sources, which may represent a more cost-effective mitigation strategy for Taiwan, potentially yielding greater air quality benefits than NO_x or SO_2 reductions alone (P.-C. Huang et al., 2024)."

R3C5: L405–408: Was there any direct measurement or observational evidence from this site to support this argument?

Response: We thank the reviewer for this important point. We acknowledge that direct OH and RO_2 measurements were not conducted during our field campaign. However, we suggest that nighttime OH and RO_2 production at the Xitou forest site is chemically plausible. Salvador et al. (2020) reported nighttime monoterpene accumulation at the same site. Monoterpene oxidation by O_3 and NO_3 leads to the formation of OH and RO_2 at night, thereby allowing the NO_3 formation pathway from $P_{RO_2\text{-}OH1}$. This part was updated in the revised manuscript in Section 3.4.2 as follows:

> (Lines 447–455) "[…] This high $P_{RO_2\text{-}OH1}$ contribution might stem from the residual pNO_3^- from daytime, or nighttime ·OH generated via reactions between O_3 and alkenes/terpenes. The latter pathway is chemically plausible at Xitou given the documented nighttime accumulation of monoterpenes at this site (Salvador et al., 2020) and the known positive correlation between monoterpene concentrations and dark OH/HO_2 radical production in forested environments, which likely proceeds via ozonolysis (Kroll et al., 2001; Aschmann et al., 2002; Kanaya et al., 2007). However, direct OH/RO_2 measurements would be needed in future campaigns to quantify the relative importance of these mechanisms. The weak night-to-night covariation between δ^{18}O-NO_3^- and CO concentrations further suggests that nighttime nitrate isotope signatures are governed by the interplay among fog processing, reduced advection, and heterogeneous chemistry, rather than transport intensity alone."

R3C6: Figure S7: A height of 10 m AGL was selected for the back-trajectory analysis, which is not commonly used. At such low altitude, surface turbulence may substantially affect the trajectories, and the results may not represent regional transport well. However, I am not an expert in meteorology, so I suggest the authors further justify this setup and discuss the associated uncertainties.

Response: We thank the reviewer for raising this concern. The initial selection of 10 m AGL was chosen to best represent the near-surface conditions directly feeding the sampling inlet. However, we acknowledge that back-trajectories initialized very close to the surface can occasionally be sensitive to localized wind shear and surface friction parameterization within the meteorological input data. To verify the robustness of our regional transport conclusions and address this concern, we conducted 24-hour backward trajectories initialized at five distinct heights spanning the surface layer to the well-mixed planetary boundary layer: 10, 50, 100, 250, and 500 m AGL. As shown in the newly added Figure S8, the calculated transport pathways are highly consistent across all five initialization heights during the studied period. This close agreement demonstrates that the synoptic-scale and regional advection pathways steering the air masses toward Xitou are robustly captured by the model, and that surface-layer turbulence parameterizations do not artificially skew the inferred origin of the plumes. Based on this verified stability, we have retained the 10 m AGL trajectory in the main text to represent the surface layer, aligning with the physical sampling height, while incorporating the sensitivity analysis results and a discussion of initialization uncertainties into the Supplementary Information.

> (Figure S7) "NOAA HYSPLIT 24-hour backward trajectories for air parcels arriving at the receptor site (23°40’12” N, 120°47’54” E) at 10 m above ground level (AGL) to reflect near-surface intake conditions. The back trajectories were calculated using the Global Forecasting System (GFS) meteorological fields at 0.25° × 0.25° spatial resolution. Trajectories are shown for arrival times of (a) 04:00 UTC (12:00 LT) and (b) 17:00 UTC (01:00 LT). Analyses were performed using the NOAA Air Resources Laboratory HYSPLIT model (https://www.ready.noaa.gov/HYSPLIT.php). The trajectory arriving at 12:00 LT on 20 and 21 April (pink and light blue) transited through agriculturally dominated lowland areas (Yunlin and Chiayi counties) prior to reaching the receptor site. Sensitivity tests using alternative initialization heights ranging from 10 m to 500 m AGL yielded consistent transport pathways (see Fig. S8), confirming that the regional-scale transport patterns described here are robust against surface-layer simulation uncertainties.”
>
> (Figure S8) “

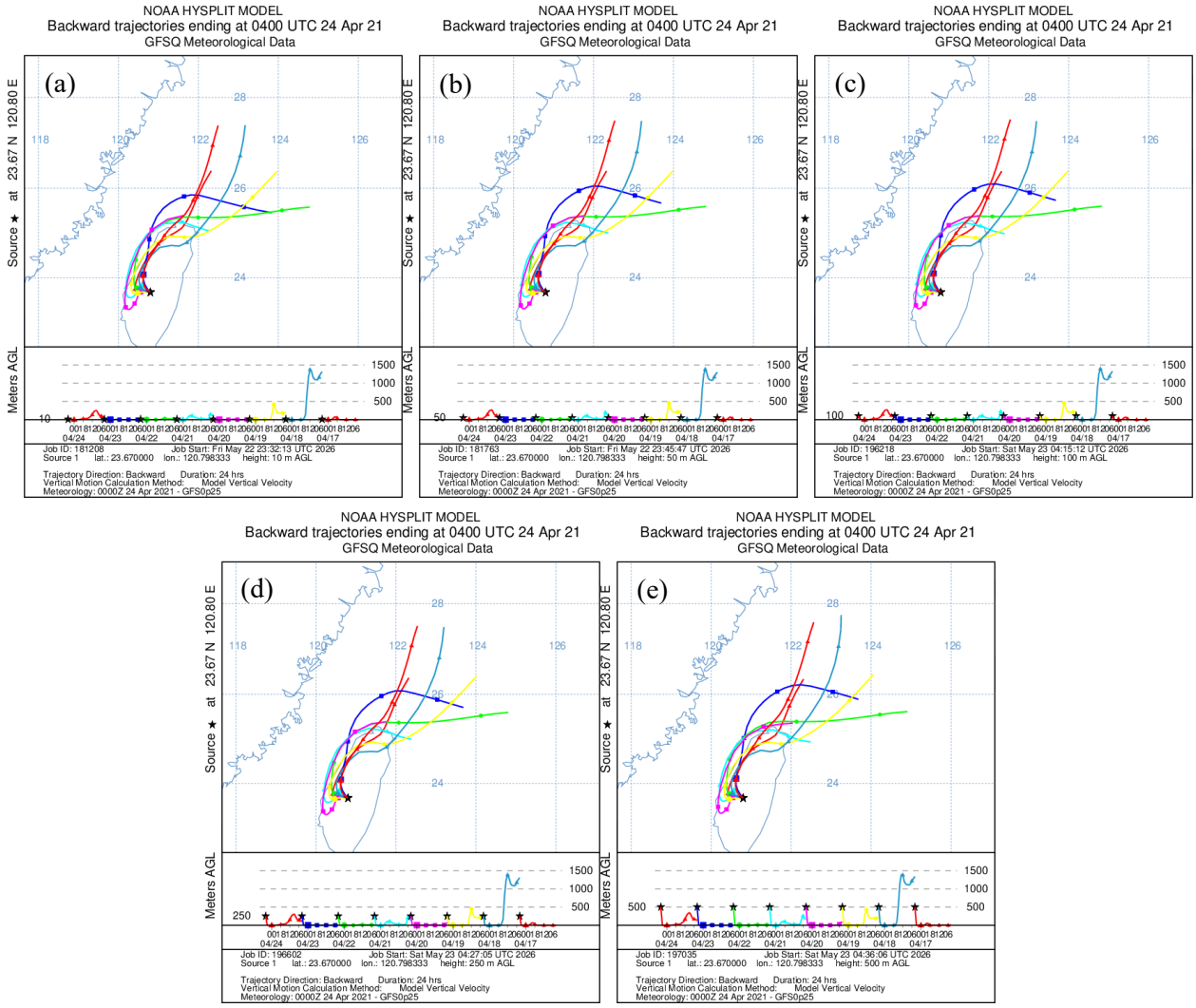


Figure S8. NOAA HYSPLIT 24-hour backward trajectories for air parcels arriving at the Xitou receptor site (23°40'12" N, 120°47'54" E) initiated at: (a) 10 m, (b) 50 m, (c) 100 m, (d) 250 m, and (e) 500 m above ground level. Calculations were driven by the Global Forecasting System (GFS) meteorological fields at 0.25° × 0.25° spatial resolution for an arrival time of 04:00 UTC (12:00 LT). The strong spatial consistency across these vertical layers demonstrates that the inferred transport pathways are robust to the choice of starting altitude."

R3C7: Figure S8: Please provide more information on how CMAQ simulates gaseous NH_3.

Response: We thank the reviewer for requesting this clarification regarding the modeling framework. We have expanded the description of the Community Multiscale Air Quality (CMAQ) model setup in both the main text and the Supplementary Information to detail how gaseous NH_3 emissions, chemical transport, and phase partitioning are handled. Specifically, the CMAQ model (v4.7.1) utilized the SAPRC99 gas-phase chemical mechanism paired with the AERO5 aerosol module. Within this framework, the thermodynamic gas-particle partitioning of the dynamic NH_3/NH_4^+ system is explicitly calculated using the embedded ISORROPIA

thermodynamic equilibrium module. This module dynamically computes the equilibrium between gaseous ammonia and particulate ammonium based on hourly variations in local ambient temperature, relative humidity, and the concentrations of competing inorganic species (such as sulfate, nitrate, and chloride). To validate this simulation configuration, we compared the CMAQ-derived particulate NH_4^+ mass concentrations against our empirical laboratory measurements. The modeled concentrations agreed with the observational data within a 20% uncertainty range, providing confidence in the simulated gaseous NH_3 baseline used to calculate the particle-phase fraction (*f*). We have updated the method section of the main text and the renumbered Figure S9 caption to reflect these technical details as follows:

> (Lines 168−176) "[…] The *f* values were derived from the Community Multiscale Air Quality (CMAQ) model (version 4.7.1), incorporating meteorological data from the Weather Research and Forecasting (WRF) model (version 3.7.1). The anthropogenic emission database was from the Taiwan Emission Data System (version 12) (Tsai et al., 2024) while biogenic VOC emissions were calculated with the Model of Emission of Gases and Aerosols from Nature algorithm (MEGAN, version 2.04) (Tsai et al., 2015). Concentration of NH_3 and NH_4^+ were simulated using the SAPRC99-AERO5 chemical mechanism, in which inorganic gas-particle partitioning of NH_3/NH_4^+ was computed by the ISORROPIA thermodynamic equilibrium module embedded within AERO5. The model-simulated pNH_4^+ mass concentrations agreed with laboratory measurements within a 20% uncertainty range (Fig. S2)."
>
> (Figure S2) "pNH_4^+ concentrations measured by fluorometric analysis versus those estimated by the Community Multiscale Air Quality (CMAQ) model following the model configuration described in Sect. 2.3.1."
>
> (Figure S9) "Figure S9. Temporal variation of (a) gaseous NH_3 and (b) particulate NH_4^+ (pNH_4^+) concentrations simulated by the Community Multiscale Air Quality (CMAQ) model following the configuration described in Sect. 2.3.1), with pNH_4^+ also shown from fluorometric analysis for comparison, and (c) the particle-phase fraction *f* (= $[pNH_4^+]$ / ($[NH_3]$ +$[pNH_4^+]$)) estimated from CMAQ-simulated concentrations."

R3C8: In the conclusion section, please more clearly highlight the uncertainties associated with the short measurement period, especially because most conclusions regarding fog impacts rely on a single fog event.

Response: We thank the reviewer for this important comment. We agree that grounding the conclusions in the context of a single fog event is critical for scientific accuracy. Accordingly, we have revised the conclusion section to explicitly state the inherent uncertainties, statistical limitations, and temporal boundaries of a one-week sampling window containing a single 26-hour fog event. We have also reframed our quantitative source apportionment values as "indicative" rather than long-term baseline averages, and explicitly outlined the necessity for future multi-event campaigns spanning different seasons to establish broader generalizability.

Crucially, we have also clarified the unique value of this specific event: while a continuous 26-hour fog event is meteorologically rare at this site, its prolonged duration effectively isolated the forest canopy from upslope urban plumes. This provided a rare, uninterrupted window to track

local chemical evolution and process-level isotopic fractionation dynamics in situ. The revised conclusion section has been modified as follows:

> (Lines 495−514) "A one-week campaign at a subtropical mountain forest in Taiwan revealed that daytime upslope transport of urban plumes consistently elevated particle concentrations above nighttime levels. Conversely, a single ~ 26-hour fog event suppressed urban input and drove progressive isotopic depletion in both δ^{15}N-NH_4^+ and δ^{15}N-NO_3^-. Simultaneously, size-resolved δ^{15}N-NH_4^+ signatures shifted from a bell-shaped distribution with a peak at accumulation-mode under clear conditions to a flattened distribution during the fog event, while a marked decline in δ^{18}O-NO_3^- indicated a transition from O_3- to RO_2-dominated nitrate formation. Because these distinct signals are easily obscured in bulk measurements, this study underscores the value of size-segregated isotope analysis. Furthermore, Bayesian source apportionment analysis showed that 50–83% of total NH_3 was consistently attributed to combustion-related sources, suggesting that targeting combustion-related NH_3 emissions could represent an effective PM mitigation approach in Taiwan.
>
> Overall, this study demonstrates that size-resolved isotope analysis provides process-level insights into nitrogen transport and transformation within a complex, high-altitude forest ecosystem. Nevertheless, certain analytical limits remain: accurate source apportionment of NH_3 and the precise quantification of discrete pNO_3^- formation pathways remain constrained by simplified equilibrium fractionation assumptions and overlapping isotopic end-members. Future research incorporating direct gas-phase isotope observations and controlled chamber studies of aqueous-phase reactions will help improve the differentiation between urban and biogenic contributions, enabling a more rigorous evaluation of targeted emission reduction frameworks. Finally, as this work constitutes a specialized case study from a short spring campaign capturing a single fog episode, multi-event observations across seasons and contrasting meteorological regimes are needed to establish the broader generality and long-term representativeness of Nr evolution."

2. Response to Reviewers' Comments

R3C9: R1C1: Similar observations and conclusions have been reported in previous studies, which is acceptable since this is what the data show. However, the authors still need to better justify the scientific significance and/or policy relevance of the study in order to satisfy ACP standards.

Response: We thank the reviewer for this comment. Regarding scientific significance, the study's contributions rest on two process-level insights that are not accessible from previous bulk or single-isotope measurements, and are therefore appropriate for ACP's scope on aerosol transformation processes:

(1) Fog-driven collapse of size-dependent isotopic structure. The characteristic accumulation-mode enrichment of δ^{15}N-NH_4^+ under clear conditions disappears during the prolonged fog episode, demonstrating that aqueous-phase re-equilibration and wet scavenging can override transport-derived isotopic size structure, a process invisible to bulk measurements and not previously characterized.

(2) Oxidation regime shift constrained by dual isotopes. By combining $\delta^{15}N$ and $\delta^{18}O$ with size resolution, we document a marked transition from O_3-influenced to RO_2-dominated nitrate formation during fog, providing observational constraints on oxidation pathway partitioning in a high-humidity subtropical mountain environment.

We present these as findings from a single well-characterized fog case study that demonstrate the mechanistic potential of the size-resolved dual-isotope framework, rather than as generalizable conclusions. Expanded measurements across multiple fog events and seasons will be needed to establish their broader representativeness, as explicitly stated in the revised Conclusions. These points are updated in the introduction of the revised manuscript as follows:

> (Lines 94−100) "While previous work at Xitou has characterized winter Nr sources (T.-Y. Chen et al., 2022), the influence of prolonged fog and stagnant atmospheric conditions on size-segregated isotopic distributions remains poorly resolved. In this case study of a one-week field campaign capturing a rare ~ 26-hour fog episode, we examine (1) how prolonged fog modifies the size-dependent isotopic structure of pNH_4^+, and (2) how the combination of $\delta^{15}N\text{-}NO_3^-$ and $\delta^{18}O\text{-}NO_3^-$ with size resolution resolves the partitioning between O_3- and RO_2-initiated nitrate formation pathways. By combining size-segregated sampling with Bayesian source apportionment, we evaluate the relative contributions of NH_3 emission sources and evaluate how fog conditions modulate pNO_3^- formation pathways."

Regarding policy relevance, the finding that combustion-related NH_3 sources dominate particulate ammonium (50–83%; robust to 88–95% across all sensitivity tests) provides independent observational support for prioritizing anthropogenic NH_3 emission controls as a cost-effective $PM_{2.5}$ mitigation strategy in Taiwan. This part was updated in the conclusion of the revised manuscript as follows:

> (Lines 500−504): "[…] Because these distinct signals are easily obscured in bulk measurements, this study underscores the value of size-segregated isotope analysis. Furthermore, Bayesian source apportionment analysis showed that 50–83% of total NH_3 was consistently attributed to combustion-related sources, suggesting that targeting combustion-related NH_3 emissions could represent an effective PM mitigation approach in Taiwan."

R3C10: R1C4: The authors proposed multiple isotope-depleting mechanisms during the fog event. However, further evidence is needed to demonstrate that these lower isotopic values do not normally occur simultaneously under non-fog conditions. This would help establish the uniqueness of the fog event and strengthen the scientific significance of the study.

Response: We thank the reviewer for this constructive suggestion. We agree that low values of individual isotopic tracers occur outside the fog period, as discussed in Sections 3.3.1 and 3.4.1 in response to R1C4. However, the simultaneous co-depletion of all three isotopic tracers ($\delta^{15}N\text{-}NH_4^+$, $\delta^{15}N\text{-}NO_3^-$, and $\delta^{18}O\text{-}NO_3^-$) was unique to the fog episode in this campaign. During non-fog periods when $\delta^{15}N\text{-}NH_4^+$ was low (19N and 21D), neither $\delta^{15}N\text{-}NO_3^-$ nor $\delta^{18}O\text{-}NO_3^-$ showed correspondingly low values. When $\delta^{15}N\text{-}NO_3^-$ was low during a non-fog period (20N), $\delta^{15}N\text{-}NH_4^+$ remained within the range typical of clear conditions. Similarly, non-fog periods with relatively low $\delta^{18}O\text{-}NO_3^-$ were not accompanied by concurrent depletion of the nitrogen isotopic

tracers. These single-tracer anomalies during clear periods are attributable to individual source or transport effects, as discussed in Sections 3.3.1 and 3.4.1.

In contrast, during the fog episode, the decrease of $\delta^{18}O\text{-}NO_3^-$ from 70.44‰ (18D) to ~34‰ (18N and 19D) reflects a shift from O_3- to RO_2-dominated nitrate formation that is mechanistically coupled to fog-driven suppression of urban oxidant input, a process distinct from source region effects alone. The simultaneous depletion and its monotonic progression from 18D through 19D are consistent with a coherent set of co-occurring mechanisms, including atmospheric stagnation, wet scavenging, aqueous-phase re-equilibration, and an oxidation regime shift, which rarely occurred in combination during non-fog low-value periods.

We acknowledge that with only one fog episode in this campaign, a statistical demonstration of uniqueness is not feasible, consistent with our reframing of this study as a case study throughout the revision. We have added the following sentence to Section 3.4.2 to clarify this point.

> (Lines 463−471) "[…] Together, these foggy conditions favor locally derived low-$\delta^{18}O$ RO_2 cycling as the predominant oxidant driving nitrate formation. While low values of individual isotopic tracers were occasionally observed during non-fog periods, attributable to source foodprints effects or transport-induced fractionation (Sect. 3.3.1 and 3.4.1), the fog episode represents the unique period during this campaign where simultaneous depletion across all three tracers ($\delta^{15}N\text{-}NH_4^+$, $\delta^{15}N\text{-}NO_3^-$ and $\delta^{18}O\text{-}NO_3^-$) occurred with coherent temporal progression. This multi-tracer convergence supports the concurrent operation of several fog-induced processes: intense atmospheric stagnation, progressive wet scavenging, aqueous-phase re-equilibration with isotopically depleted gas-phase species, and a mechanistic shift toward RO_2-dominated oxidation, all of which are mechanistically coupled to fog conditions rather than to individual source or regional transport effects."

R3C11: R1C6: I suggest the authors tone down the claims regarding "novel" observational findings, since only one fog event was observed during the measurement period.

Response: We thank the reviewer for this comment. We acknowledge that claims of novelty should be toned down, given that only one fog event was observed during the measurement period. We have carefully reviewed the revised manuscript and confirmed that the word "novel" appears only in the response to R1C6, not in the manuscript itself. We have also moderated phrases with similar connotations in the revised manuscript as follows:

Original: "These findings emphasize the importance of controlling urban NO_x and combustion-related NH_3 emissions and demonstrate how size-resolved isotope analysis elucidates aerosol evolution along transport pathways."

> Updated: (Lines 29−32) "Although based on a short-term campaign capturing a single fog episode, this case study highlights the value of size-resolved isotopic approaches for characterizing reactive nitrogen transport and evolution under contrasting meteorological conditions, providing important mechanistic insights into processing in complex environments."

Original: "Xitou, a representative cloud forest in central Taiwan, serves as an ideal natural laboratory to address these gaps."

Updated: (Lines 77−78) “Xitou, a representative cloud forest in central Taiwan, offers a suitable field setting to investigate these gaps.”

We also revised the conclusion to emphasize that this study should belong to a case study:

(Lines 495−514) “A one-week campaign at a subtropical mountain forest in Taiwan revealed that daytime upslope transport of urban plumes consistently elevated particle concentrations above nighttime levels. Conversely, a single ~ 26-hour fog event suppressed urban input and drove progressive isotopic depletion in both δ^{15}N-NH_4^+ and δ^{15}N-NO_3^-. Simultaneously, size-resolved δ^{15}N-NH_4^+ signatures shifted from a bell-shaped distribution with a peak at accumulation-mode under clear conditions to a flattened distribution during the fog event, while a marked decline in δ^{18}O-NO_3^- indicated a transition from O_3- to RO_2-dominated nitrate formation. Because these distinct signals are easily obscured in bulk measurements, this study underscores the value of size-segregated isotope analysis. Furthermore, Bayesian source apportionment analysis showed that 50–83% of total NH_3 was consistently attributed to combustion-related sources, suggesting that targeting combustion-related NH_3 emissions could represent an effective PM mitigation approach in Taiwan.

Overall, this study demonstrates that size-resolved isotope analysis provides process-level insights into nitrogen transport and transformation within a complex, high-altitude forest ecosystem. Nevertheless, certain analytical limits remain: accurate source apportionment of NH_3 and the precise quantification of discrete pNO_3^- formation pathways remain constrained by simplified equilibrium fractionation assumptions and overlapping isotopic end-members. Future research incorporating direct gas-phase isotope observations and controlled chamber studies of aqueous-phase reactions will help improve the differentiation between urban and biogenic contributions, enabling a more rigorous evaluation of targeted emission reduction frameworks. Finally, as this work constitutes a specialized case study from a short spring campaign capturing a single fog episode, multi-event observations across seasons and contrasting meteorological regimes are needed to establish the broader generality and long-term representativeness of Nr evolution.”

R3C12: R1C8: I suggest briefly introducing the relevant mechanisms when defining the terminology, since many new terms are introduced in this section. Otherwise, the discussion remains difficult to follow.

Response: We appreciate the reviewer for this suggestion. To improve readability and clarity, we have compiled a comprehensive summary table (Table 1) in the revised manuscript. This table categorizes the six distinct HNO_3 formation pathways alongside their theoretical δ^{18}O-NO_3^- isotope ranges calculated using an established mass-balance framework (Walters and Michalski, 2016). The text and table have been updated in Section 2.3.2 of the revised manuscript as follows:

(Lines 192−195) “[…] Nighttime analysis focused on $P_{RO_2\text{-het}}$ (50−69‰) and $P_{O_3\text{-het}}$ (73−102‰), while retaining $P_{RO_2\text{-OH1}}$ to account for residual daytime nitrate that persists into the night. The reaction sequences and resulting δ^{18}O-NO_3^- ranges for each pathway are summarized in Table 1. Sensitivity analyses regarding pathway exclusion are detailed in Supplementary Description S4, Tables S3 and S4.

(Lines 197−200) “Table 1. Summary of the six HNO_3 formation pathways evaluated in this study, defined by their primary oxidant (RO_2 or O_3) and terminal mechanism (OH or heterogeneous reactions), with the corresponding $\delta^{18}O\text{-}NO_3^-$ ranges calculated using a mass-balance approach assuming no kinetic isotope fractionation (Walters and Michalski, 2016).”

Pathway	Reaction sequence	$\delta^{18}O\text{-}NO_3^-$ (‰)
$P_{RO_2\text{-}OH1}$	$NO + RO_2 \rightarrow NO_2 + RO;\ NO_2 + OH\left(\delta^{18}O\text{: } -15 \text{ to } 0‰\right) \rightarrow HNO_3$	33−49
$P_{RO_2\text{-}OH2}$	$NO + RO_2 \rightarrow NO_2 + RO;\ NO_2 + OH\left(\delta^{18}O\text{: } 38 \text{ to } 61‰\right) \rightarrow HNO_3$	50−69
$P_{RO_2\text{-}het}$	$NO + RO_2 \rightarrow NO_2 + RO;$ $NO_2 + O_3 \rightarrow NO_3 + O_2;\ NO_3 + NO_2 \rightarrow N_2O_5; N_2O_5 + H_2O \rightarrow 2HNO_3$	50−69
$P_{O_3\text{-}OH1}$	$NO + O_3 \rightarrow NO_2 + O_2;\ NO_2 + OH\left(\delta^{18}O\text{: } -15 \text{ to } 0‰\right) \rightarrow HNO_3$	55−81
$P_{O_3\text{-}OH2}$	$NO + O_3 \rightarrow NO_2 + O_2;\ NO_2 + OH\left(\delta^{18}O\text{: } 38 \text{ to } 61‰\right) \rightarrow HNO_3$	73−102
$P_{O_3\text{-}het}$	$NO + O_3 \rightarrow NO_2 + O_2;$ $NO_2 + O_3 \rightarrow NO_3 + O_2;\ NO_3 + NO_2 \rightarrow N_2O_5; N_2O_5 + H_2O \rightarrow 2HNO_3$	73−102

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