Response to Reviewer 1

We appreciate the anonymous reviewer for the thoughtful reviews and comments. We have carefully considered the suggestions and revised the manuscript accordingly. The reviewer comments are in blue, our comments are in black, and modifications to the manuscript are in red.

General response

This work investigated the driving factors of nitrate in PM_1 using ML approach. In fact, numerous of studies have revealed the nitrate formation mechanism using thermodynamic analysis, box model analysis, as well as ML. This work is not novel enough for understanding the nitrate formation in East Asia, and the measurement dataset is limited extent.

Thank you for this assessment. We agree that the original manuscript did not state the scope and novelty of the study clearly enough. In the revision, we no longer present the analysis as establishing a broadly transferable nitrate-formation mechanism. Instead, we frame the work as a campaign-scale interpretation of observed wintertime PM_1 nitrate variability in Seoul, constrained by the available measurements and supported by XGBoost-SHAP, RH-history, ALWC, campaign-regime, and thermodynamic sensitivity diagnostics.

We also revised the manuscript to distinguish established nitrate-relevant processes from the campaign-specific contribution of this study. RH effects, temperature effects, and gas-particle partitioning are treated as established atmospheric processes used to interpret this Seoul winter case, while SHAP values are described as predictor attributions within the fitted model rather than chemical source apportionment. The specific responses below describe how the text was revised to address NO_x/VOC chemistry, model inputs and validation, RH interpretation, and measurement limitations related to HNO_3/NH_3, aerosol pH, viscosity, and wet-particle morphology.

We would also like to state positively what we believe this study still contributes within its revised, campaign-specific scope. Beyond confirming established processes, the value of this work is the integrated, observation-constrained characterization of a specific and reproducible wintertime process window in Seoul: the persistence of PM_1 nitrate enhancement through the late-morning RH-decline period, interpreted jointly through XGBoost-SHAP predictor attribution, reverse-mode ISORROPIA-II aerosol-water diagnostics, RH-history classification, and back-trajectory analysis applied to high-time-resolution HR-ToF-AMS composition. We do not claim a new chemical mechanism or a transferable formation pathway. Rather, we provide a measurement-grounded account of when, and under which coupled meteorological and aerosol-water conditions, wintertime nitrate accumulates in this megacity, together with an explicit statement of the gas-phase (HNO_3, NH_3) and aerosol pH/phase-state measurements that future campaigns would need to close the remaining mechanistic gaps. We hope this clarified

framing addresses the novelty concern while keeping all conclusions within the limits of the available observations.

Comment 1

From the perspective of nitrate formation mechanisms, nitrate formation is not solely related to NO_x. In fact, nitrate formation results from the combined action of multiple reactions. Each of these reactions is influenced by numerous factors, which is why the factors affecting nitrate formation are highly complex. The parameters considered in this manuscript are far from sufficient to assess the primary drivers of nitrate formation. For example, simulation results based on the box model suggest that VOCs are also important precursors for nitrate formation, but they were not considered in this study.

We agree that nitrate formation cannot be attributed solely to NO_x and that the variables used in this study are insufficient to resolve the full nitrate-formation mechanism, including VOC-related pathways. We revised Section 2.2 to describe the XGBoost inputs as available measured, reanalysis, and trajectory predictors used to interpret observed PM_1 nitrate variability, not as a complete chemical mechanism.

We also softened the NO_2 interpretation. NO_2 is now described as the strongest statistical predictor and measured reactive-nitrogen proxy in this winter dataset. VOC-derived reservoirs such as PAN and particle organic nitrates are acknowledged as unmeasured, and their expected photochemical dependence is linked to the seasonal comparison rather than treated as resolved in the winter model.

Manuscript edits:

1. **Line 87:** "Briefly, for predicting nitrate formation, the NO_2 and O_3 from AirKorea were utilized to represent the concentration of nitrate precursor and titration agent, respectively. The temporal variables including "hour of the day (HoD)" and "day of the week (DoW)" were also used to represent patterns in human activities. For the variables related to photochemical and heterogeneous formation, meteorological features include RH, temperature, solar radiation wind speed, wind direction, and solar radiation were taken from a nearby meteorological monitoring site (37.60° N, 127.0° E) within the Korea Meteorological Administration (KMA) network." **After:** "Briefly, for predicting nitrate concentration, the NO_2 and O_3 from AirKorea were used as measured gas-phase predictors related to nitrate precursor availability and oxidant/titration conditions, respectively. The temporal variables including "hour of the day (HoD)" and "day of the week (DoW)" were also used to represent recurring patterns in human activities. Meteorological predictors related to photochemical conditions, heterogeneous processes, and ventilation included RH, temperature, wind speed, wind direction, and solar radiation were taken from a nearby meteorological monitoring site (37.60° N, 127.0° E) within the Korea Meteorological Administration (KMA) network."

2. **Line 193:** "NO_2, the primary precursor of nitrate, was identified as the most significant contributor, accounting for 31.6% of nitrate formation during winter (Fig. 2a)." **After:** NO_2 showed the largest SHAP relative contribution (31.6%) to predicted PM_1 nitrate variability, indicating that NO_2 was the strongest statistical predictor in this winter dataset (Fig. 2a), and the remaining SHAP attribution was distributed among meteorological and temporal predictors."
3. **Line 202:** "The observed association between NO_2 and nitrate, supported by strong correlations and consistent diurnal patterns, confirms NO_2 as a key precursor and highlights its central role in wintertime nitrate formation." After: "These winter observations indicate that measured NO_2 availability was closely linked to PM_1 inorganic nitrate variability. VOC-derived reactive-nitrogen reservoirs, including PAN and particle organic nitrates, were not measured, so their contributions cannot be quantified here. However, prior ground-based studies suggest that, because these reservoirs are photochemically driven, their influence may be more relevant during photochemically active seasons with limited wet removal, such as spring and autumn in urban Seoul (Lee et al., 2013)."

Comment 2

Why were these specific parameters included in the ML model? Could the author provide the training and testing results for the machine learning model, as well as the results of the model's transferability validation? How were the results for RH increase and decrease in Figure 3 derived? Were they obtained through a sensitivity analysis of the machine learning model?

We agree. We added the predictor rationale and 10-fold model performance ($R^2 = 0.954 \pm 0.008$, MAE = 0.566 $\pm$ 0.022 μg m^{-3}). We also replaced the broad transferability claim with a campaign-specific statement of wintertime PM_1 nitrate controls in Seoul.

We also clarified that the RH-increase/RH-decrease results in Fig. 3 are observational subsets based on an 8-hour rolling RH slope, not an ML perturbation experiment.

Manuscript edits:

1. **Supplementary material Line 89 Added:** "The nitrate XGBoost model showed stable 10-fold cross-validation performance ($R^2 = 0.954 \pm 0.008$, MAE = 0.566 $\pm$ 0.022 μg m^{-3}), supporting its use for within-campaign interpretation."
2. **Line 654 Added:** "These RH-increasing and RH-decreasing groups are observational subsets classified from the measured RH time series (Text S2)."

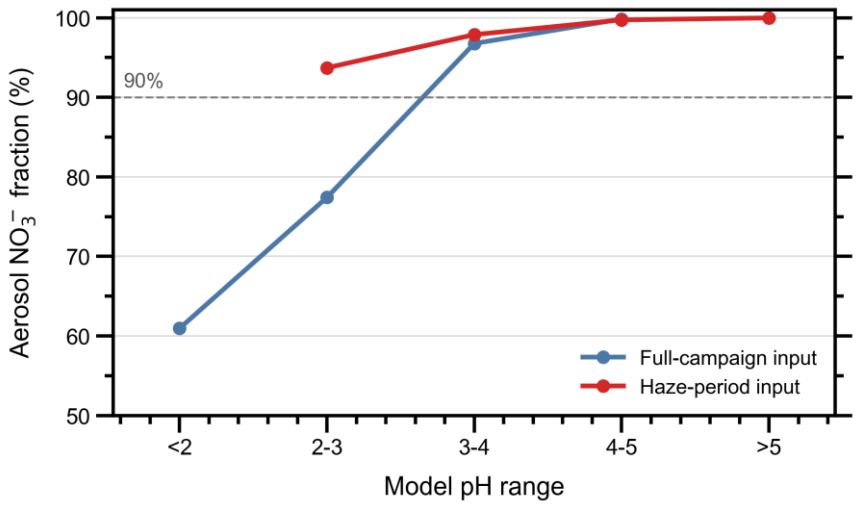
Comment 3

The author uses $TNO_3 = pNO_3 + HNO_3$ to describe total nitrate. This relationship is not accurate. This is because nitrate undergoes gas-particle partitioning, and this process is dynamic, potentially influenced by a variety of factors such as RH, temperature, and the chemical composition of particulate matter. Furthermore, these factors also affect the pH of the ALWC and particulate matter. The paper does not discuss changes in pH or how they affect TNO_3. In addition, nitrate formation will also enhance ALWC, while the authors did not consider this effect.

We agree. We revised the manuscript to keep $TNO_3 = HNO_3 + pNO_3^-$ only as a mass definition and to state that quantitative TNO_3 closure, aerosol pH, and HNO_3/pNO_3 partitioning require co-located HNO_3 and NH_3 measurements.

We also clarified that reverse-mode ALWC includes measured particle nitrate, so the ALWC-NO_3 relationship is interpreted as composition-water coupling rather than a one-way ALWC effect on nitrate.

As an additional check for the pH and partitioning issue, we include a bounded forward-mode ISORROPIA-II diagnostic using representative full-campaign and haze-period particle-composition inputs. Because forward mode requires gas-plus-particle precursor inputs, and HNO_3 and NH_3 were not measured, we prescribed a small reservoir ensemble: NH_3 at 1x, 1.5x, and 2x the representative NH_4 level; HNO_3-equivalent nitrate at the representative particle NO_3 level plus 0, 2, and 5 μg m^{-3}; RH at 35%, 50%, and 65%; and temperature from -10 to 5 °C. The HNO_3 range was included because KORUS-AQ work found South Korean aerosol formation to be sensitive to HNO_3, especially in polluted regions (Ibikunle et al., 2024). The diagnostic is included in the response to show the expected pH-dependent partitioning behavior, but is not added to the manuscript or used as an ambient reconstruction of HNO_3, NH_3, or pH.



Response Figure 1. Bounded ISORROPIA-II scenario diagnostic linking scenario-output pH to modeled nitrate partitioning. Points are forward-mode sensitivity cases with prescribed NH_3/HNO_3 reservoirs, not measured campaign pH or reconstructed ambient HNO_3/NH_3. The diagnostic is used only to show how pH-sensitive partitioning behaves within the stated assumption space.

The check shows the expected increase in modeled aerosol NO_3^- fraction with model pH. We therefore keep the manuscript interpretation bounded to measured particle nitrate and identify pH effects, HNO_3/pNO_3 repartitioning, and nitrate-ALWC feedback as future measurement needs.

Manuscript edits:

1. **Line 135: Added:** "Reverse-mode ISORROPIA-II calculations were therefore used as a relative ALWC diagnostic based on measured particle composition, ambient temperature, and RH."
2. **Line 267: Added:** "Because the reverse-mode ISORROPIA-II calculation uses measured particle-phase inorganic composition, including nitrate, the ALWC estimate includes water associated with particulate nitrate. We therefore interpret the ALWC-NO_3 relationship as composition-water coupling, not as an independent one-way effect of ALWC on nitrate."
3. **Line 276:** "This creates a thermodynamic positive feedback loop: the initial photochemical nitrate formation lowers the mutual deliquescence relative humidity (MDRH) of the aerosol mixture." **After:** "This pattern is consistent with composition-dependent water retention: increasing nitrate fraction can lower the effective RH required for particle water retention in a mixed aerosol."
4. **Line 279:** "Operating in tandem with potential phase hysteresis, this sustained aqueous volume allows continuous HNO_3 partitioning, preventing immediate crystallization and driving the 10 AM peak." **After:** "Together with possible RH-history hysteresis, this retained particle water can support continued HNO_3 partitioning during the observed morning nitrate increase."
5. **Line 289:** "The continued accumulation of nitrate mass across size bins during this period (Fig. S3b) demonstrates that even as this morphological shift occurs, the particles retain sufficient ALWC in a metastable state to facilitate continuous HNO_3 gas-to-particle partitioning." **After:** "The continued accumulation of nitrate mass across size bins during this period (Fig. S3b) is consistent with particle-water retention inferred from the ALWC diagnostics, but does not directly determine ambient particle phase state."

Comment 4

Line 169: How do the authors calculate NOR?

We added the definition at first use. NOR was calculated from molar concentrations.

Manuscript edits:

Line 177 Added: "The nitrogen oxidation ratio (NOR) was calculated as $NOR = [NO_3^-] / ([NO_3^-] + [NO_2])$ using molar concentrations."

Comment 5

Line 173-174: This is a general conclusion, which have been widely proved by other studies. Line 179-184: Conclusions here seems to be based on speculation. Throughout the text, there are too many such speculative conclusions; the author needs to provide more evidence.

We agree that the opening synthesis of Section 3.2 was too general. We revised it to present the joint role of precursor abundance and meteorology as established context, and to state the campaign-specific SHAP result more narrowly.

We also removed broad "production rate" and "outcome" language. The nonlinear RH and temperature responses are now framed as statistical associations that are examined with the process-level diagnostics in the following sections.

Similar mechanistic statements elsewhere in the manuscript were revised in their specific sections. RH-related, AMS-Dva, and temperature-related interpretations are now linked directly to the available measurements, with explicit boundaries where HNO_3, NH_3, N_2O_5, aerosol pH, viscosity, and wet-particle morphology observations are unavailable.

Manuscript edits:

1. **Line 181:** "These results demonstrate that wintertime nitrate formation is jointly controlled by precursor abundance and meteorological conditions. Our integrated analysis quantitatively resolved the relative contributions of these factors, revealing that NO_2 is the dominant driver, explaining 31.6% of the observed variability in nitrate concentrations. RH (15.9%), solar radiation (14.9%), and temperature (11.6%) were also significant contributors. While precursor availability sets the upper bound for potential formation, meteorological conditions strongly regulate the actual production rate, especially when precursor levels are already high." **After:** "These results indicate that, during the Seoul winter campaign, PM_1 nitrate variability was linked to measured precursor availability and meteorological conditions. In the XGBoost-SHAP analysis, NO_2 had the largest relative contribution to the model prediction (31.6% ± 0.6%), followed by RH (15.9% ± 0.9%), solar radiation (14.9% ± 0.7%), and temperature (11.6% ± 0.7%). Because meteorological predictors can covary, these percentages are treated as relative predictor rankings within the fitted model; chemical-source or pathway apportionment would require additional mechanistic constraints."
2. **Line 188:** "This nonlinearity likely reflects the complex interplay between meteorology and atmospheric physicochemical processes. Together, these results illustrate that wintertime nitrate production is the outcome of both direct precursor input and secondary modulation by varying meteorological conditions." **After:** "These nonlinear associations are examined below using RH-history, ALWC, and thermodynamic sensitivity diagnostics."

3. **Line 223:** "The positive correlation between relative humidity (RH) and particulate nitrate is well established, primarily due to the enhancement of heterogeneous and aqueous-phase pathways such as N_2O_5 hydrolysis under high-RH conditions" **After:** "The positive correlation between RH and particulate nitrate reflects coupled effects of aerosol liquid water, HNO_3 partitioning, and heterogeneous or aqueous pathways such as N_2O_5 hydrolysis under high-RH conditions"

Comment 6

The author points out here that changes in RH are sufficient to cause nitrate deliquescence. Is this narrow change in RH sufficient to cause nitrate deliquescence? Furthermore, the author assumes here that sulfates and nitrates are externally mixed, which is not actually the case. Internally mixed particles account for a significant proportion. For internally mixed particles, their deliquescence behavior differs from that of pure sulfates or nitrates, and they do not have a precise deliquescence point.

We agree. We replaced the pure-salt threshold framing with a mixed-particle RH-history interpretation, treating the literature ranges as reference bounds rather than Seoul-specific phase-transition thresholds (Sun et al., 2018).

We also added an RH-history diagnostic based on morning peak RH, post-peak RH minimum, ALWC-based liquid-duration during the drying interval, and nitrate growth. This diagnostic is used as supporting evidence for the RH-history interpretation, not as a direct measurement of ambient particle phase state. We also corrected the AMS interpretation to state that Dva represents dried residual-particle sizing because the HR-ToF-AMS sampled through an inlet dryer.

Manuscript edits:

1. **Line 250:** "Under strict equilibrium thermodynamics, the concurrent drop in daytime RH would induce immediate particle efflorescence (crystallization), expelling liquid water and shutting down the aqueous volume required for this freshly formed HNO_3 to partition into the aerosol phase. However, our ML and observational analysis strongly suggest that internally mixed urban aerosols persist in a metastable, semi-liquid state well below their theoretical mutual deliquescence RH due to phase hysteresis (Davis et al., 2015; Tang and Munkelwitz, 1993). In this metastable state, the retained ALWC serves as a highly concentrated, high-ionic-strength absorptive reservoir. This allows the newly generated, photochemically produced HNO_3(g) to continuously partition into the particle phase, bridging the gap between gas-phase production and aerosol-phase accumulation, and driving the 10 AM peak despite drying conditions." **After:** "Decreasing RH can reduce particle water, but internally mixed nitrate-sulfate-organic haze particles may remain aqueous or partially aqueous during drying, depending on RH history and particle composition. Reported urban haze phase-transition ranges include solid-aqueous equilibrium states for urban haze particles at 60-79% RH and 55-77% RH during hydration, with

aqueous phases retained during dehydration until about 55% RH and 50% RH, respectively (Sun et al., 2018). During 17 January-16 February 2018, measured RH ranged from 15.2% to 95.9%; haze mornings had a median morning peak RH of 64.3%, followed by a post-peak minimum RH of 43.6% before 11:00. We therefore interpret the morning nitrate enhancement as consistent with RH history, mixed-particle hygroscopicity, and gas-particle partitioning, while treating the literature phase-transition ranges as reference bounds rather than direct ambient phase-state measurements. Under this interpretation, retained ALWC can provide a concentrated absorptive reservoir that allows newly produced $HNO_3(g)$ to partition into the particle phase during drying, linking daytime gas-phase production with aerosol-phase nitrate accumulation."

2. **Line 282:** "Aerosol size distribution microphysics derived from the HR-ToF-AMS further substantiate this morphological transition (Fig. 3). During RH-decreasing periods, the peak vacuum aerodynamic diameter (Dva) of nitrate-containing particles shifted from ~350 nm to ~300 nm. Because the AMS measures Dva, which is highly sensitive to particle morphology and density, this reduction should not be interpreted merely as physical compaction of the dry mass. Rather, it provides strong observational evidence of partial efflorescence. As spherical liquid droplets begin to dehydrate and form semi-solid or crystalline inclusions, their dynamic shape factor (χ) increases (i.e., they become more irregular). An increased shape factor results in higher aerodynamic drag within the AMS aerodynamic lens, causing the particle to be measured at a smaller Dva even if the absolute dry mass is actively increasing (Decarlo et al., 2004; Canagaratna et al., 2007)." **After:** "The HR-ToF-AMS sampled through an inlet dryer; particle time-of-flight size distributions therefore represent dried non-refractory PM_1 residual-particle modes rather than wet ambient droplet morphology (Canagaratna et al., 2007). During the RH-decline period, nitrate-mode Dva shifted from approximately 350 to 300 nm. Dva depends on particle mass, density, and dynamic shape factor (Canagaratna et al., 2007; DeCarlo et al., 2004), so this shift should not be interpreted as direct evidence of wet ambient particle shape or phase state. Instead, it indicates a change in the dried nitrate-containing residual-particle mode, with possible contributions from material-density changes, dry-residue morphology, and/or smaller fresh particles."
3. **Line 296:** "In contrast to nitrate, sulfate did not show a significant response to dynamic RH changes (Fig. S17b). This is likely due to the higher deliquescence point of sulfate-containing particles (~80%) compared to ammonium nitrate (Sun et al., 2018). The average RH during the campaign was 47.7 ± 16.3%, and 48.9 ± 14.5% during 7–10 AM—sufficient for nitrate deliquescence but typically below the threshold for sulfate. Consequently, RH-driven phase transitions played a lesser role in modulating sulfate chemistry, as evidenced by the lack of differential RH contributions between increasing and decreasing RH periods (Fig. S17c)." **After:** "Unlike nitrate, sulfate did not exhibit a distinct dependence on RH tendency in the SHAP analysis (Fig. S17c). This contrast is expected because sulfate is nonvolatile in the particle phase, whereas nitrate can vary through coupled aerosol-water changes and HNO_3-pNO_3 partitioning during morning drying. We therefore interpret the nitrate response using RH

history and mixed-particle water retention, rather than campaign-average RH or separate pure-salt deliquescence thresholds."

4. **Added Figure S18:**

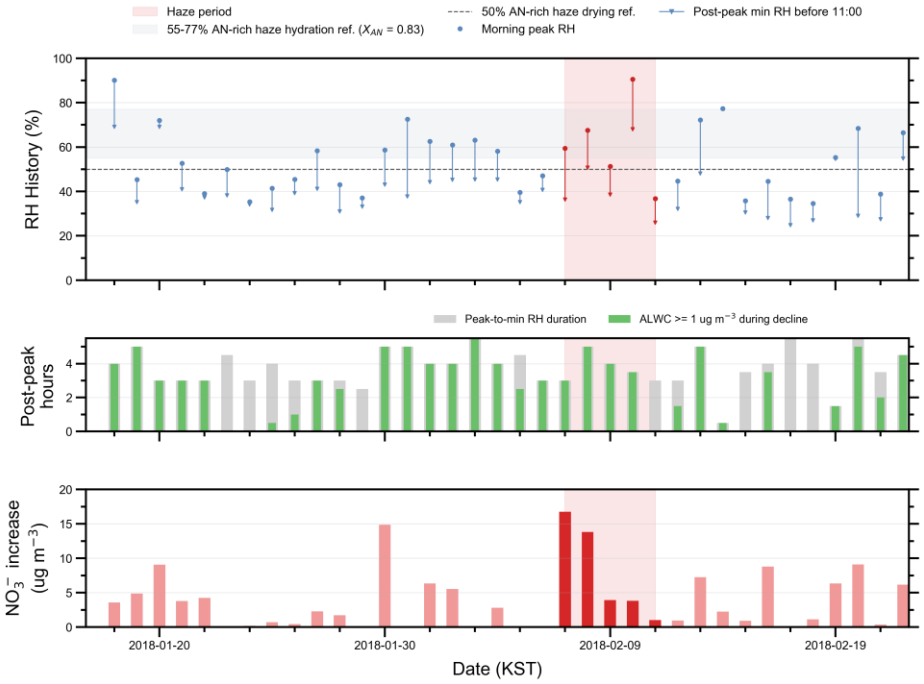


Figure S18. RH-history diagnostic for haze and non-haze mornings. The RH panel compares each day's morning peak RH with the post-peak minimum RH before 11:00; the minimum was required to occur after the peak. The shaded blue band and dashed line show Sun et al. (2018) AN-rich haze reference ranges for hydration and drying retention, used as literature bounds rather than Seoul-specific thresholds. The middle panel shows the peak-to-minimum RH duration and the portion of that drying interval with ISORROPIA-II ALWC ≥ 1 μgm^{-3}. The bottom panel shows the morning NO_3^- increase, calculated as the 06:00-11:00 peak minus the 03:00-06:00 mean background.

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