Response to the referee #1

Understanding the chemistry of reactive nitrogen in pristine environments, such as the Tibetan Plateau, is crucial for advancing our knowledge of atmospheric chemistry. This study undertook a challenging field campaign, collecting $PM_{2.5}$, total particle, and soil samples to investigate the unusual enrichment of nitrite in coarse particles and its potential sources. While the effort in sample collection and extensive analysis is commendable, the presentation of the data in this paper lacks clarity, and the interpretation of the results is, in my opinion, not entirely sound.

After reviewing this manuscript, I remain unconvinced by the authors' arguments, as significant gaps in explanation persist. Given the high publication standards of Atmospheric Chemistry and Physics (ACP), I believe the current manuscript does not meet the criteria for publication without substantial revisions.

Response: We thank the reviewer for your time and thoughtful evaluation of our manuscript. We appreciate the acknowledgment of the efforts involved in our field campaign and sample analysis. We also recognized and took seriously the concerns raised regarding the clarity of data presentation and the interpretation of our results. But before going with details of response, we wanted to first clarify that probably due to the organization and writing in the original manuscript, our intentions may have not been clearly conveyed. In this manuscript, the objective was not to draw a definitive conclusion on the source/origin of particulate nitrite, instead we wanted to present this finding and discuss what are the potential sources, as reflected by the title "**On the presence of high nitrite (NO_2^-) in coarse particles at Mt. Qomolangma**". We thought the finding, i.e., the presence of nitrite exclusively in the coarse particle is interesting and puzzling, and tried to explore the possible sources/origins and implications of this finding. We realized that it is difficult to discern the exact origin/source of the observed coarse particle nitrite given current dataset available, we hypothesized that long-range transport of pollutants from South Asia and wind-blown soil together may explain the observations, though additional investigations are necessary to discern the exact mechanisms how these sources contribute to nitrite in TSP, or if there are other sources. We are making these points clearer in the revised manuscript. Please review our point-by-point responses to your comments with more details in the following point-to-point responses.

Here are my major concerns:

1. Uncertainties in ion concentrations of $PM_{2.5}$ and total particles. The comparison of ion concentrations between TSP and $PM_{2.5}$ is particularly interesting. It clearly shows that nitrite ions are present only in TSP, while $PM_{2.5}$ contains none. However, what stands out is that despite $PM_{2.5}$ being a subset of TSP, some $PM_{2.5}$ samples occasionally show higher ion concentrations (in $\mu g/m^3$) than their corresponding TSP samples. For instance, the April 30 $PM_{2.5}$ sample appears to contain more nitrate than the TSP sample.

I suspect this discrepancy may stem from analytical uncertainties in ion concentration measurements or uncertainties in the blank corrections, but this issue has not been addressed in the manuscript. Understanding these uncertainties is crucial, especially since the ion composition of coarse aerosols is determined by subtracting two similar measurements.

I also suggest that the authors directly present the ion concentrations of coarse aerosols. While this may sometimes result in negative values, it would provide a clearer picture of the uncertainties associated with ion concentration measurements.

Response: Thanks for the valuable and insightful suggestions. Indeed, there might be some degree of uncertainties given the measurements themselves and conversion of liquid concentrations to air mass concentrations. The latter are subject to further uncertainties given the parameters of the sampling instruments (e.g., sampling flow rate). In the revised manuscript, we have added the uncertainties regarding the ionic concentrations in $PM_{2.5}$ and TSP, as outlined in Section 2.3 (lines 158-188):

2.3 Ionic concentration analysis and uncertainty estimation

Water-soluble inorganic ions (WSIs, including Na^+, NH_4^+, K^+, Mg^{2+}, Ca^{2+}, Cl^-, NO_2^-, NO_3^-, and SO_4^{2-}) in TSP (entire filter, ~16.6 cm^2) and $PM_{2.5}$ (1/32 section, ~13.0 cm^2) were extracted using 20 mL of Milli-Q ultrapure water (18.2Ω cm) in an ultrasonic bath at room temperature for 30 min. Note the TSP filters were cut to fit the filter holder from the standard Whatman quartz-fiber filters which were also used as the $PM_{2.5}$ filters. After filtration through a 0.22 μm pore size syringe filter which was pre-cleaned with ultrapure water, the filtrate was subjected to inorganic species analysis using ion chromatography (Dionex Aquion) (Zhang et al., 2020). Blank filters were pretreated and measured the same as real samples, and the limits of detection (LOD) was calculated as 3 times of standard deviations of blanks (Fang et al., 2015). In general, Na^+ in the blank filters is comparable to samples, a well-known issue for the Whatman quartz filter which is high in Na^+ blank. Therefore, in this study, we discarded the Na^+ data. The volatile components (i.e., NH_4^+, NO_2^-, NO_3^-) and K^+ in blank filters are low but several times higher than the detection limits; SO_4^{2-}, Mg^{2+} and Ca^{2+} in blank are significantly higher than the respective LOD but lower than samples by at least five times (the lower end). All reported concentrations of each ion were blank corrected as follows:

$$C_i = \frac{(\rho_{sample} - \rho_{blank}) \times V_{water} \times F}{V_{air}}$$

with C_i representing the ambient concentrations of specie i in air (ng m^{-3} or μg m^{-3}), ρ_{sample} and ρ_{blank} are the concentrations determined by the ion chromatography (ng mL^{-1}), V_{water} is the volume of ultrapure water used for extraction (20mL), V_{air} is the volume of air sampled for each $PM_{2.5}$ or TSP filter, F is the ratio of particulate matter collection area for $PM_{2.5}$ or TSP filters to the filter area used for extraction.

The overall uncertainty in ion concentration was estimated according to the law of error propagation, accounting for the sampling air volume (3% for $PM_{2.5}$ samples and 1% for TSP samples as provided by the manufactures), the extraction of water volume (~0.3% for pipetting from the manufacture Eppendorf), the blanks, and the analytical uncertainty from ion chromatography and calibration, assuming that these factors are independent. The analytical uncertainty for water-soluble ions concentration determination using ionic chromatography has been extensively assessed in our laboratory, with values typically <5% for all inorganic species at concentration of 500 ng mL^{-1}. The combined uncertainty about the ionic concentrations in $PM_{2.5}$ and TSP are shown in Table S1. In general, TSP samples are associated with relatively high overall uncertainty compared to $PM_{2.5}$ samples, perhaps due to the relatively high blank variability due to the low mass loading in TSP.

In the revised manuscript, as suggested, we have also presented the ion compositions in the coarse-mode particulate in new Figure 2. We acknowledge that in some cases, the ions concentrations in $PM_{2.5}$ is higher than that in TSP filters, as indicated by the discontinuous time series of certain ions,

i.e., SO_4^{2-}. Nevertheless, this abnormal phenomenon has no significant impact on the key findings and interpretations in the manuscript. For example, the predominance of nitrite in coarse particulate matter remains unambiguous and significant declines of secondary inorganic ions after April 30th in response to the shift of air mass origins remains well-supported.

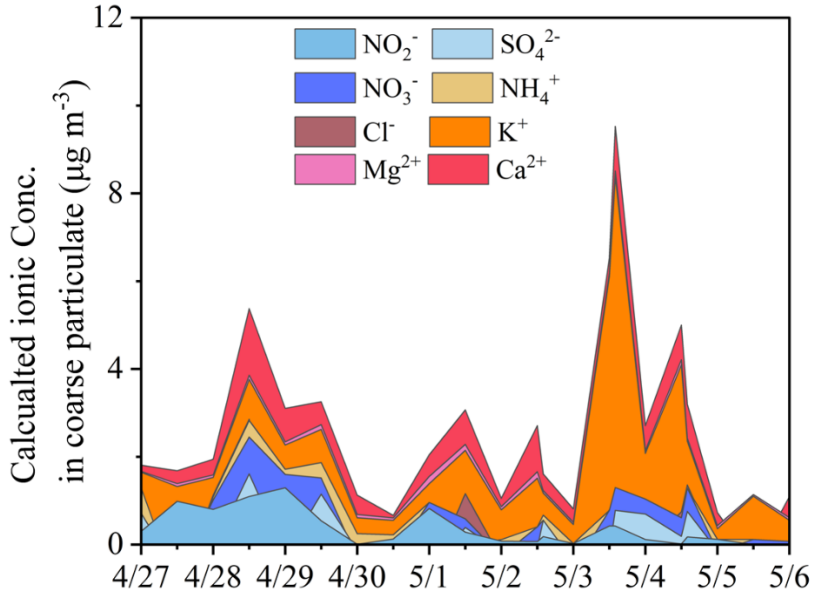

Figure 2. The estimated chemical compositions and time series of mass concentrations of chemical species (NO_2^-, SO_4^{2-}, NO_3^-, Ca^{2+}, etc.) in coarse-mode particulate during the springtime campaign (calculated as the differences between TSP and $PM_{2.5}$). Discontinuities in the time series were observed for certain species (e.g., SO_4^{2-}), likely resulting from relatively lower concentrations in the TSP samples compared to those in the corresponding $PM_{2.5}$ samples. This discrepancy is likely attributed to the propagated uncertainties involved in the concentration analysis, sampling approach and blank corrections.

2. Missing evidence: mass balance in coarse particles. I would like to see a little bit more discussion in section 4.3 when the authors attempted to attribute the observed nitrite to lofted dust, maybe as simple as mass balance calculations. For example, if the observed nitrite is indeed coming from soil, to get ~1 ug/m^3 of nitrite from soil, how much soil do you need in the air giving average soil nitrite concentration of < 100 ng/g? About 10 g/ m^3. Then, does the observed TSP concentration support your hypothesis?

Similarly, nitrate/nitrite ratio in soil also do not fully support authors' hypothesis, the authors argue that it is likely nitrate/nitrite distributed in particles of different sizes but there is no evidence supporting this, nor is there any previous work mentioned such effect. Therefore, I am not fully convinced by the existing evidence that soil is the main source of particle nitrite.

Response: Thank you for your valuable suggestion. We have included a simple mass balance-based estimation in Section 4.3 (The potential effects of lofted dust) to further assess the potential contribution of resuspended surface soil to the observed nitrite (NO_2^-) concentrations in TSP. The result indicates that, under justified reasonable assumptions, soil nitrite can fully explain the TSP nitrite in the second period of the observations, i.e., after April 30th. For elevated levels of TSP

nitrite before the April 30th, biomass burning would be necessary to be included. This in fact reconciles our conclusion that biomass burning and soil together may explain the observations.

Below we listed our detailed assessments on the potential contribution of soil to TSP nitrite and our revisions inspired by the suggestion. The assessments are as follows (lines 541-570): "We also noted that NO_2^- in surface soil is significantly lower than NO_3^-, by on average 35 times (Table 1), contrasting with the chemical compositions in TSP. It is important to note that during the complex dust generation and aerosolization processes, water-soluble ions exhibit significant chemical enrichment relative to that in parent soil (Wu et al., 2022; Gao et al., 2023). For example, Wu et al., 2022 reported that contents of nitrate in sand dust-derived PM_{10} is higher than the original soil samples by 2~80 times, while sulphate can be up to 500 times higher than the original soil. Although the enrichment of nitrite has not been evaluated to the extent of our knowledge, we propose that the observed discrepancy of nitrate/nitrite ratio between TSP and surface soil can be reconciled if the nitrite is enriched more efficiently than nitrate during dust aerosolization, i.e., by a factor of 30.

To further assess the potential contribution of resuspended soil to elevated nitrite in TSP, we conducted a rough estimation based on laboratory investigations from Wu et al. (2022). First, the springtime TSP concentrations observed in the nearby QOMS station is used (65.1 ± 50.9 μg m^{-3}; Liu et al., 2017), as TSP mass concentrations surrounding the Base camp were not available. Second, we assumed the nitrite content in soil-derived coarse particles to be similar to that of nitrate (0.2%) in laboratory-generated dust aerosol from natural sandy and gravel soils (Wu et al., 2022), which is comparable to the soil texture of the Rongbuk Valley. This assumption can also explain the comparable amounts of nitrite and nitrate observed in TSP samples in this study. Results shown that the concentrations of nitrite in TSP can be approximately 130 ± 102 ng m^{-3}, on the same order with the observed nitrite concentration in TSP after April 30th (147 ± 145 ng m^{-3}) but substantially lower than that before April 30th (625 ± 457 ng m^{-3}). Therefore, we speculate that the resuspension of surface soil may account for the observed TSP nitrite after April 30th, whereas the biomass burning and soil together co-contributed to the high TSP nitrite before April 30th. This is also consistent with the shift of air mass origins, which clearly indicated that airmasses before April 30th is significantly impacted by the biomass burning emissions, and after April 30th airmasses primarily originated from clean regions. While we acknowledge that this simplistic estimation is subject to substantial uncertainty, it provides a first-order assessment supporting the hypothesis that wind-blown soil dust contributes to coarse-mode particulate nitrite. Previously, wind-blown mineral dust has been verified as a potential source for aerosol water-soluble ions (Engelbrecht et al., 2016; Wu et al., 2022; Wu et al., 2012) and dust aerosol is also recognized as one of the important aerosol types over TP (Pokharel et al., 2019)."

The above assessments were based on several considerations. First, recent studies have indicated that due to the aerosol-soil fractionation during dust emission, the contents of water-soluble ions in soil-derived dust aerosol are significantly higher than the parent soil (Wu et al., 2022; Gao et al., 2023). Wu et al. (2022) conducted a detailed investigation of the chemical composition of water-soluble ions in soil-derived aerosols using a laboratory dust-generation system, which ensured consistency in particle size distribution and chemical composition between the generated particles and ambient dust. Their results serve as a reliable reference for estimating the potential contribution of specific components in soil-derived aerosols. Their results shown that the enrichment varied

significantly among the water-soluble ions. For example, contents of nitrate in sand dust-derived PM_{10} is higher than the original soil samples by 2~80 times (on average 22 times), while sulphate can be up to 500 times higher than the original soil. Unfortunately, the enrichment of nitrite was not evaluated.

Second, in the present study, the ratio of nitrate to nitrite in surface soil is on average 30 (Table 1 in the main text), whereas the nitrate is comparable to the nitrite in the TSP samples. To reconcile this discrepancy, we propose that nitrite may experience more efficient enrichment than nitrate during aerosolization, i.e., nitrite was enriched ~30 times more than nitrate. Moreover, Wu et al. suggested that the nitrate contents in the sand dust-derived PM_{10} is on average 0.2%. Since nitrate was determined to be comparable to the nitrite in the TSP samples, the content of nitrite in the soil-derived coarse particle is also assumed to be 0.2% in the our estimation. We acknowledge although the simplistic estimation is associated with significant uncertainty, it provides a way to assess the potential contribution of surface soil-derived nitrite to the observed high nitrite in TSP.

Third, the springtime TSP mass concentrations in the nearby QOMS stations (65.1 ± 50.9 μg m^{-3}) were used, as there were no available reports regarding the TSP mass concentrations at the Base Camp. We estimated that the concentrations of nitrite in TSP can be approximately 130 ± 102 ng m^{-3}, on the same order with the observed nitrite concentration in TSP after April 30th but substantially lower than that before April 30th. Therefore, we suggest that resuspension of surface soil is a potential contributor to the TSP nitrite after April 30th. However, for the higher nitrite levels observed before April 30th, additional sources, such as biomass burning are likely required. This interpretation is supported by the corresponding shift in air mass origins before and after April 30th, with the earlier period showing significant influence from biomass burning emissions.

3. Isotopic results do not seem to support authors' argument. The isotopic results also do not support the authors' argument. The O^{17} signal in soil samples range from 1.4‰ to 7.3‰ but in TSP the O^{17} is 0-1‰, a clear discrepancy. $\delta^{18}O$ also are significant different – 2‰ to 18‰ in the soil but lots of negative values in the TSP. I do not think this can be simply explained by isotope exchange with water because such isotope exchange always occurs: we need more evidence to believe that exchange never happens when the particles are on the surface, then once it was lifted into the air, within hours (typical lifetime of coarse particles) the exchange suddenly occurred.

Response: Thanks for raising this important question. We agree that oxygen isotopic exchange between nitrite and water molecule occurs not only in the atmosphere but also in surface soils. The significant $\Delta^{17}O$ difference observed between nitrite and nitrate in surface soils (3.7‰ vs. 9.6‰) suggests that isotope exchange is indeed occurring in the soil environment as well. Moreover, the hygroscopic nature of dust aerosol enables the absorption of water and thus facilitate the exchange process between TSP nitrite and aerosol water. According to your suggestion, the discussion is revised as follows (lines 495-540):

In addition, the similarity in $\delta^{15}N$ of NO_2^- between TSP (-7.3 ± 3.1‰) and the surface soil (-10.3 ± 3.0‰) also likely supports that locally emitted surface soil may contribute to the observed high levels of TSP NO_2^-. But one should note that the oxygen isotopes ($\delta^{18}O$ and $\Delta^{17}O$) of TSP NO_2^- were significantly lower compared to that in soil NO_2^-, indicating that the original soil NO_2^- oxygen isotope may have been modified after resuspension. This discrepancy could be explained by the potential oxygen isotope exchanges between TSP NO_2^- and aerosol liquid water (fractionation effect

of $^{18}\varepsilon_{eq} \approx 16‰$ at local temperature, T = 270K, $^{18}\varepsilon_{eq} = -0.12\ T + 48.79$; Buchwald and Casciotti, 2013), which tend to deplete both the $\delta^{18}O(NO_2^-)$ and $\Delta^{17}O(NO_2^-)$.

We noted the oxygen isotope exchange process between NO_2^- and water also occurs in surface soil. Previous study indicated that in high-altitude arid regions of TP (i.e., >5000m), denitrification process dominated the surface soil NO_2^- production, accounting for ~75% (Wang et al., 2019). Soil NO_2^- generated from denitrification process is expect to inherit the $\Delta^{17}O$ signatures of substrate NO_3^-. In this study, the surface soil $\Delta^{17}O(NO_3^-)$ were positive with average values of 9.6‰ (Table 1). The positive soil $\Delta^{17}O(NO_3^-)$ have been observed on arid environments (Wang et al., 2016; Herath et al., 2023), such as desert soil, where the low water moisture favors the preservation of atmospherically derived NO_3^-. One could estimate that soil $\Delta^{17}O(NO_2^-)$ derived from the denitrification would be 9.6‰, nitrite from other sources (e.g., nitrification) should possess zero $\Delta^{17}O$, thus in total nitrite in soil should possess $\Delta^{17}O$ of ~7.2‰ (estimated as $0.75\times\Delta^{17}O(NO_3^-)$). However, the determined soil $\Delta^{17}O(NO_2^-)$ (3.8‰) is significantly lower compared to the estimated soil $\Delta^{17}O(NO_2^-)$, indicating the occurrence of exchange process between soil water and NO_2^-, which would reduce the soil $\Delta^{17}O(NO_2^-)$ to some extent. The exchange process between soil water and NO_2^- is particularly evident in west slope of Rongbuk Valley, where soil $\Delta^{17}O(NO_2^-)$ is as low as 1.5‰ while soil $\Delta^{17}O(NO_3^-)$ is on average 10.3‰. Soil $\Delta^{17}O(NO_2^-)$ should be erased to near-zero if exchange process between soil water and NO_2^- was efficient. Therefore, the fact that the observed soil $\Delta^{17}O(NO_2^-)$ remain above 0‰ indicates unfavorable conditions for the oxygen isotope exchange process, likely due to the extremely low soil moisture content (~1%) in surface soil (Ma et al., 2023).

Upon resuspension into atmosphere, the soil-derived dust aerosols usually exhibited a certain degree of hygroscopicity (Tang et al., 2016; Chen et al., 2020), allowing the absorption of water molecule onto dust aerosol. For example, the aerosol water was determined to account for ~20% of the total PM_{10} mass during Saharan dust plumes (Cardoso et al., 2018). Based on laboratory experiment, Tang et al., 2019 reported that Asian dust also exhibited substantial hygroscopic property and revealed that the water-soluble inorganic ions, such as Cl^-, SO_4^{2-} and NO_3^- played a critical role in the absorption of water molecules on dust aerosol (Tang et al., 2019). In the present study, the SO_4^{2-} and NO_3^- account for ~30% of the total mass of water-soluble ions in TSP, implying the potential uptake of water vapor on aerosol surface. In addition to water-soluble ions, the hygroscopicity of mineral dust also depend on the surface areas, and wind-blown dust experiences a substantial increase in surface area after being lifted into the atmosphere, enhancing its capacity for water uptake (Chen et al., 2020; Seisel et al., 2004). The hygroscopicity of dust aerosol is expected to accelerate the oxygen isotope exchanges between NO_2^- and aerosol liquid water. The atmospheric water vapor $\delta^{18}O$ isotope in TP is determined to be significantly negative (approximately -35‰ to -15‰ at a remote site in TP with altitude of ~ 4200m) (Yu et al., 2015). Similarly, the oxygen isotope exchange between NO_2^- and H_2O would also homogenize and erase original soil $\Delta^{17}O(NO_2^-)$ signals, because aerosol liquid water is characterized by negligible $\Delta^{17}O$ values (Luz and Barkan, 2005). Consequently, isotope exchange with aerosol water would further reduce both $\delta^{18}O$ and $\Delta^{17}O$ of TSP NO_2^-, effectively masking the original isotopic signature inherited from surface soil.

In brief, an extensive field investigation revealed active denitrification process in dryland surface soil at elevations above 5000m from Tibetan Plateau (similar to the soil texture around our sampling site), accounting for 75% of surface soil nitrite production (Wang et al., 2019). Surface soil nitrite

produced from denitrification process inherit $\Delta^{17}O$ of nitrate, since this process should follow the mass-dependent fractionation law. Based on the field investigations in Wang et al., 2019, we estimate the surface soil NO_2^- should be associated with relatively higher $\Delta^{17}O$ values (~7.2‰, 9.6‰×0.75, assuming nitrite from other sources, e.g., nitrification, possess zero $\Delta^{17}O$) than our determined values (3.7‰). Therefore, the occurrence of oxygen atom exchange between nitrite and water molecule in surface soil may also occur, resulting in relatively low $\Delta^{17}O$ values in surface nitrite. The exchange process maybe particular evident in west slope soil, given the significant lower surface soil $\Delta^{17}O(NO_2^-)$ relative to $\Delta^{17}O(NO_3^-)$ (1.5‰ versus 10.3‰). Note that if the oxygen exchange process was sufficiently efficient, the surface soil $\Delta^{17}O(NO_2^-)$ should be erased to be negligible. Therefore, the determined positive surface soil $\Delta^{17}O(NO_2^-)$ probably indicated unfavorable conditions for the oxygen isotope exchange process may due to the extremely low soil moisture content (~1%) in surface soil (Ma et al., 2023).

Second, the mineral dust aerosols have been shown to exhibit hygroscopic properties, which would facilitate the oxygen exchange process between nitrite and water molecule, once emitted into atmosphere (Kumar et al., 2009; Shi et al., 2008; Tang et al., 2019). It is suggested that the water-soluble inorganic ions, especially sulfate, nitrate and ammonium majorly determine the hygroscopic properties of mineral dust aerosol (Tang et al., 2016; Shi et al., 2008; Tang et al., 2019), and these processes can result in a water content of up to 20% of the total PM_{10} mass as observed in Saharan dust plumes (Cardoso et al., 2018). In the present study, the sulfate and nitrate account for ~30% of the total mass of water-soluble ions, implying the potential uptake of water vapor on aerosol surface. We suggested that the hygroscopic properties may facilitate the oxygen exchange between TSP NO_2^- and water molecule, upon resuspension into atmosphere.

4. More discussion needed for air mass from different regions. The back-trajectory analysis clearly shows that the field campaign sampled air from two distinct regions, with corresponding differences in ion concentrations and $d^{15}N$ values. However, the discussion of these differences is too simplistic. It would be beneficial for the authors to separately analyze how the aerosols from each period differ and to explore in greater depth how nitrite concentrations and isotopic compositions varied between them, as the differences are quite significant.

Additionally, while the authors suggest that long-range transport is unimportant for the nitrite budget, their argument in the final section contradicts this claim. To strengthen the manuscript, the discussion should remain consistent and logically cohesive.

Response: Thank you for the valuable suggestion. We have expanded Section 4.2 (Potential effect of biomass burning emissions in South Asia via long-range transport) to separately analyze and compare the aerosol characteristics, nitrite concentrations, and isotopes before and after April 30th. We also clarified our interpretation of long-range transport of biomass burning emissions, emphasizing that while it likely plays a significant role in the observed high TSP NO_2^- before April 30th, its contribution to TSP NO_2^- is minor after April 30th. The potential impact of anthropogenic and biomass burning pollutant via long-range transport was explored in-depth and the section 4.2 was modified to keep consistency according to your suggestions. In sum, we were not trying to state that biomass burning is not important, instead we acknowledge its contribution to TSP nitrite in the first half of the sampling campaign. We have made this clearer in the revision (lines 433-476).

In this study, the mass concentrations and compositions of water-soluble ions, including SO_4^{2-}, NO_3^-, NH_4^+ and TSP NO_2^-, also varied substantially throughout the springtime campaign (Fig.1). The

potential effect of South Asian pollutants on our observations was explored by analyzing the air masses origins during the sampling period. As shown in Figure 5, during the first-half of the campaign (i.e., before April 30th), air masses predominately originated from or passed through northern India and Nepal with intensive human activities and extensive biomass burning (represented by the dense red dots in Figure 5), indicating the potential impact of South Asia pollutants on aerosol loadings of TP. Correspondingly, elevated concentrations of secondary inorganic ions (i.e., NH_4^+, NO_3^- and SO_4^{2-}) in TSP and $PM_{2.5}$ were observed before April 30th, which are comparable to the values of previous reports in QOMS station when arrived air masses experiencing severe biomass burning emissions (Bhattarai et al., 2023; Lin et al., 2021). Meanwhile, elevated levels of TSP NO_2^- (625±457 ng m^{-3}) was observed during this biomass burning-impacted period, and $\delta^{15}N(NO_2^-)$ values exhibited substantial variability. TSP samples collected during daytime of April 27th and night of April 28th are associated with high NO_2^- concentrations and $\delta^{15}N(NO_2^-)$ values. In comparison, TSP NO_2^- in other samples were significantly ^{15}N-depleted. Assuming that biomass burning emission accounted for the two high $\delta^{15}N(NO_2^-)$ samples (Bhattarai et al., 2023), the observed relatively low $\delta^{15}N(NO_2^-)$ in other samples before April 30th likely indicated the potential contribution from additional emission sources.

From May 1st to May 6th, air masses originated from the inside of the TP or surroundings and none of the fire hotspots was detected at the whole TP and along the air mass trajectories (Figure 5), potentially excluding the influence of biomass burning and anthropogenic emissions during this period. Accompanied by this significant shift in air mass origins, the concentrations of NH_4^+, NO_3^- and SO_4^{2-} in both $PM_{2.5}$ and TSP, as well as the TSP NO_2^- apparently decreased. In particular, NH_4^+ in most $PM_{2.5}$ and TSP samples were below the detection limit during this period. Furthermore, K^+ in $PM_{2.5}$, a common tracer for biomass burning, declined from 269 ± 432 ng m^{-3} before April 30th to 22 ± 12 ng m^{-3} after April 30th, with the difference being statistically significant ($p < 0.05$). Meanwhile, the average TSP NO_2^- also declined significantly from 625 ± 457 ng m^{-3} before April 30th to 147 ± 145 ng m^{-3} after April 30th. Note substantially high levels of TSP NO_2^- were also observed on several days after April 30th, i.e., during daytime of May 3rd (~ 400 ng m^{-3}). Although the TSP NO_2^- concentrations varied in wide range after April 30th, the $\delta^{15}N(NO_2^-)$ was relatively stable and comparable to that determined before April 30th (except for the two high $\delta^{15}N(NO_2^-)$ samples).

In short summary, these results likely suggest the significant impact of South Asia pollutants (i.e., biomass burning emissions) through long-range transport on the TSP NO_2^-, especially for samples collected before April 30th. However, it is difficult for the long-range transport to explain why NO_2^- is predominately present in coarse particles, as fine mode particle is typically easier to be transported in principle. While further studies need to be conducted to find out the exact reasons, one possibility is that the size partition of fine particle NO_2^- toward the coarse mode range during transport. Similar size shifts of NO_3^- have been observed and used to explain the enrichment of NO_3^- in coarse-mode aerosols in marine environments (Matsumoto et al., 2009). In addition, the coarse mode particles in this study contain more alkaline species (Figure 2), which makes nitrite more stable in TSP during the transport or more stably exist in TSP after being uptake when the polluted air masses reaching TP. For the period after April 30th, since air masses originated from clean regions with little to no biomass burning sources, other sources of nitrite might be required.

5. Implications to AOC is weak. Since the source of nitrite remains unclear, the discussion on how lofted soil influences atmospheric chemistry is weak. The authors provide no data on atmospheric dust concentrations and do not address the transport potential of soil particles, which is likely limited due to their larger size. As a result, the final section lacks convincing supporting evidence.

Response: Thank you for the valuable suggestion. We have revised this section to focus more on the potential atmospheric implications of elevated coarse-mode nitrite observed at Mt. Qomolangma, and the influence of lofted soil on atmospheric chemistry was reshaped accordingly. We now highlight its potential contribution to local photochemistry through HONO and NOx formation, supported by estimated HONO levels derived from observed [pN(III)]/[HONO] ratios. We also clarified that while these effects may be spatially limited due to the rapid deposition of coarse particles, they could still significantly impact local atmospheric oxidation capacity (AOC) in this pristine high-altitude environment, especially considering the ubiquity of the potential sources, i.e., the frequent biomass burning episodes through long range transport from South Asia (Bhattarai et al., 2023) and frequent dust activities in TP (Long et al., 2025).

Unexpectedly high levels of NO_2^- associated with coarse-particle were observed in the pristine environment at Mt. Qomolangma in the spring, 2022. After examining the potential contributions of various NO_2^- sources with assistance from air mass back-trajectory and isotope analyses, we suggest that both soil-derived nitrite and long-range transport of pollutants from South Asia may contribute to coarse-particle NO_2^- during spring at Mt. Qomolangma. This is also consistent with previous reports showing that dust and biomass burning emission through long-range transport from South Asia are the predominant contributors to the springtime aerosol loadings over TP (Zhao et al., 2020; Pokharel et al., 2019). The nitrite concentrations and isotopes further indicated that soil-derived nitrite likely serves as a baseline source of atmospheric NO_2^-, maintaining the background levels of TSP NO_2^- at this pristine site, reflected by the relatively stable isotopes when soil-derived nitrite predominated. In addition, air masses originating from South Asia would result in elevated levels of NO_2^- observed before April 30th by bringing additional biomass burning and anthropogenic pollutants, as evidenced by the more varied isotopes before April 30th compared to after that day when air mass origins shifted from South Asia to central and north Tibet. However, the detailed mechanisms of nitrite enriched on the coarse particle remain unknown and need further explorations.

In the atmosphere, photolysis of particle nitrite can produce OH radical and NO, the latter is essential for the formation of atmospheric oxidants and secondary aerosols (Figure 6). Moreover, the elevated levels of particle NO_2^- may serve as an important HONO source through the gas-to-particle partition process (Vandenboer et al., 2014a), and the thermodynamic equilibrium between particulate nitrite and HONO ([pN(III)]/[HONO] ratio) is primarily governed by the particle acidity and liquid water content (LWC) in theory (Fountoukis and Nenes, 2007; Vandenboer et al., 2014a; Chen et al., 2019). Based on the observed TSP NO_2^- and estimated ratio of [pN(III)]/[HONO] (from 4.8 to 10.6, Text S2), we can estimate the potential level of atmospheric HONO if the partition ever occurs at this site (Vandenboer et al., 2014b), and result indicates HOHO would be at 8 ~ 15 pptv, on the same order with the observations in the background atmosphere at a central Tibetan site (i.e., ~ 30 pptv at Namco (Wang et al., 2023)). Given that TSP concentrations usually reach maximum during spring over TP, i.e., 65 ± 51 μg m^{-3} at the nearby QOMS station (Liu et al., 2017), our findings suggest that the coarse-particle may serve as a potential source of atmospheric HONO and NO_x assuming the TSP are associated with nitrite. Although the coarse-particle tend to deposit rapidly within hours,

their potential to influence local atmospheric chemistry remains important to some extent, particularly considering the frequent dust events in TP (loose arid/semiarid surface, sparse vegetation, and strong winds. Long et al., 2025) and the ubiquity of long-range transport of biomass burning emissions from South Asia during this season. The impact of the TSP nitrite on the budget of NOx, HONO and OH radicals especially in the background atmosphere could be investigated using regional or global atmospheric transport model, once the detailed mechanism regarding the sources and chemistry of TSP nitrite been elucidated. In summary, our results highlight the need for further investigation into the sources, partitioning, and chemical reactivity of aerosol-phase nitrite, particularly in the pristine Tibetan Plateau, where even small inputs of NO_x or HONO can disproportionately affect oxidant budgets and reactive nitrogen cycling.

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