Response to the referee #3

This manuscript, entitled "On the presence of high nitrite (NO_2^-) in coarse particles at Mt. Qomolangma" investigated the soil-derived particulate composition on TP, specifically focused on nitrite, and concluded that this is a major source of HONO on TP. Although I appreciate their work, the manuscript has strong intrinsic weaknesses in the method used in measurement as well as the data interpretations, leaving many large holes in this work. Therefore, this work cannot be accepted in the current form for ACP publication.

Response: We sincerely appreciate the considerable time and effort the referee devoted to reviewing our manuscript. We acknowledge that methodological and data interpretation required clarification and improvement. In response, we have made substantial revisions throughout the manuscript to address the concerns. In the revised version, we have clearly described the field sampling procedures and laboratory analyses. Furthermore, we have improved the data interpretation in accordance with your suggestions, as well as those of the other referees. Please find our detailed, point-by-point responses below. We believe these revisions have significantly improved the scientific rigor and clarity of the manuscript. We respectfully invite you to review the revised manuscript, in which we have carefully addressed all major and minor concerns raised.

Here are my major remarks.

First, let's focus on the methods section. In brief, the sampling method has never been validated in both the lab and field for the TSP nitrite isotopic analysis. The soil extraction method with MQ water is problematic for nitrate and other ions, although it's been proven to be more effective for nitrite. Below are the specific comments for the methods.

Response: Thank you for this helpful suggestion. In the revised manuscript, we have provided a more detailed description regarding the field sampling and laboratory analysis. We would like to emphasize that the filter-based sampling for ambient aerosol and subsequent isotopic analysis of aerosol components is a well-established and widely accepted approach in the scientific community. Numerous peer-reviewed studies have successfully applied this method for isotopic characterization of sulphate, nitrate, ammonium and other water-soluble species in atmospheric particles, and we have followed these established protocols in previous studies. First, the filter-based sampling approach has been widely used for particle nitrite and other water-soluble inorganic ions analysis (i.e., Bhattarai et al., 2023; Bhattarai et al., 2019; Nie et al., 2012; Vernier et al., 2022). After sampling, TSP and $PM_{2.5}$ filter samples were immediately wrapped in pre-baked aluminum foil and stored in frozen until analyzed to minimize potential loss of nitrite. Milli-Q ultrapure water (18.2 MΩ cm) was used to extract the water-soluble inorganic components, and the concentration was determined using ion chromatography. Second, azide method was used for the nitrite isotope analysis following Casciotti et al., 2007. This method has been extensively validated and optimized in our laboratory (Zhou et al., 2022; Zhang et al., 2025), ensuring accuracy and reproducibility of the isotopic measurements in this study.

Regarding the soil ions extraction method: in this study, our goal was to investigate the potential importance of wind-blown dust on the observed high nitrite in TSP. Nitrite in soil was extracted using Milli-Q purewater for the following reasons (1) ultrapure water has been shown to be effective for soil nitrite determination (Homyak et al., 2015); (2) nitrite in TSP and $PM_{2.5}$ samples were also extracted using ultrapure water, ensuring consistency in comparing water-soluble nitrite in surface soil and aerosol. Moreover, the use of Milli-Q water extraction is a common practice in studies investigating the hygroscopic properties of mineral dust (usually with surface soil as surrogate for laboratory-generated dust aerosol) and the sources of water-soluble ions in dust aerosols in arid and semi-arid environments (Gaston et al., 2017; Tang et al., 2019; Chen et al., 2020; Wu et al., 2022).

Line 117, the mountain and valley wind can be from all directions. It is hard to say "upwind" or "downwind" in these areas.

Response: Thank you. We agree that using terms like “upwind” or “downwind” can be ambiguous in mountainous regions due to complex topography and variable wind patterns. However, in the Rongbuk Valley, long-term observations have revealed that the local wind system is characterized as a typical katabatic wind pattern, i.e., the strong down-slope wind along the glacier (i.e., the katabatic or glacier wind) begins in the afternoon, reaches the maximum around sunset and maintains until midnight, and then the wind starts from the opposite direction (i.e., up-slope) (Ye and Gao, 1979; Zhu et al., 2006; Zou et al., 2008; Song et al., 2007). In addition, field observations also indicated a prevailing southeasterly wind direction during the springtime campaign (Figure S1 in supporting information). During sampling, to minimize the influence of local anthropogenic activities on sampling, the instruments were strategically set in the southeast (upwind direction) and approximately 100 m away from the living space of the Base Camp. Therefore, we think it makes sense to use the term of “upwind and downwind” here.

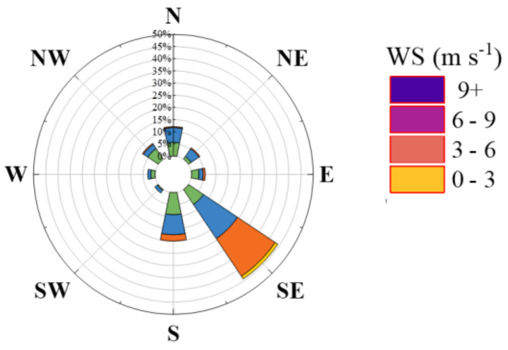

Figure S1. The wind direction and wind speed (WS, m s^{-1}) at the sampling site during the “Earth Summit Mission-2022” scientific expedition in 2022.

Line 126, while the authors claim the Whatman quartz filter collects TSP, no test has been done for HONO collection on the quartz filter. There was no method validation.

Response: Thank you. We would like to clarify that our sampling strategy was specifically designed for the collection and analysis of particulate water-soluble ions, including nitrite (NO_2^-), and not for gaseous HONO collection. One can’t collect HONO with a filter alone, instead in the literature HONO is collected using a denuder system (Chai et al., 2019; Chai et al., 2020). That filters won't collect HONO is also consistent with our observations that the $PM_{2.5}$ filter samples didn’t show any detectable nitrite, i.e., if HONO can be collected in filters, we would expect nitrite to be present in both $PM_{2.5}$ and TSP samples. Therefore, we are confident that the detected nitrite originates from particulate matter rather than gaseous HONO artifacts.

Lack of explanation. Line 130, how is "filter face velocity of approximately 0.288 m s-1." derived?

Response: Thank you for pointing this out. Sorry for the confusion, here we wanted to state the sampling flow (30 L min^{-1}). In the revised manuscript, we have deleted this sentence.

Additionally, details on "surface soil" collection are needed.

Response: Thank you for pointing this out. Additional details on the surface soil sampling have been added to the revised manuscript (lines 152-157):

Surface soil samples (0-5 cm depth, n = 9) were collected in May, 2023 from the east slope, west slope and south sides of the Rongbuk valley. A polytetrafluoroethylene (PTFE) shovel was used to collect soil. The collected soil was immediately transferred to clean plastic bags, sealed and kept frozen. Soil samples were transported into laboratory using a cold chain. Upon arrival at our laboratory, the soil samples were passed through a 60-mesh screen (~0.25 mm) to remove larger particles and thoroughly homogenized prior to chemical and isotopic analysis.

Lines 182-183, based on the statement here, it is confusing why the authors used the suggested $\Delta^{17}O = 0$ for N7373 and N23 from Albertin et al., but only use their own measured $\Delta^{17}O$ value of N10219.

Response: Thank you. The two nitrite standards N7373 and N23 are known with zero $\Delta^{17}O$ as measured by different labs, we just used the Albertin et al. 2021 study to support this, i.e., $\Delta^{17}O = 0$ for the two standards. While for N10219, which is measured with a negative $\Delta^{17}O$ in Albertin et al. 2021, but this value has not been calibrated (note, for zeros one doesn't need to calibrate but for non-zero values calibration are necessary to get the true values). Albertin et al. 2021 just measured N10219 using the azide method and adopted the values of -8.77‰ using the equation of $\Delta^{17}O = \delta^{18}O - 0.52 * \delta^{17}O$ when their measured $\delta^{18}O$ is most close to the true value. Instead, in our lab, as described in our previous publication early this year (Zhang et al., 2025), we developed a technique for measuring $\Delta^{17}O$ of nitrite international standard by oxidation them (and a normal nitrite with $\Delta^{17}O$=0) into nitrate using O_3 and corrected the $\Delta^{17}O$ transfer during the oxidation reaction, and adopted (-9.3 ± 0.2) ‰ as the true value. The normal nitrite with $\Delta^{17}O = 0$ was processed in parallel to quantify $\Delta^{17}O$ transfer during the O_3 oxidation of nitrite following Vicars and Savarino (2014). In the revised manuscript, we have made revision to avoid further confusion (lines 207-217): "The $\Delta^{17}O$ values of the three international references have not been certified. To address this, a series laboratory experiments was conducted to determine the true values of three international references in our laboratory (Zhang et al., 2025). In brief, each nitrite international reference was oxidized into NO_3^- by O_3 produced from commercial ozone generator. A parallel flow of O_3 was also used to convert a normal KNO_2 salt ($\Delta^{17}O$=0) into NO_3^- to quantify the $\Delta^{17}O$ transfer during O_3 oxidation, following the approach of Vicars and Savarino (2014). Based on these experiments, the $\Delta^{17}O$ of RSIL-N7373 and RSIL-N23 are determined to be negligible, consistent with previous findings (Albertin et al., 2021), while the $\Delta^{17}O$ of RSIL-N10219 is determined to be (-9.3 ± 0.2) ‰ in our laboratory."

Lines 189-191, although the ion-exchange method has been verified and used for nitrate ion preconcentration for isotopic analysis, no test has been done for nitrite. Before using the method for field samples, is there supposed to be a lab test for different concentrations and different solution environments?

Response: Thank you for pointing out this. The ion exchange resin method follows the same principle for all soluble ions, which means nitrite and nitrate will all be enriched and then eluted the same. In practice, sure the method needs to be verified. We have done two steps of verification. First, we tested the resin method using nitrite standards, and found there are no significant differences in nitrogen and oxygen isotopes of nitrite standards treated with and without the resin method. Second, for field samples with known amount of nitrite, once the samples were treated by the resin and then measured by mass spectrometer via the azide method, we used the peak sizes of the produced N_2 and O_2 in the mass spectrometer to estimate the recovery of nitrite using a size calibration curve which is established by repeating measurements of nitrite samples with known amounts varying from 30 nmol to 200 nmol. The result indicates ~100 % yield within analytical uncertainty. In the revised manuscript, we have added the above contents in Supporting Information Text S1 as follows: "The performance of the ion-exchange preconcentration for nitrite isotope analysis was evaluated prior to soil nitrite pretreatment in our laboratory. Briefly, 1 mL of nitrite standards (500 nmol mL^{-1}) was diluted to 150 mL and processed following the standard nitrate preconcentration protocol (Erbland et al., 2013). The isotopic analysis demonstrated the ion-exchange method was also effective for nitrite enrichment, with the differences in $\delta^{15}N$ and $\delta^{18}O$ values before and after passage through the ion-exchange resin being less than 1.6‰ across 6 replicates. Moreover, for field samples with known nitrite amount that measured by ion chromatography, the samples were subjected to resin treatment and then analyzed using the mass spectrometry after reduction into N_2O via the azide method. The peak sizes of the resulting N_2 and O_2 gases were used to estimate nitrite recovery via a calibration curve, which is established by repeating measurements of nitrite samples with known amounts varying from 30 nmol to 200 nmol. Results indicated a recovery rate of approximately 100% within analytical uncertainty."

Line 199, it is confusing why 6 days was chosen as the HYSPLIY modeling duration. Why and what fire spots need to be identified?

Response: Thank you for this comment. There are growing body of evidence showing that the anthropogenic emissions and biomass burning emissions in South Asia can come across Himalaya and be transported to the TP region (Zhao et al., 2020; Kang et al., 2019; Zhang et al., 2023; Bhattarai et al., 2023; Lin et al., 2021). Accordingly, air mass back trajectory was modelled to assess the potential impact of pollutants from South Asia via long-range transport.

We apologize for the confusion caused by the earlier incorrect statement regarding the HYSPLIT modeling duration. The backward trajectory analysis in our study used a 3-day run time, not 6 days as previously stated. The choice of a 3-day window is consistent with previous studies investigating transboundary pollutant transport from South Asia to the Tibetan Plateau (Lin et al., 2021; Bhattarai et al., 2023). In this study, the modelled air mass trajectories showed that before April 30th, 2022, the air masses generally originated from South Asia and passed through regions with intensive biomass burning activity, generally in agreement with previous reports (Lin et al., 2021; Cong et al., 2015; Bhattarai et al., 2023). To further assess the potential impacts, fire spots along the modeled trajectories were identified using MODIS fire data. This helps evaluate whether biomass burning emissions could contribute to the elevated water-soluble inorganic ions levels in TSP and $PM_{2.5}$ observed at the site before April 30th. We have clarified this rationale in the revised manuscript.

Lines 164-169, the work used MQ water for soil extraction. While the MQ water may be more effective for nitrite extraction, it has not been proved to be effective for nitrate (and other ions) *extraction.*

Response: Thank you. In brief, one of our primary objectives was to investigate the potential importance of wind-blown dust on the observed high nitrite in TSP. For this purpose, we prioritized the accurate determination of soil nitrite, for which ultrapure Milli-Q water has been shown to be more effective (Homyak et al., 2015). While we acknowledge that ultrapure water may not be optimal for extracting all ions, including nitrate, our focus was specifically on nitrite. Please also refer to our response to your first comment.

The authors suggested that the soil-originated TSP nitrite is an important source of atmospheric HONO, but without measuring atmospheric HONO concentration and its isotopic composition. Without seeing the connections between the TSP nitrite and atmospheric HONO in concentration and isotopes, the conclusion hardly makes any sense.

Response: Thank you. But we wanted to first clarify that, we have not concluded "*TSP nitrite is an important source of atmospheric HONO*". In the manuscript, we just speculated that the high level of nitrite in TSP may sever as a potential source of HONO under appropriate conditions which favors partitioning nitrite ion to HONO in the gas phase. However, whether this thermodynamic partitioning process actually occurs under ambient conditions requires further investigation.

But we wanted to note, our speculation is grounded in the well-established thermodynamic gas–particle phase partitioning of semi-volatile species. Given the pKa of nitrous acid (~3.5) and its pH-dependent Henry's law constant, particulate nitrite can dynamically exchange with gaseous HONO in the atmosphere (Park et al., 1988; Chen et al., 2019; Acker et al., 2008). Previous studies have suggested that high levels of particle-phase nitrite can act as a source of atmospheric HONO through the thermodynamic partitioning process (Li et al., 1994; Chen et al., 2019; Wang et al., 2015; Lammel et al., 1988). Our observation of elevated nitrite in TSP, therefore points to a plausible pathway for HONO production. We have revised the manuscript to more clearly state that this is a hypothesis or speculation supported by thermodynamic theory and prior literature, but make no conclusion on this topic.

Next, the data interpretation for the major statement and conclusion is problematic.

Lines 319-328, it is questionable to use the $\delta^{15}N$ isotopic fractionation factor of snow nitrate photolysis to explain the aerosol nitrate photolysis. In order to determine the role of a potential source in the observed isotopic signatures, one would need to use a source apportionment model to quantify the contribution of each source. Simply comparing the $\delta^{15}N$ values between the nitrite and nitrate doesn't yield any meaningful interpretation and can lead to the invalid statement.

Response: Thank you. We agree that applying the δ^{15}N fractionation factor derived from snow nitrate photolysis to aerosol nitrate may involves uncertainty, as the physical and chemical conditions may differ between snowpack and atmospheric aerosols. However, we note that the photo-induced isotopic fractionation effects during nitrate photolysis in snow and aerosol phase are likely consistent, since in principle it is the difference of zero-point energy (ZPE) between $^{14}NO_3^-$ and $^{15}NO_3^-$ that determines the fractionation effect (Frey et al., 2009; Miller and Yung, 2000). This is why the modeled nitrogen fractionation constant of nitrate in snow by taking the different ZPE (Frey et al., 2009) but no consideration of the snow matrix is very similar, if not identical, to that measured in lab snow nitrate photolysis experiments.

Now back to the statements in our manuscript, the original comparison was intended to refer to existing snow nitrate photolysis fractionation factors as a qualitative framework for interpreting the isotopic difference between nitrite and nitrate observed in our aerosol samples. Theoretical and field studies indicated that $\delta^{15}N$ in NO_2^- from nitrate photolysis will be strongly depleted compared to parent NO_3^- (Frey et al., 2009; Peters et al., 2014). Therefore, the $\delta^{15}N$ similarity between nitrite and nitrate may suggest particulate nitrate photolysis is unlikely the main contributor to the TSP nitrite. Additionally, if particulate nitrate photolysis had played a major role during our sampling campaign, we would expect nitrite to be present in $PM_{2.5}$ samples as well, but not only accumulated in coarse particles. Last, we did not want to do any quantitative assessments here due to the dataset available.

There is not sufficient quantitative analysis on why HONO uptake and NO_2 uptake, and conversion to nitrite pathways are not important. The conclusion that these two pathways are not important was merely based on the assumption or experience that HONO and NO_2 concentrations are very low, while the work did not measure these concentrations. As such, this is a very weak conclusion lacking direct evidence. The work did not show any isotopic evidence, even though there is literature available. What is the lifetime of nitrite on coarse particles, and what is the lifetime on fine particles? Why is nitrite more abundant on coarse particles than fine particles? What is the state of the knowledge? There is no explanation on this at all.

Response: Thank you for your detailed and constructive comments. We acknowledge that our initial discussion of the potential roles of atmospheric HONO and NO_2 uptake pathways was insufficient supported by quantitative analysis and direct observational evidence. Again, we have to state that in-situ measurements on HONO and NOx were not conducted due to logistical and technical limitations of operating such instrumentation at ~5200 m elevation.

Our conclusion regarding the minor contributions of HONO and NO_2 uptake was primarily based on previous field measurements of HONO and NO_2 conducted in the background region of central Tibetan Plateau (i.e., Namco station, ~4700 m, from April to June 2019. Wang et al., 2023) and on general atmospheric chemistry knowledge (Ye et al., 2023). The average mixing ratio of HONO at Namco site was 30 pptv, and the average mixing ratio of NO_2 was 143 pptv (Wang et al., 2023). Given that our study site of the Base Camp of Mt. Qomolangma is situated at a higher elevation (~ 5200m) and in a more remote environment with fewer anthropogenic and natural sources (i.e., soil microbial activity), one would expect the concentrations of these precursors to be even lower than that observed at Namco (Wang et al., 2023). Under such conditions, the high observed levels of particulate NO_2^- (up to 1300 ng m^{-3}) are unlikely to be explained solely by NO_2 heterogeneous reactions or HONO uptake. Moreover, if HONO uptake and NO_2 heterogeneous reactions occurs, nitrite on fine particles should also be detected due to the large surface area and greater potential for heterogeneous reactions, while our observations indicated NO_2^- only exists in coarse particles. Therefore, we believe our analysis is likely sufficient to rule out their significance in our case.

Moreover, we did not incorporate isotopic measurements (e.g., $\delta^{15}N$ of NO_2 or HONO) in our study, although such approaches maybe helpful to identify nitrite formation pathways. Note that the $\delta^{15}N$ isotopes of HONO and NO_2, which is a function of the emission sources and N fractionation effect associated with their atmospheric chemistry, may vary significantly among various atmospheric conditions. We agree that incorporating isotopic evidence of concurrent particulate NO_2^-, HONO and NO_2 would have significantly strengthened our interpretation, and we will investigate this for future research efforts.

Table 2 summarized previous reports on the concentrations of nitrite in TSP and $PM_{2.5}$ samples, and the corresponding formation mechanisms. In this study, we observed nitrite exclusively accumulated in TSP, with no detectable levels on $PM_{2.5}$. If the atmospheric chemistry or processes, such as HONO or NO_2 uptake were a dominant formation mechanism, one would expect nitrite to appear in the fine particle fraction, due to their higher surface-area-to-volume ratio and more efficient uptake capacity. The wind-blown dust or biomass burning emissions via long-range transport are more likely responsible for the observed nitrite. The precise mechanisms remain uncertain and merit further investigation through combined field measurements, laboratory experiments, and modeling studies.

Also, there are several major mistakes in the discussion (Lines 339-347). Specifically, photoenhanced conversion from NO_2 *to* NO_2^- *is not "photocatalysis";* NO_2 *uptake coefficient varies in a significant range, and* $1*10^{-5}$ *or higher is more typically. For example, see Scharko et al. 2017 ES&T (10.1021/acs.est.7b01363). Also, a typo, "initialed" indicates the recklessness of the manuscript.*

Response: Thank you for pointing the mistake out. We have corrected the term "photocatalysis" and now refer to the process more accurately as "photo-enhanced conversion" in the revised manuscript. The typographical error "initialed" has also been corrected to "initiated". In addition, we have thoroughly proofread the manuscript to eliminate similar oversights and improve overall clarity and precision.

Regarding the NO_2 uptake coefficient, we agree that it can vary significantly depending on environmental conditions and the nature of the surface. As noted, Scharko et al., 2017 reported NO_2 uptake coefficients on soil surface as high as 1×10^{-5}, whereas lower values (e.g., $<1\times10^{-6}$) are usually reported on mineral dust or salt surfaces, as summarized in Table 1 of Xuan et al., 2025. Since mineral dust and local soil dominate the TSP composition on the Tibetan Plateau (Kang et al., 2016; Liu et al., 2017; Pokharel et al., 2019), we adopted a lower uptake coefficient in our estimation to reflect the prevailing surface properties. We have clarified this rationale in the revised manuscript and added relevant references to support this choice.

For the investigation of the influence of biomass burning, the authors only did a single back trajectory for each day, which is not sufficient to determine the relative contributions of different air masses transported from different regions. Furthermore, there is no systematic tracer analysis for biomass burning influences. Therefore, the statement in Lines 371-374 is not valid.

Response: Thank you for this valuable comment. We acknowledge that using a single back trajectory per day may has limitations in fully capturing the variability and relative contributions of different air masses. In supporting information, we have now provided four backward trajectories per day at six-hour intervals to provide a more comprehensive view of air mass origins and improve the robustness of our trajectory analysis (Figure S3). The updated backward trajectories also clearly indicate that before April 30th, 2022, the air masses predominately originated from South Asia and likely bring biomass burning pollutants, while air masses predominately originated from clean regions, such as North and Central TP from May 1st to May 5th, 2022. This shift in air mass source regions is consistent with a significant decrease in potassium concentrations (K^+) in $PM_{2.5}$, a common tracer for biomass burning. Specifically, K^+ levels 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 ($p < 0.05$).

Additionally, we agree that the lack of systematic tracer analysis weakens the interpretation regarding biomass burning influences. However, satellite-based fire spot data (MODIS) and K^+ in $PM_{2.5}$ indicated the potential impact of biomass burning emissions for samples collected before April 30th. Furthermore, we have noted this limitation in the revised manuscript and suggested that future studies include more chemical tracers such as levoglucosan to better constrain biomass burning influences.

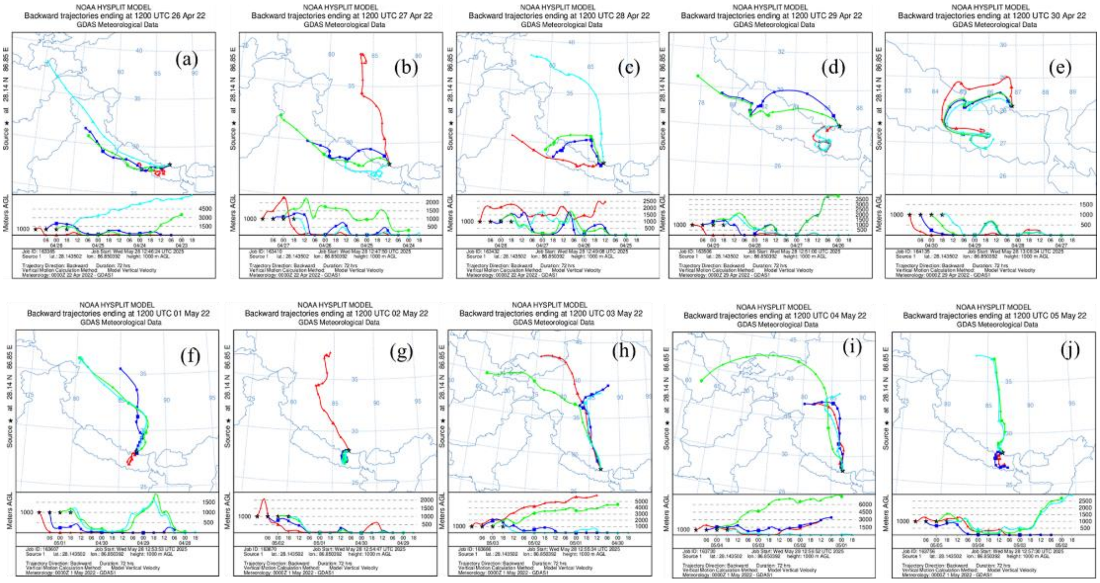

Figure S3. Similar to Figure 4 in main text. The air-mass backward trajectory was modeled at a 6 h interval each day (panels a-j)

Additionally, based on lines 375-381, the authors speculate biomass burning smoke won't contribute to the coarse mode particulate nitrite, so why would the authors still track the air mass? Additionally, it is inaccurate to state that "particle nitrite has not yet been detected in biomass burning plumes," according to the cited literature. These works just didn't measure the nitrite concentrations for some reason, one of which is the detection limit, and nitrite is orders of magnitude smaller than nitrate.

Response: Thank you for this insightful comment. We have revised this section to reflect a more consistent interpretation throughout the manuscript. Specially, our results indicated that biomass burning and anthropogenic pollutants from South Asia through long-range atmospheric transport may contribute to the observed TSP nitrite before April 30th. There are growing body of evidence showing that the anthropogenic emissions and biomass burning emissions in South Asia can come across Himalaya and be transported to the TP region (Zhao et al., 2020; Kang et al., 2019; Zhang et al., 2023 and references therein). The modelled air mass back trajectories further showed that air mass before April 30th, mainly originated from Nepal and northern India, where extensive fire activities occurred (Figure 3). Therefore, the long-range transport of polluted air could contribute to the observed high water-soluble inorganic ions levels in TSP and $PM_{2.5}$ at Mt. Qomolangma before April 30th during the springtime campaign in this study. However, the specific mechanisms responsible for the predominance of nitrite in the coarse particle mode remain unclear,

Moreover, the description regarding "particle nitrite has not yet been detected in biomass burning plumes" was removed accordingly to improve the accuracy of our discussion.

The interpretation of why nitrite in TSP has a higher $\delta^{18}O$ *is invalid. The author states that aerosol water abundance is at least 3 orders of magnitude larger than* NO_2^- *based only on ISOROPIA modeling results without measurement. As we know, the modeled results are highly uncertain and are greatly influenced by the arbitrary input parameters. More important question— What is the hygroscopic property of the soil-derived TSP? What is the relationship between "aerosol liquid water" and TSP? Is the water really in fine particles or coarse particles? Moreover, if the O exchange between water and nitrite is important in aerosol (again, fine particle or coarse particle?), is it supposed to be important in soils? Finally, what is the explanation for the* $\Delta^{17}O$ *of soil nitrite compared to the TSP? Unfortunately, there is no clear discussion on this.*

Response: Thank you for raising these important points and for the opportunity to clarify our interpretation. We acknowledge the uncertainty associated with estimating aerosol liquid water content (ALWC) solely through thermodynamic modeling. In the absence of direct ALWC measurements, thermodynamic equilibrium models such as ISORROPIA II offer a relatively reliable approach for estimating ALWC. Thermodynamic equilibrium model such as ISORROPIA II have been widely used to predict the aerosol acidity and ALWC (Fountoukis and Nenes, 2007), although the outputs are associated with uncertainty. Note that the performance of ISORROPIA II has been validated in term of water uptake measurements in laboratory experiments over a wide range of atmospherically relevant conditions (Fountoukis and Nenes, 2007). While our study did not include direct measurements of TSP hygroscopicity or aerosol water content, previous research has shown that mineral dust can become hygroscopic once emitted into atmosphere, especially when coated with soluble salts (Nie et al., 2012; Lau et al., 2006; Pathak et al., 2009; Tang et al., 2019; Chen et al., 2020). Moreover, Cardoso et al., 2018 estimated that aerosol water could constituted 20 %–30 % of the total aerosol mass in PM_{10} during dust period through a comprehensive ion mass balance approach. These studies suggested that the aerosol water could also be associated with coarse particles, consistent with the prediction from the thermodynamic equilibrium model.

We agree the oxygen isotopic exchange between nitrite and water molecule also occurs in surface soil, which can explain the significant difference of $\Delta^{17}O$ values between nitrite and nitrate in surface soil (3.7‰ versus 9.6‰). First, an extensive field investigation revealed active denitrification process in dryland surface soil at elevations above 5000m from Tibetan Plateau, accounting for 75% of surface soil nitrite (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) than our determined values (3.7‰). Therefore, we suggest the presence of oxygen atom exchange between nitrite and water molecule in surface soil, which should reduce the $\Delta^{17}O$ of nitrite, may contribute to the 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 fully efficient, the surface soil $\Delta^{17}O(NO_2^-)$ should be erased to be negligible. Therefore, the determined positive surface soil $\Delta^{17}O(NO_2^-)$ 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). Moreover, these hygroscopic transformations of the mineral dust aerosols may enhance water uptake (Tang et al., 2016; Kok et al., 2023), which would facilitate the oxygen exchange process between aerosol nitrite and water molecule. It is suggested that the water-soluble inorganic ions, especially sulfate, nitrate and ammonium play an important role in the hygroscopic properties of mineral dust aerosol (Lau et al., 2006; Pathak et al., 2009; Tang et al., 2019). 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 coarse-mode particle surface.

Accordingly, we suggest that the oxygen isotopic exchange between nitrite and water molecule can also explain the observed near-zero TSP $\Delta^{17}O(NO_2^-)$ while soil $\Delta^{17}O(NO_2^-)$ is positive. In the revied manuscript, we added the comparison of $\Delta^{17}O$ between soil nitrite and TSP nitrite according to your and other referee's comments (lines 495-520):

"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), such as desert soil, where the low water moisture content 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‰ and 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 fraction 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 exhibit 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.”

Additionally, what is the Ionic balance for the water-soluble ions in TSP and $PM_{2.5}$?

Response: Ionic balance for the water-soluble ions in TSP and $PM_{2.5}$ are provided in Result section of the revised manuscript (lines 283-294):

“Figure 3 presents the ion balance of measured water-soluble ions in $PM_{2.5}$ throughout the campaign as well as in TSP collected before and after April 30th, respectively, to highlight the significant decline in TSP NO_2^-. There is a strong correlation between cation and anion equivalents in $PM_{2.5}$ samples ($R^2 = 0.70$), whereas the correlations decreased in TSP samples ($R^2 = 0.46$ before April 30th and 0.49 after that, respectively). The nanogram-equivalent weight of cation is significantly higher than that of anions for all samples, with ratio of cation to anion equivalent of ~ 1.5 for $PM_{2.5}$, ~1.9 for TSP collected before April 30th, ~4.2 for TSP collected after May 1st. Clearly, the declines in TSP NO_2^- after April 30th results in relatively higher cation/anion equivalent ratio. The slopes of the correlation lines exceed unity for $PM_{2.5}$ and TSP samples, indicating the alkaline nature of aerosol. The observed deficiency of anion can be attributed to the presence of carbonates (i.e., $CaCO_3$), which can dissolve in water during extraction to release CO_3^{2-} and/or HCO_3^- despite the relatively low solubility (Zhang et al., 2021). “

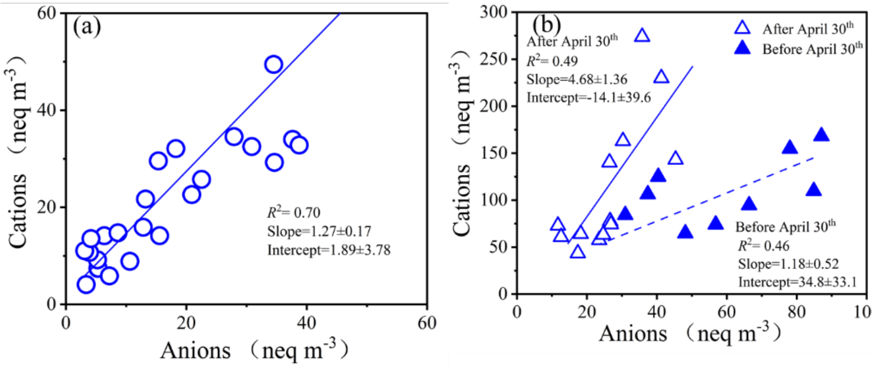

Figure 3. Ion balance for $PM_{2.5}$ (a, blue circles) and TSP samples (b, blue hollow triangles: samples collected after April 30th; blue solid triangles: samples collected before April 30th). Concentrations are expressed in nanogram-equivalent weight per cubic meter (neq m^{-3}).

Additionally, many vague statements prevent me from understanding the key points significantly.

Line 209, what do you mean by "general decline"? Why use 5/1 as the cut-off date? Is there any statistical analysis?

Line 212-213, “negligible” is vague.

Line 214, what is the point of discussing the K^+ in $PM_{2.5}$ that decreases? Why K^+ in TSP didn't show the same trend? Instead, the highest K^+ occurs after 5/1?

Line 215, "comparable" here is not supported by any numerical evidence.

Lines 220-222, which figure or summarized data shows "average daytime concentrations of the inorganic species were generally higher than those at night"?

Lines 217-218, what does the Ca^{2+} and Mg^{2+} data tell you in addition to "smaller degree"? No point can be achieved.

In the paragraph starting with Line 228, the first sentence "The variations of WSIs in TSP generally followed a similar pattern to that in $PM_{2.5}$ (Figure 1)" is an invalid statement as some species have very different trends between TSP and $PM_{2.5}$, such as K^+.

Response: Thank you for the insightful and detailed comments. The key point of our findings is the unexpectedly high levels of nitrite, which are predominately accumulated in coarse-mode particulate. We have revised the result section to clarify the points raised and avoid vague or unsupported statements.

Line 209 ("general decline"): We have replaced the vague term "general decline" with a more precise description, including quantified changes in concentration levels and corresponding time periods. The cut-off date of April 30th was initially chosen based on observed considerable shifts in atmospheric conditions of water-soluble ions and air mass origins, which we now explain more clearly in the revised text. Additionally, we have included a statistical analysis to support the significance of the observed differences in water-soluble ions before and after this date.

Line 212-213 ("negligible"): The term "negligible" has been replaced with specific quantitative values and statistical results to clarify the extent of variation and support the conclusion.

Line 214 (K^+ trends in $PM_{2.5}$ vs TSP): It is well-documented that biomass bringing emissions in South Asia can come across the Himalayas and contribute significantly to the aerosol loadings over the HTP especially during spring (Bhattarai et al., 2023; Zhao et al., 2020; Kang et al., 2019; Zhang et al., 2023). Out results indicated that there is a substantial decline in water-soluble inorganic ions in $PM_{2.5}$ after April 30th in response to the shifts of air masses origins. Before April 30th, air masses mainly originated from or passed through northern India and Nepal with intensive human activities and numerous fire hotspots, while from May 1st to May 6th the air masses originated from the inside of the TP with rare open fires. Therefore, the time series of K^+ in $PM_{2.5}$, a chemical tracer of biomass burning source, was also discussed in this study. In comparison, K^+ in TSP may originated from both biomass burning and crustal emissions (Lin et al., 2021), which may explain the different trend of K^+ in $PM_{2.5}$ vs TSP during the springtime campaign.

Line 215 ("comparable"): We have included the actual numerical data or range of values to support the statement, allowing readers to directly assess the degree of similarity.

Lines 217–218 (Ca^{2+} and Mg^{2+}): We have revised speculative sentence and focused on data-supported findings.

Lines 220–222 (daytime versus nighttime concentrations): We have now referenced the specific figure and provided summarized data (mean ± SD) in the main text or supplemental showing the comparison between daytime and nighttime concentrations for the relevant ions, along with statistical significance where applicable.

Paragraph starting at Line 228 (TSP vs $PM_{2.5}$ trends): We agree with the reviewer that the blanket statement was inaccurate. This sentence has been revised to reflect that "Some water-soluble inorganic ions in TSP showed similar variation trends with that in $PM_{2.5}$ throughout the campaign, while others, such as TSP K^+, exhibited divergent behavior”. We provide specific examples and refer to the relevant figure (Figure 1 in the main text) for clarity.

This section 3.1 was revised as follows (lines 239-258): “Figure 1 displays the chemical compositions of water-soluble inorganic ions, their corresponding time series and fractional contributions in TSP and $PM_{2.5}$. Throughout the campaign, substantial variations of total WSIs in $PM_{2.5}$ and TSP were observed. For $PM_{2.5}$ samples, the mass concentrations of total WSIs before April 30th were higher than that from May 1st to May 6th (4.1 ± 1.7 versus 1.7 ± 0.6 $\mu g\ m^{-3}$; $p < 0.05$). The cut-off date of April 30th was initially selected based on observed significantly declines in concentrations of water-soluble ions and the shifts in air mass origins (presented in section 4). This decline of total WSIs after April 30th was predominately driven by significant reductions in secondary inorganic species, i.e., SO_4^{2-}, NO_3^- and NH_4^+, with the magnitude by more than 60%. In particular, NH_4^+ in $PM_{2.5}$ was on average (322 ± 243) ng m^{-3} before April 30th whereas NH_4^+ in $PM_{2.5}$ collected during daytime and nighttime of May 1st were 1 ng m^{-3} and 3 ng m^{-3}, respectively; and NH_4^+ in $PM_{2.5}$ extractions collected from May 2nd to May 6th was below the detection limit. Therefore, the fractional contribution of secondary inorganic species in $PM_{2.5}$ also decreased (Figure 1d). Similarly, K^+ in $PM_{2.5}$, a good tracer of biomass burning (Ma et al., 2003), also declined significantly after April 30th (269 ± 432 versus 22 ± 12 ng m^{-3}; $p < 0.05$). The elevated concentrations of WSIs before April 30th (4.1 ± 1.7μg m^{-3}) are comparable to previous reports at QOMS station (4.2 ± 2.2 μg m^{-3}) in the spring (Lin et al., 2021). In comparison, concentrations of Ca^{2+} and Mg^{2+}, tracers of wind-blown dust (Wang et al., 2002), decreased by less than 20% after April 30th. In general, SO_4^{2-}, NO_3^-, and Ca^{2+} are the most abundant species in $PM_{2.5}$, accounting for the majority of the mass of total WSIs. In addition, no clear diurnal variation of water-soluble inorganic ions in $PM_{2.5}$ was observed in this study (Figure S2).”

Line 239, “similar areas of filters were extracted,” doesn’t necessarily ensure the accuracy of the comparison because the sampling setups are different.

Response: Thank you for the insightful comment. We acknowledge that differences in sampling design between TSP and $PM_{2.5}$ (e.g., flow rates) may impact the accuracy of direct comparisons. To reduce variability introduced during the extraction process, we used similar filter areas and identical volume of ultrapure water for extraction. It is important to note that, for the same filter area, $PM_{2.5}$ filters generally contained higher aerosol mass loadings than TSP, which likely resulted in higher concentrations of water-soluble inorganic ions in $PM_{2.5}$ extracts. Despite this, nitrite concentrations in $PM_{2.5}$ extracts consistently remained below detection limits, supporting the robustness of our conclusion regarding the absence of nitrite in $PM_{2.5}$.

Vague statement in Lines 282-283, “High soil NO_2^- and NO_3^- concentrations were observed on the west and east…”. “high” here is vague.

Response: Thank you. Revised into: “The soil NO_2^- content on the west slope of Rongbuk Valley (on average 124.7 ng g^{-1}) is higher than that observed on the east and south sides (75.3 ng g^{-1} and 48.3 ng g^{-1}, respectively). Throughout the text, similar revisions have been made in other parts of the manuscript to ensure clarity and precision. We thank you again.

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