

Response to Reviewer Comments #1

The authors have addressed most of the concerns I raised earlier and significantly improved the quality of the manuscript. They acknowledged the limitations of their results and implications, which, in my opinion, have strengthened their discussion. After reviewing the revised manuscript, I find this version to be much improved, and I have only a few minor comments:

Response: We sincerely appreciate the considerable time and effort the referee devoted to reviewing our manuscript again. We have revised the manuscript accordingly to address the points raised.

1. I suggest emphasizing the importance and current knowledge gaps regarding atmospheric nitrite and HONO in the opening paragraph, rather than focusing on glacier retreat. The latter seems less directly relevant to your study than, for example, the observation by Wang et al. of unexpectedly high HONO levels at Namco.

Response: Thank you for this suggestion. We agree that emphasizing the importance and current knowledge gaps related to atmospheric nitrite and HONO would better align with the focus of our study. As such, we have revised the opening paragraph accordingly, particularly referencing the unexpected high HONO levels observed by Wang et al. 2023 at Namco station. The opening paragraph is now revised into:

“The Tibetan Plateau (TP), known as the “Third Pole”, represents one of the most climate-sensitive regions on Earth (Yao et al., 2012). Over recent decades, the TP has experienced significant and rapid climate warming, primarily driven by increasing aerosol loadings and greenhouse gas concentrations due to its geographic proximity to East Asia and South Asia with intensive anthropogenic emissions (Kang et al., 2019; Lau et al., 2010; Lüthi et al., 2015). Atmospheric oxidation capacity (AOC) regulates secondary aerosol formation and trace gases removal, including CH₄ (Wang et al., 2023; Ye et al., 2023; Ye et al., 2016; Andersen et al., 2023), therefore acting as a critical link between atmospheric pollution and regional climate warming. Previous studies have suggested that strong solar radiation, high ozone (O₃) and relatively high water vapor dominate the relatively strong AOC over the TP (Lin et al., 2008). Recent field campaign further highlighted the rapid reactive nitrogen cycling, with N(III) species (i.e., HONO) as the intermediate, also plays an important role in maintaining the strong AOC in TP (Wang et al., 2023). For example, Wang et al. reported high-than-expected HONO ($\sim 30 \pm 13$ pptv) in the Namco station, a representative background site in the central TP, with identified HONO sources including NO₂ heterogeneous conversion, soil emission and particulate nitrate photolysis (Wang et al., 2023). Incorporating the observed HONO into model simulations approximately doubled the estimated OH abundance compared to simulations without HONO constraints. However, a detailed HONO budget analysis indicated these three dominant sources could not account for the observed daytime HONO levels at the background site, implying the existence of additional, yet unidentified, HONO sources.”

2. Reactions R2 and R3 do not appear to be referenced in the main text. Please consider adding relevant citations or removing them if unnecessary.

Response: Thank you. This is caused by adding/removing references in different versions, we should have been more carefully reviewing these details after each revision. We have removed in the newly submitted revision.

3. Since you propose that nitrite sources differ before and after May 1—mainly from soil after May 1, and a mix of soil and long-range transport before—Section 4.3 would be more consistent if you compare only the post-May 1 samples to the soil signature. For instance, the $\delta^{15}\text{N}$ value of “soil-derived nitrite” should be $-8.0 \pm 0.7\text{‰}$ rather than $-7.3 \pm 3.1\text{‰}$.

Response: Thank you for this valuable suggestion. The sentence was revised accordingly:

“In addition, the comparable $\delta^{15}\text{N}$ values of NO_2^- between TSP collected after May 1st ($-8.0 \pm 0.7\text{‰}$) and the surface soil ($-10.3 \pm 3.0\text{‰}$) also supports that locally resuspended surface soil is an important contributor to the observed high levels of TSP NO_2^- .”

Response to Reviewer Comments #3

Here is a major question that needs to be addressed. The work reported isotopic signatures of nitrate. Was this nitrate also extracted MQ water? Is it necessarily a complete extraction? If not, is there any fractionation?

Response: We sincerely appreciate the considerable time and effort the referee devoted to reviewing our manuscript again.

For nitrate extraction from the Whatman quartz filters using Milli-Q ultra-pure water, we have tested the extraction efficiency using a double-extraction method: after the initial 20 mL Milli-Q ultra-pure water extraction in an ultrasonic bath for 30 min at room temperature, an additional 20 mL of ultra-pure water was used to re-extract the same filter. The nitrate concentration in the second extract was generally less than 3% of that in the first, indicating that the initial extraction removed nearly all of the nitrate in the filter. Therefore, ultrasonic extraction with Milli-Q ultra-pure water provides near-complete extraction of nitrate from Whatman quartz filters, and thus there should be negligible effects of isotope fractionation.

For soil nitrate isotope analysis, a 2M KCl solution was used (5 g soil with 15 mL 2M KCl) following the method of Fang et al. (2015). The N and O isotopic signatures were determined using the denitrifier method, in which nitrate in the KCl extract is converted to N₂O gas and subsequently analyzed for its isotopic signature. Since the nitrate isotope in surface soil is invoked in this version of manuscript, the method for soil nitrate isotope analysis was included according to your suggestion. Thank you once again for your valuable feedback.

Reference

Fang Y, Koba K, Makabe A, et al. Microbial denitrification dominates nitrate losses from forest ecosystems[J]. Proceedings of the National Academy of Sciences, 2015, 112(5): 1470-1474