Point-by-point response letter

Note: The black font are comments from the referees, and the red font are authors' responses as well as the related change clarifications.

(1) Detailed response to comments from referee #1:

General Comments

This paper uses the closed-loop integration method, together with OMI and TROPOMI satellite observations and ground-based monitoring data, to systematically analyze the spatiotemporal variations of tropospheric NO_2 column concentrations, surface concentrations, and boundary transport fluxes over the Tibetan Plateau from 2005 to 2024. Given the important role of the Tibetan Plateau in regional and global climate systems, this study focuses on the key issue of transboundary NO_2 transport. A random forest approach is also introduced to assess the relative contributions of the westerlies and the South Asian monsoon at different altitude layers to the transport flux. Overall, the results contribute to a deeper understanding of regional pollutant transport processes and their controlling factors and provide references for studies on cross-regional coordinated emission reductions. This study employs a series of advanced datasets and innovative methods, and the relevant conclusions hold scientific value and provide useful reference. This paper fits the scope of ACP, and I recommend publishing it on ACP after some issues have been addressed.

Some aspects could be further refined to enhance the readability and rigor of the manuscript.

1. I noticed that the authors did not include a section on the validation of the machine learning results in the main text, although I saw it in the appendix. I believe this is a crucial part and the validation parameters (such as R2 and RMSE) should be presented in the main text. I recommend including a table in the main text to demonstrate this.
   Response: Thank you for your comment. The validation parameters of the model (e.g., R^2 and RMSE) are now presented in the main text. A detailed summary is provided in Section 2.2.1 (Table 2).

1. There are minor grammatical and expression issues in the manuscript, which are suggested to be revised to improve overall readability.
   Response: Thank you for your comment. We have carefully revised the manuscript to improve readability and ensure rigor.

Specific comments:

1. Page 2, Line 25: “…currently operated in TP, Taking Tibet as an example…” → “…currently operated in TP. Taking Tibet as an example…”
   Response: Corrected as suggested..

1. Page 5, Line 29: “To ensure strict temporal and spatial consistency, …” should be “To ensure strict spatiotemporal consistency, …”.
   Response: The text has been revised accordingly.

1. Page 6: The formula formatting here is incorrect; the author should adjust it. The same issue appeared in the supplement.
   Response: The formula formatting has been corrected in both the main text and the Supplement.

1. Page 7, Line 1: “…some grids in this region that previously exhibited mixed fluxes shift from internal efflux to external influx…” The definition of “mixed fluxes” is not sufficiently clear.

1. Clarify the meaning of “mixed fluxes” in the text, for example, whether it refers to net fluxes close to zero or to spatiotemporal alternation.
   Response: Thank you for your comment. The relevant section has been substantially revised. The ambiguous term “mixed fluxes” has been removed and replaced with a clearer description: *“some boundary grid cells transition from net efflux in the non-monsoon period to net influx during the monsoon season, thereby forming a more continuous influx structure.”*

1. Page 8, Line 2: A large number of references are cited (Guo et al., 2022; Kong et al., 2023; Zheng et al., 2024; Zhang et al., 2025), but the specific phenomena or mechanisms corresponding to each reference are not clearly indicated in the text.

1. Clearly indicate the specific phenomena or conclusions corresponding to each reference when citing, to avoid citation stacking.
   Response: Thank you for your comment. The relevant section has been substantially revised. The text has been largely rewritten, and several redundant citations have been removed to avoid citation stacking.

1. Page 8, Lines 16-21: The text repeatedly mentions “high-value” or “high-value source regions” → it is recommended to define it upon first occurrence. Is it referring to PSCF values above a certain threshold? Please clarify.
   Response: Thank you for your comment. “High PSCF value regions” are used in this study as a qualitative description of areas with relatively higher PSCF values, indicating locations with stronger potential source contributions. We have standardized the related terminology in the revised manuscript to avoid

ambiguity. Please see Lines 12 to 21 on page 8.

1. Page 9, Line 2: "…pivotal role over TP…" → "pivotal role of the TP" is more accurate.
   Response: Corrected as suggested.

1. Section 3.3 Local emissions and regional transport of NO_2 over the TP

1. Page 9, Line 7: "…have exhibited exhibited distinctly different spatiotemporal evolution patterns…" → "…have exhibited distinctly different spatiotemporal evolution patterns…".
   Response: The repeated word has been removed, and the sentence has been corrected.

1. Page 9, Lines 16-17: "…resulting in further spatial expansion of NO_2 hotspots(Choudhary et al., 2021) …" → "…resulting in further spatial expansion of NO_2 hotspots (Choudhary et al., 2021) …".
   Response: A space has been added before the parenthesis.

1. Page 9, Line 22: "one of cleanest regions" → "one of the cleanest regions"
   Response: Corrected as suggested.

1. **Page 14, Line 38:** "…pristine background region, the TP is increasingly…" → "…pristine background region, is increasingly…"
   Response: Corrected as suggested.

Figure comments

1. Figs. 3–5: The figures and text are a bit small. Making both bigger would improve readability.
   Response: Thank you for your comment. We have carefully reviewed all figures in the manuscript and adjusted their size and text formatting to improve readability throughout the paper.

(2) Detailed response to comments from referee #3:

The authors analysed the transport of NO2 over the Tibetan Plateau using OMI and TROPOMI observations in a ML-model that they have published before. They found a seasonality and a spatial gradient in the NO2 columns over the TP with increasing NO2 levels.

The topic can be interesting but I currently miss several aspects in this research: The chemical lifetime of NO2 was not studied, validation of the results is missing and the effect on air quality is not very convincing. Below I will give a more detailed account of my concerns.

Response: We sincerely thank the reviewer for their thorough and insightful comments. We have conducted a comprehensive revision of the manuscript in response, with detailed replies provided below.

When presenting transport of NO2 a discussion of its chemical lifetime is expected. The relative short lifetime of hours for NO2 distinguishes it from a passive tracer and also from CO. The transport of NO2 was modelled based only on concentrations and wind fields, while the chemical lifetime was not taken into account. If the temperature was take into account in the ML model, at least a simple link could have been made with the chemical lifetime, which is temperature dependent.

Response: We acknowledge that the relatively short chemical lifetime of NO_2 distinguishes it from long-lived tracers such as CO, and thus its cross-regional transport must be interpreted with caution. In the original manuscript, fluxes were calculated using monthly mean NO_2 concentrations, which smoothed the daily variability of this short-lived gas and could potentially underestimate transport differences. To better account for NO_2's short lifetime, the revised manuscript calculates fluxes using daily mean NO_2 concentrations during OMI overpasses (local time approximately before 13:30), with monthly statistics used to examine seasonal variability. While the use of daily data slightly adjusted some flux values, the overall transport patterns and scientific conclusions remain consistent, and the long-term seasonal features are represented more robustly. This improvement not only enhances the physical plausibility of the analysis but also aligns the methodology with the chemical lifetime characteristics of NO_2, thereby increasing the reliability and interpretability of the results.

Furthermore, previous studies indicate that NO_2 exhibits a chemical lifetime of 1.5–15 hours over India, which can extend to 20–24 hours over Bangladesh and Myanmar (Graham et al., 2024); in the typical planetary boundary layer, the lifetime is approximately 8 hours, and it may persist for several days in the upper troposphere. Lower free radical concentrations can further extend the effective lifetime to approximately 2–4 days (Ehhalt et al., 1992; Beirle et al., 2011; Zien et al., 2014). Despite its short lifetime, multi-day backward trajectories (e.g., 3 days) have been successfully used in previous studies to identify transport pathways (Meena et al., 2012; Sharma et al., 2023; Prakriti et al., 2024; Zhang et al., 2025a). In comparison, this study employs a 2-day backward trajectory, representing a more conservative time window for tracing air mass origins, while remaining consistent with the 2–4 days timescales commonly used in previous studies.

It is important to clarify that our transport framework does not rely on a simple coupling of concentration and wind fields. Wind field correction is a key step in flux calculation, during which multiple physically relevant factors are considered, including temperature distributions across different pressure levels, GEOS-Chem simulated air mass quantities, and NO_2 vertical profiles. These variables are used to weight and adjust both the wind and concentration fields, thereby providing a more physically consistent representation of atmospheric transport while accounting for thermodynamic conditions affecting NO_2 distribution.

Finally, following the reviewer's suggestion, temperature was incorporated as a predictor in the random forest model, enabling the model to capture the response of NO_2 to temperature variations and its relationship with chemical lifetime. The atmospheric lifetime of NO_2 is highly sensitive to environmental conditions. As the altitude in the troposphere increases, the lifetime of NOx extends, and the reduced concentration of radicals further prolongs the residence time of NO_2 (approximately 2 to 4 days), thereby enhancing its interregional transport efficiency (Liu et al., 2024; Zien et al., 2014). During the transport process, a fraction of NO_2 is converted into reservoir species (e.g., peroxyacetyl nitrate, PAN), which are subsequently re-released during subsidence, modulating the spatial distribution of NO_x (Zien et al., 2014). Moreover, temperature variations affect the rate at which NO_2 is converted into PAN and other reservoir species, thereby further influencing the efficiency of these processes. By incorporating temperature into the model, we are able to account for its effect on the chemical lifetime of NO_2. Please see the details in lines 4 to 20 on page 11 and Figure 1 in this section.

In summary, these improvements allow for a more comprehensive consideration of NO_2's short lifetime and temperature dependence, thereby enhancing the accuracy of the transport analysis and the robustness of the results.

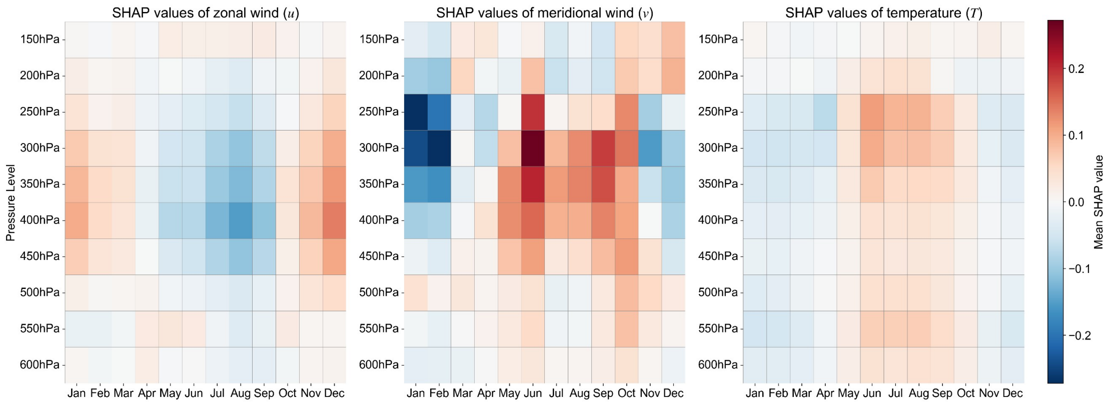

Fig. 1. SHAP-based contributions of *u*, *v*, and *T* to NO_2 transport flux variations across the southwestern Tibetan Plateau boundary, with red and blue indicating positive and negative contributions, respectively.

The authors show that the incoming flux and outgoing flux over the TP is more or less equal, which is puzzling. Does this mean that there is no relevant chemical loss, or is the loss coincidently equal to the local emissions over the TP, or do other chemical processes (conversion with PAN) play a role? Maybe the chemical lifetime is especially long for this region? These questions are not answered or even discussed.

Response: We thank the reviewer for the comment. Regarding your point that the incoming and outgoing fluxes over the Tibetan Plateau are "more or less equal" (corresponding to lines 30–32 on page 7 of the original manuscript), we have re-examined the text and recognize that the previous statement was not sufficiently clear and may have caused misunderstanding. Our intention was not to suggest that the incoming and outgoing fluxes are nearly identical, but rather to highlight that their rates of change (i.e., growth rates) are comparable. This apparent similarity may be influenced by the temporal averaging applied (e.g., monthly mean NO_2 concentrations smoothing short-term transport signals), which can partially mask instantaneous differences.

Furthermore, we emphasize that the incoming and outgoing fluxes over the Tibetan Plateau do not cancel each other, and the net flux is not close to zero. The net flux at different boundaries (influx - efflux) exhibits clear regional variations, as shown in Figs. 2 of this response and Figure 4 of the revised manuscript.

Given that this section has been substantially revised in the manuscript, the previous statement that may have caused ambiguity has been removed (please see lines 11 to 16 on page 7). It should be noted that the revised fluxes are diagnosed using NO_2 concentration fields at satellite overpass times together with corrected wind fields, representing instantaneous boundary fluxes. Because these observations correspond to a relatively short time window (i.e., the satellite overpass), chemical conversion of NO_2 is unlikely to produce significant effects at this timescale, and the fluxes primarily reflect instantaneous atmospheric transport. Further discussion of chemical processes during transport can be found in lines 4 to 20 on page 11.

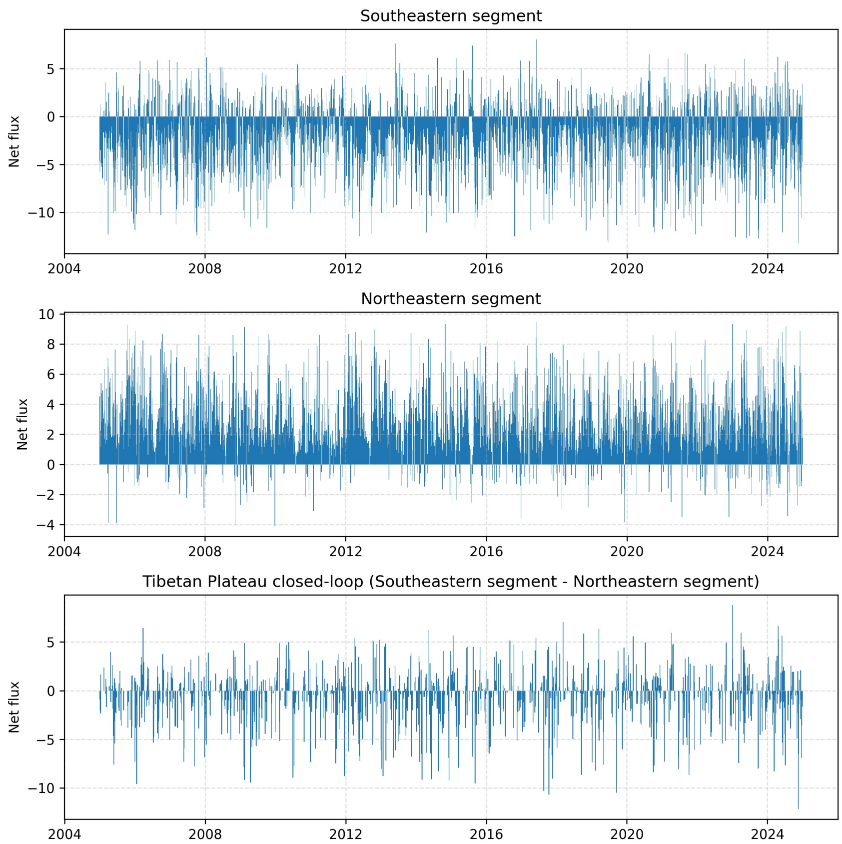

Fig. 2. Net fluxes across the southwestern, northeastern, and enclosed boundaries of the Tibetan Plateau from 2005 to 2024.

It is difficult to judge the reliability of the results when no validation is presented. There are apparently 30 sites with in-situ observations that could be used for comparison at ground level of the model, but they are largely ignored. I am also surprised that the model has layers from 1000 hPa (in steps of 25 hPa) as shown in Figure 9, while the TP has typically ground pressure levels of less than 600 hPa, which put the Figure in a completely different perspective.

Response: We thank the reviewer for this valuable comment. In the initial configuration, the model vertical levels were defined starting from 1000 hPa with 25 hPa intervals. However, due to the high elevation of the Tibetan Plateau, surface pressure is typically below approximately 600 hPa. This apparent discrepancy arises because the wind field data used to train the model covered a spatial domain slightly larger than the Tibetan Plateau. This was necessary to compensate for the relatively coarse grid resolution of ERA5 (0.25°) and to ensure that key dynamic signals of the South Asian monsoon and mid-latitude westerlies were captured. As illustrated in Fig. 3 of this response, the purple solid line indicates the geographic boundaries of the Tibetan Plateau, while the red dashed line shows the spatial coverage of the wind fields used for model training. The moderately expanded domain includes some low-altitude regions (e.g., areas around ~70°E) where surface pressure approaches 1000 hPa, leading to vertical levels that do not perfectly match the high-altitude surface conditions of the Plateau. In the revised manuscript, we optimized the spatial extent of the wind field inputs to better match the Plateau's topography, resulting in vertical structures that more realistically represent the atmospheric layers relevant to the study region. Please check the detailed in Lines 19 to 20 on page 5.

Moreover, we fully agree that ground-based observations are valuable for model evaluation. To explore the potential relationship between surface measurements and flux variations, we selected two monitoring sites relatively close to the boundary for analysis (see Figs. 4–5 of this response). Taking Chamdo as an example, the ground station is located approximately 200 km from the boundary flux grid point. Analysis of high-pollution days (surface NO_2 exceeding the 90th percentile) shows that surface concentrations and net incoming fluxes exhibit broadly similar trends under certain conditions. This suggests that surface observations may partially reflect cross-regional high-input events. However, since surface measurements primarily capture near-surface concentrations, the stations are relatively distant from the boundary, cross-regional transport involves losses, and the flux is calculated based on NO_2 vertical distributions and wind fields from the surface to the upper troposphere, representing transport of the entire atmospheric column, this correspondence involves some uncertainty and should be interpreted with caution for quantitative validation purposes.

To systematically assess the reliability of our flux calculation approach, our team previously conducted similar studies in the Yangtze River Delta and Beijing regions, combining vehicle-based differential optical absorption spectroscopy (DOAS) measurements with high-resolution flux simulations to evaluate both local and regional transport characteristics (Huang et al., 2020; Zhang et al., 2025b). The results indicate that the method can reasonably capture regional transport features and is validated under similar conditions. Collectively, these analyses support the applicability of our flux calculation approach for investigating cross-regional transport over the Tibetan Plateau.

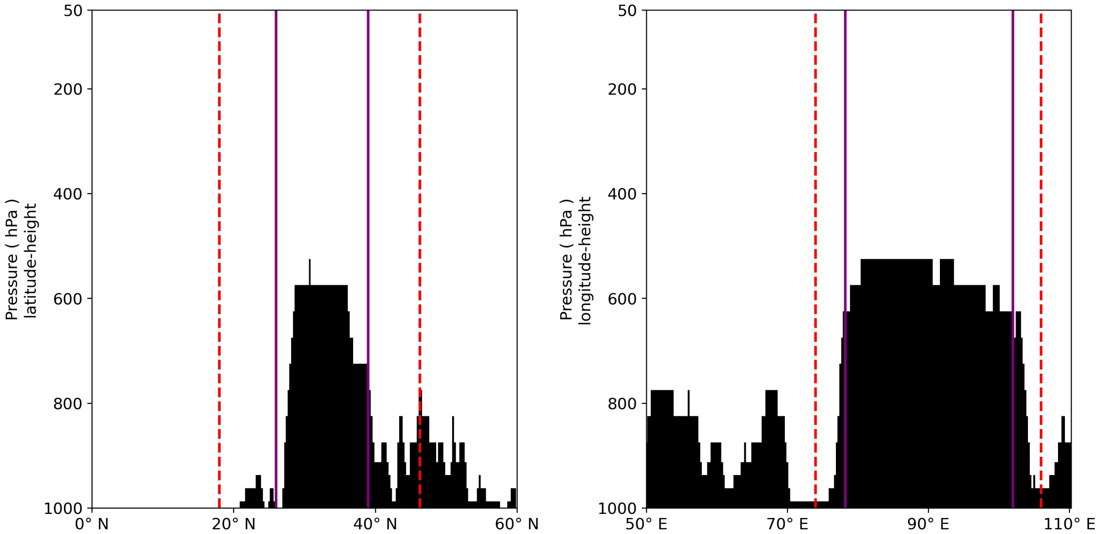

Fig. 3. Schematic of latitude–height and pressure–height profiles over the Tibetan Plateau. The purple line indicates the Plateau region, and the red dashed line marks the wind field range selected in the model, which initially included low-altitude areas. In the revised manuscript, the region was optimized. The topography is approximately derived from the range of NO_2 profiles and is for reference only.

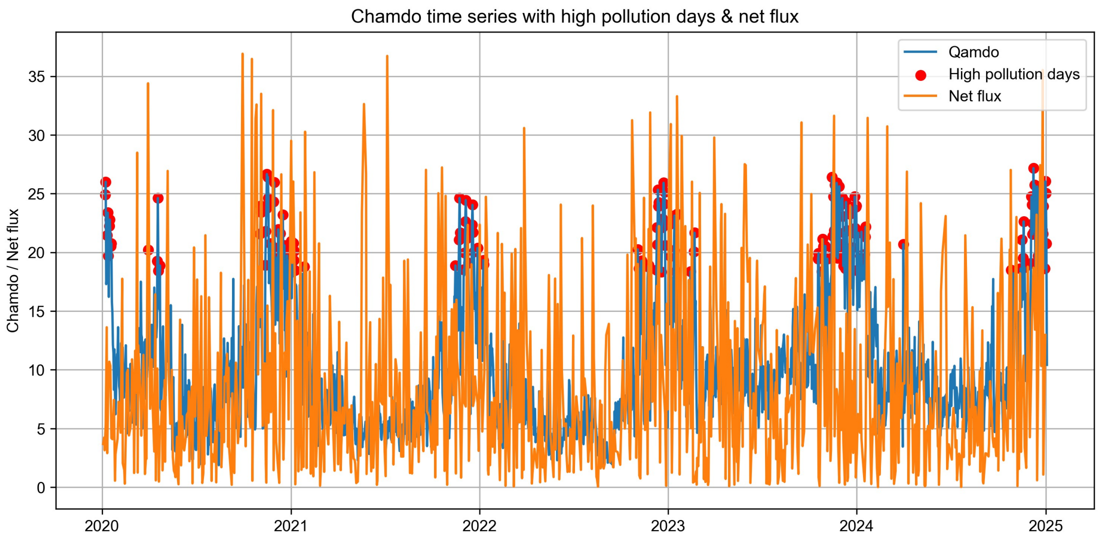

Fig. 4. Relationship between net NO_2 transport flux and surface NO_2 concentrations during high-pollution episodes in Chamdo

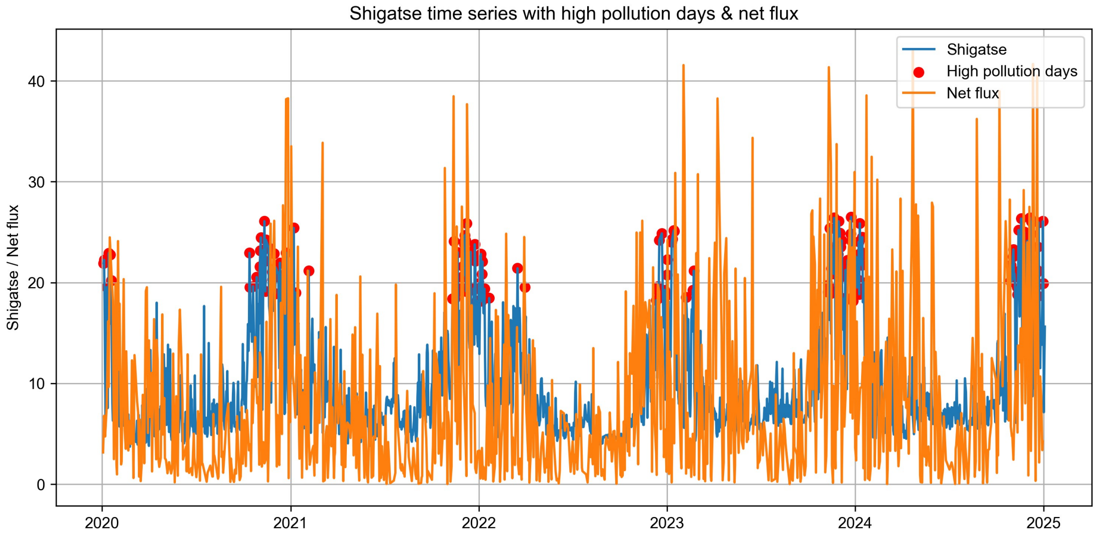

Fig. 5. Relationship between net NO_2 transport flux and surface NO_2 concentrations during high-pollution episodes in Shigatse

The conclusion that this study is important for air quality is difficult to understand. Figure 1 shows concentrations that are too low to be visible with the used colour scale. The authors claim that the ground observations are showing a decreasing trend but are unreliable. I would like to know why they are considered unreliable. The authors also conclude that the transport from polluted areas takes place on higher levels in the atmosphere. Would this mean that they do not affect the surface level? On top of this, the receptor area of the NO2 transport in China has already high concentration levels due to local emissions, so the effect would also be very limited for this region.

Response: We thank the reviewer for this valuable comment. First, regarding the reviewer's concern that the NO_2 concentrations in Fig. 1 of the main text appear visually low and difficult to discern, we agree that this issue primarily arises from technical limitations in the original color scale range and colormap design, which did not adequately represent the variability of low NO_2 values over the Tibetan Plateau. In the revised manuscript, we have accordingly adjusted both the color scale range and the binning scheme. Specifically, the mean NO_2 column density over the Tibetan Plateau during 2005–2024 is 5.7×10^{14} molecules cm^{-2}, whereas the corresponding mean value over India is approximately 2.0×10^{15} molecules cm^{-2}, about 3.5 times higher. In addition, South Asia exhibits pronounced localized emission hotspots; for example, the Indo–Gangetic Plain shows mean values of up to 3.6×10^{15} molecules cm^{-2}, approximately 6.3 times higher than those over the Tibetan Plateau. This strong spatial gradient further enhances the visual contrast, making the Tibetan Plateau appear relatively lower in comparison.

Regarding the description of the ground-based observational data, the expression in the original manuscript was not sufficiently accurate. We did not intend to suggest that the ground-based observations over the Tibetan Plateau are "unreliable," but rather to point out that, due to the sparse population in this region, monitoring stations are mainly located in urban areas, leading to certain limitations in their spatial representativeness. At the same time, as shown in Supplementary Fig. S13, atmospheric dynamical processes may provide conditions for the transport of nitrogen oxides to the near-surface layer, and such influences may occur in regions not covered by the existing urban monitoring sites. Therefore, the current ground-based observations may not fully reflect the regional background conditions. We have revised the relevant descriptions in the manuscript to avoid ambiguity and over-interpretation. Please see details in lines 2–11 on page 14.

In addition, regarding China as a receptor region, we agree that local emissions have already led to relatively high NO_2 concentrations. Under this context, the contribution of external transport to absolute concentrations may be relatively limited; however, it remains important for understanding inter-regional pollutant transport processes. During the pre- and post-monsoon periods, NO_2 emissions from South Asia can be transported in large quantities to the Tibetan Plateau and its downstream regions via mid-tropospheric wind fields. Orographic uplift and the complex terrain of the Tibetan Plateau favor the formation of transport pathways in the middle and upper troposphere. In this process, the Tibetan Plateau acts as a key "relay station" along the regional transport pathway (see Figs. 6–10 of this response). Therefore, the revised manuscript focuses the relevant discussion on the transport processes and dynamical characteristics within the Tibetan Plateau region. Please see Section 4.2 of the main text.

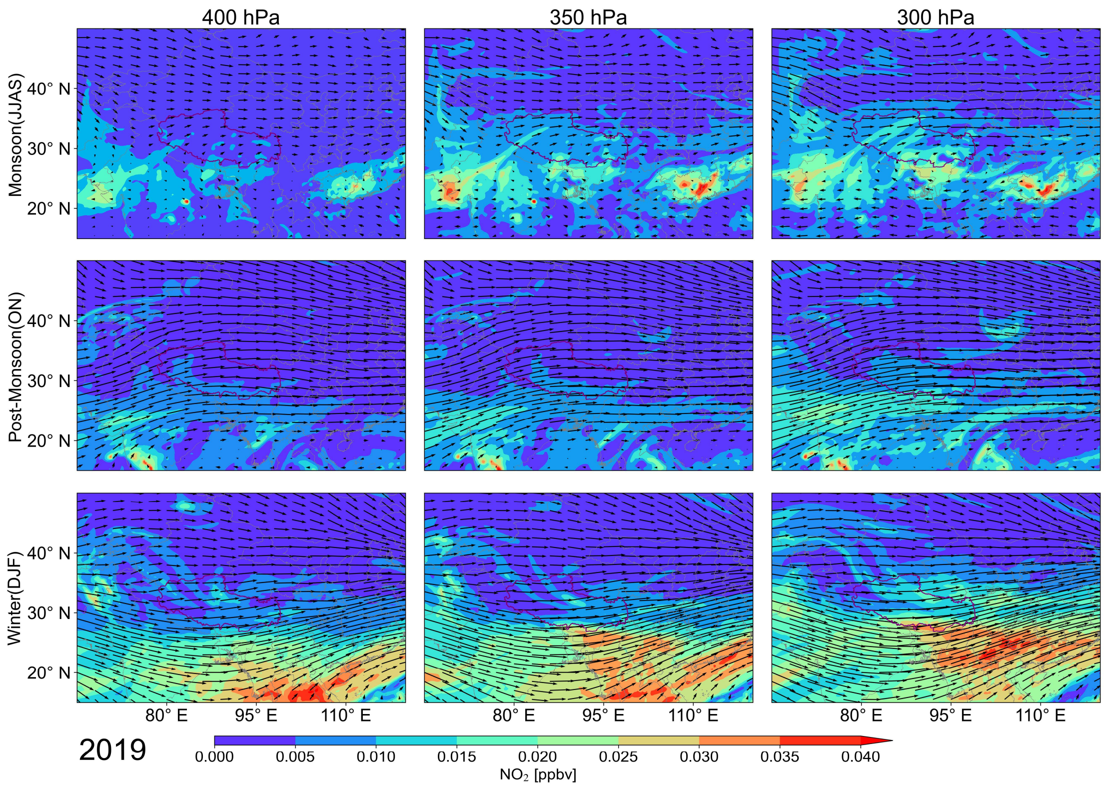

Fig. 6. NO_2 concentration and wind fields over the Tibetan Plateau at 400, 350, and 300 hPa during the monsoon, late-monsoon, and winter seasons of 2019. NO_2 concentrations are from GEOS-CF and wind fields from ERA5.

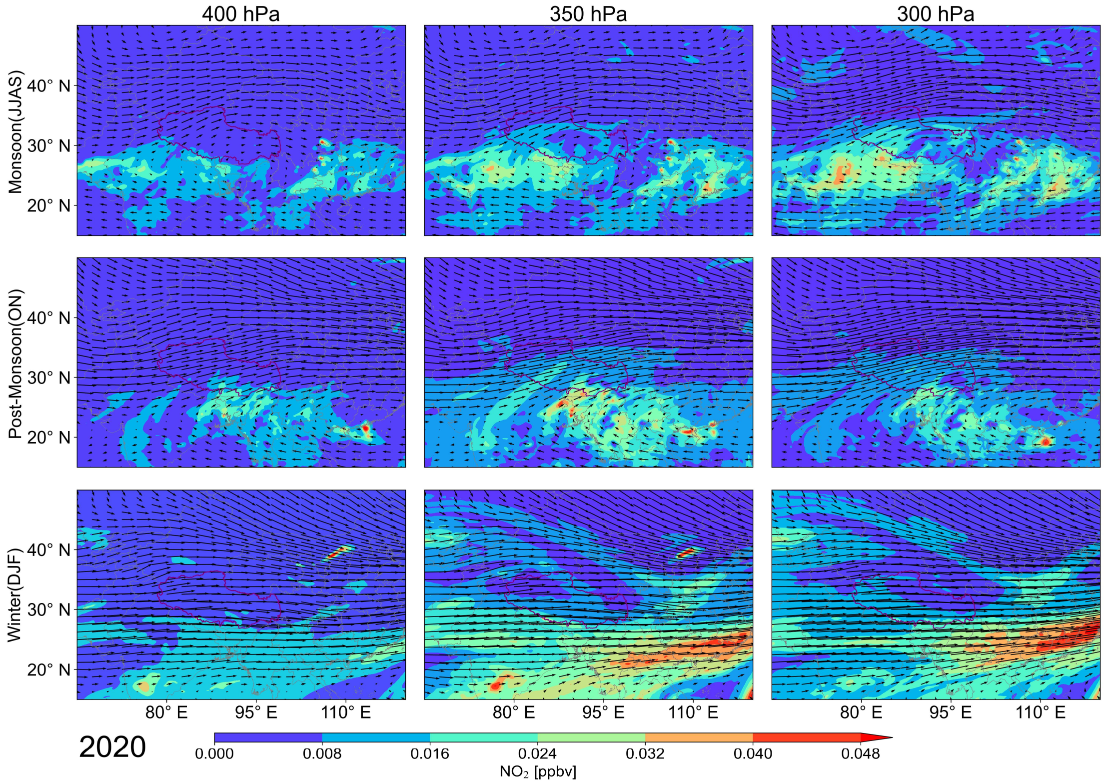

Fig. 7. NO_2 concentration and wind fields over the Tibetan Plateau at 400, 350, and 300 hPa during the monsoon, late-monsoon, and winter seasons of 2020. NO_2 concentrations are from GEOS-CF and wind fields from ERA5.

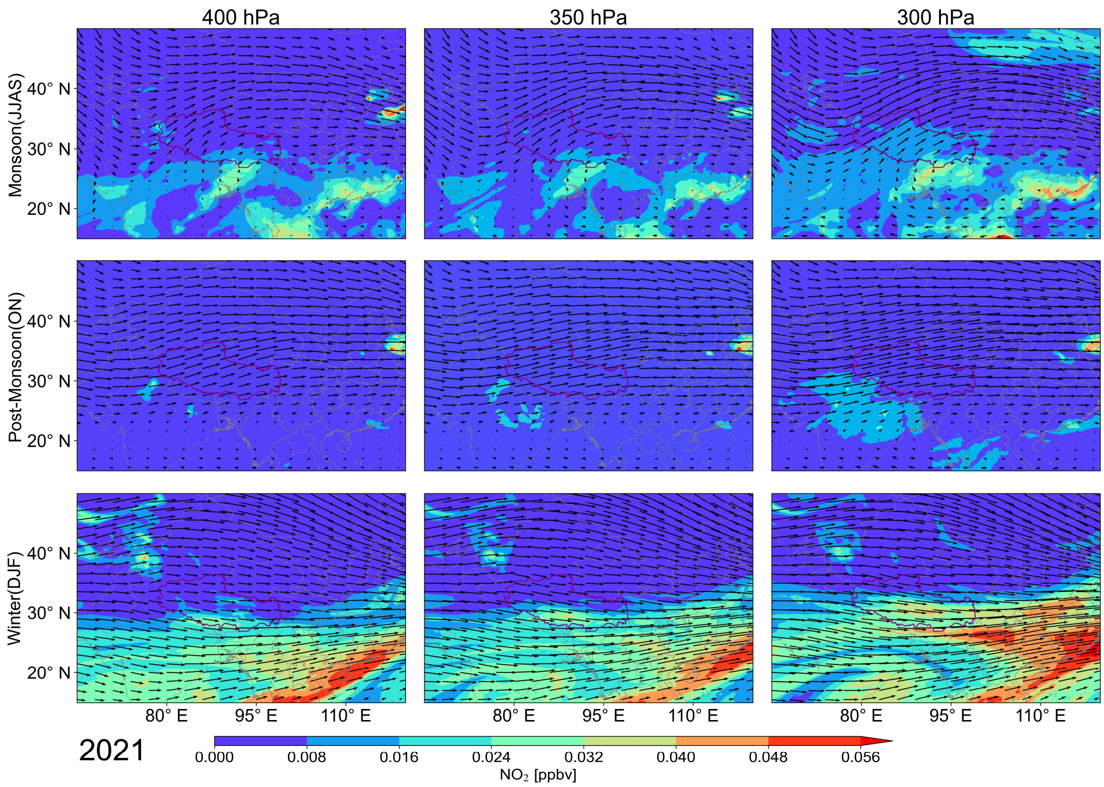

Fig. 8. NO_2 concentration and wind fields over the Tibetan Plateau at 400, 350, and 300 hPa during the monsoon, late-monsoon, and winter seasons of 2021. NO_2 concentrations are from GEOS-CF and wind fields from ERA5.

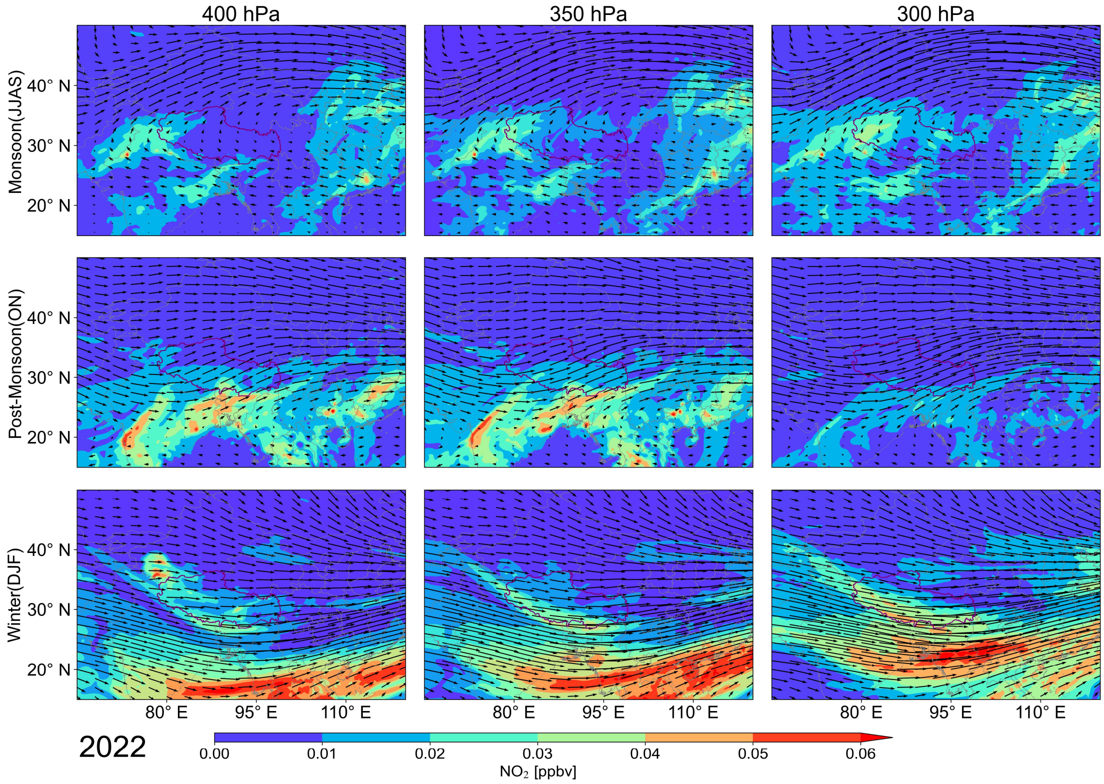

Fig. 9. NO_2 concentration and wind fields over the Tibetan Plateau at 400, 350, and 300 hPa during the monsoon, late-monsoon, and winter seasons of 2022. NO_2 concentrations are from GEOS-CF and wind fields from ERA5.

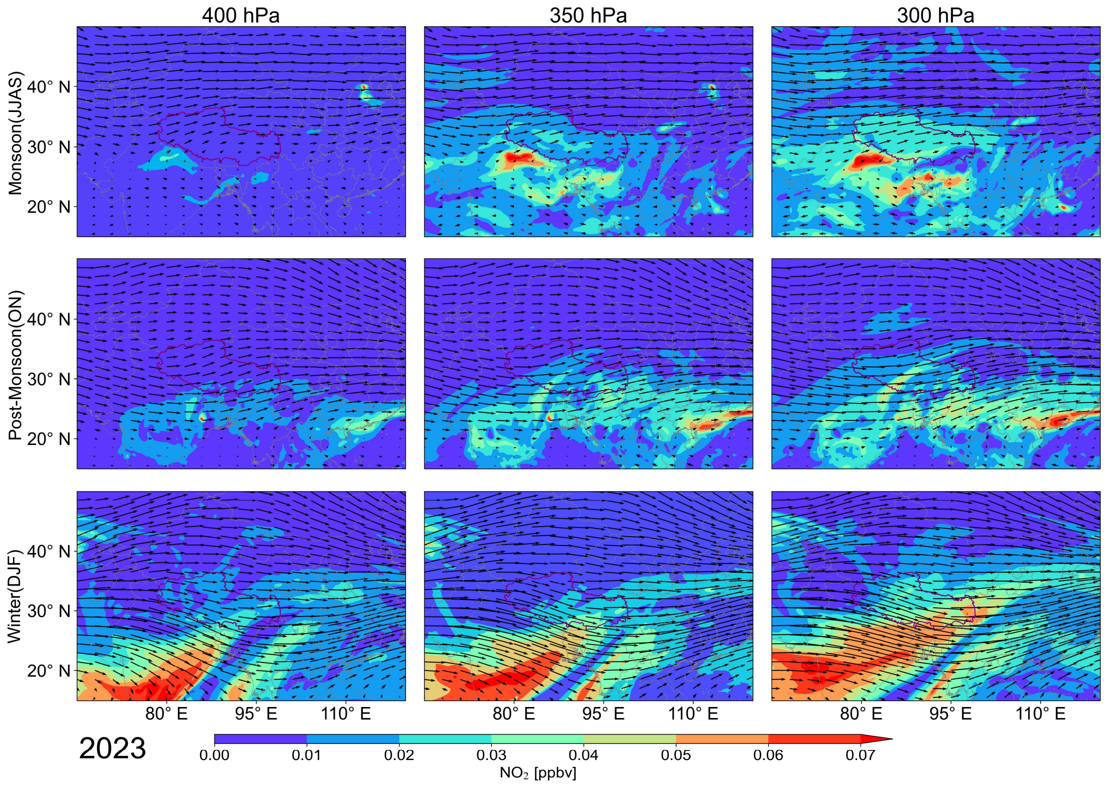

Fig. 10. NO_2 concentration and wind fields over the Tibetan Plateau at 400, 350, and 300 hPa during the monsoon, late-monsoon, and winter seasons of 2023. NO_2 concentrations are from GEOS-CF and wind fields from ERA5.

While focus of this study is the TP, this region is completely white in Figure 1 with no visible NO2 concentrations. There are also no other figures of observed concentrations either from satellite or the ground. This provides no reference for any conclusions about the relevance for air quality.

Response: We thank the reviewer for pointing out that NO_2 concentrations over the Tibetan Plateau appear relatively low in Fig. 1 of the main text and for raising concerns regarding its implications for air quality. As correctly noted by the reviewer, NO_2 concentrations over the Tibetan Plateau are generally low. However, satellite observations (OMI) indicate a continuous increasing trend over the past two decades, from a mean concentration of 3.6×10^{14} molecules cm^{-2} in 2005 to 5.5×10^{14} molecules cm^{-2} in 2024, representing an increase of approximately 52.8%. Supplementary Materials S2 and S3 further provide a more comprehensive analysis of satellite and ground-based observations over the Tibetan Plateau, offering a more complete comparison and discussion. To better characterize the sources and contributions of NO_2 over the Tibetan Plateau, we consider both local emissions and long-range transport from South Asia. Accordingly, we have revised the analysis to focus on the transport mechanisms and dynamical characteristics of NO_2 in the middle and upper troposphere.

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