



Supplement of

**Cross-regional NO₂ transport over the Tibetan Plateau (2005–2024):
bidirectional flux dynamics, seasonal drivers, and
environmental implications**

Zhenda Sun et al.

Correspondence to: Youwen Sun (ywsun@aiofm.ac.cn) and Cheng Liu (chliu81@ustc.edu.cn)

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Section S1. Detailed Methodology

S1.1 The closed-loop integral method

The closed-loop integral method is employed to quantify the NO₂ transport flux across the TP, as defined in Equations (1)-(3). A schematic overview of the procedure is provided in Fig. S1.

$$Flux_{in} \approx -\sum VCD(S_i) \cdot \omega_i \cdot \cos \beta_i \cdot \Delta S_i, \quad \beta_i > 90^\circ \quad (1)$$

$$Flux_{out} \approx \sum VCD(S_i) \cdot \omega_i \cdot \cos \beta_i \cdot \Delta S_i, \quad \beta_i < 90^\circ \quad (2)$$

$$Flux_{net} = Flux_{in} - Flux_{out} \quad (3)$$

Here, VCD(S_i) is the NO₂ column along segment S_i, and β_i denotes the angle between the wind vector and the boundary normal vector **n**. β_i > 90° identifies boundary external influx (import), while β_i < 90° denotes internal efflux (export). ΔS_i is the boundary integration step. The net flux is calculated as total influx minus efflux, with positive (negative) values indicating net import (export).

S1.2 Regression model for trend analysis

In this study, a unified regression framework was applied to the long-term time series of tropospheric NO₂ column concentrations, surface concentrations, and transport fluxes, in order to capture their secular trends and seasonal cycle characteristics (Yin et al., 2022). The model integrates a linear term with third-order Fourier components, as formulated in Equation (4).

$$Y(t) = A_0 + A_1 t + A_2 \cos\left(\frac{2\pi t}{12}\right) + A_3 \sin\left(\frac{2\pi t}{12}\right) + A_4 \cos\left(\frac{4\pi t}{12}\right) + A_5 \sin\left(\frac{4\pi t}{12}\right) \quad (4)$$

The modeled variable $Y(t)$ was decomposed into a linear component representing long-term evolution and harmonic terms describing seasonal variability. The parameters A_0 and A_1 denote the intercept and linear growth rate, respectively, while A_2 - A_5 account for the periodic seasonal cycle. The interannual rate of change was estimated as A_1/A_0 , with associated uncertainties expressed as mean ± standard deviation.

S1.3 Wind field correction and uncertainty

To reduce vertical non-uniformity errors, we apply a NO₂-weighted vertical wind correction, with wind-field uncertainty quantified using Equations (5)-(7).

$$\tau_j(z_j) = \frac{x_j(z_j) \cdot Airmass_j(z_j)}{\sum_i x_j(z_j) \cdot Airmass_j(z_j)} \quad (5)$$

$$\omega_{j,avg} = \sum_i \omega(z_i) \cdot \tau_j(z_j), \quad \theta_{j,avg} = \sum_i \theta(z_i) \cdot \tau_j(z_j) \quad (6)$$

$$\sigma_{\omega z} = \sqrt{\sum_i [\tau_j(\omega_{j,avg} - \omega(z_i))]^2}, \quad \sigma_{\theta z} = \sqrt{\sum_i [\tau_j(\theta_{j,avg} - \theta(z_i))]^2} \quad (7)$$

Where $\omega(z_i)$, $x_j(z_j)$, and $Airmass_j(z_j)$ represent the wind field vector, NO₂ VMR concentration, and air mass at height z_j along the j segment of the integration path, respectively. $\omega_{j,avg}$ is the weighting averaged wind field along the j segment of the integration path, and $\sigma_{\omega z}$ and $\sigma_{\theta z}$ are the uncertainties in the corrected wind field and wind direction, respectively.

Section S2. Surface NO₂ over TP

Using ground-based observations from the CNEMC network during 2015-2024 at major cities across TP, we systematically analyzed the spatiotemporal characteristics of surface NO₂

concentrations. The corresponding results are presented in Fig. S14. The dataset covers most major urban areas across the TP, and we selected 12 populous cities for analysis, with the data subjected to rigorous missing-value filtering and seasonal anomaly screening to ensure temporal continuity and representativeness. Results show that surface NO₂ concentrations across Tibet generally exhibit a distinct seasonal pattern of higher levels in winter and lower levels in summer, with winter monthly means being typically a few times higher than those in summer. This seasonal contrast is particularly pronounced in densely populated cities such as Lhasa and Qamdo. In contrast, the long-term tendencies of surface and column NO₂ diverge: satellite-derived column NO₂ shows an increasing trend, while surface measurements decline, highlighting a discrepancy between columnar loading and near-surface concentrations.

Trend analysis indicates that, except for Shannan where the data were unstable and thus excluded, the remaining cities all exhibit significant negative trends ($p < 0.01$), with the most pronounced decline observed in Tibet. Qamdo shows the strongest annual decrease of $-6.28 \pm 0.66 \text{ \% yr}^{-1}$, while the decline in Lhasa is relatively moderate at $-4.17 \pm 0.55 \text{ \% yr}^{-1}$. Conversely, Aba Prefecture shows a slight upward trend of $-1.87 \pm 0.78 \text{ \% yr}^{-1}$. Among all cities, Lhasa exhibits the largest seasonal amplitude, with winter surface NO₂ concentrations more than twice those in summer; in contrast, Nyingchi and Guoluo show the smallest seasonal difference. The onset of the decreasing trends is not fully synchronized across cities: for example, Qamdo experienced a sustained decline after 2015, whereas Shannan did not show a clear negative trend until after 2022. These discrepancies are likely related to differences in the timing of local energy structure adjustments and the implementation of vehicle emission control policies (Wu et al., 2017; Liu et al., 2023; Abdelwahab et al., 2024).

Section S3. Spatiotemporal patterns of NO₂ variability

Based on the analysis of OMI satellite data from 2005 to 2024, the tropospheric NO₂ vertical column density (VCD) over Tibet and its surrounding regions exhibits pronounced spatial heterogeneity, with the spatial patterns depicted in Fig. 1 and Fig. S15. The 20-year mean NO₂ VCD over the TP is $6.47 \times 10^{15} \text{ molec cm}^{-2}$, significantly lower than that over major South Asian countries such as India and Nepal, reflecting the relatively clean atmospheric background of the TP. However, intra-TP gradients are also evident. The 20-year mean NO₂ VCD values for the seven prefecture-level cities within Tibet are as follows: Ngari, $5.73 \times 10^{15} \text{ molec cm}^{-2}$; Lhasa, $7.23 \times 10^{15} \text{ molec cm}^{-2}$; Nagqu, $5.81 \times 10^{15} \text{ molec cm}^{-2}$; Nyingchi, $7.94 \times 10^{15} \text{ molec cm}^{-2}$; Shigatse, $6.53 \times 10^{15} \text{ molec cm}^{-2}$; Shannan, $1.03 \times 10^{15} \text{ molec cm}^{-2}$; and Qamdo, $5.57 \times 10^{15} \text{ molec cm}^{-2}$. Among these, relatively high NO₂ VCDs are observed in southeastern regions such as Nyingchi and Lhasa, which may be influenced by pollution accumulation or regional transport processes facilitated by topographic channels. Previous studies have confirmed that pollutants such as CO and HCHO in this region are strongly affected by transboundary transport from northeastern India (e.g., Assam) (Sun et al., 2021; Lin et al., 2024; Xu et al., 2024), and NO₂, as a chemically related species, is likely to be transported together, exhibiting similar spatial patterns.

The temporal trend analysis indicates a coherent seasonal cycle of NO₂ across the TP. Most regions display a characteristic pattern with minima in winter and maxima in summer, controlled by the intensity of photochemical reactions and the development of the planetary boundary layer. A linear regression applied to deseasonalized monthly NO₂ VCD series yields correlation coefficients (R) ranging from 0.65 to 0.94, suggesting that the trend signal is both statistically

1 significant and robust. All subregions exhibit increasing NO₂ concentrations over 2005-2024. The
2 TP-wide mean growth rate is $2.79 \pm 0.14 \text{ \% yr}^{-1}$, with regional trends spanning from $1.47 \pm$
3 $0.16 \text{ \% yr}^{-1}$ in Shannan to $3.41 \pm 0.21 \text{ \% yr}^{-1}$ in Qamdo, highlighting a clear east–west contrast in
4 the pace of enhancement.

5 However, OMI detected anomalously elevated NO₂ values during the summers of 2023 and
6 2024. To assess whether these deviations reflect actual atmospheric changes or potential
7 sensor-related issues, we compared the corresponding OMI pixels with matched TROPOMI
8 observations, with the relevant results shown in Fig. S16. The OMI–TROPOMI difference maps
9 indicate that, relative to 2022, the proportion of pixels exhibiting positive biases increased
10 substantially in 2023 and 2024, suggesting that the summertime NO₂ enhancement is unlikely to
11 be driven entirely by physical processes and is more likely to be partly influenced by the gradual
12 decline in OMI performance. This phenomenon is consistent in direction with recent assessments
13 reporting possible sensor aging. Although OMI has been in service for nearly two decades and has
14 experienced a certain degree of performance degradation, its long-term continuous record remains
15 irreplaceable for investigating regional-scale trends and multi-decadal atmospheric changes.
16 Moreover, a discrepancy exists between surface and column concentrations, likely reflecting the
17 differing contributions of boundary-layer processes, regional transport, and emission structures
18 (Zhang et al., 2025a; Zhang et al., 2025b). It is worth noting that previous studies have reported a
19 rapid increase in local NO_x emissions in high-altitude cities in recent years, which has also, to
20 some extent, contributed to the divergent changes between surface and column NO₂
21 concentrations (Xu et al., 2025). CO shows a similar pattern, but with a smaller divergence (see
22 Figs. S17-S18).

Figures

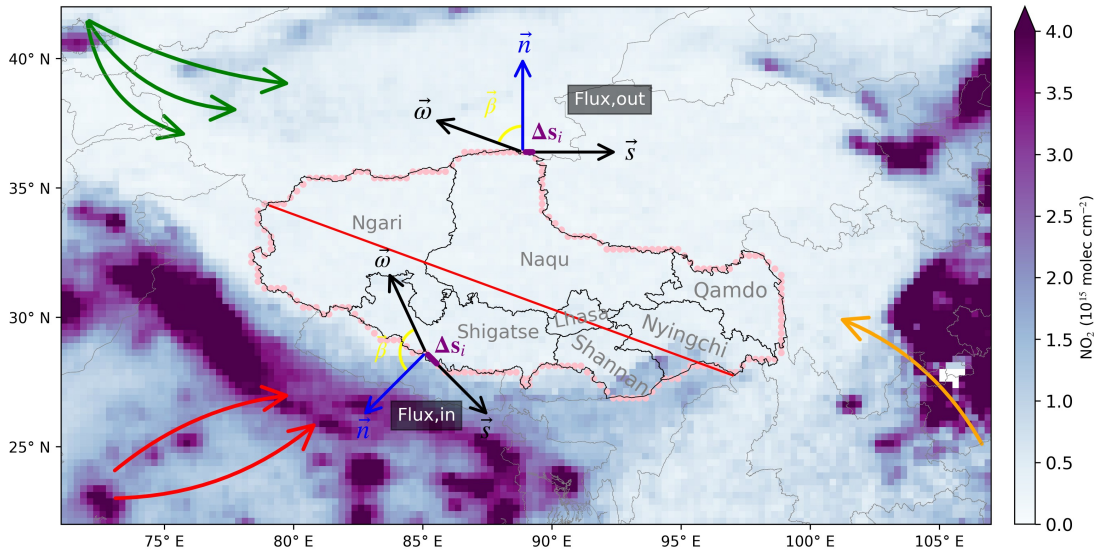


Fig. S1. Illustration of the closed-loop integral method for NO_2 flux calculation. Pink points indicate the closed loop. The black arrow ω denotes the wind field vector, s is the integration step vector, and the blue arrow n shows the boundary normal vector. β is the angle between ω and n , while Δs represents the integration step length. Green arrows indicate the westerly circulation over the plateau, red arrows denote the South Asian monsoon, and orange arrows show the East Asian monsoon.

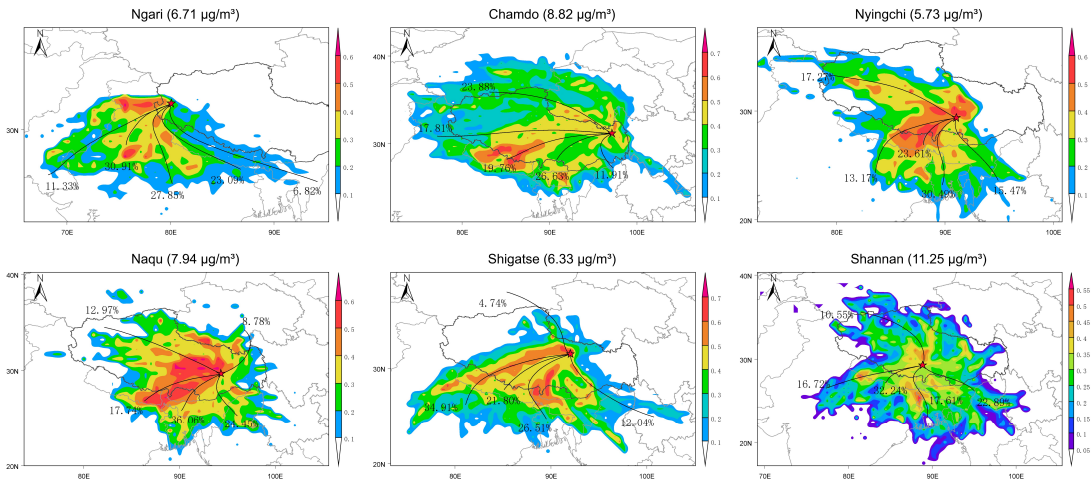


Fig. S2. PSCF-derived potential source regions and clustered back trajectories for TP, categorized into the monsoon periods.

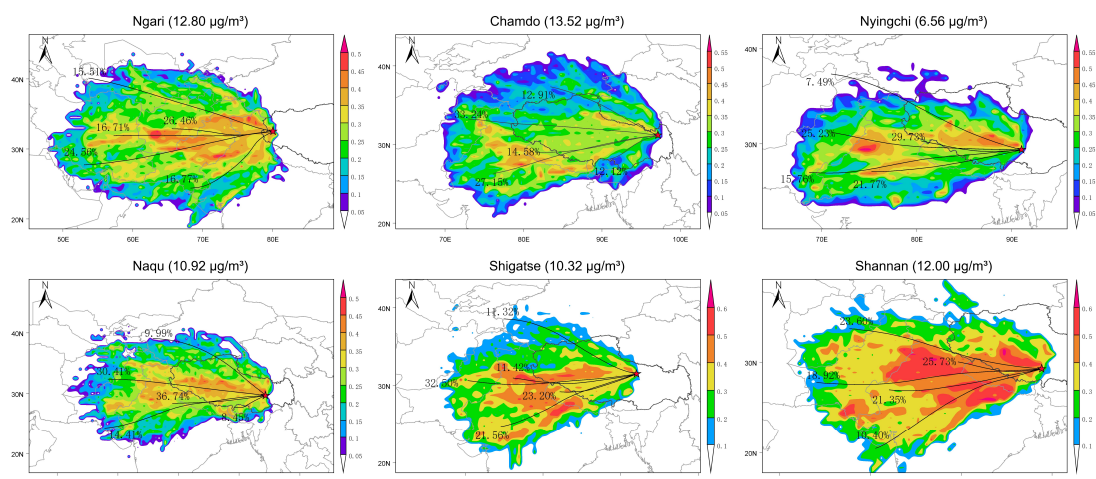


Fig. S3. PSCF-derived potential source regions and clustered back trajectories for TP, categorized into the westerly periods.

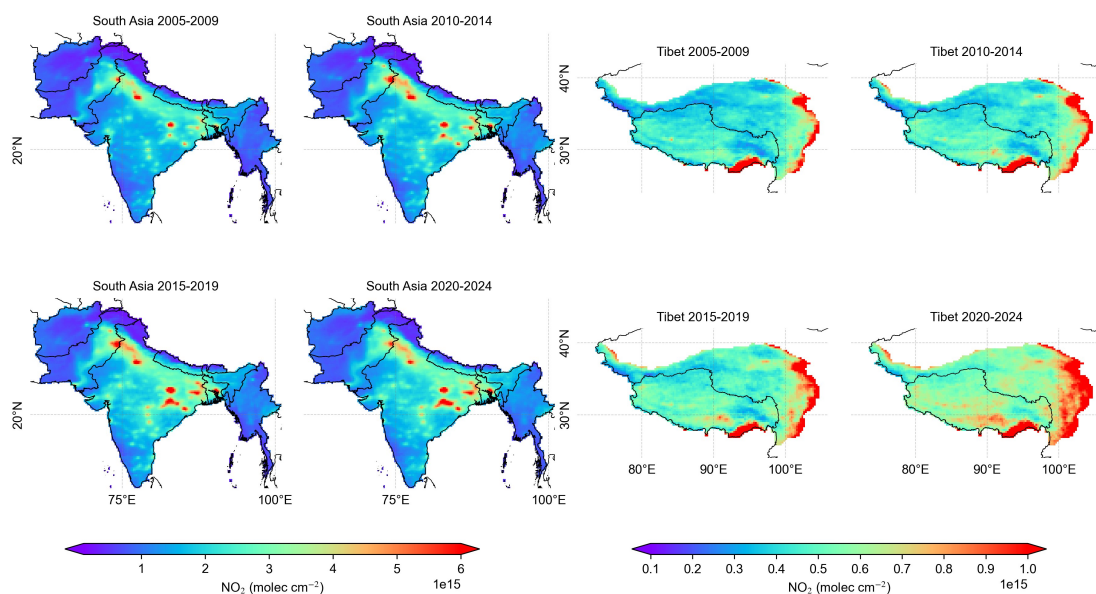
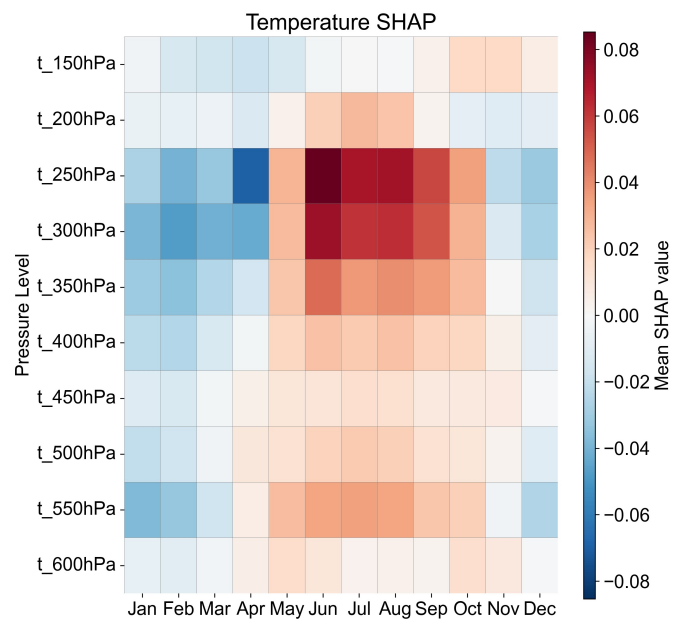


Fig. S4. Five-year grouped changes in NO₂ column concentrations over South Asia and the TP from 2005 to 2024.



Fig. S5. Annual NO_x emissions by sector (energy, transport, agriculture, and industry) across TP, derived from EDGAR and aggregated over all pixels within administrative boundaries.

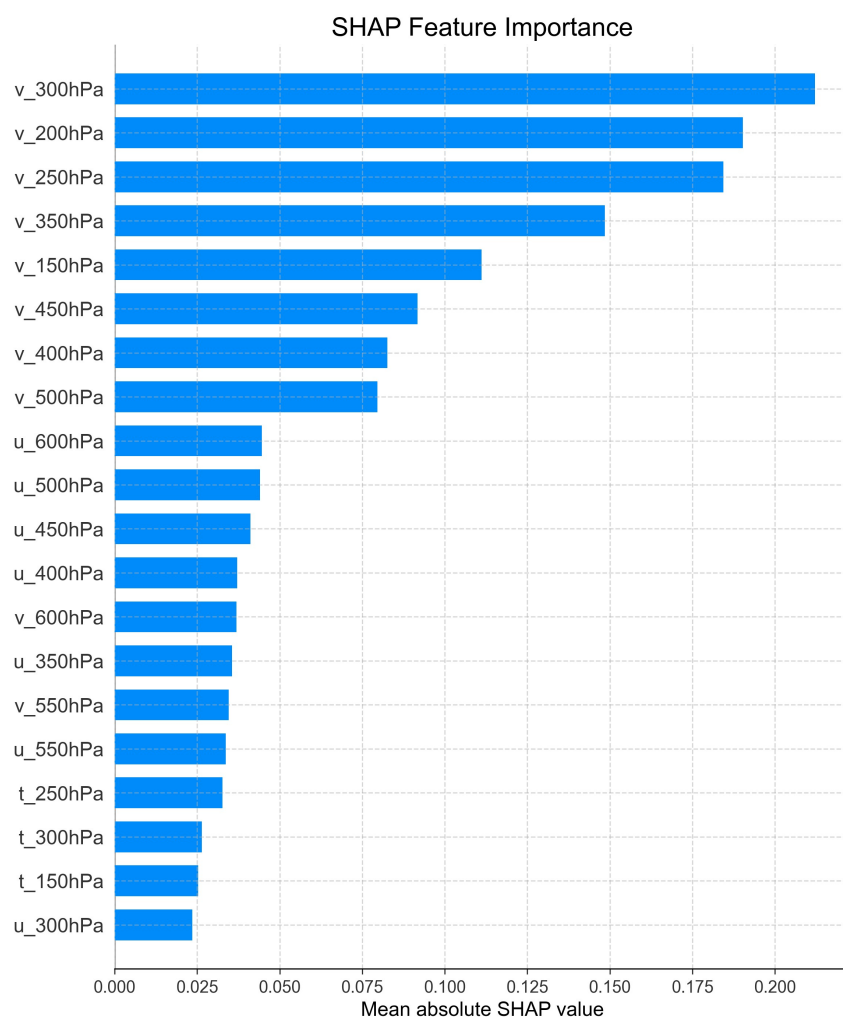
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3 **Fig. S6.** HAP-based contributions of temperature (150-600 hPa) to NO₂ transport flux variations across the
 4 southwestern TP boundary, with red and blue indicating positive and negative contributions, respectively.

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2 **Fig. S7.** Random forest–derived importance of U- and V-wind and temperature in explaining NO₂ transport flux
3 variability across the southwestern boundary of the TP.

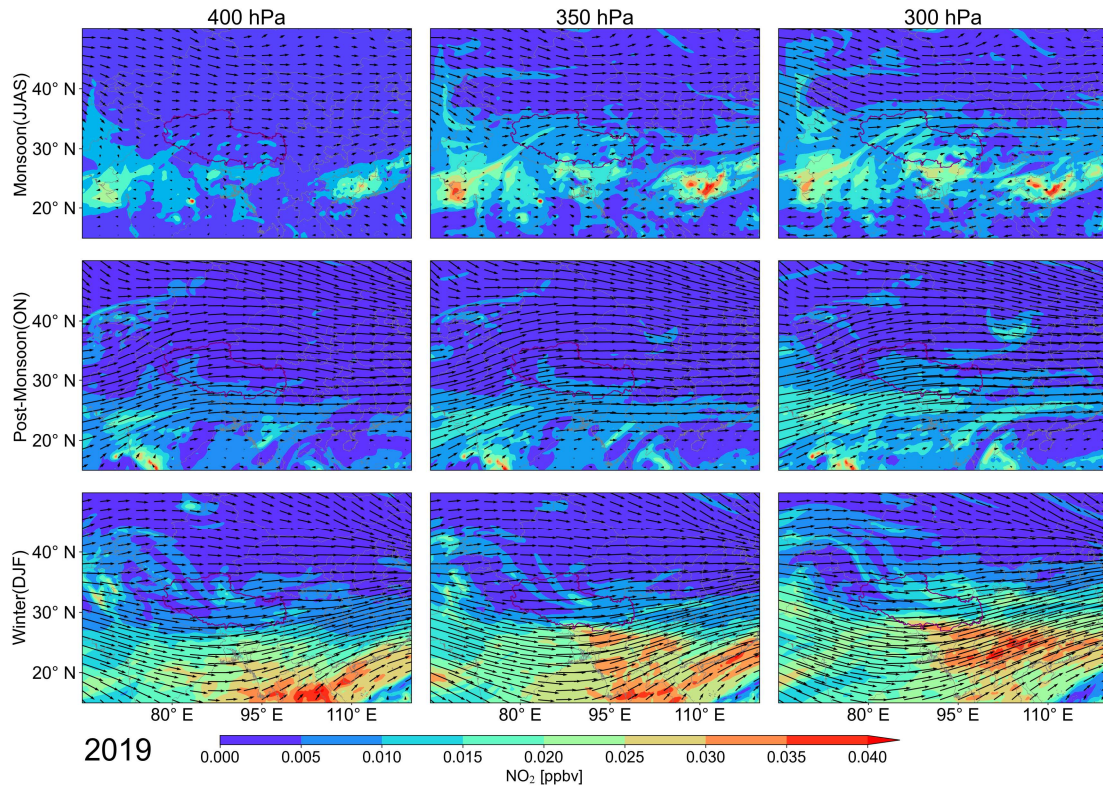


Fig. S8. NO₂ concentration and wind fields over the Tibetan Plateau at 400, 350, and 300 hPa during the monsoon, late-monsoon, and winter seasons of 2019. NO₂ concentrations are from GEOS-CF and wind fields from ERA5.

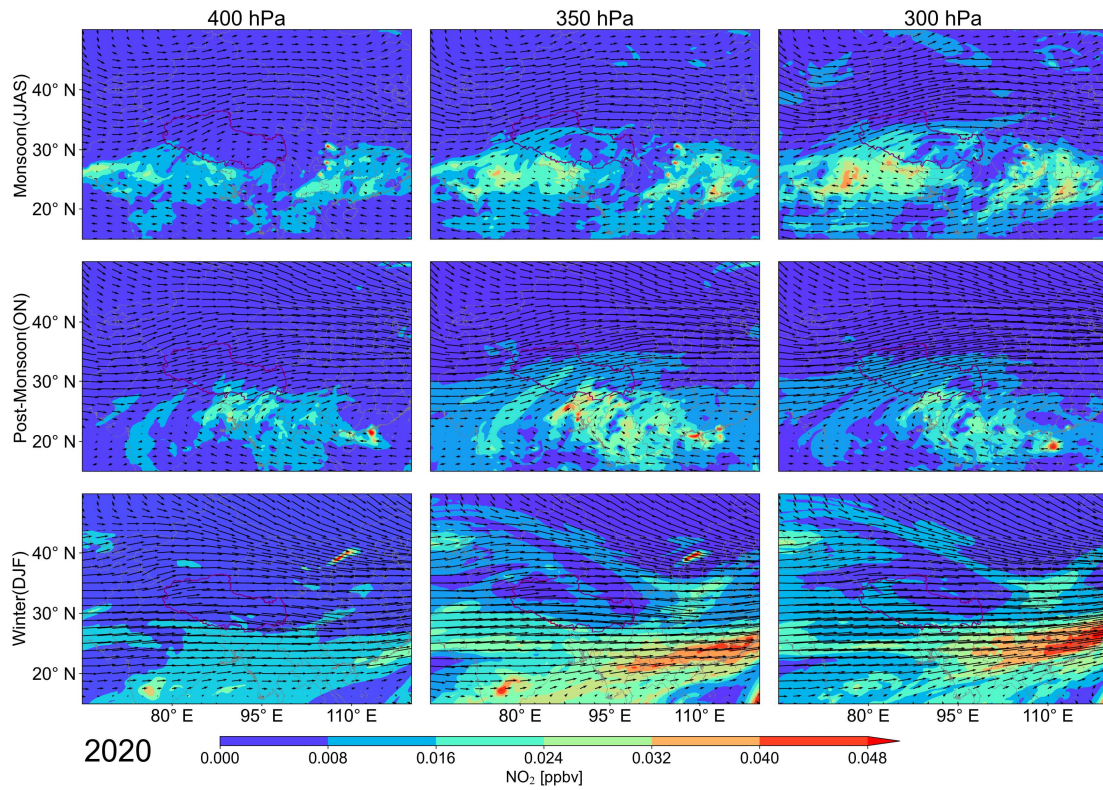


Fig. S9. NO₂ concentration and wind fields over the Tibetan Plateau at 400, 350, and 300 hPa during the monsoon, late-monsoon, and winter seasons of 2020. NO₂ concentrations are from GEOS-CF and wind fields from ERA5.

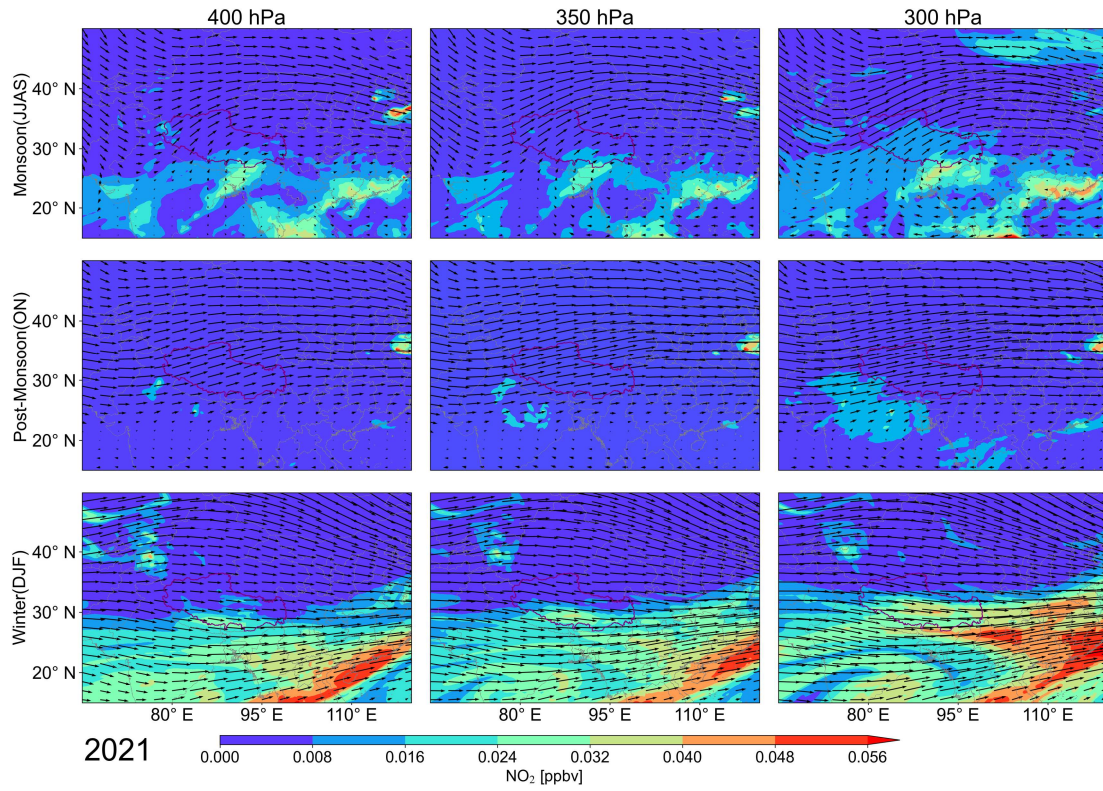


Fig. S10. NO₂ concentration and wind fields over the Tibetan Plateau at 400, 350, and 300 hPa during the monsoon, late-monsoon, and winter seasons of 2021. NO₂ concentrations are from GEOS-CF and wind fields from ERA5.

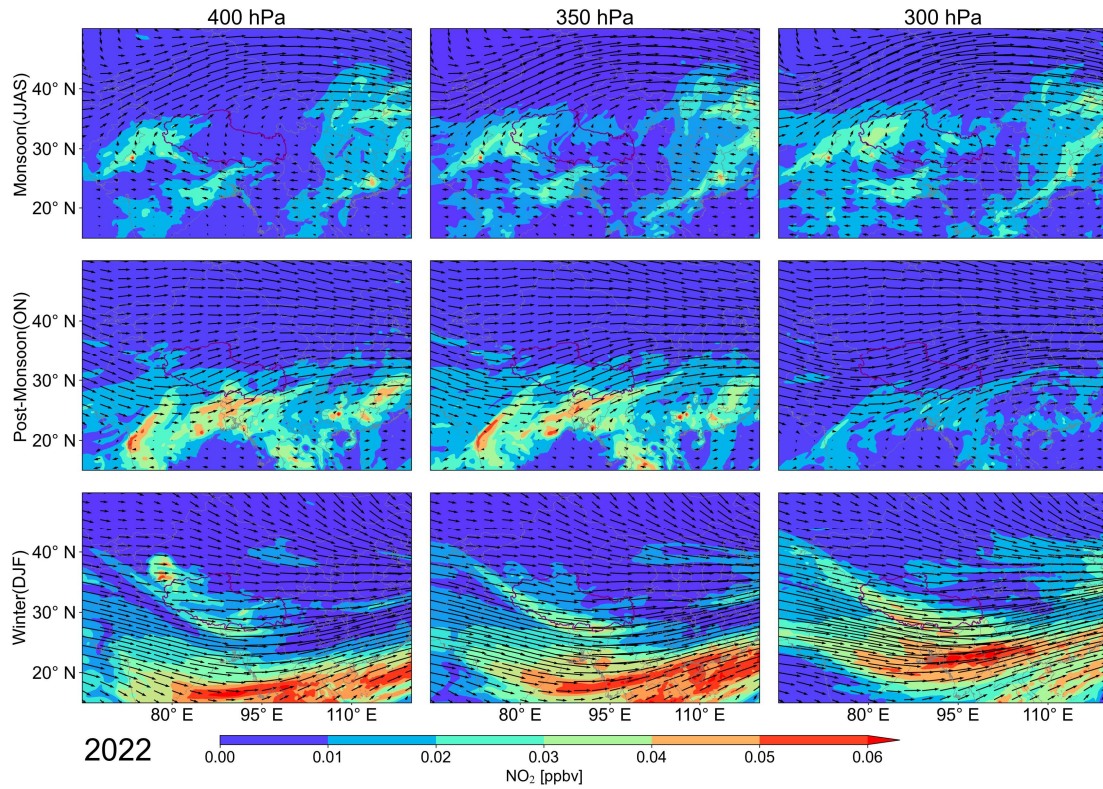


Fig. S11. NO₂ concentration and wind fields over the Tibetan Plateau at 400, 350, and 300 hPa during the monsoon, late-monsoon, and winter seasons of 2022. NO₂ concentrations are from GEOS-CF and wind fields from ERA5.

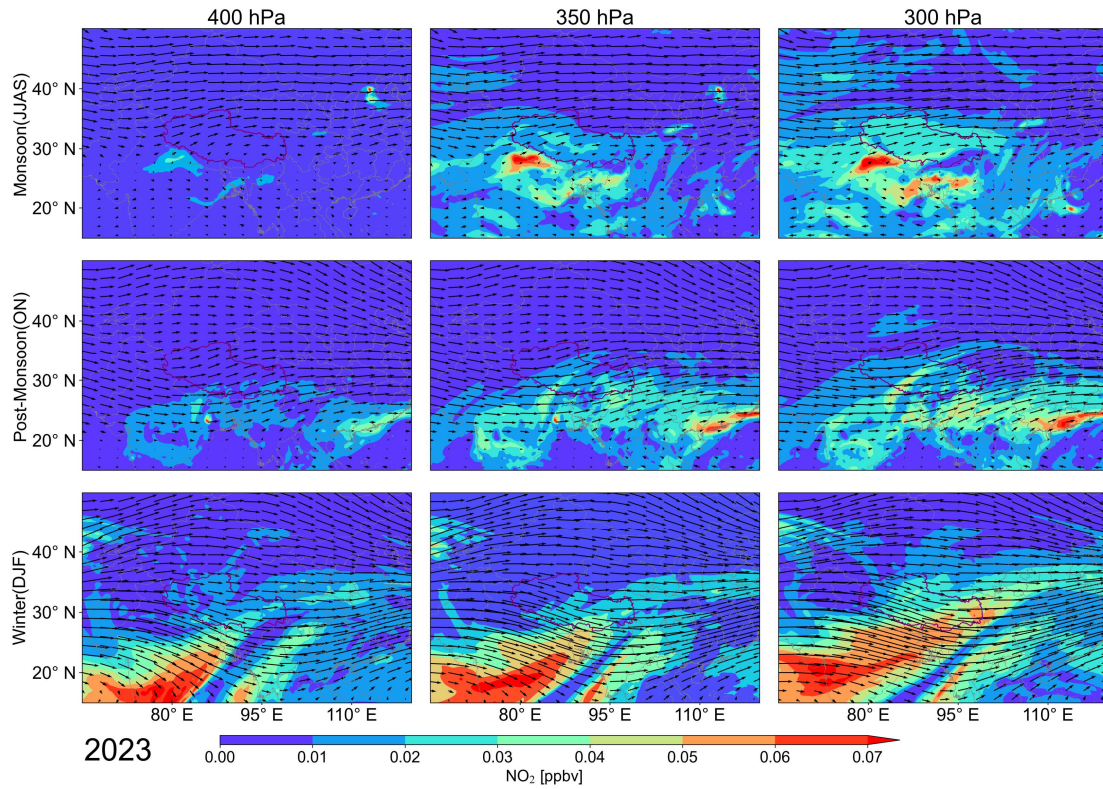


Fig. S12. NO₂ concentration and wind fields over the Tibetan Plateau at 400, 350, and 300 hPa during the monsoon, late-monsoon, and winter seasons of 2023. NO₂ concentrations are from GEOS-CF and wind fields from ERA5.

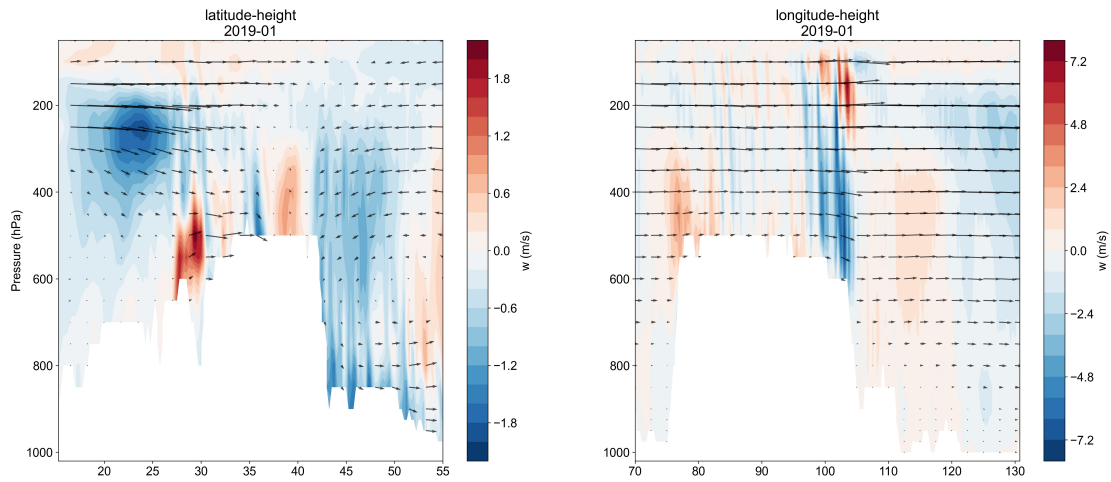


Fig. S13. Vertical Mixing over the Plateau Derived from ERA5 Reanalysis.

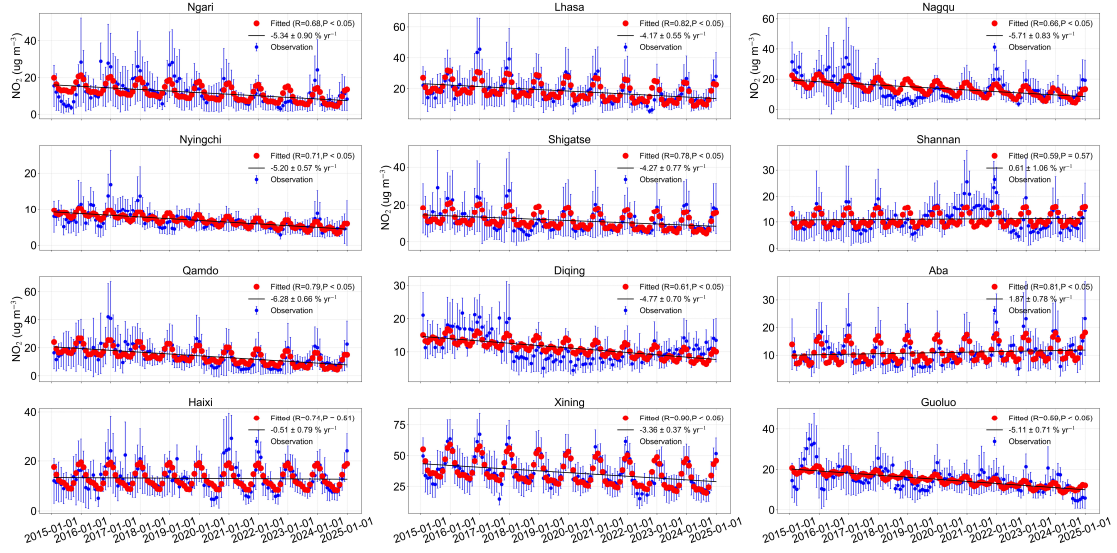


Fig. S14. Temporal evolution of NO_2 concentrations observed by CNEMC from 2015 to 2024. Monthly means (blue) with $\pm 1\sigma$ variability are shown alongside the fitted seasonal (red) and long-term (black) components.

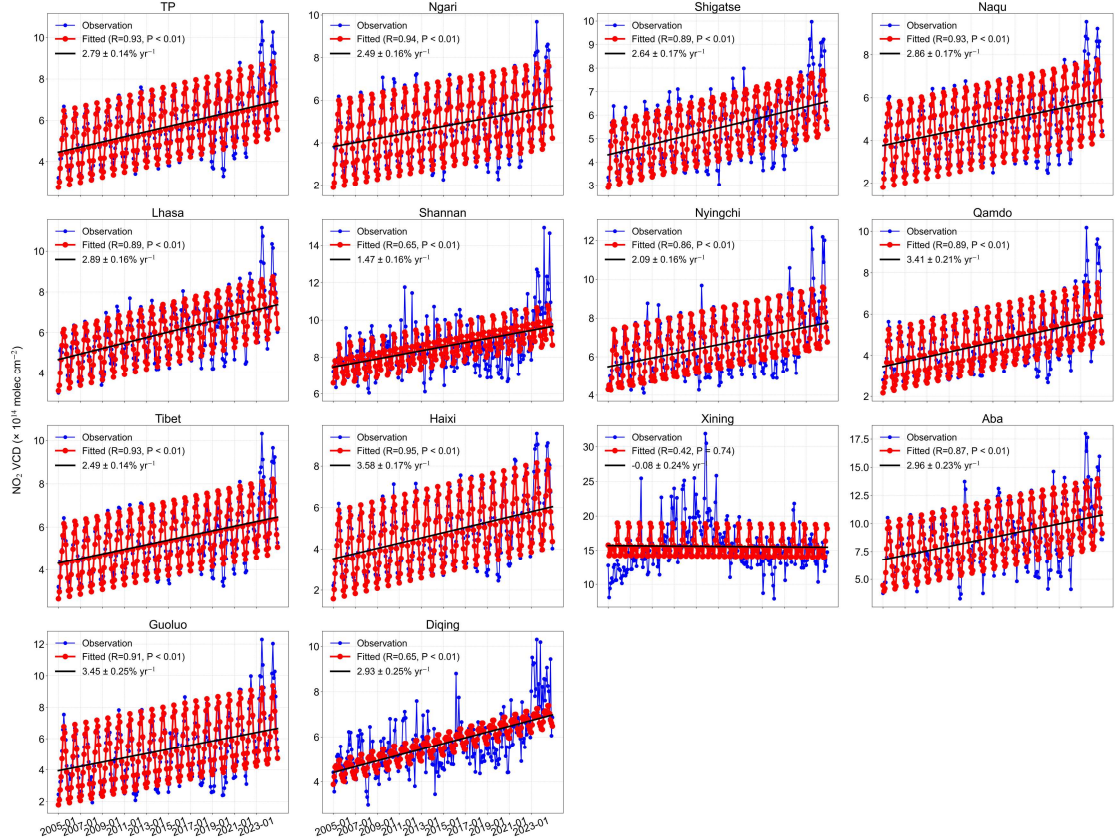
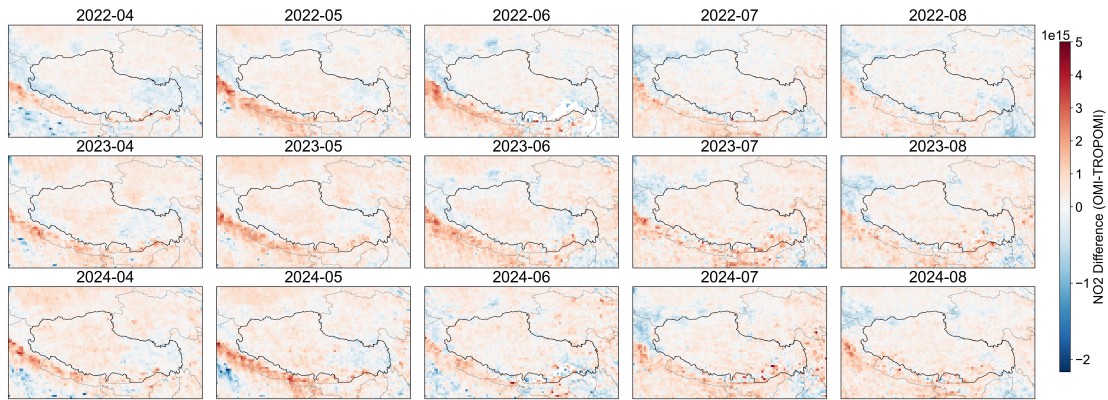


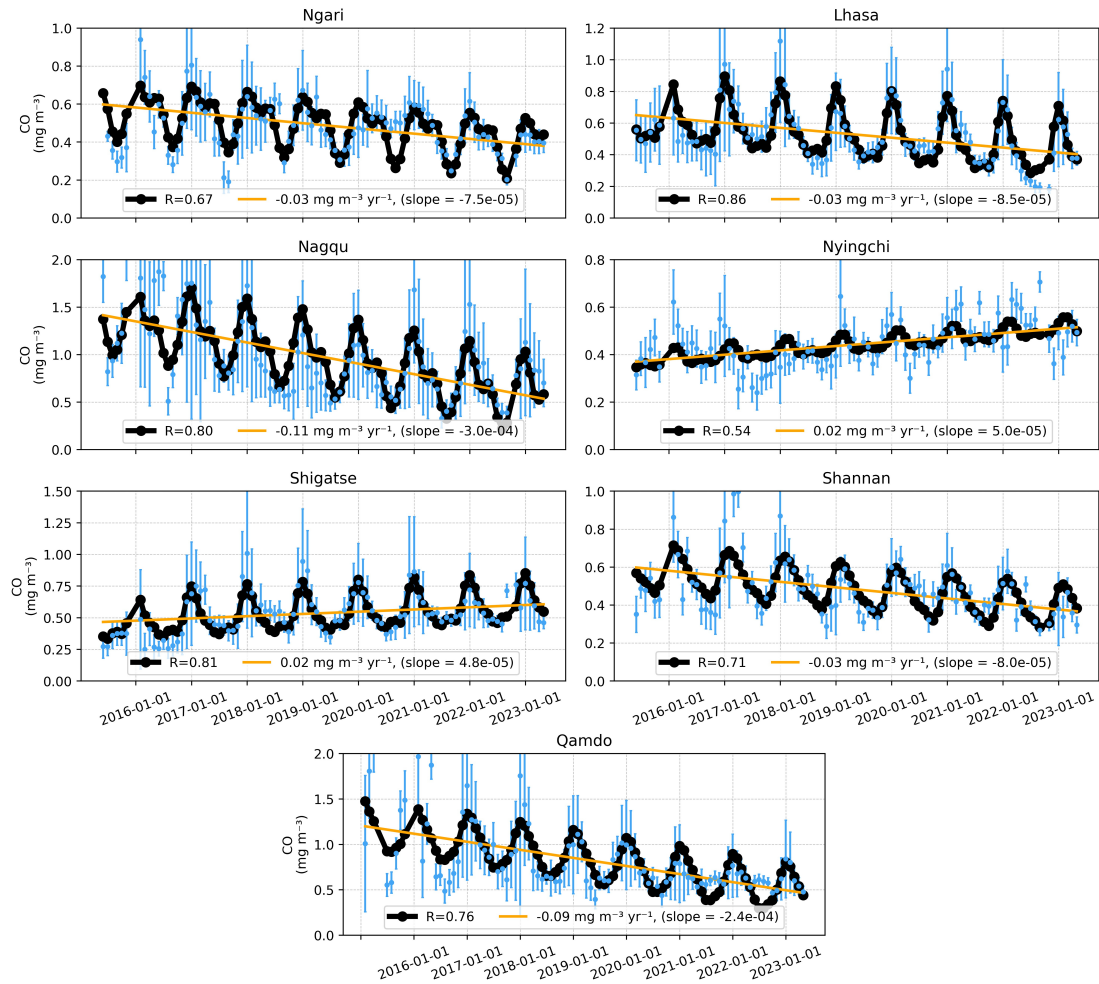
Fig. S15. Long-term NO_2 VCD (2005–2024) over the TP and selected prefecture-level cities, averaged across all pixels within each administrative boundary. Blue dots show monthly mean concentrations, red curves indicate fitted seasonal cycles, and black curves represent interannual trends.

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Fig. S16. Pixel-wise differences between OMI and TROPOMI during April–September, 2022–2024



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Fig. S17. Inter-annual variations of CO observed by CNEMC from 2015 to 2024. Blue dots represent the monthly mean CO concentration, with vertical error bars indicating the 1 σ standard deviation. The seasonal trend (black line) and inter-annual trend (orange line) are fitted using a seasonal cycle model.

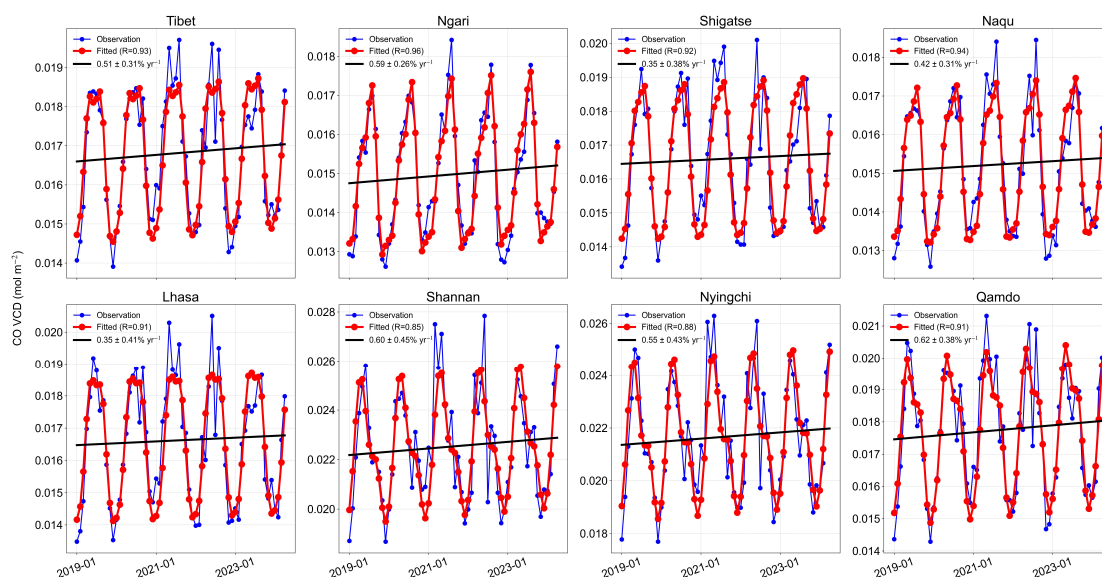


Fig. S18. Long-term fitted CO column concentrations (2019-2024) across the TP and seven representative prefecture-level cities, averaged over all pixels within their administrative boundaries.

Tables

Table S1. Uncertainties in mean wind speed and wind direction after wind-field correction for NO₂ transport flux.

Year	Averaged Wind Speed Uncertainty (m/s)				Averaged Wind Direction and Its Uncertainty (°)			
	MAM	JJA	SON	DJF	MAM	JJA	SON	DJF
2005	3.58	2.13	3.15	4.85	21.01	36.06	14.48	16.01
2006	3.46	1.95	2.77	4.74	16.77	39.58	14.57	15.79
2007	3.23	2.14	3.00	4.64	21.62	40.00	14.47	16.23
2008	3.50	2.10	3.27	5.03	19.84	41.35	13.92	15.59
2009	3.81	2.13	3.32	4.86	23.96	31.27	13.19	15.20
2010	3.71	2.02	3.32	4.59	16.48	38.61	15.54	16.16
2011	3.54	1.99	2.99	5.24	19.80	39.56	14.62	15.90
2012	3.88	2.16	3.26	5.37	20.87	38.70	13.74	16.23
2013	3.34	1.98	3.02	4.85	22.15	39.98	16.04	16.45
2014	3.66	2.08	3.08	5.37	19.22	36.77	12.68	16.41
2015	3.46	2.08	3.10	4.13	16.04	38.24	12.66	16.57
2016	3.38	2.04	3.25	4.82	13.74	33.90	14.58	15.54
2017	3.71	1.99	3.15	4.86	20.74	39.77	12.70	15.95
2018	3.11	1.99	3.11	4.96	15.86	42.05	12.76	15.31
2019	3.89	2.19	3.26	4.15	17.50	35.16	15.36	16.62
2020	3.32	2.19	3.35	4.69	20.01	35.58	12.77	17.45
2021	3.76	2.18	3.30	4.75	18.03	35.48	14.20	15.55
2022	3.32	1.84	3.08	4.77	18.80	35.98	14.48	17.23
2023	3.38	2.03	2.73	4.70	14.77	36.49	12.39	15.48
2024	3.40	2.28	2.89	4.75	20.16	38.30	15.21	15.60

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