



1                   **Quantifying transboundary transport flux of CO over the**  
2                   **Tibetan Plateau: variabilities and drivers**

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22                  **Abstract:**

23                  The Tibetan Plateau significantly impacts regional and global climate systems due to its  
24                  unique geographical location and complex environmental processes. This study investigates the  
25                  variability and driving force of transboundary transport flux of carbon monoxide (CO) over the  
26                  Tibetan Plateau from May 2018 to April 2024. The transport CO fluxes were calculated with a  
27                  closed-loop integral method using the TROPOMI, ERA5, and GEOS-CF data products. The  
28                  results show that the external influx and internal efflux of CO over the Tibetan Plateau in each  
29                  year are relatively close and have similar seasonal characteristics. High levels of CO flux occur in  
30                  late autumn to winter, and low levels occur in summer. In most cases, CO flux maximizes in  
31                  November, December or January, and minimizes in July or August. The month to month  
32                  variability during late autumn to winter is greater than that in summer. The Tibetan Plateau has  
33                  experienced an increase of  $0.65 \text{ t s}^{-1} \text{ yr}^{-1}$  in external influx, while the internal efflux has slightly  
34                  decreased by  $-0.39 \text{ t s}^{-1} \text{ yr}^{-1}$ . The magnitude of the increase in external influx in the southwestern  
35                  segment is greater than in the northeastern segment. Conversely, the magnitude of the decrease in  
36                  internal efflux in the northeastern segment is greater than in the southwestern segment. The source  
37                  attribution results reveal that the external input of CO into the Tibetan Plateau mainly comes from



1 South Asia. The increase in external influx of CO in recent years over the Tibetan Plateau are  
2 potentially linked to the rapid rise in CO concentrations from South Asia.

### 3 **1 Introduction**

4 The Tibetan Plateau, often referred to as the "Third Pole of the Earth", is characterized by its  
5 extensive snow, glaciers, permafrost, and seasonal frozen ground. Due to the complex interactions  
6 among atmospheric, cryospheric, hydrological, geological, and environmental processes, this  
7 region profoundly impacts global climate and water cycle systems. The Tibetan Plateau plays a  
8 crucial role in the global climate system and serves as a critical indicator of regional and global  
9 climate change (Qian et al., 2011; Li et al., 2017; Bibi et al., 2018; Gao et al., 2019). This plateau  
10 and its surrounding regions experience various atmospheric circulation patterns, including the  
11 Indian summer monsoon, winter westerlies, and the East Asian monsoon (Yao et al., 2012; Chen  
12 et al., 2020). These circulation patterns are essential in shaping regional climate and pollutant  
13 transport. In particular, the southwestern Tibetan Plateau is influenced by the South Asian  
14 monsoon. During summer, the intensified monsoon transports warm, moist air masses and  
15 pollutants, such as aerosols and particulate matter, from the South Asian subcontinent to the  
16 Tibetan Plateau. Owing to the uplift effect of the plateau's topography, these airflows descend into  
17 the southern and southwestern regions after crossing the Himalayas, delivering rainfall and  
18 accumulating pollutants. Conversely, the northeastern Tibetan Plateau are impacted by the East  
19 Asian monsoon. In winter, cold air from Central and Northern Asia flows into the plateau,  
20 carrying pollutants from these areas. In summer, the East Asian monsoon brings moist air and  
21 pollutants from the East Asian coast into the eastern Tibetan Plateau, affecting local air quality  
22 (Ramanathan et al., 2005; Xu et al., 2009; Kaspari et al., 2011; Qian et al., 2011; Lüthi et al., 2015;  
23 Cong et al., 2015; Zhang et al., 2015; Wang et al., 2019; Wu et al., 2020; Li et al., 2021; Sun et al.,  
24 2021). Surrounding the Tibetan Plateau, densely populated Eurasian countries are experiencing  
25 rapid economic development, leading to increased emissions of air pollutants. Consequently, these  
26 regions have become some of the most polluted areas globally, and the pollutants can be  
27 transported over long distances to the Tibetan Plateau via the Asian monsoon and westerly  
28 circulation (Lawrence and Lelieveld, 2010). In addition to the rising industrial and agricultural  
29 emissions within the Tibetan Plateau, these external pollutants can also significantly impact its  
30 ecosystem and climate (Ji et al., 2015; Sun et al., 2021; Yin et al., 2022).

31 Carbon monoxide (CO) is one of the most significant atmospheric pollutants, primarily  
32 resulting from incomplete combustion of fossil fuels, biomass burning, and the oxidation of  
33 methane and non-methane hydrocarbons. This pollutant can indirectly exacerbate global warming  
34 by participating in the formation and reactions of other greenhouse gases in the atmosphere. CO is  
35 predominantly removed from the atmosphere through reactions with hydroxyl radicals (OH)  
36 (Holloway et al., 2000; Heald et al., 2003; Luo et al., 2013; Martínez-Alonso et al., 2020). With a  
37 lifetime ranging from several weeks to months, CO can persist in the atmosphere for extended  
38 periods, undergoing both horizontal and vertical transport. Consequently, CO is frequently  
39 employed as a tracer for studying pollutant transport dynamics (Holloway et al., 2000;  
40 Gloudemans et al., 2006; Jeong and Hong, 2021). Given its unique chemical properties and  
41 significant climatic effects, studying CO flux, variability, and driving factors offers valuable  
42 insights into the atmospheric conditions over the Tibetan Plateau. Furthermore, CO sources may



1 vary across different regions, understanding their contributions to the variability of CO flux over  
2 the Tibetan Plateau is essential for developing effective management strategies.

3 To investigate transboundary transport flux of CO over the Tibetan Plateau, this study  
4 employs a comprehensive array of data products, including TROPOMI, ERA5, and GEOS-CF. By  
5 integrating a closed-loop integral method with a regression model, we elaborate a top-down  
6 approach based on satellite remote sensing to estimate CO flux across the Tibetan Plateau. This  
7 quantitative analysis encompasses transport flux of CO along the Tibetan Plateau boundary  
8 (hereafter the closed-loop flux), spanning from the surface at 1000 hPa to the stratosphere at 50  
9 hPa, over a nearly six-year time series from May 2018 to April 2024. Our analysis emphasizes  
10 seasonal and inter-annual variabilities of the closed-loop CO flux. We further split the closed-loop  
11 CO flux into sub-fluxes arising from southwestern and northeastern segments, enabling us to  
12 quantify the impacts of surrounding areas on the Tibetan Plateau. This study aims to elucidate the  
13 spatiotemporal variabilities and the driving forces of the CO transport flux over the Tibetan  
14 Plateau, thereby providing scientific evidence for a deeper understanding of the Tibetan Plateau's  
15 role in global climate and environmental dynamics.

16 In the subsequent section, we present an overview of the Tibetan Plateau territory, providing  
17 a concise description of the dataset utilized, along with the methodologies employed for the  
18 closed-loop integral calculation and trend regression model. The third section delves into the  
19 spatiotemporal dynamics of the CO total column and transport flux over the Tibetan Plateau,  
20 highlighting the trend fitting outcomes. The fourth section elucidates the driving forces behind the  
21 variability of transboundary transport CO flux. The study culminates in the fifth section with a  
22 synthesis of our findings and conclusions.

## 23 **2 Methodology and dataset**

24 This section introduces geographical description of the Tibetan Plateau, the closed-loop  
25 integral method used to derive external influx and internal efflux of CO, the regression model used  
26 for trend analysis and the dataset involved.

### 27 **2.1 Geographical description of the Tibetan Plateau**

28 We have conducted an analysis of both the external influx and internal efflux of CO across  
29 the Tibetan Plateau. The external influx refers to the quantity of CO that is transported from  
30 regions outside the Tibetan Plateau into its boundaries. Conversely, the internal efflux denotes the  
31 amount of CO that is generated within the Tibetan Plateau and dispersed to areas beyond its  
32 confines. As shown in Figure 1, we bifurcated the Tibetan Plateau's geographical boundary into  
33 two segments. The first segment encompasses the southwestern Tibetan Plateau, which is  
34 significantly influenced by the South Asian monsoon. The substantial topographic rise in this area  
35 intensifies the monsoon's impact (Huang et al., 2023). The second segment is situated in the  
36 northeastern Tibetan Plateau, where the westerlies and the East Asian monsoon play a pivotal role.  
37 There is a marked geographical disparity between these two segments.

38 For comparison, we annotate the adjacent regions around the Tibetan Plateau. We categorize  
39 these regions into three broad zones: the western, central, and eastern zones. The western zone  
40 predominantly comprises Himachal Pradesh, Uttarakhand, and Ladakh. The central zone is  
41 characterized by Nepal, Sikkim, and Bhutan, while the eastern zone includes Assam, Sagaing, and  
42 Kachin State, among others. A visual representation of these regional demarcations is presented in



1 Figure 1.

## 2 **2.2 Dataset description**

3 We utilize a comprehensive dataset encompassing the total column of CO measured by the  
4 TROPOMI instrument, the vertical profile of CO from the GEOS-CF system, meteorological data  
5 extracted from the ERA5 reanalysis, and atmospheric air-mass simulated by the GEOS-Chem  
6 model.

7 TROPOMI is a push broom imaging spectrometer on the ESA Sentinel-5 platform, providing  
8 daily global coverage of CO total column at 13:30 local time (LT) (Veefkind et al., 2012;  
9 Landgraf et al., 2020). We used TROPOMI Level 2 CO and filtered the TROPOMI data according  
10 to the method of Landgraf et al. (2020), i.e., we removed all pixels with a TROPOMI quality mark  
11 below 0.5, leaving only data with no clouds or only low-altitude clouds. For convenience of  
12 calculation, we resampled the CO data product in space and time to match the spatiotemporal  
13 resolution of the meteorological field.

14 ERA5 is the fifth generation of atmospheric reanalysis dataset for global climate from  
15 ECMWF (European Centre for Medium-Range Weather Forecasts). It provides hourly global  
16 atmospheric, land surface and ocean wave estimates since 1950 and is produced by the Copernicus  
17 Climate Change Service (C3S) of ECMWF (Hersbach et al., 2020). We extracted the wind vectors  
18 that coincident with the Sentinel-5's overpass time with a vertical resolution of 50 hPa from 50  
19 hPa to 900 hPa and 25 hPa from 900 hPa to 1000 hPa.

20 Since measurement-based CO profile is not available, the vertical CO profile from the  
21 GEOS-CF system is used to correct vertically non-uniform distribution of CO concentration and  
22 wind field. The GEOS-CF system is a near-real-time high resolution ( $0.25^\circ \times 0.25^\circ$ ) global 3D  
23 coupled chemical and meteorological modeling system developed by NASA's Global Modeling  
24 and Assimilation Office (GMAO) (Keller et al., 2021). Since 2018, GEOS-CF has provided global  
25 CO vertical profiles at 23 pressure levels (from 1000 to 10 hPa) on an hourly basis.

26 The air-mass dataset used for calculating CO flux comes from the simulations of  
27 GEOS-Chem model version 12.2.1 (<https://doi.org/10.5281/zenodo.2580198>, International  
28 GEOS-Chem Community, 2019) (Long et al., 2015). The model is driven by assimilated  
29 meteorological data obtained from the Goddard Earth Observing System (GEOS) of NASA's  
30 Global Modeling and Assimilation Office (GMAO) (Bey et al., 2001). The GEOS-FP dataset has  
31 a horizontal resolution of  $0.25^\circ$  latitude  $\times$   $0.3125^\circ$  longitude and includes 47 vertical levels from  
32 the surface to 0.01 hPa. Surface meteorological variables and planetary boundary layer height  
33 (PBLH) are provided with a 1-hour interval. We used a nested grid version of GEOS-Chem with a  
34 horizontal resolution covering the East Asia region ( $70\text{--}140^\circ$  E,  $15\text{--}55^\circ$  N), with boundary  
35 conditions derived from a global simulation at a resolution of  $2^\circ$  latitude  $\times$   $2.5^\circ$  longitude (Lee  
36 and Park, 2022).

## 37 **2.3 The closed-loop integral method for CO flux calculation**

38 We use the closed-loop integral method from Shaiganfar et al. (2017) to calculate the CO  
39 flux along the Tibetan Plateau boundary. With this closed-loop integral method, the external  
40 influx ( $Flux_{in}$ ) and internal efflux ( $Flux_{out}$ ) of CO across the Tibetan Plateau can be  
41 calculated as equations (1) and (2), respectively. The calculation methodology is illustrated in  
42 Figure 1.



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

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

$$3 \quad Flux, net = Flux, in - Flux, out \quad (3)$$

4 Where  $VCD(S_i)$  represents CO total column locates at the path  $S_i$ , which represents the  $i^{\text{th}}$   
 5 segment of the path  $S$ .  $\beta_i$  is the angle between the wind field vector  $\omega$  and the boundary normal  
 6 vector  $n$ . If  $\beta_i > 90^\circ$ , it indicates an external influx, representing the quantity of CO that is  
 7 transported from regions outside the Tibetan Plateau into its boundaries. if  $\beta_i < 90^\circ$ , it indicates  
 8 an internal efflux, representing the amount of CO that is generated within the Tibetan Plateau but  
 9 dispersed to areas beyond its confines.  $\Delta S_i$  represents the integration step size.  $Flux, net$  calculated  
 10 as the difference between  $Flux, in$  and  $Flux, out$  represent the net CO flux across the Tibetan  
 11 Plateau boundary. If  $Flux, net > 0$ , it means that the portion transported from outside regions  
 12 into Tibetan Plateau is larger than the portion transported from Tibetan Plateau to the outside  
 13 regions, and vice versa for  $Flux, net < 0$ .

#### 14 2.4 Wind field correction and uncertainty

15 ERA5 provides wind field components  $u$  and  $v$  along with their uncertainties  $\sigma_u$  and  $\sigma_v$ .  
 16 Therefore, the wind speed  $w_{spd}$  and wind direction  $w_{dir}$  can be calculated as equation (4),

$$17 \quad w_{spd} = \sqrt{u^2 + v^2}, \quad w_{dir} = 180 + \frac{180}{\pi} \text{atan2}(u, v) \quad (4)$$

18 The uncertainty in the wind field can be calculated using the error propagation formulas (5) and  
 19 (6),

$$20 \quad \sigma_{wspd} = \sqrt{\sum_i \left[ \left( \frac{\partial w_{spd}(zi)}{\partial u(zi)} \times \sigma_{u(zi)} \right)^2 + \left( \frac{\partial w_{spd}(zi)}{\partial v(zi)} \times \sigma_{v(zi)} \right)^2 \right]} \quad (5)$$

$$21 \quad \sigma_{wdir} = \sqrt{\sum_i \left[ \left( \frac{\partial w_{dir}(zi)}{\partial u(zi)} \times \sigma_{u(zi)} \right)^2 + \left( \frac{\partial w_{dir}(zi)}{\partial v(zi)} \times \sigma_{v(zi)} \right)^2 \right]} \quad (6)$$

22 Where  $\sigma_{wspd}$  and  $\sigma_{wdir}$  are the uncertainties in wind speed and wind direction, respectively.

23  $\sigma_{u(zi)}$  and  $\sigma_{v(zi)}$  are the uncertainties in  $u(zi)$  and  $v(zi)$ ,  $\frac{\partial w_{spd}(zi)}{\partial u(zi)}$  and  $\frac{\partial w_{spd}(zi)}{\partial v(zi)}$  are the partial

24 derivatives of  $w_{spd}(zi)$  with respect to  $u(zi)$  and  $v(zi)$ ,  $\frac{\partial w_{dir}(zi)}{\partial u(zi)}$  and  $\frac{\partial w_{dir}(zi)}{\partial v(zi)}$  are the partial  
 25 derivatives of  $w_{dir}(zi)$  with respect to  $u(zi)$  and  $v(zi)$ , respectively. The  $zi$  represents the height  
 26 of the wind field.

27 Since CO concentration and wind field are distributed non-uniformly along with the vertical  
 28 height, a vertically averaged wind field is needed for flux calculation to minimize errors caused by  
 29 these non-uniformities. In order to do so, we first convert the volume mixing ratio (VMR) of CO  
 30 at each altitude into mass concentration (the product of vertical profile and atmospheric air mass)  
 31 via equation (7). We then take it as the weighting function to correct the original wind field via  
 32 equation (8) (Shaiganfar et al., 2017; Huang et al., 2020). Meanwhile, we calculate the uncertainty  
 33 in the wind field following the method of Huang et al. (2020).

$$34 \quad \tau_j(z_j) = \frac{x_j(z_j) \cdot Airmass_j(z_j)}{\sum_i x_i(z_j) \cdot Airmass_j(z_j)} \quad (7)$$

$$35 \quad \omega_{j, avg} = \sum_i \omega(z_i) \cdot \tau_j(z_j), \quad \theta_{j, avg} = \sum_i \theta(z_i) \cdot \tau_j(z_j) \quad (8)$$



$$\sigma_{\omega z} = \sqrt{\sum_i [\tau_j (\omega_{j, avg} - \omega(z_i))]^2}, \sigma_{\theta z} = \sqrt{\sum_i [\tau_j (\theta_{j, avg} - \theta(z_i))]^2} \quad (9)$$

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

## 6 2.6 Regression model for trend analysis

7 We establish a regression model expressed as equations (10) and (11) to simulate the seasonal  
 8 and inter-annual variabilities of transboundary transport CO flux along the Tibetan Plateau  
 9 boundary. This model consists of a third-order Fourier series and a linear function. We refer to Sun  
 10 et al. (2021) for detailed description of its resampling methodology.

$$Y^{org}(t) = Y^{mod}(t) + \varepsilon(t) \quad (10)$$

$$Y^{mod}(t) = A_0 + A_1 t + A_2 \cos\left(\frac{2\pi t}{365}\right) + A_3 \sin\left(\frac{2\pi t}{365}\right) + A_4 \cos\left(\frac{4\pi t}{365}\right) + A_5 \sin\left(\frac{4\pi t}{365}\right) \quad (11)$$

$$d\% = \frac{Y^{org}(t) - Y^{mod}(t)}{Y^{mod}(t)} \times 100 \quad (12)$$

11 where  $Y^{org}(t)$  and  $Y^{mod}(t)$  are the original and the fitted time series of CO flux, respectively.  
 12  $A_0$  is the intercept,  $A_1$  is the annual growth rate, and  $t$  represents the number of days since May  
 13 2018. The  $\varepsilon(t)$  is the residual between the original and the fitted results. The coefficients  $A_2 - A_5$   
 14 describe the seasonal cycle and  $A_1/A_0$  is the inter-annual trend. Deviations of CO fluxes from their  
 15 average seasonal values, known as seasonal enhancements, are calculated according to Equation  
 16 (12).

## 17 3 Variability of CO total column and transboundary transport flux

### 18 3.1 Variability of CO total column

19 We average the CO total column along the Tibetan Plateau boundary and investigate its daily,  
 20 seasonal and inter-annual variabilities. The results show that the daily mean CO total column  
 21 ranges from  $7.75 \times 10^{17}$  molec  $\text{cm}^{-2}$  to  $15.61 \times 10^{17}$  molec  $\text{cm}^{-2}$ , while the seasonal mean ranges  
 22 from  $10.06 \times 10^{17}$  molec  $\text{cm}^{-2}$  to  $12.01 \times 10^{17}$  molec  $\text{cm}^{-2}$ . As shown in Figure 2, high levels of  
 23 CO total column in all seasons are observed in the southeastern segment of the Tibetan Plateau  
 24 boundary, which is adjacent to southeastern Asia.

25 We employ the regression model to fit the CO total columns averaged along the closed-loop,  
 26 the southwestern, and the northeastern segments of the Tibetan Plateau boundary. The fitting  
 27 results in Figure 3 show that the model is capable of accurately capturing and replicating the  
 28 seasonal and inter-annual variabilities of CO total column averaged using all the three manners.  
 29 The correlation coefficient ( $r$ ) achieved are 0.84, 0.90, and 0.75, while the root mean square error  
 30 ( $RMSE$ ) is  $0.54 \times 10^{17}$  molec  $\text{cm}^{-2}$ ,  $0.41 \times 10^{17}$  molec  $\text{cm}^{-2}$ , and  $0.76 \times 10^{17}$  molec  $\text{cm}^{-2}$  for the  
 31 closed-loop, the southwestern, and the northeastern segments, respectively. Satellite observations  
 32 and the fitted results show distinct seasonal characteristics. Elevated levels of CO total column are  
 33 observed along the closed-loop, northeastern, and southwestern segments from late spring to  
 34 summer, while lower levels occur during autumn and winter. The closed-loop, southwestern, and  
 35 northeastern segments all exhibit a bimodal seasonal cycle. The closed-loop and southwestern  
 36 segment have a pronounced peak in late spring and a minor peak in early autumn, whereas the



1 northeastern segment has a minor peak in late spring and a pronounced peak in early autumn.  
2 Notably, significant fluctuations in the CO total column averaged along the closed-loop,  
3 southwestern, and northeastern segments occur in autumn and winter, with more substantial  
4 fluctuations in autumn than in winter. Over the past six years, the CO total column averaged along  
5 the closed-loop, southwestern, and northeastern segments have shown an upward annual trends of  
6  $0.68 \times 10^{15}$  molec cm<sup>-2</sup> yr<sup>-1</sup>,  $0.55 \times 10^{15}$  molec cm<sup>-2</sup> yr<sup>-1</sup>, and  $0.78 \times 10^{15}$  molec cm<sup>-2</sup> yr<sup>-1</sup>,  
7 respectively.

### 8 **3.2 Variability of transboundary transport CO flux**

9 Seasonal cycles of the external influx and internal efflux of CO across the closed-loop, the  
10 southwestern and northeastern segments of the Tibetan Plateau from May 2018 to April 2024 are  
11 shown in Figure 4. Summary of the corresponding statistics are tabulated in Table 1. The results  
12 show that the external influx and internal efflux of CO across the closed-loop, the southwestern,  
13 northeastern segments in each year are relatively close and have similar seasonal characteristics.  
14 High levels of CO flux occur in late autumn to winter, and low levels occur in summer. In most  
15 cases, CO flux maximizes in November, December or January, and minimizes in July or August.  
16 The month to month variability during late autumn to winter is greater than that in summer. For  
17 the closed-loop of the Tibetan Plateau, the CO external influx varies between 7.90 t s<sup>-1</sup> - 32.73 t s<sup>-1</sup>  
18 and the internal efflux varies between 7.84 t s<sup>-1</sup> - 29.61 t s<sup>-1</sup>. In comparison, the external influx in  
19 the southwestern segment fluctuates between 3.12 t s<sup>-1</sup> and 20.89 t s<sup>-1</sup>, while the internal efflux  
20 ranges from 1.15 t s<sup>-1</sup> to 8.86 t s<sup>-1</sup>. In the northeastern segment, the external influx varies between  
21 3.42 t s<sup>-1</sup> and 11.84 t s<sup>-1</sup>, and the internal efflux spans from 5.47 t s<sup>-1</sup> to 22.69 t s<sup>-1</sup> (Table 1).

22 We applied an inter-annual regression model to fit the external influx, internal efflux and net  
23 flux of CO averaged along the closed-loop, southwestern and northeastern segments of the Tibetan  
24 Plateau. The fitting results in Figure 5 show that the model is capable of accurately capturing and  
25 replicating the seasonal and inter-annual variabilities of all kinds of fluxes, yielding high  
26 correlation coefficients (*r*) and low root mean square errors (*RMSE*). For the closed-loop CO flux,  
27 the external influx and net flux show slight positive trends, with inter-annual growth rates of 0.65 t  
28 s<sup>-1</sup> yr<sup>-1</sup> and 1.03 t s<sup>-1</sup> yr<sup>-1</sup>, respectively. In contrast, the internal efflux displays a slight negative  
29 trend of -0.39 t s<sup>-1</sup> yr<sup>-1</sup>. In the southwestern segment, the external influx and internal efflux exhibit  
30 similar variabilities, with annual mean values of 11.76 t s<sup>-1</sup> and 4.41 t s<sup>-1</sup>, respectively. The annual  
31 growth rates for external influx and internal efflux are 0.52 t s<sup>-1</sup> yr<sup>-1</sup> and -0.06 t s<sup>-1</sup> yr<sup>-1</sup>,  
32 respectively. In comparison, the external influx and internal efflux in the northeastern segment  
33 exhibit notable differences, with average values of 5.94 t s<sup>-1</sup> and 13.17 t s<sup>-1</sup>, and annual growth  
34 rates of 0.15 t s<sup>-1</sup> yr<sup>-1</sup> and -0.28 t s<sup>-1</sup> yr<sup>-1</sup>, respectively. The annual growth rates for net fluxes in  
35 the southwestern and northeastern segments are 0.59 t s<sup>-1</sup> yr<sup>-1</sup> and 0.42 t s<sup>-1</sup> yr<sup>-1</sup>, respectively.

36 This suggests that in recent years, the Tibetan Plateau has experienced an increase in external  
37 influx, while the internal efflux has slightly decreased. The magnitude of the increase in external  
38 influx in the southwestern segment is greater than in the northeastern segment. Conversely, the  
39 magnitude of the decrease in internal efflux in the northeastern segment is greater than in the  
40 southwestern segment.

### 41 **3.3 Uncertainty of CO flux calculation**



1 From May 2018 to April 2024, the external influx and internal efflux of CO averaged along  
2 the Tibetan Plateau were  $17.70 \text{ t s}^{-1}$  and  $17.56 \text{ t s}^{-1}$ , respectively, resulting in a net influx of  $0.13 \text{ t}$   
3  $\text{s}^{-1}$ . These estimates are based on TROPOMI overpasses (13:30 local time (LT)). Extrapolating to  
4 a full year, the external influx, internal efflux, and net influx are estimated to be  $558.19 \text{ Tg yr}^{-1}$ ,  
5  $553.77 \text{ Tg yr}^{-1}$ , and  $4.10 \text{ Tg yr}^{-1}$ , respectively. These values are comparable to CO emission  
6 estimated by Borsdorff et al. (2020) for Mexico and Leguijt et al. (2023) for African cities  
7 (Borsdorff et al., 2020; Leguijt et al., 2023).

8 The complex terrain of Tibetan Plateau, with its significant fluctuations in elevation and  
9 ground albedo, coupled with the variable mixture of the Asian monsoon and local valley winds,  
10 greatly increases the uncertainty of calculating CO flux over the Tibetan Plateau.  
11 TROPOMI-based CO flux calculation over the Tibetan Plateau are influenced by various factors  
12 (Shaiganfar et al., 2011; Shaiganfar et al., 2017; Tan et al., 2019; Huang et al., 2020). Here we  
13 identify two primary sources of uncertainty: CO total column and the closed-loop wind field.

14 From May 2018 to April 2024, daily CO total column over the Tibetan Plateau varied from  
15  $4.02 \times 10^{17} \text{ molec cm}^{-2}$  to  $64.43 \times 10^{17} \text{ molec cm}^{-2}$ . The averaged standard deviation varied from  
16  $0.16 \times 10^{17} \text{ molec cm}^{-2}$  to  $3.46 \times 10^{17} \text{ molec cm}^{-2}$ , corresponding to error margins from 0.81% to  
17 17.89%. The average error in CO total column along the Tibetan Plateau's closed-loop was 4.27%.

18 Wind speed and direction uncertainties significantly affect CO flux calculations. The Tibetan  
19 Plateau's complex terrain exacerbates wind variability. Using formulas (5) and (6), and (9) in  
20 Section 2.3, we calculated the uncertainties from the corrected wind field across 22 levels from  
21 1000 hPa to 50 hPa. The averaged wind speed and direction uncertainties were  $3.39 \text{ m s}^{-1}$  and  
22  $54.55^\circ$ , respectively. Wind speed uncertainty exhibited clear seasonal fluctuations. In spring  
23 (MAM), uncertainties ranged from  $0.65$  to  $0.84 \text{ m s}^{-1}$ , while summer (JJA) saw a wider range of  
24  $0.90$  to  $1.15 \text{ m s}^{-1}$ . The lowest uncertainties were observed in autumn (SON), ranging from  $0.53$  to  
25  $0.65 \text{ m s}^{-1}$ . In winter (DJF), uncertainty values were slightly higher, between  $0.65$  and  $0.71 \text{ m s}^{-1}$ .  
26 Wind direction uncertainty also varied seasonally, with values between  $18.10^\circ$  and  $26.41^\circ$  in  
27 spring, increasing to  $24.17^\circ$  to  $34.09^\circ$  in summer. Autumn presented the lowest directional  
28 uncertainty, ranging from  $16.50^\circ$  to  $20.62^\circ$ , while winter values were comparable to spring, at  
29  $19.07^\circ$  to  $23.91^\circ$  (Table 2).

30 The average uncertainties for the corrected wind speed and direction were  $0.78 \text{ m s}^{-1}$  and  
31  $22.15^\circ$ . In calculation of the closed-loop CO flux, considering only the uncertainty in wind speed  
32 results in an average error of 6.99% in CO flux, while accounting solely for wind direction  
33 uncertainty leads to an average error of 11.03%. The total error induced by both the wind field and  
34 CO total column can be calculated using the error propagation equation. The uncertainty in CO  
35 flux caused by these factors amounts to 13.81%.

#### 36 **4 Factors driving the variability of transboundary transport CO flux**

##### 37 **4.1 Differences between southwestern and northeastern segments**

38 For the closed-loop flux, both external influx and internal efflux exhibit significant  
39 fluctuations in winter from 2020 to 2023, followed by a rapid decline. Although the net flux also  
40 exhibited considerable fluctuations during this period, its seasonal variability was less pronounced  
41 compared to the external influx and internal efflux. The net flux is positive from January to May  
42 and in August and December, but turns negative in June, July, and from September to November.  
43 Specifically, the external influx in the southwestern segment consistently exceeds the internal



1 efflux, whereas in the northeastern segment, the external influx is lower than the internal efflux.  
2 Furthermore, the external influx in the southwestern segment closely resembles that of the internal  
3 efflux in the northeastern segment, particularly during summer and early autumn when the rates of  
4 decline and increase are most pronounced. Conversely, during winter and early spring, the internal  
5 efflux in the southwestern segment aligns closely with the external influx in the northeastern  
6 segment. Additionally, in summer, the external influx in the northeastern segment surpasses the  
7 internal efflux in the southwestern segment.

8 In the southwestern segment, a significant increase in flux is noted during autumn, followed  
9 by a marked decline in late winter. The net flux variations are more intricate, yet they generally  
10 remain positive. The averaged net CO flux is recorded at  $7.36 \text{ t s}^{-1}$ , with an annual growth rate of  
11  $-0.59 \text{ t s}^{-1} \text{ yr}^{-1}$ , typically exhibiting low values during the summer. Notably, from December to  
12 June in each year, the net flux experiences three distinct peaks. The first peak is observed during  
13 late autumn and winter, attributed to increased burning activities, such as winter heating and  
14 agricultural burning, alongside the enhanced transport of strong westerly airflow. These conditions  
15 contribute to elevated flux levels. Furthermore, cold winter temperatures inhibit the diffusion and  
16 dilution of pollutants, facilitating their accumulation in localized areas (Kunhikrishnan et al.,  
17 2004). The second peak occurs in spring, influenced by several factors, including rising  
18 temperatures, melting ice and snow, and the onset of plant growth. These conditions promote the  
19 release of pollutants into the atmosphere (Assessment, 2004; Hung et al., 2022). Additionally,  
20 frequent climate fluctuations may destabilize the emission and transport processes of these  
21 pollutants. The third peak is observed around June, coinciding with the strengthening of the South  
22 Asian monsoon. This seasonal shift brings increased moisture and airflow, which aids in diluting  
23 pollutants and facilitating their transport over long distances (Yu et al., 2017; Bian et al., 2020;  
24 Huang et al., 2023).

25 In the northeastern segment, the external influx peaks in both summer and winter. In winter,  
26 there is a marked increase in external influx, followed by two declines in early and late spring.  
27 This behavior is likely influenced by the dynamics of the East Asian monsoon across different  
28 seasons and varying intensities of burning activities throughout the year. The internal efflux  
29 displays a more complex pattern, featuring three peaks and subsequent declines in winter, March,  
30 and June, which correspond to the three peaks of net flux observed in the southwestern segment.  
31 Similarly, the net flux variabilities in the northeastern segment are quite intricate. The average net  
32 CO flux is  $-7.23 \text{ t s}^{-1}$ , with an annual growth rate of  $-0.42 \text{ t s}^{-1} \text{ yr}^{-1}$ . The peak and trough values  
33 occur around autumn, specifically in late autumn to early winter for peaks and late autumn for  
34 troughs. This downward trend mirrors that of the internal efflux. Additionally, we observed that: (1)  
35 The CO flux transported through the southwestern segment into the Tibetan Plateau, along with its  
36 annual growth rate over the past six years, accounted for approximately 66.44% and 77.61% of the  
37 total CO flux within the Tibetan Plateau. This indicates that the external influx to the Tibetan  
38 Plateau and its changing trend are predominantly influenced by the southwestern segment. While  
39 CO transported through the southwestern segment into the Tibetan Plateau is on the rise, the  
40 internal efflux transported through the northeastern segment is experiencing a decline; (2) Around  
41 June each year, we observe minor seasonal peaks in the external influx and net flux across the  
42 southwestern segment, as well as in the internal efflux across the northeastern segment. This  
43 phenomenon may result from the combined effects of atmospheric circulation and the South Asian  
44 summer monsoon mechanism. The atmospheric flow prior to the onset of the South Asian



1 monsoon transports pollutants such as CO from South Asia to the Tibetan Plateau, leading to  
2 increased CO concentrations and fluxes in the region (Yu et al., 2017; Huang et al., 2023). This  
3 seasonal peak also highlights the intricate interactions between atmospheric circulation and the  
4 monsoon system in South Asia during the pollutant transmission process. (3) The southwestern  
5 segment exhibits distinct pollutant transport differences compared to the northeastern segment.  
6 For example, the averaged external influx across the southwestern segment reaches as high as  
7  $11.99 \text{ t s}^{-1}$ , while the averaged internal efflux across the northeastern segment is only  $4.60 \text{ t s}^{-1}$ .  
8 This is primarily attributed to a higher level of rapid industrialization and urbanization in  
9 Southeast Asia than in Tibetan Plateau, resulting in higher pollutant emissions (including CO) in  
10 the region.

11 Eastern China is predominantly downwind of the Tibetan Plateau, where the wind flow and  
12 atmospheric stability in the upper atmosphere predispose the eastern region to act as a receiving  
13 area for pollutants from the Tibetan Plateau. The Tibetan Plateau functions as a high-altitude  
14 natural barrier, effectively limiting the in-depth spread of pollutants (Ji et al., 2015). However, the  
15 rapid industrialization and urbanization in East China have led to high local pollutant emissions.  
16 Influenced by atmospheric circulation patterns and complex topography, East China experiences  
17 strong convection and large-scale circulatory systems. While most pollutants are recirculated and  
18 deposited within the region, significant transport occurs towards South Korea, Japan, and the  
19 North Pacific. A smaller fraction may also reach the eastern edge of the Tibetan Plateau. (Zhang et  
20 al., 2015; Yan and Bian, 2015).

#### 21 4.2 Spatiotemporal distribution of CO

22 We have analyzed spatiotemporal distribution of CO total column from May 2018 to April  
23 2024 over the Tibetan Plateau and its surrounding regions. CO total columns were averaged on  
24 both annual and seasonal timescales. Specifically, we estimated the average of CO total column  
25 for winter (December to February), the pre-monsoon period (March to May), the monsoon period  
26 (June to September), and the post-monsoon period (October to November). Seasonal variations  
27 were assessed by subtracting the mean annual CO total column from their seasonal averages. The  
28 resulting data are presented in Figures 6. Additionally, we analyzed the six-year average spatial  
29 distribution and correlation of CO total column concentrations across the Indian states of  
30 Himachal Pradesh, Uttarakhand, and Ladakh, as well as Nepal and Assam. The corresponding  
31 results are presented in Figure 7.

32 The results indicate that CO concentrations over the Tibetan Plateau are consistently lower  
33 than those in South Asia throughout the year. CO levels in South Asia are particularly elevated,  
34 especially during winter and spring, when the disparity is most pronounced. In summer, CO over  
35 the Tibetan Plateau disperses across a broader area. Notably, we observed that CO concentrations  
36 over the Tibetan Plateau are significantly higher than the annual average during the monsoon  
37 season. Specifically, the average CO concentration over the Tibetan Plateau increased by  $0.90 \times$   
38  $10^{17} \text{ molec cm}^{-2}$  compared to the annual mean, whereas in India and Nepal, CO concentrations  
39 decreased by  $-2.62 \times 10^{17} \text{ molec cm}^{-2}$  and  $-0.36 \times 10^{17} \text{ molec cm}^{-2}$ , respectively. These findings  
40 suggest that CO pollutants from South Asia are transported into the Tibetan Plateau during the  
41 South Asian monsoon. Over the past six years, the CO total column in northwestern India, Nepal,  
42 and Assam has exhibited relatively high growth rates, with annual increases of  $1.10 \times 10^{15} \text{ molec}$   
43  $\text{cm}^{-2} \text{ yr}^{-1}$ ,  $1.69 \times 10^{15} \text{ molec cm}^{-2} \text{ yr}^{-1}$ , and  $1.64 \times 10^{15} \text{ molec cm}^{-2} \text{ yr}^{-1}$ , respectively.



1           The increase of CO over the Tibetan Plateau during summer may be influenced by CO influx  
2 driven by the South Asian monsoon, as well as various meteorological factors. High temperatures  
3 and intense solar radiation in summer raise the atmospheric mixing layer height, facilitating the  
4 easier dispersion of pollutants in the atmosphere (Yang et al., 2004; Huang et al., 2023).  
5 Additionally, summer precipitation and wind speed affect the spatial distribution and transport  
6 pathways of pollutants. Elevated temperatures and strong convective conditions enhance vertical  
7 mixing and horizontal transport, resulting in more extensive and rapid diffusion of pollutants  
8 across the Tibetan Plateau (Zhang et al., 2020; Sun et al., 2021).

9           During winter and spring, CO concentrations rise significantly in South Asian regions,  
10 including Nepal, Bhutan, Assam in India, and parts of Myanmar. This increase is likely driven by  
11 intensified human activities, such as biomass burning, which is common in these areas during  
12 these seasons for heating and agricultural waste disposal, leading to substantial CO emissions. The  
13 southern Tibetan region, bordering northern Assam, serves as a key pathway for pollutant  
14 transport due to its distinct plain topography. Studies have confirmed that persistent organic  
15 pollutants, HCHO, and other contaminants are transported along the Yarlung Tsangpo River valley  
16 into the Tibetan Plateau (Sheng et al., 2013; Xu et al., 2024). TROPOMI remote sensing data  
17 further show that CO pollutants infiltrate the Tibetan Plateau through this region.

18           Despite the varying geographical, climatic conditions, and emission sources in these regions,  
19 the changes in CO concentrations are interrelated. CO levels tend to be lower in high-altitude  
20 areas, such as Himachal Pradesh, Uttarakhand, and Ladakh. Nepal, particularly the Kathmandu  
21 Valley, faces unique geographic and climatic challenges due to its dense population and  
22 concentration of industrial activities, exacerbating air pollution in the region (Islam et al., 2020).  
23 The Kathmandu Valley's encirclement by high mountains makes it especially vulnerable to  
24 pollution. In contrast, Assam's plains are heavily influenced by the monsoon and high humidity,  
25 which promotes the diffusion and deposition of pollutants. Despite these regional differences, CO  
26 concentrations in these areas show strong correlations with levels in the Ali region, Nyingchi City,  
27 Shannan City, and Shigatse City on the Tibetan Plateau, with correlation coefficients ( $r$ ) ranging  
28 from 0.52 to 0.71. These correlations suggest that long-distance pollutant transport, influenced by  
29 meteorological conditions such as monsoons, may link these regions to similar or shared pollution  
30 sources (Carrico et al., 2003).

31           The rapid increase in CO concentrations from South Asia, particularly India and Nepal, is  
32 closely linked to the rise in flux over the southwestern Tibetan Plateau, driven primarily by  
33 industrialization, agricultural activities, and population growth in the region. The topography of  
34 the Tibetan Plateau, combined with the monsoon system, facilitates the transport of pollutants  
35 from South Asia to the plateau. At the same time, the plateau's capacity to absorb CO plays a  
36 critical role in modulating regional fluxes. Furthermore, China's domestic CO emissions have  
37 significantly decreased due to policy controls and economic restructuring. The Tibetan Plateau has  
38 long been regarded as an atmospheric background, with local anthropogenic emissions deemed  
39 negligible (Yao et al., 2012; Zheng et al., 2018; Kang et al., 2019; Sun et al., 2021). Overall, there  
40 is a dual trend of increasing external influx and decreasing internal efflux, with the concentration  
41 of CO received by the Tibetan Plateau from South Asia exceeding the influence of emissions from  
42 inland China.

#### 43   **4.3 Transboundary transport pathway**



1 Spatial distribution of CO across four seasons around the Tibetan Plateau is shown in Figure  
2 8, and Figure 9 presents the atmospheric circulation patterns at 200 hPa and 500 hPa, including  
3 mean horizontal wind vectors and latitude-height and longitude-height distributions. Significant  
4 seasonal variations in CO concentration are observed in the Tibetan Plateau and surrounding areas,  
5 primarily influenced by atmospheric circulation patterns, pollutant source strength, and deep  
6 convection activities. The interplay among these factors contributes to the complex dynamics of  
7 CO distribution, revealing the intricate relationship between local emissions and regional  
8 meteorological conditions.

9 The south Asian summer monsoon transports a substantial amount of air from the surface to  
10 the stratosphere, characterized by southwesterly winds in the lower troposphere and an  
11 anticyclonic circulation in the upper troposphere (Abe et al., 2013). This anticyclonic system,  
12 dominant in the upper troposphere and lower stratosphere, significantly affects CO distribution in  
13 the Tibetan Plateau by enhancing the transport of tropospheric pollutants into the stratosphere  
14 (Huang et al., 2023). The high-altitude terrain of the Tibetan Plateau amplifies this process,  
15 facilitating vertical lifting of air. Surface pollutants are transported to the upper troposphere  
16 through the Asian summer monsoon anticyclone and become confined within the South Asian  
17 High's anticyclonic system (Randel et al., 2010; Bian et al., 2012; Bian et al., 2020; Huang et al.,  
18 2023). This dynamic leads to a significant increase in CO concentration in the upper troposphere  
19 and lower stratosphere. During the summer monsoon season, the southwesterly monsoon winds  
20 carry substantial pollutants into the plateau, strongly influenced by intense deep convection. These  
21 winds uplift CO from the southern plateau and disperse it across the region (Fu et al., 2006).  
22 Large-scale deep convection plays a crucial role in lifting CO from upwind source regions to  
23 higher altitudes. While some CO returns to the source region, a portion is transported to the  
24 Tibetan Plateau by upper-level southwesterly winds.

25 In contrast, under dry winter monsoon conditions, CO can be transported to the  
26 Himalayan-Tibetan Plateau via the westerlies. The northwesterly flow rapidly conveys CO  
27 pollutants from the Northern Hemisphere to the Tibetan Plateau and its surrounding regions  
28 (Zhang et al., 2015; Sun et al., 2021). This winter flow typically introduces strong cold air, causing  
29 intense surface cooling upon entering the plateau and resulting in descending air currents. This  
30 process enhances local circulation, exacerbating CO accumulation (Liu et al., 2003; Zhang et al.,  
31 2015). Additionally, the plateau's topographical features influence CO distribution, as stable  
32 atmospheric stratification limits vertical dispersion, leading to accumulation in the lower  
33 troposphere.

34 However, the amount of CO transported to the plateau is also influenced by the location and  
35 intensity of sources, air mass trajectories, and transport timing (Yao et al., 2012; Zhang et al., 2015;  
36 Kang et al., 2019; Sun et al., 2021). Variations in CO concentration depend not only on seasonal  
37 atmospheric circulation patterns but also on the distribution and intensity of pollution sources and  
38 the frequency and strength of deep convection activity. These complex interactions lead to  
39 significant seasonal changes in CO concentrations.

## 40 **5 Conclusions**

41 In this study, we utilized data products of TROPOMI, ERA5, and GEOS-CF, along with the  
42 closed-loop integral method, to quantify transboundary transport flux of CO over the Tibetan



1 Plateau. The variabilities and driving forces of external influx, internal efflux, and net flux of CO  
2 over the closed-loop, southwestern, and northeastern segments of Tibetan Plateau were analyzed.

3 The closed-loop CO concentration along the Tibetan Plateau boundary shown significant  
4 spatiotemporal variations, with daily means ranging from  $7.75 \times 10^{17}$  to  $15.61 \times 10^{17}$  molec cm<sup>-2</sup>  
5 and seasonal means from  $10.06 \times 10^{17}$  to  $12.01 \times 10^{17}$  molec cm<sup>-2</sup>, and with high levels in the  
6 southeastern segment adjacent to southeastern Asia. The closed-loop, southwestern, and  
7 northeastern segments exhibit a bimodal cycle, peaking in late spring and early autumn, with  
8 significant autumn fluctuations and less variability in winter. During the South Asian monsoon,  
9 CO concentrations increased by  $0.90 \times 10^{17}$  molec cm<sup>-2</sup> in the Tibetan Plateau, decreased by  $1.78$   
10  $\times 10^{17}$  molec cm<sup>-2</sup> in India, and decreased by  $0.36 \times 10^{17}$  molec cm<sup>-2</sup> in Nepal compared to the  
11 annual average. A strong correlation and synchronization of CO concentrations were observed  
12 between the South Asian border region and the Tibetan Plateau. Over the past six years, CO total  
13 columns in Tibet, India, and Nepal exhibited growth trends of  $0.54 \times 10^{15}$  molec cm<sup>-2</sup> yr<sup>-1</sup>,  $0.86 \times$   
14  $10^{15}$  molec cm<sup>-2</sup> yr<sup>-1</sup>, and  $1.17 \times 10^{15}$  molec cm<sup>-2</sup> yr<sup>-1</sup>, respectively. These trends are notably  
15 higher than the growth observed in Tibet. Over six years, growth trends of  $0.68 \times 10^{15}$  molec cm<sup>-2</sup>  
16 yr<sup>-1</sup>,  $0.55 \times 10^{15}$  molec cm<sup>-2</sup> yr<sup>-1</sup>, and  $0.78 \times 10^{15}$  molec cm<sup>-2</sup> yr<sup>-1</sup> were observed for each segment.

17 Transboundary transport flux of CO in the Tibetan Plateau is high in late autumn and winter,  
18 and low in summer. Six-year averaged external influx, internal efflux, and net flux are  $17.70$  t s<sup>-1</sup>  
19 and  $17.56$  t s<sup>-1</sup>,  $0.13$  t s<sup>-1</sup>, respectively. The external influx shows a slight positive trend of  $0.67$  t s<sup>-1</sup>  
20 yr<sup>-1</sup>, while net flux increases at  $1.07$  t s<sup>-1</sup> yr<sup>-1</sup>, contrasted by a minor decline in internal efflux of  
21  $-0.40$  t s<sup>-1</sup> yr<sup>-1</sup>. In the southwestern segment, external influx and internal efflux show comparable  
22 variability, with annual means of  $11.76$  t s<sup>-1</sup> and  $4.41$  t s<sup>-1</sup>, and growth rates of  $0.52$  t s<sup>-1</sup> yr<sup>-1</sup> and  
23  $-0.06$  t s<sup>-1</sup> yr<sup>-1</sup>, respectively. Conversely, the northeastern segment exhibits significant differences,  
24 with average influx and efflux of  $5.94$  t s<sup>-1</sup> and  $13.17$  t s<sup>-1</sup>, and growth rates of  $0.15$  t s<sup>-1</sup> yr<sup>-1</sup> and  
25  $-0.28$  t s<sup>-1</sup> yr<sup>-1</sup>, respectively. In summary, these trends indicate an increase in external influx and a  
26 slight decrease in internal efflux across the Tibetan Plateau, with significant regional differences in  
27 CO fluxes; the southwestern segments serves as the primary contributor to external influx,  
28 exhibiting considerable seasonal changes, while the eastern segments shows lower external influx  
29 than internal efflux, indicating a net efflux.

30 We assessed the uncertainties of wind speed and direction across 22 layers from 1000 hPa to  
31 50 hPa, obtaining average uncertainties of  $3.39$  m s<sup>-1</sup> for wind speed and  $54.55^\circ$  for wind direction,  
32 with corrected averages of  $0.78$  m s<sup>-1</sup> and  $22.15^\circ$ , respectively. The uncertainty in wind speed  
33 accounts for an average error of 6.99%, while the uncertainty in wind direction contributes an  
34 average error of 11.03%. The average error in CO total column along the Tibetan Plateau's  
35 closed-loop was 4.27%. Using the error propagation equation, the total uncertainty in CO flux  
36 from both factors is calculated to be 13.81%.

37 In conclusion, we quantified the CO flux over the Tibetan Plateau and found a significant  
38 seasonal trend, with an increasing external influx in recent years. Specifically, the southwestern  
39 segments of the Tibetan Plateau represent the primary source of CO, demonstrating an upward  
40 trend potentially associated with the rapid increase in CO concentrations from South Asia.  
41 Conversely, CO transmission to the eastern segments is declining, likely due to decreased  
42 emissions and the plateau's inherent capacity to absorb CO. The unique geographical position of  
43 the Tibetan Plateau makes it crucial for observing and investigating transboundary transport of



1 regional atmospheric pollutants, providing a scientific basis for understanding global pollutant  
2 transport mechanisms and informing environmental protection policies.

3 **Data availability.** The TROPOMI CO dataset of this study is available for download at  
4 <https://scihub.copernicus.eu/> (last access: 16 June 2024). ERA5 hourly wind data are available  
5 download at <https://cds.climate.copernicus.eu/> (last accessed: 1 June 2024). GEOS-CF simulations  
6 are available for download at <https://gmao.gsfc.nasa.gov/> (last accessed: 12 April 2024).  
7 GEOS-Chem simulations in this study are available on request from Youwen Sun  
8 (ywsun@aiofm.ac.cn)

9 **Author contributions.** ZS carried out the data analysis and prepared the paper with input from all  
10 coauthors. YS designed the study. HY conducted the GEOS-Chem simulations and, along with  
11 XL, YS, ZP, CYL, and CL, provided constructive comments. HY also offered valuable insights  
12 into the data analysis.

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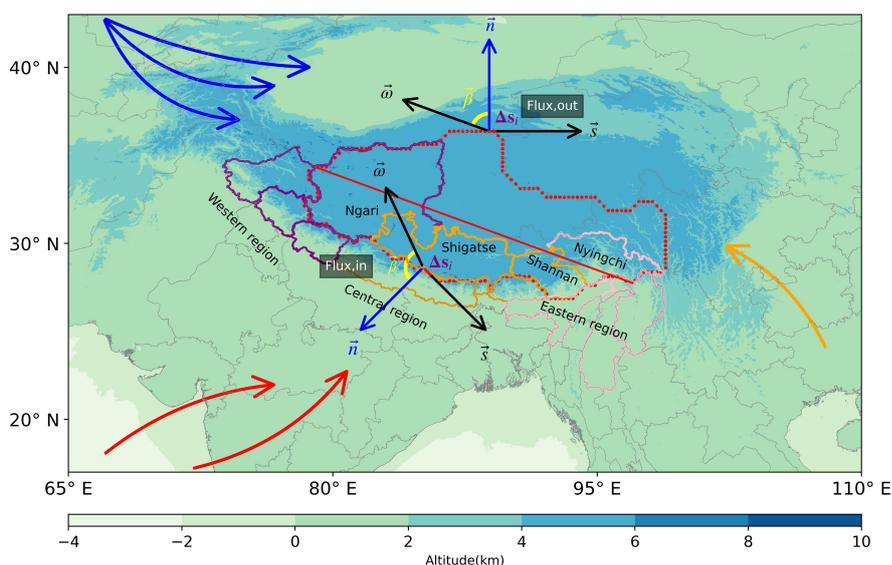
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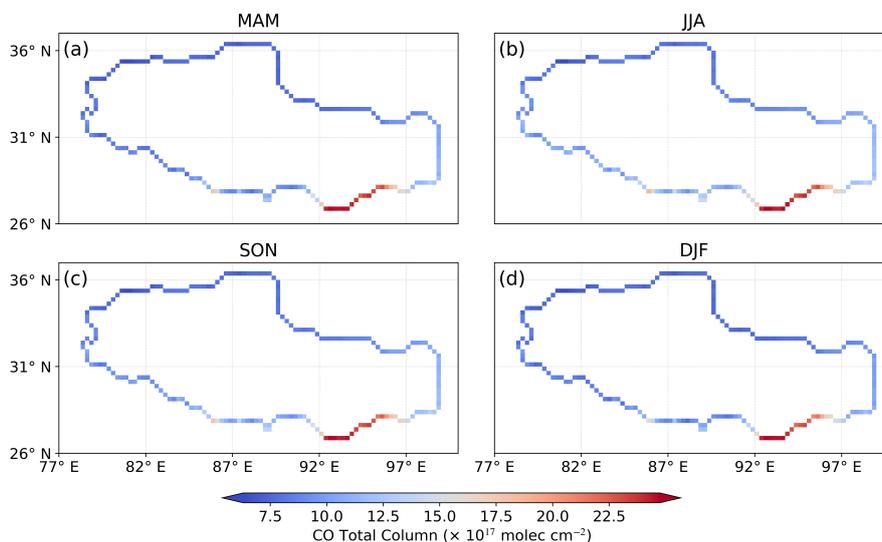
24 **Figures**



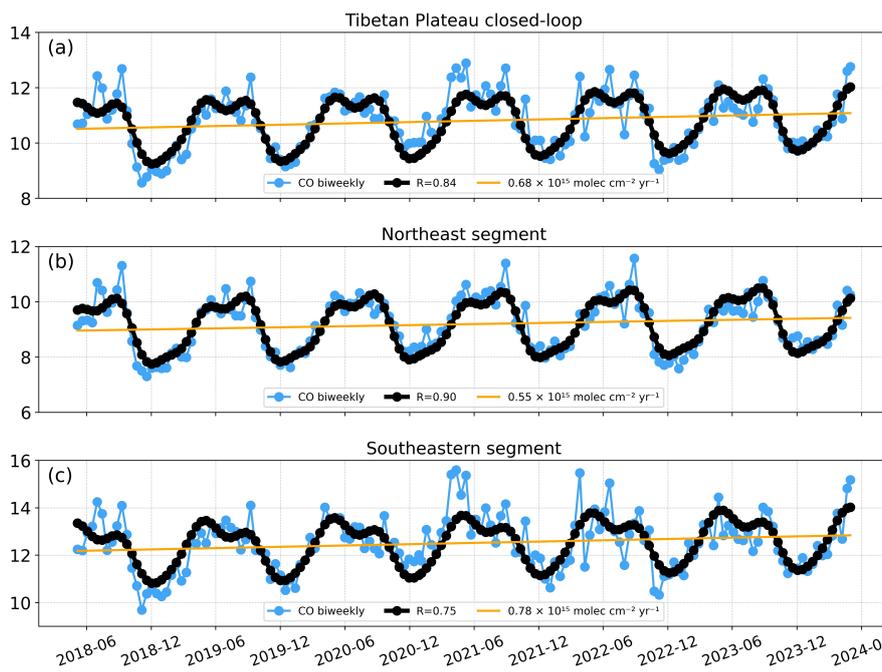
25  
 26 **Fig. 1.** Geographical description of the Tibetan Plateau and demonstration of the closed-loop integral method for  
 27 CO flux calculation, with red points indicating the closed-loop. Surrounding areas are categorized into western (in



1 purple), central (in orange), and eastern (in pink) regions. Within the Tibetan Plateau, highlighted areas include  
 2 Ngari Prefecture, Shigatse City, Shannan City, and Nyingchi City. Elevation data is sourced from NOAA NCEI.  
 3 The red bold line split the closed-loop of the Tibetan Plateau into southeastern and northeastern segments.



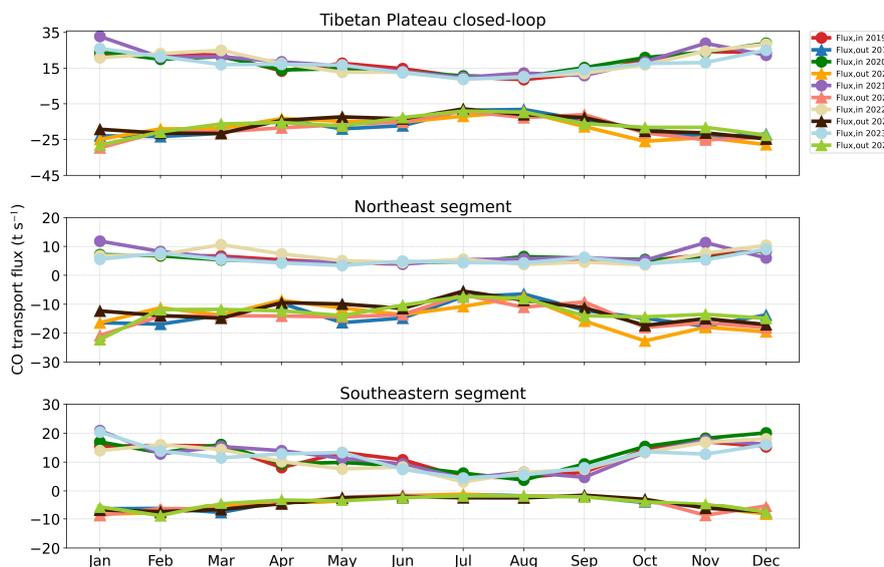
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 5 **Fig. 2.** Seasonal average of CO total column over the Tibetan Plateau, which is derived from data collected across  
 6 all days from May 2018 to April 2024, and is categorized by spring, summer, autumn, and winter.



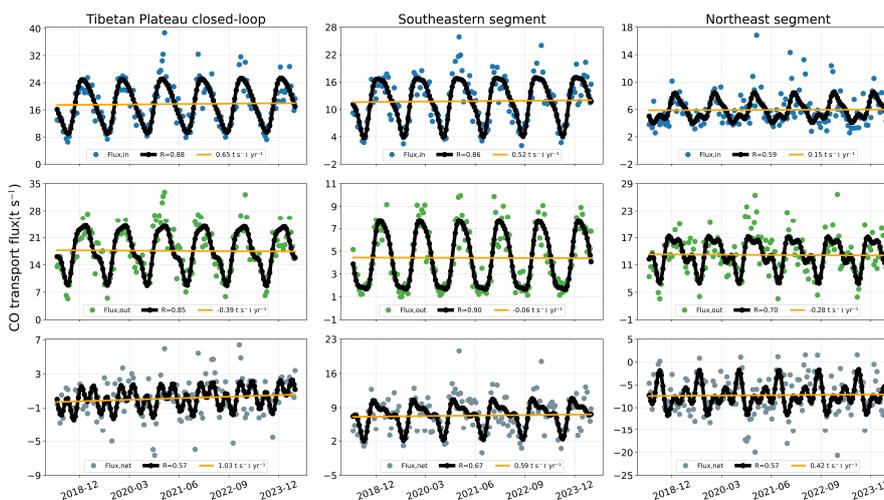
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 8 **Fig. 3.** Panels (a), (b), and (c) depict the inter-annual variabilities of CO concentrations in the closed-loop of the



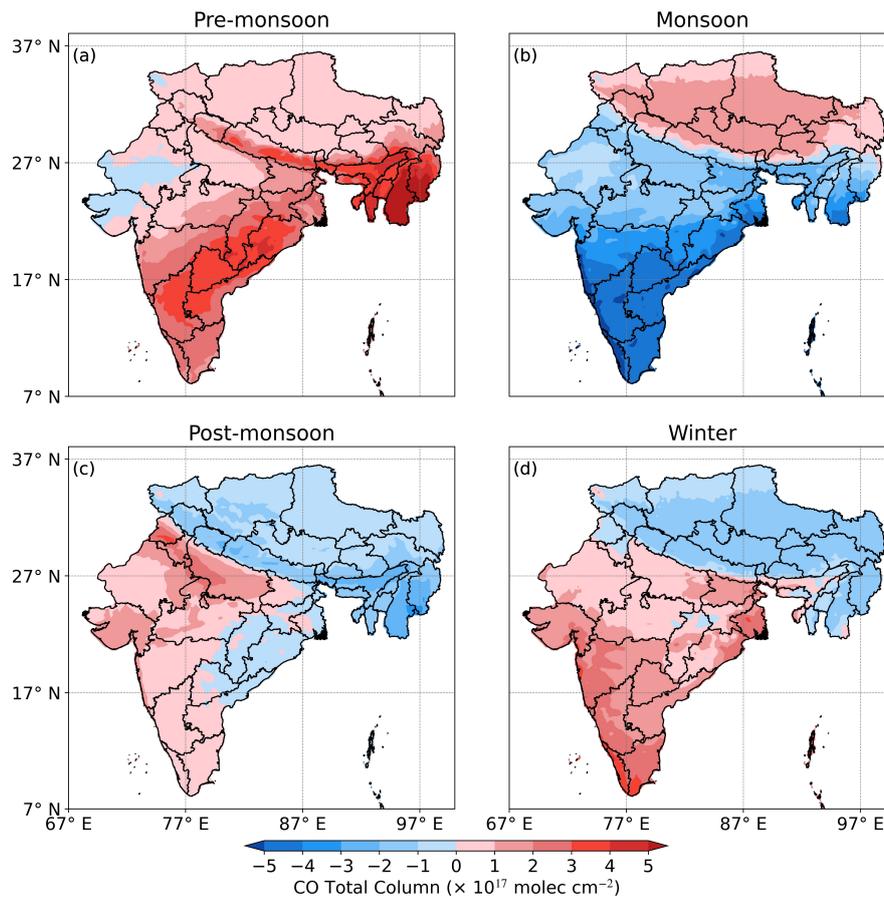
1 Tibetan Plateau, as well as the northeastern and southwestern segments, from May 2018 to April 2024. Blue dots  
 2 represent biweekly averaged CO total column. The figures illustrate the seasonal trend (black line) and  
 3 inter-annual trend (orange line) fitted by the seasonal cycle model.



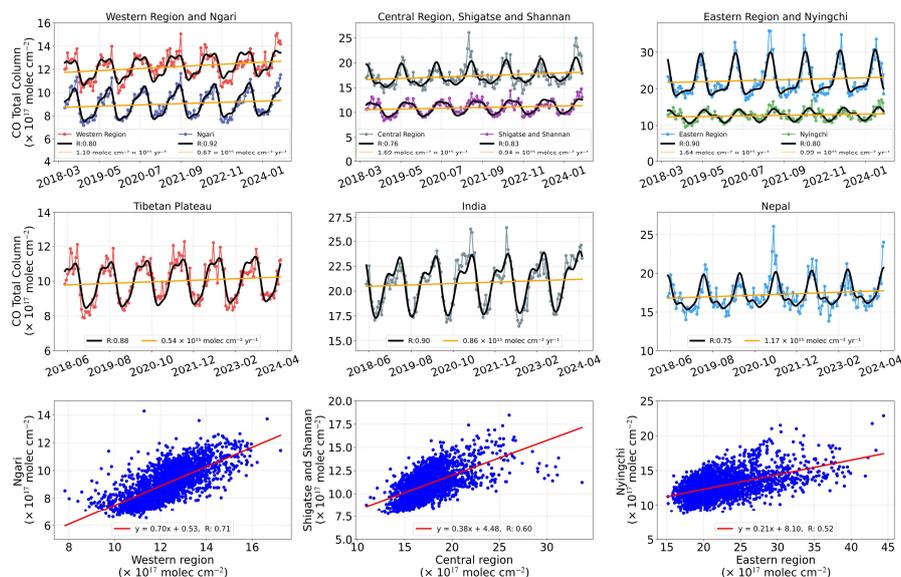
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 5 **Fig. 4.** Monthly averaged external influx and internal efflux of CO over the closed-loop, the southwestern, and the  
 6 northeastern segments of the Tibetan Plateau. Results are presented based on five complete years (2019–2023).



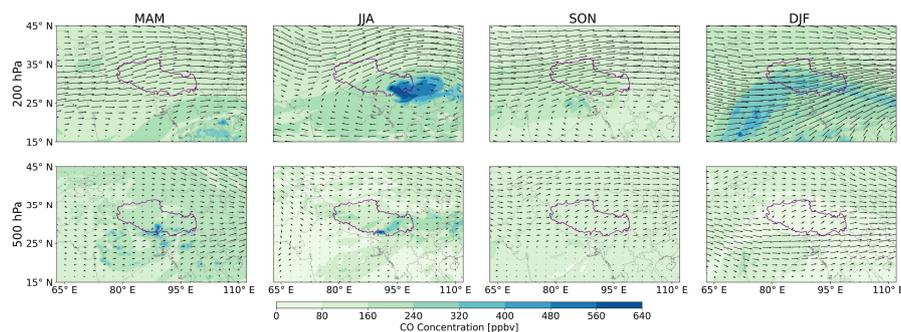
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 8 **Fig. 5.** Inter-annual variabilities of external influx, internal efflux and net flux of CO over the closed-loop, the  
 9 southwestern, and the northeastern segments of the Tibetan Plateau from May 2018 to April 2024. Blue dots  
 10 represent external influx, green dots indicate internal efflux, and gray dots show net flux. The seasonal trend (black  
 11 line) and inter-annual trend (orange line) are fitted using the seasonal cycle model.



1  
2 **Fig. 6.** CO total columns over the Tibetan Plateau and south Asia during the pre-monsoon, monsoon,  
3 post-monsoon, and winter. The data are collected from May 2018 to April 2024.

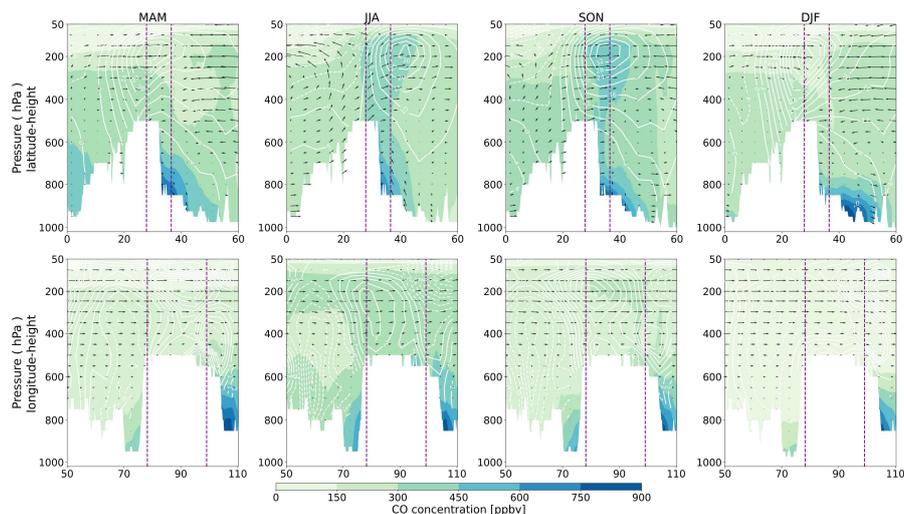


1  
 2 **Fig. 7.** Seasonal, inter-annual variabilities, and correlation analysis of CO concentrations over the western, central,  
 3 and eastern regions outside the Tibetan Plateau, and over the Ngari, Shigatse, Shannan, and Nyingchi within the  
 4 Tibetan Plateau. The red, dark blue, gray, purple, blue, and green dots in the figure represent CO concentrations in  
 5 the western region outside the Tibetan Plateau, Ngari, the central region outside the Tibetan Plateau, Shigatse,  
 6 Shannan, the eastern region outside the Tibetan Plateau, and Nyingchi, respectively.



7  
 8 **Fig. 8.** Spatial distribution of CO concentration surrounding the Tibetan Plateau across different seasons, alongside  
 9 mean horizontal wind vectors at 200 hPa and 500 hPa, represented by arrows. The study area is outlined in purple.  
 10 The CO spatial distribution data is available from GEOS-CF, while the meteorological fields are derived from  
 11 ERA5.

12  
 13



1

2 **Fig. 9.** The first row shows the latitudinal–altitude distribution of CO concentrations in different seasons averaged  
3 over the range 50°–110° E (positions correspond to different columns). The white contours at intervals of  $18 \text{ m s}^{-1}$   
4 represent the westerly (solid) and easterly (dashed) mean meridional winds; the white areas represent the terrain,  
5 and the arrows represent the wind vectors (vertical speed units are  $10^{-4} \text{ hPa s}^{-1}$ , zonal wind units are  $\text{m s}^{-1}$ ); the  
6 study area is marked by the purple dashed line. The second row is calculated from the longitude–altitude angles,  
7 averaged over the range 27°–33°N. Here the white contours represent the southerly (solid) and northerly (dashed)  
8 mean zonal winds, and the horizontal component of the wind vector is the meridional wind ( $\text{m s}^{-1}$ ). The  
9 meteorological fields are from ERA5.



1 **Tables**

2 **Table 1.** Statistics of the external influx and internal efflux of CO across the closed-loop, the southwestern and  
3 northeastern segments of the Tibetan Plateau from May 2018 to April 2024.

Type		Year							
		2018	2019	2020	2021	2022	2023	2024	
Seasonal cycle (monthly mean)	Tibetan	Flux,in max/min (t/s)	22.69/7.90 (12/8)	24.21/8.54 (11/8)	28.88/10.24 (12/8)	32.73/9.77 (1/7)	28.33/8.69 (12/7)	25.95/8.88 (1/7)	23.12/18.40 (3/4)
		Flux,out max/min (t/s)	22.45/7.89 (12/8)	23.56/8.16 (11/8)	27.80/9.37 (12/8)	29.61/9.10 (1/7)	24.61/7.84 (12/7)	28.08/9.05 (1/7)	22.18/16.65 (3/1)
	southwestern segment	Flux,in max/min (t/s)	15.21/3.57 (12/8)	17.04/4.66 (11/8)	20.09/3.77 12/8	20.89/4.35 (1/7)	17.98/3.12 (12/7)	20.36/4.48 (1/7)	16.69/11.10 (3/1)
		Flux,out max/min (t/s)	7.18/1.25 (11/7)	8.21/1.19 (12/7)	8.41/1.15 (1/7)	8.79/1.47 (1/6)	7.56/1.49 (12/9)	8.86/1.71 (2/7)	7.14/5.89 (1/3)
	northeastern segment	Flux,in max/min (t/s)	7.89/3.71 (11/10)	8.48/3.88 (12/6)	8.79/4.46 (12/7)	11.84/3.85 (1/6)	10.59/3.63 (3/10)	9.01/3.42 (12/5)	6.80/4.33 (1/4)
		Flux,out max/min (t/s)	15.97/5.84 (12/8)	17.63/6.47 (11/8)	22.69/7.35 (10/8)	20.82/6.73 (1/7)	17.34/5.47 (10/7)	22.21/7.34 (1/7)	16.30/9.52 (3/1)

4 **Table 2.** Uncertainties in the corrected mean wind speed and direction for the wind field used for calculating the  
5 closed-loop flux of CO over the Tibetan Plateau.

Year	Averaged Wind Speed Uncertainty (m/s)				Averaged Wind Direction and Its Uncertainty (°)			
	MAM	JJA	SON	DJF	MAM	JJA	SON	DJF
2018	0.78	1.15	0.60	0.67	22.31	34.09	18.10	19.07
2019	0.65	0.92	0.60	0.71	19.84	24.90	18.23	23.91
2020	0.75	0.93	0.53	0.65	23.19	24.17	13.99	22.69
2021	0.70	1.03	0.65	0.67	18.10	28.24	20.62	21.23
2022	0.84	0.90	0.59	0.68	26.41	28.05	16.50	20.02
2023	0.67	1.07	0.65	0.68	18.65	31.01	19.53	22.28
2024	0.66	-	-	-	19.41	-	-	-

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