



Trace gas atmospheric rivers: remote drivers of air pollutants

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Abstract.

Understanding how, when, and where long-range transport of pollution impacts regional and local air quality is key to developing effective air quality management strategies. Here, we propose a new approach for the characterization of the long-range transport of trace gases. We extend and modify the “atmospheric river” concept used to describe rapid long-range transport of water vapor in order to identify and analyze the transport of tropospheric ozone (O₃) and two key ozone precursors, carbon monoxide (CO), and peroxyacetyl nitrate (PAN). We apply our trace gas atmospheric river (TGARs) detection algorithm to a 14-year Tropospheric Chemistry Reanalysis (TCR-2) dataset based on global chemical data assimilation products, spanning the period 2005–2019. Over this time period, we find more than 300,000 TGARs events globally. These events account for up to 60% of total transport in some regions. We find that TGARs occur with a noticeably high frequency over mid-latitudes in both hemispheres (~20 days per 3 months). These findings highlight the important role of rapid transport events in the long-range transport of pollution.



1 Introduction

Air pollution does not respect borders. The long-range transport (LRT) of air pollution from various sources can significantly enhance background pollution levels and contribute to episodic pollution events (Holloway et al., 2000; Lin et al., 2010).

15 The importance of the role of LRT of tropospheric ozone (O_3) and its precursors in air quality has been recognized by the Task Force on Hemispheric Transport of Air Pollution (HTAP) (Janssens-Maenhout et al., 2015). With a lifetime of up to a few weeks, tropospheric O_3 can be transported hemispheric distances and significantly impact regional and local air quality. The significance of intercontinental O_3 transport on air quality has been highlighted in previous works (e.g. (Li et al., 2002; Derwent, 2004)). For example, episodic O_3 events in the western USA have been shown to be influenced by Asian emissions
 20 (Zhang et al., 2008; Lin et al., 2014; Jaffe et al., 2018; Miyazaki et al., 2022).

Tropospheric O_3 is not directly emitted at the surface but is produced by reactions of nitrogen oxide radicals (NO_x) with other precursors in the presence of sunlight (Crutzen, 1974). Certain long-lived ozone precursors, such as carbon monoxide (CO) and peroxyacetyl nitrate (PAN), can themselves be transported on hemispheric scales. For example, several studies have explored transpacific transport of O_3 and CO through observational data, satellite data, and model simulations (Heald et al.,
 25 2003; Derwent, 2004; Lin et al., 2014). The role of PAN, a long-lived reservoir for NO_x in the free troposphere, in the export of O_3 has been highlighted in numerous studies (Logan et al., 1981; Guerova et al., 2006; Zhang et al., 2008; Jiang et al., 2016; Okamoto et al., 2023; Zhai et al., 2024). Past work has highlighted the episodic nature of LRT (Atlas et al., 1992; Yienger et al., 2000; Liang et al., 2004; Lin et al., 2008; Fiore et al., 2009; Hu et al., 2019). However, to date, the long-term global climatology of extreme trace gas transport events, including their frequency, characteristics, and impact on total trace gas transport, has not
 30 been quantified.

Atmospheric rivers (ARs), narrow corridors of long-distance moisture transport, were initially conceptualized to describe water vapor transport (Zhu and Newell, 1998; Ralph et al., 2018; Guan and Waliser, 2015; Ralph et al., 2020). The definition of what constitutes an ARs event provides a framework to quantify the climatology of these events and to study their frequency and changes over decadal timescales. This concept has been adapted and applied to aerosol atmospheric rivers (AARs)
 35 (Chakraborty et al., 2021, 2022).

Here, we extend this concept to develop an approach for quantifying trace gas extreme transport events, hereafter referred to as Trace Gas Atmospheric Rivers (TGARs). We focus on tropospheric O_3 and two key O_3 precursors, CO and PAN, as represented in the Tropospheric Chemical Reanalysis version 2 (TCR-2) dataset (Miyazaki et al., 2015, 2020). In section 2, we describe the TCR-2 dataset and the TGARs algorithm. In section 3, we present results on TGARs characteristics, seasonal
 40 patterns of tropospheric transport, and the contribution of TGARs to total transport and interannual variability. In section 4, we discuss the findings. Finally, we present conclusions in section 5.



2 Methods and Datasets

2.1 Tropospheric chemical reanalysis (TCR-2) data

For the analysis of TGARs, we used the TCR-2 global chemical reanalysis dataset. TCR-2 was developed within the framework of the multi-constituent multi-model chemical data assimilation (MOMO-Chem) using an ensemble Kalman filter data assimilation technique (Miyazaki et al., 2015, 2020). The chemical reanalysis ingests various satellite measurements to estimate emissions and concentrations of numerous species. The assimilated measurements include O₃ from the Tropospheric Emission Spectrometer (TES) and Microwave Limb Sounder (MLS), CO from the Measurements of Pollution in the Troposphere (MOPITT), HNO₃ from MLS, and NO₂ from the Ozone Monitoring Instrument (OMI), the Scanning Imaging Absorption Spectrometer for Atmospheric Chartography (SCIAMACHY), and the Global Ozone Monitoring Experiment-2 (GOME-2). The TCR-2 products have been extensively validated with independent in-situ and aircraft observations (Miyazaki et al., 2019, 2021, 2022).

For the TGARs analysis, we used three chemical species (O₃, CO, and PAN) and two meteorological variables (wind components: u and v) from TCR-2. These datasets are provided with a horizontal resolution of $1.1^\circ \times 1.1^\circ$, 27 pressure levels, and a time interval of 6 hours. Each of the three chemical species plays a critical role in air pollution, but they have different chemical characteristics, such as lifetimes and sources. As a result, their vertical distributions and seasonal variations differ significantly across regions, as shown in Figure 1.

The primary sources of tropospheric O₃ are chemical production from precursors such as CO, CH₄, NO_x and volatile organic compounds (VOCs) (Crutzen, 1974; Monks et al., 2009) as well as inflow from the stratosphere. Since O₃ concentrations increase with altitude, horizontal transport in the middle and upper troposphere plays a key role in analyses of overall long-range transport. However, it is important to note that in certain cases, these analyses may be complicated by contributions from stratospheric inflows. Since CO sources are primarily at the surface, CO tends to decrease with altitude. CO can provide a clear representation of TGARs originating from anthropogenic and biomass combustion sources, as they are not significantly affected by chemical formation processes during transport. The horizontal and vertical distribution of PAN is determined by a complex interplay of emission sources, chemical reactions, and meteorological conditions. At the low temperatures typically found in the free troposphere, PAN is relatively stable and can be transported over long distances from emission regions (Fischer et al., 2014). However, at higher temperatures, PAN decomposes, releasing NO_x, which significantly affects O₃ and OH formation, thereby affecting air quality (Fischer et al., 2014). PAN concentrations peak at about 400–500 hPa, where favorable transport and production conditions lead to accumulation. By including these three different species in the TGARs analysis, we gain insight into the commonalities and species-dependent characteristics of TGARs, thereby improving our understanding of their dynamics and impacts.

2.2 Trace gas atmospheric river algorithm

The concept of ARs has been extensively used to describe narrow corridors of concentrated moisture that transport large amounts of water vapor from the tropics to higher latitudes (Zhu and Newell, 1998; Waliser and Guan, 2017; Ralph et al.,

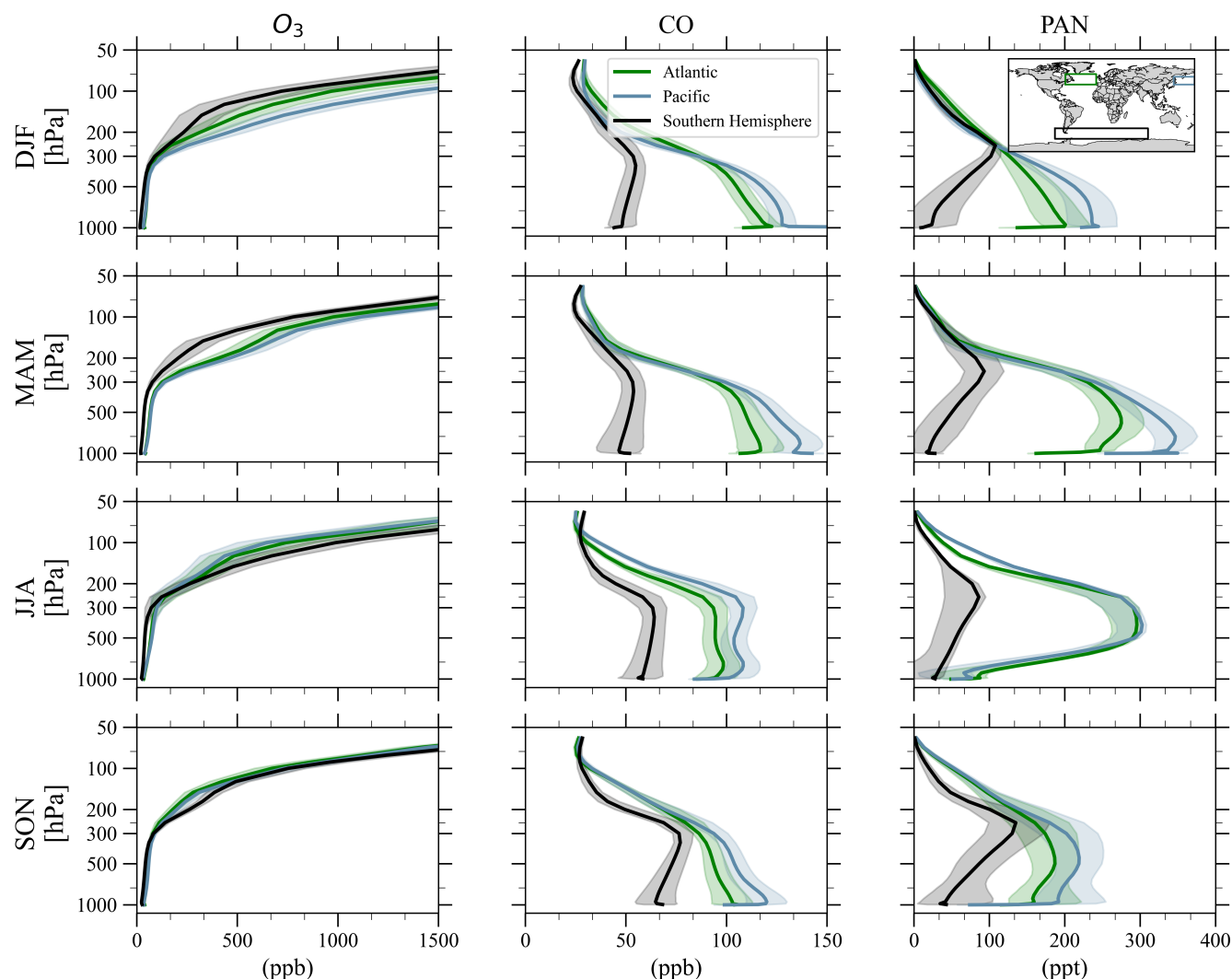


Figure 1. Regional mean vertical profiles of O_3 (ppb), CO (ppb), and PAN (ppt) concentrations from the TCR-2 chemical reanalysis for the period 2005–2019 as a function of pressure height (hPa). The results are shown for regional averages over three regions: the Atlantic (40°N – 60°N , 70°W – 10°W , green lines), the Pacific (40°N – 55°N , 140°E – 180°E , blue lines), and the Southern Hemisphere (65°S – 45°S , 90°W – 90°E , black lines) for seasonal averages in December–February (DJF), March–May (MAM), June–August (JJA), and September–November (SON). The shaded areas represent the standard deviation of $\pm 1\sigma$.

75 2018). In order to quantify the frequency and impact of these phenomena over time, definitions and detection algorithms are required. Several AR detection algorithms have been registered in the Atmospheric River Tracking Method Intercomparison Project (ARTMIP), providing intensity, direction, geometry selection, axis, and integrated vapor transport (IVT) information (Guan et al., 2010; Hagos et al., 2015; Waliser and Guan, 2017; Ralph et al., 2018; Rutz et al., 2019; Xu et al., 2020; O’Brien



et al., 2022). Building on this concept, we extend ARs to trace gases by adapting and modifying existing ARs (Guan and Waliser, 2015) and AARs (Chakraborty et al., 2021) algorithms. The following steps were implemented to detect TGARs:

(1) Integrated Gas Transport (IGT) values ($kgm^{-1}s^{-1}$) are computed by vertically integrating the six-hourly 3D wind fields (u, v) and trace gas concentrations (C , in mass mixing ratio). For O_3 , the integration is performed from the surface to 300 hPa to minimize the effects of stratospheric ozone. In contrast, CO and PAN were integrated from the ground surface to 60 hPa, since tropospheric sources mostly control these species. The integrated zonal and meridional fluxes are calculated as follows:

$$IGT(u, v)_{O_3} = \sum_{1000hPa}^{300hPa} C_{O_3} \times (u, v) \quad (1)$$

$$IGT(u, v)_{CO, PAN} = \sum_{1000hPa}^{60hPa} C_{CO, PAN} \times (u, v) \quad (2)$$

The basic steps of the TGARs detection algorithm are primarily based on the original ARs algorithm (Guan and Waliser, 2015), with minor modifications. We only consider IGT values exceeding the 85th percentile to identify transport extremes calculated separately for each grid cell. The percentile calculation is based on an overlapping 3-month window centered on each month, using all time steps from 2005 to 2019. Daily TGARs are detected at four time steps (00:00, 06:00, 12:00, and 18:00 UTC) for the 14-year period 2005–2019 for each trace gas species. The total IGT at each grid point and time step is computed as follows:

$$IGT_n = \sqrt{IGTU_n^2 + IGTV_n^2} \quad (3)$$

The IGTU and IGTV represent the vertically integrated gas mass flux in the zonal and meridional directions, respectively, for each trace gas species (n).

(2) Contiguous areas ("objects") of elevated IGT are then identified. At this stage, a candidate TGARs object is defined as a contiguous region where grid cells exceed the 85th percentile of IGT values.

(3) To retain a candidate object as a TGARs, the following criteria are applied:

- The direction of the object-mean IGT must be consistent within 45° of the object's shape orientation.
- The object must be longer than 2000 km in length and have a length-to-width ratio greater than 2. The objects that meet these criteria are referred to as TGARs.

The frequency of TGARs at each grid cell is calculated as the percentage of time steps during which TGARs are detected at that grid cell for each analysis year. The detailed methodology for the TGARs algorithm, including the relevant equations, is provided in Appendix B (Equations B1 to B8).

2.2.1 Differences between TGARs and AARs

The TGARs settings used in this study are derived from the optimized settings developed for ARs (Guan and Waliser, 2015) and AARs (Chakraborty et al., 2022). While TGARs results are often similar to ARs and AARs in terms of spatial and



seasonal variations, differences emerge in detailed spatial patterns and frequencies. For example, TGARs are less frequent, occurring about 20 days per 3 months compared to 20–40 days for AARs. In addition, AARs contribute up to 80% of the total annual transport, depending on the aerosol type (Chakraborty et al., 2022), which exceeds the contribution of TGARs (about 60%). These differences may reflect fundamental differences between trace gases and aerosols, including variations in source distributions and removal processes, such as wet deposition along ARs, which are strongly dependent on the specific properties of trace gases and aerosols. However, updates in the algorithm settings may also contribute to these discrepancies. In the AARs analysis, Chakraborty et al. (2022) uses grid-wise integrated aerosol transport (IAT) thresholds, as well as specific length and aspect ratio limits, significantly influencing event detection. Building on the optimized ARs and AARs settings, the following updates were applied to the TGARs algorithms:

- The object-average threshold based on the median IGT value, used in the AARs analysis, was removed to avoid misidentifying large regions with low IGT values as TGARs during poleward transport. Instead, a threshold based on pixel area was applied. Using a grid pixel area threshold of $1300 \times 900 \text{ m}^2$ retained more than 50% of TGARs objects, though results varied by chemical species.
- The ARs algorithm limitation to poleward transport, which was necessary for water vapor ARs, was removed. For trace gases, both polar and equatorial transport are significant due to their substantial sources in tropical and extratropical regions.



3 Results

125 3.1 TGARs characteristics and seasonal IGT pattern

Figure 2 illustrates examples of TGARs events detected using the algorithm outlined in Section 2.2. These cases demonstrate extreme transport phenomena, including O_3 transported from Asia to the western USA during an episodic event highlighted by Lin et al. (2012) (left), and CO emissions from Amazon forest fires reaching São Paulo, Brazil (right). The TGARs analysis identified a significant O_3 transport event on May 23, 2010, with a TGT value of about $0.3 \text{ kg m}^{-1} \text{ s}^{-1}$ passing over Los Angeles. Another example shows enhanced CO TGARs (about $0.02 \text{ kg m}^{-1} \text{ s}^{-1}$) on September 21, 2019, originating from Amazon fires and passing over São Paulo, consistent with observations of enhanced carbonaceous AARs from Amazon forest fires reported by Chakraborty et al. (2021). These examples highlight the potential impact of TGARs on air quality in down-wind regions. Additional examples of TGARs events, including trans-Atlantic, trans-Pacific, and polar transport, are shown in supplementary Figure S3.

135 The basic characteristics of the detected TGARs are summarized in Figure 3. TGARs were defined as IGT features with a length exceeding 2000 km, a length-to-width aspect ratio greater than 2, and a pixel-wise IGT threshold greater than 85%. Individual TGARs can be up to 10,000 km long and over 1,500 km wide, illustrating their potential to transport pollution over long distances. More extensive and wider TGARs are expected to have more intense and long-lasting impacts. For each species, the frequency of TGARs events is shown as a function of their length, width, and length-to-width ratio (Figure 3). Over the 14-year study period, a total of more than 324K TGARs events were detected for O_3 , 317K for CO, and 306K for PAN, resulting in an average of 13–14 TGARs events per 6-hour time step globally. The average length, width, and length/width ratio of the TGARs were estimated as follows: O_3 (4,059 km, 650 km, 7), CO (4,390 km, 657 km, 7), and PAN (5,951 km, 788 km, 8). These variations among species reflect differences in emission source regions and atmospheric lifetimes. Despite these differences, the TGARs for all species share a consistent characteristic: their length is approximately 6–7 times their width.

145 The spatial pattern of seasonal IGT, averaged over three-month periods, shows enhanced transport in the mid-latitudes of both hemispheres, with a distinct seasonal distribution consistent across all three species (Figure 4). The most significant IGT values are observed over the Pacific and the Atlantic in the NH and SH mid-latitudes. The highest IGT values were observed over the NH Pacific during DJF and MAM, with O_3 ranging from 0.015 to $0.020 \text{ kg m}^{-1} \text{ s}^{-1}$, CO from 0.020 to $0.025 \text{ kg m}^{-1} \text{ s}^{-1}$, and PAN from 0.015 to $0.020 \times 10^{-2} \text{ kg m}^{-1} \text{ s}^{-1}$. Over the NH Atlantic, the maximum IGT values for CO (0.012 to $0.020 \text{ kg m}^{-1} \text{ s}^{-1}$) occurred during DJF. In the SH, IGT values are generally lower, limited to $0.015 \text{ kg m}^{-1} \text{ s}^{-1}$ for O_3 and CO, and $0.010 \times 10^{-2} \text{ kg m}^{-1} \text{ s}^{-1}$ for PAN.

The IGT distributions reflect the temporal variations in both wind patterns and emission sources. Identifying the sources and spatial distribution of these precursor emissions provides essential information on the IGT of each species. CO and NO_x are primarily emitted from combustion processes and originate from natural and anthropogenic sources. Figure S1 shows emissions estimated from TCR-2 data for the period 2005–2019. CO and NO_x show distinct global patterns, with the highest concentrations found in eastern China (CO: $0.8 - 1 \times 10^{-8} \text{ kgCOm}^{-2} \text{ s}^{-1}$, NO_x : $1.5 - 3 \times 10^{-10} \text{ kgNm}^{-2} \text{ s}^{-1}$), South Asia (CO: $0.1 - 0.3 \times 10^{-10} \text{ kgCOm}^{-2} \text{ s}^{-1}$, NO_x : $0.3 - 0.5 \times 10^{-10} \text{ kgNm}^{-2} \text{ s}^{-1}$), and the central Africa (CO: $0.1 - 0.8 \times 10^{-8}$

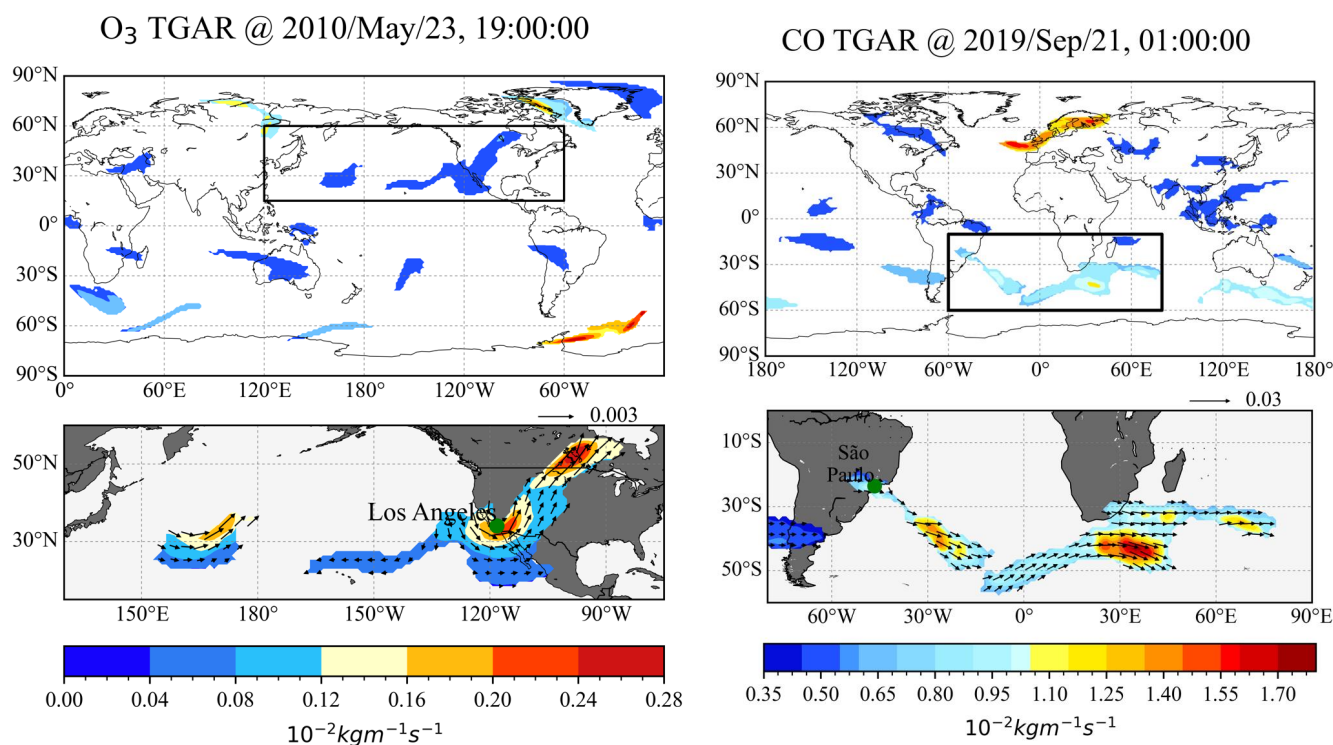


Figure 2. Snapshots of O_3 and CO TGARs events. (Left) O_3 TGARs during an episodic event on May 23, 2010, illustrating LRT from Asia to the western USA. (Right) CO TGARs originating from Amazon fires on September 21, 2019. Green dots indicate Los Angeles, USA, and São Paulo, Brazil, respectively. The rectangular black box in the global map (top panels) highlights the zoomed-in region of the TGARs object, shown in the bottom panels. The shaded colors represent the IGT magnitude ($\text{kg m}^{-1} \text{s}^{-1}$). The black arrows indicate horizontal IGT vectors.

$\text{kgCO m}^{-2} \text{s}^{-1}$, NO_x : $0.2 - 0.8 \times 10^{-10} \text{ kgNm}^{-2} \text{s}^{-1}$). The highest IGT values are typically found downwind of major emission sources, particularly over the Atlantic and Pacific Oceans, where pollutants are transported by the prevailing westerly jet. Emissions from China contribute significantly to the transport of pollutants (e.g., O_3) and precursors (e.g., CO and PAN) across the Pacific. Similarly, emissions from the USA drive the transport of pollutants across the Atlantic. Integrating the analysis of emissions and concentrations is essential for a comprehensive understanding of air pollution transport patterns and their long-term variability.

Figure 5 illustrates the climatology of TGARs frequency, defined as the number of TGARs detections at each location, with significant events (>10 days per 3 months) over the Pacific, Atlantic, and SH mid-latitudes. For O_3 , the highest frequencies of TGARs events are found at low latitudes, including the North and South Atlantic during DJF and MAM (10–18 days per 3 months), and over Asia and the Pacific during DJF (15–20 days per 3 months). CO TGARs peak in DJF over the Pacific (15–20 days per 3 months), Atlantic (10–15 days per 3 months), and Asia (15–18 days per 3 months). Frequent CO TGARs

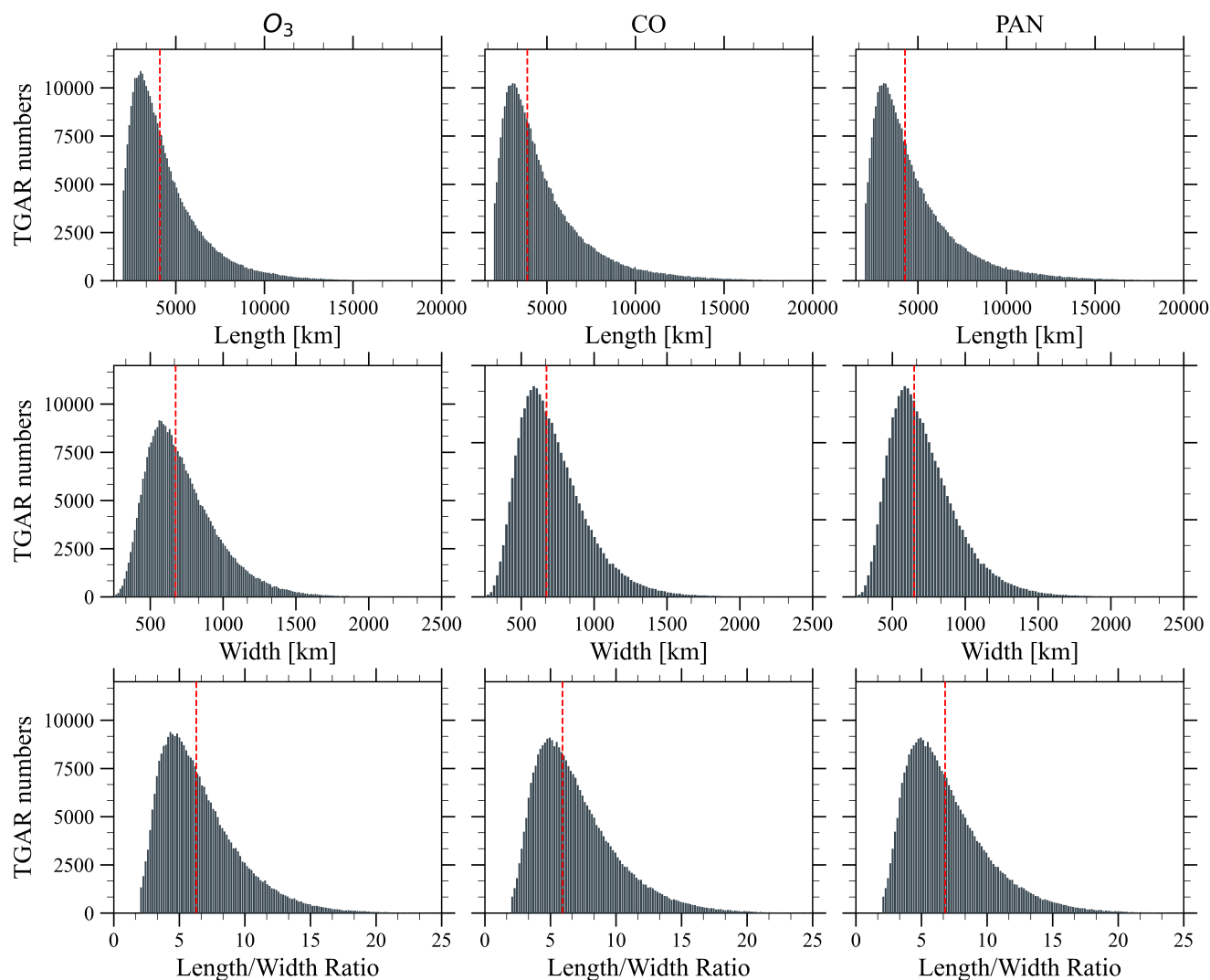


Figure 3. Histograms showing the total number of TGARs (y-axis) against three key parameters: length (km), width (km), and length/width ratio (x-axis) over the 14 years (2005–2019). The red dotted vertical line in each panel represents the median of each parameter.

also occur downwind of biomass burning regions, such as the Amazon during DJF and JJA (14–18 days per 3 months), central
 170 Africa in DJF (15–20 days per 3 months), and Southeast Asian tropical forests in DJF (12–18 days per 3 months). CO TGARs
 associated with biomass burning are common in remote SH regions (South America, Australia, and Africa), with frequencies
 of 10–18 days per 3 months. For PAN, the highest frequencies are observed in the South Atlantic and Middle East during DJF
 (15–20 days per 3 months), the North Atlantic (10–20 days per 3 months), and Indian Ocean (10–15 days per 3 months) during
 JJA, and the SH (15–20 days per 3 months) during SON. Over the Pacific, PAN TGARs occur for 5–15 days per 3 months.

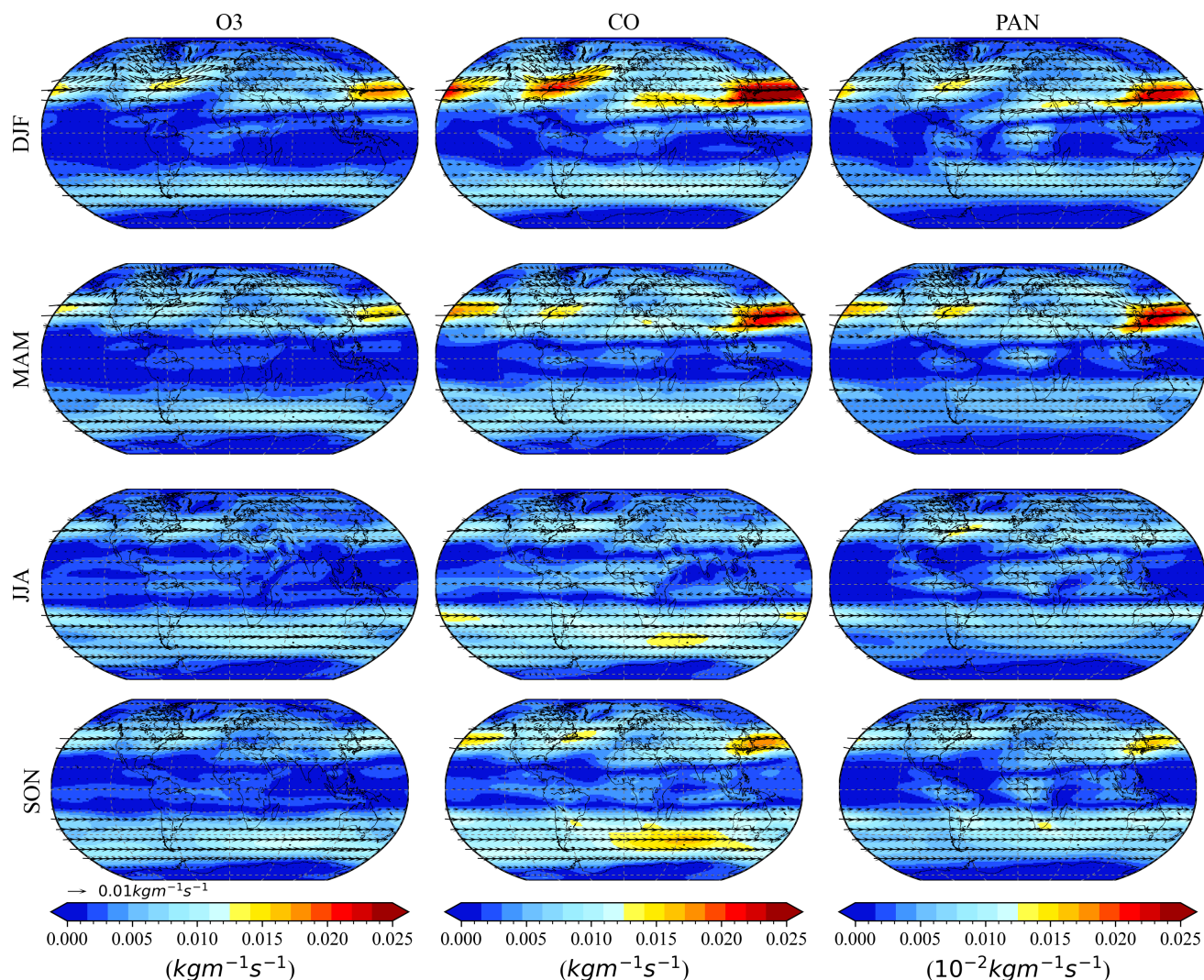


Figure 4. Global maps of seasonal IGT for O₃, CO, and PAN (left to right columns) averaged over the 2005-2019 period in December-February (DJF), March-May (MAM), June-August (JJA), and September-November (SON). The shaded color represents the IGT values. The black arrow vectors represent the direction of IGT.

175 3.2 Vertical structure of trace gases and transport

As discussed in Section 2.1, the three chemical species exhibit distinct vertical profiles (Figure 1). Unlike tropospheric O₃, which increases with altitude, CO concentrations generally peak in the lower troposphere and decrease rapidly above the upper troposphere. PAN concentrations are highest in the middle and upper troposphere due to the longer lifetime of PAN in the colder temperatures of the free troposphere compared to the lower troposphere. Since IGT estimates are based on vertical

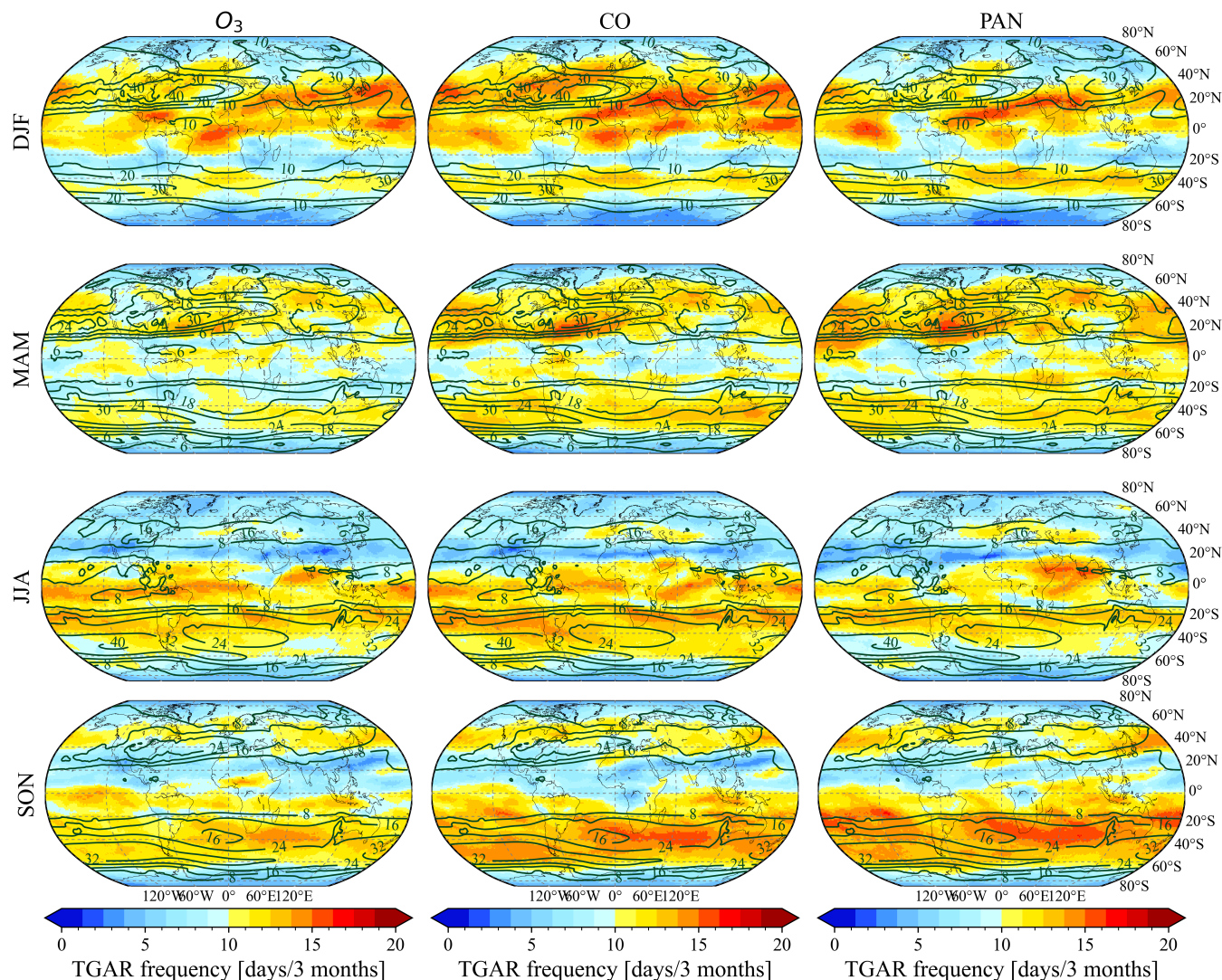


Figure 5. Global seasonal map of TGARs frequency during DJF, MAM, JJA, and SON (days per 3 months) during 2005–2019, for O_3 (first column), CO (second column), and PAN (third column) represented by shaded color. Green solid lines denote the vertically averaged horizontal wind speed (ms^{-1}).

180 integrations of horizontal transport across the troposphere, evaluating the relative contributions from different vertical levels is crucial for understanding the driving mechanisms of TGARs and their species dependence.

Overall, the relative contribution of lower tropospheric transport (below 700 hPa) to total column IGT is greater in the tropics than in the subtropics and extratropics (Figure 6). In the tropics, easterly winds and monsoon circulations dominate the horizontal transport patterns near the surface, while deep convection can transport pollutants from the surface to the free troposphere. In contrast, upper tropospheric jets are less pronounced in the tropics. In the extratropics, westerly upper tropospheric

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jets are prominent and associated with synoptic circulation and conveyor belt uplifting. In the tropics, the lower tropospheric contribution accounts for 60–80% of the total transport for CO and 40–70% for O₃ and PAN. In both the tropics and extratropics, CO has the largest lower tropospheric contribution of the three species due to the lack of significant chemical sources in the free troposphere, except for minor production from CH₄ and VOCs oxidation. In contrast, O₃ and PAN are chemically produced within the atmosphere, leading to a larger contribution of free tropospheric transport to their total IGT.

For PAN, the lower tropospheric contribution reaches 50–70% over certain tropical regions, such as Central America and equatorial Africa in DJF, and over central South America and Southern Africa in SON. These increases are consistent with biomass burning areas where PAN is not emitted directly but rapidly forms in plumes rich in NO_x and VOCs. For O₃, the lower tropospheric contribution is largest between 20°S and 20°N, with peaks over Central America and South Asia during DJF and over the central and eastern Pacific during DJF and MAM (60–70%). In the NH high latitudes, the lower tropospheric contribution for PAN is relatively higher than for other species during DJF and MAM (20–40%). This is likely due to low temperatures and poleward transport from the NH mid-latitudes.

3.3 Contribution of TGARs events to total transport

Regardless of the presence or absence of TGARs, winds transport atmospheric constituents regionally and globally. As shown in the upper panels of Figure 7, the total transport IGT, which includes contributions from both TGARs and non-TGARs events, reflects climatological wind patterns and emission distributions. Significant values are observed over the Atlantic, Pacific, and SH mid-latitudes, with different patterns and magnitudes compared to the TGARs-specific IGT patterns shown in Figure 4. The largest annual IGT values for total transport are found over the Pacific, reaching $30\text{--}40 \times 10^4 \text{ kg m}^{-1} \text{ yr}^{-1}$ for O₃, $40\text{--}60 \times 10^4 \text{ kg m}^{-1} \text{ yr}^{-1}$ for CO, and $50\text{--}60 \times 10^2 \text{ kg m}^{-1} \text{ yr}^{-1}$ for PAN. Large total IGT values are also found over the Atlantic, with $20\text{--}35 \times 10^4 \text{ kg m}^{-1} \text{ yr}^{-1}$ for O₃, $20\text{--}50 \times 10^4 \text{ kg m}^{-1} \text{ yr}^{-1}$ for CO, and $30\text{--}40 \times 10^2 \text{ kg m}^{-1} \text{ yr}^{-1}$ for PAN.

As shown in the bottom panel of Figure 7, TGARs events account for about 40–60% of the total transport in these regions, highlighting their critical role in explaining variations in tropospheric composition, especially short-term rapid enhancements. However, the significant remaining contributions from non-TGARs transport processes indicate that background transport mechanisms also play an essential role in regional air pollutant transport. The relative contribution of TGARs events to total transport varies by species and region, ranging from 40–60% in the NH Pacific, 30–55% in the NH Atlantic, and 30–60% over the SH mid-latitudes. In the tropics, total IGT transport is smaller ($5\text{--}25 \times 10^4 \text{ kg m}^{-1} \text{ yr}^{-1}$), but TGARs events contribute over 60% of the total transport in regions such as northern South America, central Africa, and Southeast Asia.

3.4 Interannual variability and long-term changes

Figure 8 shows the time series of the TGARs analysis results for the 2005–2019 period. The vertically and regionally averaged O₃ concentration is about 40–70 ppb in the NH (30°N–90°N), which is higher than the values in the tropics (30°N–30°S) and the SH (30°S–90°S), where the concentrations are limited to 45 ppb. Similarly, CO concentrations range from 70–100 ppb in the tropics and NH, while remaining below 70 ppb in the SH. For PAN, the latitudinal variation is more pronounced, with concentrations in the NH ranging from 100 to 250 ppt, followed by the tropics (100–150 ppt), and the SH showing much

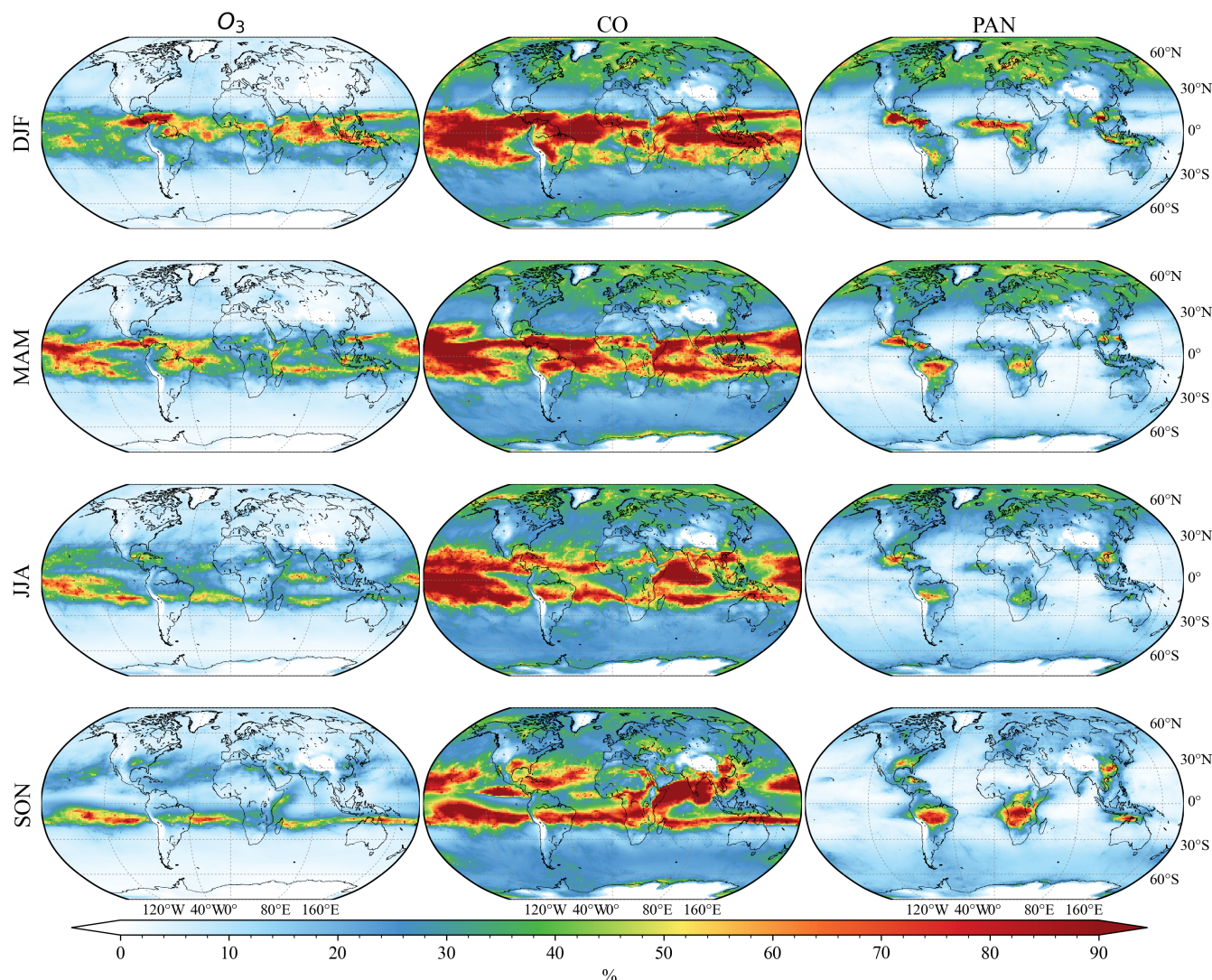


Figure 6. Spatial map of the relative contribution of the near-surface (1000–700 hPa) horizontal transport to the total tropospheric IGT for O_3 (1st column), CO (2nd column), and PAN (3rd column) for each season.

lower values, typically below 80 ppt. The NH shows slight increases in ozone and decreases in CO over the 2005–2019 period, consistent with trends reported in in-situ and satellite observations (Buchholz et al., 2021; Gaudel et al., 2024).

As shown in the right panels of Figure 8, the regional mean IGT values for O_3 have maxima of $0.01\text{--}0.025 \text{ kg m}^{-1} \text{ s}^{-1}$ in the NH and minima of less than $0.005 \text{ kg m}^{-1} \text{ s}^{-1}$ in the tropics. The CO IGT values are large across the NH and SH, ranging from 0.005 to $0.01 \text{ kg m}^{-1} \text{ s}^{-1}$, while they are small in the tropics ($<0.005 \text{ kg m}^{-1} \text{ s}^{-1}$). PAN transport is strongest in the NH ($0.05\text{--}0.09 \times 10^{-2} \text{ kg m}^{-1} \text{ s}^{-1}$), followed by the SH and the tropics ($0.03\text{--}0.05 \times 10^{-2} \text{ kg m}^{-1} \text{ s}^{-1}$). Overall, the regional

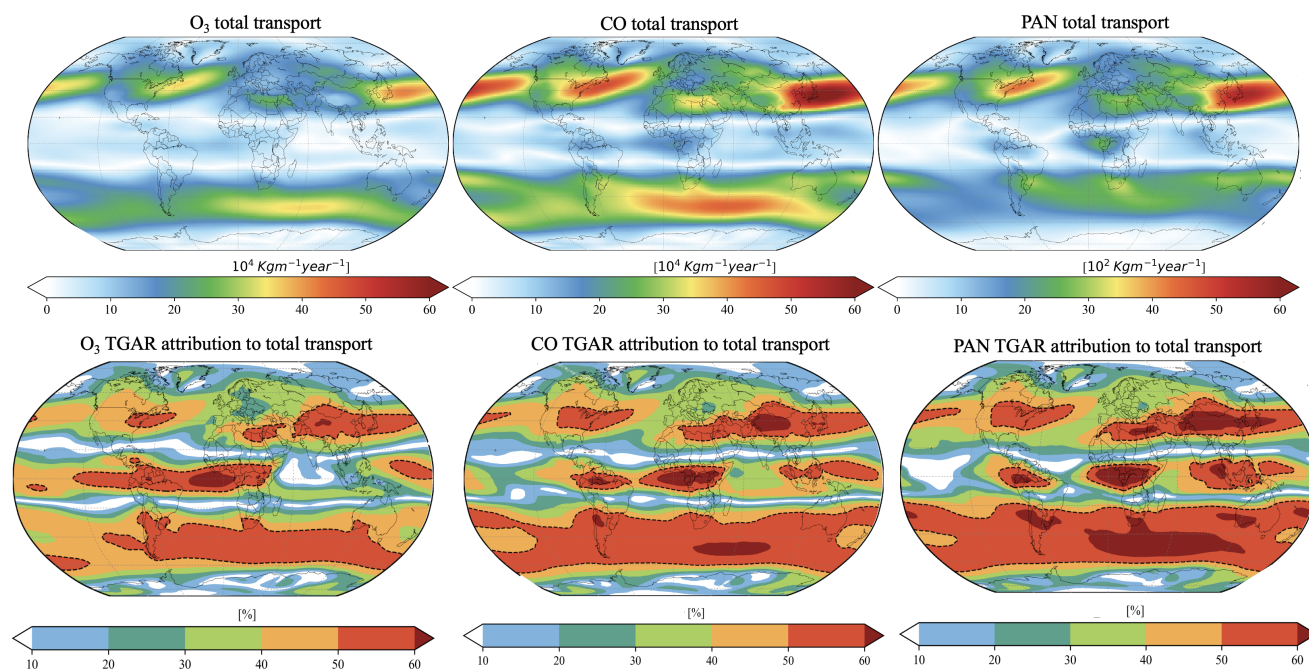


Figure 7. Spatial maps of annual IGT values for total transport (TGARs and non-TGAR events) (in $\text{kg m}^{-1} \text{yr}^{-1}$, upper panels) and the relative contribution of TGARs to total transport (in %, lower panels). From left to right, the results are shown for O₃, CO, and PAN, covering 2005–2019.

mean IGT values indicate that the primary transport of O₃, CO, and PAN occurs predominantly in the NH, followed by the SH. Meanwhile, the results show different long-term trends between concentrations and IGT. For example, IGT values for PAN in the NH show a positive trend on a regional scale, whereas no corresponding trend is found for PAN concentrations.

Figure 9 illustrates the detailed spatial patterns of the estimated annual mean concentrations and IGT trends from 2005 to 2019. Linear trends and their significance at the 95% confidence level were determined. In general, tropospheric O₃ shows an increasing trend over most regions in TCR-2, consistent with the results of previous studies using satellite data (Cooper et al., 2014; Gaudel et al., 2018) and ground-based observations (Gaudel et al., 2018; Tarasick et al., 2019; Wang et al., 2022). Numerous studies point to the regional representativeness of available observations as a significant source of uncertainty in trend analyses Wang et al. (2022). TCR-2 mitigates this by providing comprehensive records of global composition distributions, although its performance may still be limited by the limited availability of assimilated measurements (Miyazaki et al., 2019, 2020). The strongest increasing trends in tropospheric O₃ concentrations are observed over the NH high latitudes (0.1–0.5 ppb yr⁻¹), followed by North and South America (0.1–0.3 ppb yr⁻¹), northern Africa (0.05–0.2 ppb yr⁻¹), the Pacific (0.05–0.1 ppb yr⁻¹), and the Atlantic (0.05–0.15 ppb yr⁻¹). In contrast, slightly decreasing trends (–0.05 to –0.15 ppb yr⁻¹) are observed over the SH, central Africa, the Indian Ocean, the Southern Pacific, and the Tibetan Plateau. These regional O₃

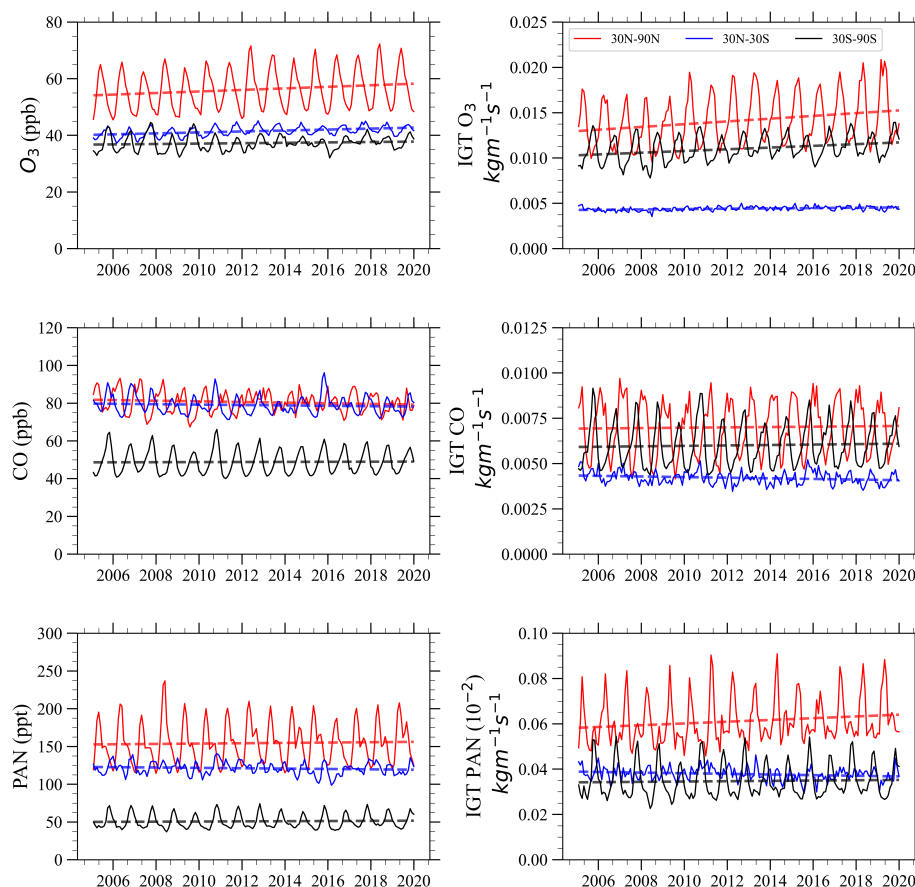


Figure 8. Time series of regional mean concentrations of O_3 , CO, and PAN (first column) and IGT values (second column). The solid lines represent concentrations and IGT for different latitudinal regions: red for 30°N – 90°N , blue for 30°N – 30°S , and black for 30°S – 90°S . The dashed lines indicate the linear trend lines for the respective regions.

variations are likely influenced by regional differences in factors such as precursor emissions and transport patterns (Parrish
 et al., 2013; Cooper et al., 2014; Archibald et al., 2020; Elshorbany et al., 2024).

A significant decreasing trend in CO concentrations was found in several regions, including East Asia (-0.6 to -0.4 ppb yr^{-1}), Europe (-0.4 to -0.1 ppb yr^{-1}), the eastern USA (-0.3 to -0.2 ppb yr^{-1}), South America (-0.6 to -0.2 ppb yr^{-1}), and central Africa (-0.2 to -0.5 ppb yr^{-1}) (Figure 9). These decreases are likely related to reduced emissions from the transportation and residential sectors in the USA and Europe and the industrial and residential sectors in East Asia, improved combustion
 efficiency in China, and changes in biomass burning patterns in Southeast Asia and Africa (Worden et al., 2013; Zheng et al.,
 2019; Zhang et al., 2020; Buchholz et al., 2021; Miyazaki et al., 2022). In contrast, increasing CO trends were found in other
 regions, including the Canadian boreal forests (0.1 – 0.3 ppb yr^{-1}), the Siberian taiga (0.2 – 0.4 ppb yr^{-1}), and Southeast Asia

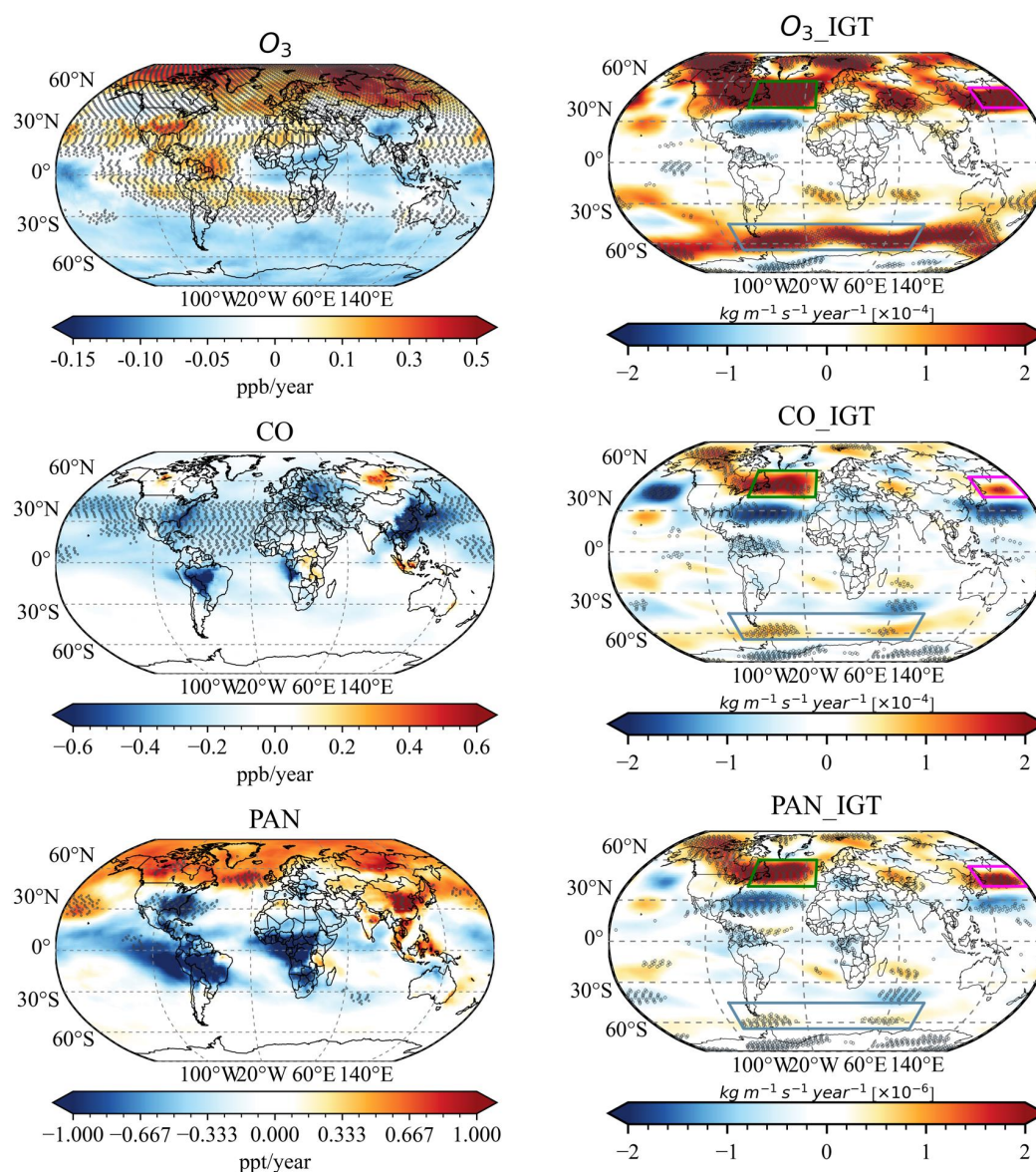


Figure 9. Spatial maps of annual trends in concentration (first column) and IGT (second column) for O₃, CO, and PAN during 2005–2019 period. Black dots indicate statistically significant trends at $p < 0.05$. Rectangular boxes highlight regions with strong IGT variability, with magenta for the Pacific, green for the Atlantic, and blue for the SH, which are the primary focus of Figure 1.

(0.4 to 0.6 ppb yr⁻¹). These increasing trends are consistent with the results of previous studies (Li and Liu, 2011; Zheng et al., 2019; Buchholz et al., 2021; Hedelius et al., 2021; Yurganov and Rakitin, 2022).



250 PAN shows distinct regional variability, with significant increases in the NH high latitudes and Asia. Regionally, the largest PAN increases are observed in eastern China ($0.8\text{--}1\text{ ppt yr}^{-1}$), India ($0.5\text{--}0.75\text{ ppt yr}^{-1}$), Southeast Asia ($0.6\text{--}1\text{ ppt yr}^{-1}$), Canada ($0.5\text{--}0.8\text{ ppt yr}^{-1}$), the Russian boreal forest ($0.2\text{--}0.9\text{ ppt yr}^{-1}$), the North Atlantic ($0.4\text{--}0.7\text{ ppt yr}^{-1}$), and the Pacific ($0.2\text{--}0.6\text{ ppt yr}^{-1}$). These patterns may be driven by regional variations in VOCs and NO_x emissions, as well as meteorological conditions. Additionally, the increasing frequency and intensity of wildfires in the NH (Talucci et al., 2022; Descals et al., 2022; Ponomarev et al., 2023) significantly impact both local and regional trends in atmospheric composition (Heald et al., 2003; Zhang et al., 2008; Lin et al., 2012). For instance, increasing NO_x emissions in China (Zhang et al., 2007; Miyazaki et al., 2022) are likely driving enhanced local photochemistry and elevated PAN concentrations (Qiu et al., 2021). These elevated PAN levels may then be transported from Asia and contribute to higher ozone concentrations over the USA (Zhai et al., 2024). In contrast, PAN concentrations show decreasing trends over central and southern Africa, the eastern USA, and northern and central South America. These trends highlight substantial changes in global PAN distributions over the past few decades, driven by a combination of anthropogenic and natural processes. Our analysis using TCR-2 complements previous studies (Payne et al., 2017; Fischer et al., 2014) that have examined global PAN distribution and their attributions. Future research could combine TCR-2 data with observations from additional long-term satellite instruments, such as those from the Cross-track Infrared Sounder (CrIS) on the SNPP and JPSS series platforms (Payne et al., 2022) and the Infrared Atmospheric Sounding Interferometer (IASI) on the MetOp platforms (Franco et al., 2018), to further investigate long-term trends in PAN concentrations and their driving factors.

The IGT trend map (Figure 9, second column) shows a significant positive trend in the mid-latitudes of both hemispheres. Within these latitude bands, the maximum positive IGT trend for O_3 ranges from $1\text{--}2 \times 10^{-4}\text{ kg m}^{-1}\text{ s}^{-1}\text{ year}^{-1}$ over the Pacific, Atlantic, and SH regions. Other regions, such as northern Canada, the Arctic, the Siberian boreal forest, and central Asia, exhibit significant positive trends. Similarly, the highest positive CO IGT trend is found over the Atlantic ($1\text{--}2 \times 10^{-4}\text{ kg m}^{-1}\text{ s}^{-1}\text{ year}^{-1}$), followed by the Pacific ($0.5\text{--}1 \times 10^{-4}\text{ kg m}^{-1}\text{ s}^{-1}\text{ year}^{-1}$) and SH ($0.2\text{--}0.5 \times 10^{-4}\text{ kg m}^{-1}\text{ s}^{-1}\text{ year}^{-1}$). In contrast, a strong negative trend in CO IGT is detected over the northern Pacific and NH subtropical Atlantic ($-2\text{ to }-1 \times 10^{-4}\text{ kg m}^{-1}\text{ s}^{-1}\text{ year}^{-1}$). For PAN, the strongest positive trends are observed over the North Atlantic ($1.5\text{--}2 \times 10^{-6}\text{ kg m}^{-1}\text{ s}^{-1}\text{ year}^{-1}$) and the Pacific ($0.5\text{--}1.5 \times 10^{-6}\text{ kg m}^{-1}\text{ s}^{-1}\text{ year}^{-1}$). Conversely, a large decreasing trend ($-2\text{ to }-1 \times 10^{-6}\text{ kg m}^{-1}\text{ s}^{-1}\text{ year}^{-1}$) is found in downwind regions of major emission sources, such as the USA and China. Overall, the IGT trends show similar patterns for the three species, although O_3 shows distinct behavior at high latitudes, likely influenced by stratospheric inflows. These results suggest that changes in circulation patterns, including TGARs frequency, are critical in driving IGT trends downstream of mid-latitude emission areas.



4 Discussion

4.1 Effect of enhanced humidity on TGARs

We also emphasize that TGARs often coincide with the transport of moist air along ARs. Humidity significantly affects the chemical production and loss of species, with effects strongly dependent on the type of trace gas or aerosol. These interactions can result in different TGARs characteristics for different trace gases and aerosols. For example, ozone chemistry is affected by increased humidity through increased production of OH radicals. For PAN, both temperature and humidity affect its decomposition, with higher humidity generally increasing OH concentrations and PAN removal efficiency. In addition, TGARs may also be accompanied by strong lightning activity, which produces NO_x , PAN, and ozone. In contrast, CO is relatively stable and less affected by water vapor during transport. As a result, high concentrations of air pollutants generated upstream of TGARs may not be fully preserved during their transport. Understanding the role of water vapor and humidity in chemical transformation and removal processes will be an important consideration in future work in evaluating the applicability of the original ARs concept to various chemical species and aerosols in the context of studying their LRT.

4.2 Impacts on local surface air quality

Assessing the relative importance of local chemical production impacts versus the remote impacts of TGARs landfall can provide valuable insights for local environmental policymaking. A significant portion of the world's population lives in coastal areas, where TGARs often reach, raising concerns about their potential impact on human health via LRT. The TGARs framework here could be used in future work to evaluate the extent that TGARs influence surface air quality in coastal and other populated areas.

4.3 Drivers of long-term trends

While this study has characterized major TGARs patterns, it is crucial to understand their role and driving mechanisms. Long-term changes in atmospheric composition are influenced by various factors, including local chemical production and loss, as well as LRT. Among these, LRT via TGAR could play a significant role in shaping trend patterns in atmospheric composition, but its relative importance has not been fully explored.

A detailed quantification of the relative importance of LRT by TGARs would provide valuable insights into the observed trends in atmospheric composition. While TGARs provide a horizontal perspective on vertical column quantities, interpreting their three-dimensional structures is essential for understanding the underlying drivers and trends. Vertical mixing, influenced by processes such as warm convection belts, links near-surface, and free tropospheric variability, while near-surface transport is shaped by local circulations. Future studies should investigate the relative contributions of different vertical layers, the exchange processes between these layers, and their impact on near-surface concentrations to provide a more comprehensive understanding of TGARs and their role in atmospheric composition trends.



4.4 Remaining uncertainty and limitation

- 310 The chemical reanalysis data (TCR-2) used in this study was produced by the assimilation of a comprehensive set of satellite data products. However, significant uncertainties remain in this dataset, especially for lower tropospheric concentrations, due to remaining model errors. Validation against independent datasets, including aircraft, satellite, and ozonesonde observations, revealed positive biases in surface O_3 over polluted regions and an underestimation of CO (Miyazaki et al., 2020). The PAN fields have so far been validated using only a limited set of aircraft measurements.
- 315 The TGARs structure may also be influenced by finer-scale atmospheric variability, which is not fully resolved in TCR-2. Improving the spatial resolution of the input data is expected to play a critical role in improving the representation of TGARs spatial patterns, especially near regions with strong wind shear and vertical transport driven by convection. While refinement of reanalysis data remains essential, satellite observations of column or partial column concentrations could provide a complementary approach to TGARs analysis if they provide dense daily spatial coverage. In the absence of detailed vertical
- 320 profiles from satellite observations, assumptions about the vertical structure would have to be made. Dense spatial maps from current and future satellite observations could significantly improve the understanding of TGARs. These data could help to delineate the spatial extent of TGARs trajectories and provide valuable insights into their regional and global characteristics.



5 Conclusions

This study developed a TGARs analysis framework, extending the concepts of water vapor ARs (Guan et al., 2010; Guan and
 325 Waliser, 2015, 2019) and Aerosol ARs (AARs) (Chakraborty et al., 2021, 2022). The framework utilized global chemical data
 assimilation products from TCR-2 for tropospheric O_3 and two key precursors, CO and PAN, over the period 2005–2019. It
 provides a novel analytical basis for studying trace gas extreme event transports, identifying global climatology and evaluating
 long-term variations in TGARs and their relative importance to total trace gas transport.

The IGT of tropospheric O_3 , CO, and PAN shows distinct geographic and seasonal patterns, with maximum IGT values
 330 occurring along the mid-latitude westerly jets, especially in the spring (MAM) season. Over the 14 years (2005–2019), more
 than 300,000 TGARs events were identified globally for O_3 , CO, and PAN. The most frequent TGARs events, occurring 10–
 20 days per three months, were observed downstream of major biomass burning regions and across the Pacific, Atlantic, and
 SH midlatitudes. These events account for 40–50% of the total transport (TGARs and non-TGARs). In the tropics, despite
 weaker IGT values compared to those in the extratropics, TGARs represent a significant long-range transport pathway, with
 335 occurrences of 5–20 days per three months contributing up to 50–60% of total transport. Conversely, TGARs play a relatively
 minor role at high latitudes. The variability in TGARs occurrence is driven by a combination of atmospheric circulation,
 precursor emissions, and chemical processes.

PAN is a critical precursor for transporting pollutants, and its distribution has been studied using satellite instruments such
 as TES, CrIS, and IASI. However, understanding of PAN distributions remains limited due to the relatively recent availability
 340 of global PAN datasets. Our analysis using the TCR-2 dataset reveals significant regional variability in PAN trends. The most
 pronounced increases in PAN concentrations are observed in East Asia, likely due to increasing VOCs and NO_x emissions.
 In contrast, substantial decreases in PAN are observed in North and South America and Europe, corresponding to decreasing
 precursor emissions. The increase in PAN concentrations over East Asia indicates enhanced photochemical production and
 regional export of pollutants, which could affect downwind regions through long-range transport. In addition, our analysis
 345 highlights the notable increase in tropospheric O_3 and decrease in CO, particularly in the NH. These changes are associated
 with evolving emissions and the influence of LRT events, underscoring the interrelated dynamics of regional and global air
 quality.

The TGARs algorithm presented in this study provides a new tool to aid in understanding the global distribution and variabil-
 ity of air pollutants, particularly their contribution to short-term extreme pollution events. When combined with in-situ surface
 350 observations, the TGARs framework can provide valuable insights into the relative contributions of LRT to near-surface air
 quality events. This is particularly important for coastal urban areas, where air quality issues present major challenges to public
 health and environmental sustainability. Future research can enhance these insights by leveraging high-resolution data from
 geostationary satellites, such as the Geostationary Environmental Monitoring Spectrometer (GEMS) and the Tropospheric
 Emissions and Pollution Monitor (TEMPO). These data can be used either directly or through integration into data assimi-
 355 lation systems to refine our understanding of TGAR dynamics with greater accuracy. Such efforts are critical for developing



targeted strategies to effectively mitigate the impacts of air pollution and for advancing air quality management practices in regions most affected by pollution transport.



Appendix A: Empirical formula to calculate the zonal and meridional transport (A1-A4), total annual transport (A5), and fraction of total annual transport (A6-A8).

$$360 \quad \overline{IGTU}(\phi, \lambda) = \frac{1}{N} \sum_{i=1}^N IGTU(\phi_i, \lambda_i) \quad (A1)$$

$$\overline{IGTV}(\phi, \lambda) = \frac{1}{N} \sum_{i=1}^N IGTU(\phi_i, \lambda_i) \quad (A2)$$

Where N is the number of times TGARs were detected at each grid point. Meanwhile, the total IGT over time T is estimated as follows:

$$365 \quad Total \overline{IGTU}(\phi, \lambda) = \int_0^T IGTU(\phi, \lambda, t) dt \quad (A3)$$

$$Total \overline{IGTV}(\phi, \lambda) = \int_0^T IGTU(\phi, \lambda, t) dt \quad (A4)$$

$$Total_annual_TGARs_IGT(\phi, \lambda) = \sqrt{(Total \overline{IGTU}(\phi, \lambda))^2 + (Total \overline{IGTV}(\phi, \lambda))^2} / 14 \quad (A5)$$

Where dt is the duration between each time step (6 h).

370 Then, we project the TGARs IGT onto the total IGT to map in the same direction. To achieve this, we used the following formula:

$$\beta_{All_events} = \tan^{-1}(Total \overline{IGTU}(\phi, \lambda) / Total \overline{IGTV}(\phi, \lambda)) \quad (A6)$$

$$\beta_{TGARs_events} = \tan^{-1}(Total \overline{IGTU}(\phi, \lambda) / Total \overline{IGTV}(\phi, \lambda)) \quad (A7)$$

375 Finally, the fraction of total annual transport accounted by TGARs events detected over a grid cell at latitude ϕ and longitude λ as follows:

$$Fraction = Total_annual_TGARs_IGT(\phi, \lambda) \times \cos(\beta) / Total_annual_IGT(\phi, \lambda) \quad (A8)$$

Where β is computed as $\beta = \beta_{All_events} - \beta_{TGARs_events}$



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DW, BG. Software: MR and BG. Visualization: MR. Original draft: preparation: MR, KM, and VHP. Writing - review, and editing: MR,
KM, VHP, BG, and DW

Competing interests. The contact author has declared that neither they nor their co-authors have any competing interest

Data availability. TCR-2 data can be obtained from <https://tes.jpl.nasa.gov/tes/chemical-reanalysis/>

385 *Code availability.* Code is available from the authors upon request.

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