Supporting Information for:
“Evaluating stratospheric tropical width using tracer concentrations”
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6. Supplemental Information

6.1 Data source

| Data Source | Temporal Resolution | Spatial resolution (lat x lon) | Vertical resolution* | Fields                      |
|-------------|---------------------|-------------------------------|----------------------|----------------------------|
|             |                     |                               | Tropopause | Stratosphere   |                              |
| FR-WACCM    | Daily               | 1.875° x 2.5°                 | 9          | 29             | [CH₄], [N₂O], \(\omega\), \(u\) |
| FR-WACCM    | Monthly             | 1.875° x 2.5°                 | 9          | 29             | [CH₄], [N₂O], [SF₆] \(\omega\), \(u\) |
| CLAES 3AT V9 | Daily              | 495km                         | 4          | 31             | [N₂O]                        |

* For the tropopause, this is the number of model levels between 70-300 hPa when \(p₀ = 1000\) hPa. For the stratosphere, this is the number of model levels between 1-200 hPa.

Table S1: Data sources used in this study.

6.2 Model description
The Community Earth System Model (CESM1) (Hurrell, 2013), is a coupled global climate model that consists of interactive atmosphere, ocean, land and sea-ice components. The atmospheric component of CESM in this paper is the Whole Atmosphere Community Climate Model (WACCM) which extends to ~140 km or 5.1 x 10⁶mb (Marsh, et al., 2013; Garcia, Smith, Kinnison, de la Camara, & Murphy, 2017), modelling atmospheric chemistry and physics from the lower troposphere to the lower thermosphere at a horizontal resolution of 1.9° latitude by 2.5° longitude. The model version used in this study has a gravity wave update to the model specification in Garcia, Smith, Kinnison, de la Camara, & Murphy (2017), with a repeating QBO and no solar variability.
6.3 Removing the anthropogenic trend by scaling

The anthropogenic increase is calculated as relative to a baseline of the temporal mean of tracer concentrations from 30N-30S at 500mb. The relative anthropogenic increase of N₂O, SF₆ and CH₄ per month is obtained by dividing the trend over 1995-2024 at 500mb, \( \frac{f_{500mb}}{N₂O_{500mb}} \), by this baseline defined as \( N₂O_{500mb} \), as follows:

\[
I_A = \frac{f_{500mb}}{N₂O_{500mb}}, \quad \text{units} = \text{month}^{-1}
\]

The anthropogenic trend is removed by scaling down each month’s stratospheric distribution, \( N₂O_{\text{strat.m}, \theta, \phi, p} \), at each \( \theta, \phi, p \). The anthropogenic increase \( I_A \) is incremented by \( \frac{n}{12} \) for each subsequent month, where \( n \) is the index for the 360 months in the time period 1995-2024:

\[
N₂O_{\text{strat,n}} = N₂O_{\text{strat,n}} \times \left(1 - \frac{n}{360} I_A\right)
\]

where \( N₂O_{\text{strat,n}} \) is month \( n \)'s stratospheric distribution without the anthropogenic trend and \( \theta, \phi, p \) are longitude, latitude, pressure. This process is illustrated in Figure S2.

We also check the trend at the mean tropical tropopause height, which reflects transport across the tropopause (Figure S3). The trend at 500mb has been chosen to remove the anthropogenic trend because it is sufficiently high up in the troposphere to not be confused with ground sources that may not escape the near-surface boundary layer.

6.4 PDF Method Description

(Sparling, 2000) and (Neu, Sparling, & Plumb, 2003) show that the subtropical edge can be located by a minimum in the PDF of tracer concentrations at each pressure level. For example, a scatter plot of tracer concentrations against latitudes at each pressure level, for all longitudes and days in the time period under consideration can be produced (see Figure S4a,c). The PDF, \( P(\chi) \), is obtained by horizontally integrating through the scatter plot. Laying the scatter plot alongside its PDF in Figure S4b,d makes clear that flat, tightly-scattered regions in the scatter plot correspond to maxima in \( P(\chi) \) and steeply sloped, loosely-scattered regions correspond to minima in \( P(\chi) \). As noted in earlier work, the PDF is multi-modal: in the winter hemisphere, the peaks correspond to the polar vortex, surf zone and tropics; the subtropical valley lies between the surf zone and tropics (Figure S4b). In the summer hemisphere, peaks in the PDF correspond to the summer midlatitude airmass and the tropics, between which the subtropical valley lies (Figure S4d).

The tracer concentrations bounding either side of the subtropical valley are demarcated as follows: we call these bounds \( \chi^* \) and delineate them with brown lines in Figure S4b,d. The observations that fall within \( \chi^* \), marked for the summer hemisphere in Figure S4c, are used to generate a second PDF \( P(\phi|\chi^*) \) by vertically integrating through the scatter plot in the range of \( \chi^* \). This PDF gives us the most probable latitude at which tracer concentrations, within the
range of $\chi^*$, exist (Figure S5). We have tested the PDF method we apply for consistency with earlier work by running it on CLAES N2O data (Roche, 1996) and find very similar results to the published study of (Sparling, 2000) (see Figure S4- S5 versus their Figure 3-4). Small discrepancies are obtained, likely because they were using Version 7 or older of the dataset, which is no longer available (we used the current version, 3AT Version 9).

6.5 Additional figures

**Figure S1** – *(a)* Illustration of the removal of the anthropogenic increase of N$_2$O at 500mb. The trend is expressed as a percentage of the temporal mean (1995-2024) of N$_2$O over the tropics, 30N-30S. The zonal-mean N$_2$O profile (blue) and the scaled down distribution with the anthropogenic increase (red), relative to a baseline at 500mb 1995-2024 from Figure S1a in: *(b)* 12-1999 and *(c)* 12-2014.

**Figure S2** – Same as Figure S1(a) but at 100mb, the pressure level of the tropical tropopause.
FIGURE S3 – The PDF method explained at 15mb: reproducing Figure 3 in (Sparling, 2000) with Version 9 of CLAES 3AT, which is the best available current dataset. The top row (ab) is the winter, southern hemisphere, August 17 – September 16 1992 and the bottom row (cd) is the summer, northern hemisphere July 19 – August 10 1992. (ac) The left panels show scatter plots of concentrations against latitude. (bd) The right panels show the probability density function (PDF), oriented horizontally to emphasise that each bar is the horizontal integration through the corresponding scatter plot in the left column. The text labels identify the key dynamical regions of the stratosphere. The summer hemisphere scatterplot in (c) illustrates the region of latitudes that fall within the tracer boundaries $\chi^*$ that have been identified from the minimum in the tracer PDF in (d).
FIGURE S4 – Latitudes at which tracer concentrations fall within the boundaries $\chi^*$ identified in Figure S3bd are used to make the corresponding PDF $P(\phi|\chi^*)$. $P(\phi|\chi^*)$ is calculated by vertically integrating the scatterplot in Figure S3ac within the tracer concentration boundaries ($\chi^*$) (Sparling, 2000). The identified subtropical edges here are at 34° N (which compares to 33.5° N found in Sparling (2000) Fig 4) in the summer hemisphere and 19° S (16° S in Sparling (2000) Fig 4) in the winter hemisphere.
FIGURE S5 - Same as Figures S3bd-4, but for October 1991.

FIGURE S6 – Distribution of (a) CH₄ and (b) N₂O concentrations at 73 mb averaged over all days in January, illustrating wave motion-induced intrusions of subtropical air into the tropics and into the midlatitudes.
Figure S7 – Ensemble-mean timeseries 1995-2025 of GWL (green) and steepest gradient widths (black) at (ab) 5mb and (de) 73mb. The ensemble standard deviation is indicated in shaded error bars. The correlation coefficients calculated on the de-seasonalised timeseries are stated in the title. Ratio of the ensemble variance of GWL widths to steepest gradient widths at (c) 5mb and (f) 73mb for the northern (red on left y-axis) and southern hemisphere (blue on right y-axis). The line where the ratio equals one has been plotted horizontally as a visual aid to determine which method has a larger ensemble variance. (g) Temporal-mean ensemble mean GWL and steepest gradient widths on a pressure x latitude plot with shaded error bars representing the ensemble standard deviation. (h) Ratio of the ensemble variance at all pressure levels between temporal-mean GWL and steepest gradient for the northern (red) and southern hemisphere (blue). The line where ratio equals one has been plotted vertically in red as a visual aid to determine which method has a larger ensemble variance.
Figure S8 – Same as Figure 4a but over the full 30-years timeseries (1995-2024). The definitions of the tropical mean used for the 1σ method are overlaid with GWL widths.
FIGURE S9 – Comparing tracer-dependent metrics against each other and between the two tracers CH₄ and N₂O for (ab) January 2005 and (cd) October 2005. Tropical widths measured by the 1σ (blue), GWL (green) and PDF (maroon) metrics for (ac) CH₄ and (bd) N₂O using two different monthly averages: mean over all days’ measured width (line-symbol) and monthly average output (dashed-line). Only widths measured above the tropical tropopause (white line) have been shown. In January (ab), monthly GWL widths at upper stratospheric pressure levels (at 1mb) are not shown as the ensemble standard deviation is very high.
**FIGURE S10** – Same as Figure 7, but for CH$_4$. 
FIGURE S11 – Annual-mean, ensemble-mean time series of modeled tropopause height-based metrics in the northern hemisphere over 1995-2024.
**FIGURE S12** – Same as Figure 10, but for the following months (in period 1995-2024): Apr-June (NH) and Oct-Dec (SH).
Bibliography for Supplemental Information

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