



An improved observation-based tropopause-relative coordinate system based on tracer climatologies

Sophie Bauchinger¹, Andreas Engel¹, Hans-Christoph Lachnitt², Andreas Zahn³, Harald Bönisch³, Peter Hoor², and Tanja J. Schuck⁴

¹Institute for Atmosphere and Environment, Goethe University Frankfurt, Frankfurt, Germany

²Institute for Atmospheric Physics, University of Mainz, Mainz, Germany

³Institute of Meteorology and Climate Research, Karlsruhe Institute of Technology, Karlsruhe, Germany

⁴Institute for Isotope Research, University of Groningen, Groningen, Netherlands

Correspondence: Sophie Bauchinger (bauchinger@iau.uni-frankfurt.de)

Abstract. Trace gas observations can be used to define chemical tropopauses based on differences between stratospheric and tropospheric characteristics. Notable examples are O_3 and N_2O , which have a well-defined positive and negative gradient in the lower stratosphere, respectively. We propose a vertical coordinate relative to the chemical tropopause that can be derived directly from trace gas measurements. Taking advantage of the extensive global coverage of regular ozone soundings, we develop an ozone-based tropopause using an observation-based climatology expressed as mixing ratios relative to the tropopause, $\Delta\Theta(O_3)$. We apply this new coordinate to two exemplary data sets in the extratropical upper troposphere and lower stratosphere. Compared to the dynamic tropopause, the short-term variability of 2D-binned N_2O is reduced by 18% and that of SF_6 by up to 38% in the two data sets. This reflects the implicit incorporation of transport and mixing processes in $\Delta\Theta(O_3)$. In both data sets, $\Delta\Theta(O_3)$ also compresses the vertical extent of observations, indicating that air masses as high $\Delta\Theta(dyn)$ are chemically more related to lower-laying air. Overall, we show that $\Delta\Theta(O_3)$ provides a chemically consistent vertical coordinate for larger observational data sets, facilitating studies of smaller-scale processes without requiring model reanalysis data.

1 Introduction

The upper troposphere and lower stratosphere (UTLS) is a highly variable region where a multitude of processes influence the chemical composition and dynamics on timescales ranging from minutes to decades. Changes in the chemical and dynamical structure of the UTLS have far-reaching implications for atmospheric composition and climate, affecting for example the transport of greenhouse gases and ozone-depleting substances, the Brewer-Dobson circulation, the residence time of trace gases in the stratosphere and the radiative properties of the atmosphere (Gettelman et al., 2011). While the observational basis in the UTLS has increased significantly in the last decades, in situ measurements in the tropopause region remain sparse for many trace gases. Sensible categorisation of these observations remains an important prerequisite for studying this region. Potential benefits include improved investigations of cross-tropopause transport, better comparisons between independent observational data sets and improved validation of chemistry-climate models.



- In the stratosphere, potential temperature (Θ) is widely used as a vertical coordinate because it eliminates effects from short-term adiabatic processes. Close to the tropopause, the relative position to the tropopause ($\Delta\Theta_{TP}$) becomes a determining factor for many local characteristics, particularly for short-lived substances (Pan et al., 2004; Hoor et al., 2004; Hegglin et al., 2006).
- 25 The location of the tropopause varies substantially in response to temperature, circulation patterns and other dynamic processes. Expressing observations in tropopause-relative coordinates therefore reduces a large fraction of this variability by grouping together air masses with similar dynamical histories. This approach has become particularly valuable in the consolidation of multiple data sets (Millán et al., 2023) and can enable the investigation of small changes in the atmospheric composition and circulation which would otherwise remain obscured.
- 30 Existing tropopause definitions capture different aspect of the troposphere-stratosphere transition. Thermal definitions characterise the vertical temperature structure of the atmosphere, while dynamical definitions based on potential vorticity (PV) or its gradient characterise the dynamical separation between tropospheric and stratospheric air (e.g. Kunz et al. (2011); Turhal et al. (2024)). More recent approaches, such as the relative-humidity-gradient method proposed by ?, provide complementary ways of identifying the tropopause region by taking into account both diabatic and adiabatic changes across the UTLS.
- 35 The calculation of $\Delta\Theta_{TP}$ for observational data sets can introduce challenges. Applying these definitions to individual aircraft measurements requires vertical profiles to evaluate gradients or the modelling of dynamical parameters such as PV. Limited temporal and spatial resolution and mismatches between modelled and observed atmospheric structures can introduce errors, including systemic biases in tropopause properties or unresolved double tropopauses (Birner et al., 2002; Xian and Homeyer, 2019; Tegtmeier et al., 2020; Meng et al., 2021).
- 40 Chemical tropopause definitions provide a fundamentally different approach by using the well defined changes in chemical composition across the tropopause region to distinguish between tropospheric and stratospheric air. Existing approaches include fixed or seasonally varying thresholds and tracer-tracer correlations, which exploit differences in chemical lifetimes and histories to characterise air mass origin and mixing (Fischer et al., 2000; Hoor et al., 2002; Pan et al., 2004; Hegglin et al., 2009). In contrast to definitions that require profile and/or model data, distinguishing tropospheric and stratospheric air masses
- 45 can then be based on individual trace gas observations.
- Only a limited number of studies combine chemical information with tropopause-relative coordinates. An example was presented by Zahn et al. (2004), who derived a seasonally varying O_3 threshold from sonde observations at Hohenpeißenberg. Building on this concept, in Sprung and Zahn (2010) a relative height coordinate referenced to the thermal tropopause using the same O_3 climatology was utilised. However, the applicability of this climatology is limited to a narrow latitudinal band, as
- 50 only data from a single station was used. A more generally applicable framework requires reference distributions that account for spatial and seasonal variability.
- Extending this approach, we construct climatological tracer distributions as a function of month, equivalent latitude and tropopause-relative potential temperature. *Equivalent latitude* (eql.) uses the adiabatic conservation properties of potential vorticity to reduce dynamical variability associated with reversible meridional excursions of air masses induced e.g. by planetary
- 55 waves. This better represents the characteristics of an air parcel along its natural transport pathways (Butchart and Remsberg,



1986). After selecting the corresponding climatological profile for a certain month and equivalent latitude, these distributions can subsequently be used to match an observed tracer measurement to the representative tropopause-relative distance, $\Delta\Theta_{TP}$.

To allow an unambiguous mapping between mixing ratio and $\Delta\Theta_{TP}$, suitable reference tracers must exhibit sufficiently distinct tropospheric and stratospheric mixing ratios and a monotonic lower stratospheric profile. The change in composition across the profile must be significantly larger than the natural variability from chemical processes and than the measurement uncertainty. We focus on the two stratospheric tracers O_3 and N_2O , which exhibit particularly suitable vertical distributions and are governed by very different processes in the lower stratosphere.

O_3 mixing ratios increase rapidly above the tropopause as a consequence of photochemical production in the stratosphere. The extensive global record of O_3 soundings provides a robust statistical basis for constructing a climatology. However, O_3 distributions exhibit a pronounced seasonality and latitude dependence, while processes such as stratosphere-troposphere exchange, the polar vortex and ozone depletion introduce additional variability (Bethan et al., 1996; Hegglin et al., 2009; Petetin et al., 2016). These effects limit the applicability of fixed threshold values and motivate a climatology-based approach (Zahn et al., 2004; Thouret et al., 2006).

N_2O , in contrast, is emitted predominantly at the Earth's surface with a broad distribution of emissions and has a long lifetime of 116 ± 9 years (Prather et al., 2015; SPARC, 2013). It is therefore relatively well mixed in the troposphere, while photolysis in the middle and upper stratosphere leads to decreasing mixing ratios with altitude. In principle, the comparatively simple lower stratospheric distribution of N_2O makes it particularly well suited for deriving a tropopause-relative coordinate. However, N_2O is also generally harder to measure with the precision and accuracy required and considerably fewer measurements exist in the UTLS. The increases in tropospheric background values also need to be taken into account for longer observation periods.

Together, these tracers provide complementary test cases. O_3 offers excellent observational coverage, whereas N_2O provides a chemically simpler distribution with an oppositely signed gradient. Demonstrating that the proposed framework is applicable to both suggests a general applicability of this method to tracers with different chemical characteristics.

In this study, we introduce a tropopause coordinate that combines the advantages of tropopause-relative potential temperature coordinates with the chemical information derived from observations. We construct climatologies of O_3 and N_2O expressed relative to the dynamical 3.5 PVU tropopause, representing typical tracer distributions as a function of equivalent latitude, month, and tropopause-relative potential temperature. By comparing a individual observations to the corresponding climatological profile, we derive the $\Delta\Theta_{TP}$ that is most representative of the measured tracer mixing ratio. This coordinate can subsequently be used equivalently to conventional tropopause-relative coordinates without requiring model-derived tropopause information for each individual measurement. We characterise the resulting climatologies and demonstrate the application of $\Delta\Theta(O_3)$ to observational data sets.

Section 2 describes the observational data used to construct and apply the climatologies. Section 3 presents the methodology and characterises the resulting climatologies, while Section 4 discusses the application of $\Delta\Theta(O_3)$ to observational data sets. The final section summarises the findings and conclusions.



2 Global measurement data

- 90 Observations of O_3 and N_2O strongly differ in their availability and global coverage. O_3 is measured regularly by ozone sondes launched across the world, along with broad coverage through aircraft observations and satellite data. N_2O has primarily been measured during research campaigns, leading to limited data availability in space and time. We choose to simplify the O_3 data set by focussing solely on O_3 sondes, while for N_2O we will be using both balloon and aircraft campaign data due to the sparsity of global coverage otherwise.
- 95 The trace gas data is combined with ERA5 reanalysis data with an initial resolution of 6 hours, 1° horizontally and 137 vertical levels corresponding to an approximate vertical spacing of 500 m (Hersbach et al., 2020). For each ascent / flight, meteorological parameters were interpolated onto the measurement position and time using potential temperature mapping. For each observation, these include the equivalent latitude of the measurement, as well as the potential temperature of the local thermal and dynamical (3.5 PVU surface) tropopause. The potential temperature distance to the tropopause, $\Delta\Theta_{TP}$,
100 is then calculated as the difference between the potential temperature of the observation and of the local tropopause at the corresponding coordinates. Interpolated reanalysis data have been made available by Lachnitt (2025).

2.1 Ozone sonde data

- Numerous stations across the world carry out regular ozone sonde observations, many of which are accessible through the World Ozone and Ultraviolet Data Center (WOUDC) (WMO/GAW Ozone Monitoring Community, 2025). To create a global
105 climatology, we use data from 31 stations across all latitudes from 1980 to 2023, chosen based on the availability of consistent time series over multiple decades. The temporal availability varies depending on the station, which is shown in detail together with the geographical distribution in Appendix A. We evaluate the O_3 mixing ratio in ppb by relating the partial pressure in mPa over air pressure in hPa multiplied by a factor of 1.656×10^4 .

- The ozone sonde measurement distribution is shown as a zonal mean over equivalent latitude in the upper panel of Fig. 1.
110 The figure shows that stratospheric measurements are unevenly distributed across latitude. There is a clear maximum in measurement density in the northern hemispheric mid-latitudes.

2.2 Nitrous oxide data

- For the last decades, measurements of N_2O were largely limited to whole air samples, cryogenic samplers and the deployment of gas chromatographs and tunable diode laser instruments onboard sporadic research aircraft missions. Instruments capable
115 of measuring N_2O continuously at high frequency have now become more common, however the global availability of high-resolution data is limited (Lebegue et al., 2016). An overview of N_2O observations from 1987–2018 has been previously given by Krysztofiak et al. (2023b). The data used in this study are in large parts based on this data set and excludes particularly dynamic cases and outliers. We extend the dataset by the full time period of IAGOS CARIBIC-2 N_2O flask measurements analysed by a GC-ECD (Gas Chromatograph - Electron Capture Detector) setup (Schuck and Obersteiner, 2024), as well as

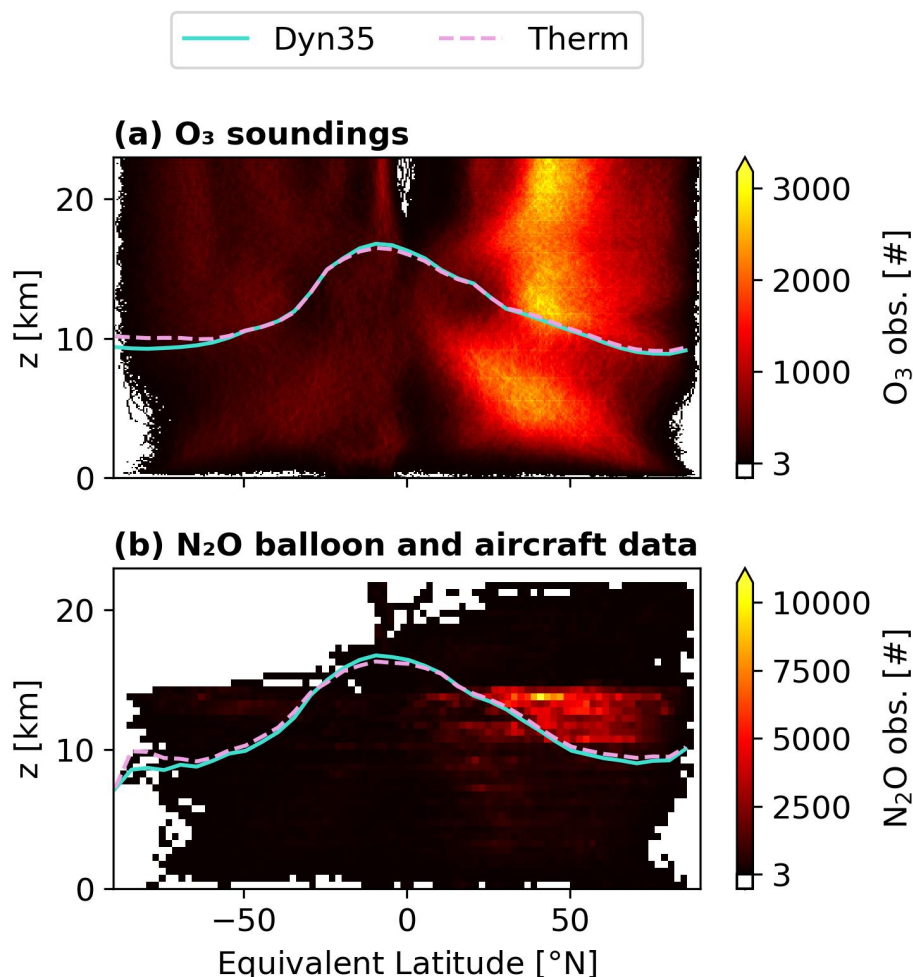


Figure 1. Global observational density for O_3 and N_2O . (a) O_3 data from soundings in 0.5° equivalent latitude (eql) and 100 m altitude (z) bins. (b) N_2O from balloon and aircraft in bins of 2° in eql. and 200 m in z . The lines indicate the mean altitude of the dynamic tropopause / 3.5 PVU surface (solid) and thermal (dashed) tropopause for the observations included. In the tropics, the procedure outlined by Sprenger et al. (2003) is used to evaluate a dynamical tropopause in regions where the dynamical tropopause is not well defined / above the 380 K isentrope.

120 additional data from research missions SouthTrac (HALO, 2021) and PHILEAS (HALO, 2025) aboard the German research aircraft HALO. Appendix B outlines all campaigns and relevant instrumentations that are used in the present study.

The overall increase in atmospheric abundance and specifically tropospheric background values of N_2O need to be accounted for when analysing longer observational time periods. The background reference is obtained by fitting a trend line to global monthly mean N_2O data from NOAA/GML (Dutton et al., 2022). We detrend N_2O measurements by dividing by the tropo-



125 spheric background at the time of observation to obtain N_2O *rel. to BGD* [ppb/ppb]. Pollution events then result in values > 1
 and air in the stratosphere < 1 , decreasing with altitude.

Most N_2O observations are located in the northern hemisphere, primarily over North America and large parts of Europe. However, the total data available is limited. This leads to poor coverage especially in the southern hemisphere, which can be seen clearly in panel (b) of Fig. 1. Additionally, most campaigns in this data set do not reach far into the stratosphere, making
 130 stratospheric observations of N_2O rare. With most measurements originating from balloon and aircraft campaigns investigating specific processes, the data set may additionally be biased towards less typical states of the atmosphere.

2.3 Independent aircraft data

We later apply the newly derived chemical tropopause based on O_3 to independent data sets. The O_3 climatology is based purely on sounding data, therefore independent data sets includes the IAGOS-CARIBIC project and the PHILEAS campaign onboard
 135 the HALO aircraft. We use N_2O , O_3 and SF_6 as test substances from each of these. During CARIBIC (Civil Aircraft for the Regular Investigation of the Atmosphere Based on an Instrument Container), the N_2O and SF_6 measurements are obtained from whole air samples taken every 30 to 60 minutes and subsequent laboratory-based analyses using a GC-ECD (data see Schuck and Obersteiner (2024), for the measurement methods see Schuck et al. (2009)). O_3 observations during CARIBIC-2 are obtained by the Fast Airborne Ozone instrument (FAIRO), a UV-photometer with a time series resolution of 0.25 Hz (for
 140 data see Zahn et al. (2024), for meta data Obersteiner (2024)), integrated over whole air sampling times.

The HALO campaign PHILEAS (Probing High Latitude Export of air from the Asian Summer Monsoon) took place in August/September 2023 with data available upon request via the HALO database (HALO, 2025). The campaign comprised eleven scientific flights around north-western America and the northern Pacific, several flights in Europe, and two transfer flights from central Europe during August and September 2023. High-resolution in situ measurements of SF_6 , N_2O and O_3 are
 145 available throughout these flights. N_2O was measured with a time resolution of 1 s by the University of Mainz airborne QCL-spectrometer instrument (UMAQS) (Müller, 2015) and O_3 by the FAIRO instrument. SF_6 was measured with a time resolution of 1 min on the ECD channel of the Gas chromatograph for Observation of Stratospheric Tracers (GhOST). Similarly to N_2O , SF_6 data were detrended referencing a tropospheric background time series from NOAA/GML (Dutton et al., 2017). While the reference data and SF_6 from CARIBIC air samples are reported on the WMO X2014 scale, data from the GhOST instrument
 150 are reported on the SIO-05 scale. We therefore apply a conversion factor of 1.0049 ± 0.002 (WMO/SIO ratio) to make absolute numbers comparable (Prinn et al., 2018).

3 Methods

3.1 Constructing climatological reference profiles

To create a climatology that can serve as the basis for chemical tropopause definition, we first calculate the “typical” profiles of
 155 O_3 and N_2O relative to a reference tropopause. In a previous analysis we found the dynamical tropopause at the 3.5 PVU surface



to best describe the chemical composition change across the extratropical tropopause (Bauchinger et al., 2025). Calculations were additionally carried out in reference to the thermal tropopause as defined by the WMO (1957), however this analysis will focus on the dynamic tropopause only. Leveraging the benefits of potential temperature as vertical coordinate in the stratosphere, we therefore express the climatology in terms of potential temperature distance to the 3.5 PVU surface, $\Delta\Theta(\text{dyn})$.

160 Trace gas observations are combined with ERA5 reanalysis data, including the corresponding equivalent latitude and dynamical tropopause information. $\Delta\Theta_{TP}$ is calculated as the difference between the potential temperature of the observation and the local tropopause. The observations are then separated into equivalent latitude ranges and months before being binned vertically into 5 K intervals of $\Delta\Theta(\text{dyn})$. Statistics are calculated only for bins containing at least three observations. As the resolution of measurements is not uniform across campaigns and instruments, we apply a weighted binning algorithm to avoid
 165 bias towards highly resolved data sets. Each measurement is weighted by the inverse of the number of observations from the same flight or balloon ascent within a given bin. This ensures that densely sampled individual profiles do not disproportionately influence the climatological mean.

The seasonal and latitudinal dependence of O_3 and N_2O mixing ratios requires an appropriately resolved climatology. For O_3 , sufficient observations are available to construct monthly climatologies for 10° equivalent latitude bands. As broadly
 170 distributed emissions of N_2O lead to only a small hemispheric difference (Ishijima et al., 2010) and with the overall scarcity of the data set, we choose to construct a monthly climatology for 30° equivalent latitude bands for N_2O . For both substances, the analysis is restricted to the extratropics (i.e. polewards of 30° equivalent latitude), where data coverage reaches deeper into the stratosphere, the higher number of ozone sonde stations provide a better statistical basis and the dynamical tropopause is well defined.

175 Figure 2 graphically shows how observations of O_3 can be used to derive a value of $\Delta\Theta(\text{O}_3)$. A measurement of O_3 from an independent data set is compared to a reference profile of climatological O_3 versus $\Delta\Theta(\text{dyn})$ within the same month and latitudinal band. The algorithm then returns an interpolated value of $\Delta\Theta$ that corresponds to the particular measurement given. For regions and times without suitable reference profiles, a NaN value is returned. For O_3 values below/above the minimum/maximum O_3 mixing ratios included in the climatology, $\Delta\Theta(\text{O}_3)$ can also not be calculated, however a flag indicating
 180 tropospheric/stratospheric characteristics of the measurement is returned. With a suitable climatology, this methodology can be used with different reference tropopause definitions and tracer substances. In addition to the dynamic 3.5 PVU tropopause, we created a climatology referencing the thermal tropopause as supplementary data. Python code for the application of this tropopause can also be found in the supplementary data.

3.2 N_2O climatology

185 To create a climatological reference, N_2O data from relevant balloon and aircraft missions were combined and the weighted mean profiles evaluated per month and 30° equivalent latitude range. Figure 3 shows the monthly mean profiles over $\Delta\Theta(\text{dyn})$ in the northern and southern mid-latitudes in Panels (a) and (c), respectively. The difference in data coverage can be seen here through data gaps in the southern hemispheric profiles. Profiles for the remaining equivalent latitude ranges are shown in Appendix C, further visualising the data scarcity in the southern hemispheric stratosphere.

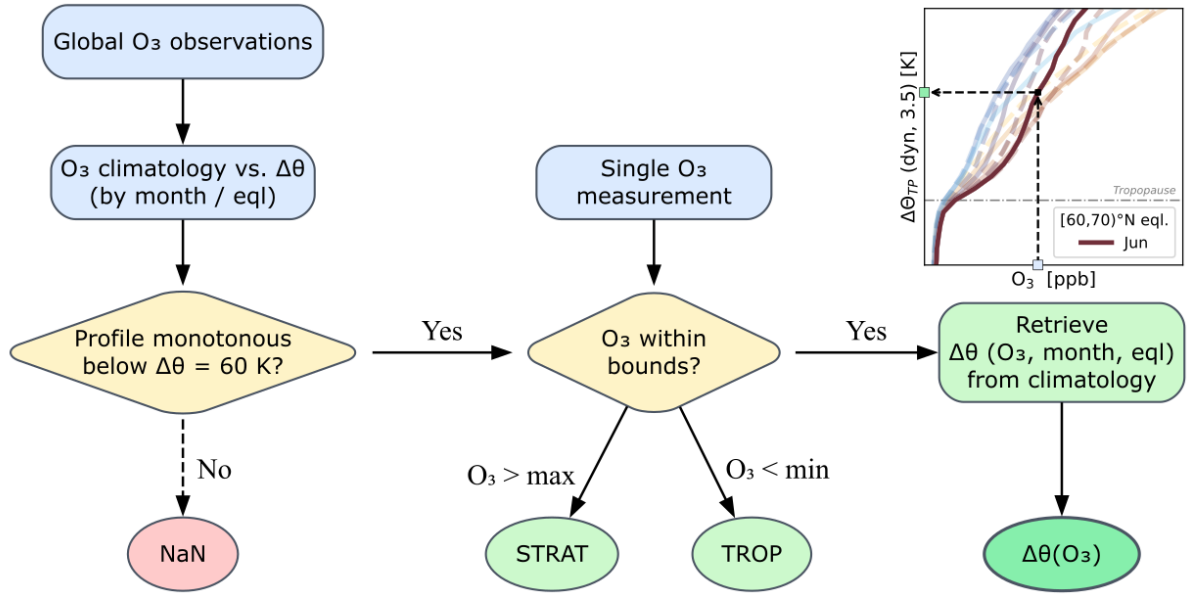


Figure 2. Illustration of the O_3 climatology creation and application to observations. Sonde observations are used to create a climatology of O_3 versus $\Delta\theta(dyn)$, which then serves as a reference to calculate $\Delta\theta(O_3)$. The top right shows how a measurement of $O_3 = 930$ ppb in June at an equivalent latitude between $60\text{--}70^\circ\text{N}$ leads to a derived $\Delta\theta(O_3) = 45$ K by referencing the climatological profile.

190 Referencing the discussions of processes governing the N_2O distributions in the stratosphere, we expect N_2O mixing ratios to monotonically decrease with $\Delta\theta$ in the stratosphere. However, the climatological profiles evaluated from the available data set show significant irregularities in every equivalent latitude range. The smoothest profiles are available in the northern hemispheric mid-latitudes, where most measurements are located. This behaviour is consistent across different coordinate systems: θ , pressure, altitude, $\Delta\theta(therm)$ or $\Delta\theta(dyn)$ with varying thresholds and with smaller/larger equivalent latitude
 195 ranges all show these irregularities.

As the derivation of $\Delta\theta(N_2O)$ requires monotonic behaviour in the stratosphere, we cannot use irregular profiles directly. We fit the data to a suitable function exhibiting the required behaviour, choosing a negative hyperbolic tangent of the form

$$[N_2O] = a + b \times \tanh(-\Delta\theta \times c + d), \quad (1)$$

with fit parameters a, b, c and d . This function was chosen for its monotonic decreasing behaviour with increasing $\Delta\theta$, and
 200 its limits of ± 1 as $\Delta\theta \mapsto \mp\infty$. In panels (b) and (d) of Fig. 3, the resulting fits are shown for mid-latitudinal data. Months without a valid fit are indicated by a diamond in the legend. In these cases there is either not enough data, the fitting function does not converge or the result is not of sufficient quality ($R^2 < 0.8$).

The gradient varies depending on the month, with stronger lower-stratospheric gradients during warmer months. In the northern hemispheric mid-latitudes, a gradual transition between months can be seen clearly, however this relationship cannot
 205 be investigated in other regions due to data gaps. Among the fitted profiles, the strongest gradients appear in the southern

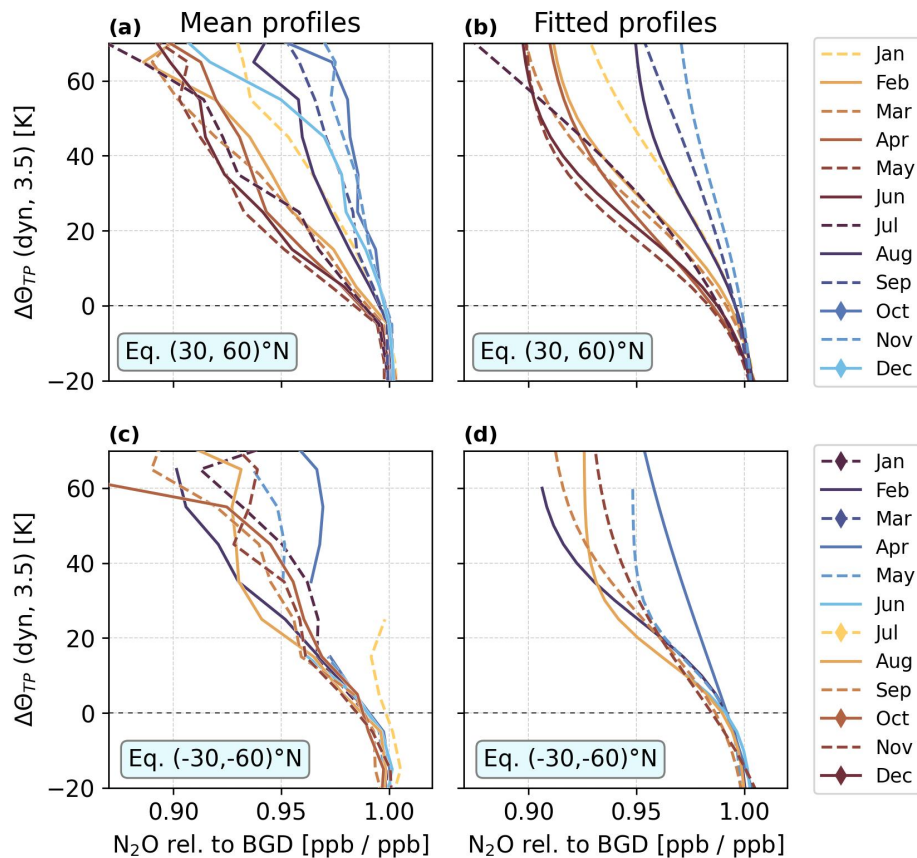


Figure 3. Monthly mean profiles for equivalent latitude range (a-b) [30,60]°N and (c-d) (-60,-30]°N. Panels (a,c) shows observational data binned into 5 K bins in potential temperature. Panels (b,d) show the best fit function for each profile. Diamonds in the legend indicate that no best fit could be evaluated for that month.

hemispheric high latitudes. However, for many months the stratospheric gradient becomes very small or approaches zero for some parts of the stratospheric profile. To determine a representative $\Delta\Theta_{TP}$ based on N_2O observations, the profile therefore needs to exhibit a sufficiently strong decrease throughout the lower stratosphere. For small gradients, the resulting $\Delta\Theta_{TP}$ is ambiguous, particularly when measurement uncertainties are of the same order of magnitude as the change in N_2O across the profile. The average total errors reported for the campaign data used for establishing a climatological relationship often reach several ppb and, in some cases, tens of ppb. This can be compared with the magnitude of the gradients in the lowermost stratosphere, which, for example, range from -0.14 ppb K^{-1} in November to -0.61 ppb K^{-1} in May when converted from relative to absolute N_2O mixing ratios using a representative tropospheric background value of 338 ppb. For a bin spacing of 5 K, these gradients correspond to decreases in N_2O mixing ratio of approximately 0.7–3.1 ppb between adjacent bins, which is comparable in magnitude to the measurement errors.



The factors influencing the quality of the N_2O profiles are multi-faceted. One limitation is the overall number of data points that create the statistical basis. To investigate the impact of sampling availability, we constructed a subsampled climatology of O_3 , randomly selecting data to match the number of N_2O measurements for each bin. This was repeated with different random states 10 times. One example showing downsampled profiles compared with the original sampling can be seen in Appendix D. For all samples, we found increasing irregularities in the O_3 profiles, where the full data set appeared smooth. This suggests that the measurement density of N_2O is likely to be a driving factor for the profile irregularities. Additional factors such as the impact of natural variability, measurement precision and absolute calibration uncertainty compared to the often small stratospheric gradient of N_2O are likely to further impact the deviated profiles.

We have shown that while N_2O can be used as a tracer for distinguishing between troposphere and stratosphere, a climatological tropopause definition and altitude scale based on N_2O is currently not feasible with the data set used here. Advances in regular and precise monitoring of N_2O mixing ratios in the UTLS and expanding the current data set may change this in the future. Additionally, the use of satellite data may allow the construction of a monotonic climatology, but here the vertical resolution is expected to reduce the sharpness of the tropopause.

3.3 O_3 climatology

After constructing the O_3 climatology for monthly equivalent latitude ranges of 10° , we investigate the latitudinal dependence and seasonality of the resulting profiles. Figure 4 shows the monthly O_3 mixing ratio at three distances to the tropopause ($\Delta\Theta = 0, 30, 60 \text{ K}$) for the equivalent latitude ranges $80\text{--}90^\circ$ and $50\text{--}60^\circ$ in both hemispheres. Southern hemispheric data are offset by six months so that corresponding seasons are aligned. A more comprehensive version, including additional equivalent latitude bands and tropopause-relative distances, is shown in Appendix E.

The figure illustrates pronounced differences between hemispheres and equivalent latitude bands, highlighting the need for a climatology that is sufficiently resolved in both season and equivalent latitude. The amplitude of the seasonal cycle increases with distance from the tropopause, while differences between equivalent latitude bands likewise become more pronounced. In the southern polar region, ozone-depleting processes lead to a pronounced spring-time minimum at $\Delta\Theta(\text{dyn}) = 60 \text{ K}$, whereas the corresponding northern hemispheric climatology does not show a comparable spring-time minimum.

In the $80\text{--}90^\circ\text{S}$ equivalent latitude band in October, O_3 mixing ratios at $\Delta\Theta = 60 \text{ K}$ are lower than at 30 K . This indicates that the corresponding profile is not monotonic across this vertical range. It is therefore not possible to derive a unique value of $\Delta\Theta(\text{O}_3)$ from the climatological O_3 profile in this region. Profiles exhibiting this behaviour therefore need to be systematically identified and appropriately flagged before deriving $\Delta\Theta(\text{O}_3)$.

To provide an unambiguous value of $\Delta\Theta$ for a given observation, the corresponding profile needs to be monotonic in the lower stratosphere. For each profile, we identify the first occurrence of a non-monotonic value in increasing $\Delta\Theta$. The results are shown in Fig. 5. Grid cells without a value remain monotonic up to the upper boundary of the climatology of 80 K .

Almost all grid cells of month $\times 10^\circ$ equivalent latitude grid cells in the northern extratropics remain monotonic throughout the climatology. In the tropics, however, non-monotonic behaviour frequently begins as low as 30 K above the tropopause (see supplementary data), reflecting the combination of complex dynamics and a substantially smaller observational data set. Non-

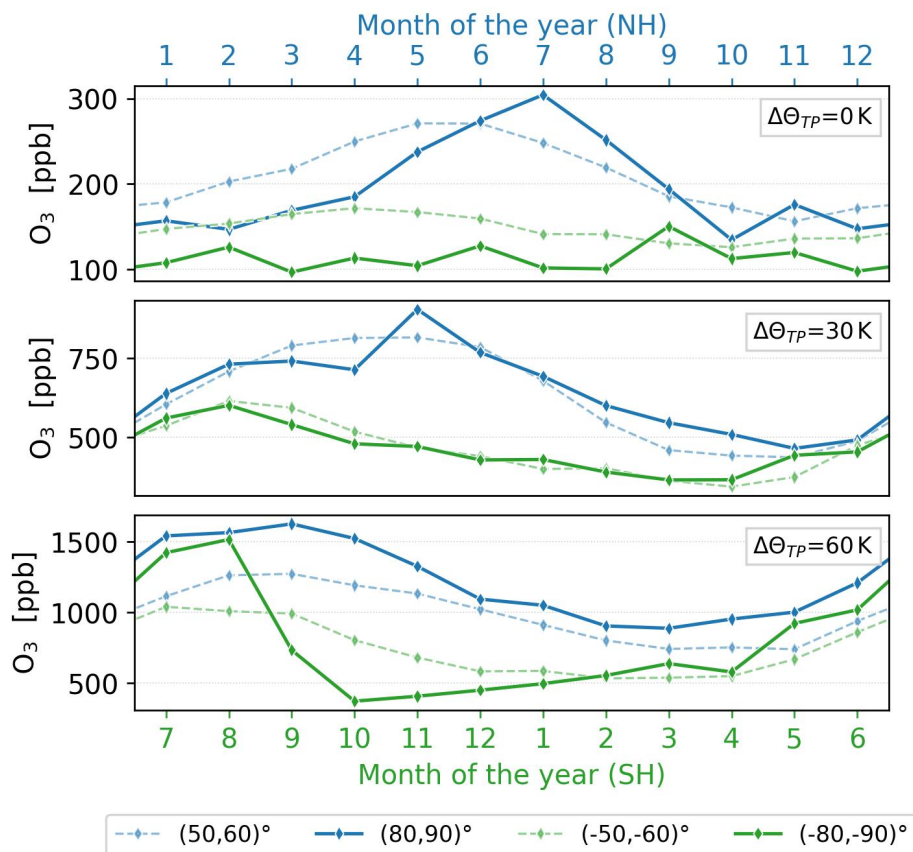


Figure 4. Hemispheric difference of O_3 measurements at selected distances to the dynamical (3.5 PVU) tropopause within mid- and high-latitude equivalent latitude ranges. $\Delta\Theta_{TP}$ values represent bins of 5 K centred at the displayed value. Southern hemispheric data was shifted by 6 months to visually match seasons.

monotonic behaviour also increases in the southern polar region during spring and early summer, where ozone-hole formation and associated springtime chemistry lead to decreases in O_3 at 30–50 K above the tropopause.

Profiles for which non-monotonic behaviour starts below 60 K above the extratropical tropopause are considered unsuitable for deriving a representative $\Delta\Theta(O_3)$. These regions are highlighted in yellow and orange in Fig. 5. This threshold ensures that the climatology remains applicable throughout most of the lowermost stratosphere, therefore these grid cells are excluded from the climatology and applications of the climatological tropopause do not return a valid value of $\Delta\Theta(O_3)$. Overall, 17 of the 144 total profiles (12%) had to be removed from the climatology. For all remaining grid cells, only parts of the profile are kept where an unambiguous relationship between O_3 and $\Delta\Theta(dyn)$ could be evaluated. For all other months and regions, a clear relationship between O_3 and $\Delta\Theta(dyn)$ allows $\Delta\Theta(O_3)$ to be derived for independent O_3 observations. The resulting $\Delta\Theta(O_3)$ can be used analogously to other tropopause-relative coordinates.

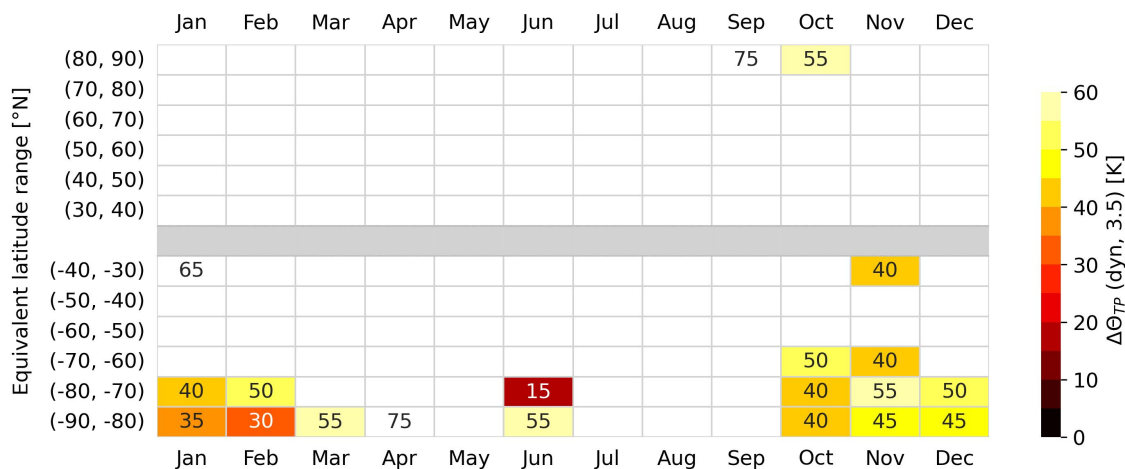


Figure 5. First occurrence of a decreasing value in the climatological profiles of O_3 versus potential temperature relative to the dynamical 3.5 PVU tropopause. Data is binned into month $\times$ 10° equivalent latitude $\times$ 5 K in $\Delta\Theta_{TP}$. Grid cells without a value are strictly monotonically increasing across the full vertical range (up to 80 K above the tropopause). Highlighted in colour are fields with non-monotonic behaviour below 60 K above the tropopause.

260 4 Results and discussion

4.1 Applying $\Delta\Theta(O_3)$ to IAGOS CARIBIC-2 project (2005-2020)

The climatological $\Delta\Theta(O_3)$ is now applied to SF_6 measurements from the IAGOS-CARIBIC flask samples between 2005 and 2020 in the northern extra-tropics. The data is distributed across 335 flights with highest observational density in the mid-latitudes along typical trans-continental flight paths. The samples are collected primarily during the cruise phase of long-haul flights, which corresponds to a typical altitude of 10–12 km. Similarly to N_2O , the SF_6 data are detrended to account for the long observation period and are expressed as mixing ratio relative to the tropospheric background, $SF_6 \text{ rel. to BGD}$ [ppt/ppt]. We now evaluate how the distribution of SF_6 changes depending on the chosen tropopause-relative coordinate. Statistics are calculated for bins of $2.5^\circ N$ in equivalent latitude and 2.5 K in both $\Delta\Theta(dyn)$ and $\Delta\Theta(O_3)$, where the reference O_3 is based on integrated values from a higher resolution instrument. For bins containing at least three data points, the mean SF_6 mixing ratio and standard deviation are calculated and shown in Fig. 6.

A notable difference between the two coordinates is the reduced vertical extent of $\Delta\Theta(O_3)$. As a consequence, the same observations also occupy fewer vertical bins. While the dynamic tropopause places CARIBIC observations up to 60 K above the tropopause, the O_3 -based coordinate reaches only around 30 K for the same observations. Note that this result does not relay information on the physical separation between observation and local tropopause. It does, however, suggests that the O_3 distribution does not support the large dynamical distance from the tropopause shown by $\Delta\Theta(dyn)$. As the CARIBIC data set is sampled predominantly at flight altitudes of 10–12 km, large values of $\Delta\Theta$ are expected to occur mainly under atypical

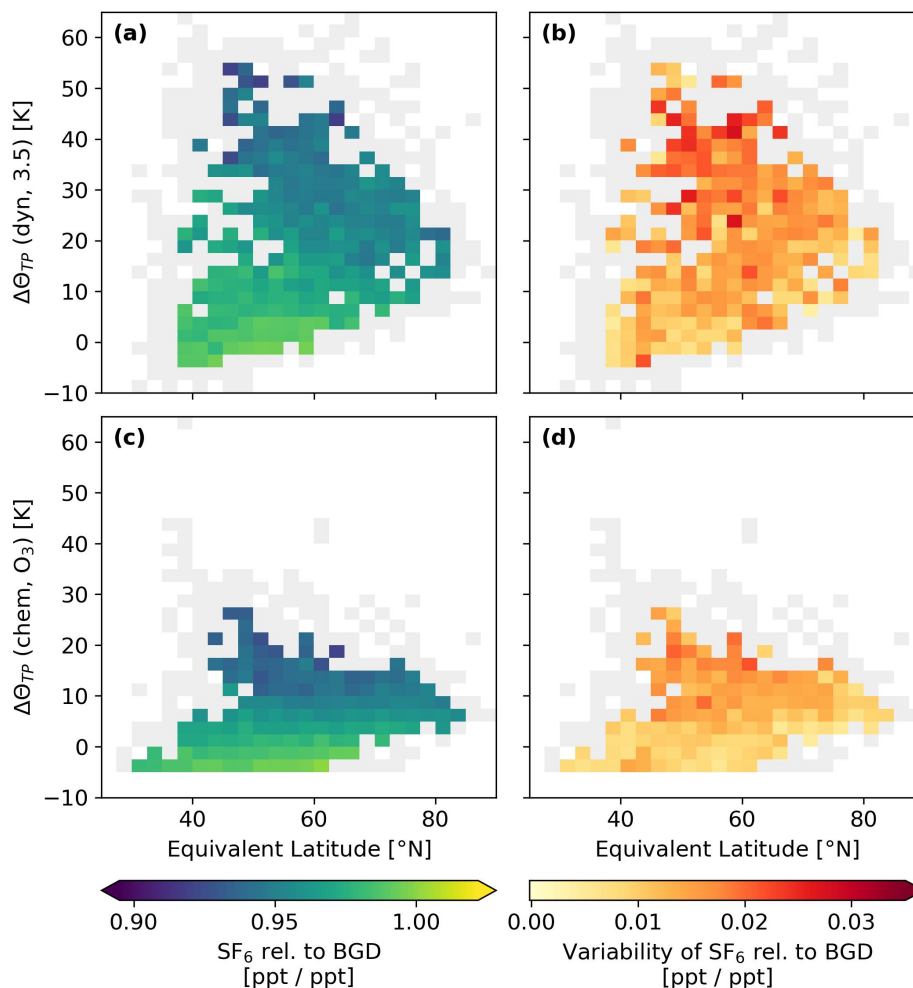


Figure 6. 2D-binned SF_6 measurements from IAGOS-CARIBIC flask samples relative to (top row) dynamical 3.5 PVU-surface tropopause and (bottom row) derived O_3 -TP climatological tropopause. On the left the mean values of each bin are shown, with the corresponding bin variability on the right hand side. Data were binned on a grid of 2.5 K in $\Delta\Theta_{TP}$ and 2.5 °N in equivalent latitude. Bins with fewer than three data points are indicated in grey.

conditions, such as (1) unusually low tropopause heights, for example due to tropopause folds at the subtropical and polar jets; (2) strong potential temperature gradients above the tropopause, for example above regions of strong convective or strong frontal uplift; or (3) unusually high potential temperatures at the observation point, for example due to exceptionally strong meridional excursions of tropical air masses.

Note further that the SF_6 -binned mean as well as the standard deviation display a tropopause-following structure when changing coordinates to $\Delta\Theta(\text{O}_3)$. This indicates that $\Delta\Theta(\text{dyn})$ does not fully capture the dynamical variability affecting the



chemical distribution further above the tropopause. Referencing to the chemical gradient of O_3 reduces the variability as shown in Fig. 6 (b,d) since O_3 and SF_6 are linearly correlated in the UTLS.

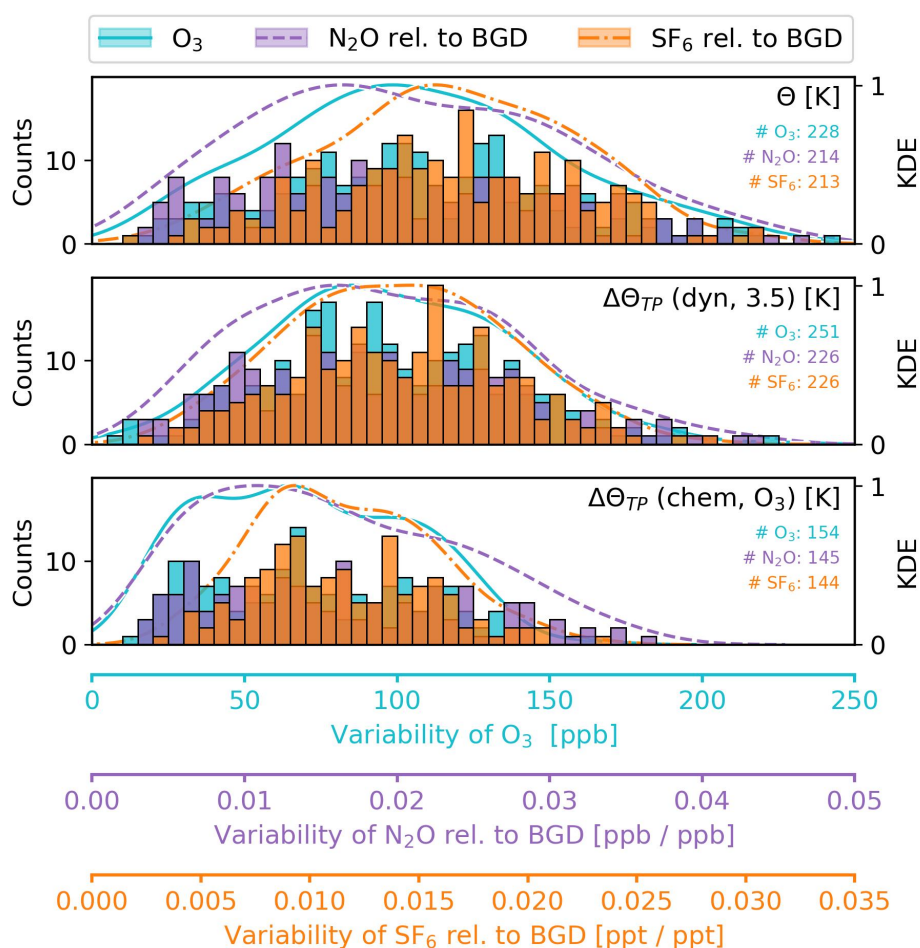


Figure 7. Histogram and kernel density estimation of binned 2D variability in IAGOS-CARIBIC. Shown are detrended SF_6 , detrended N_2O and O_3 . Data were binned in 2.5° equivalent latitude over 2.5 K bins in potential temperature (top, Θ), potential temperature relative to the modelled dynamical tropopause / 3.5 PVU surface (middle, $\Delta\Theta_{TP}(dyn)$) and derived potential temperature relative to the chemical O_3 -tropopause (bottom, $\Delta\Theta_{TP}(chem)$).

285 To further investigate this, Fig. 7 compares the binned variability of N_2O , SF_6 and O_3 for the vertical coordinates Θ , $\Delta\Theta(dyn)$ and $\Delta\Theta(O_3)$. The chemical tropopause has a smaller vertical extent than the dynamical tropopause, resulting in fewer populated bins for the same vertical spacing. The number of bins for each tracer and coordinate is indicated in the figure and kernel density estimates (KDEs) are included to facilitate comparison between panels. Note that tracer distributions within a panel should not be compared directly as measurement availability differs between tracers.



290 For all three trace gases, variability is lowest when using $\Delta\Theta(\text{O}_3)$. Compared with $\Delta\Theta(\text{dyn})$, the mean binned variability decreases by 18% for N_2O and SF_6 and by 28% for O_3 . The maximum variability decreases by 14%, 18% and 28% for N_2O , SF_6 and O_3 , respectively. The reduction in O_3 is expected because O_3 is used to derive the chemical coordinate; we therefore include this result for completeness. Overall, the reduction of variability is much larger when changing from $\Delta\Theta(\text{dyn})$ to $\Delta\Theta(\text{O}_3)$ than when changing from Θ to $\Delta\Theta(\text{dyn})$.

295 The reduced binned variability indicates that $\Delta\Theta(\text{O}_3)$ provides a more chemically coherent representation of the sampled air than the dynamic tropopause. A comparison with larger observational data sets would be necessary to investigate how well this reflects climatological conditions.

4.2 Applying $\Delta\Theta(\text{O}_3)$ to PHILEAS campaign (Aug-Sep 2023)

We apply the same methodology to the PHILEAS data set to assess how well the results from the CARIBIC climatology can be generalised. In contrast to the regular CARIBIC flask observations, PHILEAS provides high-resolution in situ measurements obtained during a focused field campaign. The campaign focused on the outflow from the Asian Summer Monsoon, which transports large amounts of polluted air into the extratropical upper troposphere and stratosphere (Riese et al., 2024; Jesswein et al., 2025). Consequently, the sampled air masses are expected to be strongly influenced by transport, mixing and pollution. The distance to the local dynamical tropopause may not fully represent the chemical character of the sampled air, whereas the chemical tropopause is implicitly sensitive to these effects.

305 We separate this data set in 2.5° equivalent latitude and 2.5 K vertical bins, analogous to the previous example (see Appendix F for the binned SF_6 data from PHILEAS). The chemical tropopause similarly compresses the vertical range, which similarly reduces the number of bins and maximum sampled distance from 80 K in $\Delta\Theta(\text{dyn})$ to 55 K in $\Delta\Theta(\text{O}_3)$ for the same observations. As in the CARIBIC data set, the O_3 mixing ratios observed at the largest values of $\Delta\Theta(\text{dyn})$ are more representative of air located substantially closer to the tropopause. This indicates that those large dynamical distances are not reflected in the chemical composition.

315 Figure 8 shows the binned variability for 2.5° equivalent latitude and 2.5 K vertical bins, analogous to the CARIBIC analysis. The reduction in binned variability is consistent across all three trace gases. Compared to $\Delta\Theta(\text{dyn})$, the mean binned variability decreases by 38% for N_2O , 57% for O_3 and 11% for SF_6 . The reduced vertical extent likewise leads to fewer populated bins for the same spacing. The maximum variability decreases by 37% for N_2O , 43% for SF_6 and 63% for O_3 .

320 Despite fundamental differences in geographical and temporal coverage and resolution, the CARIBIC and PHILEAS data sets show the same systematic behaviour. In the first example, measurements collected over 15 years were combined, while in the PHILEAS example only August and September of a single year are present. The climatological representation obtained from the CARIBIC data stands in contrast to the PHILEAS data, which represents a single season under conditions heavily influenced by the Asian Summer Monsoon. This difference in temporal sampling needs to be considered when comparing the results from both data sets, as atmospheric variability itself changes substantially throughout the year. Nevertheless, both data sets show the same systematic reduction in variability when using $\Delta\Theta(\text{O}_3)$ instead of $\Delta\Theta(\text{dyn})$. In both cases, $\Delta\Theta(\text{O}_3)$ reduces the tropopause-relative vertical extent of the observations, sharpens stratospheric gradients and reduces binned vari-

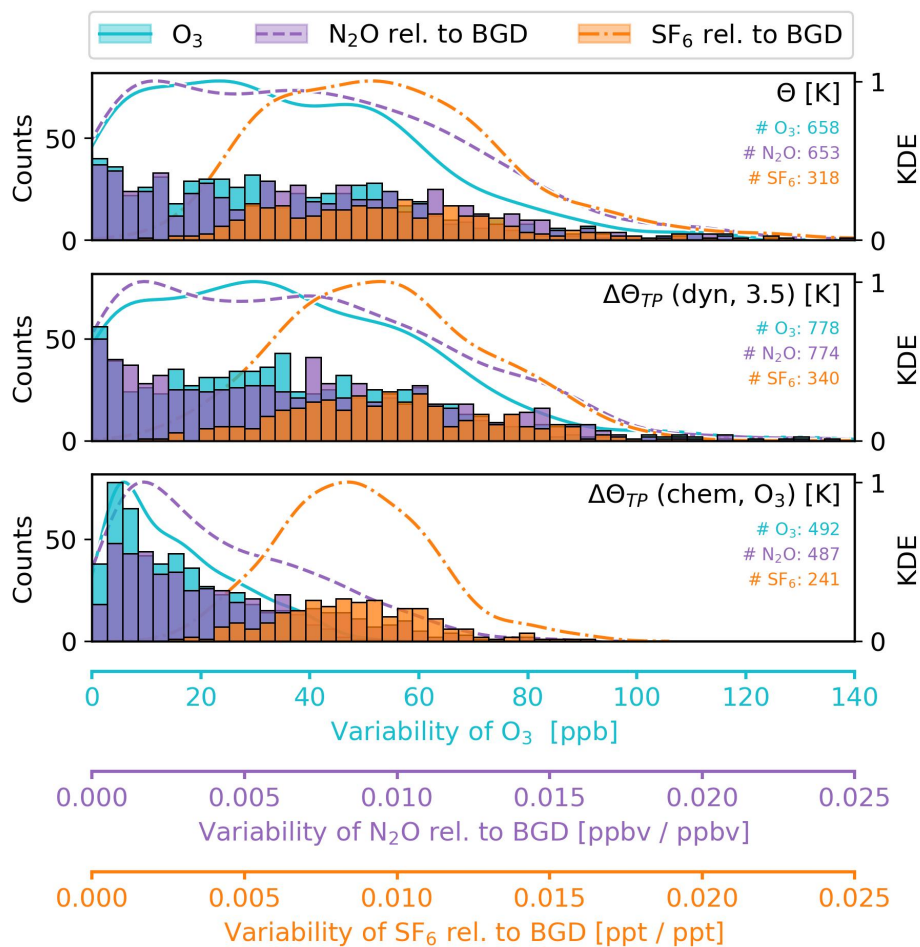


Figure 8. As Fig. 7, for PHILEAS data.

abilities for all trace gases investigated. These consistent results indicate that the climatological O_3 -based tropopause provides a more chemically coherent vertical coordinate than the dynamic tropopause across substantially different observational data sets.

5 Conclusions

Trace gas observations in the extratropical tropopause region are difficult to consolidate due to the strong impact of local dynamics on their chemical composition. Conventional tropopause-relative coordinates often require atmospheric profiles or model data, which are limited in availability or may introduce mismatches between observations and modelled atmospheric structures. We show that, with a robust climatology, a representative potential temperature distance to the chemical tropopause, $\Delta\Theta(O_3)$, can be derived from individual O_3 observations. The coordinate is based on the climatological relationship between



O₃ and potential temperature distance to the dynamical tropopause (3.5 PVU surface), but its application to individual observations does not require a local vertical profile or modelled tropopause. This provides an observation-based vertical coordinate
 335 that is sensitive to the chemical characteristics of the sampled air while retaining the benefits of a tropopause-relative frame-
 work. We apply $\Delta\Theta(\text{O}_3)$ to independent measurements of SF₆, N₂O and O₃ from the IAGOS-CARIBIC project and the HALO
 aircraft campaign PHILEAS. In both data sets, $\Delta\Theta(\text{O}_3)$ systematically compresses the vertical extent of the observations com-
 pared to $\Delta\Theta(\text{dyn})$, indicating that distance from the local dynamical tropopause does not fully capture the chemical structure
 of the sampled air. The new coordinate leads to a reduction in binned variability of 18% for N₂O and SF₆ (and 28% for O₃) in
 340 CARIBIC, while reductions of 11% for SF₆ and 38% for N₂O (and 57% for O₃) were found for PHILEAS.

These results indicate that $\Delta\Theta(\text{O}_3)$ accounts for aspects of transport and mixing that are not fully represented by distance
 from the local dynamical tropopause alone. O₃, as a passive dynamic tracer in the lowermost stratosphere, reflects the effects of
 the dynamics on the distribution of species. Tracers with a systematic relationship to O₃, such as N₂O and SF₆, consequently
 show reduced dynamical variability when expressed as a function of $\Delta\Theta(\text{O}_3)$. The compression of the vertical extent further
 345 indicates that large potential temperature distances from the dynamical tropopause do not necessarily correspond to distinct
 chemical conditions. Organising observations according to $\Delta\Theta(\text{O}_3)$ therefore provides a means of consolidating measurements
 with more consistent chemical characteristics, facilitating studies of atmospheric composition in which short-term dynamical
 variability needs to be distinguished from longer-term changes.

Thermal, dynamical and chemical tropopause definitions characterise different aspects of the troposphere-stratosphere transi-
 350 tion. While existing approaches capture these characteristics, applying them consistently to individual observations may require
 local atmospheric profiles or model data. Our approach instead uses an observation-based climatology to assign a representative
 $\Delta\Theta_{TP}$ from individual O₃ measurements. Compared with chemical tropopause definitions based on fixed thresholds or local
 atmospheric structure, our method accounts for seasonal and latitudinal variation in the relationship between O₃ and distance
 from the dynamical tropopause and provides a consistent vertical coordinate across the extratropical lower stratosphere. The
 355 consistent behaviour in the regular but lower-resolution CARIBIC observations, combining measurements over 15 years, and
 the high-resolution PHILEAS campaign, sampling August and September 2023, demonstrates that the effect is not restricted
 to a particular sampling strategy or temporal coverage.

The strong seasonal and latitudinal dependence of O₃ necessitates monthly resolution for each 10° equivalent latitude range.
 We found that 88% of the climatologically averaged profiles in the extratropics are monotonic in the lower stratosphere and
 360 describe an unambiguous relationship between O₃ and $\Delta\Theta(\text{dyn})$. Irregularities in the tropics and parts of the southern polar
 region during spring and summer prevent reliable inversions, limiting applicability of the O₃-based coordinate in these regions.
 N₂O is expected to provide a suitable monotonic chemical structure in the lower stratosphere, but available observations are
 too sparse and irregular to construct a robust climatological coordinate. This demonstrates the importance of the availability,
 quality and representativeness of observations for this approach. The monthly resolution in 10° equivalent latitude bands
 365 should also be taken into account when applying this methodology to higher resolution data. In the southern polar regions,
 chemical interactions related to ozone-hole formation further limit the applicability of the coordinate. Other tracers may provide



complementary information, depending on their atmospheric distribution, interactions and observational coverage. Improved regular and continuous N₂O measurements in the UTLS may allow this implementation the methodology in the future.

Overall, $\Delta\Theta(\text{O}_3)$ provides a practical, observation-based vertical coordinate for consolidating extratropical UTLS measurements across different dynamical states. The main advantage of this methodology lies in the applicability to individual observations without requiring corresponding local profiles or reanalysis data, while retaining information on their chemical characteristics. It thus provides a complementary framework for investigating atmospheric composition, transport and mixing in observational data sets.



Appendix A: WOUDC station overview

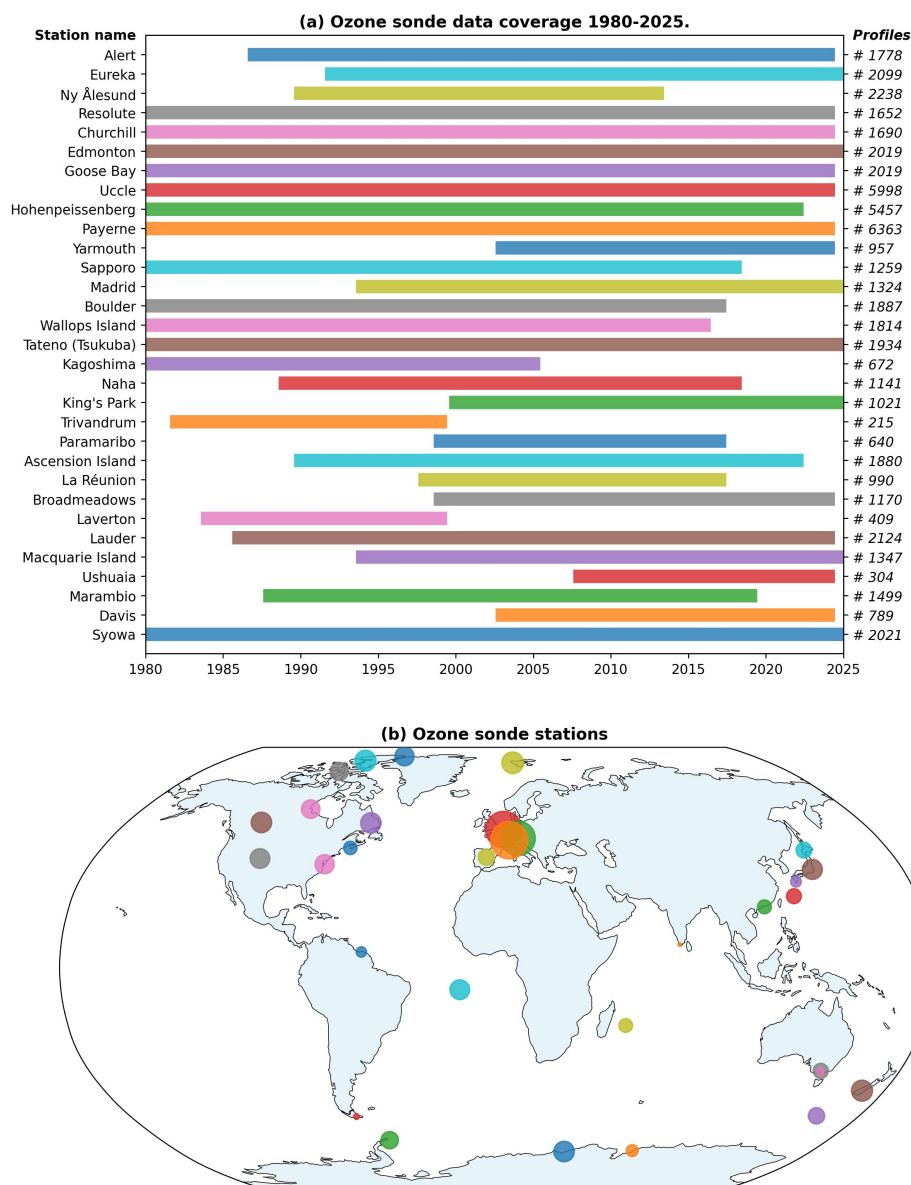


Figure A1. Overview of available ozone sonde measurements from 31 globally distributed stations. Shown are (a) the availability and number of profiles per station over time and (b) the geographical distribution of stations. Stations are sorted by latitude (top=north) in panel (a) and the colour mapping is the same across panels. Marker sizes in (b) are indicative of the number of profiles available.



375 Appendix B: N₂O data sources

Table B1. N₂O data sources and instrumentation overview.

FTIR = Fourier transform infrared spectrometry

GC-ECD: Gas Chromatograph-Electron Capture Detector

IR abs. spec. = Infrared absorption spectrometry

QCL = Quantum Cascade Laser

Campaign	Years	Nr. of pts	Instrument	Platform	Instrument type
AASE	1988-1989	195	WAS	ER2	Flask samples
OMS	1989-1999	299	ALIAS-2	Balloon	IR abs. spec.
	1989-1999	112	FT	Balloon	FTIR
	1989-1999	50	MkIV	Balloon	FTIR
	1989-1999	5	SLS	Balloon	Limb Sounder / Radiometer
PEMWESTA	1991	26702	DACOM	DC8	IR abs. spec.
TRACE-A	1992	22443	DACOM	DC8	IR abs. spec.
PEMWESTB	1994	9548	DACOM	DC8	IR abs. spec.
POLARIS	1997	1563	ACATS	ER2	GC-ECD
	1997	73	ALIAS-2	Balloon	IR abs. spec.
	1997	3340	SLS	Balloon	Limb Sounder / Radiometer
ACCENT	1999	1343	ACATS	WB57	GC-ECD
SOLVE THESEO 2000	1999-2000	47	ACATS	Balloon	GC-ECD
	1999-2000	4088	ACATS	ER2	GC-ECD
	1999-2000	978	ASUR	DC8	Radiometer
	1999-2000	56324	DACOM	DC8	IR abs. spec.
	1999-2000	4	SLS	Balloon	Limb Sounder / Radiometer
TRACE-B	2001	18208	DACOM	DC8	IR abs. spec.
CNESODInvalidation	2001-2002	3487	SPIRALE	Balloon	IR abs. spec.
SPURT	2001-2003	45903	TRISTAR	LearJet 35	QCL-Spectrometer
BOS	2002-2007	108	ACATS	Balloon	GC-ECD
	2002-2007	21	AWAS	Balloon	Radiometer
	2002-2007	290	FT	Balloon	Cryogenic flask samples
SCIAVALUE	2002-2003	349	ASUR	Falcon	FTIR
SOLVEII	2003	26371	DACOM	DC8	IR abs. spec.
ENVISATvalidation	2003-2006	5703	SPIRALE	Balloon	IR abs. spec.



Campaign	Years	# pts	Instrument	Platform	Instrument type
PAVE	2005	170	ASUR	DC8	Radiometer
	2005	11610	DACOM	DC8	Flask samples
AVEHouston2	2005	346	AWAS	WB57	IR abs. spec.
CRAVE	2006	581	AWAS	WB57	flask samples
	2006	963	PANTHER	WB57	GC-ECD
CARIBIC	2006-2020	7385	WAS	Lufthansa A340	Flask + GC-ECD
TC4-DC8	2007	102	DACOM	DC8	IR abs. spec.
	2007	861	GC	WB57	GC-ECD
ARCTAS	2008	17199	DACOM	DC8	IR abs. spec.
Start08	2008	1360	QCLS	GV-HIAPER	QCL-Spectrometer
	2008	20339	UCATS	GV-HIAPER	IR abs. spec.
SCOUTO3	2008	4943	SPIRALE	Balloon	IR abs. spec.
Hippo	2009-2011	104891	QCLS	GV-HIAPER	QCL-Spectrometer
	2009-2011	378	UGC	GV-HIAPER	IR abs. spec.
STRAPOLATE	2009	3471	SPIRALE	Balloon	IR abs. spec.
GloPac	2010	2652	ACATS	GlobalHawk	GC-ECD
MACPEX	2011	14649	ALIAS	WB57	IR abs. spec.
ENRICHED	2011	2503	SPIRALE	Balloon	IR abs. spec.
ATTREX	2011-2014	15733	UCATS	GlobalHawk	GC-ECD
DC3	2012	16756	DACOM	DC8	IR abs. spec.
ESMVal	2012	16423	TRIHOP	HALO	QCL-IR abs. spec.
TACTS	2012	15889	TRIHOP	HALO	QCL-IR abs. spec.
Airtoss	2013	5562	UMAQs	Learjet	QCL-Spectrometer / continuous
GWcycle	2013	5834	UMAQs	Falcon	QCL-Spectrometer / continuous
Polstracc	2015-2016	45700	TRIHOP	HALO	QCL-IR abs. spec.
KORUSAQ	2016	58476	DACOM	DC8	IR abs. spec.
POSIDON	2016	1436	PANTHER	WB57	GC-ECD
ORCAS	2016	22746	QCLS	GV-HIAPER	QCL-Spectrometer
ATOM	2016	13321	UCATS	DC8	GC-ECD
StratoClim	2017	6680	COLD	M55 Geophysica	QCL-Spectrometer
Wise	2017	44370	UMAQs	HALO	QCL-Spectrometer / continuous
Southtrac	2019	48852	UMAQs	HALO	QCL-Spectrometer / continuous
Phileas	2023	507992	UMAQs	HALO	QCL-Spectrometer / continuous



Appendix C: Global N₂O monthly mean profiles and best fits

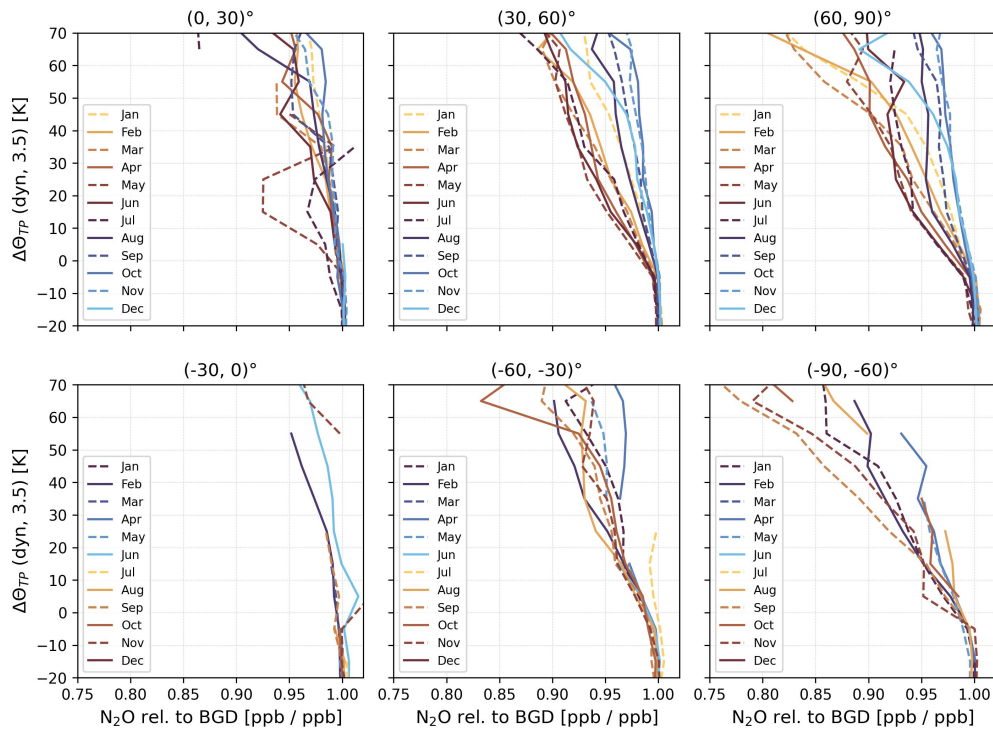


Figure C1. Monthly mean profiles N₂O per 30° equivalent latitude range (boundaries shown in the axis titles) and month. Data is binned vertically in potential temperature distance to the local 3.5 PVU surface $\Delta\Theta_{TP}(dyn, 3.5) = 5$ K.

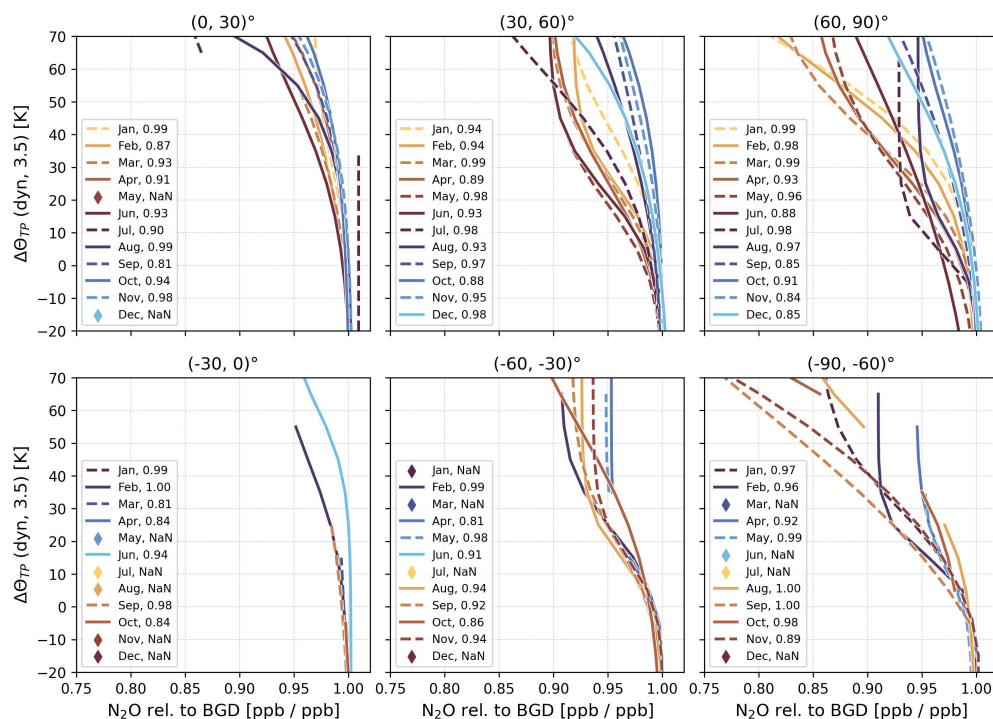


Figure C2. Climatological fitted profiles of N_2O for equivalent latitude bins of 30° and per month. The values shown in the legend indicate the R^2 quality of fit parameter for the respective month. Diamonds in the legend indicate that no best fit could be evaluated.



Appendix D: Subsampled O₃

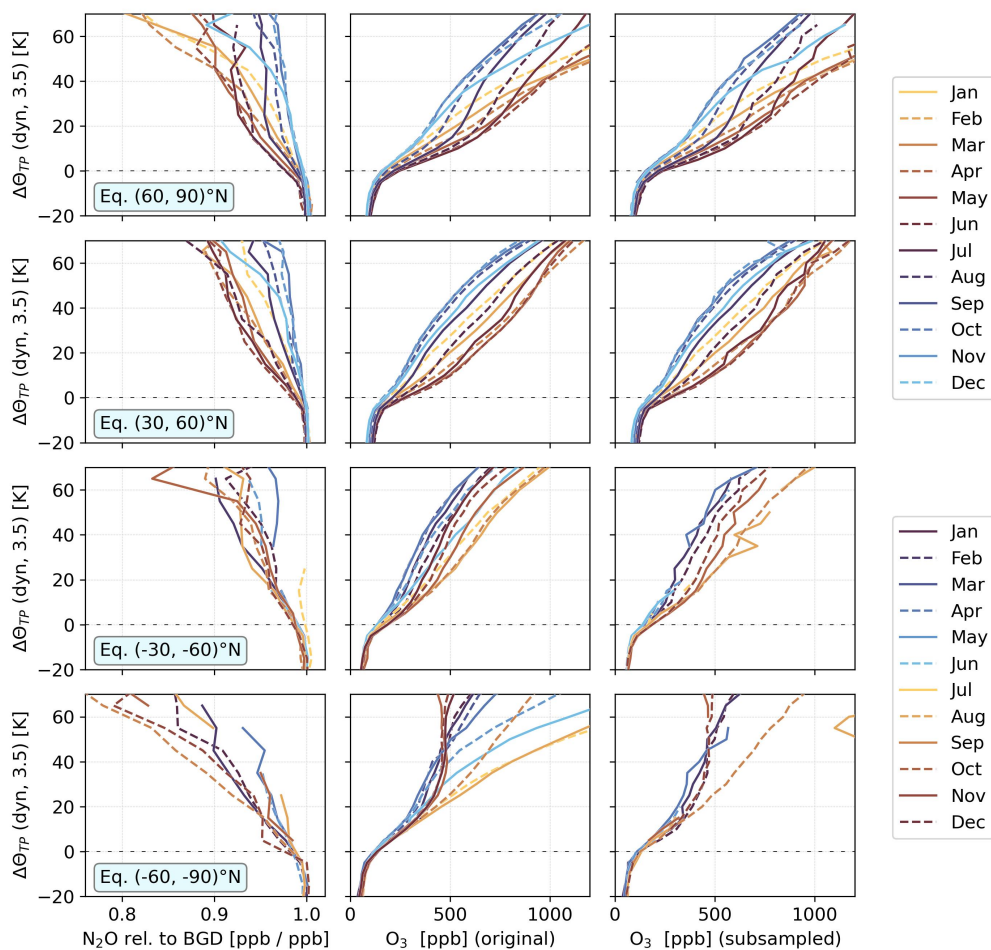


Figure D1. N₂O profiles (left column), O₃ profiles at original sampling density (middle column) and subsampled O₃ profiles to match N₂O observational density (right column) for equivalent latitude ranges shown as text box in the left column: First row = (60,90), second = (30,60), third = (-30,-60), fourth = (-60,-90). O₃ data were subsampled after binning (equivalent latitude = 30° vs. month vs. $\Delta\Theta_{TP}(dyn) = 5$ K) with numpy random seed = 31.



Appendix E: O₃ seasonality / hemispheric differences

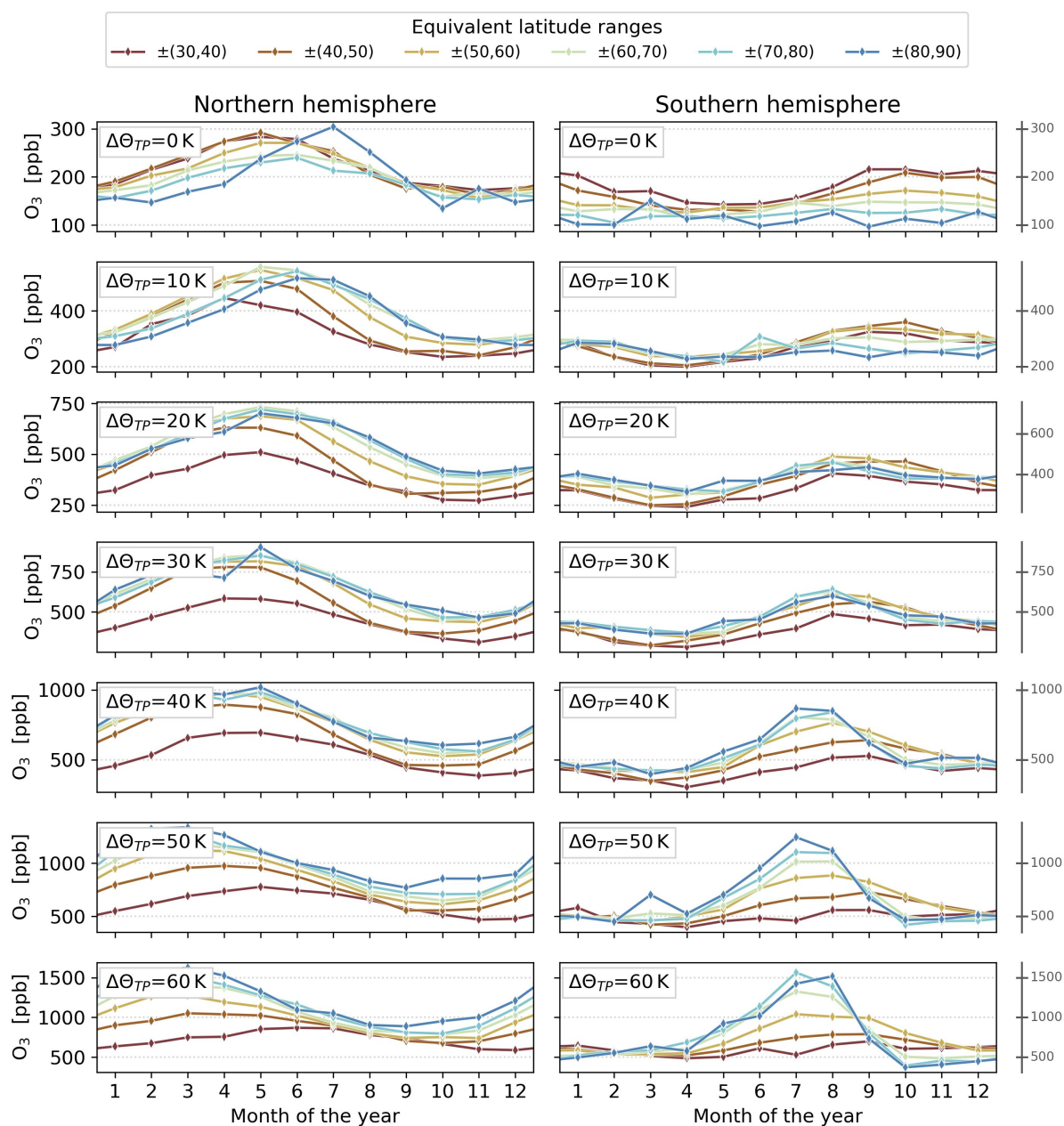


Figure E1. Seasonality of O₃ at different displacement values from the tropopause and equivalent latitude ranges. Note that y-axis ranges vary across panels. Values of $\Delta\Theta_{TP}$ indicate the midpoint of the corresponding 5 K bin in potential temperature distance to the dynamic tropopause (3.5 PVU surface).



Appendix F: PHILEAS: Binned SF_6

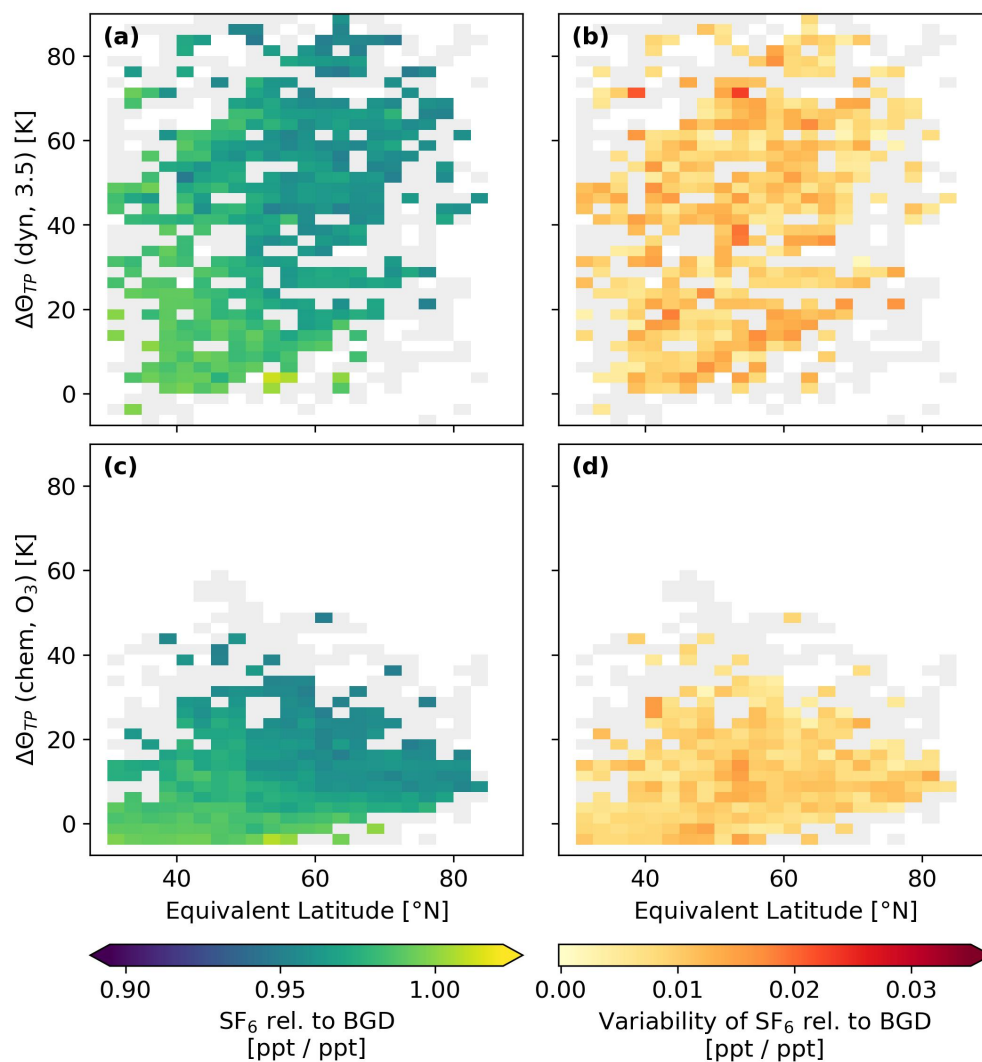


Figure F1. As Fig. 6 but for PHILEAS SF_6 data from the GHoST instrument.



380 *Code and data availability.* Climatological data for the ozone tropopause and Python scripts for the application can be accessed at <https://doi.org/10.5281/zenodo.22112145>. The ozone sonde data can be accessed through the World Ozone and Ultraviolet Data Center (WOUDC) at <https://woudc.org/>. The zonal mean N₂O data set as compiled by Krysztofiak et al. (2023a) is available at <https://doi.org/10.25326/428> with individual data sets available upon request from the author. Interpolated ERA5 reanalysis data for campaigns and O₃ sondes are available at <https://doi.org/10.5281/zenodo.15076519>. The whole-air-sampler data of the IAGOS-CARIBIC project are available at <https://doi.org/10.5281/zenodo.8188548>. The observational data of the HALO flights during the PHILEAS campaign are available upon request via the HALO database (<https://halo-db.pa.op.dlr.de/mission/138>).

Author contributions. HCL contributed ERA5 model data interpolated to observations. SB, TS and AE initiated and conceptualised the core research goals and together with AZ, HB and PH contributed valuable ideas in discussions. SB performed the analysis and prepared the manuscript with contributions from TS and AE. All authors contributed via discussions and comments.

390 *Competing interests.* At least one of the (co-)authors is a member of the editorial board of Atmospheric Chemistry and Physics.

Acknowledgements. This research has been supported by the Deutsche Forschungsgemeinschaft (grant no. SCHU 3258/3-1 – project ID 501095243) and DFG collaborative research programme “The Tropopause Region in a Changing Atmosphere” TRR 301 – project ID 428312742). This research was supported by the German Research Foundation (DFG) within the Priority Program HALO SPP 1294 under project number 316646266 and EN 367/19-1. IAGOS-CARIBIC data were created with support from the European Commission, national agencies in Germany (BMBF), France (MESR) and the UK (NERC), and the IAGOS member institutions (<https://www.iagos.org/organisation/members>).



References

- Bauchinger, S., Engel, A., Jesswein, M., Keber, T., Bönisch, H., Obersteiner, F., Zahn, A., Emig, N., Hoor, P., Lachnitt, H.-C., Weyland, F., Ort, L., and Schuck, T. J.: The extratropical tropopause – trace gas perspective on tropopause definition choice, *Atmospheric Chemistry and Physics*, 25, 14 167–14 186, <https://doi.org/10.5194/acp-25-14167-2025>, 2025.
- 400 Bethan, S., Vaughan, G., and Reid, S. J.: A comparison of ozone and thermal tropopause heights and the impact of tropopause definition on quantifying the ozone content of the troposphere, *Quarterly Journal of the Royal Meteorological Society*, 122, 929–944, <https://doi.org/10.1002/qj.49712253207>, _eprint: <https://onlinelibrary.wiley.com/doi/pdf/10.1002/qj.49712253207>, 1996.
- Birner, T., Dörnbrack, A., and Schumann, U.: How sharp is the tropopause at midlatitudes?, *Geophysical Research Letters*, 29, 45–1–45–4, <https://doi.org/10.1029/2002GL015142>, _eprint: <https://onlinelibrary.wiley.com/doi/pdf/10.1029/2002GL015142>, 2002.
- 405 Butchart, N. and Remsberg, E. E.: The Area of the Stratospheric Polar Vortex as a Diagnostic for Tracer Transport on an Isentropic Surface, *Journal of the Atmospheric Sciences*, 43, 1319–1339, [https://doi.org/10.1175/1520-0469\(1986\)043<1319:TAOTSP>2.0.CO;2](https://doi.org/10.1175/1520-0469(1986)043<1319:TAOTSP>2.0.CO;2), 1986.
- Dutton, G., Elkins, J. W., Hall, B., and NOAA, E.: Earth System Research Laboratory Halocarbons and Other Atmospheric Trace Gases Chromatograph for Atmospheric Trace Species (CATS) Measurements, <https://doi.org/10.7289/V5X0659V>, 2017.
- Dutton, G., Hall, B., Dlugokencky, E., Lan, X., Nance, D., Gentry, M., Madronich, M., and NOAA Global Monitoring Laboratory: Nitrous
 410 Oxide (N₂O) hemispheric and global monthly means from the NOAA/GML halocarbons program., <https://doi.org/10.15138/GMZ7-2Q16>, 2022.
- Fischer, H., Wienhold, F. G., Hoor, P., Bujok, O., Schiller, C., Siegmund, P., Ambaum, M., Scheeren, H. A., and Lelieveld, J.: Tracer correlations in the northern high latitude lowermost stratosphere: Influence of cross-tropopause mass exchange, *Geophysical Research Letters*, 27, 97–100, <https://doi.org/10.1029/1999GL010879>, _eprint: <https://onlinelibrary.wiley.com/doi/pdf/10.1029/1999GL010879>, 2000.
- 415 Gettelman, A., Hoor, P., Pan, L. L., Randel, W. J., Hegglin, M. I., and Birner, T.: The extratropical upper troposphere and lower stratosphere, *Reviews of Geophysics*, 49, <https://doi.org/10.1029/2011RG000355>, 2011.
- HALO: Mission: SouthTRAC (Southern hemisphere TRANsport Composition and dynamics), <https://doi.org/10.17616/R39Q0T>, 2021.
- HALO: Mission PHILEAS (Probe High Latitude Export of air from the Asian Summer Monsoon), <https://doi.org/10.17616/R39Q0T>, 2025.
- Hegglin, M. I., Brunner, D., Peter, T., Hoor, P., Fischer, H., Staehelin, J., Krebsbach, M., Schiller, C., Parchatka, U., and Weers, U.: Measurements of NO, NO_y, N₂O, and O₃ during SPURT: implications for transport and chemistry in the lowermost stratosphere, *Atmospheric Chemistry and Physics*, 6, 1331–1350, <https://doi.org/10.5194/acp-6-1331-2006>, 2006.
- 420 Hegglin, M. I., Boone, C. D., Manney, G. L., and Walker, K. A.: A global view of the extratropical tropopause transition layer from Atmospheric Chemistry Experiment Fourier Transform Spectrometer O₃, H₂O, and CO, *Journal of Geophysical Research: Atmospheres*, 114, <https://doi.org/10.1029/2008JD009984>, _eprint: <https://onlinelibrary.wiley.com/doi/pdf/10.1029/2008JD009984>, 2009.
- 425 Hersbach, H., Bell, B., Berrisford, P., Hirahara, S., Horányi, A., Muñoz-Sabater, J., Nicolas, J., Peubey, C., Radu, R., Schepers, D., Simmons, A., Soci, C., Abdalla, S., Abellan, X., Balsamo, G., Bechtold, P., Biavati, G., Bidlot, J., Bonavita, M., De Chiara, G., Dahlgren, P., Dee, D., Diamantakis, M., Dragani, R., Flemming, J., Forbes, R., Fuentes, M., Geer, A., Haimberger, L., Healy, S., Hogan, R. J., Hólm, E., Janisková, M., Keeley, S., Laloyaux, P., Lopez, P., Lupu, C., Radnoti, G., de Rosnay, P., Rozum, I., Vamborg, F., Villaume, S., and Thépaut, J.-N.: The ERA5 global reanalysis, *Quarterly Journal of the Royal Meteorological Society*, 146, 1999–2049, <https://doi.org/10.1002/qj.3803>, _eprint: <https://onlinelibrary.wiley.com/doi/pdf/10.1002/qj.3803>, 2020.
- 430



- Hoor, P., Fischer, H., Lange, L., Lelieveld, J., and Brunner, D.: Seasonal variations of a mixing layer in the lowermost stratosphere as identified by the CO-O₃ correlation from in situ measurements, *Journal of Geophysical Research: Atmospheres*, 107, ACL 1–1–ACL 1–11, <https://doi.org/10.1029/2000JD000289>, eprint: <https://agupubs.onlinelibrary.wiley.com/doi/pdf/10.1029/2000JD000289>, 2002.
- Hoor, P., Gurk, C., Brunner, D., Hegglin, M. I., Wernli, H., and Fischer, H.: Seasonality and extent of extratropical TST derived from in-situ CO measurements during SPURT, *Atmospheric Chemistry and Physics*, 4, 1427–1442, <https://doi.org/10.5194/acp-4-1427-2004>, 2004.
- Ishijima, K., Patra, P. K., Takigawa, M., Machida, T., Matsueda, H., Sawa, Y., Steele, L. P., Krummel, P. B., Langenfelds, R. L., Aoki, S., and Nakazawa, T.: Stratospheric influence on the seasonal cycle of nitrous oxide in the troposphere as deduced from aircraft observations and model simulations, *Journal of Geophysical Research*, 115, D20 308, <https://doi.org/10.1029/2009JD013322>, 2010.
- Jesswein, M., Lauther, V., Emig, N., Hoor, P., Keber, T., Lachnitt, H.-C., Ort, L., Schuck, T., Strobel, J., Van Luijt, R., Volk, C. M., Weyland, F., and Engel, A.: Tracing elevated abundance of CH₂Cl₂ in the subarctic upper troposphere to the Asian Summer Monsoon, <https://doi.org/10.5194/egusphere-2024-3946>, 2025.
- Krysztofiak, G., Catoire, V., Dudok De Wit, T., Kinnison, D. E., Ravishankara, A. R., Brocchi, V., Atlas, E., Bozem, H., Commane, R., D’Amato, F., Daube, B., Diskin, G. S., Engel, A., Friedl-Vallon, F., Hintsä, E., Hurst, D. F., Hoor, P., Jegou, F., Jucks, K. W., Kleinböhl, A., Küllmann, H., Kort, E. A., McKain, K., Moore, F. L., Obersteiner, F., Ramos, Y. G., Schuck, T., Toon, G. C., Viciani, S., Wetzels, G., Williams, J., and Wofsy, S. C.: N₂O Temporal Variability from the Middle Troposphere to the Middle Stratosphere Based on Airborne and Balloon-Borne Observations during the Period 1987–2018, *Atmosphere*, 14, 585, <https://doi.org/10.3390/atmos14030585>, 2023a.
- Krysztofiak, G., Catoire, V., Witt, T. D. d., Kinnison, D., Ravishankara, A. R., Brocchi, V., Elliot Atlas, Bozem, H., Commane, R., D’Amato, F., Daube, B., Diskin, G. S., Engel, A., Friedl-Vallon, F., Hintsä, E., Hurst, D. F., Hoor, P., Jegou, F., Jucks, K. W., Kleinböhl, A., Küllmann, H., Kort, E., McKain, K., Moore, F. L., Obersteiner, F., Ramos, Y. G., Schuck, T., Toon, G. C., Viciani, S., Wetzels, G., Williams, J., and Wofsy, S. C.: N₂O mixing ratio zonal mean and trend in the UTLS from aircraft and balloon measurements, <https://doi.org/10.25326/428>, 2023b.
- Kunz, A., Konopka, P., Müller, R., and Pan, L. L.: Dynamical tropopause based on isentropic potential vorticity gradients, *Journal of Geophysical Research*, 116, D01 110, <https://doi.org/10.1029/2010JD014343>, 2011.
- Lachnitt, H.-C.: Consistent meteorological (ERA5) and chemical (CLaMS) information across airborne measurements over the globe between 1997 and 2023, <https://doi.org/10.5281/zenodo.15076520>, 2025.
- Lebague, B., Schmidt, M., Ramonet, M., Wastine, B., Kwok, C. Y., Laurent, O., Belviso, S., Guemri, A., Philippon, C., Smith, J., and Conil, S.: Comparison of nitrous oxide (N₂O) analyzers for high-precision measurements of atmospheric mole fractions, *Atmos. Meas. Tech.*, pp. 1221–1238, <https://doi.org/10.5194/amt-9-1221-2016>, 2016.
- Meng, L., Liu, J., Tarasick, D. W., and Li, Y.: Biases of Global Tropopause Altitude Products in Reanalyses and Implications for Estimates of Tropospheric Column Ozone, *Atmosphere*, 12, 417, <https://doi.org/10.3390/atmos12040417>, 2021.
- Millán, L. F., Manney, G. L., Boenisch, H., Hegglin, M. I., Hoor, P., Kunkel, D., Leblanc, T., Petropavlovskikh, I., Walker, K., Wargan, K., and Zahn, A.: Multi-parameter dynamical diagnostics for upper tropospheric and lower stratospheric studies, *Atmospheric Measurement Techniques*, 16, 2957–2988, <https://doi.org/10.5194/amt-16-2957-2023>, 2023.
- Müller, S.: Untersuchung von Mischungs- und Transportprozessen in der oberen Troposphäre - unteren Stratosphäre basierend auf in-situ Spurengasmessungen, Ph.D. thesis, Johannes Gutenberg-Universität Mainz, Mainz, <https://doi.org/10.25358/openscience-1807>, 2015.
- Obersteiner, F.: FAIROmeta, <https://doi.org/10.5281/zenodo.11275355>, 2024.
- Pan, L. L., Randel, W. J., Gary, B. L., Mahoney, M. J., and Hintsä, E. J.: Definitions and sharpness of the extratropical tropopause: A trace gas perspective, *Journal of Geophysical Research D: Atmospheres*, 109, 1–11, <https://doi.org/10.1029/2004JD004982>, 2004.



- Petetin, H., Thouret, V., Fontaine, A., Sauvage, B., Athier, G., Blot, R., Boulanger, D., Cousin, J.-M., and Nédélec, P.: Characterising tropospheric O₃ and CO around Frankfurt over the period 1994–2012 based on MOZAIC–IAGOS aircraft measurements, *Atmospheric Chemistry and Physics*, 16, 15 147–15 163, <https://doi.org/10.5194/acp-16-15147-2016>, 2016.
- Prather, M. J., Hsu, J., DeLuca, N. M., Jackman, C. H., Oman, L. D., Douglass, A. R., Fleming, E. L., Strahan, S. E., Steenrod, S. D., Søvde, O. A., Isaksen, I. S. A., Froidevaux, L., and Funke, B.: Measuring and modeling the lifetime of nitrous oxide including its variability, *Journal of Geophysical Research: Atmospheres*, 120, 5693–5705, <https://doi.org/10.1002/2015JD023267>, <https://agupubs.onlinelibrary.wiley.com/doi/pdf/10.1002/2015JD023267>, 2015.
- Prinn, R. G., Weiss, R. F., Arduini, J., Arnold, T., DeWitt, H. L., Fraser, P. J., Ganesan, A. L., Gasore, J., Harth, C. M., Hermansen, O., Kim, J., Krummel, P. B., Li, S., Loh, Z. M., Lunder, C. R., Maione, M., Manning, A. J., Miller, B. R., Mitrevski, B., Mühle, J., O’Doherty, S., Park, S., Reimann, S., Rigby, M., Saito, T., Salameh, P. K., Schmidt, R., Simmonds, P. G., Steele, L. P., Vollmer, M. K., Wang, R. H., Yao, B., Yokouchi, Y., Young, D., and Zhou, L.: History of chemically and radiatively important atmospheric gases from the Advanced Global Atmospheric Gases Experiment (AGAGE), *Earth System Science Data*, 10, 985–1018, <https://doi.org/10.5194/essd-10-985-2018>, 2018.
- Riese, M., Hoor, P., Rolf, C., and Kunkel, D.: Quasi-horizontal transport of Asian summer monsoon air during the PHILEAS campaign in summer and autumn 2023, Tech. Rep. EGU24-6647, Copernicus Meetings, <https://doi.org/10.5194/egusphere-egu24-6647>, conference Name: EGU24, 2024.
- Schuck, T. and Obersteiner, F.: IAGOS-CARIBIC whole air sampler data (v2024.01.12), <https://doi.org/10.5281/zenodo.10495039>, 2024.
- Schuck, T. J., Brenninkmeijer, C. A. M., Slemr, F., Xueref-Remy, I., and Zahn, A.: Greenhouse gas analysis of air samples collected onboard the CARIBIC passenger aircraft, *Atmospheric Measurement Techniques*, 2, 449–464, <https://doi.org/10.5194/amt-2-449-2009>, publication Title: Atmos. Meas. Tech Volume: 2, 2009.
- SPARC: SPARC Report N°6. Lifetimes of Stratospheric Ozone-Depleting Substances, Their Replacements, and Related Species, SPARC Report 6, SPARC, <https://www.aparc-climate.org/publications/sparc-reports/sparc-report-no-6/>, 2013.
- Sprenger, M., Croci Maspoli, M., and Wernli, H.: Tropopause folds and cross-tropopause exchange: A global investigation based upon ECMWF analyses for the time period March 2000 to February 2001, *Journal of Geophysical Research: Atmospheres*, 108, <https://doi.org/10.1029/2002JD002587>, 2003.
- Sprung, D. and Zahn, A.: Acetone in the upper troposphere/lowermost stratosphere measured by the CARIBIC passenger aircraft: Distribution, seasonal cycle, and variability, *Journal of Geophysical Research: Atmospheres*, 115, <https://doi.org/10.1029/2009JD012099>, <https://onlinelibrary.wiley.com/doi/pdf/10.1029/2009JD012099>, 2010.
- Tegtmeier, S., Anstey, J., Davis, S., Dragani, R., Harada, Y., Ivanciu, I., Pilch Kedzierski, R., Krüger, K., Legras, B., Long, C., Wang, J. S., Wargan, K., and Wright, J. S.: Temperature and tropopause characteristics from reanalyses data in the tropical tropopause layer, *Atmospheric Chemistry and Physics*, 20, 753–770, <https://doi.org/10.5194/acp-20-753-2020>, 2020.
- Thouret, V., Cammas, J.-P., Sauvage, B., Athier, G., Zbinden, R., Nédélec, P., Simon, P., and Karcher, F.: Tropopause referenced ozone climatology and inter-annual variability (1994–2003) from the MOZAIC programme, *Atmospheric Chemistry and Physics*, 6, 1033–1051, <https://doi.org/10.5194/acp-6-1033-2006>, 2006.
- Turhal, K., Plöger, F., Clemens, J., Birner, T., Weyland, F., Konopka, P., and Hoor, P.: Variability and trends in the potential vorticity (PV)-gradient dynamical tropopause, *Atmospheric Chemistry and Physics*, 24, 13 653–13 679, <https://doi.org/10.5194/acp-24-13653-2024>, 2024.
- WMO: Definition of the tropopause, *WMO Bulletin*, 6, 137, <https://library.wmo.int/idurl/4/42003>, 1957.



- WMO/GAW Ozone Monitoring Community, W. M. O.-G. A. W. P. W.-G. O. a. U. R. D. C. W.: OzoneSonde Dataset. A list of all contributors is available on the website., <https://doi.org/10.14287/10000008>, 2025.
- Xian, T. and Homeyer, C. R.: Global tropopause altitudes in radiosondes and reanalyses, *Atmospheric Chemistry and Physics*, 19, 5661–5678, <https://doi.org/10.5194/acp-19-5661-2019>, 2019.
- 510 Zahn, A., Brenninkmeijer, C. A. M., and Van Velthoven, P. F. J.: Passenger aircraft project CARIBIC 1997-2002, Part I: the extratropical chemical tropopause, *Atmos. Chem. Phys. Discuss.*, 4, 1091–1117, <https://doi.org/10.5194/acpd-4-1091-2004>, publication Title: Atmos. Chem. Phys. Discuss Volume: 4, 2004.
- Zahn, A., Obersteiner, F., Gehrlein, T., Neumaier, M., Dyroff, C., Förster, E., Sprung, D., Van Velthoven, P., Xueref-Remy, I., Ziereis, H., Bundke, U., Gerbig, C., Scharffe, D., Slemr, F., Weber, S., Hermann, M., Cheng, Y., and Bönisch, H.: IAGOS-CARIBIC MS files
- 515 collection (v2024.10.28), <https://doi.org/10.5281/zenodo.14000090>, 2024.