Public justification (visible to the public if the article is accepted and published):

Dear authors,

Thank you for addressing the reviewer comments. There are some minor comments pending before the manuscript can be accepted for publication, see below.

Thank you for reviewing our revised manuscript. We have revised the manuscript accordingly.

L54 … with near global coverage on seasonal timescales or …with near global coverage every 3 months

L54: We revised the sentence to "with near global coverage every three months".

L60 please add Waters et al 2006 10.1109/TGRS.2006.873771 after MLS

L60: We added the reference Waters et al 2006 after MLS.

L100 what is the precision and accuracy for Methane

L100 could you just say 660 ppt?

L100: We revised the text as follows: "The precision and accuracy are 660 ppt and less than 2% for CO, and 1200 ppt and 5-20% for CH_4, respectively (Schmidt et al., 2024)."

L127 even though you explained how the tropopause was calculated in the response to reviewers; it needs to be reflected here.

L127: we revised the text as follows: "Here, we focus on eight CH_4 and CO profiles to compare with ACE-FTS measurements and to highlight their contrasting structures near the tropopause, identified from collocated temperature profiles using the WMO definition (Fig. 1)."

L214: Please add the tropopause line to figure 3. The one shown Fig 2 for air core. And one derived for ACCLIP (note that the AIRCORE and ACCLIP related tropopauses seem to be at different heights).

We have added the tropopause for both AirCore and ACCLIP to Figure 3. The difference in tropopause height reflects the different geographic regions sampled: the AirCore campaign took place over the Tibetan Plateau, whereas ACCLIP sampled the western Pacific.

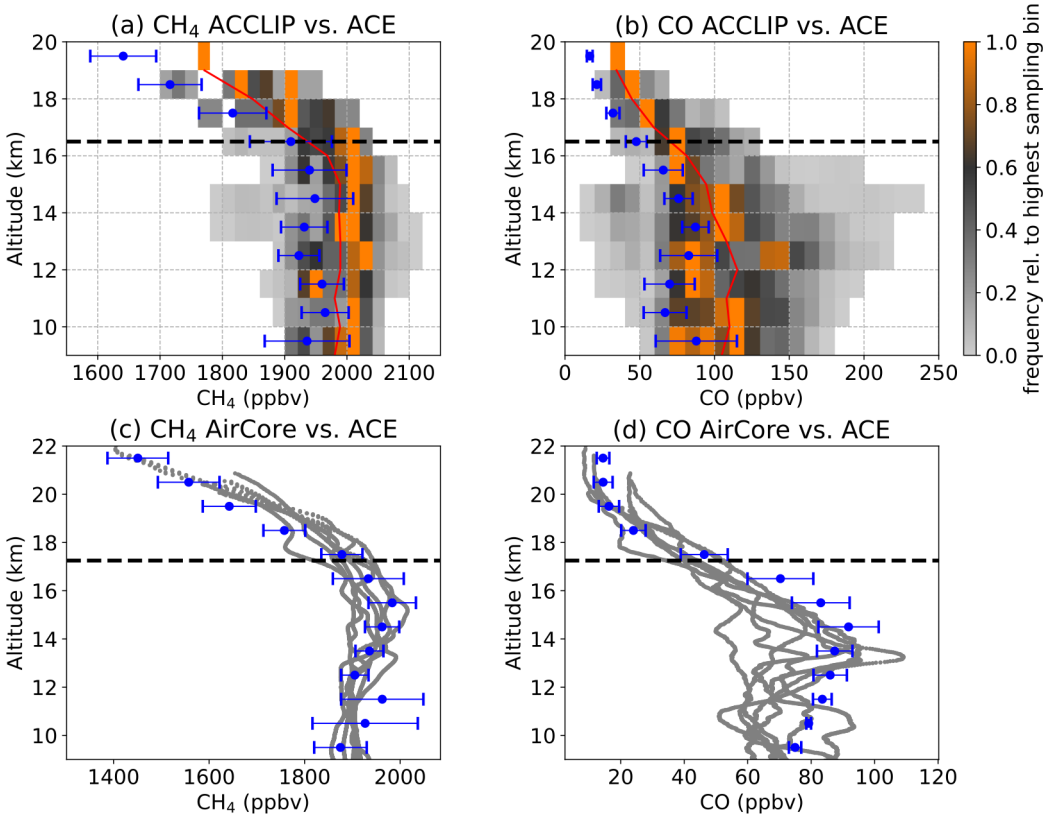


Figure 3: Comparison of ACE-FTS measurements with in-situ observations in the ASM region. (a) Layer-normalized relative frequency distributions of CH_4 measured during the ACCLIP campaign, with horizontal and vertical bin sizes of 20 ppbv and 0.5 km, respectively. The red line shows the mean CH_4 mixing ratio for each vertical bin level. The blue dots represent the mean vertical CH_4 profile measured by ACE-FTS, and the blue error bars indicate one sigma standard deviation at each level. The black dashed line indicates the mean tropopause determined from the collocated temperature profiles using the WMO definition; (b) Same as (a), but for CO. (c) Comparison between the AirCore CH_4 profiles (gray lines) and the mean ACE-FTS profile (blue dots and error bars, as in panel a). (d) Same as (c), but for CO.

L230. The stream function doesn't seem to be capturing the ASM anticyclone, or at least the enhancements on CO. I think the levels perhaps need to be fine-tuned per height (Pan et al use 4 and 25 at 150hPA). Please confirm that you are using the correct levels.

Figure 4: In the original Figure 4b, the stream function contours shown at 100 hPa, as in Figure 4a. We have revised Figure 4b to show the contours at 150 hPa, using levels of 4, 14 and 25 following Pan et al. The revised contours better delineate the ASM anticyclone and its spatial correspondence with the CO enhancement.

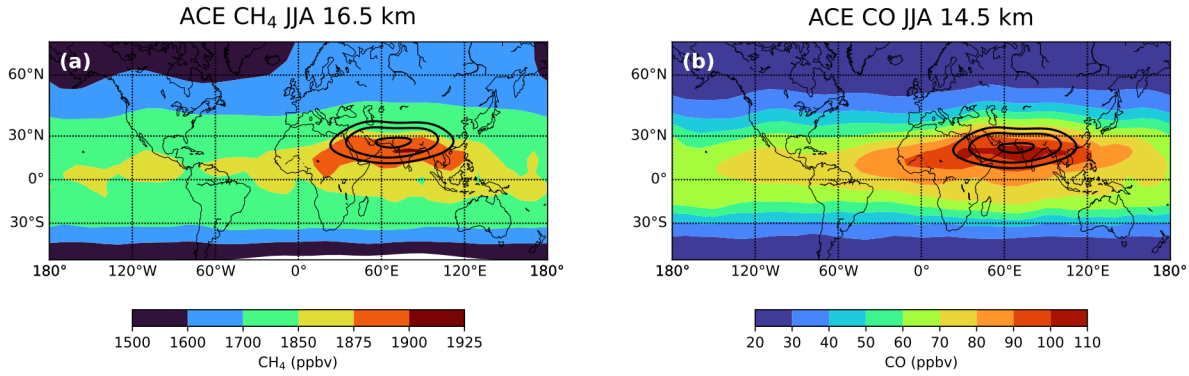


Figure 4: Horizontal distributions of JJA climatological mixing ratios from ACE-FTS measurements during 2005-2023. (a) CH_4 at 16.5 km and (b) CO at 14.5 km. The black solid contours show selected stream function levels ($[4, 14, 25] \times 10^6\ m^2\ s^{-1}$; based on the ERA5 data) at the corresponding altitudes, averaged over JJA during 2005-2023, to indicate the climatological location and horizontal extent of the ASM anticyclone. Note that the JJA map predominantly represents measurements over the midlatitudes in June, the northern subpolar region in July, and the tropics in August (see Fig. 1), rather than spatially uniform sampling throughout JJA.

Figure 6. Please use a regular scaling for the colorbars (i.e., every 10 for panel a, and every 4 for panel b, for example). In panel a, the jumps between colors are not even linear. In panel b, the white region encompasses a really broad region. It seems that you are cherry picking your results.

We have revised Figure 6 to use uniformly spaced contour levels, with intervals of 10 ppbv in panel (a) and 4 ppbv in panel (b). This removes the broad white region, providing a more consistent representation of the data.

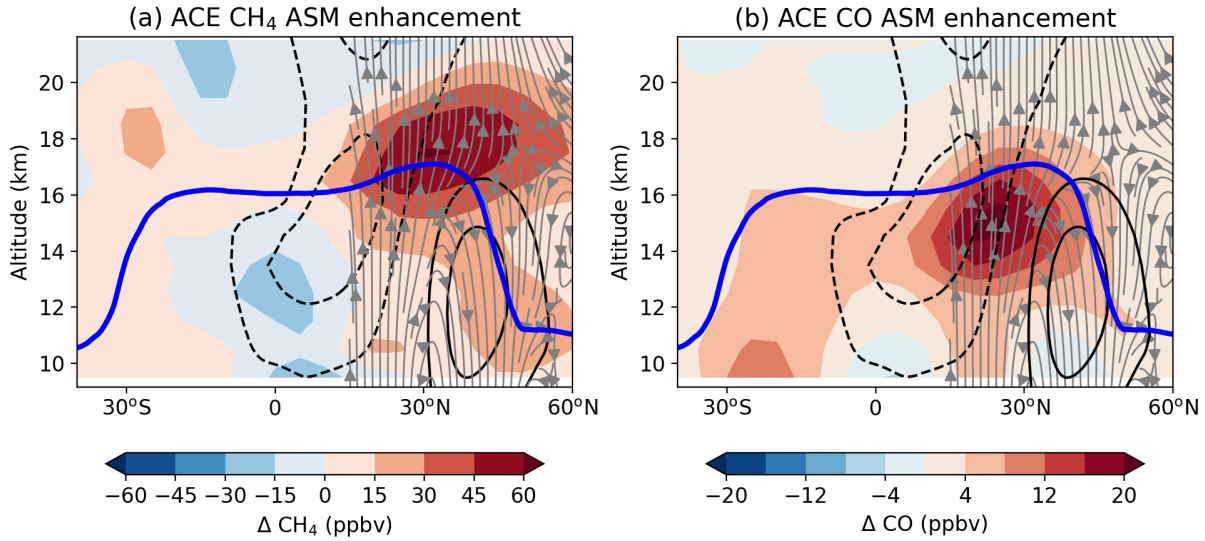


Figure 6: Meridional cross sections of the ASM enhancement in (a) CH_4 and (b) CO derived from ACE-FTS observations for JJA during 2005-2023, defined as the mean mixing ratio over 20°-120°E minus the background mean over all remaining longitudes. Blue lines indicate the lapse-rate tropopause provided by Hoffmann et al., 2022. Solid and dashed black contours represent westerly (10 and 20 m/s) and easterly winds (-10 and -20 m/s), respectively. Gray streamlines, shown only between 15-60°N to cover the ASM region and higher latitude potentially influenced by ASM transport, depict the circulation in the latitude-altitude plane and are constructed from the meridional wind (m/s) and vertical velocity (mm/s). All meteorological fields are averaged over 20-120°E during JJA 2005-2023 based on ERA5 reanalysis. Note that the vertical velocity is converted from pressure-coordinate vertical velocity using the hydrostatic relation.

Reviewer 1

Review of egusphere-2026-2644: “Methane in the Asian Monsoon Anticyclone and global UTLS behavior observed in ACE-FTS satellite data“ by You Yi et al.

We thank the reviewer for the positive comments and valuable suggestions. We have revised the manuscript and please see the track-changes file for these revisions. All line numbers in our responses refer to the clean version of the revised manuscript.

The manuscript by You Yi and colleagues addresses the impact of the Asian Summer Monsoon (ASM) on the distribution of methane in the upper troposphere and lower stratosphere (UTLS). For this the authors use satellite data from ACE-FTS to describe the vertical and horizontal distribution of methane as well as the seasonal cycle of methane in this region. They show that methane has its maximum at higher altitudes than a comparable tracer, CO, and that the decrease of methane concentrations into the stratosphere starts at higher altitudes than for CO. This is attributed to the longer lifetime of methane. Their results are supported by in situ observations from AirCore and aircraft measurements. Together with data for surface methane emissions, they can also demonstrate the effectiveness of the lifting process within the ASM – while emissions peak during summer, the peak methane concentrations in the UTLS occur in autumn. The authors also analyze trends based on the ACE-FTS data which are in line with previous findings.

Methane is a highly relevant constituent in the atmosphere with increasing importance in discussions centered around climate change. Analysis of the global distribution and sources of methane are of relevance and fit well into the scope of ACP. The manuscript addresses an interesting question, particularly based on observational data. Such data is often required to validate chemistry climate models and might also be relevant for trend studies. Therefore, the paper will be a great addition to the scientific literature. In my opinion, the manuscript is overall already in a good shape. The scientific reasoning is sound, the figures are appropriate and fit to the story. The overall thematic flow of the paper is clear and comprehensible. However, this is the case if the reader is already in an expert in the topic. For all others, I think the manuscript would benefit from more information and extended discussion at several

points. This is also my main concern on which I will give some examples below. Otherwise, I think the paper is on a good path to be published and I recommend a publication after minor revision. I choose minor revision here because my comments are mainly about providing more information and not about a new or extended analysis or about reasoning.

My major concern is that although all relevant information is touched upon in each section, the often very brief presentation of the content gives the impression that the authors have reduced the material too much. Since this occurs in many places throughout the manuscript, the overall impression is that important information necessary for understanding and proper contextualization is missing or treated too superficially. Some examples are given below:

We have expanded the relevant sections, as detailed in our responses to the specific comments below.

1. In the introduction, I am missing a sentence or two about methane sources, especially in South-East Asia to emphasize why this region is so important for the global methane budget not only from a transport perspective but also from an emission perspective. This could be part of the first paragraph (l. 28ff) or of the paragraph starting in line 43.

We added two sentences to describe the methane sources in south-east Asia and emphasize the strong seasonality of the sources.

In lines 40-42, “South and Southeast Asia are major CH_4 source regions, with emissions dominated by agriculture (including rice cultivation and livestock), waste, wetlands. Emissions from these sources increase during the ASM season, likely due to enhanced rainfall (Yu et al., 2023; East et al., 2024; Subramanian et al., 2026).”

2. Since CO is also widely discussed throughout the manuscript, it might deserve a small introductory paragraph, at least to contrast it to methane in the introduction.

We added some introductions of ACE-FTS CO related studies in lines 62-75. “Previous studies have used ACE-FTS observations to investigate the regional transport signals and large-scale tracer variability. Park et al (2008) documented enhanced CO and other tropospheric tracers within the Asian monsoon anticyclone, providing evidence of

convective transport and chemical isolation. CO is an effective tracer of vertical transport because its free-tropospheric lifetime (~1 month) is substantially longer than typical deep convective transport timescales. Enhanced CO in the UTLS can therefore indicate recent deep convective transport of polluted air, while correlations between CO and other trace gases can help identify source characteristics and common transport pathways.

Park et al. (2013) examined the global distributions and seasonal variability of CO and hydrocarbons in the UTLS based on ACE-FTS measurements. These tracers exhibited semi-annual cycle in the tropical upper troposphere related to seasonally varying sources and deep tropical convection, and a maximum tied to the Asian monsoon anticyclone. Above the tropical tropopause, these species reveal a strong annual cycle related to annual variation of the upward branch of BDC."

3. The paragraph about ACE-FTS (l. 47ff) would benefit from some references. There are several statements in this five lines which could be filled with references. Also it would be good to inform the reader about other methane (and CO) related studies with ACE-FTS to put the results from this study more in perspective.

We added references Bernath et al., 2005 and Nassar et al., 2005 in line 56 and line 58, respectively. We added the paragraph describing the ACE-FTS methane and CO related studies on line 62-78.

"Previous studies have used ACE-FTS observations to investigate the regional transport signals and large-scale tracer variability. Park et al (2008) documented enhanced CO and other tropospheric tracers within the Asian monsoon anticyclone, providing evidence of convective transport and chemical isolation. CO is an effective tracer of vertical transport because its free-tropospheric lifetime is substantially longer than typical deep convective transport timescales. Enhanced CO in the UTLS can therefore indicate recent deep convective transport of polluted air, while correlations between CO and other trace gases can help identify source characteristics and common transport pathways.

Park et al. (2013) examined the global distributions and seasonal variability of CO and hydrocarbons in the UTLS. These tracers exhibited semi-annual cycle in the tropical upper

troposphere related to seasonally varying sources and deep tropical convection, and a maximum tied to the Asian monsoon anticyclone. Above the tropical tropopause, these species reveal a strong annual cycle related to annual variation of the upward branch of BDC. More recently, Sheese et al. (2025) used ACE-FTS measurements, including CH_4, to quantify the contributions of methane oxidation and circulation changes to stratospheric water vapor trends. To address the limited tropical sampling, they supplemented ACE-FTS observations with model simulations and applied multiple linear regression (MLR) to estimate CH_4 trends entering the stratosphere. ”

4. Line 62: What is the wavenumber 0.02 cm-1? Please specify.

We have revised the sentence in lines 89-91. “The Atmospheric Chemistry Experiment–Fourier Transform Spectrometer (ACE-FTS), onboard the Canadian SCISAT satellite launched in 2003, is a high-resolution infrared Michelson spectrometer with a spectral resolution of 0.02 cm^{-1} that measures solar occultation transmission spectra between 2.2–13.3 μm (Bernath et al., 2005).”

5. Line 68: Here you mean effective horizontal resolution? What is meant with oversampled at 1 km?

The ~3 km resolution refers to the effective vertical resolution, rather than the horizontal resolution. By “oversampled on a 1 km grid”, we mean that the retrieved CH_4 profiles are reported on a vertical grid with 1 km spacing, which is finer than the effective vertical resolution of approximately 3 km. We have revised the sentence to clarify this point in lines 95-96.

“This study uses the recent version of level 2 data, version 5.2 (v5.2), which provides interpolated data on a 1 km grid (Boone et al., 2023).”

6. Is there a specific reason why no measurement error is given for the AirCore data? (section 2.2.1).

We added one sentence to describe the measurement uncertainty in lines 123-124. “The average measurement uncertainty of CH_4 and CO are less than 10 ppb and 5 ppb below 20 km.”

7. Figure 1: the lapse-rate tropopause shown in this figure is derived from which data? Does this lapse-rate vary much between the individual flights? I asked whether I would like to know whether the differences between methane and CO which are discussed here might become even more prominent when all profiles would be plotted in a tropopause relative coordinate system.

In original figure 1 (now figure 2), the lapse-rate tropopause was determined from the temperature profiles measured by the radiosondes launched together with the AirCore, following the WMO definition. The tropopause altitude does vary but not too much (17.5 km$\pm$ 0.5 km) among the individual flights, because all flights were conducted at the same location in August. Consequently, plotting the profiles relative to the tropopause produces minor changes.

8. In section 3 it is often mentioned that ERA5 data has been used. Since this occurs at multiple occasions, I would suggest to include a small subsection in Section 2 about the ERA5 data used in this study.

We have added a paragraph describing the ERA5 reanalysis used in the study on section 2.3.

Lines 152-159: “Meteorological fields from ERA5, the fifth-generation global reanalysis produced by the European Centre for Medium-Range Weather Forecasts (ECMWF; Hersbach et al., 2020), are used to characterize the circulation associated with the observed CH_4 and CO distributions. We use ERA5 monthly pressure-level data provided on a regular 0.25° latitude-longitude grid, with 37 standard pressure levels ranging from 1000 to 1 hPa. The time periods are from 2005 to 2023, matching the period of the ACE-FTS analysis. ERA5-based lapse-rate tropopause heights are obtained from Hoffmann and Spang (2022). This product identifies the lapse-rate tropopause using the World Meteorological Organization

(WMO) definition in 1957. These heights provide a reference for interpreting the observed CH_4 and CO distributions relative to the tropopause."

9. In Figure 3 the black lines show a stream function, how is this calculated? Which years are used, which values of the stream function are shown here and does this explain why the lines are only evident in the region of the ASM?

The horizontal stream function is calculated from ERA5 zonal and meridional winds using the Python package windspharm. The stream function is calculated by

$$\psi = \nabla_h^{-2}\zeta$$

Here, ψ is stream function, unit $m^2\ s^{-1}$; ζ is relative vorticity, unit s^{-1}.

The stream function was averaged over JJA during 2005-2023. The selected contour levels are [4, 14, 25]$\times 10^6$ $m^2\ s^{-1}$, chosen to illustrate the climatological location and horizontal extent of the ASM anticyclone, following the approach of Pan et al (2024). doi: 10.1073/pnas.2318716121.

We revise the Figure 4 (original Figure 3) caption to include the details. "Figure 4: Horizontal distributions of JJA climatological mixing ratios from ACE-FTS observations during 2005-2023. (a) CH_4 at 16.5 km and (b) CO at 14.5 km. The black solid contours show selected stream function levels ([4, 14, 25]$\times 10^6$ $m^2\ s^{-1}$; based on the ERA5 data) at the corresponding altitudes, averaged over JJA during 2005-2023, to indicate the climatological location and horizontal extent of the ASM anticyclone. Note that the JJA map predominantly represents measurements over the midlatitudes in June, the northern subpolar region in July, and the tropics in August (see Fig. 1), rather than spatially uniform sampling throughout JJA."

10. Also in Figure 3 it is not fully clear to me why 16.5 and 14.5 km have been chosen here for methane and CO? The text says that the maximum of CH4 is around 16 km, and for CO 14 km. From Figure 4 would follow that the maximum for CH4 is more 15.5 km and for CO 13.5 km to 14 km.

The levels of 16.5 km for CH_4 and 14.5 km for CO were selected because the horizontal enhancement associated with the ASM anticyclone is particularly clear at these levels. The sampling density in the ASM region is greater at higher altitudes than at lower altitudes. The heights of approximately 16 km for CH_4 and 14 km for CO quoted in the text were intended as approximate descriptions of the peak altitudes, rather than precise values. Given the 1 km vertical grid spacing in ACE-FTS profiles, we choose the level of 16.5 km for CH_4 and 14.5 km for CO where the maximum is mostly clear.

11. The lapse rate tropopause in Figure 4: has this been calculated by yourself? If so how and on which ERA5 data (pressure level, model level, monthly mean, daily, hourly data, horizontal and temporal resolution)?

The lapse rate tropopause shown here is downloaded from https://datapub.fz-juelich.de/slcs/tropopause/ (Hoffmann et al., 2022). We revise the Fig. 5 (original Fig. 4) caption to clearly state the data source. "Figure 5: Meridional cross-sections of JJA climatological mixing ratios over the ASM region (20–120°E), derived from the horizontal maps shown in Fig. 4. Observed (a) CH_4 mixing ratios and (b) CO mixing ratios from ACE-FTS. The white dashed lines denote the JJA climatological lapse-rate tropopause obtained from ERA5 reanalysis during 2005-2023 provided by Hoffmann et al., 2022." We have also introduced the tropopause product in sect. 2.3.

12. In Figure 5, the gray lines show the stream function: what is meant with composite here? Do I assume correctly that the data source is again ERA5? And did you calculate the vertical wind from the Lagrangian tendency of the pressure (omega)?

The gray lines are streamlines constructed from the meridional wind and vertical velocity. Our use of "composite" was imprecise. We intended to describe the combination of these two velocity components used to generate the streamlines. Both components are based on ERA5 data and averaged over 20-120°E during JJA 2005-2023. The vertical velocity was converted from the ERA5 pressure vertical velocity ($\omega = \frac{dP}{dt}$) using the hydrostatic relation

$w = -\frac{\omega}{\rho g}$. We have revised the caption to clarify the meaning of the gray streamlines and added the vertical velocity conversion statement.

"Figure 6: Meridional cross sections of the ASM enhancement in (a) CH_4 and (b) CO derived from ACE-FTS observations for JJA during 2005-2023, defined as the mean mixing ratio over 20°-120°E minus the background mean over all remaining longitudes. Blue lines indicate the lapse-rate tropopause provided by Hoffmann et al., 2021. Solid and dashed black contours represent westerly (10 and 20 m/s) and easterly winds (-10 and -20 m/s), respectively. Gray streamlines, shown only between 15-60°N to cover the ASM region and higher latitude potentially influenced by ASM transport, depict the circulation in the latitude-altitude plane and are constructed from the meridional wind (m/s) and vertical velocity (mm/s). All meteorological fields are averaged over 20-120°E during JJA 2005-2023 based on ERA5 reanalysis. Note that the vertical velocity is converted from pressure-coordinate vertical velocity using the hydrostatic relation."

13. Why are there only gray lines between ~15°N to 60°N? And finally, I would have expected that th the upwelling (tropics) and the downwelling (extratropics) would be evident in the stratosphere, but the arrows along the streamlines all point upward. Is this related to the region and time?

The gray streamlines are restricted to approximately 15-60°N because we focus on the ASM region and the higher-latitude regions influenced by ASM transport. They are constructed from the ERA5 meridional wind and vertical velocity averaged over 20-120°E and depict the circulation in the latitude-altitude plane within this longitude sector.

The tropical upwelling and extratropical downwelling mentioned by the reviewer characterize the Brewer-Dobson circulation, which is generally described using the zonal-mean residual circulation. Our streamlines instead represent the 20-120°E longitudinal mean wind field. Park et al. (2007) pointed out the upward circulation above the tropopause inside ASM anticyclone in the extra-tropical region. The upward structure shown in the Fig. 6 (original Fig. 5) therefore depends on the selected region and period.

14. The blue line is again the ERA5 lapse-rate tropopause? And the contour lines for the horizontal wind show magnitudes of +10 m/s and +20 m/s (solid) and -10 m/s and -20 m/s (dashed)?

Yes. We revised the Fig. 6 (original Fig. 5) caption to clearly describe the variable shown in the figure. "Figure 6: Meridional cross sections of the ASM enhancement in (a) CH_4 and (b) CO derived from ACE-FTS observations for JJA during 2005-2023, defined as the mean mixing ratio over 20°-120°E minus the background mean over all remaining longitudes. Blue lines indicate the lapse-rate tropopause provided by Hoffmann et al., 2021. Solid and dashed black contours represent westerly (10 and 20 m/s) and easterly winds (-10 and -20 m/s), respectively. Gray streamlines, shown only between 15-60°N to cover the ASM region and higher latitude potentially influenced by ASM transport, depict the circulation in the latitude-altitude plane and are constructed from the meridional wind (m/s) and vertical velocity (mm/s). All meteorological fields are averaged over 20-120°E during JJA 2005-2023 based on ERA5 reanalysis. Note that the vertical velocity is converted from pressure-coordinate vertical velocity using the hydrostatic relation."

15. In Figure 7, OLR data is shown? Where does this come from and what exactly is shown here? At some places it seems that multiple lines are evident and not just one line for 220 K.

OLR data is obtained from Clouds and Earth's Radiant Energy System Energy Balanced and Filled Top-of-Atmosphere Monthly means data product. The product uses reprocessed NOAA-20 data using GMAO MERRA-2 as the meteorology starting with April 2022 for fluxes and VIIRS clouds and scene type determination. The lines shown here are from 190 W/m^2 to 220 W/m^2 with interval of 10 W/m^2. We have added the OLR data description in the Section 2.3 in lines 159-164.

"Monthly mean outgoing longwave radiation (OLR) for 2005-2023 is obtained from the Clouds and the Earth's Radiant Energy System (CERES) Energy Balanced and Filled top-of-atmosphere Edition 4.2 product (Loeb et al., 2017, 2024). We us the all-sky Top-of-Atmosphere (TOA) outgoing longwave flux, provided on a 1° latitude-longitude grid in W/m^2. Regions with OLR less than 220 W/m^2 are used as a proxy for deep convection. For

comparison with ACE-FTS observations in the tropics, climatological OLR fields are calculated for February, April, August, and October."

The OLR data shown in the figure are the values less than 220 K (OLR<=220 K) to indicate deep convection. We choose the February for DJF, April for MAM, August for JJA and October for SON season to keep consistent with ACE-FTS observations in the tropics. We revised the caption to fully illustrate the data used in the figure.

"Figure 8: Climatological horizontal distributions of CH_4 at 16.5 km for (a) DJF, (b) MAM, (c) JJA and (d) SON seasons derived from ACE-FTS observations gridded at 5° latitude by 10° longitude, averaged over the period 2005-2023. The white contours represent outgoing longwave radiation (OLR) from 190 W/m^2 to 220 W/m^2 with interval of 10 W/m^2, with lower values used as proxy for enhanced deep convection. Note that in the tropics, DJF, MAM, JJA and SON primarily represent observations in February, April, August and October, respectively. The OLR data are monthly means for February, April, August, and October in panels (a)-(d), respectively, to match the tropical ACE-FTS sampling."

16. For the discussion around and the data in Figure 8, the region is extended to 15°S- 30°N and the entire zonal direction. How well does this represent the ASM region?

The domain used in Fig.9 (15°S-30°N, encompassing all longitudes) is intended to examine the potential influence of ASM transport on the broader tropical CH_4 distribution, rather than to represent the ASM region itself.

17. Also for Figure 8 and the surface emission data: Can you include some more information about this surface data set, so that is not necessary to go the Saunois et al. paper.

We added one paragraph in section 2.3 to describe the surface flux in lines 166-176.

"The CH_4 surface flux dataset provided by Saunois et al. (2025) is constructed by combining bottom-up emission inventories and model estimate for individual source and sink categories. xThe prior flux draws on Emission Database for Global Atmospheric Research (EDGAR v7; Crippa et al., 2023) for anthropogenic emissions, with extension based on Food

and Agriculture Organization (FAO) agriculture data and includes an alternative configuration using the Greenhouse gas and Air pollutant Interactions and Synergies (GAINS) model for fossil fuel emissions (Höglund-Isaksson et al., 2020). Biomass-burning emissions are obtained from Global Fire Emission Database (GFED) v4.1 (van der Werf et al., 2017), and wetland emissions are represented by the monthly ensemble mean of 11 models reported by Saunois et al. (2020). Additional components include termite emissions, ocean emissions from Weber et al. (2019), geological emissions from Etiope et al. (2019), and soil methane uptake from the MeMo model (Murguia-Flores et al., 2018). These components are assembled into gridded surface flux fields for use as prior inputs to atmospheric inversions, with temporal variability determined by the underlying inventories and models."

18. Is the LOTUS regression model the one available here on this web page: https://usask-arg.github.io/lotus-regression/. Can you provide further details about the model in your text, especially, on your setup (proxies and input data).

We added the model description in the Section 2.4 in lines 178-193.

"To estimate the trends and reconstruct monthly methane time series in the tropics, we apply the Long-term Ozone Trends and Uncertainties in the Stratosphere (LOTUS) regression code (SPARC/IO3C/GAW, 2019), following the standard multiple linear regression (MLR) framework described by Dubé et al. (2023). The model is fitted separately to the available observations in each latitude-altitude bin:

$$\begin{aligned} CH_4(t) = {} & \beta^{(2)}(t) + \beta_{trend} \times linear(t) + \\ & \beta^{(2)}_{qboa} \times QBO_{(a)}(t) + \beta^{(2)}_{qbob} \times QBO_{(b)}(t) + \\ & \beta_{solar} \times F_{10.7}(t) + \beta_{enso} \times ENSO(t) + \\ & \beta_{aod} \times AOD(t) + R(t) \end{aligned} \quad (1)$$

Here, each β is a regression coefficient for each term, the superscripts are the number of highest seasonal harmonic for a given term, $\beta^{(2)}(t) = const + A_1 \sin(2\pi t) + B_1 \cos(2\pi t) + A_2 \sin(4\pi t) + B_2 \cos(4\pi t)$, A_i and B_i are the sine and cosine coefficients, respectively, with

subscripts $i = 1$ and $i = 2$ denote the annual and semi-annual cycles. Time t is expressed in years. $QBO_{(a)}(t)$ and $QBO_{(b)}(t)$ are the first two principal components of the Singapore zonal winds, $F_{10.7}(t)$ is the solar flux at 10.7 cm, ENSO (t) is the multivariate ENSO index, and $R(t)$ is the residual. These predictors are further described in SPARC/IO3C/GAW (2019). Dubé et al (2023) applied this standard MLR framework to estimate stratospheric N_2O trends from ACE-FTS observations. ”

19. Figure 10 b): which bin size has been used here for this plot (in delta z and delta lat)? And could you provide potentially one sentence why the trend is larger at the tropical tropopause than above? Is this upwelling related or has this to do with the strength of lateral mixing?

A possible explanation is that CH_4 near the tropical tropopause responds more directly to the accelerating tropospheric increase, whereas air at higher reflects an earlier tropospheric CH_4 trends. This can lead to a smaller trend at higher altitudes over the same analysis period. From Figure 10 a), CH_4 accelerates beginning 2020. Lateral mixing may also contribute, however, the trend alone cannot distinguish its contribution from that of vertical transport. We have added the following sentence to acknowledge this interpretation in lines 373-376. “The larger trend near the tropical tropopause than above may reflect a more immediate response to the accelerating tropospheric CH_4 increase, whereas the response at higher altitude is delayed and smoothed by stratospheric transport and mixing”

20. ERA5 data should be listed in the Data availability section.

We added one sentence in lines 456-457. “ERA5 data can be downloaded from https://cds.climate.copernicus.eu/datasets/reanlysis-era5-pressure-levels-monthly-means?tab=download”

Reivewer 2

Review of „Methane in the Asian Monsoon Anticyclone and global UTLS behavior observed in ACE-FTS satellite data“ by Yi et al.

We thank the reviewer for the positive comments and valuable suggestions. We have revised the manuscript and please see the track-changes file for these revisions. All line numbers in our responses refer to the clean version of the revised manuscript.

General comments:

The paper presents an analysis of ACE-FTS CH4 and CO data related to the Asian monsoon region. The data are compared climatologically to AirCore and aircraft campaign data, and general agreement is found. The distributions are then analysed relative to their vertical and horizontal confinement by the Asian monsoon anticyclone. In a second part, the paper moves to analysis of global distributions of methane (also from ACE-FTS), and studies its seasonality in the tropics and its global trends. Main findings are that CO is more strongly confined due to its shorter chemical lifetime, while methane is transported out of the Asian monsoon anticyclone vertically, and to the North and into the tropics. A tropical stratospheric tape recorder of methane is claimed to be found, with its positive phase starting in October, and its negative phase starting in August. Methane trends are found to show hemispheric asymmetry in the lower stratosphere which is related to a weakened shallow branch of the Brewer-Dobson circulation in the Northern hemisphere relative to the Southern hemisphere.

In general, the paper is well written, the data analysis seems to be thorough (except for the restrictions I make below), and the figures are instructive. Nevertheless, I have two general comments.

First, the specific observational restrictions of ACE-FTS seem to be widely ignored in the data analysis. As an occultation instrument in a near-polar orbit, ACE-FTS is not able to cover all latitudes at the same time. In particular, the low latitudes (between 20S and 20N) are probed during four months of the year only: February, April, August and October. Other latitudinal regions are probed at other times of the year. The authors are well aware of this fact, nevertheless they worked with seasonal mean data when producing global maps or global distributions. This might produce some confusion and, in particular, some artificial structures. A warning in the text would be required, at least. While I can live with this procedure (mainly since it has been done the same way many times before and it wouldn‘t be fair to critizise the authors for an approach that has been more or less the common - however not exact - way to proceed), I have more severe problems with deriving a tape recorder signal from just four data points over the year. As I understand, the related figure (Fig. 9) is constructed from a climatology constructed from all years covered by ACE-FTS. Only four data points (Feb, Apr, Aug, Oct) are really probed, all other data in between seem to come from interpolation(?). Deriving a tape recorder signal and assessing its uplift velocity from these four data points seems to be quite far-fetched to me. I have made suggestions below how to improve on this analysis.

We acknowledge that the ACE-FTS sampling limitation affects the interpretation of the global seaosnal distribution and the tropical annual cycle.

In order to make the point explicit, we have added several revisions based on the suggestions below. We added one figure in Section 2 showing the monthly latitudinal coverage to tell the readers that the ACE-FTS measurements in tropics are mainly in February, April, August and October per year. We have also clarified in the text and relavant figure captions that the seasonal distributions represent composite of available observations rather than all months seasonal means. These clarifiation highlights the need for cautious interpreting the global maps and zonal mean distributions. We have also expanded the introduction of previous studies using ACE-FTS measurements to place our approach and its limitations in context. By doing so, we found evidences to support our current treatment to the global maps.

For methane tape recorder analysis, we have followed the reviewer's suggestion by applying a regression fit to the observed time series at different altitudes. The reconstructed seasonal cycle is presented alongside the original four-month observations to assess the consistency of the altitude-dependent phase progression. We also acknowledge that the tape recorder is hardly derived from only four-month observation. Details of the regression analysis and its implications for the inferred upward propagation and its rate are provided in our reponse to the specific comment on Fig. 9.

Second, I think this work needs to be put better into context of previous work wrt several aspects. The Asian monsoon anticyclone and its specific transport capabilities have be analysed in great detail in recent years. The authors should refer in more detail to this previous work, sharpen their statements by doing this, and demonstrate more clearly what their specific new findings are. I have listed a number of publications below that came immediately to my mind, but there are certainly far more.

We have incorporated the suggested references and expanded the Introduction and Discussion to better place our findings in the context of previous research on transport associated with the Asian monsoon anticyclone.

We have also clarified the specific observational contribution of this study: characterizing the methane enhancement near the tropopause, its vertical position relative to the CO enhancement, and its extension above the tropopause and toward higher latitudes. We now discuss these findings in relation to previously identified transport mechanisms, distinguishing the observational evidence provided here from the understanding established by earlier studies. The corresponding revisions are detailed in our responses to the specific comments below.

Specific comments:

1. Abstract, line 15: What do you refer to when saying "relative maximum"? Vertically, horizontally?

We revised the sentence to “In the ASM, a maximum in methane concentration occurs near the tropopause, at a higher altitude than the corresponding carbon monoxide maximum.” to clarify the relative vertical positions of the methane and carbon monoxide enhanced layers in line 16.

2. line 35: Where do the methane enhancements (“rich in methane …”) come from? What are the sources and where are they?

We added two sentences to describe the methane sources in south-east Asia and emphasize the strong seasonality of the sources.

In lines 40-42, “South and Southeast Asia are major CH_4 source regions, with emissions dominated by agriculture (including rice cultivation and livestock), waste, and wetlands. Emissions from these sources increase during the ASM season, likely due to enhanced rainfall (Yu et al., 2023; East et al., 2024; Subramanian et al., 2026).”

3. line 38: Please cite also more recent publications, of e.g. Vogel et al. (see my suggestions below).

We have added a citation to Vogel et al., 2026 in line 47.

line 42: Where does this statement come from? Can you cite review papers or assessments where this deficiency is stated?

We agree that the original wording was too broad and insufficiently supported. Our intention was to motivate a specific question arising from the transport mechanisms discussed above, rather than to make a general assessment of deficiencies in existing knowledge. We have therefore replaced the original statement with:

Lines 49-51: “Given the strong seasonality of regional methane emissions and the efficient transport of boundary pollution into the UTLS by the ASM, an important question is how these emissions and transport processes together contribute to the global distribution and seasonal variability of UTLS methane. ”

4. line 51: It needs to be stated here that ACE-FTS measures rarely in the tropics, only during four months per year.

We revised the text to acknowlede the limited tropical sampling of ACE-FTS. In lines 55-56, we revised the sentence to: “provides information on seasonal variability (although tropical observations are limited to four months per year: February, April, August, and October), interannual variability, and long-term trends (Bernath et al., 2005).”

5. line 66: Four months per year are hardly enough to derive a seasonal cycle (see more detailed comment below).

We agree with the reviewer that four months of observations per year are insufficient to fully characterize the seasonal cycle in the tropics. Our original wording was therefore too strong. We have revised the text to explicitly acknowledge the limited tropical sampling of ACE-FTS.

We cite a previous study comparing seasonally sampled ACE-FTS observations with MLS satellite data. This comparison showed reasonable agreement in large-scale spatial patterns despite the limited tropical sampling of ACE-FTS.

In lines 58-61, “The limited tropical sampling of ACE-FTS has been examined in Park et al (2013) by comparing its CO measurements with those of Microwave Limb Sounding (MLS). They found reasonable agreement in the large-scale spatial and temporal patterns of seasonal mean CO, supporting the use of ACE-FTS observations to characterize these features despite the sparse sampling.”

Moreover, we cite a recent study employing a regression approach similar to that used later in our analysis to better characterize the tropical CH_4 features.

In lines 75-78, “More recently, Sheese et al. (2025) used ACE-FTS measurements, including CH_4, to quantify the contributions of methane oxidation and circulation changes to stratospheric water vapor trends. To address the limited tropical sampling in tropics, they supplemented ACE-FTS observations with model simulations and applied multiple linear regression (MLR) to estimate CH_4 trends entering the stratosphere.”

6. line 69: This validation refers certainly to an earlier data version. Which one? Same for comparisons to MIPAS.

We revised the sentence to point out the earlier version validation in lines 96-99.

“Validation of the earlier v2.2 methane profiles showed differences of ~5 % below 35 km and ~10–15 % at 40–60 km relative to Halogen Occultation Experiment (HALOE) satellite data , and a negative bias of approximately 10% at 8-20 km relative to Michelson Interferometer for Passive Atmospheric Sounding (MIPAS) (De Mazière et al., 2008).”

7. line 77: For the tropics and the ASM, this means that DJF is actually February, MAM is actually May, JJA is actually August, and SON is actually October. The temporal coverage shifts for other latitude bands. Seasonal averaging masks this effect. How far is this accounted for in the global maps and zonal mean distributions?

Our original analysis grouped all available observations within each three-month period without explicitly accounting for differences in temporal coverage across laitutde bands. We agree that these composites should not be interpreted as uniformly sampled three-month seasonal means, and that differences in sampling month can affect the interpretation of both the global maps and the zonal mean distributions.

First, we explicitly emphasize the sampling limitations.

We have therefore added monthly sampling for the latitude bands from 2005 to 2023 shown in figure 1 to clarify the temporal coverage underlying each composite.

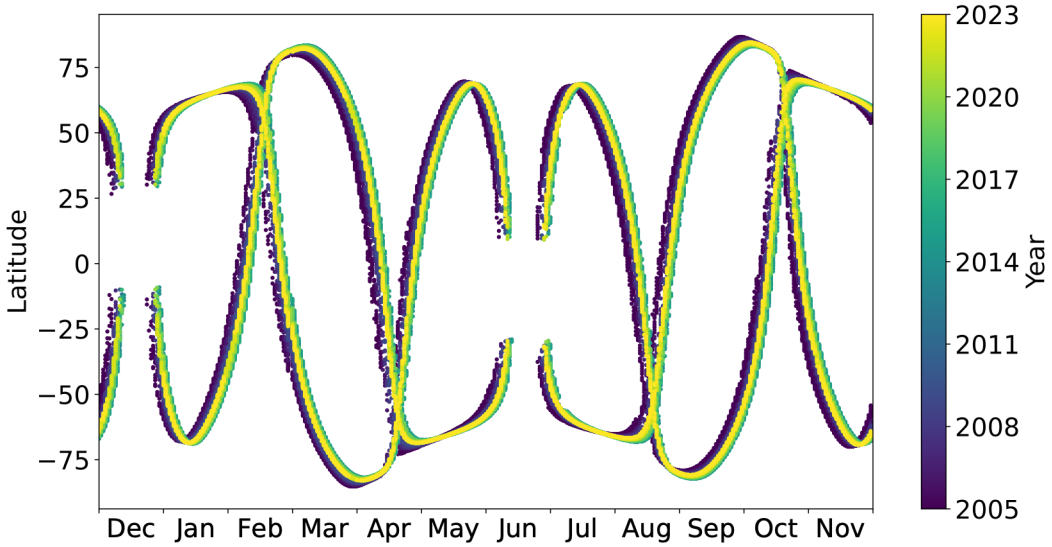


Figure 1: Latitudinal coverage of the ACE-FTS measurements for 2005-2023.

We have also revised the text and figure captions to describe the results as composites of available observations within the DJF, MAM, JJA, and SON periods to emphasize the seasonality in tropics and ASM region are only four months each year. These changes make the sampling limitations explicit.

No correction for latitude-dependent differences in temporal sampling was applied to the climatological global maps or zonal mean distributions. Our use of three-month observational composites follows previous ACE-FTS climatological studies (e.g. Park et al. 2008, 2013). The global map and zonal mean distributions were constructed from all available observations within each three-month period. We have revised the text and figure captions to clarify that they represent composites of available observations rather than uniformly sampled seasonal means, and that differences across latitudes may partly reflect differences in sampling month. See lines 231-233, 243-245, 259-261.

8. line 91 to 95: How do you explain the enhancements in the tropopause region, but not below, in the troposphere? Does This mean that the air was not uplifted locally (and vertically), but tropopause air was transported horizontally before the observation, from areas with enhanced CO and CH_4 amounts?

Yes, we examined the vertical structure of the methane profiles in our previous study. The enhanced methane concentrations near the tropopause were primarily associated with airmasses originating from Pakistan approximately two weeks before the measurements, where TROPOMI also observed high XCH_4 values. In addition, evidence of a stratospheric intrusion in the middle troposphere was identified at the campaign site (37.8N, 95.3E). More details are provided in Yi et al. (2024). doi: 10.1029/2024GL109256.

9. line 120: The selected profiles should not be called “collocated”, since they are within a wide spatial span, and they have a vague temporal coincidence only. The term “collocated” is mostly used within profile-to-profile validation, while your comparisons have the character of climatological comparisons, looking for typical profile shapes.

We agree that the term “collocated” is not appropriate in this context. We have therefore removed the word “collocated” in line 199.

10. Fig. 2: It is interesting to see that the bulge in the ACCLIP mean CO profile (red line, top right panel) comes from a bimodal distribution of CO, hinting towards some inter-annual variability. Have you looked into this fact in more detail?

The ACCLIP CO profiles were collected only in August 2022. I’m not sure that the bimodal distribution reflects the interannual variability since CO has relatively short lifetime. The bimodality more likely reflects variability in air-mass origins, convective influence, and transport history among the individual flights. We have not investigated these factors in detail, as they are beyond the scope of the present study, and therefore refrain from attributing the bimodality to specific process.

11. Fig. 2, figure caption: what does “near the location of AirCore campaign” exactly mean? As described in the text lines 117-121? Please specify.

We rewrite the caption to be clear. “Figure 3: Comparison of ACE-FTS measurements with in-situ observations in the ASM region. (a) Layer-normalized relative frequency distributions of CH_4 measured during the ACCLIP campaign, with horizontal and vertical bin sizes of 20 ppbv and 0.5 km, respectively. The red line shows the mean CH_4 mixing ratio for each vertical

bin level. The blue dots represent the mean vertical CH_4 profile measured by ACE-FTS, and the blue error bars indicate one sigma standard deviation at each level; (b) Same as (a), but for CO. (c) Comparison between the AirCore CH_4 profiles (gray lines) and the mean ACE-FTS profile (blue dots and error bars, as in panel a). (d) Same as (c), but for CO."

12. Line 148 and Fig.3: The description "near global climatological distributions of CH4 and CO in JJA" is not wrong, but not fully correct either. ACE-FTS samples different latitudes at different times, which means that for JJA e.g. tropics are observed in August, while Northern midlatitudes are observed in June, and Northern (sub-)polar regions in July. The averaging over a season provides the wrong impression that the distributions in Northern high latitudes are observed at the same time as those in the tropics. I am fully aware that this is a property of ACE-FTS observations that cannot be avoided. Nevertheless, at least a sentence or two should explaining these specific observational details should be provided.

We added one sentence in lines 231-233. "Note that the JJA map is a composite primarily consisting of measurements over the tropics in August, the northern subpolar region in July, and the midlatitudes in June, as shown in Fig. 1."

We also added one sentence on Fig. 4 (original Fig. 3) caption. In lines 243-245, "Note that the JJA map predominantly represents measurements over the midlatitudes in June, the northern subpolar region in July, and the tropics in August (see Fig. 1), rather than spatially uniform sampling throughout JJA."

13. Fig. 4: Same comment as for Fig. 3

We added the same sentence on Fig. 5 (original Fig. 4) caption. In lines 259-261, "Note that the JJA map predominantly represents measurements over the midlatitudes in June, the northern subpolar region in July, and the tropics in August (see Fig. 1), rather than spatially uniform sampling throughout JJA."

14. Line 153: What do you refer to with "more pronounced"? Relatively? In percent per latitude? Or do you mean that CO spreads farther longitudinally outside the AMA? Please clarify!

By “more pronounced”, we intended to describe the distinct spatial contrast between enhanced CO within the ASM anticyclone and the surrounding air, in both the meridional and zonal directions. We revise the sentence in lines 234-236. “The gridded results reveal clear enhancements of both CO and CH_4 within the ASM anticyclone, with CO showing a larger relative enhancement with respect to the surrounding background.”

15. Line 174-175 and Fig.5: I assume these enhancements refer also to JJA? This should be mentioned in the text and the figure caption.

We revise the sentence in lines 263-264. “The JJA enhancements of CO and CH_4 inside the anticyclone are further quantified in Fig. 6 by subtracting the profiles outside the anticyclone (longitudes outside of 20-120°E) from those inside (longitudes 20-120°E).”

We revised the Fig. 6 (original Fig. 5) caption to clearly describe the variable shown in the figure. “Figure 6: Meridional cross sections of the ASM enhancement in (a) CH_4 and (b) CO derived from ACE-FTS observations for JJA during 2005-2023, defined as the mean mixing ratio over 20°-120°E minus the background mean over all remaining longitudes. Blue lines indicate the lapse-rate tropopause provided by Hoffmann et al., 2021. Solid and dashed black contours represent westerly (10 and 20 m/s) and easterly winds (-10 and -20 m/s), respectively. Gray streamlines, shown only between 15-60°N to cover the ASM region and higher latitude potentially influenced by ASM transport, depict the circulation in the latitude-altitude plane and are constructed from the meridional wind (m/s) and vertical velocity (mm/s). All meteorological fields are averaged over 20-120°E during JJA 2005-2023 based on ERA5 reanalysis. Note that the vertical velocity is converted from pressure-coordinate vertical velocity using the hydrostatic relation.”

16. Line 180: Eddy-shedding transport towards the North-East is discussed in detail by Vogel et al., 2014 (ACP, 14, 12745-12762, 2014; doi: 10.5194/acp-14-12745-2014) and other colleagues of this team more recently (e.g. Clemens et al., 2022, doi:10.5194/acp-22-3841-2022). Respective references should be added.

We have added Vogel et al (2014) and Clemens et al (2022) to acknowledge previous work on transport from the Asian monsoon anticyclone into the extratropical lower stratosphere through eastward eddy shedding in lines 270-272.

“Previous studies have shown that eastward eddy shedding from the Asian monsoon anticyclone facilitates the transport of tropospheric trace gases into the lower stratosphere at northern mid- and high latitudes (Vogel et al., 2014; Clemens et al., 2022).”

17. Fig.6: It should be emphasised more strongly that here, we look at a zonal mean distribution covering all longitudes, while before, e.g. in Fig. 4, the data were from the region of the AMA only. Further, the same comment regarding seasonal coverage applies here as for Figs.3 and 4.

We have revised the Fig. 7 (original Fig. 6) caption to emphasize the zonal mean and sampling limitation.

“Figure 6: Climatological meridional cross sections of zonal mean CH4 for the (a) DJF, (b) MAM, (c) JJA and (d) SON seasons derived from ACE-FTS observations, averaged over period 2005-2023. White dashed lines denote the lapse-rate tropopause provided by Hoffmann et al., 2022. Unlike the AMA regional distributions in Fig. 5, these panels show the full zonal mean. Note that in the tropics, DJF, MAM, JJA and SON primarily represent observations in February, April, August and October, respectively.”

18. Lines 201-203: The CH4 maximum near the tropical tropopause is interesting, as it cannot come from strictly vertical transport of air masses at the same latitudes, but must involve meridional transport. Vogel et al., 2019 (ACP, 19, 6007–6034, 2019, https://doi.org/10.5194/acp-19-6007-2019) have looked into this in great detail (compare their Fig. 14). Reference to this publication should be made and your findings should be discussed in the light of this publication.

We have added a discussion of Vogel et al., 2019, particularly the transport pathways illustrated in their Fig. 14 in lines 299-303.

"Vogel et al. (2019) described isentropic transport from the Asian monsoon anticyclone into the tropics during the upward spiraling within a few months (see their Fig. 14). These transport processes provide a pathway for air mass originating within the anticyclone and in adjacent tropical regions to enter the tropical pipe. This transport pathway may contribute to the observed SON enhancement of CH_4 near and above the tropical tropopause."

19. Line 209/210: Reference to Vogel et al., 2019 should be made.

Reference has been added in lines 308-310. "During the boreal autumn season, the chemical composition within the anticyclone can be transported into the tropics as the anticyclone breaks up (Vogel et al., 2015, 2019; Orbe et al., 2015)."

20. Line 216 - 218: It should be made clear at this point that the CH4 surface fluxes are not derived form ACE-FTS data in this work, but stem from another publication and are based on other data.

We have revised the text in lines 318-321. "We further examine the role of ASM transport in shaping CH_4 seasonality in the tropics using the seasonal variability of CH_4 surface fluxes obtained from Saunois et al. (2025) and the area of low outgoing longwave radiation (OLR) as a proxy for deep convection over 15°S – 30°N (Fig. 9). The surface flux estimates are independent of the ACE-FTS observations used in this study."

We also added one paragraph in section 2.3 to describe the surface flux in lines 166-176.

"The CH_4 surface flux dataset provided by Saunois et al. (2025) is constructed by combining bottom-up emission inventories and model estimate for individual source and sink categories. The prior flux draws on Emission Database for Global Atmospheric Research (EDGAR v7; Crippa et al., 2023) for anthropogenic emissions, with extension based on Food and Agriculture Organization (FAO) agriculture data and includes an alternative configuration using the Greenhouse gas and Air pollutant Interactions and Synergies (GAINS) model for fossil fuel emissions (Höglund-Isaksson et al., 2020). Biomass-burning emissions are obtained from Global Fire Emission Database (GFED) v4.1 (van der Werf et al., 2017), and wetland emissions are represented by the monthly ensemble mean of 11 models reported by Saunois et al. (2020). Additional components include termite emissions, ocean

emissions from Weber et al. (2019), geological emissions from Etiope et al. (2019), and soil methane uptake from the MeMo model (Murguia-Flores et al., 2018). These components are assembled into gridded surface flux fields for use as prior inputs to atmospheric inversions, with temporal variability determined by the underlying inventories and models."

21. Line 221-223: As demonstrated by Vogel et al., 2019, the transport from the Asian monsoon anticyclone (AMA) does already happen when the AMA is still intact, and not only after its break-up. Please discuss your findings versus this publication.

We revised the sentences in lines 324-327 to fully describe the transport pathway.
"The localized CH_4 maxima can be transported isentropically from the Asian monsoon anticyclone into the tropics (Vogel et al., 2019). This transport, together with further redistribution of monsoon air during the seasonal break-up of the anticyclone and convective transport at other longitudes, leads to the global UTLS maximum in the tropics in SON."

22. Fig. 9 and related text (lines 231-239): This figure and the related discussion is a bit tricky due to the limited sampling of ACE-FTS at low latitudes (as the authors indicate in line 234). Within the latitude band 20S-20N, there are indeed only ACE-FTS data available for the month February, April, August, and October). Indeed we find the positive and negative maxima of the anomalies for these months. How have the data for the other months been generated? By interpolation? If so, it is very bold to derive a tape recorder and assess its uplift velocity from interpolated data. I recommend strongly to reconsider inclusion of this figure. At least, these statements need to be checked with CH4 data that are available from satellite observation all year round (e.g. HALOE, MIPAS). Or, as an alternative approach, apply a regression fit to time series (as shown in Fig. 10, left panel) at various stratospheric altitudes and derive the tape recorder and its uplift velocity from the phase shift between the altitude levels.

We clarify that Fig.10 (original Fig.9) was constructed using observations from February, April, August and October only; no CH_4 values were generated for the other months by

interpolation. We recognize that the continuous contour shading could give the impression of continuous monthly coverage.

Following the reviewer's suggestion, we apply the Long-term Ozone Trends and Uncertainties in the Stratosphere (LOTUS) regression code (SPARC/IO3C/GAW, 2019), following the standard multiple linear regression (MLR) framework described by Dubé et al. (2023). The model is fitted separately to the available observations in each latitude-altitude bin:

$$CH_4(t) = \beta^{(2)}(t) + \beta_{trend} \times linear(t) + \beta^{(2)}_{qboa} \times QBO_{(a)}(t) + \beta^{(2)}_{qbob} \times QBO_{(b)}(t) + \beta_{solar} \times F_{10.7}(t) + \beta_{enso} \times ENSO(t) + \beta_{aod} \times AOD(t) + R(t) \quad (1)$$

Here, each β is a regression coefficient for each term, the superscripts are the number of highest seasonal harmonic for a given term, $\beta^{(2)}(t) = const + A_1 \sin(2\pi t) + B_1 \cos(2\pi t) + A_2 \sin(4\pi t) + B_2 \cos(4\pi t)$, A_i and B_i are the sine and cosine coefficients, respectively, with subscripts $i = 1$ and $i = 2$ denote the annual and semi-annual cycles. Time t is expressed in years. $QBO_{(a)}(t)$ and $QBO_{(b)}(t)$ are the first two principal components of the Singapore zonal winds, $F_{10.7}(t)$ is the solar flux at 10.7 cm, ENSO (t) is the multivariate ENSO index, and $R(t)$ is the residual. These predictors are further described in SPARC/IO3C/GAW (2019). Dubé et al (2023) applied this standard MLR framework to estimate stratospheric N_2O trends from ACE-FTS observations.

The above method description is presented on section 2.4.

The reconstructed monthly CH_4 time series are presented in appendix (Fig. A1).

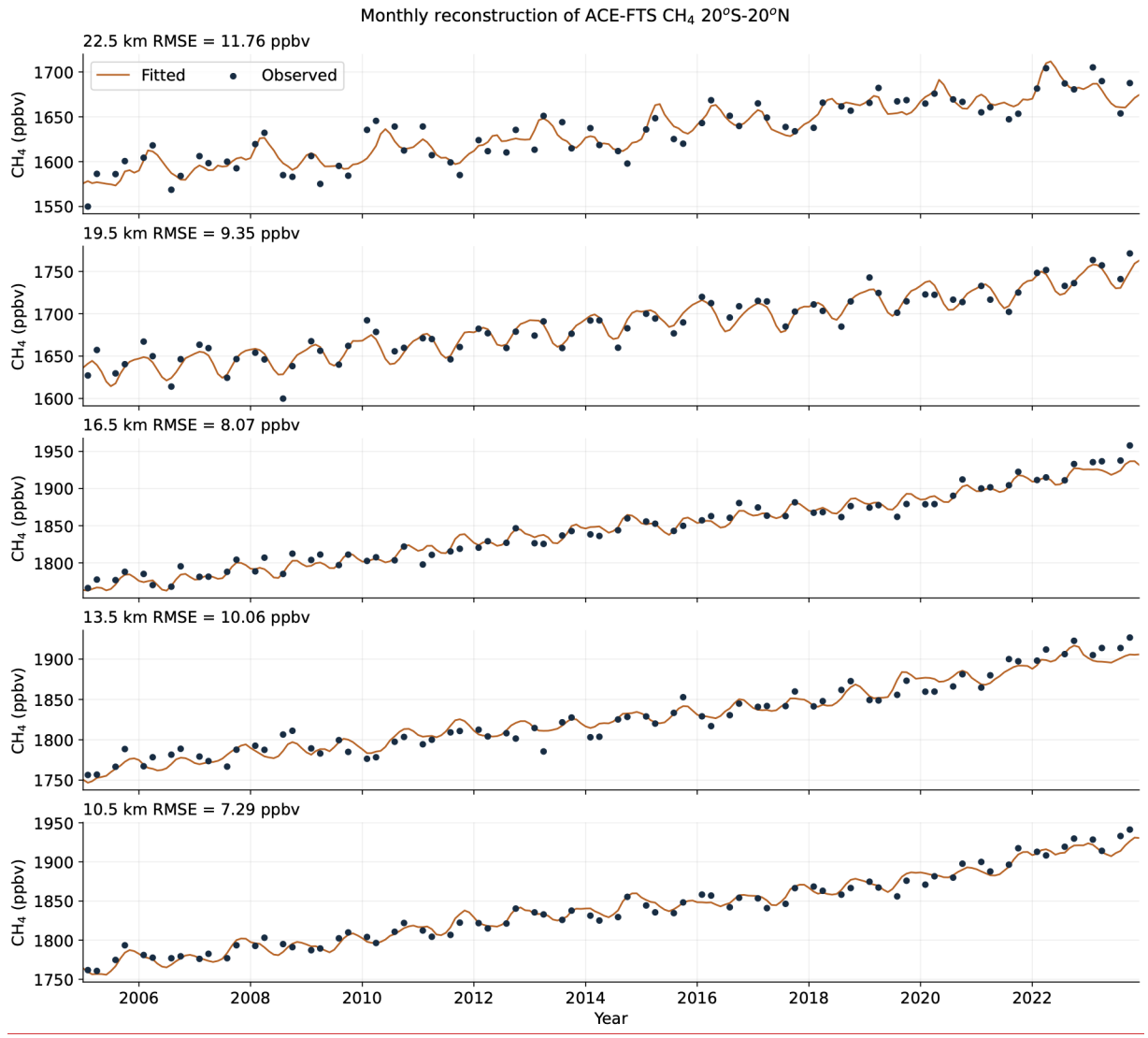


Figure A1: Monthly reconstruction of ACE-FTS CH_4 mixing ratios averaged over 20°S-20°N at selected altitudes during 2005-2023. Black dots represent observations available in February, April, August, and October. Red lines show monthly reconstructions from regression fits. Regressions were fitted separately at each altitude using only available observations. The root-mean-square error (RMSE) between fitted and observed values is indicated above each panel.

In lines 440-442, "The regression fits, obtained using the method described in Sect. 2.4, reproduce the long-term increase and the main seasonal variations in ACE-FTS CH4 observations averaged over 20°S–20°N during 2005–2023 at selected altitudes (Fig. A1)."

We then calculate the climatological seasonal anomalies of CH_4 based on the reconstructed monthly time series (Fig. 10b). We also rewrite the section to clearly state that direct

association from upper troposphere to lower stratosphere is poorly defined by the lack of ACE-FTS tropical measurements between October-February.

In lines 335-352, “Variations in the tropical CH_4 annual cycle and links between the upper troposphere and lower stratosphere are studied in Fig. 10. Here two versions of seasonal CH4 anomalies (deviations from the annual mean at each altitude) over 20°N-20°S are shown, based on (a) the four monthly ACE-FTS samples in August, October, February and April, and (b) monthly reconstruction using annual and semi-annual harmonic fits (method described in Sect. 2.4).

The observations show the boreal autumn (October) positive anomaly extends up to and across the tropopause, consistent with the seasonal peak shown in Fig. 7. In the lower stratosphere, there is a predominant annual cycle with a maximum in February and minimum in August, and largest amplitude over 18-20 km. The timing and altitude maximum of this variability suggests a link to the annual cycle in upwelling in the tropical lower stratosphere (BDC, with maximum in boreal winter) acting on the background vertical gradients in CH_4 above the tropical tropopause, e.g. Fig. 7. This is similar to the seasonal behavior observed for carbon monoxide and ozone (with opposite sign) in the tropical lower stratosphere (Randel et al., 2007).

The timing of the lower stratosphere CH_4 maximum in boreal winter may also suggest some upward propagation of the CH_4 maximum near the tropopause in October, as suggested by the white dashed lines in Fig. 10 (with a slope of 8 km/year, corresponding to the calculated upwelling velocity). However, this direct association from upper troposphere to lower stratosphere is poorly defined by the lack of ACE-FTS tropical measurements between October-February.”

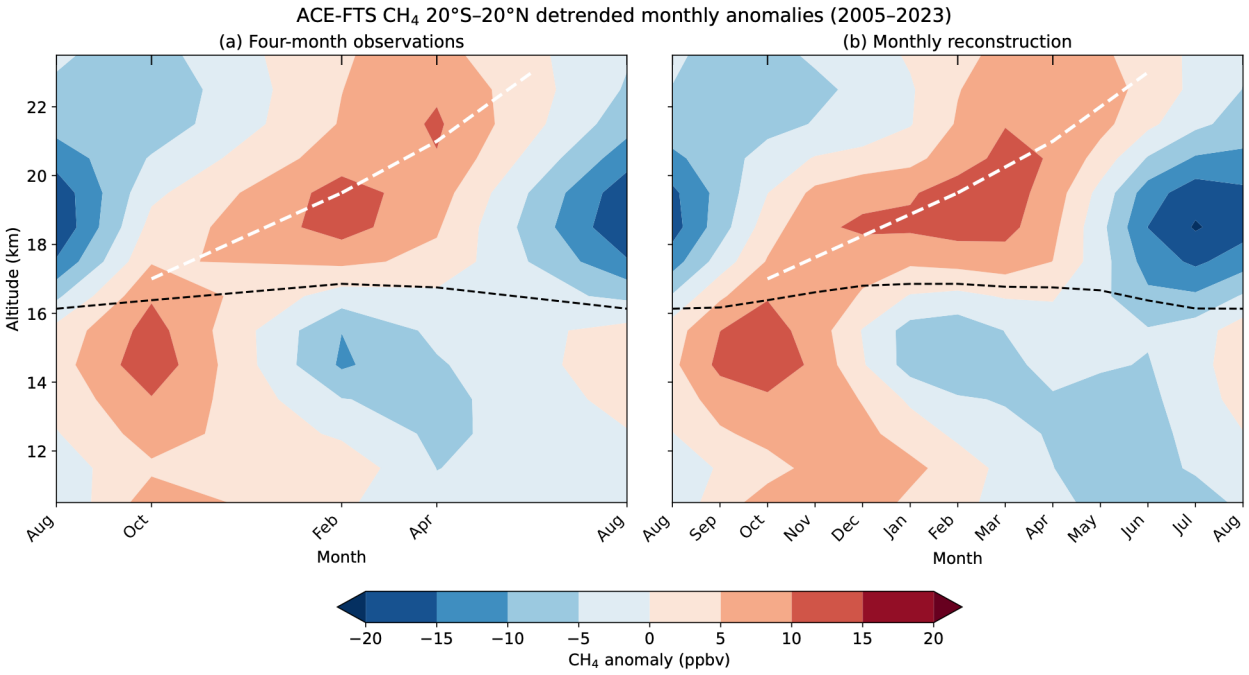


Figure 10: Climatological detrended monthly anomalies of ACE-FTS CH_4 averaged over 20°S-20°N during 2005-2023, derived from (a) observations in August, October, February and April (b) monthly reconstructions using regression fit including annual and semi-annual cycle. At each altitude, the linear trend is first removed from both observations and reconstructed values. Anomalies are then calculated relative to each year's mean of the four detrended observation months in (a) and the 12 detrended reconstructed months in (b), before averaging each month across the 19 years. Black dashed lines indicate the climatological tropopause height. White dashed lines in (a) trace the maximum anomalies at each altitude to indicate the possible upward progression of positive anomalies. The same white dashed lines are overlaid in (b) for comparison. August is repeated at the right edge to show continuity across the annual cycle.

We also revised the abstract regarding the section to "Positive anomalies occur in the tropical lower stratosphere in boreal winter, consistent with the upwelling branch of the Brewer–Dobson circulation (BDC)." in lines 22-24.

We also revised the discussion regarding the section to "Large positive anomalies in the tropical lower stratosphere in the boreal winter suggests a possible role for tropical upwelling associated with the BDC. However, direct association of the positive anomalies from upper troposphere to lower stratosphere is poorly defined by the lack of ACE-FTS tropical measurements between October-February." in lines 424-427.

23. Line 252: How do these trends fit to tropospheric methane trends from other studies?

Schmidt et al (2024) reported CH_4 trend is 6.41 ppb/year between 2012-2018 and increases significantly to 15.12 ppb/year since 2019. Converting to relative units (assuming a background of ~1900 ppb), these correspond to approximately 3%/decade and 7%/decade, respectively. Our Fig. 11b shows broad positive trends of +3-4%/decade in the global troposphere, which is very close to the trend reported by Schmidt et al. (2024). We have added a sentence (lines 371-372) in the revised manuscript to clarify this comparison. "Schmidt et al. (2024) reported a tropospheric methane trend of 6.41 ppb/yr during 2012-2018, increasing to 15.12 ppb/yr after 2019."

24. Lines 255 -260: The hemispheric asymmetry of trends of the Brewer-Dobson circulation has been analysed in a number of papers. It was also observed by e.g. Mathieu et al., 2014 (doi:10.1038/nature13857) and Haenel et al., 2015 (doi:10.5194/acp-15-13161-2015), analysed in detail by Stiller et al., 2017 (https://doi.org/10.5194/acp-17-11177-2017), and finally confirmed to appear in CCMI model simulations as well by Ivaniha et al., 2026 (10.1029/2025JD045069), tracing it back to QBO-BDC interactions. A more thorough referencing of previous work is desirable here.

We have expanded the discussion and added the suggested references to better acknowledge previous observational and modelling work on hemispheric asymmetries in stratospheric transport trends. In particular, we added the related analysis from Stiller et al. (2017) and the role of QBO-related variability demonstrated by Ivaniha et al. (2026). We have also revised the concluding statement to acknowledge these mechnisms and to avoid attributing the observed CH_4 trend asymmetry to a specific change in BDC strength or structure without further dynamical diagnostics.

In lines 379-387, "This pattern is consistent with previously reported hemispheric asymmetries in stratospheric tracer and age-of-air trends after 2000 for other long-lived stratospheric tracers (e.g. Mahieu et al., 2014; Haenel et al., 2015; Han et al., 2019; Strahan et al., 2020; Prignon et al., 2021; Dube et al., 2023, 2025) and with reanalysis-driven simulations indicating relative aging of air in the NH compared to the SH (Ploeger and Garny,

2022). Stiller et al. (2017) attributed much of the observed age-of-air trend asymmetry to a southward shift of subtropical transport barriers. More recently, Ivaniha et al. (2026) confirmed a southward shift of the tropical pipe in CESM1-WACCM and linked it to nonlinear Quasi-Biennial Oscillation (QBO) effects. These studies suggest that changes in stratospheric transport, including circulation shifts and QBO-related variability, may contribute to the hemispheric asymmetry in our CH_4 trends."

25. Summary and discussion: I recommend to put the findings of this work in closer context to findings of previous publications, some of which I have listed above. In particular, there is rich literature on transport out of the anticyclone to the North-East (lines 275-280) and into the tropics (lines 281-286) and on the hemispheric asymmetry of BDC trends (lines 290-294) that should be considered in the discussion. By doing so, the authors should sharpen their statements and demonstrate more clearly what their specific new findings are.

We have revised all three paragraphs to place our findings more clearly within the existing literature and distinguish our observational contribution from previously established transport mechanisms.

For transport into the northern extratropical lower stratosphere, we now discuss eastward eddy shedding and quasi-horizontal poleward transport, citing Ploeger et al. (2013, 2017) and Vogel et al. (2014, 2015). We clarify that our contribution is the observed contrast between CH_4 enhancements extending northward and above the tropopause and CO enhancements concentrated below the tropopause within the anticyclone. These distributions provide additional observational evidence consistent with established pathways.

In lines 408-413, "The methane distribution is consistent with previous study on transport from the ASM anticyclone into the northern extratropical lower stratosphere (Ploeger et al., 2013, 2017; Vogel et al., 2014, 2015; Clemens et al., 2022). These studies identify eastward eddy shedding and quasi-horizontal poleward transport along isentropic surface as pathways by which air within the ASM anticyclone reaches the extratropical lower

stratosphere. Our observations provide additional tracer evidence consistent with these transport pathways revealed by the simulation results."

For transport toward the tropics, we have incorporated previous findings on the dispersal of anticyclone air and its subsequent transport into the tropical stratosphere (Vogel et al., 2015, 2019; Garny and Randel, 2016; Ploeger et al., 2017). We now place the observed autumn broadening of enhanced CH_4 toward the tropics and the apparent upward progression of its seasonal anomaly in this context. Our specific contribution is the characterization of these CH_4 spatial and seasonal patterns to support the transport mechanism.

In lines 420-424, "This evolution is consistent with simulations showing a late-season broadening of monsoon-source tracers toward the tropics (Vogel et al., 2015) and dispersal of anticyclone air across the tropics (Garny and Randel, 2016). Previous studies have identified pathways involving diabatic ascent and quasi-horizontal transport into the tropical stratosphere, followed by further ascent within the tropical pipe (Ploeger et al., 2017; Vogel et al., 2019)."

For the hemispheric asymmetry of stratospheric trends, we have added Mahieu et al. (2014), Haenel et al. (2015), Stiller et al. (2017), and Ivaniha et al. (2026). We have broadened the discussion to include circulation shifts and QBO-related transport variability, rather than attributing the asymmetry solely to a weakened NH shallow branch. We emphasize that the ACE-FTS CH_4 trends provide complementary tracer evidence, while not independently identifying the underlying dynamical mechanism.

In lines 431-435, "The spatial pattern is consistent with post-2000 trends in other long-lived tracers and reanalysis-driven simulations reported in recent studies (Mahieu et al., 2014; Haenel et al., 2015; Ploeger and Garny, 2022). They may reflect variability in the strength and spatial structure of the BDC, including shifts in subtropical transport barriers and QBO-related modulation of transport and mixing (Stiller et al., 2017; Ivaniha et al.,2026)."

These revisions clarify that the contribution of this work lies in documenting CH_4 distributions, seasonality, and trends and interpreting them in relation to established stratospheric transport mechanisms.