

Validation of ACE-FTS satellite data in the upper troposphere/lower stratosphere (UTLS) using non-coincident measurements

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CO, O₃, and H₂O data in the upper troposphere/lower stratosphere (UTLS) measured by the Atmospheric Chemistry Experiment Fourier Transform Spectrometer (ACE-FTS) on Canada's SCISAT-1 satellite are validated using aircraft measurements. In the UTLS, validation of chemical trace gas measurements is a challenging task due to small-scale variability in the tracer fields, strong gradients of the tracers across the tropopause, and scarcity of measurements suitable for validation purposes. Two alternative methods for the validation of the satellite data are introduced, which avoid the usual need for coincident measurements: tracer-tracer correlations, and vertical profiles relative to the tropopause height. Both largely reduce geophysical variability and thereby provide an "instantaneous climatology", allowing measurement comparison with non-coincident data which yields information about the precision, and a statistically meaningful error-assessment of the ACE-FTS satellite data. We found that the ACE-FTS CO and lower stratospheric O₃ agree with the aircraft measurements within ±10% and ±5%, respectively. The ACE-FTS O₃ in the UT exhibits a high bias of up to 40%. H₂O indicates a low bias with relative differences of around 20% in the LS and 40% in the UT, respectively. When taking into account the smearing effect of the vertically limited spacing between measurements of the ACE-FTS instrument, the errors decrease by 5–15% around the tropopause. The ACE-FTS instrument hence offers unprecedented precision and vertical resolution in the UTLS, that will allow a new global perspective on UTLS tracer distributions.

1 Introduction

The upper troposphere/lower stratosphere (UTLS) has recently attracted major research interest in atmospheric science due to its key role in chemistry-climate coupling. In order to characterize UTLS tracer distributions and to detect future changes, tracer measurements with global coverage are needed. Satellites are the only means by

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which this task can be accomplished, however, their capability to measure accurately in the UTLS is limited, and the validation of the measurements difficult.

The restrictions are imposed by the dynamical and chemical structure of the UTLS. Dynamical variability in the tropopause region induced by Rossby wave activity is high, and length scales of the associated features in the tracer fields are small – less than 1 km in the vertical, and 100 km in the horizontal. Tracer mixing ratios also exhibit a strong gradient across the tropopause because it acts as a transport barrier (Pan et al., 2004; Hoor et al., 2004; Hegglin et al., 2006). Ultimately, the remote sensing technique used determines the achievable vertical and horizontal resolution of the measurements and hence the capability of the instrument to resolve the given geophysical small-scale variability. The Atmospheric Chemistry Experiment Fourier Transform Spectrometer (ACE-FTS) on Canada's SCISAT-1 satellite (Bernath et al., 2005) has provided accurate measurements of numerous chemical species throughout the stratosphere and into the UT since February 2004, with a vertical spacing between measurements of less than 1 km at the lowest retrieval altitudes. The measurements suggest a high potential for studies related to the upper troposphere/lower stratosphere (UTLS).

In the stratosphere and the mesosphere, the ACE-FTS satellite measurements are being validated by comparison to balloon-borne or independent satellite data which are approximately coincident in time and space. Coincidence criteria are defined in various ways, but typically the measurements have to be taken within several hours and at locations no further apart than around 500 km. As pointed out by Walker et al. (2005), however, it can be difficult to find a sufficient number of coincident measurements for a statistically meaningful validation. Furthermore, this validation method does not account for the variability in tracer distributions produced by the geophysical variability found within the defined time and length scales. As illustrated in the following example, the use of coincident measurements is especially an issue in the UTLS, where geophysical variability is large and a strong gradient in chemical tracers is found across the tropopause. Figure 1 provides a cross section of ECMWF potential vorticity at 6° W on 10 November 2001 at 6:00 UTC. The 2 PVU (1 PVU=1 potential

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vorticity unit= $10^{-6} \text{ m}^2 \text{ s}^{-1} \text{ K kg}^{-1}$) tropopause surface shows strong undulations and even profiles fulfilling the coincidence criteria exhibit large differences in tropopause height. Figure 2 shows two vertical profiles of CO mixing ratios measured in-situ by an aircraft instrument during the same dynamical situation and taken within 8 hrs and a distance of 400 km. As can be seen, the error derived for the measurements in the tropopause region is as large as 50%, though this is due to the geophysical variability and not a measurement error (the profiles are taken by the same instrument). Further complicating a statistically meaningful comparison is the sharp tracer gradient at the tropopause, which causes sampling errors not to be normally distributed. In this study, we introduce two alternative methods for the validation of the ACE-FTS CO, O₃, and H₂O measurements in the UTLS: tracer-tracer correlations, and vertical profiles relative to the tropopause height. These strongly reduce geophysical variability and help in assessing the quality and vertical information content of the satellite data. The paper is structured as follows. Section 2 provides the description of the ACE-FTS satellite and SPURT aircraft measurements. In Sect. 3, the new validation methods are introduced, which are used in Sect. 4 to validate the ACE-FTS measurements. A summary is given in Sect. 5.

2 Data description

2.1 ACE-FTS satellite data

The Atmospheric Chemistry Experiment Fourier Transform Spectrometer (ACE-FTS) is the primary instrument on SCISAT-1, a Canadian-led satellite mission for remote sensing of the Earth's atmosphere. ACE-FTS features high resolution (0.02 cm^{-1}) and broad spectral coverage in the infrared (750 to 4400 cm^{-1}). The instrument operates almost exclusively in solar occultation mode (Bernath et al., 2005). The SCISAT-1 satellite was launched into low Earth circular orbit (650 km) with high inclination (74°) on 12 August 2003. In solar occultation mode, this orbit provides seasonally varying

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coverage of the globe, with an emphasis on mid-latitudes and the polar regions. Up to 30 occultation events (sunrises or sunsets viewed by the orbiting satellite) occur per calendar day. Science operations for the ACE-FTS began in February 2004. Retrievals for the ACE-FTS employ a nonlinear least squares global-fit approach (Boone et al., 2005). In this study we validate ACE-FTS version 2.2 H₂O, CO, as well as the “version 2.2 O₃ update” results in the UTLS between February 2004 and January 2007. ACE-FTS results are provided on two altitude grids, a 1-km grid common to all occultations and a “retrieval grid” that varies from occultation to occultation. The retrieval grid contains values at the measurement altitudes, unless there are multiple measurements within a layer on the 1-km grid, in which case it provides a single value at the center of the 1-km grid layer. For this study, we use the measurements on the retrieval grid.

The altitude spacing between measurements varies over the course of the year, governed primarily by the beta angle (the angle between the satellite orbit plane and the Earth-Sun vector) corresponding to the occultation. The ACE-FTS instrument collects measurements every 2 s. The rate of change of altitude decreases with increasing beta angle, leading to higher vertical sampling for larger beta angles. At low altitudes, refraction effects and clouds also impact the measurement spacing. Figure 3 shows the probability density function of the vertical spacing between altitudes on the retrieval grid as a function of height for all measurements between 5 and 50 km. The altitude spacing in the UTLS varies from about 3 km to less than 1 km. Note, however, that the vertical resolution of the ACE-FTS is limited by its field-of-view. The instrument has a 1.25 mrad input aperture, which subtends an altitude range of 3–4 km at the tangent point (the point of closest approach to the Earth for a solar ray measured by the instrument). We focus on the validation of the UTLS measurements between ± 5 km of the tropopause, i.e. between approximately 5 and 15 km altitude or 500 and 100 hPa. The validation of the stratospheric and mesospheric ACE-FTS version 2.2 data (with ozone updates) is published along with this paper in a special issue of Atmos. Chem. Phys. on validation of ACE (for CO: Clerbaux et al., 2007¹, for O₃: Dupuy et al.,

¹Clerbaux, C., George, M., Turquety, S., et al.: CO measurements from the ACE-FTS mis-

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2007², for H₂O: Carleer et al., 2007³). Earlier comparisons of the version 1.0 ACE-FTS O₃ data with GOMOS satellite data (Fussen et al., 2005) yielded good agreement between the two data sets with differences mostly lower than 10% between 15 and 45 km. Comparisons with POAM III and SAGE III data yielded similar results (Walker et al., 2005), with HALOE an agreement of ±5% between 15 and 35 km (McHugh et al., 2005). In the same study the ACE-FTS H₂O data indicated a high bias of around 20% at altitudes below 20 km. A first validation of the version 1.0 CO by Jin et al. (2005) using ODIN satellite data yielded excellent agreement, and a comprehensive analyses of these data was provided by Clerbaux et al. (2005). Version 2.2 level data were furthermore compared to the AURA-MLS satellite data: the MLS O₃ showed a low bias in the lower stratosphere of 10% to 20%, and the H₂O good agreement except at lowest levels around 100 hPa with differences increasing up to 30% (Froidevaux et al., 2006).

2.2 SPURT aircraft data

As reference data set, we use in-situ high-resolution and high-precision CO, O₃, and H₂O measurements from the SPURT (German acronym for “trace gas transport in the tropopause region”) aircraft campaign carried out seasonally between November 2001 and July 2003 in the Northern Hemisphere over Europe and covering a latitude range between 30° N and 80° N. For each season, approximately 32 flight hours or 24 000 data points were obtained. An overview of the campaign can be found in Engel et al. (2006). Detailed descriptions of the CO, O₃, and H₂O measurements can be found

sion: validation using ground-based, airborne and satellites observations, Atmos. Chem. Phys. Discuss., submitted, 2007.

²Dupuy E., Walker, K. A., Kar, J., et al.: Validation of ozone measurements from the Atmospheric Chemistry Experiment (ACE), Atmos. Chem. Phys. Discuss., in preparation, 2007.

³Carleer, M., Boone, C. D., Walker, K. A., et al.: ACE-FTS Water vapor validation, Atmos. Chem. Phys. Discuss., in preparation, 2007.

in Hoor et al. (2004), Hegglin et al. (2006), and Krebsbach et al. (2006), respectively. Note that these data were obtained during earlier years than the ACE-FTS data, hence interannual variability in UTLS tracer distributions might add an uncertainty to the error assessment. However, the primary aim of this study is to introduce and show the potential of the new validation methods.

2.3 Derived meteorological products

For each measurement point of the two data sets we derived the thermal tropopause height according to the WMO-definition, i.e. the lowest level at which the lapse rate drops to 2 K km^{-1} or less, and the average lapse rate between this level and all higher levels within 2 km does not exceed 2 K km^{-1} (WMO, 1957), using the Goddard Earth Observing System Model, Version 4 (GEOS-4) by interpolation of the model fields onto the exact measurement location in time and space. The GEOS-4 analyses are described by Bloom et al. (2005). A Physical Space Statistical Analysis Scheme is used. The GEOS-4 data used here are provided on 55 hybrid (σ /pressure) model levels from the surface to 0.01 hPa. The horizontal grid is 1.0° latitude by 1.25° longitude. Six-hourly average fields are provided centered at 00:00, 06:00, 12:00 and 18:00 UTC. Besides the standard meteorological variables, GEOS-4 products include an extensive set of fields from the model and assimilation system, including PV calculated internally in the model. Further information on the derived meteorological products can be found in Manney et al. (2007⁴).

We need to mention that for the method presented here it is crucial to derive the thermal tropopauses for each data set with the same model. This is because different models vary in their vertical resolution and capability of reproducing the atmosphere's temperature structure and tropopause heights, which represent additional error sources.

⁴Manney, G. L., Daffer, W. H., Zawodny, J. M., et al.: Solar occultation satellite data and derived meteorological products: sampling issues and comparisons with Aura MLS, J. Geophys. Res., available at <http://mls.jpl.nasa.gov>, submitted, 2007.

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3 New validation methods

3.1 Tracer-tracer correlations

Sufficiently long-lived species exhibit compact correlations (Plumb and Ko, 1992), which reduce day-to-day variations and provide an “instant climatology”. Compact correlations are – depending on the respective lifetimes of the tracers used – not necessarily linear, as is the case for the O₃-CO and O₃-H₂O correlations; instead, they exhibit a strong curvature, which can be used to identify the chemical transition between the troposphere and the stratosphere (Pan et al., 2007 and references therein). Apart from recent troposphere-to-stratosphere transport events, which produce distinct and nearly linear mixing lines (Fischer et al., 2000), the compactness of these correlations is relatively high and their shape distinct due to their strong dependency on the location of the tropopause, so that we can use them to gauge the precision of the ACE-FTS measurements.

3.2 Vertical profiles relative to the tropopause height

Another method for reducing the effects of geophysical variability in UTLS tracer measurements, and for obtaining fields suitable for comparison of non-coincident measurements, is the use of tracer vertical profiles relative to the tropopause height (cf. Hoor et al., 2004; Pan et al., 2004; Hegglin et al., 2006). These profiles are more compact and show less scatter than data plotted in geometric altitude coordinates, furthermore they reveal a sharp gradient between tropospheric and stratospheric tracer mixing ratios which can be used to test the vertical resolution and information content of the ACE-FTS data. Since this method further allows us inclusion of all measurements, not just the coincident ones, one can get much better statistics.

In order to apply this method to the ACE-FTS and SPURT data, we first calculate the distance from the thermal tropopause of each measurement point. We then calculate the CO, O₃, and H₂O mean mixing ratios and their standard deviations (σ) for every

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1 km altitude bin between -6 km and 6 km relative to the tropopause height. We also derive the relative differences (rds) between the SPURT ($mean_{spurt}$) and the ACE-FTS mean tracer profiles ($mean_{ace}$) using

$$rds = 100 - (mean_{spurt}/mean_{ace}) \times 100, \quad (1)$$

5 and their uncertainties (u) calculated according to the general rules of error propagation:

$$u = (\sigma_{spurt}/mean_{spurt} - \sigma_{ace}/mean_{ace}) * 100. \quad (2)$$

4 Results and discussion

4.1 SPURT versus ACE-FTS tracer-tracer correlations

10 Figure 4 shows the $CO-O_3$ and H_2O-O_3 correlations for the ACE-FTS and the SPURT measurements for the latitude range between 30° and 90° N and for different seasons. The agreement between the correlations of the two data sets is remarkable, despite the fact that the measurements were carried out in different years. In general, the SPURT data lie well within the range of the ACE-FTS data, and the limited spread (or scatter)

15 of the ACE-FTS indicates a very good precision of the instrument. Some features such as the “high-heel” shape found in the spring H_2O-O_3 correlation, or the relatively broad transition between the troposphere and the stratosphere in the summer $CO-O_3$ correlation, are evident in both data sets.

4.2 SPURT versus ACE-FTS vertical profiles

20 The validation of the ACE-FTS CO , O_3 , and H_2O data using vertical profiles relative to the tropopause height is presented in Figs. 5 to 7. In order to minimize the uncertainties caused by a potential latitudinal dependency of the mean tracer profiles, we now only use data between 40° N and 60° N. The comparison between satellite and aircraft data

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yields the best result for CO. Relative differences are smaller than $\pm 10\%$, and the uncertainties of the relative differences further indicate that the two data sets are not significantly different. This result is comparable to the 16% relative difference derived from a validation using MOZAIC aircraft data in the UTLS by Clerbaux et al. (2007). The same can be said for O₃ in the LS with $\pm 5\%$ (cf. Dupuy et al., 2007), however, in the UT, the relative differences increase to around 40%, and indicate a persistent high bias of the ACE-FTS O₃. This might be due to a general sensitivity problem of satellites when it comes to detect and accurately measure low mixing ratios below atmospheric layers with very high mixing ratios. H₂O shows the largest relative differences of about 20% in the LS, and 40% in the UT, with the uncertainties of the relative differences suggesting a systematic low bias of the ACE-FTS measurements, at least for the middle to upper troposphere. However, H₂O is the most variable tracer in the troposphere among the species presented here, and indeed comparison of the ACE-FTS data with H₂O data from the ER-2 aircraft during the POLARIS campaign indicate rather a high bias in the UT (not shown). This inconsistency should be further investigated by comparing the ACE-FTS to other datasets in addition to the ones of SPURT and ER-2. Recall that the vertical resolution of the ACE-FTS is limited by its field-of-view. Due to this effect, it is difficult to resolve structure at better resolution than 3 km. As shown in Fig. 3, there is often a significant altitude oversampling in the vicinity of the tropopause, which may reduce the impact of the field-of-view. However, at the level of 1 km, there is additionally an inherent smoothing in the retrieval process, since forward model calculations employ data on a 1-km grid. To account (very roughly) for the limitations in the ACE-FTS vertical resolution, we smooth the SPURT profiles with a triangular function. For O₃, for example, we would apply the following smoothing

$$O_3[i] = 0.25 \times O_3[i-1] + 0.5 \times O_3[i] + 0.25 \times O_3[i+1] \quad (3)$$

where i is the altitude level of a given mean O₃ value. By smoothing the SPURT O₃ profiles with the above equation, we can gauge whether smearing effects from the

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ACE-FTS vertical resolution have a significant effect on the results.

Figure 8 shows the relative difference between the smoothed SPURT and the ACE-FTS mean O₃ profiles. The comparison with Fig. 6 indicates, that the smoothing effect accounts only for about 5–15% of the total measurement error, with largest impact just above and below the tropopause.

5 Conclusions

In this study we present the validation of ACE-FTS CO, O₃, and H₂O measurements on Canada's SCISAT-1 satellite in the upper troposphere/lower stratosphere (UTLS) using SPURT aircraft measurements. It contributes to the validation efforts of the ACE-FTS data published in this issue. Other studies (except the one focusing on CO), however, mainly focus on validation in the stratosphere and the mesosphere. In the UTLS, validation of chemical trace gas measurements is a challenging task due to small-scale variability in the tracer fields, strong gradients of the tracers across the tropopause, and scarcity of measurements suitable for validation purposes. We here suggest two alternative/complementary methods for the validation of satellite measurements in the UTLS: tracer-tracer correlations, and vertical profiles relative to the tropopause height. These methods are known to reduce geophysical variability, and thereby provide an “instantaneous climatology”, which avoids the need for coincident measurements. The climatological comparison allows to include all available measurements, not just the coincident ones, and by this yield better statistics, and more reliable information about the instrument precision.

We found that the ACE-FTS CO and lower stratospheric O₃ agree with the aircraft measurements within ±10% and ±5%, respectively. The ACE-FTS O₃ in the UT exhibits a high bias of up to 40%. H₂O indicates a low bias with relative differences of around 20% in the LS and 40% in the UT, respectively. When taking into account the smearing effect of the limited spacing between the measurements of the ACE-FTS instrument, the errors decrease by 5–15% around the tropopause. This limitation in the

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vertical resolution must be taken into account when interpreting the ACE-FTS UTLS measurements. Note, that tropopause heights derived for the different data sets used in validation studies should be derived using the same model, since differences in model characteristics may present an additional error source. The significance of the results presented here should be tested with further aircraft and satellite data. Nevertheless, the analysis indicates that the ACE-FTS offers unprecedented precision and vertical resolution in the UTLS, allowing a new global perspective on UTLS trace gas distributions. Our study shows that aircraft observations provide valuable data sets for satellite validation. By applying the methods presented here, more value can be extracted from historical data sets. It furthermore allows the comparison of satellites whose observation periods don't overlap, an issue frequently pointed out in the literature.

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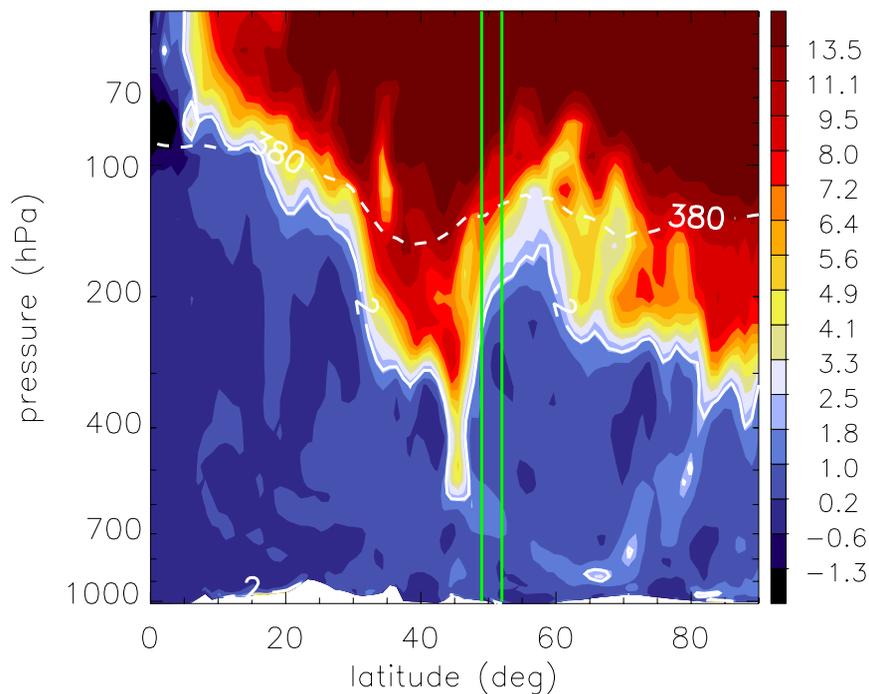


Fig. 1. Vertical cross section of ECMWF potential vorticity (colour coded) from equator to North Pole at 6° W on 10 November 2001 at 06:00 UTC. White thick line shows the dynamical tropopause (2 PVU), white dashed line the 380 K isentrope. Green lines indicate the location of two independent profiles fulfilling the coincidence criteria.

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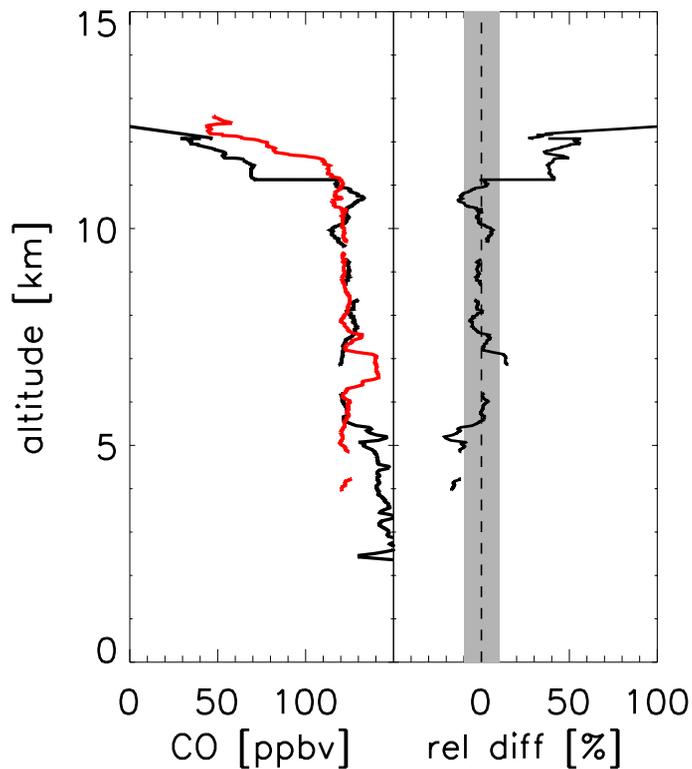


Fig. 2. Left panel: Vertical profiles of CO mixing ratios taken during ascent (black) and descent (red) of the SPURT aircraft on 10 November 2001. Right panel: Relative difference of the measured profiles. Gray shading indicates an error range of $\pm 10\%$.

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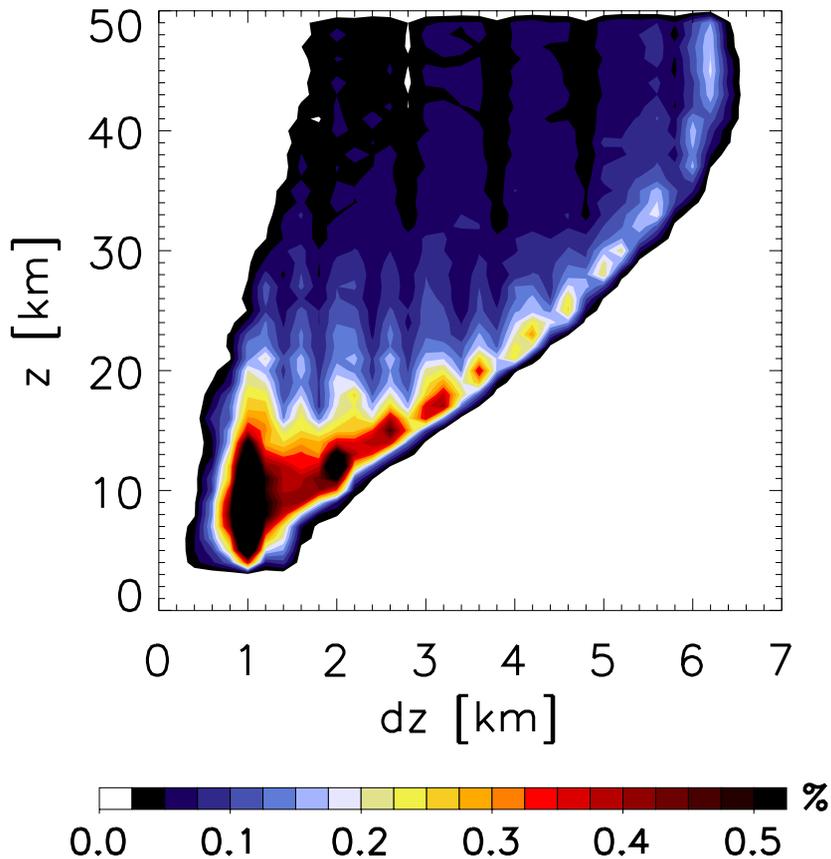


Fig. 3. Probability density function of the vertical spacing between the ACE-FTS measurements (dz) as a function of retrieval altitude. The vertical spacing in the tropopause region (between approximately 8 and 14 km) exhibits values much lower than 1 km.

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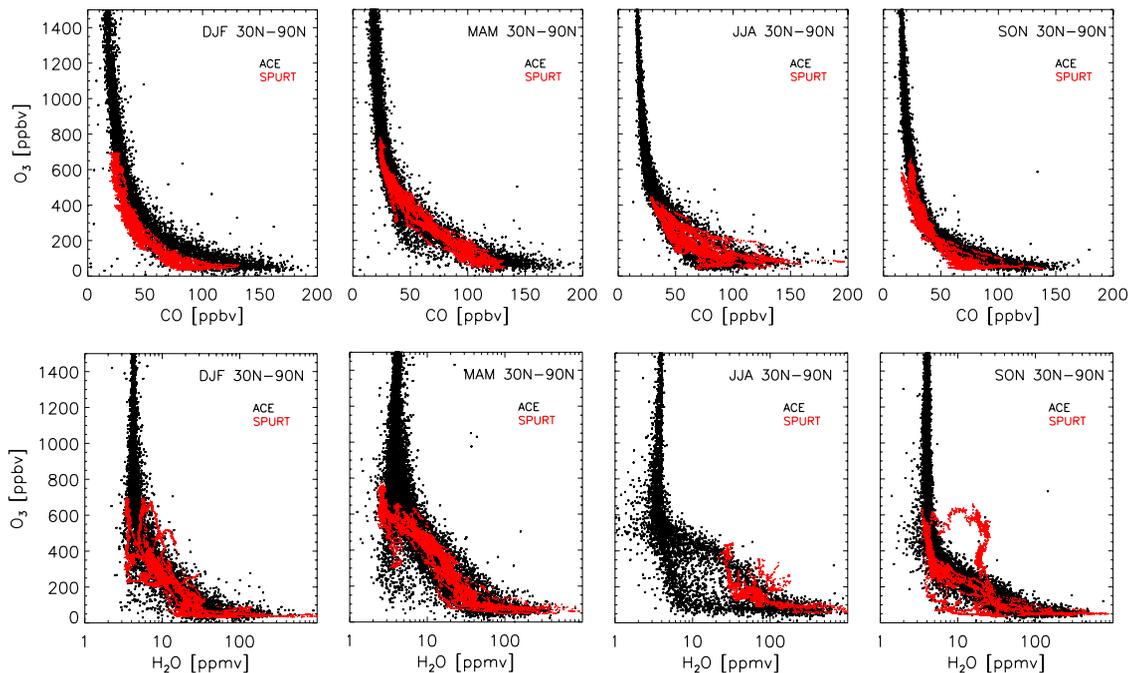


Fig. 4. CO-O₃ (upper panels) and H₂O-O₃ correlation (lower panels) for ACE-FTS satellite (black) and SPURT aircraft (red) data between 30° and 90° N. From left to right: winter, spring, summer, and autumn measurements. Note that the two data sets are obtained during different years.

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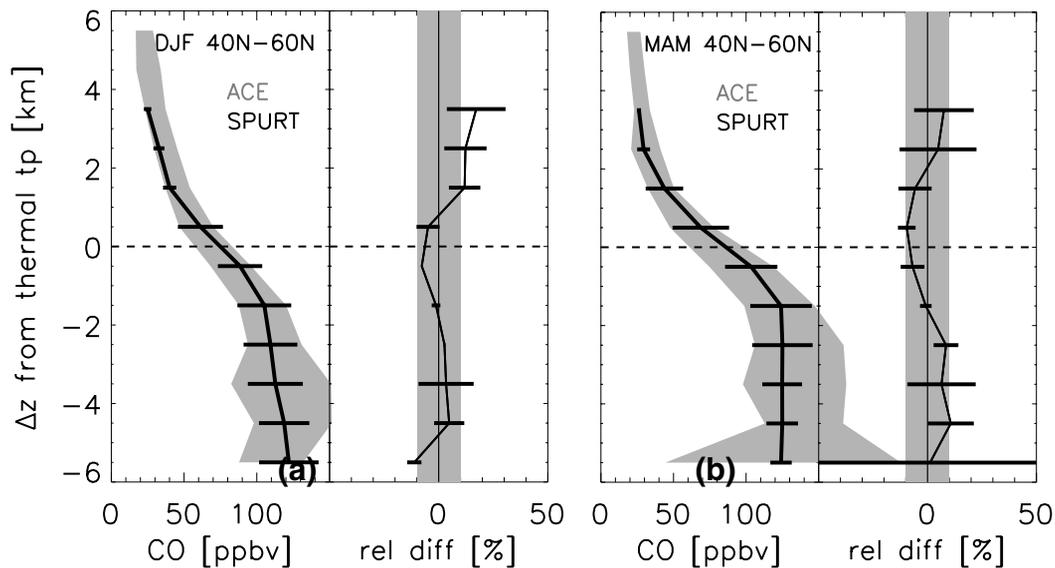


Fig. 5. Left panels: Vertical profiles of mean CO mixing ratios with standard deviations as a function of altitude relative to the thermal tropopause for SPURT (black) and ACE-FTS (gray), and for **(a)** DJF and **(b)** MAM. Right panels: Relative differences between the SPURT and ACE-FTS mean profiles (black line). The gray bar indicates $\pm 10\%$ relative difference. Horizontal bars show the uncertainties of the relative differences.

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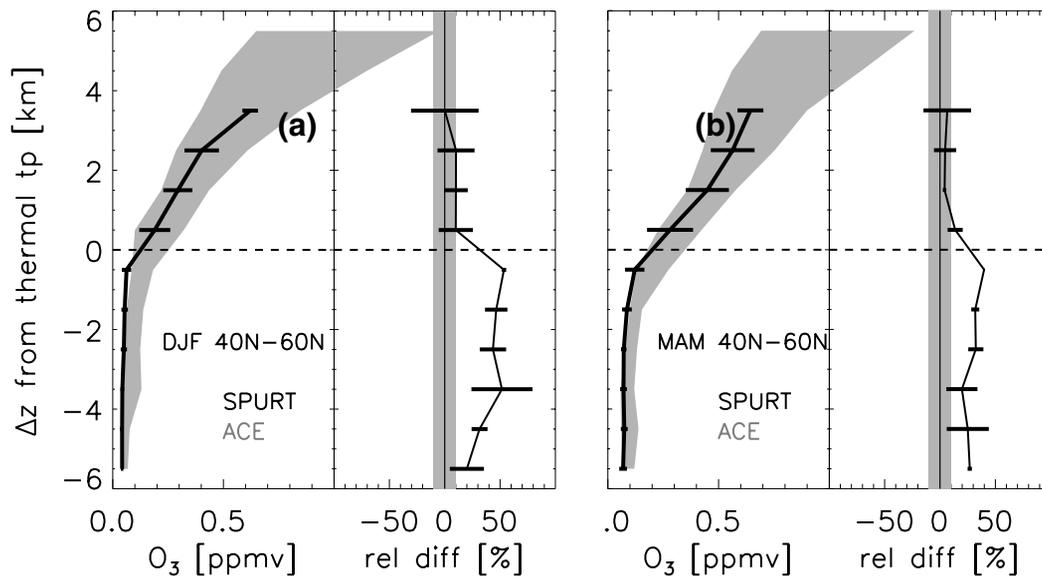


Fig. 6. Same as previous Fig. 5 but for O_3 .

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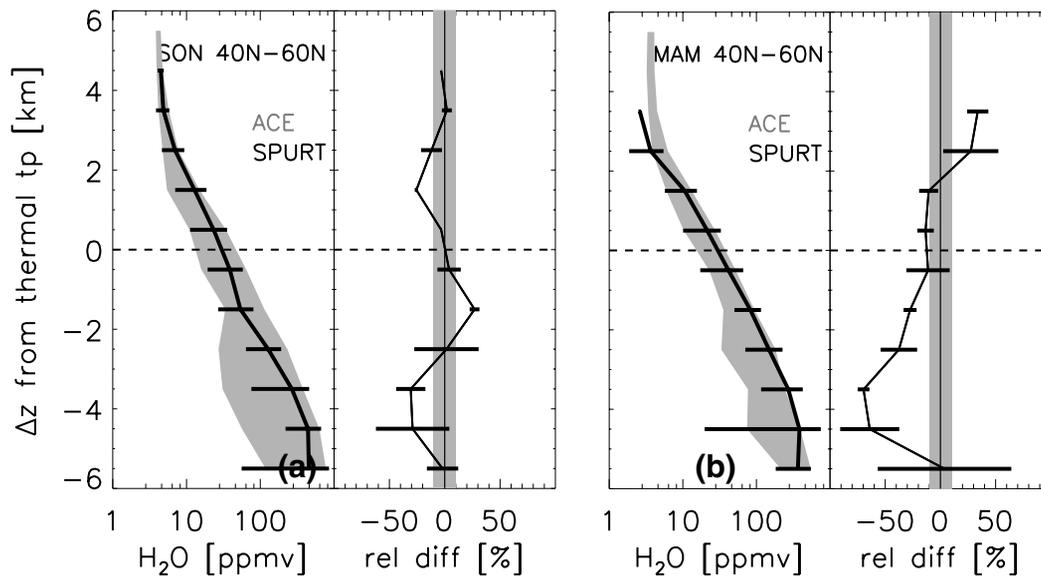


Fig. 7. Same as Fig. 5 but for H₂O and for (a) SON, and (b) MAM.

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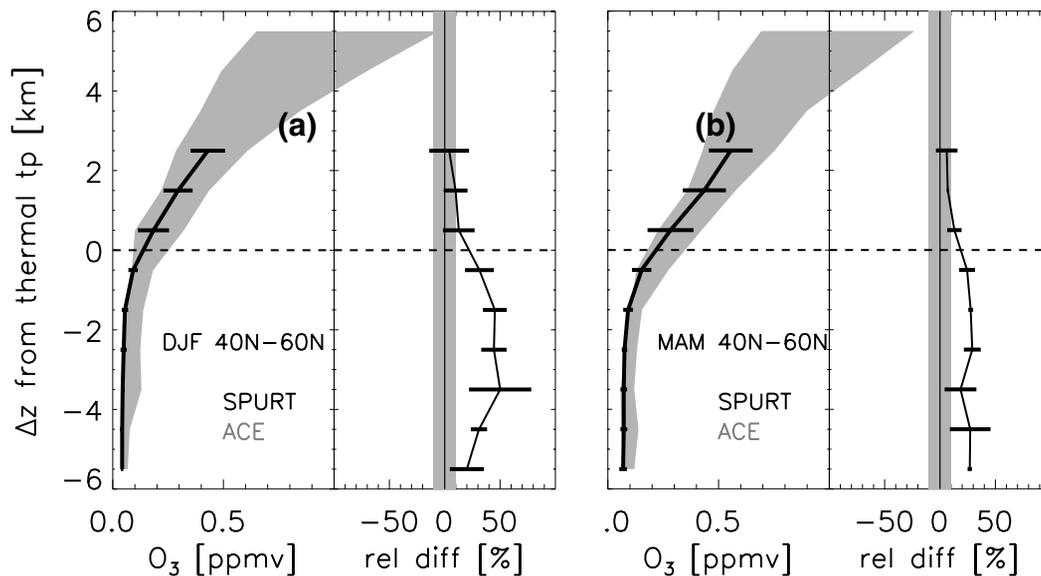


Fig. 8. Same as Fig. 6 but the SPURT vertical O_3 profile is smoothed out in order to account for the limited vertical resolution of the ACE-FTS instrument.

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