



## On sampling bias adjustment for sparsely observing satellite instruments for the example of carbonyl sulfide (OCS)

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**Abstract.** When computing climatological averages of atmospheric trace gas mixing ratios obtained from satellite-based measurements, sampling biases arise if data coverage is not uniform in space and time. Complete homogeneous spatio-temporal coverage is essentially impossible to achieve. Solar occultation measurements, by virtue of satellite orbits and the requirement of direct observation of the sun through the atmosphere, result in particularly sparse spatial coverage. In this study, a method is presented to adjust for such sampling biases when calculating climatological means. The method is demonstrated using carbonyl sulfide (OCS) measurements at 16 km altitude from the ACE-FTS (Atmospheric Chemistry Experiment Fourier Transform Spectrometer). At this altitude, OCS mixing ratios show a steep gradient between the poles and equator. ACE-FTS measurements, which are provided as vertically resolved profiles, and integrated stratospheric OCS columns are used in this study. The bias adjustment procedure requires no additional observations other than the satellite data product itself and is expected to be generally applicable when constructing climatologies of long-lived tracers from sparsely and heterogeneously sampled satellite data. In a first step of the adjustment procedure, a regression model is used to fit a 2-D surface to all available ACE-FTS OCS measurements as a function of day-of-year and latitude. The regression model fit is used to calculate an adjustment factor, which is then used to adjust each measurement individually. The mean of the adjusted measurement points of a chosen spatio-temporal frame is then used as the bias-free climatological value. When applying the adjustment factor to seasonal averages in 30° zones, the maximum spatio-temporal sampling bias adjustment was 11% for OCS mixing ratios at 16 km and 5% for the stratospheric OCS column. The adjustments were validated against the much denser and more homogeneous OCS data product from the limb-sounding MIPAS (Michelson Interferometer for Passive Atmospheric Sounding) instrument, and both the direction and sign of the adjustments were in agreement with the adjustment of the ACE-FTS data.

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## 1 Introduction

Creating climatologies of atmospheric trace gas concentrations from satellite-based measurements is usually done by collecting available observations into latitudinal and monthly/seasonal bins and calculating the respective averages (e.g. Jones et al. 2012 and Koo et al., 2017 compiled comprehensive trace gas climatologies from ACE-FTS observations). For such methods, an evenly distributed coverage, with no significant measurement gaps, is desirable to avoid the calculation of a biased mean. Satellite-based instruments, however, perform measurements only on distinct orbits, leaving spatio-temporal measurement gaps. This inhomogeneous sampling in space and time can introduce significant biases when calculating climatological averages (Aghedo et al., 2011; Toohey et al., 2013) if they are calculated in the traditional way. The magnitude of the sampling bias depends on the frequency spectrum of the spatial and temporal structure to be averaged. The bias can become particularly large when analysing data from solar occultation instruments that typically provide two measurements per orbit leading to sparse and spatially structured data coverage. The annual solar occultation sampling pattern of ACE-FTS, is shown in Figure 1a.

Recent studies by Aghedo et al. (2011), Sofieva et al. (2014), Toohey et al. (2013) and Millan et al. (2016) have investigated the effects of sampling biases for various satellite data products. Toohey et al. (2013) quantified the sampling bias for a number of satellites measuring ozone and water vapour. Depending on the trace gas, pressure level and latitude, they frequently found sampling biases as high as 20% and, in some cases, biases as high as 40% in regions with steep spatial and/or temporal gradients, such as in the vicinity of the polar vortex in both hemispheres. In an effort to quantify long-term trends in stratospheric ozone between 60°N and 60°S, Damadeo et al. (2018) used a regression model (described in Damadeo et al., 2014) to estimate the sampling biases of several solar occultation instruments. They found that these biases lead to about 1% per decade absolute percentage differences in derived ozone trends. A common attribute of all previous methods used to estimate the sampling bias is that they either use additional/multiple data products or atmospheric models that use *a priori* knowledge of atmospheric transport and chemistry. To our knowledge, to date, no method has been reported where the quantification of a sampling bias, and the adjustments made to correct for it, does not require additional independent information. Here, we present a novel approach to adjust measurements to mitigate sampling biases in climatological averages of carbonyl sulfide (OCS) measured by the solar occultation instrument ACE-FTS. The approach is suitable to be used on measurements with a seasonal cycle that is smooth enough to be represented by a low order expansion in Fourier series. Motivated by efforts to quantify the stratospheric burden of carbonyl sulfide (OCS) from ACE-FTS observations (Kloss, 2017, and Deshler et al., manuscript in preparation), we use OCS measurements from ACE-FTS. We introduce these measurements in Section 2 together with OCS measurements from Envisat-MIPAS that will be used to evaluate our method. Section 3 describes in detail the method developed to estimate and adjust for spatio-temporal sampling biases, which is then evaluated using the much denser and more homogeneous MIPAS data set in Section 4. The wider applicability of our method is discussed in Section 5.



## 2 Methods

### 2.1 ACE-FTS OCS observations

ACE-FTS is an infrared solar occultation spectrometer on the Canadian satellite SCISAT, delivering data since 2004 (Bernath et al., 2005). It measures in the spectral region from  $750 - 4400 \text{ cm}^{-1}$  ( $2.2 - 13.3 \text{ }\mu\text{m}$ ) with a spectral resolution of  $0.02 \text{ cm}^{-1}$ . From these data, mixing ratio values are derived for over 30 trace gases together with temperature and pressure in selected altitude regions. As a solar occultation spectrometer, ACE-FTS retrieves only 30 profiles per day (two per orbit, at sunrise and sunset) and thus exhibits significant data gaps in specific regions, as shown in Figure 1a. Measurements of the solar spectrum are made at tangent altitudes from 150 km down to 5 km (or cloud top) at a vertical resolution of 3 to 4 km. OCS mixing ratios are retrieved up to about 30 km altitude, above which the concentration typically drops below the detection limit.

In this study we use version 3.6 ACE-FTS OCS volume mixing ratio measurements between February 2004 and September 2016 (Boone et al. 2005, Boone et al. 2013), retrieved from microwindows in the range  $2036 \text{ cm}^{-1}$  to  $2056 \text{ cm}^{-1}$ . The average fitting error for OCS is a statistical error for the retrieval from the fitting process and is between 1% and 3%, for the period considered here. A detailed analysis of OCS from ACE-FTS version 2.2 is presented in Barkley et al. (2008).

The stratospheric OCS column is calculated by vertically integrating the stratospheric concentration profiles from the dynamical tropopause to the top of the retrieved OCS profiles, where mixing ratios decrease to zero. The dynamical tropopause is defined as 380 K potential temperature in the tropics and 3.5 PV units at latitudes poleward of  $30^\circ$ , and is calculated from ECMWF ERA-Interim data (Dee et al., 2011). Partial columns are then accumulated into  $1^\circ \times 1^\circ$  bins over the chosen time period (e.g. one season: DJF, MAM, JJA, SON). Where there is more than one partial column in any bin, the mean is calculated. Values for bins with no profiles are linearly interpolated or, close to the poles, are extrapolated from the two bins closest to the respective pole. To obtain the stratospheric burden for a particular region, respective columns are summed.

### 2.2 OCS observations by Envisat-MIPAS

The Michelson Interferometer for Passive Atmospheric Sounding (MIPAS) is a mid-infrared spectrometer on board the ESA (European Space Agency) satellite ENVISAT. It is a limb-sounding instrument, analysing the spectral radiance emitted by atmospheric trace gases. From its sun-synchronous polar orbit, MIPAS measures vertical profiles of multiple trace gases, including OCS. From 2002 – 2012 MIPAS operated in the spectral region between  $685 - 2410 \text{ cm}^{-1}$  ( $4.1 - 14.6 \text{ }\mu\text{m}$ ), at a resolution of  $0.025 \text{ cm}^{-1}$  until 2004 and then at  $0.065 \text{ cm}^{-1}$  from 2005 onwards (Fischer et al., 2008). The vertical sampling is around 3 km in the altitude range from about 5 to 150 km above the clouds. With a horizontal sampling of about 400 to 500 km along the orbit MIPAS measured 1000 vertical profiles per day from 2002 to 2004 and 1400 between 2005 and 2012, covering almost all latitudes from  $88^\circ\text{S}$  to  $88^\circ\text{N}$ . These are about 40 times as many profiles as can be provided by ACE-FTS. OCS profiles are retrieved in spectral windows between  $839 \text{ cm}^{-1}$  and  $876 \text{ cm}^{-1}$  (Glatthor



et al., 2015; Glatthor et al., 2017). The retrieval uncertainty for a single OCS scan is estimated to be 10% between 10 and 15 km, 26% at 20 km and increasing up to 195% at 40 km altitude (Glatthor et al., 2015).

### 2.3 A regression model representation of the OCS field

5 Adjusting for spatio-temporal sampling biases requires some description of the gap-free field. The field could be obtained, for example, from chemistry-transport-model output, or, as mentioned above, from a satellite data set providing higher spatial and temporal sampling. In this study, we use the sparse data themselves to create a gap-free OCS field through the application of a regression model fit. The regression model is used to fit a continuous smooth 2-D (time and latitude) surface either to OCS mixing ratios at a given altitude or to fields of OCS partial columns. The regression model is of the form:

$$OCSEst = a_0 + \sum_{i=1}^N \left[ a_{2i-1} \times \sin\left(\frac{2\pi d}{365.25}\right) + a_{2i} \times \cos\left(\frac{2\pi d}{365.25}\right) \right] \quad (1)$$

10 where the Fourier expansion in  $N$  accounts for the annual cycle in the compound of interest and  $d$  is the day of the year. To accommodate the latitudinal structure in OCS, each of the  $a_i$  coefficients are expanded in a Legendre series of index  $M$ . Values for  $N$  and  $M$  must be carefully selected to capture as much of the latitudinal and seasonal structure in OCS as possible, but must also avoid overfitting. For OCS, optimal fits were found for  $N=1$  and  $M=4$  resulting in a total of 15 fit coefficients. The output of Equation (1),  $OCSEst$  is visualized in Figure 1b. Applying fewer coefficients does not represent the OCS variability sufficiently, while applying more coefficients showed minima and maxima that are not observed in ACE-FTS as signs of  
15 overfitting.

A total of 12.5 years of ACE-FTS OCS mixing ratios at 16 km altitude are passed to the regression model to obtain the 15 fit coefficients (see Figure 1a). A different set of fit coefficients is obtained from the regression model when it is fitted to the stratospheric partial columns. Note that because the regression model provides a value for any arbitrary latitude and day of the year, it meets the ‘continuous’ requirement for  $OCSEst$ . The extent to which the regression model can capture the true underlying morphology of the latitude vs. time OCS field depends on the OCS measurement coverage: however,  
20 with too many gaps in the measurements, the regression model will be required to have lower  $N$  and  $M$  expansions and may not capture subtleties in the OCS field to avoid over-and under fitting in areas of low data coverage. As a solar occultation spectrometer with only 30 measurements per day, ACE-FTS exhibits significant data gaps in specific regions (as seen in Figure 1a) that restrict the expansions in Equation (1) to  $N=1$  and  $M=4$ .

## 3 Sampling Bias Adjustment

Using the gap-free field as described in Section 2.3 ( $OCSEst$ ), adjusted values can then be calculated as:



$$OCS_{adj} = OCS_{orig} \times \frac{\overline{OCSEst}}{OCSEst(lat, long, t)} \quad (2)$$

where  $OCS_{adj}$  is the OCS value adjusted for its representativeness of the temporal-zonal mean,  $OCS_{orig}$  is the unadjusted OCS measurement,  $\overline{OCSEst}$  is some estimate of the true OCS temporal-zonal mean, and  $OCSEst(lat, long, t)$  is the estimated OCS concentration at the location and time of the actual

of the year, it meets the ‘continuous’ requirement for  $OCSEst$ .  $OCSEst$  does not have to be quantitatively correct – any biases divide out in Equation (2). There are several options for obtaining  $OCSEst$ . The only prerequisites are that the  $OCSEst$  field represents the true underlying temporal and spatial morphology of the OSC field (though, as pointed out above, the values themselves do not need to be exact) and it needs to be continuous in so far as spatio-temporal means can be calculated from the  $OCSEst$  field without any spatio-temporal sampling gaps. The procedure for adjusting the sampling bias when calculating an average mixing ratio for a defined region over a given time period is illustrated in Figure 1. As examples, the method is explained in detail for two representative latitude-time boxes: one at 30 – 60°N for JJA (red box in Figure 1a-c) and one for 60 to 90°S for DJF (black box in Figure 1a-c).

Figure 1a shows the OCS mixing ratio values from 12.5 years of ACE-FTS observations as a function of latitude and time-of-year. The small year-to-year shifts in the latitudinal coverage of ACE-FTS causes small offsets between the traces for individual years seen in Figure 1a. The red and black boxes in Figure 1 indicate the selected time and latitude frames used to demonstrate the application of this method. The boxes were chosen as examples for the highest (red box) and lowest (black box) ACE-FTS latitude coverage. The climatological mean OCS pattern, represented as the regression model fit to the 12.5 years of ACE-FTS measurements, as a function of latitude and season, is shown in Figure 1b. Figure 2 shows the same for the OCS stratospheric columns. Values for  $\overline{OCSEst}$  for the two example spatio-temporal means, indicated by the red (JJA, 30°N-60°N) and black (DJF, 60°S-90°S) boxes in Figure 1, can be calculated analytically, without any spatio-temporal sampling bias, from the regression model fit.

ACE-FTS data ( $OCS_{orig}$ ) for 2010 are shown in Figure 1c. OCS mixing ratios from the regression model at the same latitudes and times as  $OCS_{orig}$  provide  $OCSEst(lat, long, t)$  allowing the original data to be adjusted using Equation (2). The advantage of applying Equation (2) rather than simply using  $\overline{OCSEst}$  as the zonal mean seasonal mean is that trends and year-to-year variability observed in the data set remain. Equation (2) adjusts each measurement to be more indicative of the zonal seasonal mean. Figure 1d shows the adjusted ACE-FTS data set for the example of the red box in Figure 1c. These data points, now adjusted for their representativeness of the zonal seasonal mean, can then be used to calculate a better estimate of the true zonal seasonal mean for the temporal and spatial domain of the red box. It should be noted that only derived averages are adjusted and not the individual data points. The average values should be more representative for the mean of the compound within each chosen box than without applying the adjustment method. The adjustment should not be applied to individual data points for any other purpose: clearly, the sampling bias is a systematic error type that only arises when deriving spatio-temporal averages and it does not impair the quality of individual data points at a particular location and time.



## 4 Evaluation of the adjustment procedure

### 4.1 Case study results

As seen in Figure 1a and shown in Barkley et al. (2008), OCS mixing ratios at a specific altitude (here 16km) decrease with increasing latitude. The stratospheric partial column distribution, shown in Figure 2, is quite different. Because both pressure and OCS mixing ratios rapidly decrease with height above the tropopause, the major fraction of the stratospheric OCS column resides in the few kilometres just above the tropopause and thus the significant decrease in tropopause height with latitude leads to lower partial columns in the tropics and higher values closer to the poles. For the same reason, the annual cycle and day-to-day variability of the dynamical tropopause rather than the annual cycle in OCS mixing ratios largely controls the temporal variability of the stratospheric OCS partial columns, resulting in a more variable stratospheric OCS partial column field compared to the mixing ratio distribution shown in Figure 1a, potentially confounding the adjustment procedure.

Figure 3 shows the frequency distribution of ACE-FTS OCS measurements at 16 km from 2004 to 2016 for the two chosen latitude bands and time regions. The green histograms show the distribution of the original measurements and the blue histograms show the distribution of the adjusted measurements using Equation (2). Here, all individual measurements are adjusted for biases in the seasonal zonal mean. The shifts in the mean values and contraction of the standard deviations provide useful summary metrics of the effects of the applied spatio-temporal sampling bias adjustments. The distribution of all 12 years of data between 60°S and 90°S in the southern hemispheric summer (DJF) is shown in Figure 3a. This example was chosen because it displays the highest shift of 28 pptv or 11% in the mean OCS mixing ratios after applying the adjustment. The decrease in the mean value from 293 to 265 pptv in the latitude band from 60°S to 90°S can be explained by the fact that there are large measurement gaps at the southernmost latitudes, especially in DJF, and no measurements between 85°S and 90°S. Decreasing mixing ratios towards the poles, and measurement gaps where lower mixing ratios are expected, lead to a high biased mean over the chosen box (here: black box Figure 1a) when only averaging the available measurements. The true mean over the entire box is expected to be lower than the mean of only the available data. Thus, the shift of the mean to a lower value seen in Figure 3a qualitatively represents an adjustment of the simple data average towards the true mean of OCS mixing ratios over the entire box, and therefore at least a partial remedy for the sampling bias. Because Equation (2) generally shifts each data point towards the mean of the distribution, the standard deviation of the adjusted data will be lower than the standard deviation of the original data set. This is because in the original data set both measurement uncertainties and actual variability inside the considered box add on to the resulting standard deviation. Note that the observed reduction in the standard deviation (8 pptv in our black box example) reflects neither a reduction of the statistical uncertainty associated with the derived mean, nor a reduced variability over the entire box compared to only the available data. In fact, if actual observations covering the entire box were available, then their standard deviation would most likely be higher than that of the limited data because values would vary over a wider range of mixing ratios.

The histograms in Figure 3b show the data distribution for the red box in Figure 1, i.e. between 30°N and 60°N in northern hemispheric summer (JJA). Here, the adjustment method yields only a small shift in the average of 6 pptv (1.5%) because the entire chosen latitude range is covered by ACE-FTS



measurements, which are therefore much more representative of the true mean value of the entire box compared to the previous example. For the red box, the original measurement values are more or less evenly distributed around the regression model mean, and Equation (2) shifts data towards the mean from both sides. Consequently, the reduction in the standard deviation by 32% is larger than in the previous example.

To assess whether the methodology quantitatively adjusts the sampling bias, a validation against an independent data set was performed and will be described in the following section.

#### 4.2 Quantitative evaluation based on MIPAS observations

To quantify the sampling bias arising from the sparse ACE-FTS sampling for a chosen latitude-time box, the OCS data product from the MIPAS instrument with its much denser data coverage is used. Because of the dense sampling pattern and an almost complete latitude coverage (down to 88°S), the sampling bias of MIPAS is negligible compared to that of ACE-FTS. For the best possible quantitative evaluation, the spatio-temporal box in the ACE-FTS measurements with the highest observed sampling bias is chosen: December 2009 – February 2010, 60°S to 90°S (i.e. the black box in Figure 1). We compare the average of all MIPAS observations in a particular box to the average of only those MIPAS observations that are roughly equivalent in space and time to the available ACE-FTS observations in that box (i.e. only MIPAS measurements from 1<sup>st</sup> of December 2009 to 5<sup>th</sup> of January 2010 between 60°S and 68°S are used). Figure 4 visualizes how much denser the MIPAS sampling is, compared to that of ACE-FTS in the chosen latitude-time box. It should be noted that Glatthor et al. (2017) found a systematic difference between OCS from MIPAS and ACE-FTS that is highest at 14 km altitude, with ACE-FTS measurements lower by 75 to 100 ppt. We also find the ACE-FTS mean value between 60°S and 90°S to be 115 ppt (28%) lower than the mean value of MIPAS. Therefore, relative rather than absolute mixing ratio differences are used to quantitatively describe the sampling bias in the comparison below.

Using the chosen spatio-temporal box (black box in Figure 1), we show in Figure 5 histograms of the relative frequency distributions of all MIPAS OCS mixing ratios at 16 km observed between 60°S and 90°S in DJF 2009/10 and of only those MIPAS observations roughly covering the ACE-FTS sampling locations in that particular box (i.e. only MIPAS measurements from 1<sup>st</sup> of December 2009 to 5<sup>th</sup> of January 2010 between 60°S and 68°S are used). The histograms in Figure 3a and Figure 5 look similar in terms of shape and relative position. When we compare the two histograms in Figure 5 it becomes apparent that extending the sampling space over the entire box (down to 88°S) changes the distribution by adding additional lower mixing ratio values that were measured at the southernmost latitudes. The difference between the mean values of both histograms is 46 pptv, equivalent to a relative deviation of about 11%, with the average of the full data set being lower. Thus, the difference has the same direction and magnitude as the shift in mean value when using the ‘adjusted ACE-FTS data, compared to the original ACE-FTS data (Figure 3a). For this example, the performed sampling bias adjustment of the climatological mean from ACE-FTS data appears to work not only qualitatively but also quantitatively.



### 4.3 Significance

To investigate the scientific relevance and applicability of the proposed sampling bias adjustment, climatologies for the seasonal stratospheric OCS columns and OCS mixing ratios at 16 km altitude are calculated with and without sampling bias adjustments.

Due to the satellite orbit, ACE-FTS does not measure in the latitude ranges 85°N – 90°N and 85°S – 90°S, which can lead to a higher sampling bias close to the poles compared to the tropics and mid-latitudes where mostly all latitudes are covered within each season. Additionally, OCS mixing ratios exhibit lower stratospheric variability in the tropics. Therefore, the sampling bias is higher towards the poles and lower in the tropics. For the majority of points, from 60°N to 60°S (c.f. Figure 1), the modifications made using Equation (2) has only a minimal effect and is within the measurement uncertainty calculated using the ACE-FTS error estimates (see Toohey et al., 2010, for details on ACE-FTS error estimation).

The largest difference between the seasonal mean calculated using original OCS measurements and the seasonal mean calculated using the adjusted OCS measurements occurs in the latitude band 60°S - 90°S. Figure 6 shows the seasonal mean of the stratospheric column and of mixing ratios at 16 km for this latitude band as calculated from the adjusted data set in blue and the original ACE-FTS measurements in red. The relative difference between the mean values from the original and adjusted data set varies between 0.1 and 5.1% for the stratospheric columns (for the 5.1% difference: 1.29 kg/km<sup>2</sup> instead of 1.36 kg/km<sup>2</sup>) and between 0 and 11% for OCS mixing ratios at 16km (for the 11% difference: 216 ppt instead of 195 ppt). In this particular case (Figure 5), there is a marginal impact on the amplitude of the seasonal cycle as the adjustment most significantly reduces the austral summer OCS maximum at 16 km in virtually all years. No significant trends are apparent in either the original or adjusted data. Theoretically, budget related trends in atmospheric OCS, which are independent of the season would not be affected by the adjustment method and, if present, would show up in both the original and adjusted data sets. Trends related to dynamic changes in one particular region and season would also show up in both data sets if data from that region and season existed.

## 5 Conclusion and Discussion

In this study, we present a method to adjust the spatio-temporal sampling bias in climatologies calculated from sparsely sampled satellite observations without requiring additional observational evidence beyond the data set used. Generally, the method should work for any atmospheric compound or property of which the variability follows defined seasonal and latitudinal patterns and can therefore be sufficiently well described using a regression model approach. The method has been shown to quantitatively adjust the sampling bias in seasonal 30° latitude band climatologies of OCS mixing ratios at 16 km altitude and OCS stratospheric column constructed from ACE-FTS observations. ACE-FTS with its solar occultation viewing geometry and therefore sparse and heterogeneous sampling pattern is particularly sensitive to the occurrence of a sampling bias when calculating climatologies (Toohey et al., 2013). OCS with its atmospheric variability outside the boundary layer limited to large spatial (100s of km) and temporal (i.e. seasons) scales (Barkley et al., 2008) provides an ideal tracer to investigate and demonstrate the sampling bias adjustment method. We expect the method to be applicable



in the construction of climatologies for tracers with variabilities on similar scales, including most compounds for which climatologies from ACE-FTS data have been compiled by Jones et al. (2012) and Koo et al. (2017). Even though it is important to consider the sampling pattern of satellite based measurements, which leads to a sampling bias, at least for OCS the influence of the sampling bias is too small to significantly alter the scientific conclusions of climatologies.

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## References

Aghedo, A. M., Bowman, K. W., Shindell, D. T., and Faluvegi, G.: The impact of orbital sampling, monthly averaging and vertical resolution on climate chemistry model evaluation with satellite observations, *Atmos. Chem. Phys.*, 11, 6493-6514, 2011.

Barkley, M. P., Palmer, P. I., Boone, C. D., Bernath, P. F., and Suntharalingam, P.: Global distributions of carbonyl sulfide in the upper troposphere and stratosphere, *Geophys. Res. Lett.*, 35, 2008.

Bernath, P. F., McElroy, C. T., Abrams, M. C., Boone, C. D., Butler, M., Camy-Peyret, C., Carleer, M., Clerbaux, C., Coheur, P. F., Colin, R., DeCola, P., Bernath, P. F., McElroy, C. T., Abrams, M. C., Boone, C. D., Butler, M., Camy-Peyret, C., Carleer, M., Clerbaux, C., Coheur, P. F., Colin, R., DeCola, P., DeMaziere, M., Drummond, J. R., Dufour, D., Evans, W. F. J., Fast, H., Fussen, D., Gilbert, K., Jennings, D. E., Llewellyn, E. J., Lowe, R. P., Mahieu, E., McConnell, J. C., McHugh, M., McLeod, S. D., Michaud, R., Midwinter, C., Nassar, R., Nichitiu, F., Nowlan, C., Rinsland, C. P., Rochon, Y. J.,



- Rowlands, N., Semeniuk, K., Simon, P., Skelton, R., Sloan, J. J., Soucy, M. A., Strong, K., Tremblay, P., Turnbull, D., Walker, K. A., Walkty, I., Wardle, D. A., Wehrle, V., Zander, R., and Zou, J.: Atmospheric Chemistry Experiment (ACE): Mission overview, *Geophys. Res. Lett.*, 32, 2005.
- Boone, C. D., Nassar, R., Walker, K. A., Rochon, Y., McLeod, S. D., Rinsland, C. P., and Bernath, P. F.: Retrievals for the atmospheric chemistry experiment Fourier-transform spectrometer, *Appl. Optics*, 44 (33), 7218–7231, 2005.
- 5 Boone, C. D., Walker, K. A., and Bernath, P. F.: Version 3 Retrievals for the Atmospheric Chemistry Experiment Fourier Transform Spectrometer (ACE-FTS), *The Atmospheric Chemistry Experiment ACE at 10: A Solar Occultation Anthology* (Peter F. Bernath, editor, A. Deepak Publishing, Hampton, Virginia, U.S.A., 2013), 103–127, 2013.
- Damadeo, R. P., Zawodny, J. M., and Thomason, L. W.: Reevaluation of stratospheric ozone trends from SAGE II data using a simultaneous temporal and spatial analysis, *Atmos. Chem. Phys.*, 14, 13455–13470, 2014.
- 10 Damadeo, R. P., Zawodny, J. M., Remsberg, E. E., and Walker, K. A.: The impact of nonuniform sampling on stratospheric ozone trends derived from occultation instruments, *Atmos. Chem. Phys.*, 18, 535–554, 2018.
- Dee, D. P., Uppala, S. M., Simmons, A. J., Berrisford, P., Poli, P., Kobayashi, S., Andrae, U., Balmaseda, M. A., Balsamo, G., Bauer, P., Bechtold, P., Beljaars, A. C. M., van de Berg, L., Bidlot, J., Bormann, N., Delsol, C., Dragani, R., Fuentes, M., Geer, A. J., Haimberger, L., Healy, S. B., Hersbach, H., Hólm, E. V., Isaksen, I., Kållberg, P., Köhler, M., Matricardi, M., McNally, A. P., Monge-Sanz, B. M., Morcrette, J.-J., Park, B.-K., Peubey, C., de  
15 Rosnay, P., Tavolato, C., Thépaut, J.-N. and Vitart, F., The ERA-Interim reanalysis: configuration and performance of the data assimilation system. *Q.J.R. Meteorol. Soc.*, 137: 553–597. doi: 10.1002/qj.828, 2011.
- Fischer, H., Birk, M., Blom, C., Carli, B., Carlotti, M., von Clarmann, T., Delbouille, L., Dudhia, A., Ehret, D., Endemann, M., Flaud, J. M., Gessner, R., Kleinert, A., Koopman, R., Langen, J., Lopez-Puertas, M., Mosner, P., Nett, H., Oelhaf, H., Perron, G., Remedios, J., Ridolfi, M., Stiller, G., and Zander, R.: MIPAS: an instrument for atmospheric and climate research, *Atmos. Chem. Phys.*, 8, 2151–2188, 2008.
- 20 Glatthor, N., Hopfner, M., Baker, I. T., Berry, J., Campbell, J. E., Kawa, S. R., Krysztofiak, G., Leyser, A., Sinnhuber, B. M., Stiller, G. P., Stinecipher, J., and von Clarmann, T.: Tropical sources and sinks of carbonyl sulfide observed from space, *Geophys. Res. Lett.*, 42, 10082–10090, 2015.
- Glatthor, N., Hopfner, M., Leyser, A., Stiller, G. P., von Clarmann, T., Grabowski, U., Kellmann, S., Linden, A., Sinnhuber, B. M., Krysztofiak, G., and Walker, K. A.: Global carbonyl sulfide (OCS) measured by MIPAS/Envisat during 2002–2012, *Atmos. Chem. Phys.*, 17, 2631–2652, 2017.
- Jones, A., Walker, K. A., Jin, J. J., Taylor, J. R., Boone, C. D., Bernath, P. F., Brohede, S., Manney, G. L., McLeod, S., Hughes, R., and Daffer, W. H.:  
25 Technical Note: A trace gas climatology derived from the Atmospheric Chemistry Experiment Fourier Transform Spectrometer (ACE-FTS) data set, *Atmos. Chem. Phys.*, 12, 5207–5220, 2012.



Kloss, C.: Carbonyl Sulfide in the stratosphere: airborne instrument development and satellite based data analysis, Ph.D., Chemistry Department, Bergische Universität Wuppertal, Wuppertal, 2017.

Koo, J. H., Walker, K. A., Jones, A., Sheese, P. E., Boone, C. D., Bernath, P. F., and Manney, G. L.: Global climatology based on the ACE-FTS version 3.5 dataset: Addition of mesospheric levels and carbon-containing species in the UTLS, *J. Quant. Spectrosc. Radiat. Transf.*, 186, 52-62, 2017.

5 Millan, L. F., Livesey, N. J., Santee, M. L., Neu, J. L., Manney, G. L., and Fuller, R. A.: Case studies of the impact of orbital sampling on stratospheric trend detection and derivation of tropical vertical velocities: solar occultation vs. limb emission sounding, *Atmos. Chem. Phys.*, 16, 11521-11534, 2016.

Sofieva, V. F., Kalakoski, N., Paivarinta, S. M., Tamminen, J., Laine, M., and Froidevaux, L.: On sampling uncertainty of satellite ozone profile measurements, *Atmos. Meas. Tech.*, 7, 1891-1900, 2014.

10 Toohey, M., Strong, K., Bernath, P. F., Boone, C. D., Walker, K. A., Jonsson, A. I., and Shepherd, T. G.: Validating the reported random errors of ACE-FTS measurements, *J. Geophys. Res.-Atmos.*, 115, 2010.

Toohey, M., Hegglin, M. I., Tegtmeier, S., Anderson, J., Anel, J. A., Bourassa, A., Brohede, S., Degenstein, D., Froidevaux, L., Fuller, R., Funke, B., Gille, J., Jones, A., Kasai, Y., Kruger, K., Kyrola, E., Neu, J. L., Rozanov, A., Smith, L., Urban, J., von Clarmann, T., Walker, K. A., and Wang, R. H. J.: Characterizing sampling biases in the trace gas climatologies of the SPARC Data Initiative, *J. Geophys. Res.-Atmos.*, 118, 11847-11862, 2013.

15 Figures

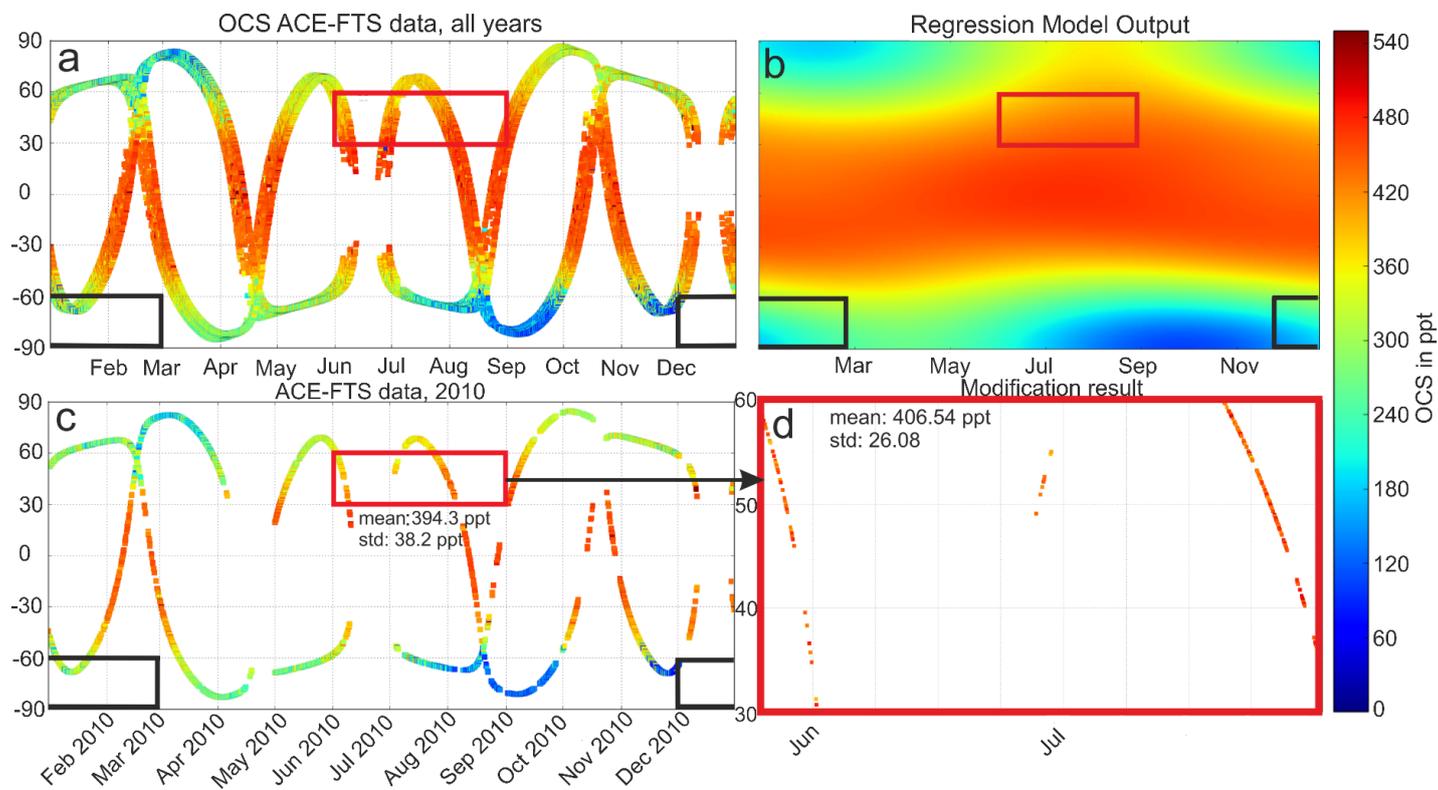


Figure 1: Schematic illustration of how the sampling bias is estimated and adjusted for OCS mean mixing ratio at 16 km altitude in any chosen time/latitude bin. Two examples are discussed in more detail in the text and are indicated by the red and black boxes. (a): All ACE-FTS measurements (2004 - 2016) as a function of day-of-year and latitude. (b): Regression model output to the ACE-FTS data of (a). (c) ACE-FTS measurements in 2010. (d): 'adjusted' data set, i.e. after the applying Equation (2) to the ACE-FTS measurements shown in (c), for the red box.

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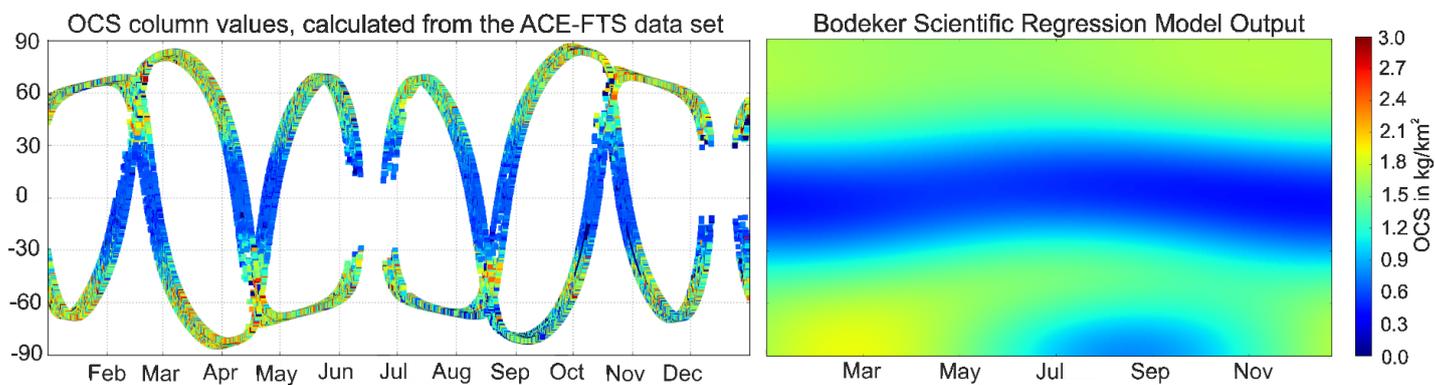


Figure 2: Stratospheric OCS column values in kg/km<sup>2</sup> which were calculated for a 1° x 1° grid, using the ACE-FTS OCS data and the resulting Regression Model Output.

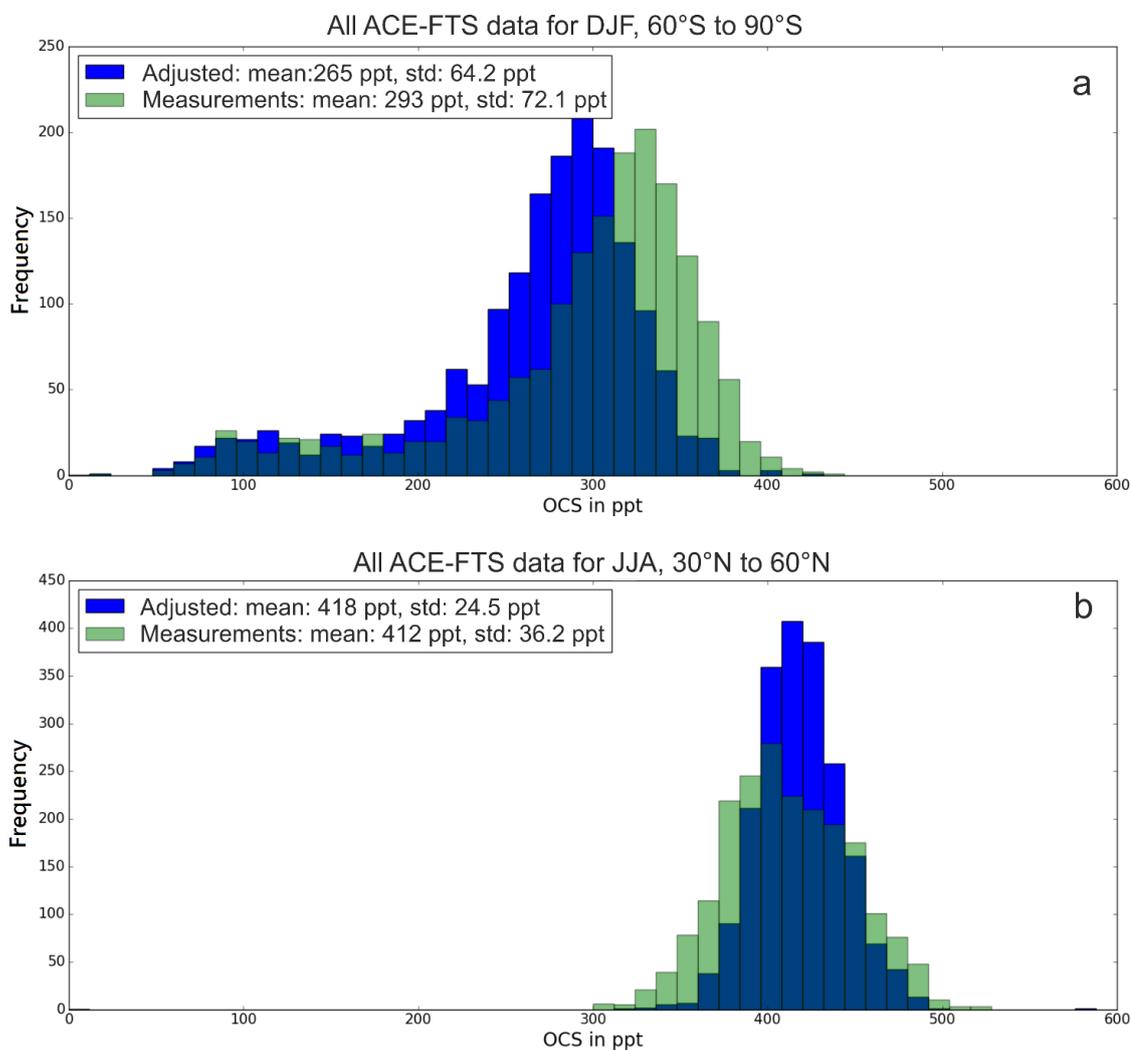


Figure 3: Comparison of the distributions and resulting mean and standard deviation values of measured (green) OCS and the 'adjusted' measurements using Equation (2) (blue) for the same time/latitude bins indicated by the black (a) and red (b) boxes in Figure 1. Histograms include all 12 years of ACE-FTS OCS mixing ratio measurements at 16km altitude.

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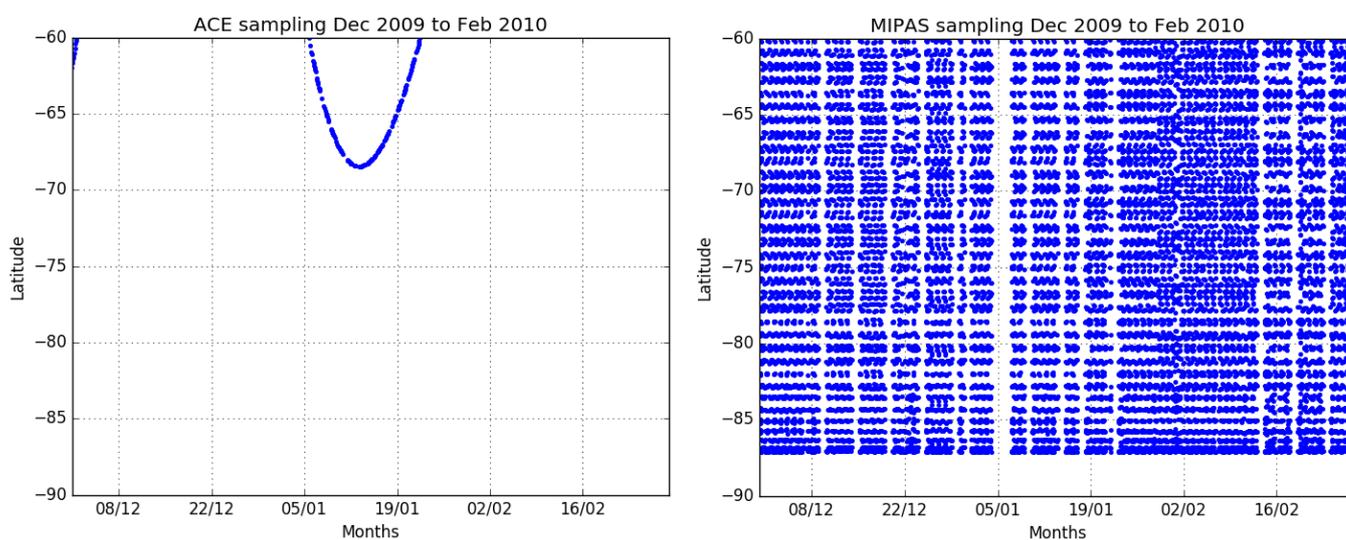
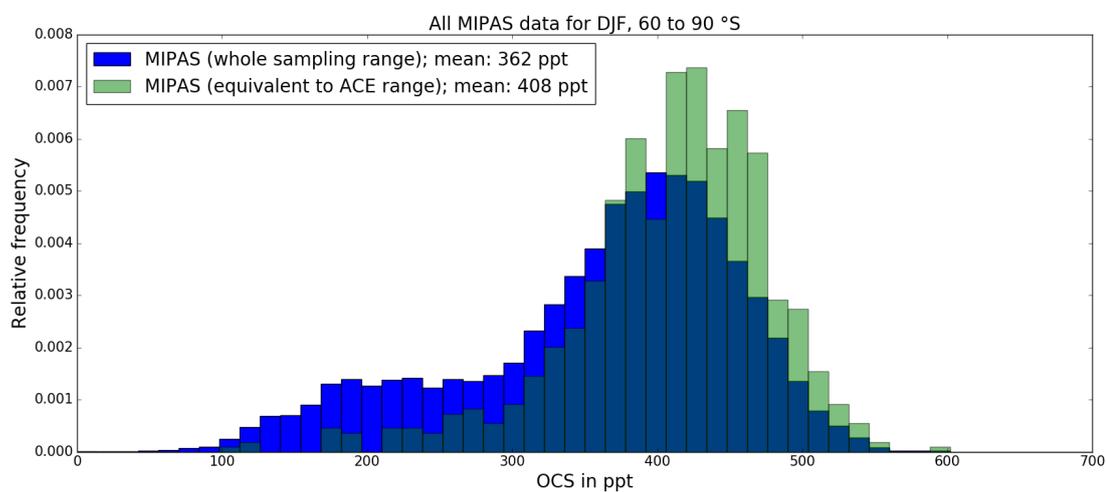


Figure 4: Sampling pattern of ACE-FTS and MIPAS between 60°S and 90°S, December 2009 to February 2010.



5 **Figure 5: MIPAS data distribution for DJF 2009 - 2010, 60°S to 90°S, for all available MIPAS OCS mixing ratio measurements at 16km altitude (blue) and for MIPAS OCS profiles in a comparable latitude and time frame as ACE-FTS measurements (green). The respective ACE-FTS plot, considering all years during DJF of ACE-FTS (to establish a reasonable statistic) is shown in Figure 3a.**

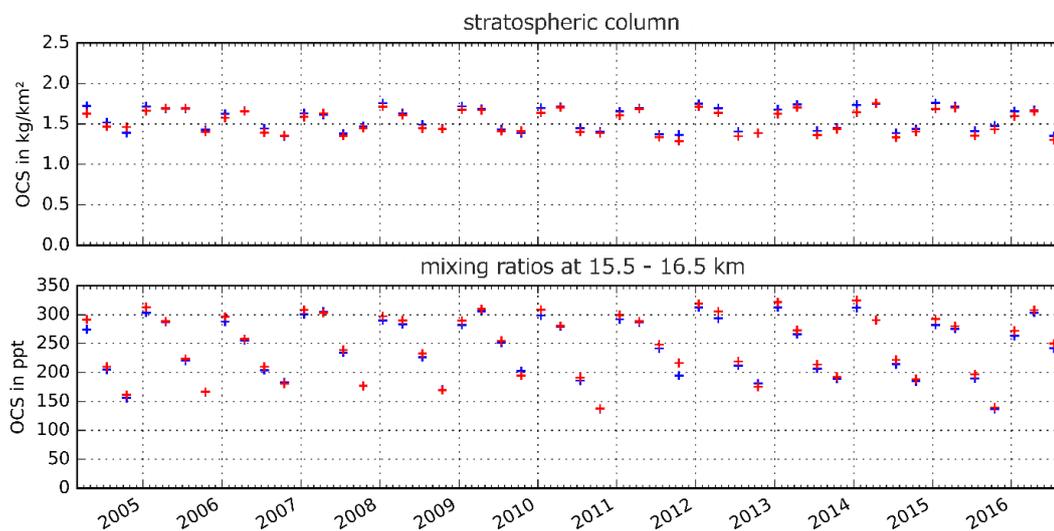


Figure 6: Comparison of the unadjusted (red) and adjusted (blue) seasonal OCS stratospheric columns and seasonal averaged OCS mixing ratio from 15.5 to 16.5 km altitude between 60°S to 90°S.