The STRatospheric Estimation Algorithm from Mainz (STREAM): Implementation for GOME-2

O3M-SAF Visiting Scientist Activity

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1 Summary

The STRatospheric Estimation Algorithm from Mainz (STREAM) determines stratospheric column densities of NO_2 which are needed for the retrieval of tropospheric columns. It is based on the total column measurements over clean, remote regions as well as over clouded scenes where the tropospheric column is effectively shielded. Weighting factors are defined to determine the influence of individual satellite measurements on the stratospheric estimate. STREAM is a flexible and robust algorithm and does not require input from chemical transport models. While originally developed as TROPOMI verification algorithm and optimized for OMI, it was successfully applied to GOME-2 as well.

Comparisons of GOME-2 tropospheric columns from STREAM and the operational product from GDP 4.7 generally reveal very similar patterns, but GDP 4.7 is affected by a systematic low bias of tropospheric columns. STREAM significantly improves these biases and results in more realistic tropospheric columns (higher medians and fewer negative results). It is thus recommended to implement STREAM in a GDP update.

2 Introduction

Nitrogen oxides (NO_x=NO+NO_2) play a key role in atmospheric chemistry, both in the stratosphere and the troposphere. The highly structured absorption bands of NO_2 in the blue spectral range make it a prime example among the trace gases retrieved by UV/visible satellite instruments such as GOME, SCIAMACHY, OMI, and the GOME-2 series.

Since the spectroscopic analysis yields total column densities of NO_2, the retrieval of tropospheric column densities requires the quantification and subtraction of the stratospheric fraction ("Stratosphere-Troposphere-Separation", STS). STS can principally be done based on coincident, independent measurements (as available for SCIAMACHY in limb geometry, see Beirle et al. (2010)) or based on Chemical Transfer Models (CTMs) (as done via data assimilation within TEMIS algorithms, e.g. Boersma et al. (2011)).

One of the first STS algorithms, however, was the reference sector method (RSM), which estimates the global stratospheric NO_2 fields from the nadir measurements themselves over a clean reference region, e.g. the remote Pacific (Richter and Burrows, 2002; Martin et al., 2002; Beirle et al., 2003). The simple RSM is based on the assumptions of a) longitudinal homogeneity of stratospheric NO_2, and b) negligible tropospheric contribution over the reference region. This procedure is quite simple, transparent, and robust. The RSM was successfully applied by different groups to different satellite instruments and generally performs well. However, the resulting tropospheric NO_2 are affected by systematic biases caused by the simplifying assumptions:

a) The assumption of longitudinal homogeneity is often justified, at least in temporal means when small scale stratospheric dynamical features cancel out. But in particular close to the polar vortex, high longitudinal variations can occur, as already discussed by Richter and Burrows (2002) and Martin et al. (2002). Thus, tropospheric columns derived by RSM can be off by more than 10^{15} molec cm^{-2} in winter at latitudes from 50° polewards, thereby affecting scientific interpretations of tropospheric column densities over North America or Northern Europe.
In order to reduce the artefacts caused by assumption (a), several modifications of the RSM have been proposed in recent years, which generally allow for zonal variations of the stratospheric estimate, while the basic approach (using nadir measurements over clean regions for STS) has been retained (e.g., Leue et al., 2001; Wenig et al., 2004; Bucsela et al., 2006; Valks et al., 2011; Bucsela et al., 2013). We refer to this group of STS algorithms as "modified RSM" (MRSM).

b) The tropospheric background column in the Pacific (or any other "clean" reference region) is very low (compared to columns over regions exposed to significant NO_x sources), but not 0. Some algorithms explicitly correct for the tropospheric background: Valks et al. (2011) assume a constant background of 0.1×10^{15} molec cm^{-2}. Bucsela et al. (2013) involve tropospheric concentration profiles from a model climatology.

MRSMs typically apply a rather conservative masking approach for potentially polluted pixels. Continents are masked out almost completely. Especially at Northern mid-latitudes, the masked area can be larger than the area used for the stratospheric estimation, and over the Eurasian continent, the STS algorithm misses any supporting measurement points over about ten thousand km. This can lead to significant errors during interpolation.

Within GDP 4.7 (and the upcoming GDP 4.8 algorithm), STS is realized by a MRSM as described in Valks et al. (2011, 2013). The existing operational GOME-2 tropospheric NO_2 product is generally of good quality and is used by the Copernicus atmospheric core service MACC. It has been successfully used within scientific studies on e.g. temporal (weekly and seasonal cycles, trends) and spatial patterns (like ship tracks) as well as emission estimates of NO_x. However, the remaining uncertainties due to the stratospheric estimation are one of the main error sources in the tropospheric NO_2 column retrieval and potentially result in systematic regional biases. Particular challenging are northern mid-latitudes in winter/spring, when the polar vortex causes strong spatial gradients in stratospheric NO_2, with large impact on the retrieved tropospheric columns over e.g. Europe (Valks et al., 2011).

In this AS activity, the STRatospheric Estimation Algorithm from Mainz (STREAM), originally developed as verification algorithm for the upcoming TROPOMI instrument, was adopted and applied to GOME-2 with the aim to implement it in a GDP update for the O3MSAF at DLR Oberpfaffenhofen.

STREAM is a MRSM as well, requiring no further model input, and can relatively easy be implemented also for the NRT data processor. It differs from the current GDP 4.7/4.8 implementation essentially in three aspects:
- In STREAM, there is no strict discrimination of “clean” versus “polluted” satellite pixels or regions. Instead, weighting factors are defined for each satellite pixel determining how far it influences the stratospheric estimate.
- In addition to “clean” regions, also satellite measurements over mid-altitude clouds, for which the tropospheric column can be considered to be effectively shielded, are used for the stratospheric estimate (by assigning them with a high weighting factor). In this respect, STREAM is similar to the MRSM of the operational NASA OMI product (Bucsela et al., 2013).
- After the initial stratospheric estimate, an additional iteration with modified weights is done, where pixels with initially negative tropospheric residues (i.e., high-biased stratospheric columns) are assigned with a higher weight, whereas pixels with high positive tropospheric residue (indicating tropospheric pollution) are weighted down.

3 STREAM

The STRatospheric Estimation Algorithm from Mainz (STREAM) stands in tradition of modified RSM, i.e. the stratospheric field is estimated directly from satellite measurements for which the tropospheric contribution can be considered to be negligible. For this purpose, measurements over remote regions without tropospheric sources are used. In addition, also cloudy measurements are considered where the tropospheric column is shielded.

Below we summarize the main STREAM (v0.92) procedure and settings. Further details can be found in Beirle et al. (2015).

STREAM consists basically of two steps:
1. In contrast to other MRSMs, no strict pollution mask is applied. Instead, weighting factors are calculated for each satellite pixel, determining how far the measured NO_2 total columns are contributing to the estimated stratospheric field (Sect. 3.2).
2. Global maps of stratospheric NO_2 are determined by applying weighted convolution (Sect. 3.3).

Before describing the STREAM algorithm, we define the investigated quantities and abbreviations used hereafter in the next section.

3.1 Terminology

With Differential Optical Absorption Spectroscopy (DOAS), so-called slant column densities (SCDs) S, i.e. concentrations integrated along the mean light path, of NO_2 are derived. SCDs are converted into VCDs (vertical column densities, i.e. vertically integrated concentrations) V via the air-mass factor (AMF) A: $V = S/A$. The AMF depends on radiative transfer (determined by viewing geometry, clouds, aerosols) and the trace gas profile. For the stratospheric column, it is basically given by viewing geometry.

Input to STREAM are total vertical column densities V^* of NO_2, which are derived from the total SCDs divided by the respective stratospheric AMFs, which basically removes the dependency on viewing angles. Over clean regions with negligible tropospheric columns, V^* is dominated by the actual total VCD. In case of tropospheric pollution, V^* underestimates the total VCD, as the AMF is in most cases smaller in the troposphere than in the stratosphere. These situations have to be excluded in the stratospheric estimate.

STREAM yields an estimate for the stratospheric VCD V_{strat} based on the assumption that V^* can be considered as proxy for V_{strat} in "clean" regions and over cloudy measurements. We define the tropospheric residue (TR) T^* as

$$T^* = V^* - V_{\mathrm{strat}}, \tag{1}$$

i.e. as the difference of total and stratospheric VCDs based on a stratospheric AMF.

Tropospheric VCDs (TVCDs), which are the final product of NO_2 retrievals used for further tropospheric research, are connected to T^* via

$$V_{\text{trop}} = T^* \times \frac{A_{\text{strat}}}{A_{\text{trop}}}. \tag{2}$$

For cloud-free satellite pixels, the ratio $\frac{A_{\text{strat}}}{A_{\text{trop}}}$ typically ranges from about 1 above clean oceans at low and mid-latitudes to $\approx$ 2-3 above moderately polluted regions, and up to >4 at high latitudes in case of low A_{trop}, when NO_2 profiles peak close to the ground.

Below we focus on the tropospheric residue T^* instead of the tropospheric VCD V_{trop}, as biases in the stratospheric estimation can directly be related (factor -1) to the respective biases in T^*, and the matter of tropospheric AMFs is beyond the scope this study.

3.2 Definition of weighting factors

STREAM estimates the stratospheric column density of NO_2 based on nadir measurements for which the tropospheric column can be considered to be negligible, either because it is low or shielded by clouds. But in contrast to other MRSMs, we do not flag the pixels as either clean or (potentially) polluted. Instead, weighting factors for individual satellite pixels determine how strongly they are considered in the stratospheric estimation. Satellite measurements which are expected to be free from tropospheric contribution get a high weight accordingly. In this section, we define the different weighting factors we apply.

3.2.1 Pollution weight

In order to estimate the stratospheric NO_2 field from total column density measurements, at first only "clean" measurements where the tropospheric column can be considered to be negligible, are considered. In cases of very high total column densities (V^*>10 $\times 10^{15}$ molec/cm^2) which clearly exceed the domain of stratospheric column densities, a tropospheric contribution is obvious, and these measurements are excluded by assigning them a weighting factor of 0.

In most cases, however, the tropospheric contribution to the total column is not that easy to determine. We thus define a pollution weight w_{pol} based on our a-priori knowledge about the spatial distribution of tropospheric NO_2, reflecting a kind of tropospheric pollution probability. Such information can be gained from long-term means of satellite measurements. Here, we use the mean tropospheric NO_2 column as derived from SCIAMACHY (Beirle and Wagner, 2012) as basis for the compilation of a "pollution proxy" P. Details on the definition of P are given in Beirle et al. (2015). The pollution weight is then defined as

$$w_{\text{pol}} = 0.1/P^3 \tag{3}$$

I.e., the higher the pollution proxy, the lower the weighting factor and the less the measurement is contributing to the stratospheric estimate. Eq. 3 is displayed in Fig. 1(a).

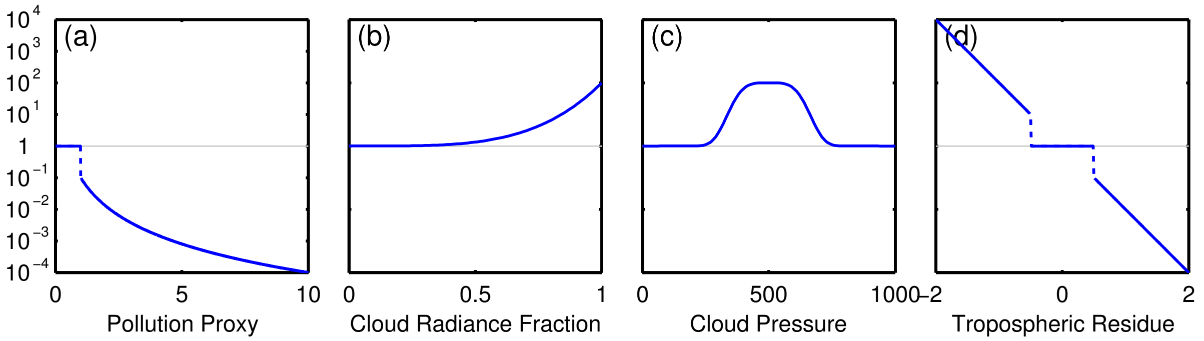

Figure 1: Definition of weighting factors (a) w_{pol} as function of the pollution proxy P (eq. 3), (b) w_{cld} as function of the cloud radiance fraction (eq. 4) for a cloud pressure of 500 hPa, (c) w_{cld} as function of the cloud pressure (eq. 4) for a cloud radiance fraction of 1, and (d) w_{TR} as function of the tropospheric residue (eq. 5).

3.2.2 Cloud weight

In addition to measurements over remote regions free of tropospheric sources, also clouded satellite measurements, where the tropospheric column is shielded, provide a good proxy for the stratospheric column density. Thus, the weighting factor w_{cld} is used to increase the weight of clouded satellite pixels. This is achieved by the following definition:

$$
\begin{aligned}
w_{\mathrm{cld}} &:= 10^{2\times w_{\mathrm{C}}\times w_{\mathrm{P}}} && \text{(a)}\\
\text{with} & \\
w_{\mathrm{C}} &:= C^4 && \text{(b)}\\
\text{and} & \\
w_{\mathrm{P}} &:= e^{\left(-\frac{1}{2}\left(\frac{p-p_{\mathrm{ref}}}{\varsigma_p}\right)^4\right)} && \text{(c)}
\end{aligned}
\qquad (4)
$$

w_{cld} (a) is composed of the terms w_{C} (b) and w_{P} (c).

w_{C} reflects the dependency on the cloud radiance fraction C. Due to the exponent of 4, only pixels with large cloud radiance fraction reach a high weighting factor and contribute strongly to the stratospheric estimation.

w_{P} describes the dependency on cloud pressure P. It is basically a modified Gaussian (with exponent 4 instead of 2, making it flat-topped) centered at $p_{\mathrm{ref}} = 500$ hPa with the width $\varsigma = 150$ hPa. I.e., only cloudy measurements at medium altitudes are assigned with a high weighting factor, while high clouds (potentially contaminated by lightning NO_x) as well as low clouds (where tropospheric pollution might still be visible) are excluded.

As both w_{C} and w_{P} yield values in the range from 0 to 1, the factor of 2 in the exponent of eq. 4(a) sets the maximum value of w_{cld} to 10^2. Eq. 4 is displayed in Fig. 1 (b) and (c).

3.2.3 Tropospheric residue weight

STREAM yields global fields of stratospheric VCDs V_{strat}, explained in detail below (sect. 3.3), which allow to calculate tropospheric residues T^* according to eq. 1. While the "true" tropospheric fields are not known, the resulting T^* can still be used in order to evaluate and improve the stratospheric estimation in a second iteration:
1. as negative column densities are non-physical, $T^*<0$ clearly indicates that the stratospheric field has been overestimated. Consequently, the affected satellite measurements should be assigned with a higher weighting factor such that they contribute stronger to the stratospheric estimate.
2. a high T^* indicates tropospheric pollution. Thus, the respective weights should be decreased.

Thus, we define a weighting factor w_{TR} as

$$w_{\text{TR}} := \begin{cases} 10^{-2\times T^*} & \text{if } |T^*| > 0.5 \times 10^{15} \text{ molec/cm}^2 \\ 1, & \text{else} \end{cases} \tag{5}$$

As T^* is defined as the difference of V^* and V_{strat} (eq. 1), i.e. two quantities of the same order of magnitude with non-negligible errors, the resulting statistical distribution of T^* inevitably includes negative values. These negative values caused by statistical fluctuations are required in the probability density function (and should not be excluded) in order to keep the mean unbiased. Thus, w_{TR} should be only applied to significant and systematic deviations of T^* from 0. This is achieved by the following:

1. in contrast to w_{cld}, which is defined for each individual satellite measurement, w_{TR} is defined based on the TRs averaged over $1^\circ\times1^\circ$ grid pixels. I.e., first the values of T^* within one grid pixel are averaged, reducing statistical noise, before eq. 5 is applied, and the resulting weight is then assigned to all satellite measurements within the grid pixel.

2. w_{TR} is only applied if the absolute value of the mean grid box T^* exceeds a threshold of 0.5×10^{15} molec/cm^2 (eq. 5).

3. w_{TR} is only applied, if a larger area is affected by systematic low or high TR, i.e. if the adjacent grid pixels exceed the threshold as well. I.e., a single outlier will not trigger w_{TR}.

w_{TR} could in principle be tuned in multiple iterations. In STREAM v0.92, one iteration is performed.

Eq. 5 is displayed in Fig. 1(d).

3.2.4 Total weight

The total weight of each satellite pixel is defined as the product of the individual weighting factors:

$$w_{\text{tot}} := w_{\text{pol}} \times w_{\text{cld}} \times w_{\text{TR}} \tag{6}$$

The concept of the combination of different weighting factors is easily extendible by further weights based on fire or flash counts in order to account for, e.g., irregular NO_x sources such as biomass burning or lightning.

3.3 Weighted convolution

Global daily maps of the stratospheric column density are derived by applying "weighted convolution", i.e. a spatial convolution which takes the individual weights for each satellite pixel into account. This approach is an extension of the "normalized convolution" presented in (Knutsson and Westin, 1993). By this weighted convolution, the stratospheric field is smoothed and interpolated at the same time. A similar approach was used by Leue et al. (2001), but only using the fitting errors of NO_2 SCDs as weights.

The algorithm is implemented as follows:

- A lat/lon grid is defined with 1° resolution. Each satellite pixel is sorted into the matching grid pixel according to its center coordinates. At the j^{th} latitudinal/i^{th} longitudinal grid position, there are K OMI pixels with the total columns $V_{ijk}(k = 1..K)$ and the weights w_{ijk}. We define

$$C_{ij} := \sum w_{ijk} \times V_{ijk} \tag{7}$$

  and

$$W_{ij} := \sum w_{ijk} \tag{8}$$

  In case of measurement gaps (i.e. $K = 0$), both C_{ij} and W_{ij} are set to 0.

  The weighted mean VCD for each grid pixel is then given as

$$V_{ij} = \frac{C_{ij}}{W_{ij}} \tag{9}$$

- A convolution Kernel G is defined (e.g. a 2D Gaussian). Spatial convolution is applied to both C and W (taking the dateline into account appropriately, i.e. i=1 and i=360 are adjacent grid pixels):

$$\overline{C} := G \otimes C \tag{10}$$

$$\overline{W} := G \otimes W \tag{11}$$

- The smoothed stratospheric VCD for each grid pixel as derived from weighted convolution is then given as

$$\overline{V}_{ij} := \frac{\overline{C}_{ij}}{\overline{W}_{ij}} \tag{12}$$

The degree of smoothing is determined by the definition of the convolution Kernel G. Generally, information on the stratospheric column over polluted regions should be taken from clean measurements at the same latitude. Thus, σ_{lon} has to be sufficiently large, while σ_{lat} has to be low as gradients in latitudinal dimension should be mostly conserved. For high latitudes, however, the longitudinal extent of the Kernel has to be small enough as well in order to be able to resolve the strong gradients caused by the polar vortex.

In order to meet these requirements, we implement the convolution in the following way:

- Two convolutions are performed, based on a large ($\sigma_{\text{lon}} = 50°$, $\sigma_{\text{lat}} = 10°$) and a small ($\sigma_{\text{lon}} = 10°$, $\sigma_{\text{lat}} = 5°$) Kernel, yielding two estimates for V_{strat}, i.e. $V_{\text{strat}}^{\text{pol}}$ and $V_{\text{strat}}^{\text{eq}}$.
- The final stratospheric VCD is defined as the weighted mean of both depending on latitude ϑ:

$$V_{\text{strat}} := \cos^2(\vartheta) V_{\text{strat}}^{\text{eq}} + \sin^2(\vartheta) V_{\text{strat}}^{\text{pol}} \tag{13}$$

By this method, spatial smoothing is wide enough at the equator (needed to interpolate e.g. the stratosphere over Central Africa), but small enough at the polar vortex.

In latitudinal direction, this procedure can cause small, but systematic biases if stratospheric NO_2 show significant latitudinal gradients on scales of σ_{lat} or smaller. To overcome this, STREAM provides the (default) option to run the weighted convolution on "latitude-corrected" VCDs. I.e., the mean dependency of V^* on latitude is determined (again over the Pacific), subtracted from all individual V_{ijk}, and added back again after the weighted convolution. By this procedure, latitudinal gradients are largely removed for the convolution (but not from the final stratospheric fields), and the systematic biases vanish.

3.4 Data processing

STREAM estimates stratospheric fields and tropospheric residues for individual orbits. For each orbit under investigation, the orbit itself plus the 7 previous and subsequent orbits are used for the calculation of V^*, weighting factors, and thus V_{strat} via weighted convolution. For the daily means presented in this study, all orbits where the orbit start date matches the day of interest are averaged.

Alternatively, STREAM can be operated in Near-Real time (NRT) mode, in which 14 previous (as subsequent orbits are not available) are included in the weighted convolution.

3.5 Performance

The performance of STREAM is investigated in depth in Beirle et al. (2015). Here we summarize the main findings:

STREAM was successfully applied to satellite measurements from GOME 1/2, SCIAMACHY, and OMI. The resulting TR over clean regions and their variability have been found to be low. However, systematic "stripes" can still appear in STREAM TR if the basic assumption that the stratospheric column varies smoothly with longitude is not given, e.g. in case of “tilted” stratospheric patterns.

STREAM results are robust with respect to variations of the algorithm settings and parameters. With the baseline settings, the errors of STREAM on a synthetic (model-based) dataset have been found to be below 0.1 $\times 10^{15}$ molec/cm^2 on average.

The emphasis of clouded observations, which provide a direct measurement of the stratospheric rather than the total column, should supersede an additional correction for the tropospheric background, which successfully worked for OMI, and, to some extent (for reasons not yet fully understood), also for SCIAMACHY and GOME-2.

STREAM was initially tested for OMI measurements and compared to the DOMINO v2 product (Boersma et al., 2011), in which STS is implemented by data assimilation. The deviation of monthly mean TR is generally low (0.1-0.2 $\times 10^{15}$ molec/cm^2). Comparison to other state-of-the-art STS schemes (including various approaches, e.g. a different MRSM for OMI (Bucsela et al., 2013) or limb-nadir-matching for SCIAMACHY (Beirle et al., 2010)) yield deviations of similar order.

The uncertainty of STS is thus generally negligible for TVCDs over polluted regions. But the remaining systematic regional patterns still contribute significantly to the uncertainty of TVCDs over “semi-polluted” regions and have to be kept in mind for emission estimates of area sources of NO_x such as soil emissions or biomass burning.

4 Application of STREAM for GOME-2 and comparison to GDP 4.7

STREAM has been applied to GOME-2 data (based on the total SCDs, stratospheric AMFs, and cloud products provided in GDP 4.7). Below, we show results for January and July 2010 exemplarily, discuss the performance of STREAM, and quantify the differences in stratospheric estimates and tropospheric residues with respect to the current GDP 4.7 (Valks et al., 2011, 2013).

Figure 2 displays the total VCD (a) and the stratospheric estimates from STREAM (b), STREAM NRT (c), and GDP 4.7 (d) for 1st of January 2010. In figure 3, the tropospheric residues, i.e. the total minus stratospheric VCD, are shown for the same day based on a simple RSM (a), STREAM (b), STREAM NRT (c), and GDP 4.7 (d). Figures 4 and 5 show the respective results for 1st of July 2010, where GOME-2 was operated in narrow swath mode, causing poor global coverage. This however does not affect STS performance of the investigated algorithms. Figures 6 and 7 display monthly mean TR for the respective algorithms, again for January and July 2010. Fig. 9 summarizes the statistical distribution of TR from different STS algorithms for selected regions. Shown are the respective medians (white dashes) and the 10th-90th (light) and 25th-75th (dark) percentiles, both for daily (narrow bars) and monthly (wide bars) means.

The TR from a simple RSM reveals a large spread over remote regions (figure 3(a)), in particular at mid and high latitudes, resulting from the simplifying assumption of zonal invariance of the stratospheric column. In the monthly means (Figs. 6(a) and 7(a)), these artefacts are reduced at midlatitudes, where fluctuations of daily stratospheric patterns cancel at large part out, while at high latitudes, significant systematic artefacts still remain, in particular in the Northern hemispheric winter, due to the asymmetry of the polar vortex.

These shortcomings of the simple RSM are largely reduced by both STREAM and GDP 4.7, which both allow for zonal variability of the stratospheric fields. The improvement is clearly visible in both daily (Figs. 3 and 3(b)-(d)) as well as monthly (Figs. 6 and 7(b)-(d)) means.

Over the Pacific, TR from RSM is on average 0 by construction. For STREAM, TR is found to be about 0.05 $\times 10^{15}$ molec/cm^2 on average, resulting from the emphasis of clouded pixels, which provide direct measurements of the stratospheric column.

On single days, the resulting TR for STREAM standard and NRT mode are slightly different, as different orbits contribute to the daily stratospheric estimate. In the monthly means, however, TR are only marginally different between standard and NRT mode. I.e., the observed daily differences are predominantly of statistical nature and cancel out in the monthly mean.

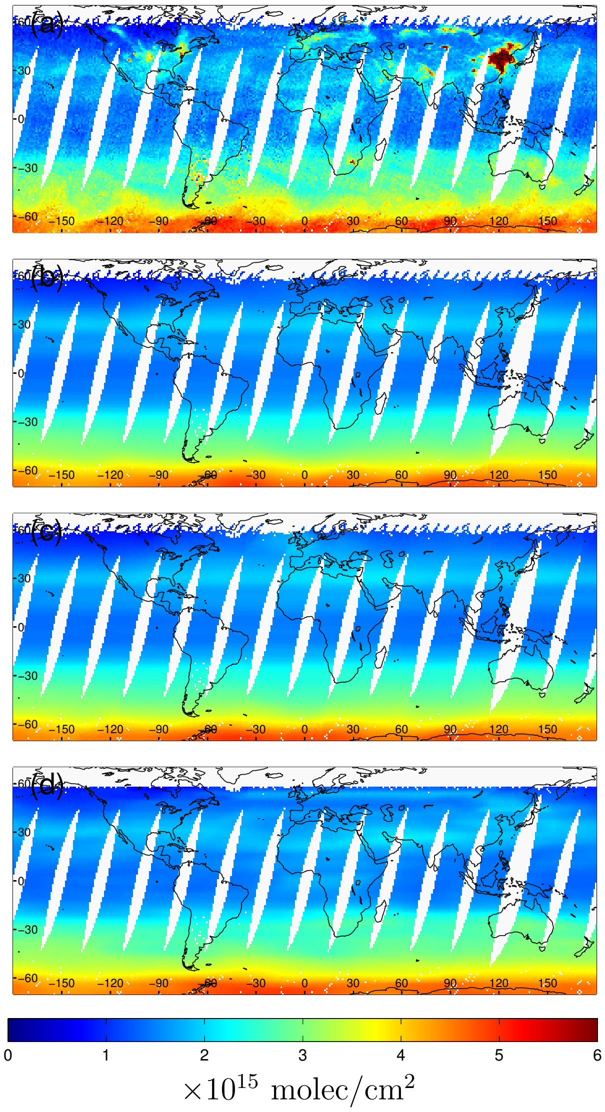

Figure 2: (a) Total NO_2 VCD (based on a stratospheric AMF) for 1 January 2010. (b)-(d) Estimates of the stratospheric VCD resulting from STREAM (b), STREAM NRT (c), and GDP 4.7 (d).

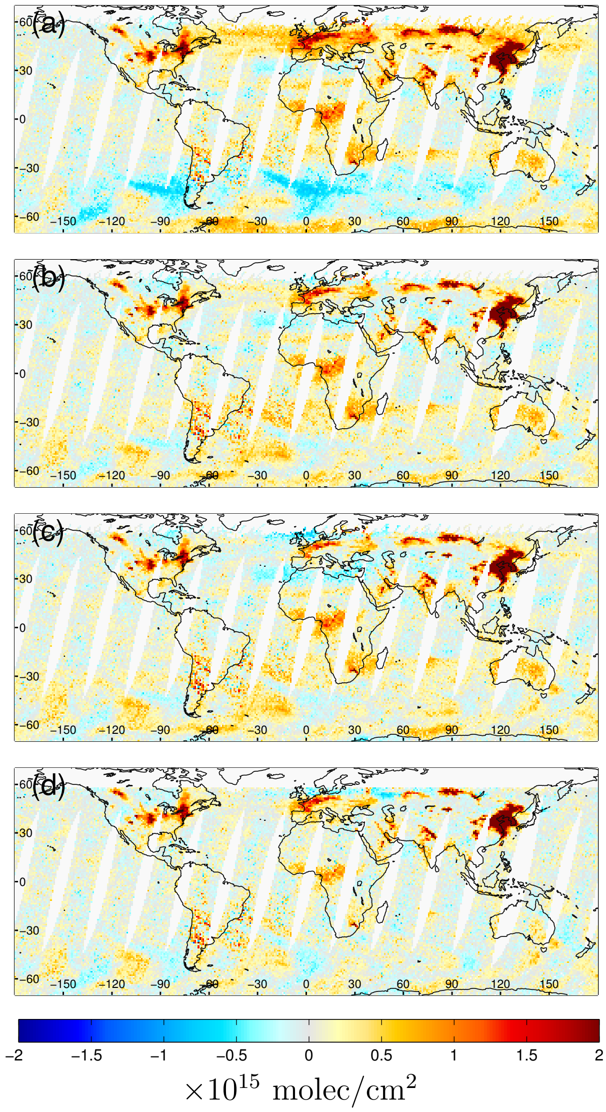

Figure 3: Tropospheric residues for 1 January 2010 resulting from RSM (a), STREAM (b), STREAM NRT (c), and GDP 4.7 (d).

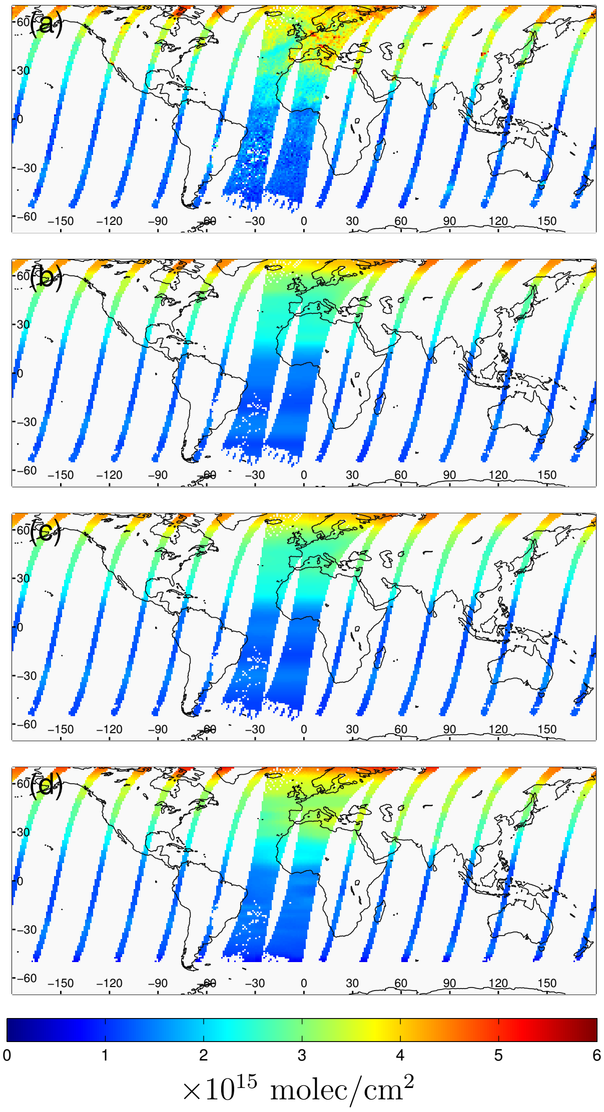

Figure 4: (a) Total NO_2 VCD (based on a stratospheric AMF) for 1 July 2010. (b)-(d) Estimates of the stratospheric VCD resulting from STREAM (b), STREAM NRT (c), and GDP 4.7(d).

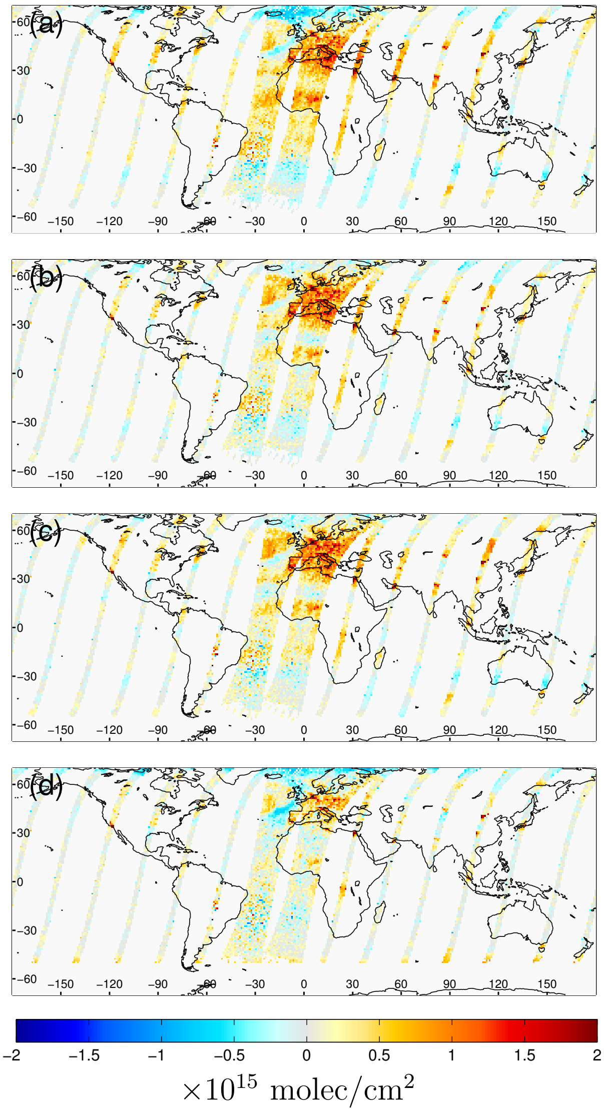

Figure 5: Tropospheric residues for 1 July 2010 resulting from RSM (a), STREAM (b), STREAM NRT (c), and GDP 4.7 (d).

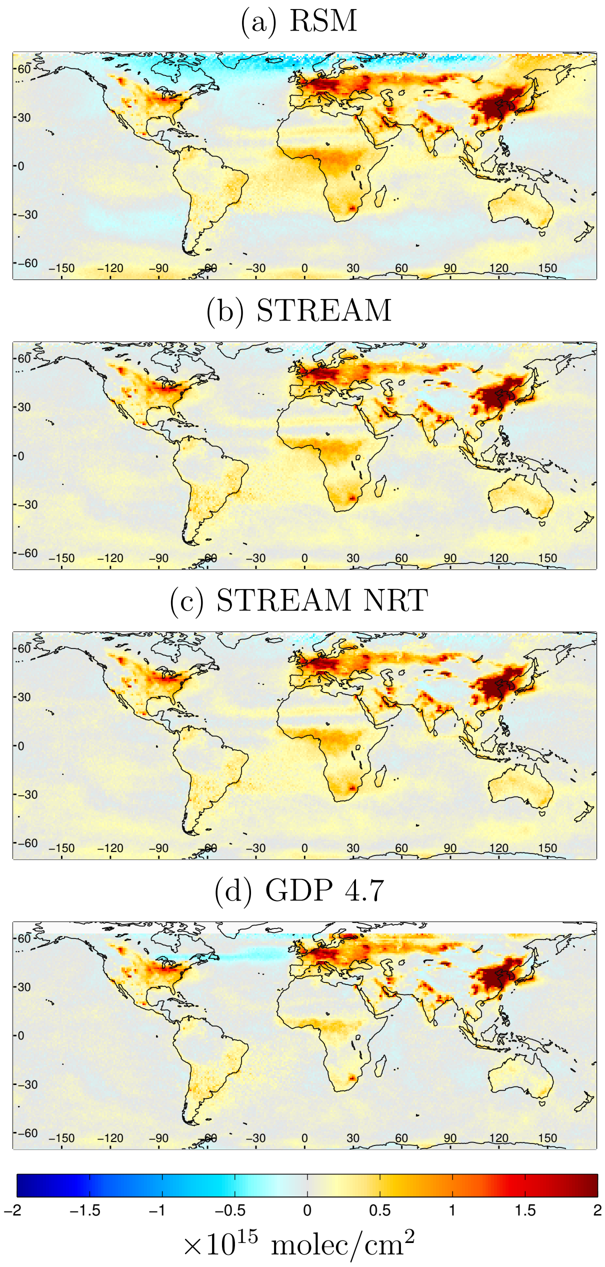

Figure 6: Monthly mean tropospheric residues for January 2010 resulting from RSM (a), STREAM (b), STREAM NRT (c), and GDP 4.7 (d).

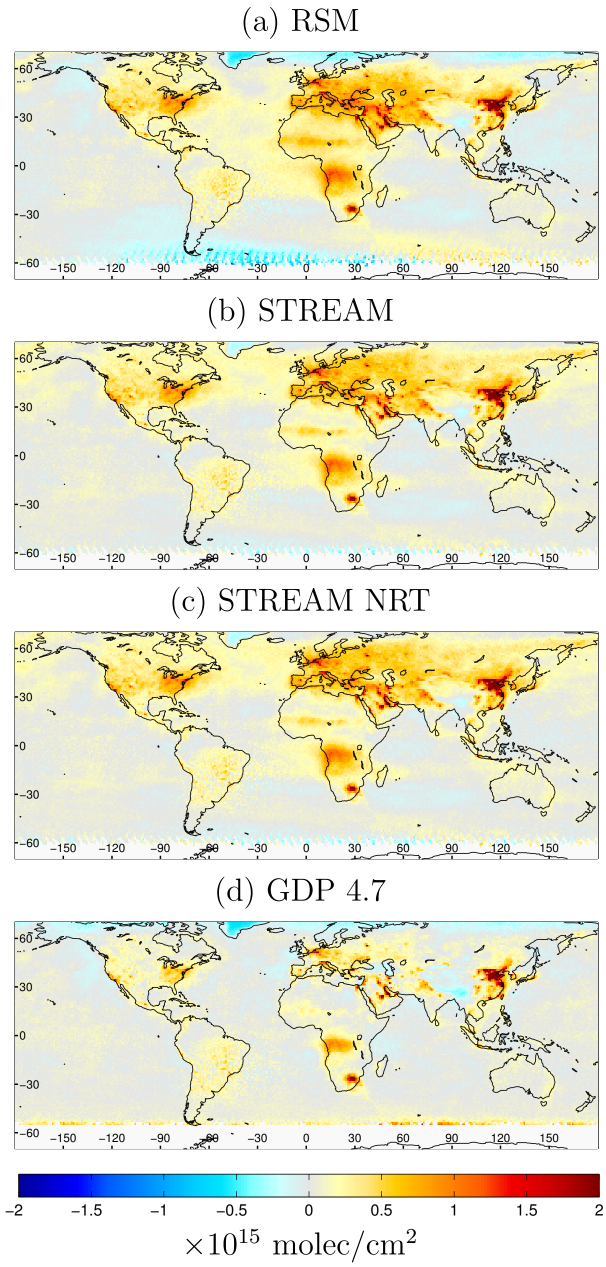

Figure 7: Monthly mean tropospheric residues for July 2010 resulting from RSM (a), STREAM (b), STREAM NRT (c), and GDP 4.7 (d).

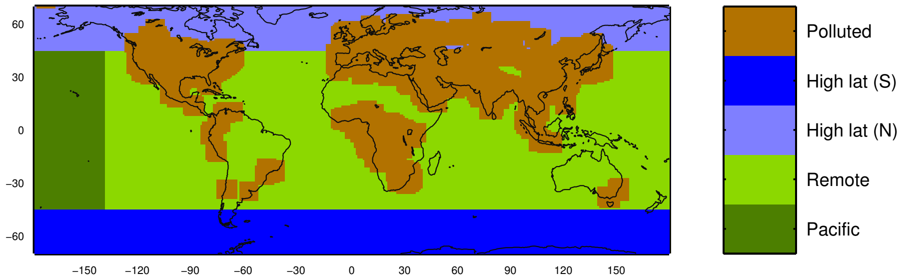

Figure 8: Regions of interest for the calculation of regional statistics of T^*.

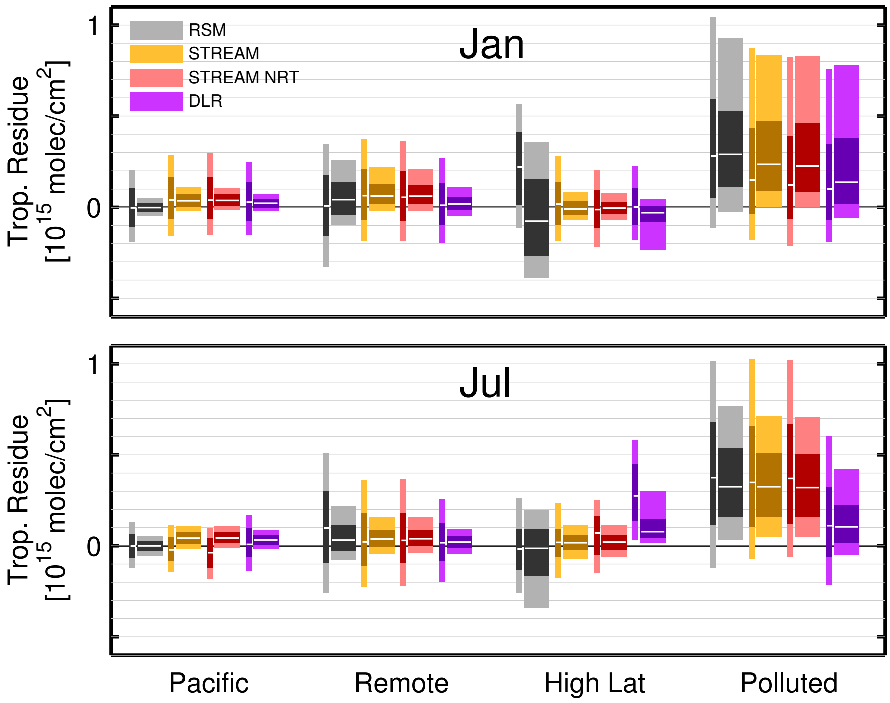

Figure 9: Regional statistics of GOME-2 tropospheric residues T^* from different algorithms for January (top) and July (bottom) 2010. Light and dark bars reflect the 10-90 and 25-75 percentiles, respectively. The median is indicated in white. Narrow bars show the statistics for the first day of the month, wide bars those of the monthly means. Regions are explained in Fig. 8. Note that "High Lat" refers to the hemispheric winter, i.e. Northern latitudes in January, but Southern latitudes in July.

In GDP 4.7, STS for NO_2 is done by a MRSM as well as described in Valks et al. (2011, 2013). Basically, polluted regions (defined by monthly mean TVCDs from the MOZART-2 model being larger than 1 $\times 10^{15}$ molec/cm^2) are masked out. Global stratospheric fields are then derived by low pass filtering in zonal direction by a 30° boxcar filter.

Overall, TRs from GDP 4.7 result in very similar patterns as STREAM. Total TRs, however, seem to be generally biased low. Over the Pacific, mean T^* is close to 0 in January, despite the applied tropospheric background correction of 0.1 $\times 10^{15}$ molec/cm^2. Over polluted regions, median TR from GDP 4.7 is systematically lower (by 0.2 $\times 10^{15}$ molec/cm^2 in July) than from STREAM, and almost 25% of all grid pixels even have TR$<$0.

Figure 10 displays the differences of the monthly mean TR from GDP 4.7 and STREAM-for January and July 2010, again pointing out the systematically lower values of GDP 4.7 TR, especially over continents, in July.

The systematic low bias of TR from GDP 4.7 probably results from moderately polluted pixels over regions labelled as "unpolluted", which still can reach TVCDs up to 1 $\times 10^{15}$ molec/cm^2 in MOZART. These measurements cause a high bias of the estimated stratospheric field around polluted regions; by the subsequent spatial low-pass filtering, this high bias is passed over to the (masked) polluted regions and results in low-biased TR. Further investigations are needed to find out why this effect is stronger in July than in January.

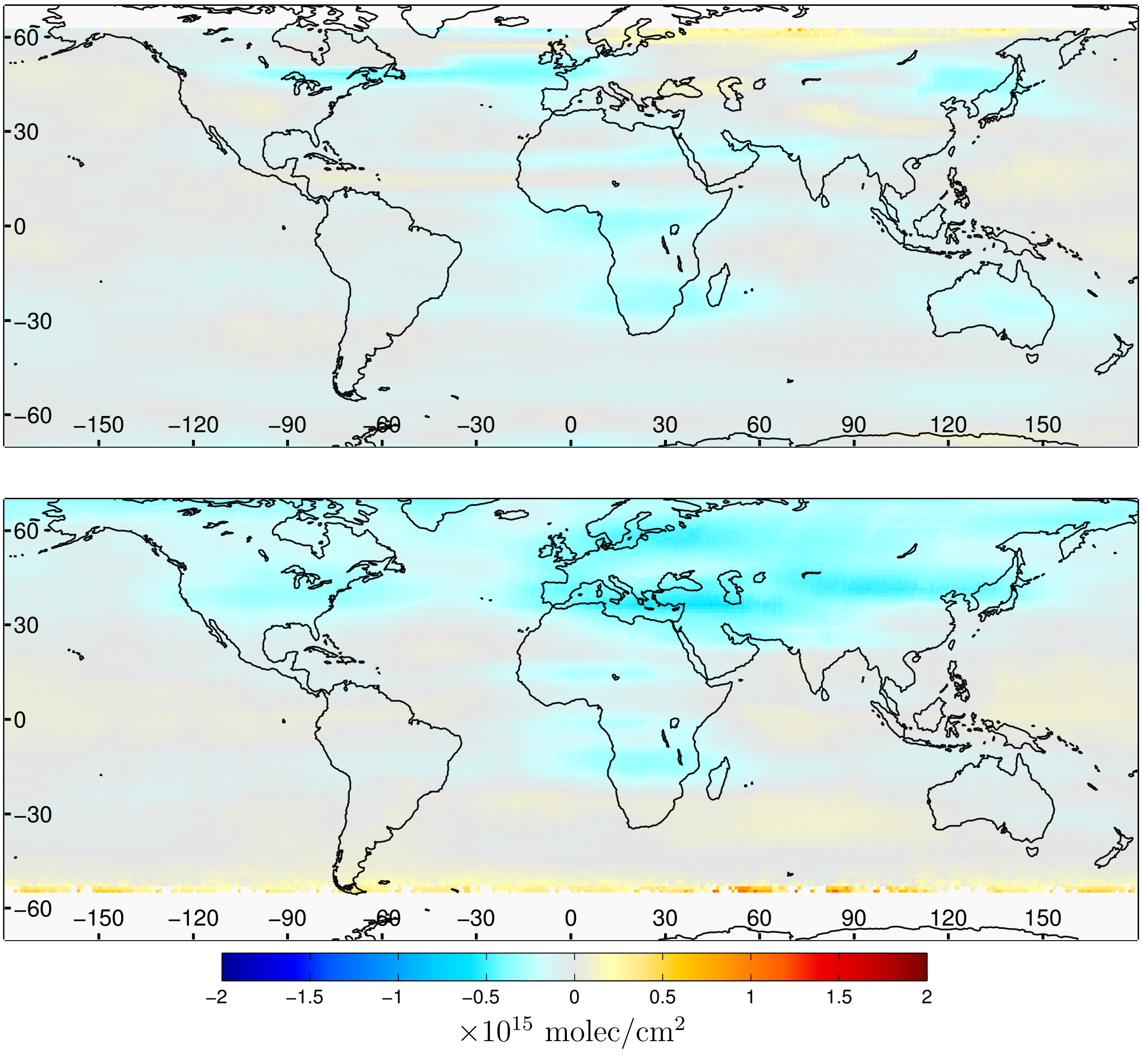

Figure 10: Monthly mean difference of tropospheric residues T^* from DLR and STREAM for GOME-2 measurements in January (top) and July (bottom) 2010.

4.1 Solar Eclipse

On 15 January 2010, STREAM resulted in extraordinary high variability of TR over the Indian ocean (Fig. 11), related to very low total VCDs (even <0) for one orbit. These artefacts turned out to be caused by the spectral analysis being deficient due to the low radiances during a solar eclipse on that day (Espenak and Anderson , 2008).

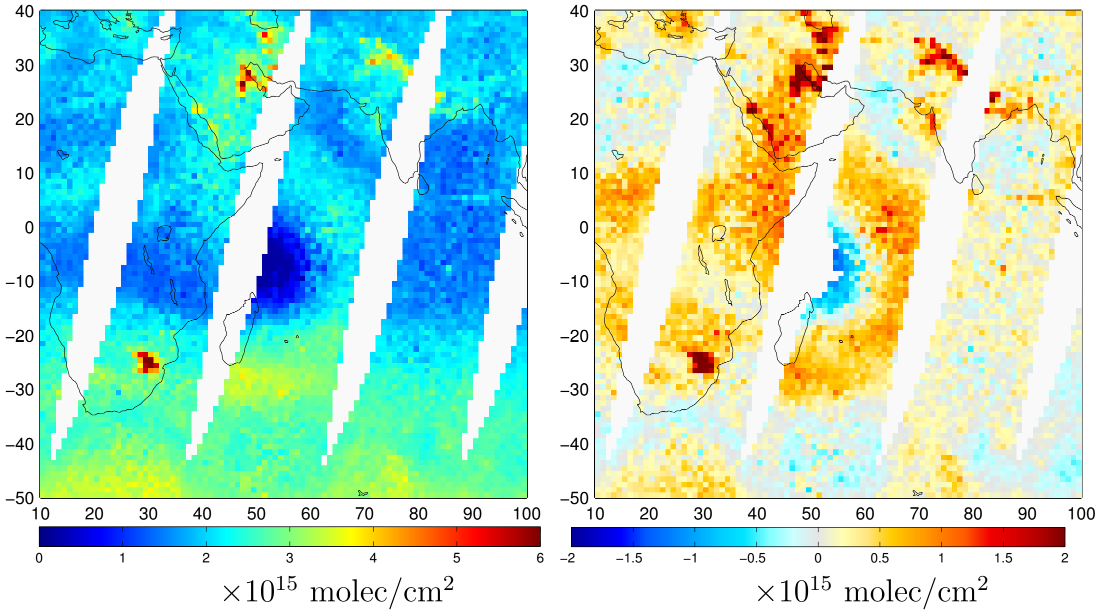

Figure 11: GOME-2 total VCD (left) and tropospheric residues T^{*} from STREAM (right) on 15 January 2010. Negative VCDs are observed East from Africa caused by a solar eclipse. Thus, TR show large artificial patterns.

Removing the affected orbit results in normal performance of STREAM for this day. We thus recommend that a screening of solar eclipses is done automatically (as done for e.g. OMI) before the stratospheric correction is performed.

5 Conclusions & recommendations

The STRatospheric Estimation Algorithm from Mainz (STREAM), developed for TROPOMI verification, was successfully applied to STS for GOME-2. It can be operated both in normal (offline) and NRT mode with similar performance (for monthly means).

Stratospheric columns are estimated based on satellite observations over remote, clean regions, and over mid-altitude clouds. The latter provide additional supporting points in the global stratospheric field over weakly polluted regions, thereby reducing potential interpolation errors. Furthermore, as these cloudy measurements directly provide the stratospheric rather than the total column, an additional correction of the tropospheric background is not required within STREAM.

Tropospheric columns resulting from STREAM have been compared to the GDP 4.7. While overall differences are low, tropospheric columns from GDP 4.7 generally reveal a low bias (e.g., almost 25% of all measurements over polluted regions are negative in July 2010). STREAM results in more realistic statistical distributions, i.e. higher medians and fewer negative results. It is thus recommended to implement STREAM in a future GDP update.

The STREAM algorithm and its potential implementation in the operational GDP has been discussed at an AS Meeting at DLR Oberpfaffenhofen on 28 May 2015. The MATLAB implementation of STREAM v0.92 for GOME-2 NRT analysis has been provided to DLR Oberpfaffenhofen on 27 October 2015.

References

Beirle, S., Platt, U., Wenig, M. and Wagner, T.: Weekly cycle of NO_2 by GOME measurements: a signature of anthropogenic sources, Atmos. Chem. Phys., 3(6), 22252232, 2003.

Beirle, S., Kühl, S., Pukite, J. and Wagner, T.: Retrieval of tropospheric column densities of NO_2 from combined SCIAMACHY nadir/limb measurements, Atmos. Meas. Tech., 3(1), 283-299, doi:10.5194/amt-3-283-2010, 2010.

Beirle, S. and T. Wagner: Tropospheric vertical column densities of NO_2 from SCIAMACHY, Available from: http://www.sciamachy.org/products/NO2/NO2tc_v1_0_MPI_AD.pdf, 2012.

Beirle, S. et al., The STRatospheric Estimation Algorithm from Mainz (STREAM), to be submitted to AMT.

Boersma, K. F., Eskes, H. J., Dirksen, R. J., van der A, R. J., Veefkind, J. P., Stammes, P., Huijnen, V., Kleipool, Q. L., Sneep, M., Claas, J., Leitao, J., Richter, A., Zhou, Y. and Brunner, D.: An improved tropospheric NO_2 column retrieval algorithm for the Ozone Monitoring Instrument, Atmospheric Measurement Techniques, 4, 1905-1928, doi:10.5194/amt-4-1905-2011, 2011.

Bucsela, E. J., Celarier, E. A., Wenig, M. O., Gleason, J. F., Veefkind, J. P., Boersma, K. F. and Brinksma, E. J.: Algorithm for NO2 vertical column retrieval from the ozone monitoring instrument, IEEE Transactions on Geoscience and Remote Sensing, 44(5), 12451258, doi:10.1109/TGRS.2005.863715, 2006.

Bucsela, E. J., Krotkov, N. A., Celarier, E. A., Lamsal, L. N., Swartz, W. H., Bhartia, P. K., Boersma, K. F., Veefkind, J. P., Gleason, J. F. and Pickering, K. E.: A new stratospheric and tropospheric NO_2 retrieval algorithm for nadir-viewing satellite instruments: applications to OMI, Atmospheric Measurement Techniques, 6(10), 2607-2626, doi:10.5194/amt-6-2607-2013, 2013.

Espenak, F., and Anderson, J., Annular and Total Solar Eclipses of 2010, NASA/TP2008214171, online available at http://eclipse.gsfc.nasa.gov/SEpubs/2010/TP214171a.pdf, 2008.

Knutsson, H. and Westin, C.-F.: Normalized and differential convolution, in Proceedings of Computer Society Conference on Computer Vision and Pattern Recognition (CVPR), IEEE, pp. 515-523., 1993.

Leue, C., Wenig, M., Wagner, T., Klimm, O., Platt, U. and Jhne, B.: Quantitative analysis of NO_x emissions from Global Ozone Monitoring Experiment satellite image sequences, J. Geophys. Res., 106(D6), 5493-5505, 2001.

Martin, R., Chance, K., Jacob, D., Kurosu, T., Spurr, R., Bucsela, E., Gleason, J., Palmer, P., Bey, I., Fiore, A., Li, Q., Yantosca, R. and Koelemeijer, R.: An improved retrieval of tropospheric nitrogen dioxide from GOME, JOURNAL OF GEOPHYSICAL RESEARCH-ATMOSPHERES, 107(D20), doi:10.1029/2001JD001027, 2002.

Richter, A. and Burrows, J. P.: Tropospheric NO_2 from GOME Measurements, Adv. Space Res., 29(11), 1673–1683, 2002.

Valks, P., Pinardi, G., Richter, A., Lambert, J.-C., Hao, N., Loyola, D., Van Roozendael, M. and Emmadi, S.: Operational total and tropospheric NO2 column retrieval for GOME-2, Atmospheric Measurement Techniques, 4, 14911514, doi:10.5194/amt-4-1491-2011, 2011.

Valks, P.,Loyola, D., Hao, N., Hedelt, P., Slijkhuis, S., and Grossi, M., Algorithm Theoretical Basis Document for GOME-2 Total Column Products of Ozone, Tropospheric Ozone, NO2, Tropospheric NO2, BrO, SO2, H2O, HCHO, OClO and Cloud Properties, DLR/GOME-2/ATBD/01, Issue 2/H, 2013

Wenig, M., Kühl, S., Beirle, S., Bucsela, E., Jähne, B., Platt, U., Gleason, J. and Wagner, T.: Retrieval and analysis of stratospheric NO2 from the Global Ozone Monitoring Experiment, J. Geophys. Res., 109(D4), D04315, 2004.