MONITORING NITROGEN OXIDES WITH SATELLITE INSTRUMENTS: HIGH RESOLUTION MAPS FROM GOME NARROW SWATH MODE AND SCIAMACHY

S. Beirle, U. Platt and T. Wagner

Institut für Umweltphysik (IUP), Im Neuenheimer Feld 229, 69120 Heidelberg, Germany
Email: beirle@iup.uni-heidelberg.de

ABSTRACT

The Global Ozone Monitoring Experiment (GOME) allows to retrieve tropospheric vertical column densities (TVCDs) of NO_2. Mean maps of the global distribution of NO_2 TVCDs reveal a clear fingerprint of anthropogenic sources due to fossil fuel combustion. However, GOME data suffer from the rather large footprint size of 320 km east-west, leading to a smearing out of the NO_2 peaks in the retrieved maps. A better spatial resolution is achieved by the GOME measurements in the "narrow swath mode" (NSM) (80*40 km^2) and especially by SCIAMACHY on ENVISAT (60*30 km^2). Here we present first mean composites of SCIAMACHY and compare them to GOME results. We demonstrate the benefit of the improved spatial resolution of GOME NSM and SCIAMACHY, i.e. a more precise knowledge of the global distribution of NO_2, allowing to localize and assign enhanced NO_2 levels directly to sources like cities, and to separate different source types. We furthermore analyze the effect of the pixel size on the retrieved TVCDs quantitatively.

1. INTRODUCTION

Nitrogen oxides ($NO+NO_2=NO_x$) are important trace gases in the troposphere with impact on human health and atmospheric chemistry, e.g. via their role in ozone production. Therefore, a better understanding of the various source strengths is desirable. The estimation of the strength of different NO_2 sources (mainly fossil fuel combustion, biomass burning, soil emissions and lightning) still exhibits high uncertainties up to one order of magnitude [1].

Satellite measurements enable a new and independent approach to the determination of trace gas emissions. The whole globe is observed with one instrument under the same conditions. Moreover, several types of measurements from different satellites can be made simultaneously (e.g. NO_2, HCHO, SO_2 column densities, fire counts and lightning activity).

Spectral data from satellite instruments (the Global Ozone Monitoring Experiment (GOME) [2] on ERS-2 and the SCanning Imaging Absorption SpectroMeter for Atmospheric CHartographY (SCIAMACHY) [3] on ENVISAT) is used to derive slant column densities of NO_2 using Differential Optical Absorption Spectroscopy (DOAS) [4]. The stratospheric fraction of the total column can be estimated in a reference sector over the pacific [5] and is subtracted from the total column to derive tropospheric slant column densities. Tropospheric vertical column densities (TVCDs) are retrieved by dividing those slant columns with a tropospheric air mass factor (AMF) [e.g. 5-8] to account for radiative transfer. Here we choose a simple approach with a place independent tropospheric AMF equal to 1 for zenith sun [5]. The advantage of this simple approach is that our results do not depend on any spatially resolved input data (like maps of ground albedo or aerosol distribution). Thus we can exclude artificial spatial patterns inherited from external data in our retrieved maps.

GOME data has successfully been used to identify and quantify different NO_x sources, i.e. anthropogenic [9, 10], biomass burning [11], lightning [12] and soil emissions [13]. However, the results from GOME observations suffer from the coarse spatial resolution of 320*40 km^2 (see Fig. 1, where the size of the standard GOME pixel is compared to the GOME NSM and SCIAMACHY pixels that are discussed below).

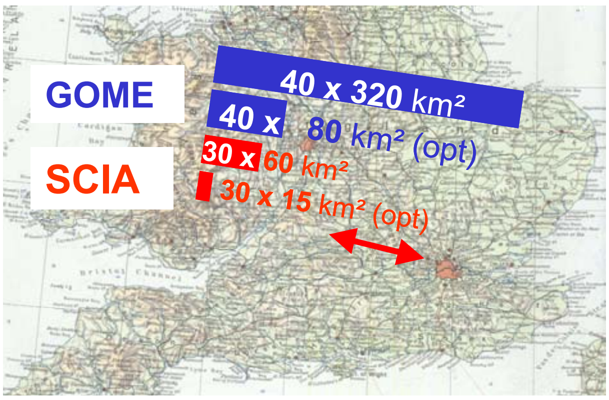

Fig. 1. Size and orientation of the different ground pixels of GOME and SCIAMACHY, displayed for England. The extent of the small SCIAMACHY pixels is comparabel to the size of large cities like London.

2. SCIAMACHY

SCIAMACHY was launched in March 2002 onboard the ESA satellite ENVISAT. In comparison to GOME, the wavelength range has been expanded to the IR, allowing the retrieval of greenhouse gases and CO. With respect to the NO_2 evaluation, SCIAMACHY has two major improvements compared to GOME: (a) new viewing geometries (limb, occultation) that allow direct measurements of the stratospheric columns and (b) much smaller nadir ground pixels (standard 60*30 km^2, up to 15*30 km^2) than those of GOME (see Fig. 1). So far we do not use the limb measurements for stratospheric estimates and concentrate on aspect (b) in this study.

Our retrieval algorithm of slant column densities is quite similar to the established settings for the GOME instrument [6, 7, 14]. The spectral range 430-450 nm was used for the DOAS [3] fitting procedure. The solar reference was taken from the daily SCIAMACHY measurements. Furthermore, the cross sections of O_3, O_4 and H_2O as well as a Ring-spectrum, and a polynomial acting as high pass filter, are fitted. The fit generally works properly. The derived column densities agree well with the results from validation measurements (see [15]).

3. GOME NARROW SWATH MODE DATA

Besides the GOME standard size mode (SSM) with a resolution of 320*40 km^2, every tenth day GOME operates in the so called "narrow swath mode" (NSM) with a spatial resolution of 80*40 km^2 (Fig. 1). A detailed study of the GOME NSM NO_2 data is presented in [16]. The long time series of GOME NSM data (from 1997 on) and the fact that the NSM backscan pixel (240*40 km^2) is comparable in size to the GOME SSM pixels allows the direct quantitative comparison of results of different pixel sizes. From this we also learn what would be qualitatively expected from SCIAMACHY measurements. Furthermore, the GOME NSM composite of NO_2 TVCDs serves as a highly resolved reference map of the past.

Here we summarize some of the results presented in [16] and complement them with respect to recent SCIAMACHY results.

4. HIGH RESOLUTION MAPS OF NO_2

4.1 SCIAMACHY results

Fig. 2 shows the mean TVCDs over Europe as seen from GOME SSM. The polluted regions in the Po valley (northern Italy) and in western Germany / Belgium / Netherlands can be identified, but further structures are

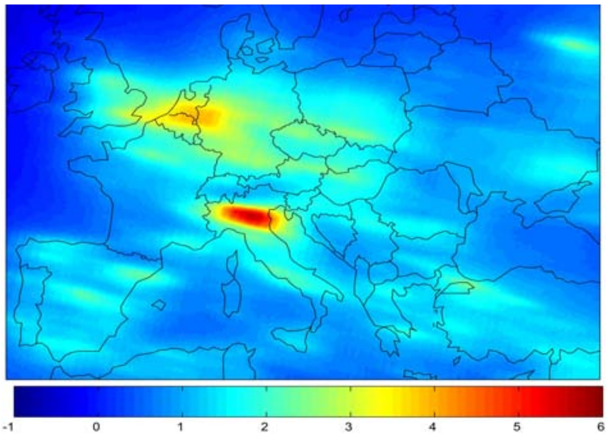

Fig. 2. Mean NO_2 TVCD (10^{15} molec/cm^2) for Europe as derived from GOME SSM data (1996-2001)

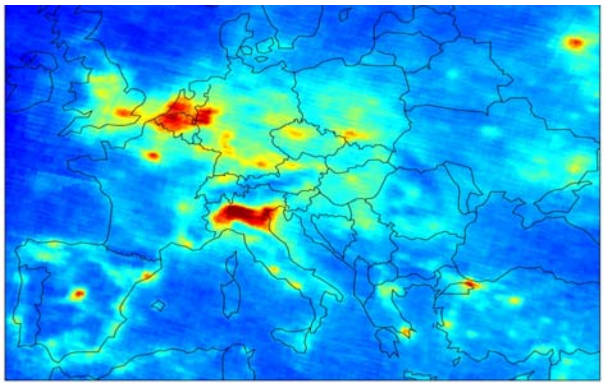
Fig. 3. Mean NO_2 TVCD for Europe as derived from SCIAMACHY (January 2003 - June 2004). Colorscale as in Fig. 2.

hard to distinguish. The direct comparison with the SCIAMACHY composite in Fig. 3 impressively illustrates the benefit of higher spatial resolution: several hot spots show up that can be directly attributed to large cities (e.g. London, Paris, Madrid, Athens, Moscow etc.).

Figs. 4 and 5 display the mean NO_2 TVCD from SCIAMACHY for the USA/Mexico and the Middle East, respectively. As for Europe, the most hot spots of enhanced TVCDs coincide with the location of large cities. However, in the US also the large coal power plant "Four corners" can be identified (marked white in Fig. 4) as already demonstrated for the GOME NSM data in [16].

In Fig. 5, also the populated region around the Nile river is clearly reflected. Furthermore, as recently demonstrated [17], even ship tracks in the Red Sea can be identified, whereas GOME data only allows to detect the highly cruised track between Sri Lanka and Indonesia that is almost parallel to GOME pixels [10].

The highly resolved spatial information as derived from SCIAMACHY (or GOME NSM) is thus of importance for the identification of specific sources, i.e. megacities,

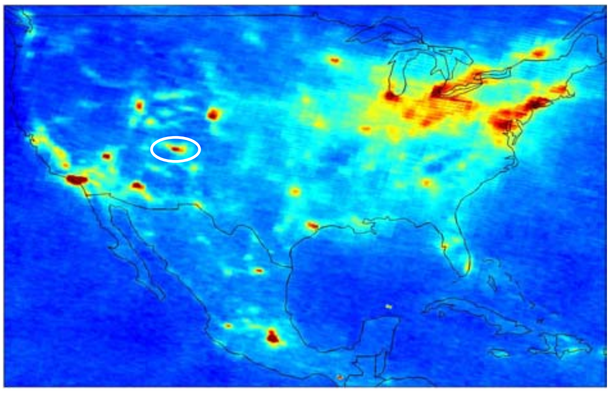

Fig. 4: Mean NO_2 TVCD for USA and Mexico as derived from SCIAMACHY (January 2003 - June 2004). Colorscale as in Fig. 2. The marked spot is associated to a large power plant ("Four corners") with >2 GW power.

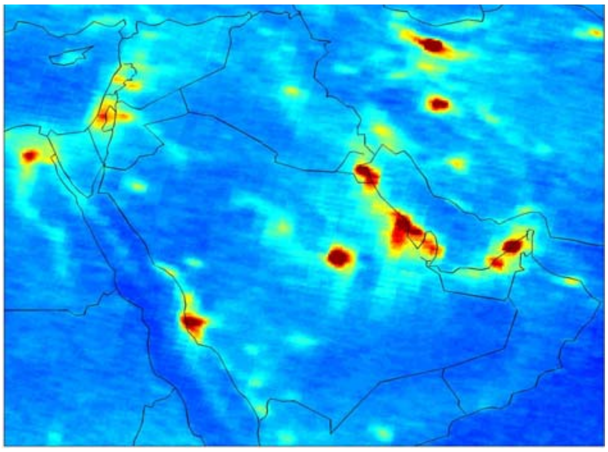

Fig. 5: Mean NO_2 TVCD for the Middle East as derived from SCIAMACHY (January 2003 - June 2004). Colorscale as in Fig. 2.

power plants or ship tracks. It can further be compared to the spatial patterns of emission inventories to check for consistency. For instance, the inventory from Wickert [18] states the Berlin NO_x emissions to be as large as those in the Ruhr region (50-100 t/km^2), whereas we find only a slight enhancement of NO_2 TVCDs over Berlin, in GOME NSM as well as in SCIAMACHY data. The same holds for other large cities that seem to be rather clean, as Hamburg (1.7 Mio. inhabitants, Germany), Warszawa (1.6 Mio, Poland) or Kyyiv (2.6 Mio, Ukraina,).

Also comparisons with the widespread EDGAR database [19] reveal some inconsistencies. For instance, the Saudi Arabian sources show only moderate source strength in EDGAR, while Jeddah, Riyadh and the cities at the Persian Gulf show relatively high TVCDs (Fig. 5). Thus, the SCIAMACHY mean composite can obviously be used to check and improve emission inventories.

4.2 Comparison with GOME NSM results

Figure 6 shows the mean NO_2 TVCD of all GOME narrow swath pixels during 1997-2002, corrected for artifacts due to the patchy temporal coverage of NSM observations (see [16]). The resulting map is compared to the global SCIAMACHY composite shown in Fig. 7.

Overall, Fig. 6 and 7 show a similar distribution of TVCDs. Nevertheless, there are some differences. The scatter of the GOME NSM TVCDs in the southern Atlantic (Fig. 6) is due to the South Atlantic Anomaly, i.e. a deformation of the magnetic field of the earth, allowing high energy cosmic radiation to hit the satellite and affect the spectral data. Obviously, this has only a week influence on the SCIAMACHY measurements (Fig. 7), where only a handful of pixels are affected.

To display further deviations between both mean composites, Fig. 8 shows their difference. Discussing the deviations, one has to keep in mind that:

(a) A slightly different reference sector for the stratospheric estimation was chosen for the SCIAMACHY retrieval of TVCDs. This is most likely the reason for the homogenous bands of e.g. apparently higher SCIAMACHY TVCDs over oceans between 0° and 30° N and especially for the lower SCIAMACHY values south from 15° S.

(b) The SCIAMACHY data enclose only 1.5 years, and is thus exposed to normal year to year fluctuations. For instance the higher SCIAMACHY TVCDs in Central Africa seem to arise from NO_x emissions above average in the biomass burning season in summer 2003.

(c) In the SCIAMACHY composite, the months January-June are represented twice, but autumn months only once. This might explain for instance the lower SCIAMACHY TVCDs over Australia, where TVCDs show a strong yearly cycle due to lightning [12].

(d) The SCIAMACHY pixels are smaller (30*60=1800 km^2) than the GOME SSM pixels (40*80=3200 km^2). Hence, it is expected that TVCDs of SCIAMACHY are higher over NO_2 hot spots with small spatial extent. Table 1 lists the maximum TVCDs for several cities as observed with different pixel sizes. As expected, the maxima are lowest for the GOME SSM (2nd column), as the large ground pixels smooth the actual TVCDs over 320 km east-west. As discussed in [16], the ratio of NSM and SSM TVCDs (5th column) depends on the homogeneity of the NO_2 distribution. It equals 1 for a totally homogenous NO_2 burden, whereas a high ratio (as in Mexico City) indicates a NO_2 hot spot of rather low extent (~50 km). The typical extent (FWHM) of the NO_2 plumes derived in [16], however, is of the order of ~100km. The expected increase of the SCIAMACHY observations due to the reduced pixel size is thus about <10%.

The measured SCIAMACHY TVCDs (4th column) are generally of the order of the GOME NSM observations

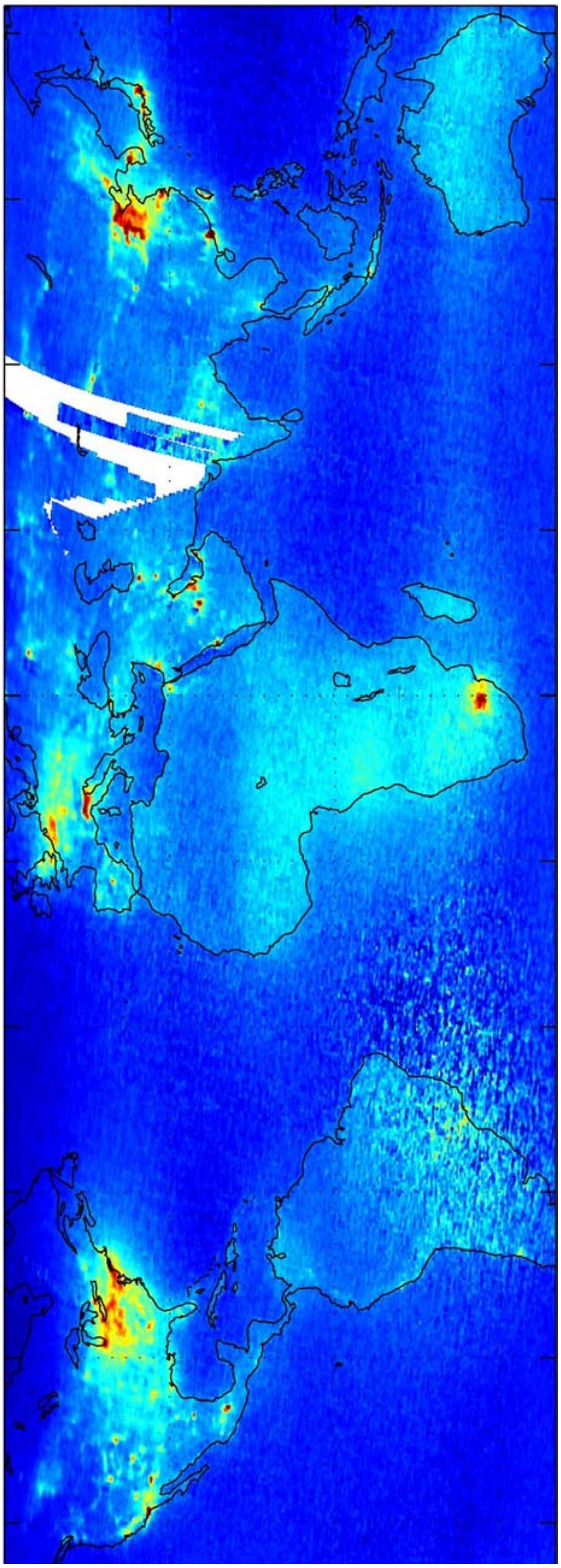

Fig.6.Mean NO_2 TVCD from GOME NSM (1997-2001). Figure taken from [16]. Colorscale as in Fig. 2.

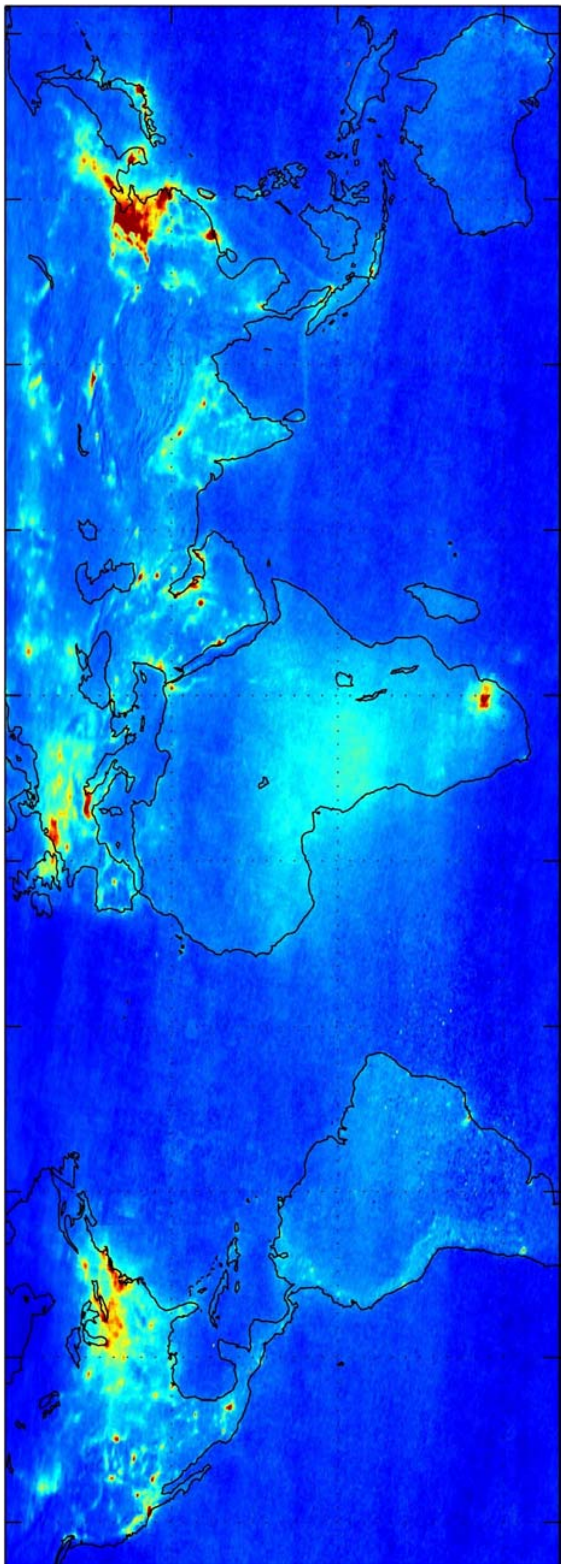

Fig. 7. Mean NO_2 TVCD from SCIAMACHY (January 2003 - June 2004). Colorscale as in Fig. 2.

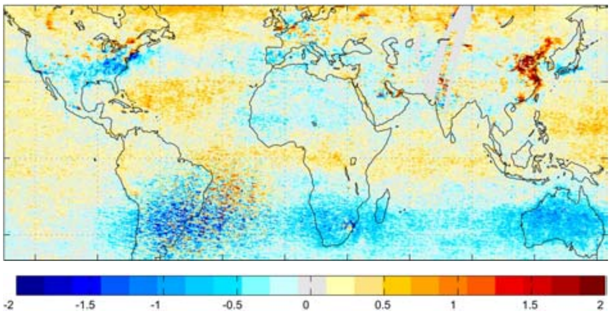

Fig. 8. Difference of mean TVCDs from SCIAMACHY (Fig. 7) and GOME NSM (Fig.6).

Table 1. Maximum TVCD of several cities for different pixel sizes.

City	VCD SSM	VCD NSM	VCD SCIA	NSM/ SSM	SCIA/ NSM
Los Angeles	8.70	22.42	17.16	2.58	0.77
New York	4.98	8.30	9.63	1.67	1.16
Mexico City	4.88	15.66	15.13	**3.21**	0.97
Ruhr Region	3.94	5.74	6.07	1.46	1.06
Milan	5.68	8.30	8.88	1.46	1.07
South Africa	6.56	9.22	8.35	1.41	0.91
Jeddah	3.18	8.44	8.17	2.65	0.97
Riyadh	4.32	9.68	12.03	2.24	1.24
Hong Kong	6.16	12.86	14.19	2.09	1.10
Shanghai	4.32	7.84	9.35	1.81	1.19
Beijing	5.70	8.32	11.80	1.46	**1.42**
Seoul	5.46	10.24	12.25	1.88	1.20
Tokyo	4.30	8.74	10.83	2.03	1.24
Istanbul	2.44	5.56	5.19	2.28	0.93
Shijiazhuang	5.86	9.60	13.07	1.64	**1.36**
Zhengzhou	4.66	6.56	13.66	1.41	**2.08**

(3rd column). In most cities, the SCIAMACHY/NSM ratio (column 6) does not exceed 1.2, and is even lower than 1 in some cases (for reasons (a), (b), (c) and/or (e)). The high ratios of ~1.4 (Beijing, Shijiazhuang) up to 2 (Zhengzhou) can not be explained solely by the reduced pixel size.
(e) The GOME NSM composite (1997-2001) is a reference for the time before the ENVISAT launch, in contrast to the recent SCIAMACHY measurements (2003-2004). Thus differences in both datasets may indicate a change in emissions. The most impressive difference of SCIAMACHY and GOME NSM composites shows up in China where it is more than $2*10^{15}$ molec/cm^2 for a large area and up to $7*10^{15}$ molec/cm^2 for 113.4°E, 35.0°N, i.e. the Chinese megacity Zhengzhou. Here also the highest ratio of 2.08 between SCIAMACHY and NSM occurs (see Table 1). This large and significant increase of TVCDs over China obviously indicates drastically increased emissions over the last years (see also [20]).

4.3 Comparison with backscan

The total swath width of SCIAMACHY observations is 960 km, i.e. 16 pixels a 60 km. These are followed by 4 backscan pixels with 240 km width each.
Clouds shield the tropospheric NO_2 column. Furthermore, the clouded part of a ground pixel is generally brighter than the cloud free part. The VCD derived from backscan measurements (i.e. the intensity weighted mean of the forescans) is thus expected to be lower than the actual mean of the forescans (see also [16]).
However, the quantitative comparison of back- and forescan for the NSM data revealed that the backscan TVCDs (with triple size) result on average in the same values as the forescan observations [16]. Here we repeat this study for SCIAMACHY data. Fig. 11 shows the correlation of averaged forescan VCDs with the respective backscan VCD. As for the NSM, we could not find systematically lower backscan VCDs, but find a 1:1 relation: the slope of the linear fit is 1.0000±0.0001.

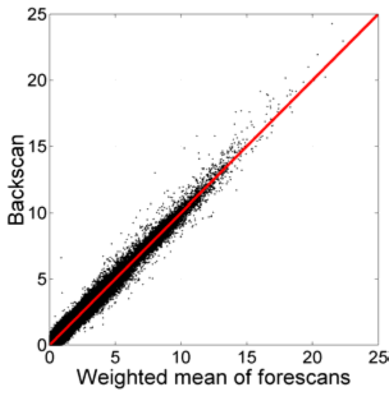

Fig. 11: Correlation of the averaged SCIAMACHY forescan VCDs (10^{15} molec/cm^2) and the respective backscan VCD.

A possible explanation for this is given in [16]: In most cases, cloud fractions for fore- and backscan pixels are quite similar, and the extreme case of a totally clouded pixel next to a totally cloud free pixel is rather rare. The maximum intensity exceeds the minimum by a factor of 6 in extreme cases, but only by 1.8 on average.
The expected underestimation of the backscan observations was quantified for the NSM pixels in [16], using the intensity information, and was found to be only about 4%, thus too small to be significantly detected in NSM data. For SCIAMACHY, however, the amount of available data is much higher; in Fig. 11 750,000 data points are plotted. Thus we should be able to detect a deviation of the slope from 1 if it exists. The fact that we do not find the slightest indication for lower backscan VCDs remains thus quite astonishing. However, it possibly could be explained by amounts of NO_2 inside the upper cloud layers or above the clouds, where its visibility is enhanced.

5. CONCLUSION

SCIAMACHY data can be used to derive TVCDs of NO_2, thus provide the continuation of the GOME dataset. Due to the improved spatial resolution as compared to GOME, sources of the scale of large cities as well as large power plants or ship tracks can clearly be identified.

The narrow swath mode of GOME allows to retrieve a global map of the distribution of NO_2 TVCDs with a similar spatial resolution as SCIAMACHY, and thus can serve as a reference for the time before the launch of ENVISAT. A quantitative comparison reveals that the largest differences occur in China, where emissions seem to have increased drastically over the last few years.

A comparison of SCIAMACHY fore- and backscan VCDs shows a 1:1 correlation, what is not expected due to the shielding effect of clouds accompanied by their brightness. The role of the NO_2 profile and adequate radiative transfer modelling for partly clouded scenes will be analyzed furthermore in future.

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