AN IMPROVED TOTAL AND TROPOSPHERIC NO_2 COLUMN RETRIEVAL FOR GOME-2

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ABSTRACT

This paper focuses on an improved algorithm for the retrieval of total and tropospheric NO_2 columns from the Global Ozone Monitoring Experiment–2 (GOME-2). A larger 425-497 nm wavelength fitting window with correction for GOME-2 slit function variations is used to determine the NO_2 slant column density. The STRatospheric Estimation Algorithm from Mainz (STREAM) is applied to determine the stratospheric column density of NO_2. A new surface Lambertian equivalent reflectance (LER) climatology based on GOME-2 observations is used for the calculation of the air mass factor (AMF). Examples of the retrieved GOME-2 total and tropospheric NO_2 columns are shown for Europe and Asia.

1. INTRODUCTION

Nitrogen dioxide (NO_2) plays significant roles in both stratospheric and tropospheric chemistry. It is strongly related to ozone destruction in the stratosphere, and is regarded as an ozone precursor and an important air pollutant affecting human health and ecosystem in the troposphere. Since the rapid economic and population growth in many developing countries, e.g. China, the emissions of nitrogen oxides have been widely increasing mainly due to the fossil fuel consumption. Therefore, a continuous global observation of tropospheric NO_2 to extract emission information is of great importance. Since the local ground-based measurements are relative sparse, the observations from satellites enable reliable global monitoring on a global scale and on long time scales. Using observations from GOME/ERS-2 and SCIAMACHY/Envisat, Richter et al. [1] illustrate the increase in tropospheric NO_2 columns over Eastern China.

The Global Ozone Monitoring Experiment–2 (GOME-2) on the MetOp-A/B satellites is continuing the long-term monitoring of atmospheric traces gases by GOME and SCIAMACHY. GOME-2 is a nadir-scanning UV-VIS spectrometer measuring the Earth's backscattered radiance and extra-terrestrial solar irradiance in the spectral range between 240 and 790 nm. The abundance of NO_2 in the stratosphere and troposphere is determined from the narrow band absorption structures of NO_2 in the back-scattered and reflected radiation in the visible spectral region.

In this work, we developed an improved retrieval algorithm of total and tropospheric NO_2 columns from GOME-2. This new algorithm will be implemented in a future version of the operational GOME Data Processor (GDP) at the German Aerospace Center (DLR). First, we describe the current operational total and tropospheric NO_2 retrieval algorithm for GOME-2 (GDP 4.8). Then the improvements in the various steps in the total and tropospheric NO_2 retrieval algorithm are presented, including the slant column retrieval, the troposphere-stratosphere separation, and the calculation of the air mass factor (AMF). Section 3 shows examples of the retrieved tropospheric NO_2 columns from GOME-2 using the improved algorithm, followed by a discussion and future outlook of the work.

2. THE METHODOLOGY

2.1 Operational total and tropospheric NO_2 retrieval for GOME-2

The operational total and tropospheric NO_2 column retrieval for GOME-2 consists of several algorithm steps [2,3]. First, the differential optical absorption spectroscopy (DOAS) method [4] is applied to determine the NO_2 slant columns from GOME-2 (ir)radiance spectra in the visible wavelength region between 425 nm and 450 nm. Then the initial total NO_2 vertical columns are calculated by an AMF conversion using the LIDORT radiative transfer model [5] and a stratospheric NO_2 profile climatology [6]. In LIDORT, the atmosphere is separated into a number of optical layers and formalised based on various inputs, such as the optical properties in each layer, the viewing geometry and the surface albedo. For cloudy scenarios, the AMF is calculated using GOME-2 cloud properties derived from the OCRA and ROCINN algorithms [7,8].

In order to separate the stratospheric and tropospheric components, a spatial filtering algorithm is applied to estimate the stratospheric NO_2 columns, which uses the nadir measurements over relatively clean areas, referred as modified reference sector method (MRSM). A pollution mask is defined to skip polluted regions with

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large tropospheric NO_2 columns. Based on MOZART-2 chemistry transport model results, the regions with monthly averaged tropospheric NO_2 columns larger than 1.0×10^{15} molec/cm^2 are classified as 'polluted'. The residual 'clean' pixels are low-pass filtered in the zonal direction.
Finally, tropospheric air mass factors are computed using monthly averaged NO_2 profiles from the MOZART-2 model and the tropospheric NO_2 columns can be determined. The calculation of the tropospheric air mass factors requires the same inputs as the AMF calculation for the initial total NO_2 vertical columns, but its dependency on these inputs as well as on the *a priori* NO_2 profile is much stronger.

2.2 Optimization of DOAS slant column algorithm

The use of an extended 425-497 nm wavelength fitting window to determine the NO_2 slant column density with the DOAS method has been analysed [9]. Theoretically the retrieval quality will be improved, since more spectral points are included in the fit. However, spectral interference with other absorbing trace gases also increases. Therefore, additional absorption cross-sections for liquid water and desert are included in the DOAS fit to reduce systematic errors, as well as Eta and Zeta polarization calibration functions for the GOME-2 instrument. In addition, the degree of polynomial (used to separate the broad and narrow band spectral structures of the spectra) has been increased from three to five, aiming at correcting for the scan angle dependent degradation of GOME-2 instrument. To account for the slit function variation of GOME-2 with time, effective slit functions are derived from GOME-2 solar irradiance spectra by adjustment to the high resolution solar reference of Chance and Kurucz [10].

Fitting window	425 – 497 nm
Cross sections	NO_2 240K (Vandaele et al., 2002) H_2O (HITRAN 2000) O_3 228K (Brion et al., 1998) O_4 (Greenblatt et al.) liquid water (Pope et al.) and sand (Richter) GOME-2 Eta and Zeta polarization vectors
Ring effect	Effective Ring spectrum (Chance and Spurr)
Polynomial	5 order
Slit function	Fitted Asymmetric Gaussian for GOME-2
Reference spectrum	Daily solar spectrum from GOME-2

Table 1. Improved DOAS setting for slant column calculation

The effect of the large fitting window on the retrieved NO_2 column for GOME-2/MetOp-A is shown in Fig. 1. The use of the larger 425-497 nm fitting window results in an overall increase in the retrieved NO_2 columns. Considering the seasonal variation and pollution condition, larger absolute increases are found over the regions with higher amount of NO_2 columns. The (sub)tropics also show relatively large changes.

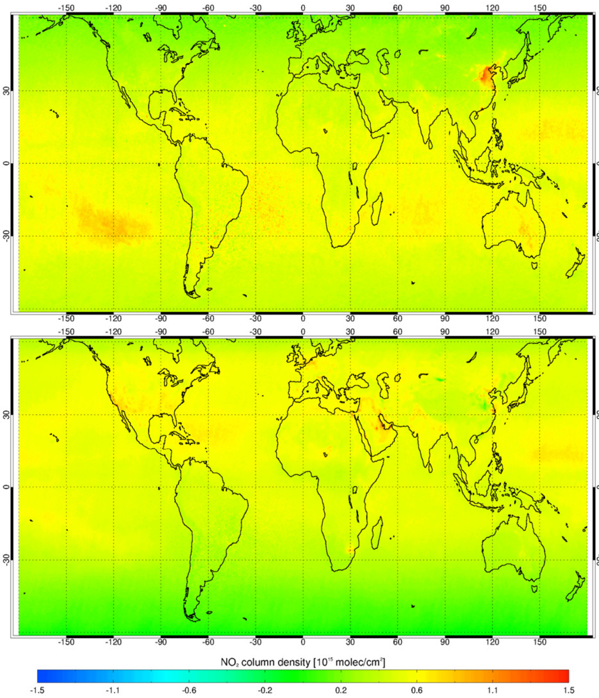

Figure 1. Monthly average difference of total NO_2 from a large 425-497 nm wavelength fitting window and the standard 425-450 nm wavelength fitting window, measured by GOME-2/MetOp-A for Januray (top) and July (bottom) 2008.

A statistical analysis has been applied to estimate the precision of the NO_2 slant columns for the standard and extended fitting windows, as shown in Fig.2. The clean tropical Pacific (20º S-20º N, 160º E-180º E) is divided into small boxes (2º×2º), and the spread of the variation of the NO_2 columns within each box is regarded as a good measure of random noise. The Gaussian distribution has a FWHM of 1.36×10^{15} molec/cm^2 for the standard fitting window and 1.04×10^{15} molec/cm^2 for the large fitting window. This corresponds to a slant column error of about 0.57×10^{15} molec/cm^2 and 0.44×10^{15} molec/cm^2, respectively. Hence, by using an extended fitting window, the random error in the individual GOME-2 NO_2 measurements has been decreased by ~25%.

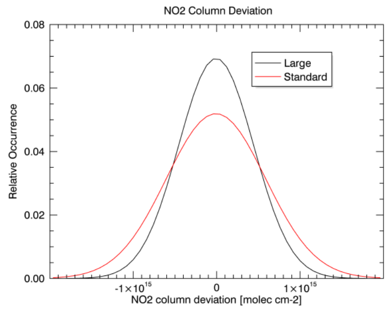

Figure 2. the Gaussian distribution of deviations with the NO_2 columns from corresponding ($2^o \times 2^o$) box mean values in the tropical Pacific region (20^o S-20^o N, 160^o E-180^o E), using a large fitting window (black) and the standard fitting window (red). Curves are normalized to have unit area and centered on zero.

2.3 Stratosphere-troposphere separation

The STRatospheric Estimation Algorithm from Mainz (STREAM) is applied to determine the stratospheric column density of NO_2 [11]. Belonging also to the MRSM, the STREAM derives weighting factors for each satellite pixel, which define the contribution from the initial NO_2 total columns to the stratospheric field. The potentially polluted pixels are weighted low, instead of being totally masked out in the currently used spatial filtering algorithm. Cloudy GOME-2 observations with cloud top pressures between 350 and 650 hPa are given higher weighting, since they directly provide information of the stratospheric columns. Additionally, if a large area of pixels suffers from large biases in the tropospheric residues, the weights will be further adjusted iteratively. The stratospheric NO_2 component is then derived by weighted convolution and subtracted from the initial total columns to get the tropospheric residues. The tropospheric residues calculated with the STREAM and the spatial filtering algorithms are compared in Fig. 3. The bias between the tropospheric residues calculated with STREAM and the current spatial filtering algorithm is relatively small above the ocean. In general, the use of the STREAM leads to an increase in the tropospheric residues, especially over continents, and this effect is larger in summer.

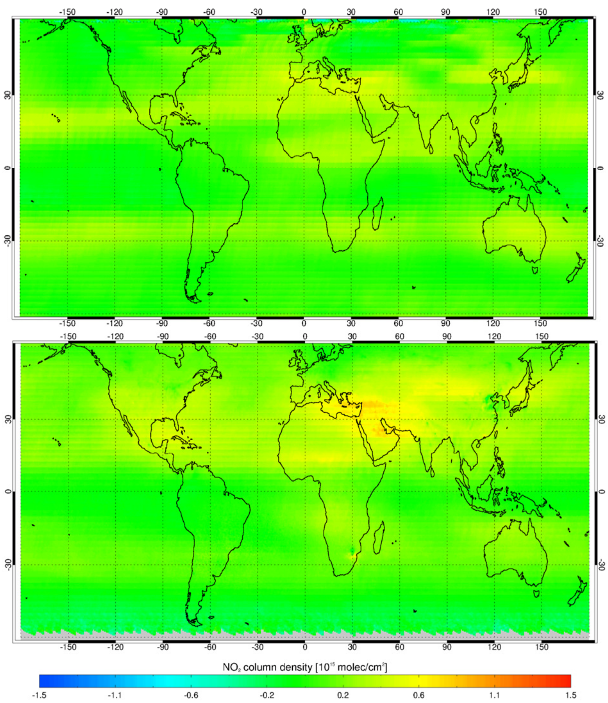

Figure 3. Monthly average difference of NO_2 tropospheric residues from STREAM and the spatial filtering algorithm, measured by GOME-2/MetOp-A for Januray (top) and July (bottom) 2008.

2.4 Surface reflectance climatology for NO_2 AMF calculation

The use of a new surface Lambertian equivalent reflectance (LER) climatology based on GOME-2 observations [12] for the calculation of the AMF has been analysed. In contrast to the currently used surface LER climatology based on GOME/ERS-2 data, the new surface LER climatology is consistent with the GOME-2 NO_2 observations (i.e. illumination conditions and instrument characteristics). The GOME-2 surface albedo climatology takes advantage of newer observations for 2007-2013, smaller footprint size, and an improved LER algorithm. The effect of the new surface albedo climatology on the tropospheric NO_2 retrieval is shown for March 2008 in Fig. 4.

Changes in surface LER have a large effect on the tropospheric AMF for NO_2. As illustrated in Fig. 4, the differences in the retrieved tropospheric NO_2 columns are consistent with the changes in the surface LER. For example, over central Europe, the GOME-2 surface LER climatology shows smaller values, and therefore a lower sensitivity to tropospheric NO_2 is assumed in the AMF calculation. This results in a decrease in the AMF and hence, an increase in the retrieved tropospheric NO_2 column from GOME-2 is found.

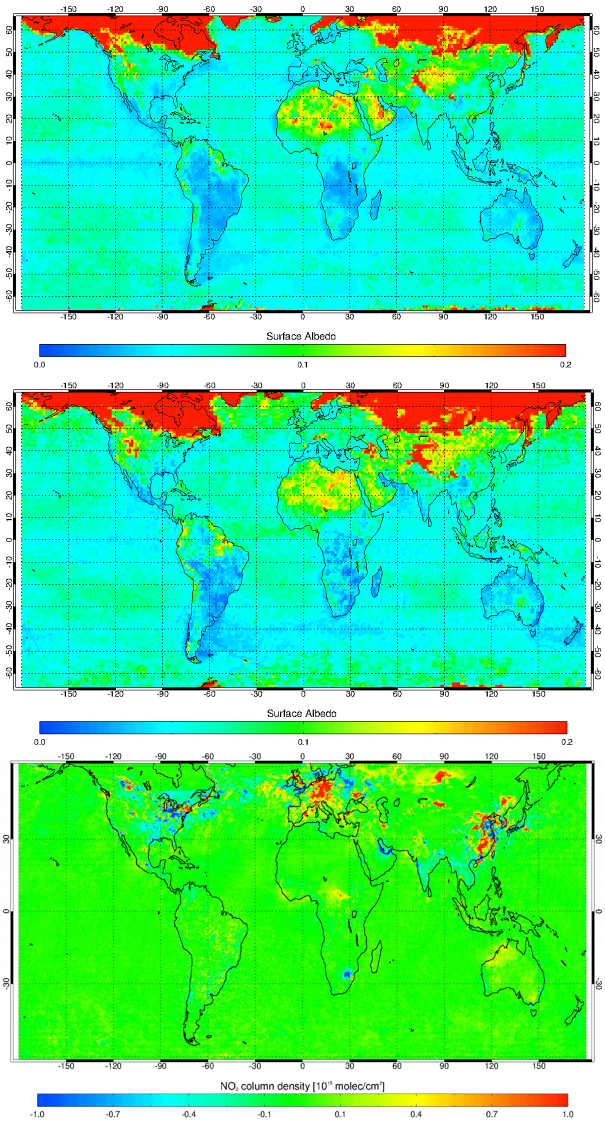

Figure 4. Map of surface LER data for 440 nm in March based on GOME-2 observations [12] (top) and GOME/ERS-2 data (middle). The bottom panel shows the difference between the tropospheric NO_2 columns for March 2008 retrieved using the new GOME-2 surface LER climatology and the old one based on GOME/ERS-2 data.

3. EXAMPLES OF GOME-2 TROPOSPHERIC NO_2

Fig. 5 shows the monthly averaged tropospheric NO_2 columns from GOME-2 for March 2008 over Europe, as retrieved with the improved algorithm and the GDP 4.8 algorithm. Both panels show the high tropospheric NO_2 columns over large urban and industrial regions, such as South-East England, Netherlands, and the northern Italy. Hotspots like Madrid, Paris, Ruhr area and Moscow are clearly visible in the GOME-2 measurements. The new algorithm generally results in larger tropospheric NO_2 columns over Europe, which is mainly related to the improved STS algorithm.

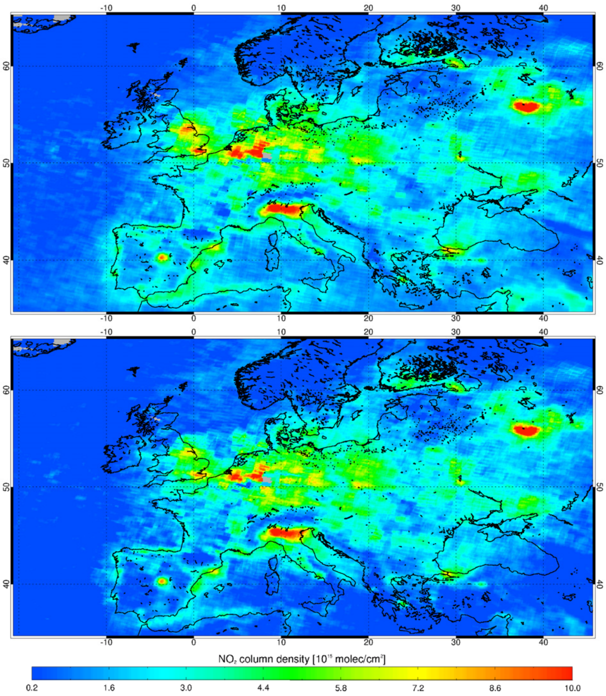

Figure 5. Monthly average tropospheric NO_2 over Europe retrieved using the improved algorithm (top) and the GDP algorithm (bottom), measured by GOME-2/MetOp-A for March 2008.

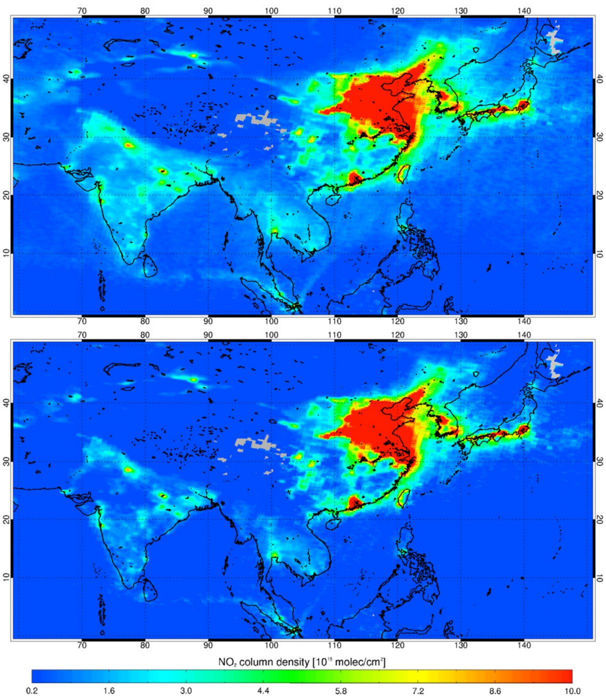

Figure 6. Monthly average tropospheric NO_2 over Asia retrieved using the improved algorithm (top) and the GDP algorithm (bottom), measured by GOME-2/MetOp-A for March 2008.

Fig. 6 shows the monthly averaged tropospheric NO_2 columns from GOME-2 for March 2008 over Asia. The coastal regions in China, Japan North Korea and South Korea show large amount of tropospheric NO_2 columns, with the appearance of hotspots like New Delhi, Guangzhou and Tokyo. A large area with extremely high NO_2 pollution is found over east China, due to the rapid development of its economy. Comparing to the GDP algorithm, the improved retrieval algorithm points out larger tropospheric NO_2 columns over Asia as well. Another feature in the improved data is the enlarged tropospheric NO_2 along the shipping lanes in the Bay of Bengal and the South China Sea [13].

4. SUMMARY

In this work, an improved algorithm for the retrieval of total and tropospheric NO_2 columns from GOME-2 has been developed. The use of the improved algorithm results in an overall increase in the retrieved NO_2 columns. In the DOAS fit, a larger 425-497 nm wavelength fitting window with correction for GOME-2 slit function variations is applied, leading to an increase in the total NO_2 columns and a decrease of the noise in the NO_2 columns. The use of new STREAM algorithm for the stratosphere-troposphere separation generally results in larger tropospheric NO_2 columns, especially over polluted continental regions. In the AMF calculation, the use of a new GOME-2 surface LER climatology has a significant effect on the retrieved tropospheric NO_2 column. The examples of the tropospheric NO_2 distribution for Europe and Asia as retrieved with the improved algorithm show increased values, comparing to the operational GDP algorithm.

In the near future, the stratospheric-tropospheric separation will be further improved for the GOME-2 instrument. For the tropospheric AMF calculation, *a priori* NO_2 profiles from new chemistry transport model runs will be used. Furthermore, comparisons with other satellite products as well as with in-situ measurements will be performed.

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