Monitoring Nitrogen Oxides in the troposphere with satellite instruments

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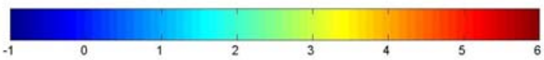
Abstract. We present the results of several studies on the identification and quantification of the different sources of NO_x (traffic, industry, biomass burning, lightning) from GOME satellite data. Furthermore, we demonstrate the potential of satellite data to estimate the mean lifetime of tropospheric NO_x.

Introduction

Nitrogen oxides ($NO+NO_2=NO_x$) are important ozone precursors in the troposphere. Global production is estimated to be of the order of 23-81 Tg [N]/yr, approx. half of it due to fossil fuel combustion (Lee et al., 1997). Further contributions arise from biomass burning and lightning.

Column densities of NO_2 are detectable from satellite platforms using differential optical absorption spectroscopy (DOAS, Platt 1994). Tropospheric vertical column densities (VCDs) can be retrieved by estimating and subtracting the stratospheric fraction (e.g. Leue et al., 2001; Martin et al., 2002). However, quantitative studies have to consider radiative transfer that is influenced by ground albedo, aerosol load, NO_2 profile and clouds.

The Global Ozone Monitoring Experiment GOME (Burrows et al., 1999) provides a time series of 8 years (1996-2003) of NO_2 column densities on a global scale with a spatial resolution of 320*40km^2. Since March 2002, SCIAMACHY provides the continuation of the GOME time series with an improved spatial resolution (60*30km^2) and new viewing geometries, allowing also direct stratospheric measurements.



Global distribution

The global coverage of LEO satellites allows the retrieval of mean maps of tropospheric NO_2. Due to the quite low lifetime of NO_2 in the troposphere, this reflects well the distribution of NO_x sources. The east-west resolution of GOME (320 km), however, causes a loss of spatial information. In contrast, the improved resolution of SCIAMACHY allows to resolve structures in the order of the size of large cities. The benefit of this resolution can be assessed using GOME observations in the narrow swath mode (NSM), i.e. 80*40km^2, operating three days per month. Fig. 1 displays the 5 year mean of the NSM observations that are corrected for temporal inhomogeneous sampling (Beirle et al., 2004(a)).

The impact of anthropogenic emissions is clearly visible. Whereas the mean of all GOME pixels only pronounces the large industrial areas in the US, Europe and east Asia, the NSM mean also allows the identification of single cities and even large power plants (Four Corners, New Mexico).

Weekly Cycle

Since human activity in western countries is governed by a seven day cycle, emissions are reduced over weekends. This is reflected in the weekly cycles of tropospheric NO_2 VCDs for different regions of the world (Fig. 2) (Beirle et al., 2003). In industrialized regions, a clear weekly cycle with lowest VCD on Sunday can be found, with reductions up to 60% (Milan). The cities in the Middle East show a shifted weekly cycle: In Israel Saturday is the day of lowest VCD due to the Sabbath being the religious day of rest. In Islamic cities there is a slight weekly effect with lowest VCD on Friday. In China, no weekly cycle can be found.

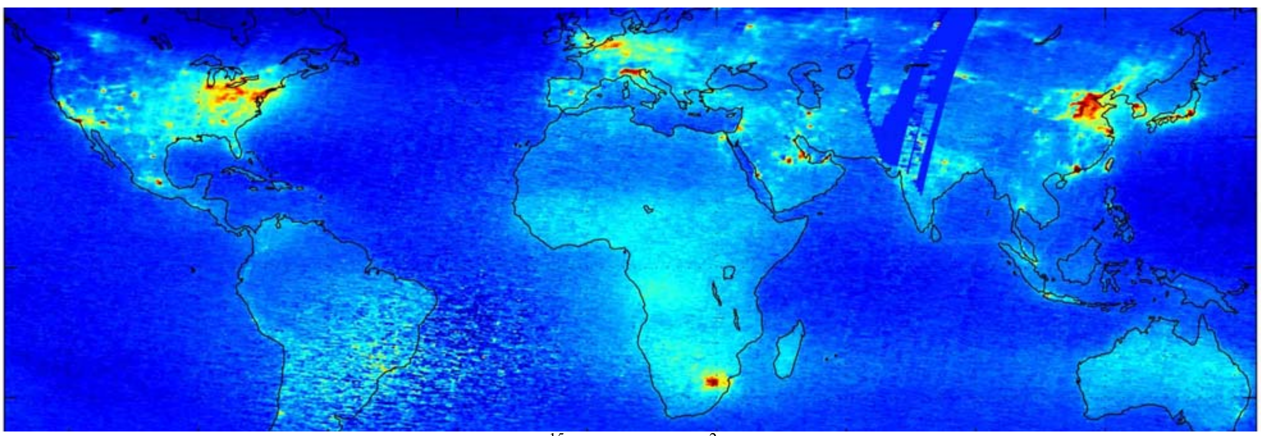


Figure 1. Global mean of tropospheric NO_2 VCD (10^{15} molecules/cm^2), using GOME narrow swath mode data with a spatial resolution of 40*80km^2, corrected for temporal inhomogeneous sampling (Beirle et al., 2004(a)).

The existence of a weekly cycle is an obvious signature of anthropogenic sources. The depth of the Sunday minimum holds information about the fraction of anthropogenic NO_2. This helps to discriminate between manmade and natural sources. A separate analysis for winter and summer months also allows to split up steady emissions (heavy industry), emissions with a seasonal cycle (heating, air conditioning) and emissions with a strong weekly cycle (industry, traffic).

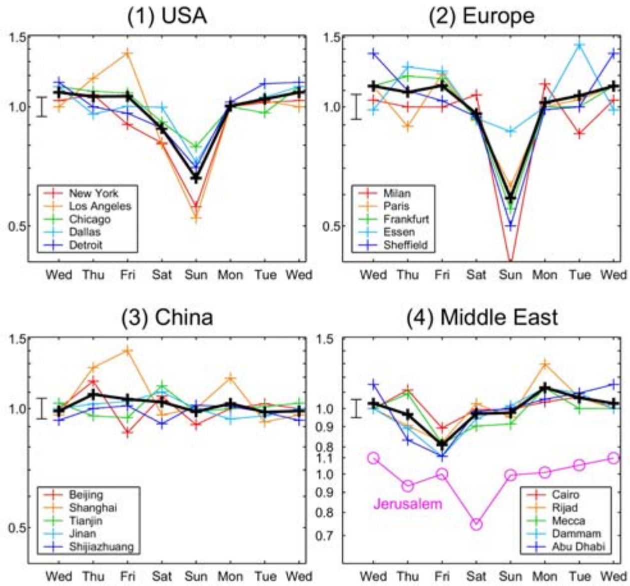


Figure 2. Weekly cycle of tropospheric NO_2 VCD for different cities (1996-2001 mean). The values are normalized on the median weekly value to be one (rel. units). Black lines are averaged curves. The errorbar to the left of every plot indicates the maximum error (standard error of the mean) of all data points.

Lightning

Lightning takes place around the world, but is distributed quite inhomogeneously. The regions of highest activity are placed over tropical land masses. In those regions, a direct estimation of NO_x produced by lightning is compli- cated by the fact that also other, probably more predomi- nant NO_x sources like industry (USA, South Africa) or re- gular biomass burning (South America, Central Africa, Indonesia) are present. So the challenge is to find a region with lightning activity, where other sources are of minor importance. Moreover, to analyze the influence of lightning with statistical methods, it is helpful to have a strong tem- poral variability, i.e. a yearly cycle. Therefore, we have chosen Central Australia, where we correlated monthly means of lightning frequency (LIS) and of the tropospheric vertical column densities of NO_2, and estimated the globally produced lightning NO_x to be approx. 2.8 (0.8-14) Tg [N]/year (Beirle et al., 2004(b)).

Biomass Burning

Several studies reported biomass burning events detected in GOME data, as well concentrating on single fire events (Spichtinger et al., 2001; Damoah et al., 2004) as considering seasonal or monthly means (Richter and Burrows, 2001; Spichtinger et al., 2004). However, any quantitative approach to estimate NO_x emissions from biomass burning is severely disturbed by the smoke and aerosol production accompanied.

Lifetime

The lifetime of tropospheric NO_2 is of the order of hours up to a day. The sharp borders of the hot spots in Fig. 1 indicate that it can't be much longer even for sites as northerly as Moscow. For a quantitative estimate of lifetimes for different regions we investigate the weekly cycle: The low Sunday emissions influence the VCD measured on Monday in the area downwind from the source regions. We compared the mean weekly cycle for Germany for instance with weekly cycles modelled for different lifetimes and retrieved the lifetime to be about 6 hours in summer and 24 hours in winter. This method requires the presence of a strong source with a pronounced weekly cycle.

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