**Atmospheric
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Weekly cycle of NO$_2$ by GOME measurements: a signature of anthropogenic sources

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Abstract. Nitrogen oxides (NO+NO$_2$=NO$_x$ and reservoir species) are important trace gases in the troposphere with impact on human health, atmospheric chemistry and climate. Besides natural sources (lightning, soil emissions) and biomass burning, fossil fuel combustion is estimated to be responsible for about 50% of the total production of NO$_x$. Since human activity in industrialized countries largely follows a seven-day cycle, fossil fuel combustion is expected to be reduced during weekends. This "weekend effect" is well known from local, ground based measurements, but has never been analysed on a global scale before.

The Global Ozone Monitoring Experiment (GOME) on board the ESA-satellite ERS-2 allows measurements of NO$_2$ column densities. By estimating and subtracting the stratospheric column, and considering radiative transfer, vertical column densities (VCD) of tropospheric NO$_2$ can be determined (e.g. Leue et al., 2001). We demonstrate the statistical analysis of weekly cycles of tropospheric NO$_2$ VCDs for different regions of the world. In the cycles of the industrialized regions and cities in the US, Europe and Japan a clear Sunday minimum of tropospheric NO$_2$ VCD can be seen. Sunday NO$_2$ VCDs are about 25–50% lower than working day levels. Metropolitan areas with other religious and cultural backgrounds (Jerusalem, Mecca) show different weekly patterns corresponding to different days of rest. In China, no weekly pattern can be found.

The presence of a weekly cycle in the measured tropospheric NO$_2$ VCD may help to identify the different anthropogenic source categories. Furthermore, we estimated the lifetime of tropospheric NO$_2$ by analysing the mean weekly cycle exemplarily over Germany, obtaining a value of about 6 h in summer and 18–24 h in winter.

1 Introduction

Over the last 150 years, the atmospheric composition has changed dramatically due to the industrial revolution. Among the various emitted pollutants nitrogen oxides (NO+NO$_2$=NO$_x$ and reservoir species) play an important role. In the troposphere they have a large impact on human health, climate and atmospheric chemistry, e.g. due to the role they play in ozone production (Atkinson, 2000) and their influence on the OH concentration ("oxidizing power", Jacob, 2003). The estimation of the strengths of the different NO$_x$ sources (industry, biomass burning, aircraft, soil emissions, lightning) still has high uncertainties (e.g. Lee et al., 1997). Nevertheless, NO$_x$ by fossil fuel combustion is estimated to account for more than 50% of the overall production.

Industrial activity and traffic in western countries are reduced during weekends leading to lower levels of emitted pollutants ("Weekend effect"; Cleveland et al., 1974; Elkus and Wilson, 1977; Cerveny and Coakley, 2002). Compared to working day levels the weekend emissions of NO$_x$, e.g. in Germany, are reduced by approximately 35% (Wickert, 2001). The degree of reduction of industrial activity and traffic may differ from country to country and even from region to region, but a Sunday minimum of NO$_x$ emissions is generally expected for all countries with a Christian tradition, celebrating Sunday as the day of rest.

Satellite measurements allow a new and independent approach to the determination of trace gas emissions. The entire globe is monitored with a single instrument under the same conditions and over long periods of time. The Global Ozone Monitoring Experiment GOME on board the ESA satellite ERS-2 provides data to determine vertical column densities (VCDs) of NO$_2$ (Wagner, 1999; Leue et al., 2001). By subtracting the estimated stratospheric fraction, tropospheric VCDs of NO$_2$ are retrieved (Leue et al., 2001; Velders et al., 2001; Wenig et al., 2001; Martin et al., 2002).
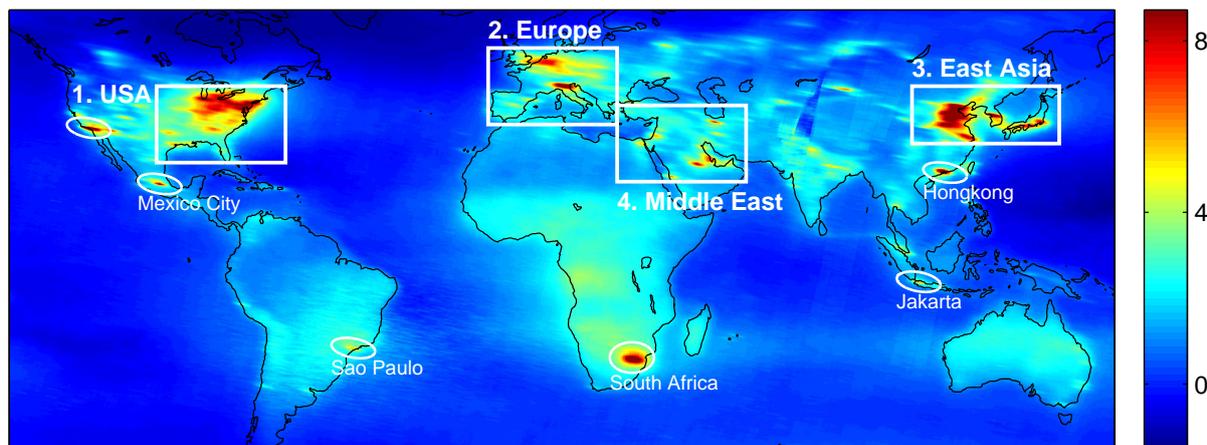
Fig. 1. Six years mean (1996–2001) of global tropospheric NO$_2$ Vertical Column Density in 10^{15} molecules/cm^2. The weekly cycle of the framed areas 1. US East Coast, 2. Europe, 3. East Asia and 4. Middle East, as well as 5. the marked individual Metropolises are considered in detail in this study.

Since the lifetime of tropospheric NO$_2$ is of the order of one day (Leue et al., 2001), enhanced VCDs directly indicate nearby sources of NO$_x$.

Remote sensing of the troposphere in principle is constrained by clouds and aerosol, and quantitative correction of radiative transfer requires additional information on surface albedo and trace gas profile (e.g. Velders et al., 2001). Furthermore, the spatial resolution of one GOME ground pixel (320×40 km^2) is rather coarse compared to the extent of individual cities. In spite of these uncertainties of individual VCD measurements, the statistical analysis of long time series of GOME data provides reliable information, for instance on the mean tropospheric NO$_2$ distribution and on features of its weekly cycle.

2   Retrieval of tropospheric NO$_2$ vertical column densities from GOME

In April 1995, the ERS-2 satellite was launched by the European Space Agency, carrying (in addition to other instruments) the Global Ozone Monitoring Experiment (GOME) (Burrows et al., 1999). The satellite operates at an altitude of about 780 km. It flies along a sun- synchronous polar orbit and crosses the equator at 10:30 (local time). GOME consists of four spectrometers measuring the radiation reflected by the earth in the UV/vis spectral range with a resolution of about 0.2–0.4 nm. Global coverage is achieved every three days. The primary objective of GOME is the retrieval of Ozone column densities, but beyond this the column densities of several trace gases can be determined by applying Differential Optical Absorption Spectroscopy (DOAS) (Platt, 1994). The retrieval of vertical column densities (VCDs) of NO$_2$ is described in detail by Wagner (1999), Leue et al. (2001) and Wenig (2001).

Since the global distribution of stratospheric NO$_2$ is much more homogeneous and less time dependent than it is in the troposphere, it is possible to estimate the stratospheric fraction of the total column (Leue et al., 2001; Velders et al., 2001; Wenig, 2001). For this study, the stratospheric column was determined in a reference sector over the remote Pacific (Richter and Burrows, 2002) and assumed to be independent from longitude. The resulting mean global distribution of tropospheric NO$_2$ generally justifies our assumptions about stratospheric homogeneity (see Fig. 1). But in some regions, especially the North Atlantic, and mainly in winter time, we obviously overestimated the stratosphere, resulting in negative tropospheric columns. This additive bias affects our study at least in the case of Europe. Its relevance will be discussed in conjunction with our results. (A recent study of stratospheric NO$_2$ from GOME is presented by Wenig et al., 2003, discussing the longitudinal homogeneity and seasonal dependency in detail.)

The difference between the total and the estimated stratospheric column represents the tropospheric fraction. For a quantitative analysis the tropospheric vertical column density has to be corrected for effects of radiation transfer in the troposphere. The absolute value of the correction factor depends on surface albedo, trace gas profile and especially the cloud cover (Richter and Burrows, 2002; Martin et al., 2002). In this study, we apply a uniform correction factor of 4 (Velders et al., 2001) to keep the analysis as simple as possible. We focus on the relations of mean VCDs of different days of the week, i.e. normalized weekly cycles. Any regional bias of the correction factor (e.g. due to a different profile shape) is multiplicative, thus not affecting our study.

The coverage of one GOME pixel is about 40 km north–south by 320 km east–west. This complicates the monitoring of individual cities: the total area of pixels containing a specific location is 80×640 km^2 (since the city center may be
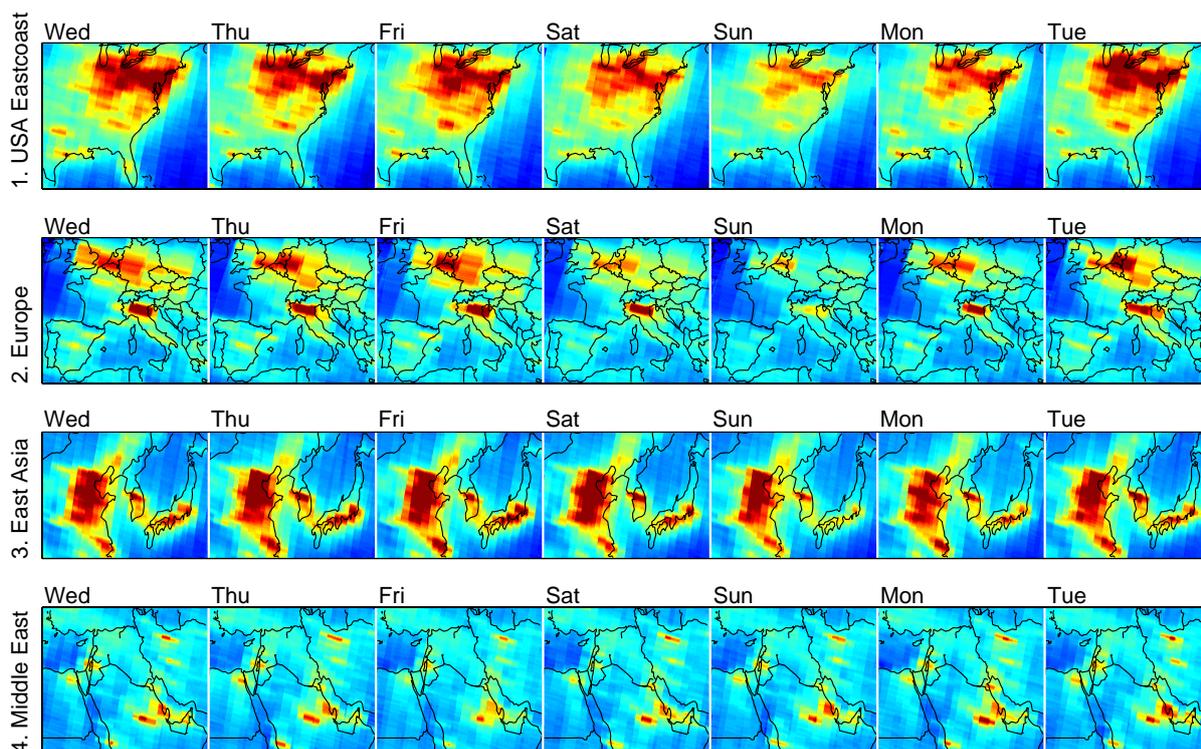
Fig. 2. Mean tropospheric NO$_2$ VCD (1996–2001) for the regions of interest, as marked in Fig. 1, for the different days of the week. Colour scale and unit are the same as in Fig. 1.

situated in the upper right corner of the GOME pixel as well as in the lower left corner), implying possible interferences with other sources.

The flight track of the ERS-2 satellite has a periodicity of 35 days. That means, that every 35 days the GOME ground pixel coordinates are identical. This circumstance may influence the analysis of weekly cycles, since 35 is a multiple of 7. Hence each day of the week is scanned with a different spatial sampling. This implies that, by monitoring a certain location, the influence of neighbouring regions differs from day to day. This effect is in general of minor importance, but may cause artefacts in the weekly cycle of tropospheric NO$_2$ VCD in a few cases (see below).

The retrieval of tropospheric information from satellite data can be strongly affected by clouds: an enhanced cloud cover might "shield" the troposphere and lead to the observation of a reduced VCD. In principle, there could even be a systematic weekly cycle of cloud cover. In fact, Cerveny and Balling Jr (1998) report a weekly cycle of precipitation. To examine if there is a weekly cycle of cloud cover we use the Heidelberg Iterative Cloud Retrieval Utilities (HI-CRU), recently presented by Grzegorski (2003). HICRU contains an improved algorithm that retrieves cloud covers using the GOME Polarization Monitoring Devices (PMD), which are broadband spectrometers with a better spatial resolution. Therefore, information on cloud cover is obtained for the same time and place as for the NO$_2$ retrieval.

By analysing the weekly cycle of the HICRU cloud cover data we found no evidence of a weekly pattern of enhanced cloud cover on Sundays. In the case of Germany, for instance, the mean cloud cover (1996–2001) is 0.450 on weekdays and 0.457 on Sundays. The weekday cloud covers (i.e. the fraction of the area covered by clouds) vary between 0.431 (Tuesday) and 0.479 (Friday); the standard deviation of the five mean weekday values is 0.018. Especially the fact that for Germany the day with the highest cloud cover shows no irregular features in the NO$_2$ VCD gives us confidence that the influence of cloud cover on our study is negligible. We did not observe enhanced cloud cover on Sundays for the US either.

3 Weekly cycle of tropospheric NO$_2$ VCD

The GOME measurements currently span more than 7 years, providing a large amount of data for statistical evaluation. Figure 1 shows the mean global distribution of tropospheric NO$_2$ VCDs for 1996–2001. Regions with high industrial activity and even individual metropolises, e.g. Mexico City, can clearly be seen. In this study, we focus on four regions of the world with enhanced NO$_2$ VCD values: 1. the eastern USA, 2. western Central Europe, 3. East Asia and 4. the Middle East, as well as 5. some individual cities, as marked in Fig. 1. Rural areas with other predominant sources of NO$_x$
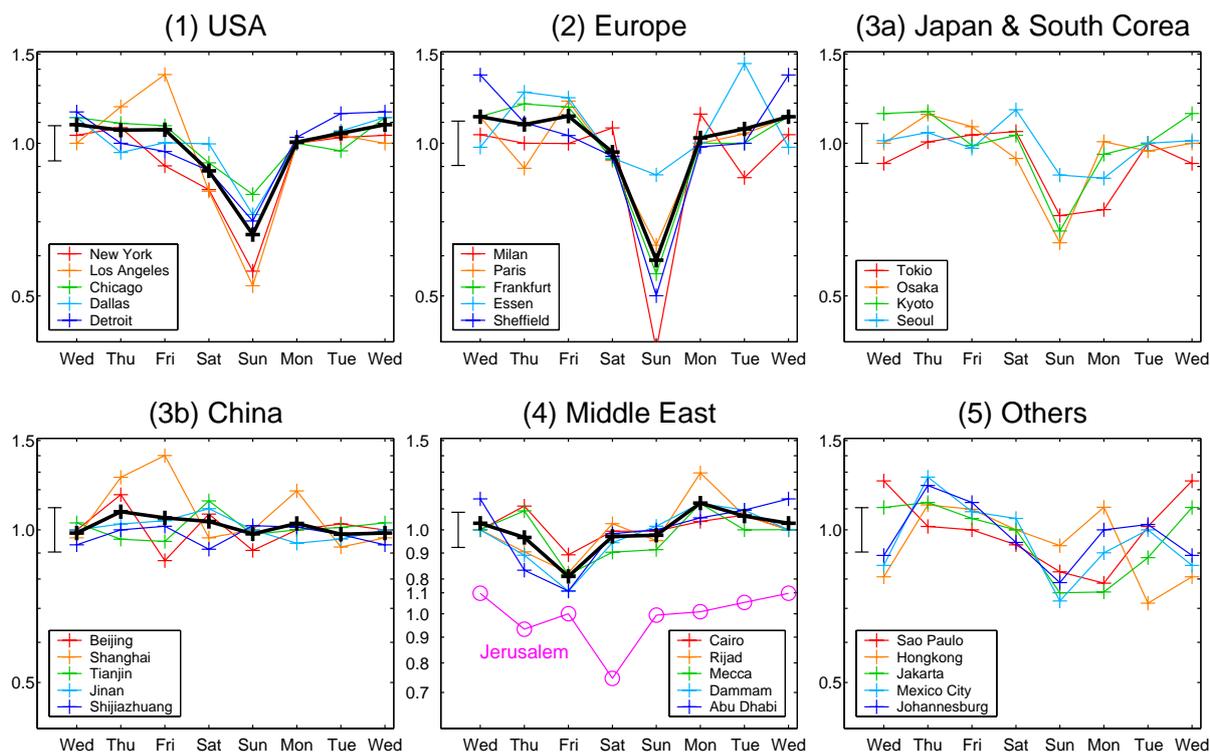
Fig. 3. Weekly cycle of mean (1996–2001) tropospheric NO$_2$ VCD for the considered locations (as marked in Fig. 1). The values are normalized with respect to the median weekly value (relative units). Black lines are averaged curves. The error bar to the left of every plot indicates the maximum relative error (standard error of the mean) of all data points. The scale is logarithmic to emphasize relative features.

(soil emissions, biomass burning, lightning) show no indication for a weekly NO$_x$ cycle.

The weekly cycle of tropospheric NO$_2$ VCD is visualized in two ways. Figure 2 shows maps of the tropospheric NO$_2$ VCD for the regions under consideration for the different days of the week (mean over 1996–2001). This provides information about the absolute source strength in the different regions. The effect of the 35 day periodicity of GOME is reflected by the stripe-like structures parallel to the ERS-2 flight direction.

In addition, Fig. 3 shows plots of weekly cycles for some selected cities to illustrate the relative reduction of the tropospheric NO$_2$ VCD during the day of rest. For each day of the week we calculated the mean (and its statistical error) of all GOME observations (1996–2001) containing the respective location. These values are normalized to the median weekly value for better comparison. (The median was chosen as reference in order to avoid a bias by the Sunday minimum: The mean of all days would shift from the normal weekday level towards the lower Sunday level, and consequently, the weekdays normalized to the mean would be higher than unity.) The normalisation allows to calculate relative errors. The error bar of each subplot is the maximum of all individual relative errors. We chose a logarithmic scale since we focus on relative changes in the weekly cycle. Furthermore, the length of the relative error bar is independent from the actual mean.

Both figures show the same general features: the level of NO$_2$ VCD is similar for the different working days, but there is a significant reduction of tropospheric NO$_2$ VCD during the weekend, especially on Sunday, for the US (1), Europe (2), and Japan (3a). In all cities considered in these regions, there is a minimum on Sunday. The NO$_2$ VCD is reduced by about 25–50% and even 60% (Milan (2)). A Sunday minimum can also be seen in Johannesburg and Mexico City (5).

In China (3b), no indications for a weekly cycle can be found in Figs. 2 or 3. China has a different economic, cultural, and religious background. Nevertheless, there is also a seven day week and a work free weekend. The absence of a weekend effect in our data probably indicates that the Chinese NO$_x$ emissions are dominated by power plants and heavy industry, operating throughout the week. On the other hand, in the US and in Europe individual transport appears to contribute significantly to the NO$_x$ sources.

The cities in the Middle East (4) show a shifted weekly cycle due to different religious traditions, i.e. other days of rest. In Israel, Saturday (Sabbath) is the day of lowest NO$_2$ VCD. In Islamic cities, there is a slight weekly effect with lowest emissions on Friday.

Besides the noticeable minimum on Sunday, there are two more days of interest in the weekly cycles of the western cities: a) Saturday, since it is also a working free day for most professions in western countries (reduced industrial activity), but a preferred day for shopping or short trips (possible
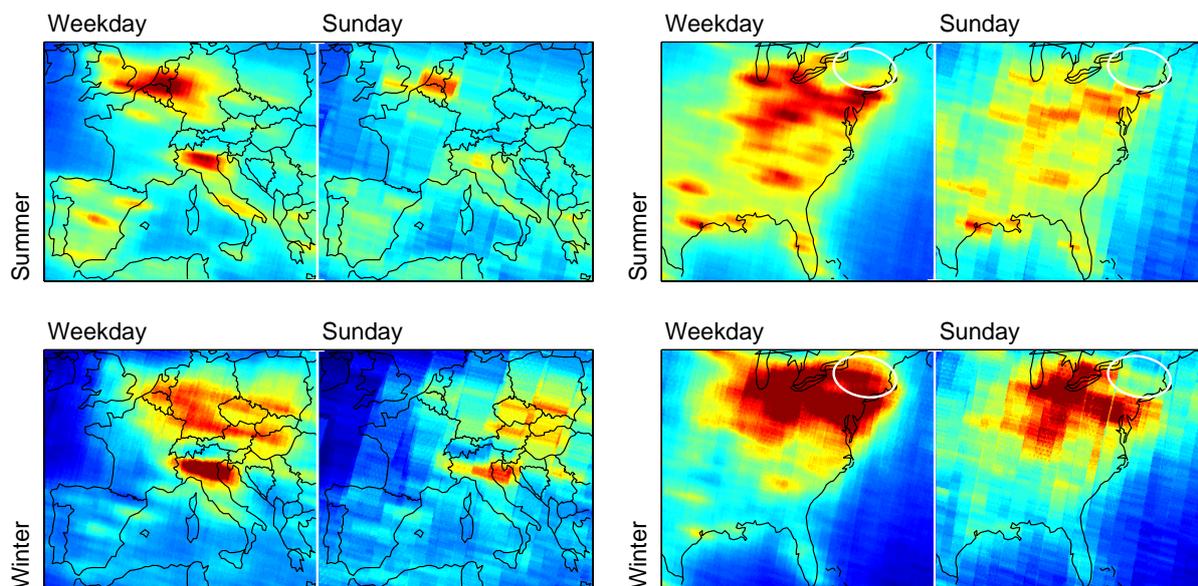
Fig. 4. Mean tropospheric NO_2 VCD (1996–2001) for Europe (left) and US east coast (right), separated for summer (upper row) and winter (lower row). The mean workday level is compared to the respective Sunday plot. Colour scale and unit are the same as in Fig. 1. The marked area in the northeastern US is discussed in the text.

increase of individual transport), and b) Monday, since it starts with relatively clean air, whereas for the other working days the measured air masses might be influenced by the pollution of the day before, as the lifetime of NO_x in the troposphere is about one day.

a) Saturday NO_2 values are slightly reduced in most western cities. But in some places they are quite high, reaching normal working day levels (e.g. Milan, Dallas, Tokyo).

b) Monday values in the US and Europe are almost on working day level, whereas there are uncommonly low levels in Tokyo and Seoul (3a) as well as in Sao Paulo and Jakarta (5).

To understand these features in the weekly cycle in detail, further information about local emissions, wind speed and direction, and lifetime of NO_2 is necessary. Another aspect is the daily cycle of emissions, especially the starting time of work in the morning which differs from region to region, whereas the local observation time of GOME is the same (about 10:30). A rather late start of human activity in the morning could possibly explain the Monday minimum in the weekly cycle for e.g. Sao Paulo. The fixed observation time also affects the level of the Sunday minimum, since on Sundays generally emissions start later (Sunday trips or shopping (in the USA) take place mainly after the GOME overpass).

The 35 day periodicity of the ERS-2 satellite flight track should be, in general, of minor influence. However, it is thought to be the most probable reason for some unusually high values, e.g. the Tuesday values in Essen or the high Wednesday values in Sheffield (Fig. 3(2)). For instance, in the Tuesday scans, the polluted regions of West Germany

(Ruhr area) and the Netherlands are largely covered by a single track of GOME pixels, whereas there are two neighbouring tracks on Wednesdays (see Fig. 2(2)). This leads to a higher observed NO_2 VCD on Tuesdays compared to Wednesdays. Also the relative high Sunday values in Essen seem to derive from this effect.

Additionally, we compared the weekday and Sunday levels of tropospheric NO_2 in Europe and the USA for summer and winter separately (Fig. 4). We observed a significant Sunday reduction for both regions and both seasons. In principle, a quantitative comparison of the weekly cycles for summer and winter should hold information about the role of heating (north-western Europe) or air-conditioning (USA). However, such a quantitative analysis seems to be not feasible yet:

– The reduction of data (3 months per year instead of 12) increases the statistical error by a factor of 2.

– Obviously, the stratospheric bias depends on season. Mainly in winter we retrieved negative tropospheric columns over the Atlantic e.g. around Ireland or east of Florida (Fig. 4, lower row).

– The lifetime of NO_x is higher in winter (see next chapter). This also influences the weekly cycle, i.e. the Sunday minimum (in winter there is more NO_x from Saturday remaining).

Nevertheless, Fig. 4 reveals some new insights. For instance, while the Netherlands/Belgium/Germany plume vanishes almost completely on Sundays in winter (Fig. 4, lower
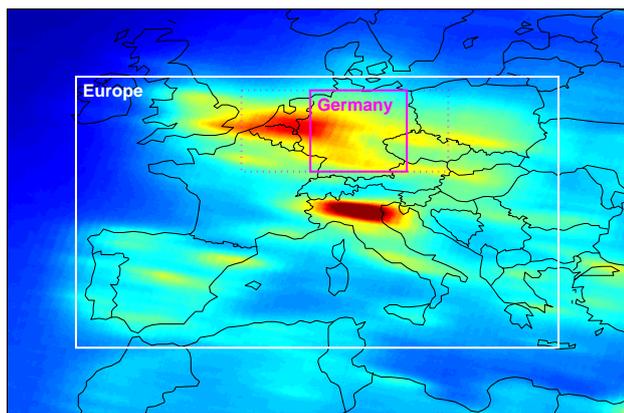
Fig. 5. Zoom of Fig. 1 on West Europe. For the lifetime estimation of tropospheric NO_x in Germany, the considered area is marked ($7°$–$14°$ E and $48°$–$54°$ N). The dotted box ($2°$–$17°$ E) indicates an alternative area for comparison (see text).

left panel), indicating transport as main source of NO_x, the VCD over Austria/Hungary/Czechia/Slovakia/Poland almost stays the same. This indicates that the predominant source of NO_x has no weekly cycle, i.e. is very probably heating. (Buildings in Eastern Europe are worse isolated, which leads to higher energy consumption for heating, Bayer et al., 2002).

Another quite astonishing result is the north-eastern region of the US between Lake Ontario and Boston (Fig. 4, marked ellipse, right panels): While this region is quite clean in summer (on weekdays as well as on Sundays), it is highly polluted on winter workdays, but much less on Sundays. This indicates a weekly cycle in NO_x — either due to source, visibility to GOME, or sink — not existing in summer. At present we do not understand this feature.

4   Lifetime estimation

The average lifetime of NO_x in the boundary layer depends on many factors like meteorological conditions, photolysis, length of night, temperature, OH and H_2O concentrations. It is of the order of one day, but varies by about one order of magnitude with time and place. Estimates of the mean lifetime for a given location and season reflect typical conditions. Furthermore, they are needed to relate NO_x emissions quantitatively to GOME VCDs. We use the weekly cycle to estimate mean lifetimes of NO_x in summer and winter for Germany exemplarily.

The principal sink of NO_x is oxidation to HNO_3 that is removed by wet or dry deposition in the troposphere (e.g. Jacob, 2003). During daytime, the conversion takes place by NO_2 directly reacting with OH radicals. At night, NO_3 is formed (by reactions of NO_2 with ozone), that itself reacts with NO_2 to N_2O_5. This product finally reacts heterogeneously with H_2O to HNO_3 (Geyer and Platt 2002). For

a mid-latitude located country like Germany, the removal in summer takes place by both mechanisms, while in winter it occurs almost solely by the second path due to low OH concentrations.

For our estimation, we analysed the influence of the low Sunday emissions on the VCD measured on Monday in the area downwind from the source regions. Therefore we consider Germany, i.e. the area $7°$–$14°$ E and $48°$–$54°$ N (Fig. 5). Since the main sources are located in the western part of Germany and the mean wind direction is eastwards, the emitted pollutants should remain within the area for at least one day. (A mean wind velocity of approx. 5 m/s corresponds to a daily transport of 432 km, whereas the east-west extension of Germany is about 700 km.) We based our analysis on some simple assumptions:

1. The ratio NO_2/NO_x does not depend on the day of week. That means, the normalized weekly cycles of NO_x and NO_2 are the same.

2. There is a constant NO_x emission during daytime (12 h), while there are no emissions at night. Saturday emissions are 66%, Sunday emissions 60% of working day level (adopted from Wickert, 2001), see Fig. 6a.

3. The chemical loss of NO_x is first order in NO_x characterised by a constant lifetime τ. We neglect possible changes of τ due to different VOC/Ozone concentrations on Sundays.

4. For the considered area, there is neither in- nor outflow of NO_x due to wind transport. By this approach we neglect the pollution in the Netherlands/Belgium in particular, that itself is subject to a weekly cycle. The error of this simplification was estimated by extending the box size (dotted rectangle in Fig. 5).

Therefore, the change of the NO_2 VCD can be described as

$$d(VCD_{NO_2})/dt = E(t) - (VCD_{NO_2})/\tau$$

$E(t)$: Emissions (in units of molecules $cm^{-2} s^{-1}$).

This differential equation was solved numerically. Figure 6b shows the retrieved average weekly cycle of German NO_2 VCD at the ERS-2 overpass time for different lifetimes τ. This is compared to the actual measurements of GOME in Fig. 6c. This comparison was performed for winter and summer data separately. The dotted lines in Fig. 6c are related to the extended area (dotted rectangle in Fig. 5). As can be seen, the actual weekly cycle is nearly unaffected by the extent of the considered area.

The Sunday reduction is underestimated in the modelled weekly cycle. This might indicate that the reduction of weekend emissions is actually more extensive than assumed. Besides, in assumption 2 (Fig. 6a) we neglected that Sunday
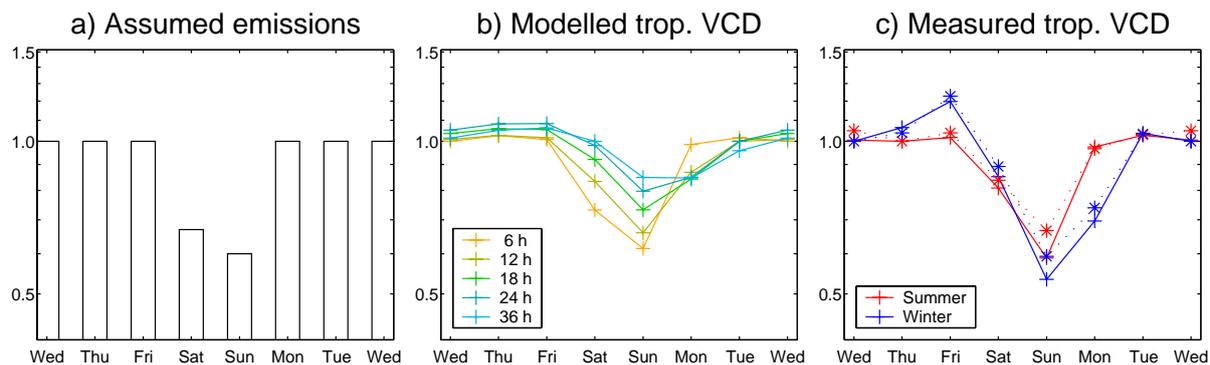
Fig. 6. Weekly cycle of NO$_2$ (modelled and measured) for Germany (as shown in Fig. 5). All plots are normalized to the median weekly value (relative units). **(a)** Assumed weekly cycle of NO$_x$ emissions (adopted from Wickert, 2001). **(b)** Modelled tropospheric VCD at ERS-2 overflight time for different lifetimes of NO$_x$. **(c)** Mean tropospheric VCD of NO$_2$ measured with GOME. Dotted curves refer to the dotted rectangle in Fig. 5.

emissions start later in the morning. Another important reason, especially for winter, is the overestimation of the stratosphere for the North Atlantic region. If we add a constant offset of, for instance, 10^{15} molec cm^{-2} (that would compensate the negative values over Ireland), the Sunday minimum would be 61% instead of 53%.

In any case, the detailed temporal pattern of the weekly cycle of the modelled VCD crucially depends on the assumed lifetime. (Fig. 6b). The weekly cycle of the summer data (Fig. 6c) shows Monday values on the normal workday level. Therefore, the summer weekly cycle is smaller than 12 h and in good agreement with a lifetime of 6 h (a shorter lifetime would result in Saturday values lower than observed, whereas a longer lifetime would show reduced Monday values). In winter, the observed Monday values are lower than the Saturday ones, indicating a lifetime of about 18–24 h (any lifetime longer than 36 h would lead to Monday values lower than Sunday values, which is not observed). Our results are quantitatively in good agreement with the GEOS-CHEM calculations presented by Martin et al., 2003. The lifetime of Germany NO$_x$ in summer is significantly shorter than in winter as expected. The winter result of 18–24 h can serve as estimate for the removal via NO$_3$/N$_2$O$_5$, since OH concentration is quite negligible in winter.

5 Conclusions

The existence of a weekly cycle is an obvious signature of anthropogenic sources. In most western industrialised areas a clear weekly cycle with a noticeable Sunday minimum can be found in tropospheric NO$_2$ VCD from GOME, adding credibility to the retrieval of tropospheric trace gas measurements from space. Regions with a different religious background show a shifted weekly cycle with minimum NO$_2$ VCD on Saturday (Israel) or Friday (cities with predominantly Islamic population). In China, no weekly cycle of tropospheric NO$_2$ could be found.

The low level of the Sunday minimum in the western cities provides information about the fraction of periodic anthropogenic sources of NO$_x$. In principle, the analysis of the weekly cycle (for different seasons) could help to discriminate manmade from natural sources (lightning, biomass burning, soil emissions) and assess the contribution of local traffic to the NO$_x$ sources.

The characteristics of the weekly cycle, i.e. the ratio of the Monday values and Sunday minima, allow a rough estimate of the mean lifetime of boundary-layer NO$_x$. We find a mean lifetime of 6 h (summer) and 18–24 h (winter), respectively, for Germany.

The quantitative interpretation of features of the weekly cycle requires background information about the considered regions. Weekly as well as daily patterns of emissions, the predominant source of NO$_x$ (traffic, heavy industry or heating) and the lifetime of tropospheric NO$_x$ differ for different levels of industrialisation, religious and cultural customs and meteorological conditions.

The statistical errors of the data points in Fig. 3 are quite small due to the large amount of data. Systematical errors due to the estimation of the stratospheric column and the large spatial extent of the GOME pixels, as described in Sect. 2, have to be borne in mind. A better spatial resolution (nominal 30 × 60 km^2) will be provided by the SCanning Imaging Absorption SpectroMeter for Atmospheric CHartographY (SCIAMACHY) onboard the ENVISAT, which is presently in its validation phase. Furthermore, due to the additional limb viewing geometry a direct estimation of the stratospheric column is possible. Time series of SCIA-MACHY data as well as ongoing GOME measurements will allow the improved analysis of weekly cycles of tropospheric NO$_2$ and will afford quantitative estimates of different types of anthropogenic emissions as well as the lifetime of tropospheric NO$_2$ for different regions of the world.

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