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Long-term changes of tropospheric NO₂ over megacities derived from multiple satellite instruments

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Abstract

Tropospheric NO₂, a key pollutant in particular in cities, has been measured from space since the mid-1990s by the GOME, SCIAMACHY, OMI, and GOME-2 instruments. These data provide a unique global long-term data set of tropospheric pollution.

5 However, the measurements differ in spatial resolution, local time of measurement, and measurement geometry. All these factors can severely impact the retrieved NO₂ columns, which is why they need to be taken into account when analysing time series spanning more than one instrument.

In this study, we present several ways to explicitly account for the instrumental differences in trend analyses of the NO₂ columns derived from satellite measurements, while preserving their high spatial resolution. Both a physical method, based on spatial averaging of the measured earthshine spectra and extraction of a resolution pattern, and statistical methods, including instrument-dependent offsets in the fitted trend function, are developed. These methods are applied to data from GOME and SCIAMACHY separately, to the combined time series and to an extended data set comprising also GOME-2 and OMI measurements.

All approaches show consistent trends of tropospheric NO₂ for a selection of areas on both regional and city scales, for the first time allowing consistent trend analysis of the full time series at high spatial resolution and significantly reducing the uncertainties of the retrieved trend estimates compared to previous studies.

20 We show that measured tropospheric NO₂ columns have been strongly increasing over China, the Middle East, and India, with values over East Central China tripling from 1996 to 2011. All parts of the developed world, including Western Europe, the United States, and Japan, show significantly decreasing NO₂ amounts in the same time period. On a megacity level, individual trends can be as large as $+27 \pm 3.7 \text{ \% yr}^{-1}$ and $+20 \pm 1.9 \text{ \% yr}^{-1}$ in Dhaka and Baghdad, respectively, while Los Angeles shows a very strong decrease of $-6.0 \pm 0.37 \text{ \% yr}^{-1}$. Most megacities in China, India, and the Middle East show increasing NO₂ columns of $+5\text{--}10 \text{ \% yr}^{-1}$, leading to a doubling to

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triplication within the observed period. While linear trends derived with the different methods are consistent, comparison of the GOME and SCIAMACHY time series as well as inspection of time series over individual areas shows clear indication of non-linear changes in NO_2 columns in response to rapid changes in technology used and the economical situation.

1 Introduction

During the first decade of the 21st century, the Earth's population living in cities, which had been growing since the industrial revolution and the birth of the anthropocene, reached over 50 % (United Nations, Department of Economic and Social Affairs, Population Division, 2012). This development is closely related to increasing growth rates of megacities, large urban agglomerations of more than 10 000 000 inhabitants, and of conurbations generally. The resulting high traffic, energy use, and industrial production make them hot-spot areas in terms of pollution (Molina and Molina, 2004).

One of the most important air pollutants in the troposphere is nitrogen dioxide (NO_2). It participates in a catalytic chain reaction producing ozone (O_3), is a key precursor of nitric acid (HNO_3) and acid deposition, and is directly hazardous to human health (Finlayson-Pitts et al., 1999; World Health Organization, 2003). Its main source is the emission of nitrogen monoxide (NO) from the anthropogenic combustion of fossil fuels (Noxon, 1978); natural sources include biomass burning, microbial activity in soils (Williams et al., 1992), which following the development of the Haber Bosch process and its use in the use of fertilisers has been modified by anthropogenic activity, and lightning (Noxon, 1976).

NO_2 and NO are often, through their day time chemistry, coupled in the troposphere: NO reacts with O_3 to form NO_2 , which in turn is photolysed to form NO . The sum of NO and NO_2 is termed NO_x . Tropospheric NO_x has a relatively short lifetime of several hours to few days (Finlayson-Pitts et al., 1999). As a result, NO_2 has its largest

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concentrations in the boundary layer close to the emission sources, making measurements of NO_2 columns well suited to infer NO_x emissions.

The strong absorption lines of the NO_2 molecule in the visible wavelength range of the spectrum facilitate the use of optical absorption spectroscopy for measuring atmospheric NO_2 abundances. Nadir viewing instruments have been deployed on satellite platforms since the mid-1990s. This has resulted in the global monitoring of NO_2 concentrations under consistent measurement conditions (Burrows et al., 2011). For measurements of tropospheric NO_2 , the nadir-viewing Global Ozone Monitoring Experiment (GOME, on board ERS-2; Burrows et al., 1999), the SCanning Imaging Absorption spectroMeter for Atmospheric CHartography (SCIAMACHY, on board ENVISAT; Bovensmann et al., 1999; Burrows et al., 1995, and references therein), the Ozone Monitoring Instrument (OMI, on board AURA; Levelt et al., 2006), and GOME's operational meteorological successor GOME-2 (on board Metop-A¹; Callies et al., 2000) provide measurements of the upwelling radiance suitable for retrieval of tropospheric NO_2 . These four instruments provide a continuous time series of tropospheric NO_2 measurements beginning in August 1995. The measured spectra are often analysed using differential optical absorption spectroscopy (DOAS; Platt and Stutz, 2008), and scientific data products of tropospheric NO_2 exist since the early 2000s (among others: Richter and Burrows, 2002; Leue et al., 2001; Martin et al., 2002; Boersma et al., 2007; Richter et al., 2005; Boersma et al., 2004; Richter et al., 2011). Long-term changes in tropospheric NO_2 amounts can thus be investigated by combining measurements from several of these four instruments.

Whilst over large areas of relatively homogeneous NO_2 signals coincident GOME and SCIAMACHY observations agree very well (Richter et al., 2005), they show considerable differences for ground scenes with steep gradients in the tropospheric NO_2 columns. This is the result of spatial smoothing, which differs depending on the ground

¹To ease notation, we will use the name “GOME-2” as shorthand for the GOME-2 instrument on board Metop-A, as the successor satellites Metop-B and Metop-C have not been launched before the end of our study period.

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5 resolution of the instruments. A schematic diagram illustrating this resolution effect is shown in Fig. 1, and Fig. 2a, b show mean $VCD_{\text{trop}} \text{NO}_2$ from GOME and SCIAMACHY measurements as an example. SCIAMACHY measurements show considerably more spatial detail and yield higher NO_2 columns over pollution hot-spots than GOME measurements. In summary, the inherent spatial heterogeneity of the NO_x fields must be considered when investigating the temporal evolution of $VCD_{\text{trop}} \text{NO}_2$ over small, localised areas.

10 There have been several studies of the temporal evolution of tropospheric NO_2 . The first reports on systematic changes have been published by Richter et al. (2005) and Irie et al. (2005). Konovalov et al. (2006) mathematically convoluted SCIAMACHY measurements, gridded to $1^\circ \times 1^\circ$, to calculate correction factors by which GOME measurements were multiplied, yielding a consistent combined GOME/SCIAMACHY dataset. van der A et al. (2008) fitted a non-linear trend function to ten years of GOME and SCIAMACHY data from megacity regions, modelling the seasonal variation by using one sinus component, and ignoring the apparent offset between the two instruments. 15 Nine years of SCIAMACHY measurements were analysed by Schneider and van der A (2012), focussing on China and Europe on a country level. Stavrakou et al. (2008) used measurements from GOME and SCIAMACHY to invert anthropogenic NO_x emissions, gridding the measurements to the coarse resolution of the used IMAGES model (5° × 5°). Konovalov et al. (2010) applied non-linear trend analysis to 13 yr of summer measurements from GOME and SCIAMACHY of European urban centres, where SCIAMACHY measurements were convoluted to the spatial resolution of GOME. Zhou et al. (2012) performed an extensive study on the impact of annual and weekly cycles and of meteorology on observed NO_2 columns over Europe using a generalized additive regression model. Several further studies have been published, focussing on 25 certain regions of the world like China (He et al., 2007; Zhang et al., 2009a, 2007), India (Ghude et al., 2008), the megacity Moscow (Sitnov, 2010), coal power stations in the United States (Kim et al., 2006), and international shipping lines (Richter et al., 2004; Franke et al., 2009). Recent studies have been focussing on the impact of NO_x

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emission reductions due to the global economic crisis and air quality legislation in the United States (Russell et al., 2012), Europe (Castellanos and Boersma, 2012), Greece (Vrekoussis et al., 2012), China (Lin and McElroy, 2011), and for shipping emissions (de Ruyter de Wildt et al., 2012).

5 In the present study, we provide an update on the global NO₂ trends reported in Richter et al. (2005), propose two alternative methods of handling the issue of different spatial resolution between GOME and SCIAMACHY measurements, and report on NO₂ trends for a number of megacities and large urban agglomerations. The manuscript is structured as follows: in Sect. 2, we describe the dataset of VCD_{trop} NO₂ which we used for this study. A general overview of the changes in tropospheric NO₂ which can be deduced from GOME and SCIAMACHY measurements alone is given in Sect. 3. In Sect. 4 we introduce a physical correction method, which super-imposes the spatial patterns of SCIAMACHY measurements on GOME data to create a consistent high resolution data set. An alternative statistical way of deriving trend estimates from the combined dataset by explicitly accounting for the instrumental differences is presented in Sect. 5. This statistic trend model is expanded to include measurements from all four available instruments, as described in Sect. 6.

2 Measurements of tropospheric NO₂

20 The GOME instrument on board the European Research Satellite 2 (ERS-2) was a nadir viewing spectrometer measuring three pixels per forward scan and one backscan. For a 960 km swath width this was the measurement mode for 90 % of the operation; the spatial resolution was 320 km and 40 km in across and along track directions, respectively. 10 % of the time the instrument had a swath width of 240 km and ground scenes of 80 × 40 km². The satellite was on a near polar orbit, crossing the equator at 10:30 LT. Global coverage was obtained every three days at the equator (Burrows et al., 1999).

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The SCIAMACHY instrument on board the ENVISAT satellite measured with a swath width of 960 km. While the along track extents of the swath were 30 km, the typical integration time of 0.25 s lead to across-track extents of 60 km for the forward pixels, which we used in this study. ENVISAT circled the Earth on a near polar orbit, crossing the equator at 10:00 LT. SCIAMACHY attained global coverage every six days (Burrows et al., 1995; Bovensmann et al., 1999; Gottwald and Bovensmann, 2011).

The Ozone Monitoring Instrument on board the Aura satellite (OMI) is a wide-field-imaging grating spectrometer, providing daily global coverage at a nadir point horizontal resolution of $13 \times 24 \text{ km}^2$ since October 2004. The GOME-2 instrument on board the Metop-A satellite covers the Earth's surface with a horizontal resolution of $80 \times 40 \text{ km}^2$ with nearly global coverage every day.

All four instruments measure spectra of extraterrestrial solar irradiance and the upwelling earthshine radiance at the top of the atmosphere. For the GOME, SCIAMACHY, and GOME-2 instruments, these spectra are subsequently analysed using differential optical absorption spectroscopy (Platt and Stutz, 2008) in the 425–450 nm wavelength window (Richter and Burrows, 2002). Absorption cross sections for O₃ (Bogumil et al., 2003), NO₂ (Bogumil et al., 2003), O₄ (Greenblatt et al., 1990), and H₂O (Rothman et al., 1992) are included in the fitting process, as well as the infilling of Fraunhofer lines and molecular absorption known as the Ring Effect (Vountas et al., 1998), an under-sampling correction (Chance, 1998), and for GOME and SCIAMACHY, a calibration function accounting for the polarisation dependency of the instrument's spectral response (Richter and Burrows, 2002). For the determination of the tropospheric NO₂ column amount from the measurements of the OMI instrument, we use the slant columns from NASA's OMI/Aura Nitrogen Dioxide (NO₂) Total & Tropospheric Column1-orbit L2 Swath $13 \times 24 \text{ km}$ (Version 3) dataset. The differences between the four instruments are summarized in Table 1.

The stratospheric contribution to the measured total slant columns SCD_{tot} has been accounted for using simulated stratospheric NO₂ fields from the Bremen 3d CTM (B3dCTM). It is a combination of the "Bremen transport model" (Sinnhuber et al.,

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2003a) with the chemistry code of the “Bremen two-dimensional model of the strato-
sphere and mesosphere” (Sinnhuber et al., 2003b; Winkler et al., 2008), which evolved
from SLIMCAT (Chipperfield, 1999). The model is driven by ECMWF ERA-Interim me-
teorological reanalysis fields (Dee et al., 2011) and runs on a combined 30 min/15 min
5 time-scale for transport and chemistry, respectively. Vertical transport is included in the
dynamics using the advection scheme developed in Prather (1986). While the hori-
zontal resolution is constant at $2.5^\circ \times 3.75^\circ$, the vertical resolution varies from ~ 1 km
at the lower stratosphere to ~ 4 km at ~ 60 km. The model runs on 28 isentropic sur-
faces from 330 K to 3402 K (about 10–65 km). Stratospheric vertical columns VCD_{strat}
10 for each satellite measurement were interpolated in space and time using smoothing
spline and linear interpolation, respectively.

Stratospheric air mass factors AMF_{strat} have been calculated with the radiative trans-
fer model SCIATRAN (Rozanov et al., 2005) using the modelled NO₂ profiles and con-
sidering the temperature dependence of the NO₂ absorption cross section by using
15 ECMWF ERA-Interim temperature fields (Dee et al., 2011). These AMF_{strat} have been
utilised to convert the modelled VCD_{strat} to SCD_{strat} . Before subtracting these modelled
 SCD_{strat} from the measured SCD_{tot} , an additive offset is applied to correct for sys-
tematic biases between the model simulations and the satellite measurements. This
matching is performed over the Pacific Ocean (180° W– 150° W), where the tropospheric
20 contribution to the total measured SCD_{tot} is estimated using a 10-yr climatology of
 VCD_{trop} taken from the Oslo CTM2 model, which has been converted to slant columns
using tropospheric air mass factors (AMF). The process of stratospheric correction is
described in detail in Hilboll et al. (2012).

Tropospheric AMFs have also been calculated with the radiative transfer model SCI-
ATRAN (Rozanov et al., 2005). The vertical distribution of tropospheric NO₂ has been
25 taken from a climatology of NO₂ mixing ratios from the MOZART2 model (Horowitz
et al., 2003), and surface spectral reflectance from GOME measurements (Koelemeij-
er et al., 2003). Both aspects are explained in detail in Nüß (2005). The AMFs have
then been spatially interpolated to a 0.125° grid. Measurements with a cloud coverage

exceeding 20 % have been filtered out using the FRESCO+ algorithm (version 6; Wang et al., 2008). Additionally, we applied an intensity filter to discard scenes with very large surface reflectivity. This is necessary as the used albedo or surface spectral reflectance climatology (Koelemeijer et al., 2003) does not account for short-term changes in reflectivity for example from snow; in addition, the FRESCO+ cloud fractions have large uncertainties over bright surfaces.

3 Changes in tropospheric NO₂ as observed by GOME and SCIAMACHY

When concatenating annual mean VCD_{trop} NO₂ from GOME (1996–2002) and SCIAMACHY (2003–2011) measurements into one 16-yr time series, many areas of the World show persistent changes. Figure 3 shows this for seven selected regions², the time series are all normalized to the 1996 value to make relative changes comparable. This figure extends the approach used to determine Fig. 1 in Richter et al. (2005), but uses a slightly updated data analysis (e.g. FRESCO+ v6 cloud screening for both instruments, a more recent version of the stratospheric correction model, and a higher spatial resolution of the gridding). All the selected spatial regions show persistent trends, which are downward in case of Western industrialized countries (United States, Europe, and Japan), and upwards for countries with developing economies like China, India, and in the Middle East. East Central China, for example, has seen a tri-
plication of tropospheric NO₂ columns over the whole time period, while NO₂ values over the central East coast of the United States have receded by more than 40%. These changes are explained by two behaviours, which have opposite impacts: continuing success in the reduction of NO_x emissions, in particular from cars and power plants, and increasing emissions from intensified use of the combustion of fossil fuels

²The North Central Indian industrial region extending between the cities of New Delhi and Lahore lies partly within an area for which GOME measurements are not available because of platform calibration made by ERS-2 in this region. Therefore, this region had to be selected quite small.

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for energy. In rapidly developing regions such as China, any reductions in specific emissions are overwhelmed by the absolute increase of energy use.

It is important to note, however, that even though the selected areas are all quite large, some of them do seem to show signs of the change in instrument between GOME and SCIAMACHY, as the value for 2003 seems to be sticking out.

The regional trends in satellite observed NO₂ columns are compared to EDGAR NO_x emission data (European Commission, Joint Research Centre (JRC) and Netherlands Environmental Assessment Agency (PBL), 2011) integrated over the same regions (Fig. 4). First comparison shows that the sign of direction of changes is the same for all regions in EDGAR and satellite observations. While generally, the trend observed in the measured NO₂ columns is more pronounced than in the EDGAR emission inventory, this is not the case for the Middle East and North Central India, which show smaller trends in the data than in the inventory. For East Central China, it is interesting to note that until 2002, the inventory does not significantly increase. Only from 2003 onward, it shows a strong positive trend. This is in contrast to the satellite observations, which show increasing NO₂ values over East Central China from the start of the observation period in 1996 onwards. At the same time, the temporal variability of the NO_x emissions is considerably lower than that of the observed NO₂ columns. This could either be the result of measurement uncertainties, mostly related to year-to-year changes in spatial and temporal sampling of the observations, or indicate inappropriate or inadequate assumptions in the bottom-up inventory. One possible issue in the direct comparison of emission inventories and satellite measurements might be found in the way emission inventories are compiled. Often, country-level emission estimates are spatially redistributed based on activity data. In regions where only limited reliable activity data are available, this might lead to inaccuracies in the spatial distribution of the emissions. On the other hand, changes in NO₂ columns and NO_x emissions are not necessarily related to each other in a linear way, particularly if changes are large (Stavrakou et al., 2008; Lamsal et al., 2011; Konovalov et al., 2010). In spite of the qualitative agreement between emission inventory and satellite observations, the unusual behaviour of

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bottom-up emissions in China before 2003 and the discrepancy between the onset of the consequences of the development in China and India apparent in the measurements but not the inventory need further investigation to identify unambiguously their origin.

5 In many regions, NO_2 pollution remains a local problem, as is apparent from plotting histograms of VCD_{trop} . Figure 5 shows the temporal evolution of the histograms for some selected regions for the SCIAMACHY time series (2003–2011). For all regions with the exception of East Central China, a large part of the area is characterized by low and moderate NO_2 levels, and only relatively small areas are affected by intense pollution. Comparing the distributions in different years, it is clearly visible that economic growth and emission control measures and the resulting increases and decreases in the observed VCD_{trop} impact not only on pollution hotspots, but on regional scales as a whole. Over East Central China, for example, the fraction of areas with exceptionally high NO_2 levels has been significantly increasing during the observing period, leading to a shrinking area which can be considered as being polluted at lower levels. Over the same period, the Central Eastern US have seen systematic increases in the fraction of low or moderately polluted areas.

4 Correcting for the difference between GOME and SCIAMACHY measurements based on physical principles

20 In order to create a consistent data set of NO_2 measurements at high spatial resolution, the difference in the area of the ground scene or spatial footprint of the measurements made by the GOME and SCIAMACHY instruments needs to be accounted for. Previous studies have either degraded the resolution of both data sets to a level where differences can be ignored or applied an ad hoc deconvolution of gridded GOME data (Konovalov et al., 2006). In this study, high spatial resolution SCIAMACHY spectra have been averaged to cover a similar area as individual GOME observations. Comparison of the high and low resolution analysis then provides an empirical correction

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function or factor, which can be applied to GOME data. For this, we make the assumption that the difference between GOME and SCIAMACHY measurements of VCD_{trop} only depends on the size difference between the two instruments' ground pixels and on season. This allows us to simulate measurements with GOME's spatial resolution using SCIAMACHY measurements by averaging over five adjacent spectra.

4.1 Method

Based on these assumptions, we calculate a monthly climatology of correction factors which describe the difference between the two instruments. To approximately match the GOME pixel size of $320 \times 40 \text{ km}^2$, we average the measured spectra from five adjacent SCIAMACHY forward scan ground pixels with an effective size of $300 \times 30 \text{ km}^2$, which yields three enlarged ground pixels per forward scan. On these enlarged ground pixels, we then perform a regular DOAS fitting procedure with the same settings as used for the regular retrieval to obtain a dataset of SCD_{tot} from reduced resolution. This dataset we further process in a fashion identical to that used for the original SCD_{tot} , which in the end yields a set of $VCD_{\text{trop}} \text{ NO}_2$ from SCIAMACHY measurements with reduced resolution.

This approach has the advantage of providing an appropriate end-to-end simulation of the effects of spatial resolution change, including the impact of change in cloud statistics on the fit, the specific orientation of the satellite ground pixels and the details of the a priori data bases used. The only issue which could not be fully simulated is the change in FRESCO+ cloud fraction, which was approximated by using the average of the individual cloud fraction values. As these values are only used for screening of cloudy scenes, the impact of this approximation is assumed to be small.

Both datasets (regular and reduced resolution) of VCD_{trop} are then gridded globally to a $\frac{1}{16}^\circ \times \frac{1}{16}^\circ$ grid and compiled into monthly averages for the months t' from January 2003 to December 2011. From these two monthly datasets, we calculate the climatological means $VCD^{\text{SCIA}}(t, \vartheta, \varphi)$ and $VCD^{\text{SCIA}}_{\text{red.res.}}(t, \vartheta, \varphi)$, where $t = 1, \dots, 12$ are the

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months from January to December, and ϑ and φ are latitudes and longitudes, respectively. From these monthly climatologies, we derive the climatological correction factors (ratios)

$$\Gamma(t, \vartheta, \varphi) = \frac{\text{VCD}^{\text{SCIA}}(t, \vartheta, \varphi)}{\text{VCD}_{\text{red.res.}}^{\text{SCIA}}(t, \vartheta, \varphi)}. \quad (1)$$

- 5 The gridded monthly averages of the GOME measurements are subsequently multiplied with the appropriate ratio grid of correction factors to yield the resolution-corrected GOME $\text{VCD}_{\text{trop}} \text{NO}_2$:

$$\text{VCD}_{\text{corr.}}^{\text{GOME}}(t', \vartheta, \varphi) = \Gamma(t(t'), \vartheta, \varphi) \times \text{VCD}^{\text{GOME}}(t', \vartheta, \varphi) \quad (2)$$

4.2 Spatial variation of Γ

- 10 The resolution correction factor Γ is a measure for the spatial heterogeneity of the observed NO_2 signal. Values of Γ larger than 1 indicate that typically observed NO_2 columns at a location are higher than in the surrounding area. Figure 6 shows the global distribution of the annual mean $\bar{\Gamma}(\vartheta, \varphi) = \frac{1}{12} \sum_t \Gamma(t, \vartheta, \varphi)$ of the resolution correction factor climatology. Over the open oceans, the signal of Γ is noisy. This is related
- 15 to the large relative errors of the very low measured $\text{VCD}_{\text{trop}} \text{NO}_2$ over the oceans, which are close to or well below the instrument's detection limit. Over land, however, and especially in regions with high spatial gradients in NO_2 emissions, such as the United States and Europe, the maxima in the gridded values for Γ clearly coincide with urban agglomerations or conurbations. A line of enhanced values of Γ is also visible
- 20 over the Atlantic Ocean from the tip of North-Western France to North-Western Spain. This coincides with a major shipping line, whose NO_2 emissions have previously been discovered in satellite imagery (Richter et al., 2004; Franke et al., 2009; de Ruyter de Wildt et al., 2012). Especially in the Western US, where NO_x emissions tend to be very sparsely distributed, local maxima in Γ can be attributed to individual sources like

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coal power stations. As it is to be expected, local minima of the correction factor are visible longitudinally adjacent to these maxima (see Fig. 7). These findings agree very well with those by Beirle et al. (2004), who applied a similar methodology to GOME measurements, using the instrument's narrow swath mode.

5 4.3 Validation of the resolution correction

In many cases, the resolution correction works extremely well. This can be evaluated using data from the period of overlapping measurements of the GOME and SCIAMACHY instruments (August 2002–June 2003). To reduce the impact of temporal sampling, only data points for which both instruments have measurements on the same day are included in the validation. For Europe, Fig. 2 compares original GOME and SCIAMACHY measurements to spatially down-sampled SCIAMACHY and resolution-corrected GOME measurements. The spatial distribution of GOME measurements is well approximated by the down-sampled SCIAMACHY measurements. Furthermore, the greater spatial detail inherent to the gridded SCIAMACHY measurements is obviously well transferred to the resolution-corrected GOME measurements.

A more detailed comparison over pollution hot spots is provided in Fig. 8, which shows the monthly mean values of $VCD_{\text{trop}} \text{NO}_2$ for the four cities Mumbai, Seoul, Mexico City, and Rome³. While in the direct comparison, there are differences of up to a factor of two, most of the corrected GOME data agree with SCIAMACHY data within 10%, independent of season. Quite often, however, the correction method actually overshoots, which leads to unreasonably high $VCD_{\text{trop}} \text{NO}_2$ for the corrected GOME measurements, especially over urban areas. As an example, the four cities Houston, Baghdad, Cairo, and Karachi are shown in Fig. 9. When performed over larger areas, a small over-correction is also observable in some months (see Fig. 10).

³Depending on the extent of the urban sprawl, the individual city regions have been defined as rectangles with edge lengths between 0.5° and 1.0°.

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In summary, one can conclude that the resolution correction method proposed here produces valid results in most cases when investigating time series of $VCD_{\text{trop}} \text{NO}_2$ over urban agglomerations and conurbations. Nevertheless the lack of a quantitative measure for the goodness of the resolution correction results in the provision of quantitative trend analysis with robust error estimation challenging. This issue will be addressed in Sect. 5.

4.4 Discussion

The apparent overcorrection in some situations is initially at least unexpected, as from the approximations made, one would expect the correction to underestimate the resolution effect, because the SCIAMACHY pixels with reduced resolution still only cover about 70 % of the ground area of a GOME measurement. There are several possible reasons for the observed differences: a problem with the approach used for resolution correction, a bias in one or both of the data sets, or real NO_2 differences, for example from the different overpass times of the two satellites having different emissions.

For most investigated areas, down-sampled SCIAMACHY measurements agree extremely well with original GOME measurements (see Figs. 8–10), showing that the derived resolution correction factor Γ should in principle well capture the differences between the two instruments which are due to their respective spatial resolutions.

As a next check, we investigated whether DOAS analysis of the spatially averaged SCIAMACHY spectra actually yields the same mean $SCD_{\text{tot}} \text{NO}_2$ as obtained from the regular measurements. The results for 4 exemplary orbits from the year 2011 are shown in Table 2. We chose the open Pacific Ocean for the first test because in this pristine region, we expect spatially homogeneous NO_2 measurements. As the measured slant columns agree almost perfectly in most cases, we conclude that for homogeneous scenes, the spatial averaging of the measured spectra does not influence the magnitude of the retrieved VCD_{trop} .

When repeating the same test for regions affected by the over-correction, e.g. the relatively isolated pollution hot spot formed by the city of Cairo in Northern Egypt, we

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capture the impact of the different instrumental resolutions very well. After applying it to GOME measurements, combined time series of tropospheric NO₂ columns do not show instrumental differences any more in many cases (Fig. 11), allowing for the first time the evaluation of long-term NO₂ trends over local hot-spots.

5 Fitting a non-linear trend model accounting for the difference between GOME and SCIAMACHY measurements

5.1 Method

An alternative approach to evaluate the combined time series for temporal changes is to use a trend model which includes a parameter allowing for adjustments between the two data sets. Such a model is not based on modelling the physical process of spatial averaging but has the advantage of providing a more rigorous error assessment. In addition, the model is not limited to GOME and SCIAMACHY but can also be applied to data from other instruments for which a physical resolution correction is not easily possible. In order to account for the systematic difference between GOME and SCIAMACHY measurements, we follow the approach proposed by Mieruch et al. (2008) for the analysis of global water vapour trends. The method is based on the work of Weatherhead et al. (1998) and Tiao et al. (1990), and explicitly accounts for the instrumental difference with a levelshift parameter in the fitting procedure.

The trend model explicitly accounts for (a) a levelshift between the two instruments and (b) for a change in the amplitude of the seasonal variation. If brief, the time series of monthly averages of NO₂ measurements $Y(t)$ is described by the equation

$$Y(t) = \mu + \omega t + \delta U(t) + S(t) + N(t) \quad (3)$$

where μ is the VCD_{trop} measurement at time $t = 0$, ω is the monthly trend component, and t is the time in months since January 1996. δ is the levelshift between GOME and SCIAMACHY measurements occurring at time $t = T_0$ (which we set to January 2003),

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and $U(t)$ is the step function

$$U(t) = \begin{cases} 0, & t < T_0 \\ 1, & t \geq T_0 \end{cases} \quad (4)$$

The seasonal component $S(t)$ has the same shape (represented by harmonic functions) and varying amplitude for the two instruments, and is modelled by

$$S(t) = (1 + (\gamma - 1)U(t)) \sum_{j=1}^4 \left(\beta_{1,j} \sin\left(\frac{2\pi jt}{12}\right) + \beta_{2,j} \cos\left(\frac{2\pi jt}{12}\right) \right) \quad (5)$$

where the term $(\gamma - 1)U(t)$ describes the amplitude change of the seasonality component at the change between the instruments. $N(t)$, finally, is the noise component, i.e. the part of the time series which cannot be explained by the underlying model.

The trend estimator $\hat{\omega}$ is calculated in a two-step procedure: first, the noise components $N(t)$ are derived by solving Eq. (3) for those estimators $\hat{\mu}$, $\hat{\omega}$, $\hat{\delta}$, and $\hat{\gamma}$ which lead to minimal $N(t)$. After subtracting the seasonal component $S(t)$, which usually has negligible effect on the estimation of the other trend parameters (Weatherhead et al., 1998), the autocorrelations are being accounted for using a linear matrix transformation. Finally, linear regression is applied to derive the estimators $\hat{\mu}$, $\hat{\omega}$, $\hat{\delta}$, and $\hat{\gamma}$ (Mieruch et al., 2008).

For the estimation of the trend error, we assume the noise process $N(t)$ to be AR(1), i.e. autoregressive with lag 1. The autocorrelation in the noise is accounted for in the error calculation as detailed in Mieruch et al. (2008). The trend is assumed to be significant iff

$$P_{H_0}(|\hat{\omega}| > 2\sigma_{\hat{\omega}}) = \text{erf}\left(\frac{|\hat{\omega}|}{\sigma_{\hat{\omega}}\sqrt{2}}\right) > 95\% \quad (6)$$

with

$$\operatorname{erf}(x) = \frac{2}{\sqrt{\pi}} \int_0^x e^{-t^2} dt \quad (7)$$

the Gauss error function.

5.2 Comparison of resolution correction factor Γ and levelshift parameter δ

5 As both the resolution correction factor Γ and the levelshift parameter δ are measures for how large the tropospheric NO_2 content at a given point is compared to its surrounding area, the two datasets should show large similarities. Indeed, as shown in Fig. 12, the maps of Γ and δ are in very good agreement. For example, large urban agglomerations, as, e.g. Madrid, Paris, Moscow, and Istanbul, are clearly visible in both datasets.

10 As the two methods are unconnected and only depend on the satellite measurements, we conclude that both techniques are legitimate approaches to overcome the issue of varying pixel sizes when combining measurements from GOME and SCIAMACHY. However, the levelshift can also account for other, not resolution related differences between the two datasets.

15 If the resolution correction method described in Sect. 4 worked perfectly well, the application of the levelshift trend model (Eq. 3) to resolution-corrected GOME measurements $\text{VCD}_{\text{corr.}}^{\text{GOME}}$ and SCIAMACHY measurements VCD^{SCIA} should yield a levelshift $\delta \equiv 0$ everywhere. In practice, the resolution correction only produces valid results when the actual tropospheric NO_2 column is considerably larger than the instrument's detection limit, as pointed out in Sect. 4.2. In Fig. 13, we compare the spatial distribution of the levelshift offset δ calculated for the original and the resolution-corrected GOME datasets. As expected, the original data show a characteristic pattern of positive and negative levelshifts δ over and adjacent to pollution hot spots, respectively, along the East and South coasts of the Japanese main island Honshu

20 and over South Korea. These patterns dissolve when the levelshift model is applied

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to resolution-corrected GOME measurements. However, the levelshift does not totally vanish. While over Japan and the Korean Peninsula, δ is reduced significantly and close to zero in the resolution-corrected dataset, the area around Shanghai in Eastern China still shows enhanced levelshifts of $10\text{--}14 \times 10^{15} \text{ molec cm}^{-2}$, which is about 40 % lower than in the original dataset. One possible explanation is the temporal changes in the spatial NO_2 patterns, which cannot be accounted for using the resolution correction factor (see Sect. 4.4). As it can be seen from Fig. 16, the largest changes in tropospheric NO_2 columns in the Shanghai area do not happen in the city itself, but rather North-West of Shanghai. Most of these changes are due to a steep increase in the years 2002–2005. This results in a decrease of the spatial NO_2 gradient over time, and consequently, the resolution correction factor calculated from the 2003–2011 climatology is too low for the stronger spatial gradients of the 1996–2002 period. As a result, a clearly visible gap between GOME and SCIAMACHY measurements remains in the time series for Shanghai, which results in the large levelshift which can be observed.

5.3 Global distribution of tropospheric NO_2 changes

The levelshift model has been applied to the combined dataset of GOME (January 1996–December 2002) and SCIAMACHY (January 2003–December 2011) measurements. The results show strong and significant changes in tropospheric NO_2 columns for large areas in North America, Europe, the Middle East, China, Japan, and India (see Fig. 14). While Western countries have experienced strongly decreasing $\text{VCD}_{\text{trop}} \text{NO}_2$ during the past 16 yr, developing countries like India, China, and in the Middle East, show strongly increasing NO_2 columns. Over the United States, large decreases can be observed for the Los Angeles metropolitan area and for large parts of the Eastern US. The areas with the largest reductions coincide with those areas already shown to be strongly affected by power plant emission reductions (see Kim et al., 2006). In the Middle East, the large increases are limited to large urban agglomerations, like the cities of Dimashq, Baghdad, Kuwait, Tehran, Isfahan, Riyadh, Jeddah, Cairo, Doha,

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and Dubai. For the regions defined in Fig. 3, the results are summarised in Table 3. All these observations are in good agreement with the general findings presented in Sect. 3, and are backed by Granier et al. (2011), who report according trends in NO_x emissions in all major emission inventories.

The results from the combined GOME/SCIAMACHY dataset can be compared to trends which are derived from GOME or SCIAMACHY measurements alone, as shown in Fig. 15. Obvious differences can be identified for North America, Europe, and China. Over the East coast of the US, the largest annual decreases in tropospheric NO_2 columns can be observed in the SCIAMACHY time series. In Europe, the GOME time series shows considerably larger decreases in $\text{VCD}_{\text{trop}} \text{NO}_2$ than both the SCIAMACHY and the combined time series. At the Chinese East coast, finally, the area showing large annual increases is considerably larger in the SCIAMACHY period than in the GOME time series. This observation is consistent with the observed increase in the fraction of highly polluted areas in China (see Fig. 5), and is backed by a comparison of absolute to relative changes, as shown in Fig. 16. While in the absolute changes, the well known area of East Central China shows the largest upward trend, the spatial distribution of the relative changes is more homogeneous, and the largest relative increases are actually seen at the margins of the area with highest NO_2 columns. Large annual increases of 10 % and more can be observed in extended areas. Apart from the Chinese coastal area, which hosts the bulk of the nation's economic activities and is known for its high levels of air pollution, other regions show large relative trends as well. Especially in Inner Mongolia and Xingjiang, and in the Ningxia, Shaanxi, and Gansu regions of central China, large relative growth rates of tropospheric NO_2 columns can be observed. As these areas still show relatively low $\text{VCD}_{\text{trop}} \text{NO}_2$, they do not stand out in the absolute annual increases.

Another interesting aspect in Fig. 16 is the large increase of NO_2 over the Yellow Sea between China and Korea. This NO_2 is attributed to result from transport of air from the polluted regions on land and gives an idea of the impact of transport on the observed NO_2 fields with impacts on pollution levels in countries downwind of China.

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Any increases in emissions from shipping also need to be considered. It is noteworthy to also point out that the effect of the transport of air pollution also limits the spatial resolution of the satellite NO₂ maps, in particular in winter.

The differences between the trends in the GOME and SCIAMACHY time periods indicate an overall problem with the assumption of linear trends in NO_x emissions and NO₂ columns. As emissions are controlled by technological, political, and economic factors, rapid, non-linear changes have to be expected. This issue has already been raised by Konovalov et al. (2010), but for most regions, the length of the time series in combination with the uncertainties of the individual values does not yet allow for reliable detection of the non-linear component of the observed changes. However, for individual time series over selected regions (i.e. Los Angeles and Athens), the non-linearity is obvious and has to be taken into account when interpreting the data.

5.4 VCD_{trop} NO₂ over megacities

The levelshift trend model facilitates the investigation of the development of individual pollution hot spots' NO₂ abundances. As it does not require the spatial down-sampling needed in previous studies (van der A et al., 2008; Konovalov et al., 2010), accurate estimation of the temporal evolution of NO₂ pollution of localized sources becomes feasible. We summarise the trend estimates for a variety of megacities in Table 4. The resulting NO₂ trend is comparable using original and resolution-corrected GOME measurements in the levelshift fit; in virtually all cases the absolute growth rates between the two datasets agree within the assumed uncertainty. The respective relative trends, however, differ considerably. This results from the resolution-corrected GOME data in the reference year 1996 having considerably higher NO₂ columns than in the original dataset. Exemplarily, time series for New York, Tehran, Mumbai, and Beijing are shown in Fig. 17.

For all four cities, the gridded VCD_{trop} NO₂ (on a $\frac{1}{16}^\circ \times \frac{1}{16}^\circ$ grid) have been averaged over an area of $1.0^\circ \times 0.5^\circ$ ($1.0^\circ \times 0.75^\circ$ in the case of Mumbai). In all cases, it is clearly visible that (a) there are significant differences between GOME and SCIAMACHY

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measurements which must not be ignored, and that (b) these differences are being addressed by the levelshift method very well. New York shows a strongly decreasing trend of $-5.3 \pm 1.7 \times 10^{14} \text{ molec cm}^{-2} \text{ yr}^{-1}$. This is considerably stronger than the $-0.3 \pm 1.7 \times 10^{14} \text{ molec cm}^{-2} \text{ yr}^{-1}$ reported by van der A et al. (2008) for the 1996–2006 time period. However, one has to note that van der A et al. did not account for instrumental differences between GOME and SCIAMACHY measurements. Figure 17 shows that VCD_{trop} measured by SCIAMACHY are significantly higher than those retrieved from GOME measurements, which leads to an underestimation of a downward trend if not accounted for. Tehran, Mumbai, and Beijing all show very pronounced upward trends of $+2.1 \pm 0.68 \times 10^{14} \text{ molec cm}^{-2} \text{ yr}^{-1}$, $+8.2 \pm 1.7 \times 10^{13} \text{ molec cm}^{-2} \text{ yr}^{-1}$, and $+8.8 \pm 2.5 \times 10^{14} \text{ molec cm}^{-2} \text{ yr}^{-1}$, respectively. These results compare well to those reported by van der A et al. (2008), who reported $+2.6 \pm 0.5 \times 10^{14} \text{ molec cm}^{-2} \text{ yr}^{-1}$, $+7 \pm 3 \times 10^{13} \text{ molec cm}^{-2} \text{ yr}^{-1}$, and $+12 \pm 2.9 \times 10^{14} \text{ molec cm}^{-2} \text{ yr}^{-1}$. The larger trends by van der A et al. (2008) for Tehran and Beijing are probably again caused by their ignoring the offset between GOME and SCIAMACHY measurements, as the jump in NO_2 values between the two instruments biases a standard trend model towards higher growth rates. Generally, we observe a convincing reduction of the trend uncertainties by using our levelshift model, as compared to the estimates presented in van der A et al. (2008), which leads to significant trends in most of the regions considered in this study.

Interestingly, the cities Guangzhou, Shenzhen, and Hong Kong in South-Eastern China do not show significant trends. While this might be expected in Hong Kong, which industrialised earlier and has an advanced level of economic development, it seems unlikely that NO_2 pollution in Shenzhen, whose population has more than doubled since 1995 (O'Donnell, 2011), has not increased. This lack of a detectable trend might be caused by unfavourable temporal sampling, due to high cloud cover, resulting in only very few measurements per month, which leads to large individual errors of the monthly average NO_2 columns, or possibly by some control measures which counteract the emissions from a rapidly increasing population.

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6 Non-linear levelshift-like model combining all four instruments

As measurements of tropospheric NO₂ from different satellite instruments are becoming available, it is necessary to find ways to analyse these data in an integrated consistent and consolidated manner. The levelshift model presented in Sect. 5 has the limitation that it can only account for one measurement per time step. Therefore, we evolved the method to be able to use measurements from all four available instruments by developing a non-linear trend model accounting for the differences in the measured VCD_{trop} from GOME, SCIAMACHY, OMI, and GOME-2 (Hilboll et al., 2013).

6.1 Method description

We model the time series $Y(t, i)$ of average NO₂ measurements made by instrument i in month t with the trend model

$$Y(t, i) = \mu_i + \omega t + \eta_i \times \sum_{j=1}^4 \left(\beta_{1,j} \sin \left(\frac{2\pi j t}{12} \right) + \beta_{2,j} \cos \left(\frac{2\pi j t}{12} \right) \right) + N(t, i) \quad (8)$$

ω is the linear trend component, which we assume to be common among all four instruments. μ_i are the offsets of the linear trend per instrument, and η_i gives the relative amplitude of the seasonality component (with $\eta_1 \equiv 1$). The optimal trend estimators ($\hat{\omega}, \dots$) are then calculated by minimizing the sum of the squared noise components $N(t, i)$, applying weights to the individual monthly averages $Y(t, i)$ to account for the varying number of instruments providing VCD_{trop} at time t . It must be noted that as a result of the multi-instrument trend model analysing multiple observations per time step, the uncertainties and accurate error estimates of the estimated trends are challenging.

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6.2 Results and discussion

As is apparent from Tables 3 and 4, the linear trend estimates $\hat{\omega}$ from the multi-instrument fit generally agree very well with those derived from the levelshift model.

Many regions do not show large differences between the instruments (e.g. New Delhi), while in other regions, these differences are strongly pronounced, as in Tehran (see Fig. 18). Reasons for this include differing spatial distributions of both the NO₂ columns themselves and other factors influencing the NO₂ retrieval, as, e.g. aerosol load, surface altitude, and surface spectral reflectance. All these quantities influence the retrieved NO₂ columns, as the differing spatial resolutions of the satellite measurements lead to instrument-dependent spatial sampling, which in turn has a significant effect on the observed NO₂ amounts or levels and their seasonality. A further issue is the different local times of the satellite measurements. As OMI measures in the early afternoon, both the actual NO₂ concentrations and the influence of surface spectral reflectance (due to different solar zenith angles) and aerosol load (due to different boundary layer height) can lead to offsets between the instruments.

This can result in varying ground amounts of retrieved NO₂ columns and varying seasonal cycles between the four instruments. For large ground scenes, the NO₂ columns will be more inhomogeneous than in small ones, leading to a stronger smearing of the high pollution peaks of, e.g., megacities. We show this exemplarily for the three cities New Delhi, New York, and Tehran, using topography (Hastings et al., 1998), population density (Center for International Earth Science Information Network (CIESIN), Columbia University, International Food Policy Research Institute (IFPRI), the World Bank, and Centro Internacional de Agricultura Tropical (CIAT), 2011), and NO_x emission (European Commission, Joint Research Centre (JRC) and Netherlands Environmental Assessment Agency (PBL), 2011) data (see Fig. 19). In New Delhi, which lies in a topographically flat region with homogeneously high population density and NO_x emissions, virtually no difference between the four instruments can be observed. Under these conditions, NO₂ can spread without barriers. The area around New York City

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is also topographically flat, but the NO_x emissions are mostly constrained to land, with the exception of shipping routes and aircraft corridors, land being only $\frac{2}{3}$ of the whole area. Therefore, emitted NO_2 can spread towards the ocean, leading to NO_2 column gradients within the observed area. This NO_2 gradient between megacity and open ocean leads to the noticeable impact of the instrument's pixel size on retrieved VCD_{trop} values. In the case of Tehran, emissions are mostly confined to the city's boundaries. Moreover, the emitted NO_2 cannot spread evenly throughout the area, because Tehran is bordered by the Alborz mountain range towards the North and East, leading to inhomogeneous NO_2 pollution in the observed area and thus to lower NO_2 columns in the case of large pixel sizes. Therefore, the NO_2 time series over Tehran show a very strong dependence on the instrument's spatial resolution.

The multi-instrument trend model presented in this section has the advantage of being suitable for the later inclusion of measurements from future satellite instruments. Under the assumption that the growth rate of the linear trend component is constant among all instruments, the model is therefore an excellent tool to assess the temporal evolution of the measured quantity. In the trend calculation, each month has the same weight, while at the same time a maximum of the available measurement data is included in the fitting procedure, and instrumental differences are accounted for to some extent.

6.3 Possible extensions to the multi-instrument trend model

As a result of the multitude of factors contributing to the magnitude and changes in tropospheric NO_2 column densities, it is well possible that change rates are not constant throughout the seasons. One way to account for this is to link temporal changes to the amplitude of the seasonality component of Eq. (8). The effect of such a model extension requires the introduction of a parameter ξ representing the rate of change of

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the seasonality component:

$$Y_i(t, i) = \mu_i + \omega t + (1 + \xi t) \times \eta_i \times \sum_{j=1}^4 \left(\beta_{1,j} \sin \left(\frac{2\pi j t}{12} \right) + \beta_{2,j} \cos \left(\frac{2\pi j t}{12} \right) \right) + N(t, i) \quad (9)$$

We find that the trend estimator $\hat{\omega}$ is of smaller magnitude than when the seasonality is kept at a fixed amplitude. The increasing (decreasing) NO₂ values are partly absorbed by a changing amplitude of the seasonality signal, which in turn leads to lower linear trend estimators $\hat{\omega}$.

In virtually all cases the signs of the linear and harmonic trend estimators $\hat{\omega}$ and $\hat{\xi}$ coincide. The observed magnitude of $\hat{\xi}$ however varies widely, depending on the region of interest. While for many regions the two estimators are similar in (relative) magnitude (e.g. Beijing, Hong Kong, New York, Po Valley), often the harmonic component $\hat{\xi}$ is significantly larger than $\hat{\omega}$, as, e.g. in Athens, Baghdad, Barcelona, and Cairo. As summer NO₂ values often show slower rates of change than winter values, this is averaged out when assessing the linear growth rates. The seasonal cycle however is strengthened by this effect, as the difference between summer and winter values becomes larger.

In a further step, the trend model needs to be extended to account for the different noise levels of the individual instruments' time series. This is common in climate time series analysis (see Mudelsee, 2010) and assures that instruments with more stable measurements have more weight in the fitting process. Usually, this is achieved by minimizing the variability-weighted squared noise components $\frac{N(t,i)}{S(i)}$ where $S(i)$ denotes the standard deviation of all measurements of instrument i . In practice, this common correction leads to a slight reduction of the retrieved linear trend estimates for many regions. As a result of the relatively high variability of SCIAMACHY measurements (when compared to OMI and GOME-2, probably caused by the significantly lower spatial coverage), the instrument has lower relative weight in the trend calculation. Since most regions show the strongest relative NO₂ changes for SCIAMACHY observations (see, e.g. Figure 20), the instrument's lower relative weight thus leads to a slight reduction

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of the calculated trend estimates. For the results shown in Tables 3 and 4, we do not apply this correction so as to make the results more comparable to those retrieved from the resolution correction factor and levelshift methods.

Finally, the uncertainties of the trend estimates $\hat{\omega}$ need to be calculated. As an analytic formulation of the error component would be extremely complex, this has to be achieved using bootstrap methods (Mudelsee, 2010).

7 Summary and conclusions

In the present study, we investigate the temporal evolution of tropospheric NO₂ columns retrieved from satellite observations during the 1996–2011 time period. For the first time, the instrumental differences between the used satellite sensors have been explicitly accounted for. To assess the robustness of this approach and the reliability of the derived linear changes or trends, we introduced two additional and complementary strategies for the derivation of instrumental and multiple instrumental trends in tropospheric NO₂.

Firstly, we spatially averaged SCIAMACHY spectra to be comparable in ground pixel size to GOME measurements, performed DOAS fits on these spectra, and calculated a resolution correction factor from these down-scaled and the regular SCIAMACHY measurements. These correction factors were shown to represent the spatial dimension of the NO₂ signal measured at a given point on the Earth, as they very well repeat the patterns observed in NO₂ emissions. The GOME measurements were then multiplied with the correction factors. While this can lead to an over-correction in some cases, we showed that generally, the corrected NO₂ time series from GOME measurements over individual city regions are brought into very good agreement with SCIAMACHY values during the time period of parallel measurements, facilitating trend analyses on a spatial resolution appropriate for SCIAMACHY observations.

Secondly, we applied a trend model similar to that developed by Mieruch et al. (2008) for the study of H₂O trends to the combined GOME/SCIAMACHY time series, which

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explicitly accounts for a spatially varying additive offset between the two instruments. The spatial pattern of these offsets was shown to be very similar to that of the resolution correction.

We calculated annual change rates in tropospheric NO₂ columns for a number of large urban agglomerations (see Table 4). Compared to previous studies, our method yields considerably lower uncertainties of the retrieved trend estimates, as changes in level and seasonality between the instruments are taken into account. This model was then evolved to be able to include measurements from GOME-2 and OMI, and to account for a trend in the amplitude of the seasonal cycle.

The results show for the period of observation that NO₂ columns over the industrialised countries (US, Europe, Japan, Australia) have been steadily decreasing, with significant trends of up to $-6\% \text{ yr}^{-1}$. On the other hand, Chinese and Indian megacities, as well as many urban centres in the Middle East, show very strong upward trends of up to $+20\% \text{ yr}^{-1}$. Trends calculated with data from all four instruments agree well with those derived from GOME and SCIAMACHY measurements alone, highlighting the consistency of the satellite observations in spite of differences in sampling, spatial resolution, and overpass time. On the other hand, trends derived from the GOME and SCIAMACHY time series independently show systematic differences. These are attributed to changes in emission trends between the two time periods, e.g. the accelerated development in China or the recent emission reductions in the US due to improved technology and economic crises.

These strong and significant changes in tropospheric NO₂ columns over megacities show the ongoing need for further instruments, which are able to continue appropriate measurements. When assessing the temporal evolution, it is imperative that instrumental differences are being considered. This is especially true for the upcoming Sentinel 5 Precursor mission, as the proposed TROPOMI instrument will have a very high spatial resolution of $9 \times 9 \text{ km}^2$. With the increasing length of the NO₂ time series available for analysis, the potential to understand the relationship between NO₂ emissions and their atmospheric abundances becomes better than ever.

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We show the “raw” NO₂ measurements from GOME (resolution-corrected), SCIAMACHY, OMI, and GOME-2 for some selected megacity regions in Fig. 20. Several interesting features can be observed from these plots. In Athens, for example, NO₂ columns seem to be increasing until about 2004, followed by some years of stagnation, which turn into a rapid downward trend 2008/2009 (Vrekoussis et al., 2012). This is attributed to the deep economic crisis affecting Greece since that time. Similarly, Chicago and Los Angeles both see downward trends, starting from stagnating and even increasing NO₂ columns before 2000, and leading to significant NO₂ decreases in following years, with the most recent years showing some sign of slowdown of the decreases. This can be attributed to both increasingly efficient emission control measures and a slowdown of economic activity, as has been reported by Russell et al. (2012).

In Beijing, summer minima in 2008 are clearly lower than in the preceding and following years. This effect has been linked to the 2008 summer Olympic Games and has been studied, e.g., by Mijling et al. (2009). Over Jeddah and Mexico City, the NO₂ columns derived from the OMI instrument are significantly lower than those from SCIAMACHY and GOME-2, especially during the summer minima. This might be an effect of the different measurement time, as OMI measures in the early afternoon as opposed to the morning for all other instruments. For Mexico City, Zhang et al. (2009b) studied the diurnal cycle of NO₂ surface concentrations, and showed that 10:00 LT morning values were about three times higher than 13:15 LT values.

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Wieters proved extremely helpful in setting up the B3dCTM model. The authors are grateful to Sebastian Mieruch, Manfred Mudelsee, and Igor Konovalov for valuable discussions on time series analysis, and thank Joana Leitão for giving valuable recommendations regarding the manuscript. Greet Maenhout from JRC proved very helpful in fixing the EDGAR emission database for 2008. GOME and SCIAMACHY radiances have been provided by ESA. OMI Level-2 data used in this study were acquired as part of the activities of NASA's Science Mission Directorate, and are archived and distributed by the Goddard Earth Sciences (GES) Data and Information Services Center (DISC). GOME-2/Metop-A Level-1 data have been provided by EUMETSAT. ECMWF ERA-Interim data were supplied by the "European Centre for Medium-Range Weather Forecasts" at Reading, UK. The service charges for this open access publication have been partially covered by the Deutsche Forschungsgemeinschaft (DFG).

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Table 1. Characteristics of the four satellite platforms used in this study.

| Instrument | Equator crossing | Global coverage | Availability period | Pixel size at nadir (km ²) |
|------------|------------------|-----------------|---------------------|--|
| GOME | 10:30 LT | 3 days | Oct 1995–Jul 2003 | 40 × 320 |
| SCIAMACHY | 10:00 LT | 6 days | Aug 2002–Apr 2012 | 30 × 60 |
| OMI | 13:30 LT | 1 day | Oct 2004– | 13 × 24 |
| GOME-2 | 09:30 LT | ~ 1 day | Jan 2007– | 40 × 80 |

Table 2. Relative difference of original SCIAMACHY measurements $SCD^{SCIA}(\vartheta, \varphi)$ and reduced-resolution SCIAMACHY measurements $SCD_{red.res.}^{SCIA}(\vartheta, \varphi)$ for four exemplary orbits from the year 2011.

| | relative difference $\frac{SCD^{SCIA}(\vartheta, \varphi)}{SCD_{red.res.}^{SCIA}(\vartheta, \varphi)} - 1$ (%) | | | |
|----------------|--|------------|------------|------------|
| orbit number | 46 222 | 47 514 | 48 821 | 50 143 |
| Latitude range | 1 Jan 2011 | 1 Apr 2011 | 1 Jul 2011 | 1 Oct 2011 |
| 90° S–80° S | –0.15 | +0.00 | n/a | +0.75 |
| 80° S–70° S | +4.1 | –16.0 | n/a | +2.6 |
| 70° S–60° S | +1.0 | +2.7 | –6.1 | +0.68 |
| 60° S–50° S | +0.90 | +0.87 | +2.3 | +0.44 |
| 50° S–40° S | +0.75 | +2.2 | +1.7 | +0.49 |
| 40° S–30° S | +0.12 | +0.59 | +0.92 | +2.3 |
| 30° S–20° S | –0.32 | +0.23 | +1.2 | +0.59 |
| 20° S–10° S | –0.34 | +0.29 | –0.54 | +0.51 |
| 10° S–0° | +0.28 | –0.29 | –0.051 | +0.35 |
| 0°–10° N | +0.75 | +1.1 | +0.054 | +0.062 |
| 10° N–20° N | +0.47 | +0.056 | +0.063 | +0.62 |
| 20° N–30° N | +0.12 | +0.086 | –0.11 | +0.36 |
| 30° N–40° N | +0.51 | +0.11 | +0.080 | +0.62 |
| 40° N–50° N | +0.61 | –0.014 | +0.38 | +0.25 |
| 50° N–60° N | +0.29 | +0.33 | +0.66 | +0.64 |
| 60° N–70° N | –16.0 | +1.2 | +0.90 | +0.22 |
| 70° N–80° N | +3.8 | –1.0 | +2.3 | –6.3 |
| 80° N–90° N | n/a | –2.4 | +0.83 | –17.0 |

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Table 3. Annual growth rate $\Delta\text{VCD}_{\text{trop}} \text{NO}_2$ from the levelshift model (Eq. 3) and the multi-instrument fit (Eq. 8) for the regions shown in Fig. 3. The relative trends have been computed relative to the 1996 annual mean.

| Region | Levelshift-model (orig. GOME data) | | Levelshift-model (rescorr. GOME data) | | Multi-instrument fit (10^{14} molec.cm ⁻² yr ⁻¹) |
|---------------------|---|-----------------------|---|-----------------------|---|
| | (10^{14} molec.cm ⁻² yr ⁻¹) | (% yr ⁻¹) | (10^{14} molec.cm ⁻² yr ⁻¹) | (% yr ⁻¹) | |
| continental US | -0.81 ± 0.14 | -1.7 ± 0.30 | -0.81 ± 0.15 | -1.6 ± 0.30 | -0.78 |
| Central-Eastern US | -2.9 ± 0.38 | -3.0 ± 0.38 | -2.9 ± 0.39 | -2.9 ± 0.39 | -2.7 |
| Western Europe | -2.9 ± 0.63 | -2.6 ± 0.56 | -3.0 ± 0.65 | -2.6 ± 0.56 | -2.6 |
| Japan | -0.49 ± 0.13 | -1.2 ± 0.33 | -0.49 ± 0.14 | -1.2 ± 0.33 | -0.49 |
| Middle East | +0.78 ± 0.08 | +4.0 ± 0.42 | +0.79 ± 0.08 | +4.0 ± 0.42 | +0.72 |
| East Central China | +10 ± 1.1 | +21 ± 2.2 | +10 ± 1.1 | +20 ± 2.2 | +8.7 |
| North Central India | +1.3 ± 0.27 | +4.0 ± 0.84 | 1.1 ± 0.27 | +4.6 ± 1.1 | +1.1 |

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Table 4. Annual growth rate $\Delta\text{VCD}_{\text{trop}} \text{NO}_2$ from the levelshift model (Eq. 3) and the multi-instrument fit (Eq. 8) for a list of megacities. The relative trends have been computed relative to the 1996 annual mean. Non-significant trends (see Eq. 6) are shown in italics.

| City | Levelshift-model (orig. GOME data) | | Levelshift-model (rescorr. GOME data) | | Multi-instrument fit (10^{14} molec $\text{cm}^{-2}\text{yr}^{-1}$) |
|--------------|--|-----------------------|--|-----------------------|--|
| | (10^{14} molec $\text{cm}^{-2}\text{yr}^{-1}$) | (% yr^{-1}) | (10^{14} molec $\text{cm}^{-2}\text{yr}^{-1}$) | (% yr^{-1}) | |
| Algiers | +0.74 ± 0.14 | +3.6 ± 0.69 | +0.83 ± 0.17 | +2.6 ± 0.53 | +0.60 |
| Athens | -2.3 ± 0.70 | -4.1 ± 1.2 | -2.1 ± 0.87 | -1.7 ± 0.71 | -2.1 |
| Baghdad | +3.5 ± 0.33 | +20 ± 1.9 | +3.9 ± 0.34 | +10 ± 0.87 | +3.2 |
| Beijing | +8.8 ± 2.5 | +6.7 ± 1.9 | +9.4 ± 2.7 | +4.8 ± 1.4 | +9.5 |
| Buenos Aires | +1.1 ± 0.48 | +3.9 ± 1.6 | <i>+0.85 ± 0.73</i> | <i>+1.1 ± 0.94</i> | +0.54 |
| Cairo | +1.9 ± 0.25 | +7.2 ± 0.93 | +2.1 ± 0.27 | +4.4 ± 0.55 | +1.7 |
| Chicago | -6.7 ± 1.8 | -4.1 ± 1.1 | -7.0 ± 2.0 | -2.5 ± 0.68 | -6.2 |
| New Delhi | +3.1 ± 0.53 | +9.3 ± 1.6 | +3.1 ± 0.56 | +5.5 ± 1.0 | +2.6 |
| Dhaka | +3.7 ± 0.52 | +27 ± 3.9 | +3.9 ± 0.50 | +13 ± 1.6 | +3.4 |
| Dimashq | +3.4 ± 0.54 | +10 ± 1.6 | +3.9 ± 0.60 | +6.0 ± 0.92 | +2.6 |
| Guangzhou | <i>+1.2 ± 2.0</i> | <i>1.3 ± 2.0</i> | +3.0 ± 2.6 | +1.6 ± 1.4 | +0.23 |
| Hong Kong | <i>-2.6 ± 1.8</i> | <i>-2.3 ± 1.6</i> | <i>-0.76 ± 2.2</i> | <i>-0.40 ± 1.2</i> | -1.1 |
| Istanbul | <i>-0.53 ± 1.1</i> | <i>-0.72 ± 1.5</i> | <i>-0.52 ± 1.3</i> | <i>-0.41 ± 1.0</i> | -0.36 |
| Jakarta | -1.4 ± 0.42 | -3.9 ± 1.1 | -1.6 ± 0.55 | -2.6 ± 0.91 | -1.2 |
| Jeddah | +1.3 ± 0.29 | +4.1 ± 0.92 | +1.6 ± 0.38 | +2.3 ± 0.53 | +1.4 |
| Karachi | +0.93 ± 0.22 | +6.8 ± 1.6 | +1.1 ± 0.29 | +3.8 ± 1.0 | +0.85 |
| Kolkata | +0.75 ± 0.22 | +3.0 ± 0.89 | +0.73 ± 0.23 | +2.0 ± 0.61 | +0.80 |
| Lagos | +0.41 ± 0.10 | +3.4 ± 0.83 | +0.40 ± 0.11 | +2.6 ± 0.73 | +0.33 |
| Lima | +0.99 ± 0.40 | +7.1 ± 2.9 | +1.2 ± 0.48 | +3.0 ± 1.2 | +1.1 |
| London | -4.4 ± 1.3 | -2.4 ± 0.71 | -4.5 ± 1.4 | -2.2 ± 0.68 | -3.0 |
| Los Angeles | -14 ± 1.7 | -6.0 ± 0.72 | -14 ± 1.9 | -3.1 ± 0.43 | -13 |
| Manila | -1.1 ± 0.18 | -5.3 ± 0.85 | -1.4 ± 0.31 | -3.6 ± 0.76 | -1.0 |
| Mexico City | <i>-0.54 ± 0.82</i> | <i>-1.2 ± 1.5</i> | <i>-0.30 ± 0.98</i> | <i>-0.22 ± 0.72</i> | -0.51 |
| Moscow | <i>-0.20 ± 0.15</i> | <i>-0.25 ± 0.19</i> | <i>+0.32 ± 1.7</i> | <i>+0.24 ± 1.2</i> | -1.4 |
| Mumbai | +8.2 ± 1.7 | 4.3 ± 0.91 | +0.91 ± 0.20 | +2.5 ± 0.53 | +0.70 |
| New York | -5.3 ± 1.7 | -2.5 ± 0.80 | -5.2 ± 2.3 | -1.4 ± 0.64 | -5.7 |
| Osaka | -1.9 ± 0.98 | -1.7 ± 0.88 | -2.1 ± 1.1 | -1.2 ± 0.61 | -2.5 |
| Paris | -4.6 ± 2.0 | -3.0 ± 1.3 | -5.8 ± 2.6 | -2.3 ± 1.0 | -5.2 |
| Riyadh | +2.0 ± 0.39 | +6.7 ± 1.3 | +2.5 ± 0.49 | +3.5 ± 0.68 | +2.0 |
| São Paolo | <i>+0.52 ± 0.46</i> | <i>-1.3 ± 1.2</i> | <i>+0.53 ± 0.58</i> | <i>+0.62 ± 0.69</i> | +0.37 |
| Seoul | <i>+0.65 ± 1.7</i> | <i>0.42 ± 1.1</i> | <i>+0.61 ± 2.1</i> | <i>+0.21 ± 0.72</i> | +1.0 |
| Shanghai | +12 ± 3.1 | +13 ± 3.3 | +13 ± 3.2 | +9.5 ± 2.4 | +9.4 |
| Shenzhen | <i>-2.4 ± 1.6</i> | <i>-1.9 ± 1.3</i> | <i>-1.8 ± 1.7</i> | <i>-1.2 ± 1.1</i> | -2.2 |
| Tehran | +2.1 ± 0.68 | +5.7 ± 1.9 | +2.5 ± 0.74 | +2.8 ± 0.84 | +2.7 |
| Tokyo | -5.2 ± 1.1 | -3.6 ± 0.79 | -5.5 ± 1.4 | -2.2 ± 0.54 | -5.4 |

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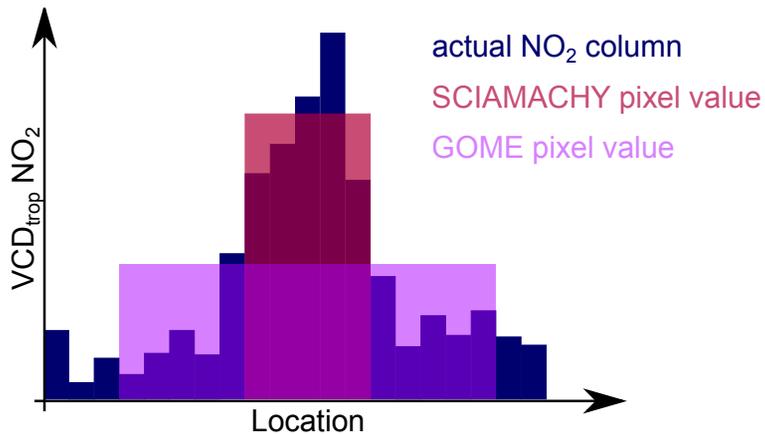


Fig. 1. Schematic view of the effect of the instrument's ground pixel size on the measured $VCD_{\text{trop}} \text{NO}_2$.

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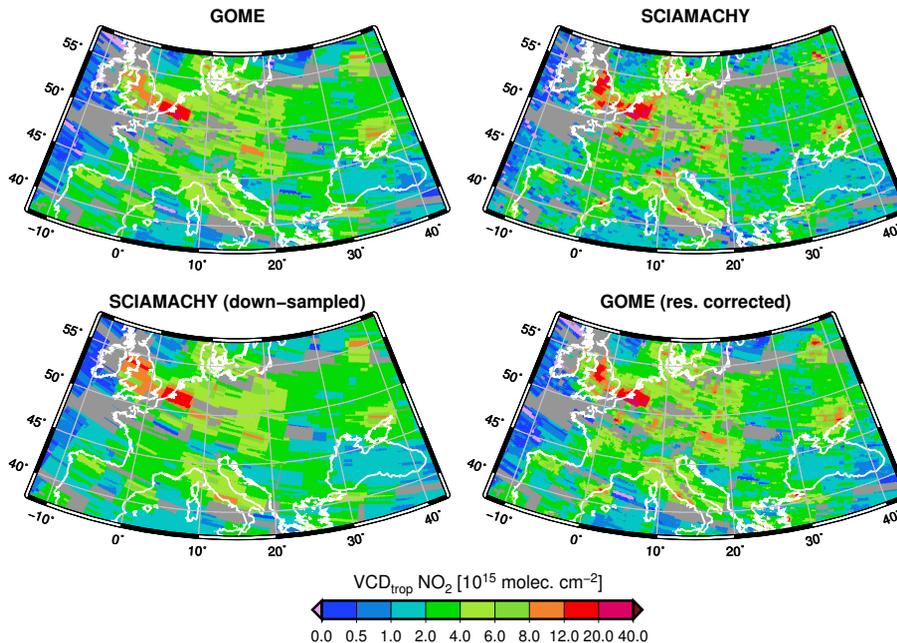


Fig. 2. Tropospheric NO₂ columns from GOME (top left), SCIAMACHY (top right), down-sampled SCIAMACHY (bottom left), and resolution-corrected GOME (bottom right) measurements for May 2003, gridded to $\frac{1}{16}^\circ \times \frac{1}{16}^\circ$. The effect of spatial smoothing can be seen in the original GOME and the down-sampled SCIAMACHY measurements for all point-like sources like cities, as the signal becomes smeared out.

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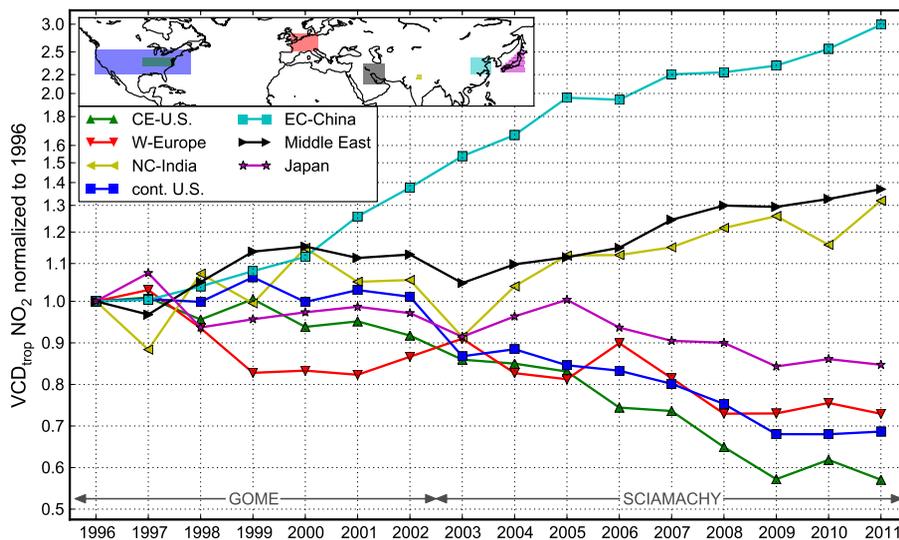


Fig. 3. Mean annual VCD_{top} NO_2 normalized to 1996, for the regions Central East coast US, Western Europe, US, East Central China, Japan, Middle East, and North Central India. Values for 1996–2002 are from GOME, values from 2003–2011 from SCIAMACHY measurements. The first five regions are defined as in Richter et al. (2005). The y-axis has been modified to make relative changes above and below 1 more comparable (values larger than one have been scaled to $y \mapsto 2 - \frac{1}{y}$).

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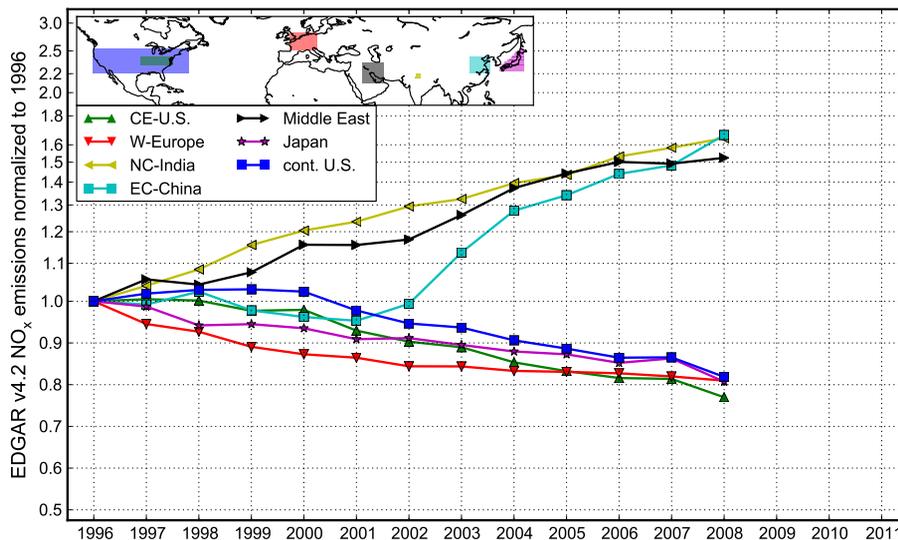


Fig. 4. NO_x emissions from the EDGAR v4.2 database, normalized to 1996, for the regions Central East coast US, Western Europe, US, East Central China, Japan, Middle East, and North Central India. The y-axis has been modified to make relative changes above and below 1 more comparable (values larger than one have been scaled to $y \mapsto 2 - \frac{1}{y}$). Since the published version 4.2 of the EDGAR database contains erroneous emission data for the year 2008 in Iran, this plot uses an updated, so far unpublished version (G. Maenhout, personal communication, 2012) for the Middle East region.

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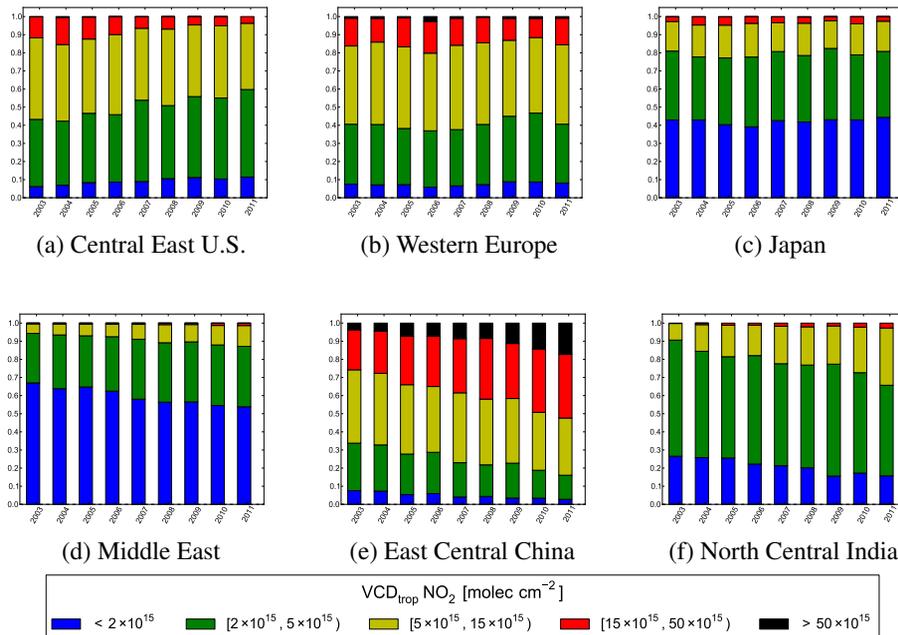


Fig. 5. Histogram of VCD_{trop} NO₂ from SCIAMACHY over the regions from Fig. 3 for the years 2003–2011. The plot shows the relative counts of background (blue), moderately polluted (green), polluted (yellow), very polluted (red), and extremely polluted (black) 1° × 1° grid cells per region.

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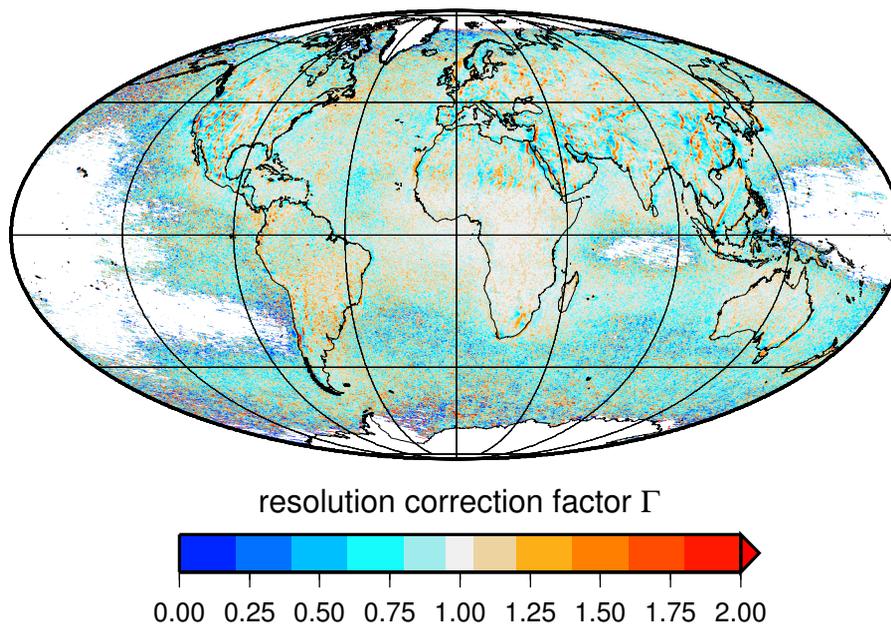


Fig. 6. Annual mean $\bar{\Gamma}$ of the resolution correction factor climatology (2003–2011). Pixels with a mean $\text{VCD}_{\text{red.res.}}^{\text{SCIA}}$ significantly lower than the instrument precision (estimated to be $10^{14} \text{ molec cm}^{-2}$) have been excluded from the plot.

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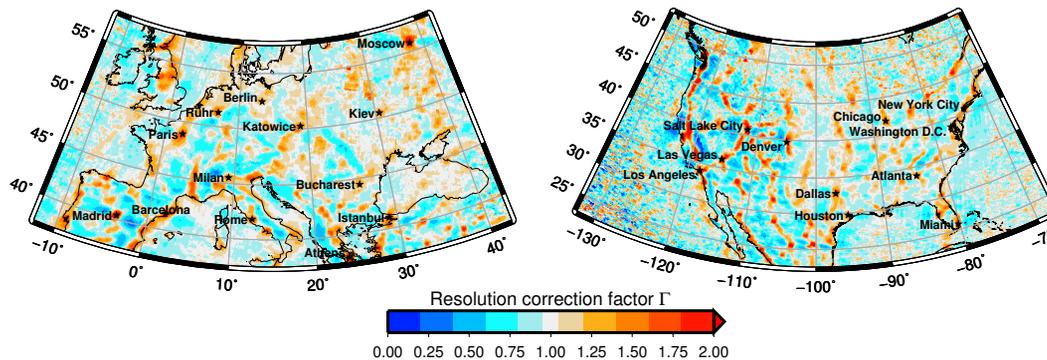


Fig. 7. Annual mean $\bar{\Gamma}$ of the resolution correction factor over the United States (right) and Europe (left).

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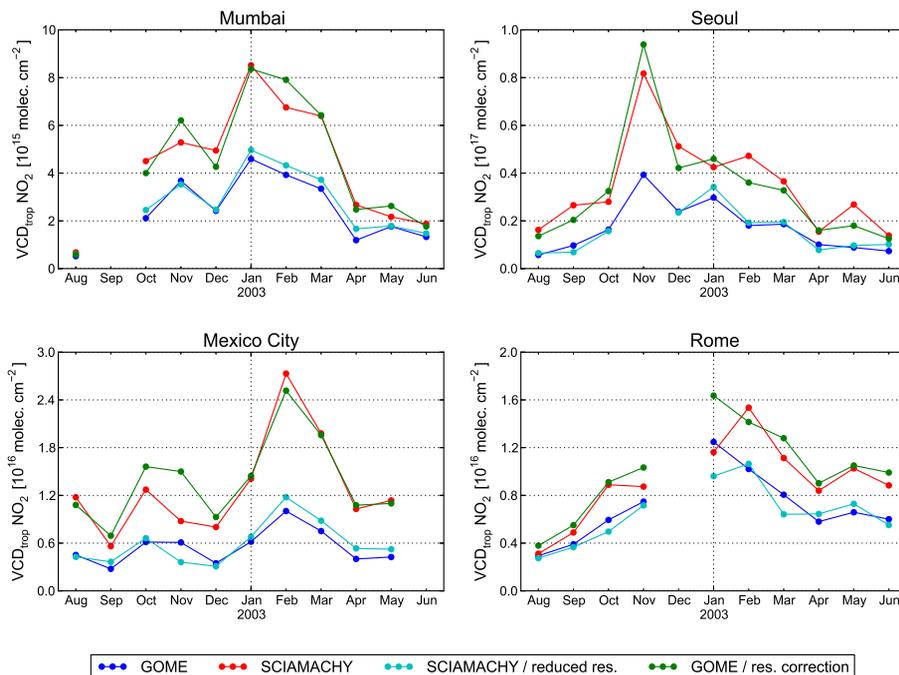


Fig. 8. Monthly averages of VCD_{trop} NO₂ over four exemplary areas showing very good agreement between corrected GOME and original SCIAMACHY measurements, for the time period August 2002–June 2003. Only those days for which both GOME and SCIAMACHY measurements for a city are available are taken into account.

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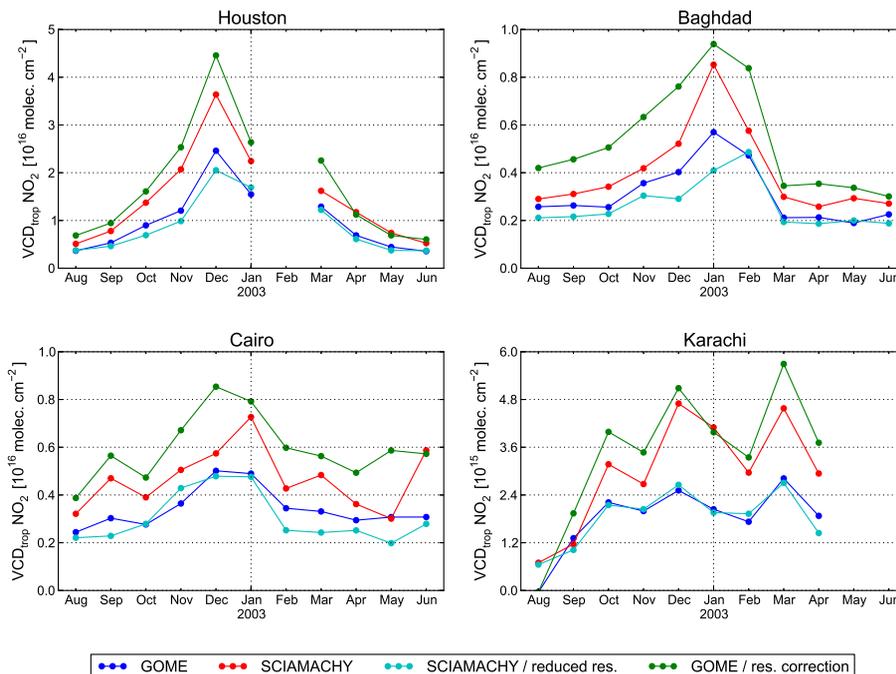


Fig. 9. Monthly averages of $VCD_{trop} NO_2$ over four exemplary areas showing a significant over-correction, for the time period August 2002–June 2003. Only those days for which both GOME and SCIAMACHY measurements for a city are available are taken into account.

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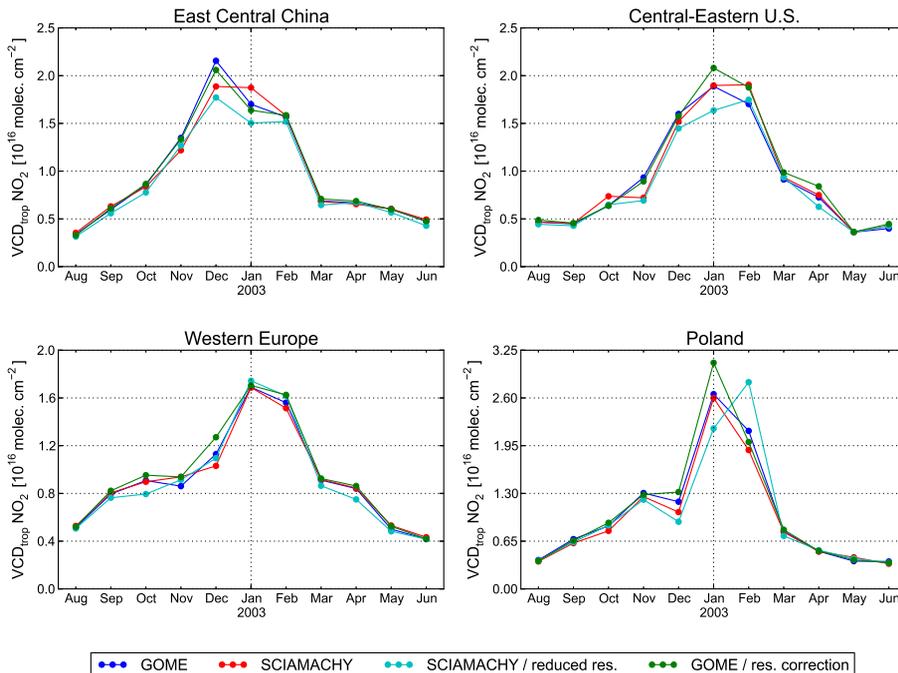


Fig. 10. Monthly averages of VCD_{trop} NO₂ over four country to continental scale areas, for the time period August 2002–June 2003. The regions are the same as used in Richter et al. (2005). Only those days for which both GOME and SCIAMACHY measurements for a city are available are taken into account.

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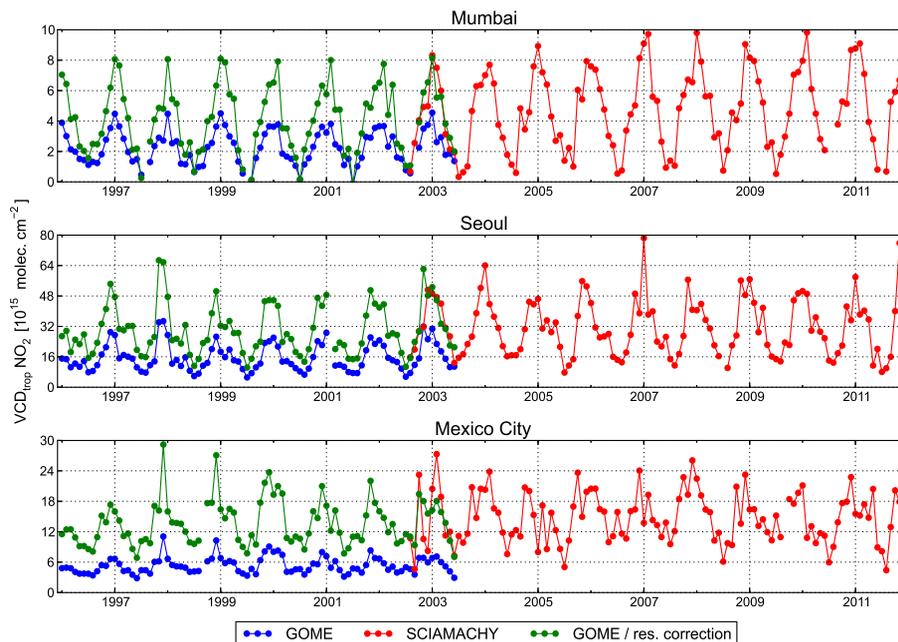


Fig. 11. Time series of original GOME (blue), resolution-corrected GOME (green), and SCIAMACHY (red) measurements of tropospheric NO_2 for the three selected megacities Mumbai, Seoul, and Mexico City.

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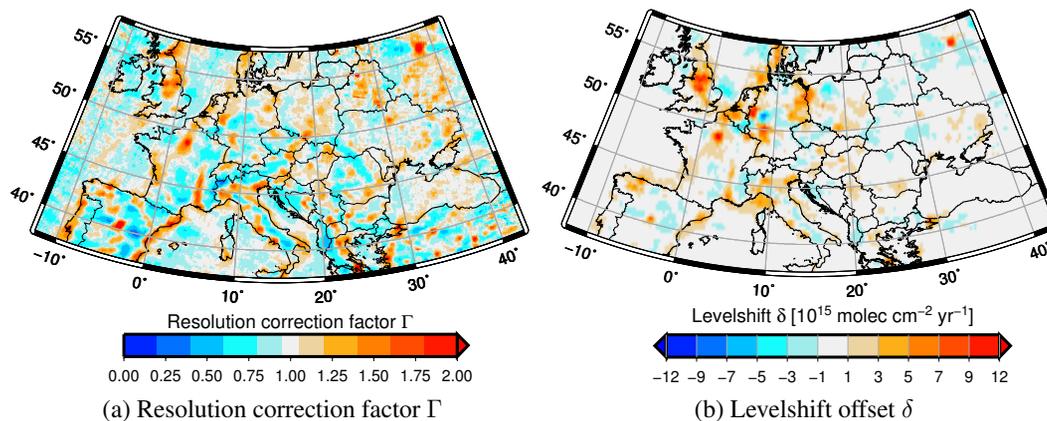
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**Fig. 12.** Comparison of resolution correction factor and levelshift offset.

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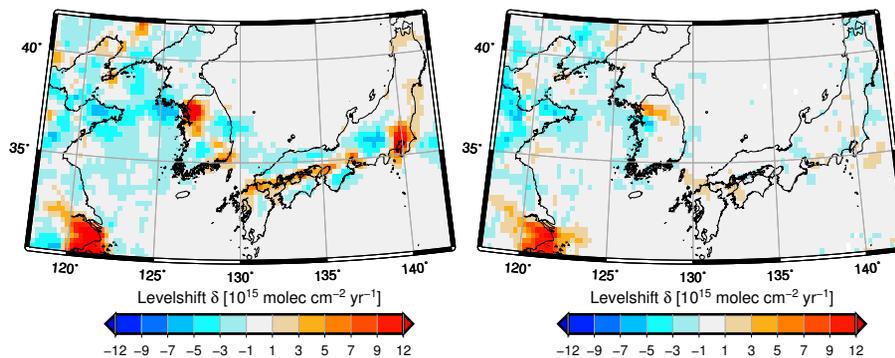


Fig. 13. Comparison of levelshift offset δ between the original (left) and resolution-corrected (right) GOME measurements and the SCIAMACHY dataset.

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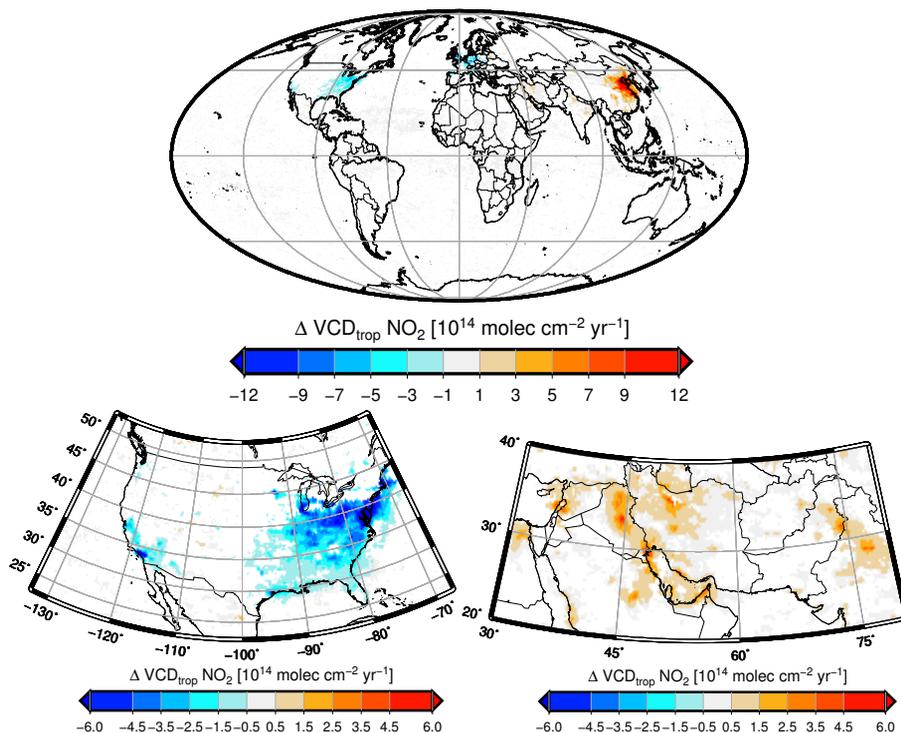


Fig. 14. Slope ω of the linear trend component of the levelshift trend model (Eq. 3), applied to monthly averages of GOME and SCIAMACHY measurements from 1996–2011, gridded to 0.25°. Those grid cells where the two-sided p-value for a hypothesis test whose null hypothesis is that the slope is zero is larger than 0.05 are excluded from the plot.

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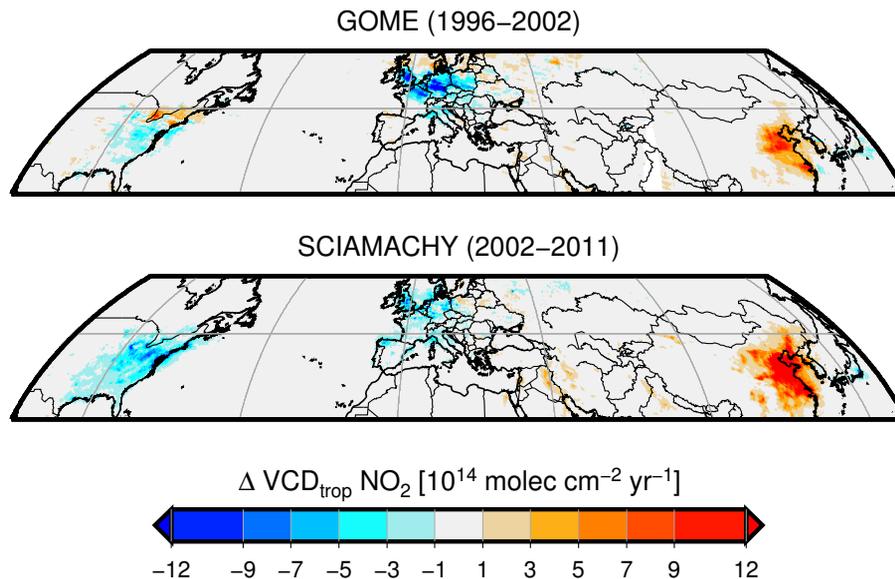


Fig. 15. Slope ω of the seasonality model (Eq. 3 without the levelshift terms $\delta U(t)$ and $(\gamma - 1)U(t)$), applied to monthly averages of GOME (top) and SCIAMACHY (bottom) measurements alone. All data have been gridded to 0.25° before performing the model fit. Contrary to Fig. 14, statistically insignificant trends have not been masked out.

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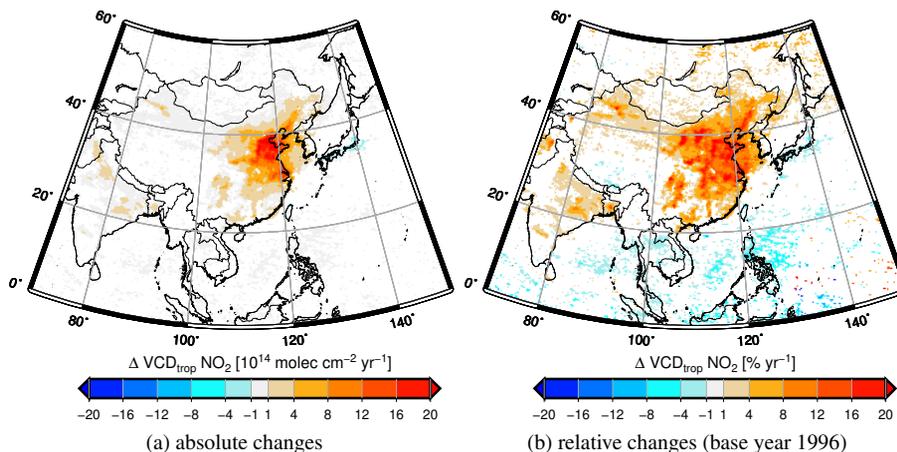


Fig. 16. Annual change in $\text{VCD}_{\text{trop}} \text{NO}_2$ over China derived from the levelshift trend model for the years 1996–2011. The maps show the statistically significant values of the linear trend component ω from the levelshift model (Eq. 3) in absolute values (left) and relative changes (right).

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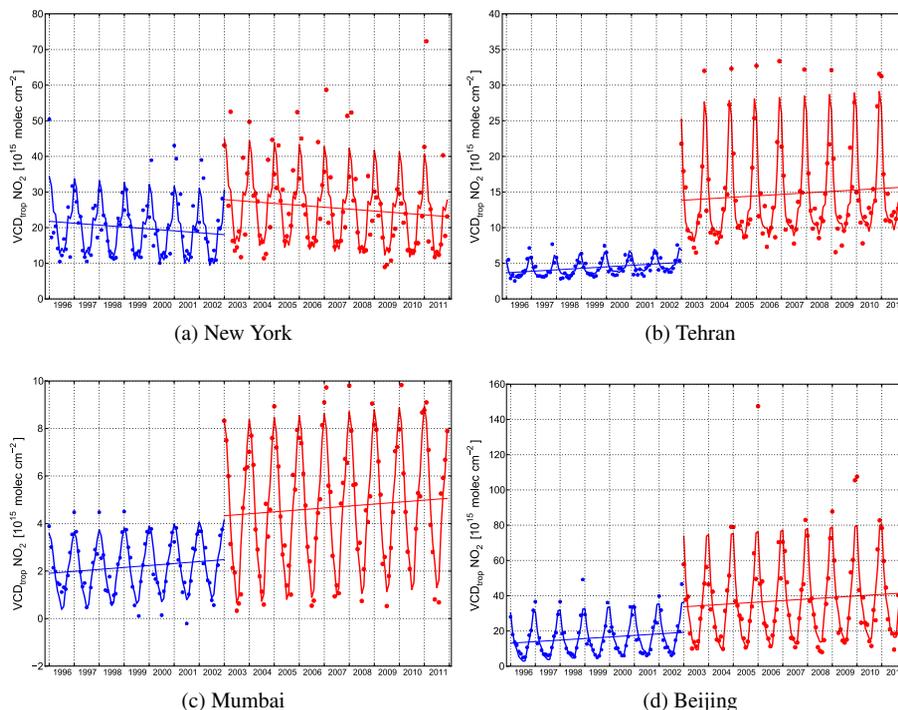


Fig. 17. Time series of monthly $\text{VCD}_{\text{trop}} \text{NO}_2$ (dots), fitted levelshift trend model (thick line), and linear component of the fit (straight line) for the time periods 1996–2002 (GOME, blue) and 2003–2011 (SCIAMACHY, red).

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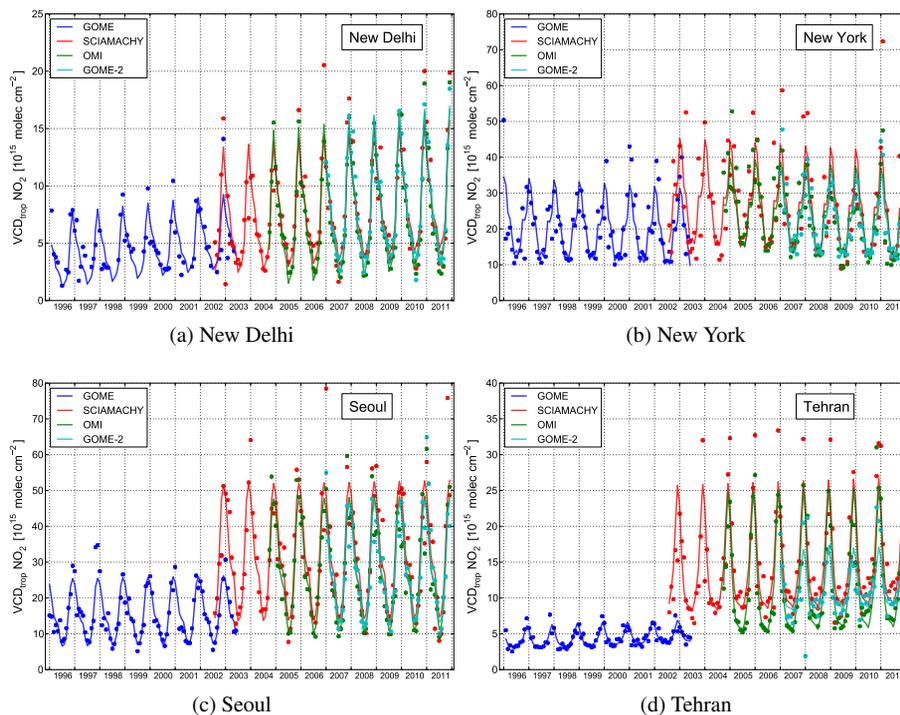


Fig. 18. Measured monthly average $\text{VCD}_{\text{trop}} \text{NO}_2$ (dots) and fitted trend function from the multi-instrument trend model (lines) for the four instruments GOME (blue), SCIAMACHY (red), OMI (green), and GOME-2 (cyan).

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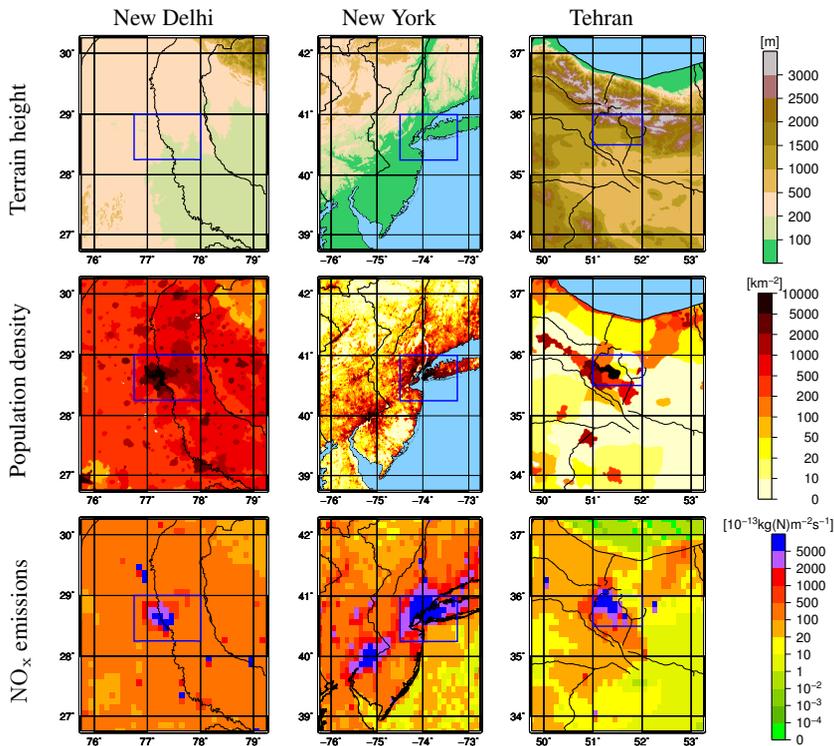


Fig. 19. Topography, population density, and NO_x emissions for the three selected megacity regions New Delhi (India), New York (United States), and Tehran (Iran). Topographic data is from the GLOBE project (Hastings et al., 1998), population density data from the GRUMP dataset (Center for International Earth Science Information Network (CIESIN), Columbia University, International Food Policy Research Institute (IFPRI), the World Bank, and Centro Internacional de Agricultura Tropical (CIAT), 2011), and NO_x emission values are from the EDGAR database (European Commission, Joint Research Centre (JRC) and Netherlands Environmental Assessment Agency (PBL), 2011).

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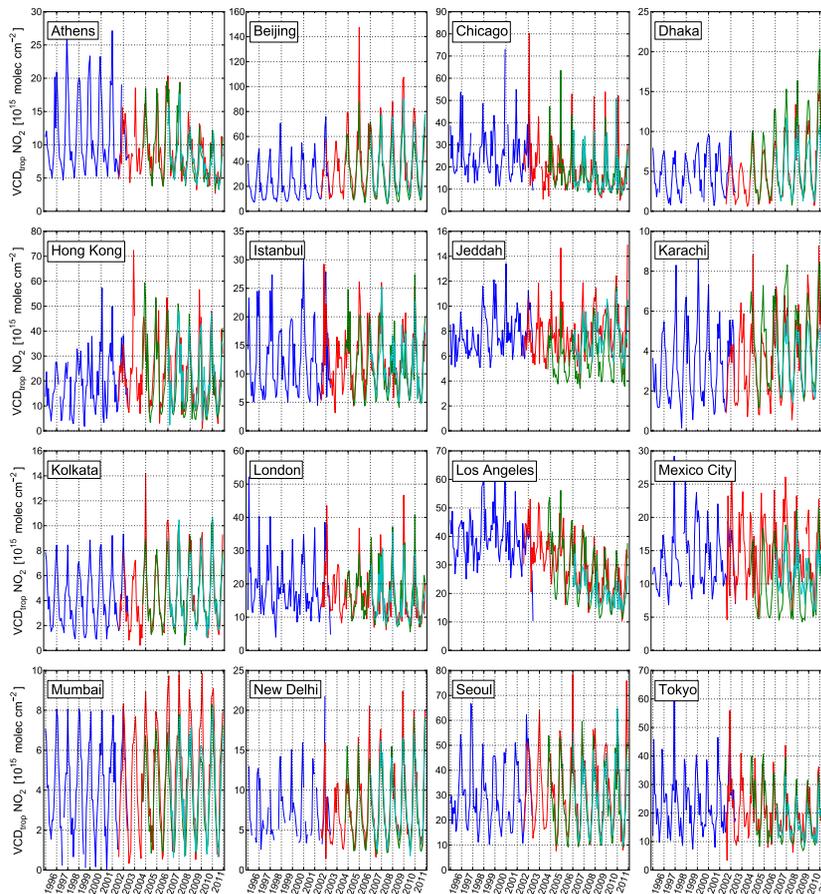


Fig. 20. Time series of monthly mean $VCD_{top} NO_2$ from GOME (blue), SCIAMACHY (red), OMI (green), and GOME-2 (cyan). The GOME measurements have been corrected for the instrument's pixel size using the mechanism presented in Sect. 4.