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Multiannual changes of CO_2 emissions in China: indirect estimates derived from satellite measurements of tropospheric NO_2 columns

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Abstract. Multiannual satellite measurements of tropospheric NO_2 columns are used for evaluation of CO_2 emission changes in China in the period from 1996 to 2008. Indirect top-down annual estimates of CO_2 emissions are derived from the satellite NO_2 column measurements by means of a simple inverse modeling procedure involving simulations performed with the CHIMERE mesoscale chemistry–transport model and the CO_2-to-NO_x emission ratios from the Emission Database for Global Atmospheric Research (EDGAR) global anthropogenic emission inventory and Regional Emission Inventory in Asia (REAS). Exponential trends in the normalized time series of annual emissions are evaluated separately for the periods from 1996 to 2001 and from 2001 to 2008. The results indicate that the both periods manifest strong positive trends in the CO_2 emissions, and that the trend in the second period was significantly larger than the trend in the first period. Specifically, the trends in the first and second periods are best estimated to be in the range from 3.7 to 8.3 and from 11.0 to 13.2 % per year, respectively, taking into account statistical uncertainties and differences between the CO_2-to-NO_x emission ratios from the EDGAR and REAS inventories. Comparison of our indirect top-down estimates of the CO_2 emission changes with the corresponding bottom-up estimates provided by the EDGAR (version 4.2) and Global Carbon Project (GCP) glomal emission inventories reveals that while acceleration of the CO_2 emission growth in the considered period is a common feature of both kinds of estimates, nonlinearity in the CO_2 emission changes may be strongly exaggerated in the global emission inventories. Specifically, the atmospheric NO_2 observations do not confirm the existence of a sharp bend in the emission inventory data time series in the period from 2000 to 2002. A significant quantitative difference is revealed between the bottom-up and indirect top-down estimates of the CO_2 emission trend in the period from 1996 to 2001 (specifically, the trend was not positive according to the global emission inventories, but is strongly positive in our estimates). These results confirm the findings of earlier studies that indicated probable large uncertainties in the energy production and other activity data for China from international energy statistics used as the input information in the global emission inventories. For the period from 2001 to 2008, some quantitative differences between the different kinds of estimates are found to be in

the range of possible systematic uncertainties associated with our estimation method. In general, satellite measurements of tropospheric NO_2 are shown to be a useful source of information on CO_2 sources collocated with sources of nitrogen oxides; the corresponding potential of these measurements should be exploited further in future studies.

1 Introduction

The rapid increase in atmospheric carbon dioxide (CO_2) concentration due to anthropogenic emissions is commonly recognized as one of the major driving forces of global warming (IPCC, 2007). Good knowledge of CO_2 anthropogenic emissions into the atmosphere and their long-term changes is therefore indispensable to any scientific study or science-based policy actions aiming at prediction and control of climate change.

Presently, CO_2 and other greenhouse gas (GHG) emissions on different temporal and spatial scales are reported in many international (e.g., Olivier et al., 2005; GCP, 2010; Ciais et al., 2010; Janssens-Maenhout et al., 2012) and national (e.g., Ohara et al., 2007; Zhao et al., 2012; Huang et al., 2011) inventories. Such inventories are based on combination of available statistical information on activities in different sectors of the economy (such as transport, industries, energy production, etc.) with emission factors for individual processes and fuel types. There is, however, evidence of inaccuracies in the available statistical information, leading to errors in corresponding emission estimates. For example, Akimoto et al. (2006) and Guan et al. (2012) discuss possible underestimation of fossil fuel CO_2 emissions in China due to inaccuracies in official information on the fossil fuel (in particular coal) consumption.

An alternative way to obtain emission estimates of CO_2 and other trace gases is provided by the inverse modeling approach (Enting, 2002). Emission estimates derived from trace gas observations by means of atmospheric models can improve knowledge of the GHG balance (e.g., Schulze et al., 2009), and help in pinpointing possible uncertainties and inconsistencies in the bottom-up inventories. For brevity, emission estimates constrained by atmospheric measurements are referred below to as "top-down" estimates, while more traditional emission estimates based on emission inventories are referred to as "bottom-up" estimates.

In the last decade, the inverse modeling approach has rapidly been gaining popularity due to the advent of satellite measurements of various trace gases in the troposphere (Burrows et al., 2011). In particular, satellite measurements of nitrogen dioxide (NO_2) have been widely used to retrieve spatial structure and trends of emissions of nitrogen oxides (e.g., Martin et al., 2003; Müller and Stavrakou, 2005; Konovalov et al., 2006, 2008; Kurokawa et al., 2009; Wang et al., 2007; Napelenok et al., 2008; Miyazaki et al., 2012), carbon monoxide (CO) measurements were found to be helpful for constraining CO emissions (e.g., Arellano et al., 2004; Pétron et al., 2004; Heald et al., 2004; Kopacz et al., 2010), and satellite observations of methane (CH_4) were used to validate and improve CH_4 bottom-up emission estimates (e.g., Bergamaschi et al., 2007, 2009; Monteil et al., 2011). It was also demonstrated (e.g., Curci et al., 2010; Lee et al., 2011; Palmer et al., 2003; Dufour et al., 2009; Millet et al., 2008; Stavrakou et al., 2009) that satellite measurements can provide useful information about emissions of several important organic gases.

Numerous studies (e.g., Rayner and O'Brien, 2001; Pak and Prather, 2001; Houweling et al., 2004; Chevallier et al., 2007; Kadygrov et al., 2009; Hungershoefer et al., 2010; Nassar et al., 2011) studied the potential of satellite CO_2 measurements as a source of information on CO_2 emissions and natural sinks. It has been demonstrated from model studies that inversions of such measurements could facilitate improvements of CO_2 emission estimates on large (continental) scales. At the same time, it has also been found that emission estimates derived from CO_2 satellite observations are especially sensitive to model and measurement errors (e.g., Houweling et al., 2010). This sensitivity reflecting the large lifetime and relatively small variability (typically, less than 5 %) of CO_2 columns in the atmosphere seriously hinders estimation of CO_2 emissions and their changes on finer (regional) scales. Similar limitations also exist in the case of CO_2 inversions based on ground-based measurements (Enting, 2002), with these measurements being more precise but more sparse than the satellite remote sensing data.

The focus of this paper is the evaluation of multiannual changes in CO_2 emissions from China. According to Boden et al. (2011) and other inventories, China has recently become the world leader in total CO_2 emissions. However, there are strong indications (see, e.g., Gregg et al., 2008; Guan et al. 2012) that the uncertainty in the available CO_2 emission inventory data for China is rather large (15–20 %). Our idea is that indirect but still useful information about the CO_2 emissions changes can be derived from satellite measurements of tropospheric NO_2 columns. Indeed, on the one hand, it was shown earlier that such measurements clearly reflect local "hot spots" and long-term changes in anthropogenic NO_x emissions in China (Richter et al., 2005; Wang et al., 2010, 2012; Zhang et al., 2007; Lin, 2012). On the other hand, both CO_2 and NO_x have many common sources associated with fossil fuel and other fuel combustion, and therefore temporal changes in anthropogenic emissions of these species should be related. Importantly, due to the relatively short lifetime of NO_x, NO_2 atmospheric observations are less affected by the long-range transport from distant emissions than CO_2 measurements, and therefore interpretation of the NO_2 observations is more straightforward and robust.

The idea to estimate CO_2 emissions by analyzing observations of co-emitted species is not new (see, e.g., Rivier et al.,

2006), and has already been exploited in several studies. For example, Brioude et al. (2012) used aircraft measurements of several species (CO, NO_y, and SO_2) to estimate CO_2 emissions from Houston. Earlier, Palmer et al. (2006) and Suntharalingam et al. (2004) used the correlation between CO and CO_2 instantaneous observations during aircraft campaigns to improve spatial allocation of NO_2 emission sources in eastern Asia, including China. Unlike the mentioned studies, we do not exploit possible correlation between atmospheric concentrations of CO_2 and co-emitted species, which in the considered specific case of NO_2 being used as a "tracer" of CO_2 emissions is probably rather small because the behavior of NO_2 in the atmosphere is strongly affected by chemical processes. Instead, the assumed relation between CO_2 and NO_x emissions is specified using data of emission inventories. To relate NO_x emissions to the NO_2 observations we employ a mesoscale chemistry–transport model and a simple inverse modeling method similar to that used in Konovalov et al. (2010) for studying NO_x emission trends in megacities. Accordingly, our estimates of CO_2 emission changes are obtained by merging the top-down estimates of NO_x emission changes and bottom-up estimates of the NO_x-to-CO_2 emission conversion factors. Such CO_2 emission estimates can therefore be considered either as indirect top-down or hybrid estimates combining the top-down and bottom-up information.

Since CO_2 emission trend estimates obtained in this study are based on observations (even though some emission inventory data are also involved), they are to a significant extent independent from similar estimates provided by emission inventories. Taking this into account, we believe that our indirect top-down (or hybrid) CO_2 emission estimates can be helpful for detecting and elucidating possible uncertainties and/or inconsistencies in alternative bottom-up emission estimates and in inventory data that are used to relate NO_x and CO_2 emissions in our analysis. Specifically, a disagreement between the hybrid and bottom-up estimates in our case may indicate (assuming that our estimates of NO_x emissions are sufficiently accurate) that either the inventory data for CO_2 emissions or the assumed relationship between CO_2 and NO_x emissions (the NO_x-to-CO_2 emission conversion factor) or both are insufficiently accurate. Note that we did not evaluate absolute values of NO_x or CO_2 emissions from China; only their temporal changes from a baseline year value are examined here. Due to this, our results are expected to be rather insensitive to poorly known biases in the modeled and measured NO_2 columns as long as these biases remain constant with time during the study period.

The data used in our analysis are described in Sect. 2. The method is outlined in Sect. 3. Our estimates of CO_2 and NO_x emission trends in China are presented in comparison with corresponding data of several emission inventories in Sect. 4. The potential uncertainties of our results and their possible implications are discussed in Sect. 5. Finally, the main findings of this study are summarized in Sect. 6.

2 Data

2.1 Data of satellite measurements

We used tropospheric NO_2 column amounts retrieved as described by Richter et al. (2005) at the Institute of Environmental Physics and Remote Sensing, University of Bremen from measurements performed by the GOME (1996–2002) (Burrows et al., 1999) and SCIAMACHY (2003–2008) (Bovensmann et al., 1999) instruments onboard the ERS-2 and ENVISAT satellites, respectively. Specifically, we used the standard monthly data products (GOME tropospheric excess NO_2 columns version 2 (Nüß, 2005) and SCIAMACHY tropospheric excess NO_2 columns version 0.7 (Richter et al., 2005) provided on a regular grid with the resolution of $0.5^\circ \times 0.5^\circ$ and $0.125^\circ \times 0.125^\circ$ in the cases of GOME and SCIAMACHY data, respectively. The same data have already been used in several previous studies aimed at evaluation of long-term changes of NO_x emissions in different regions of the world (Richter et al., 2005; He et al., 2007; Kim et al., 2006, 2009; Konovalov et al., 2008, 2010). The initial nominal horizontal resolution of the GOME measurements is $320 \times 40\,km^2$, and almost global coverage is achieved within three days. The SCIAMACHY measurements have a higher nominal horizontal resolution of $60 \times 30\,km^2$, but a longer global coverage period of about six days.

Preprocessing of the "standard" monthly datasets included several steps described below. First, the global data were regridded to a $0.5^\circ \times 0.5^\circ$ regional grid focusing on China. Second, "empty" grid cells (in which measurements are missing, e.g., because of clouds) in each of the monthly datasets were filled in by means of a temporal or spatial interpolation. Specifically, an empty grid cell in a given monthly dataset was assigned with either the average over the two nearest months or (if the data for those months are also missing) the average over the eight closest grid cells of the same month. Finally, to ensure the consistency of NO_2 columns measured with different spatial resolution, the SCIAMACHY monthly data were spatially smoothed on the longitudinal plane in the same way as in Konovalov et al. (2010) to simulate the smoothing introduced by the GOME measurements. Specifically, the smoothed c^s and original c^{or} NO_2 columns are related as follows:

$$c^s_{(i)} = \sum_{j=0}^{2m} c^{or}_{i-m+j}\,\rho_j \left[\sum_{k=0}^{2m}\rho_k\right]^{-1},$$
$$\rho_j = \exp\left[-\frac{1}{2}\left(\frac{(j-m)\cos(\varphi)}{\lambda}\right)^2\right], \quad (1)$$

where the lower subscripts specify the longitudinal grid cell index (counting along a constant latitude), m is the number of grid cells on the longitudinal plane within 320 km (the typical resolution of the GOME measurements), φ is the latitude, and λ is the effective distance scale. The value

of λ was estimated in Konovalov et al. (2010) to be of 0.85 ± 0.16 by minimizing the mean squared difference between the smoothed NO_2 columns (from SCIAMACHY) for 2003 and the original NO_2 columns (from GOME) for 2002 over 12 urban agglomerations situated in Europe and in the Mediterranean. Note that our results concerning total emissions from a very large region (such as the eastern part of China) considered in this study are found to be quite insensitive to the choice of the value of λ. As it has been shown previously (Richter et al., 2005), the GOME and SCIAMACHY data averaged over East China are sufficiently consistent (even without smoothing of the SCIAMACHY data), and their combined time series can therefore be used for evaluation of corresponding emission changes. The smoothing procedure involving a sufficiently accurate estimate of λ is more important when emissions from smaller regions (such as China's provinces) are evaluated, but this study is not aimed at obtaining accurate quantitative estimates of emissions for individual Chinese provinces.

2.2 Data of emission inventories

We used annual emission data from several emission databases, including the Emission Database for Global Atmospheric Research, version 4.2 (EDGAR v4.2), the Global Carbon Project (GCP) international inventories, the Regional Emission Inventory in Asia (REAS), and a new global data product for CO_2 emission by combustion processes (PKU-CO2) compiled at Peking University. CO_2 and NO_x emissions are available from the EDGAR v4.2 (EC-JCR/PBL, 2010) database both as the national totals and on a grid with the resolution of $0.1^\circ \times 0.1^\circ$. GCP (GCP, 2010) provides only total national carbon emission estimates. The data from the EDGAR v4.2 gridded and GCP country-averaged emission inventories cover the whole period addressed in this study (1996–2008). The methods used in the EDGAR and GCP emission inventories are described in detail elsewhere (e.g., Olivier et al., 2001; Janssens-Maenhout, 2012; Ciais et al., 2010).

The "historical" emission estimates by the REAS inventory were available (both on a grid and as the national totals) as the version 1.11 (v1.11) data (Ohara et al., 2007) for the years from 1980 to 2003 and as the version 2.1 (v2.1) data (Kurokawa et al., 2013) for the years from 2000 to 2008. In this study, we independently considered the time series of the national total emissions from REAS v1.11 and REAS v2.1 for the 1996–2001 and 2001–2008 periods, respectively. Such periods are chosen consistently with the periods used in our trend analysis (see Sects. 3 and 4.1).

The Peking University (PKU-CO2) emission inventory consists of gridded CO_2 emissions from combustion processes over the globe, using county-level fuel data in China; it was established for the year 2007, and updated with the new county-level data in China for three years only (1997, 2002, and 2008) in this study. Details can be found elsewhere (Wang et al., 2013).

The emission inventory data outlined above are considered below in comparison with corresponding estimates based on satellite measurements. Focus is given to the data of the global emission inventories (EDGAR and GCP) that have direct implications for climate change research. Data of the regional emission inventories (REAS and PKU-CO2) are used in this study mainly for sensitivity tests and as a source of supportive information. A thorough comparative analysis of the different global and regional emission inventories is beyond the scope of this paper.

2.3 Simulated data

To evaluate the relationship between the tropospheric NO_2 columns and NO_x emissions, we used the CHIMERE chemistry–transport model (www.lmd.polytechnique.fr/chimere). CHIMERE is a mesoscale Eulerian three-dimensional model that takes into account all important processes determining the evolution of nitrogen oxides in the troposphere, such as a number of gas-phase and heterogeneous chemical reactions, dry and wet deposition, advection, turbulent diffusion, and deep convection. Parameterizations of these processes in the model are described in several papers (e.g., Schmidt et al., 2001; Bessagnet et al., 2004; Hodzic and Jimenez, 2011).

The simulations were performed with a horizontal resolution of $1^\circ \times 1^\circ$ for 12 layers in the vertical (up to 200 hPa pressure level). The model domain covered the East Asia region (from 58.250° to 141.750° E and from 20.250° to 65.750° N) including continental China. Meteorological data were obtained from the WRF-ARW model (Skamarock et al., 2005; Wicker and Scamarock, 2002), which was run with a horizontal resolution of $90 \times 90\,km^2$ and driven with the NCEP Reanalysis-2 data. Chemical processes were simulated with the MELCHIOR2 chemical mechanism. Boundary conditions for gases and aerosols were taken from climatological runs of LMDZ (Lott et al., 2005; Hourdin et al., 2006) and GOCART (Ginoux et al., 2001; Chin et al., 2000, 2002), respectively. Anthropogenic emissions were specified using the EDGAR v4.2 data, biogenic emissions were based on the MEGAN global inventory (Guenther et al., 2012). Along with the "baseline" scenario, where both anthropogenic and biogenic emissions were taken into account, the model was also run with zero anthropogenic emissions in China.

In order to be consistent with satellite data, which are described in Sect. 2.1, the modeled NO_2 columns for each model grid cell are taken between 10 and 12 h of local solar time and only on days with insignificant cloud cover. Since the total cloud cover was not used in the CHIMERE simulation, we use a selection criteria based on threshold value of the radiation attenuation coefficient. Specifically, we disregarded days on which reduction of solar radiation due to

clouds was larger than 30 %. The same criterion was used in our earlier studies (Konovalov et al., 2005, 2006). The simulated NO_2 columns were smoothed in the same way as the SCIAMACHY data.

Figure 1 presents the spatial distributions of the simulated and measured NO_2 columns over China (before smoothing). It can be seen that CHIMERE reproduces the data derived from satellite measurements rather adequately, although the agreement is obviously not perfect. In particular, the model systematically underestimates the NO_2 columns over the most polluted industrial regions in the eastern part of China. Systematic underestimation of satellite measurements of NO_2 columns over China by simulations was found earlier in other studies employing different models (e.g., Ma et al., 2006; He et al., 2007; Lin et al., 2010; Lin, 2012), and was interpreted as mostly being a consequence of a corresponding bias in NO_x emission inventories for China.

3 Method

The first step of our method is the estimation of interannual changes in anthropogenic NO_x emissions. Following Konovalov et al. (2010), we define

$$C_i^j \approx \alpha_i^j E_i^{(\mathrm{NO_x})j} + C_i^{\mathrm{b}j},$$
$$i \in \{1..12\},\ j \in \{1996..2008\}, \quad (2)$$

where $\mathbf{C}$ is the monthly mean total tropospheric NO_2 column amount measured by satellites over a considered region for the month i and the year j; $\mathbf{E}^{(\mathrm{NO_x})}$ is the corresponding anthropogenic NO_x amount emitted in the same region and month; α is the sensitivity of the NO_2 columns to changes of the NO_x emissions; and $\mathbf{C}^{\mathrm{b}}$ is the "background" level of the tropospheric NO_2 column amounts, which is not related to anthropogenic emissions in the given region and month. Note that α can be interpreted as the effective lifetime of NO_2 columns with regard to both chemical and transport processes.

Similar to Eq. (2) we can define the relationship between the modeled NO_2 columns and NO_x emissions:

$$C_{\mathrm{m}i}^k \approx \alpha_{\mathrm{m}i}^k E_{\mathrm{m}i}^{(\mathrm{NO_x})k} + C_{\mathrm{m}i}^{\mathrm{b}k},$$
$$i \in \{1..12\}, k \in \{1996..2008\}, \quad (3)$$

where the subscript m denotes the modeled values corresponding to the observed ones in Eq. (2). After replacing $\mathbf{C}^{\mathrm{b}}$ and α by $\mathbf{C}_{\mathrm{m}}^{\mathrm{b}}$ and $\boldsymbol{\alpha}_{\mathrm{m}}$, respectively, in Eq. (2) and after combining Eq. (2), and Eq. (3), we can establish the following relationship between the inferred monthly emissions and those assumed in the model:

$$E_i^{(\mathrm{NO_x})j} \approx E_{\mathrm{m}i}^{(\mathrm{NO_x})k} \frac{(C_i^j - C_{\mathrm{m}i}^{\mathrm{b}j})}{(C_{\mathrm{m}i}^k - C_{\mathrm{m}i}^{\mathrm{b}k})} \zeta_{ij}^k,$$
$$i \in \{1..12\},\ j,k \in \{1996..2008\}, \quad (4)$$

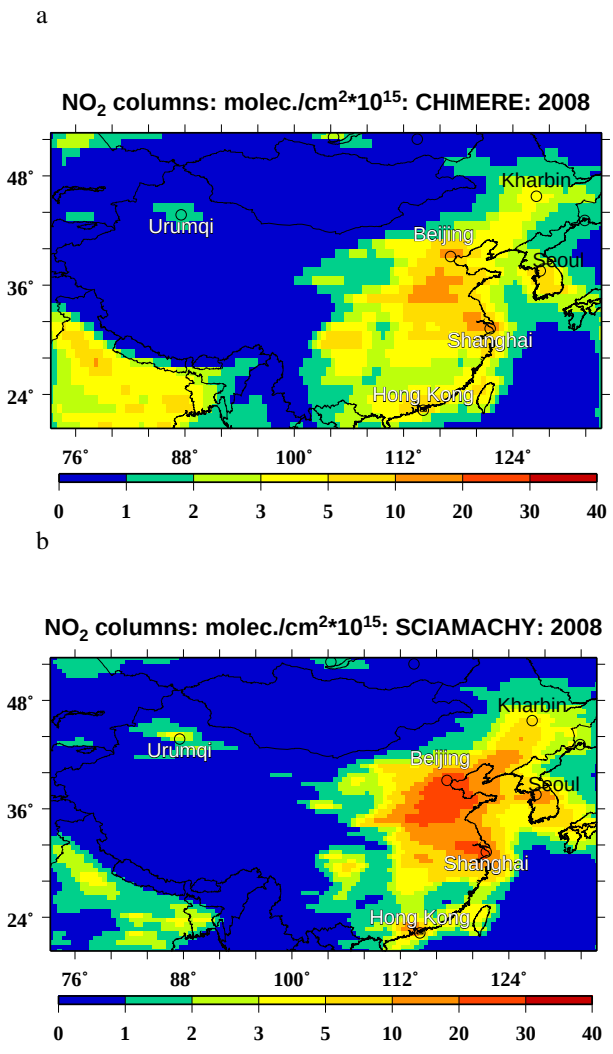


Fig. 1. Annual mean tropospheric NO_2 column amounts (molecules$\times 10^{15}$ cm^{-2}) simulated with the CHIMERE model **(a)** and derived from SCIAMACHY measurements **(b)** for the year 2008.

where

$$\zeta_{ij}^k = \frac{\alpha_{\mathrm{m}i}^k}{\alpha_{\mathrm{m}i}^j}, \quad (5)$$

and the years j and k can, in the general case, be different.

Accordingly, the estimated total annual emission, $\mathbf{E}_{\mathrm{t}}^{(\mathrm{NO_x})}$, in the year j can be expressed through the monthly emission specified in the model for the year k as follows:

$$E_{\mathrm{t}}^{(\mathrm{NO_x})j} = \sum_{i=1}^{12} \left(E_{\mathrm{m}i}^{(\mathrm{NO_x})k} \frac{(C_i^j - C_{\mathrm{m}i}^{\mathrm{b}j})}{(C_{\mathrm{m}i}^k - C_{\mathrm{m}i}^{\mathrm{b}k})} \zeta_{ij}^k \right),$$
$$j,k \in \{1996..2008\}. \quad (6)$$

We evaluated $\mathbf{C}_{\mathrm{m}}$ and $\mathbf{C}_{\mathrm{m}}^{\mathrm{b}}$ by means of the CHIMERE chemistry–transport model under following assumptions:

1. The impact of NO_x emitted in a previous month on the NO_2 columns in a given month is negligible (note that in principle this impact could be reflected in $\mathbf{C}_m^b$).

2. The transport of NO_x into a considered region from outside is also negligible.

3. The long-term changes in natural NO_x emissions (from soil, wildfires, and lightning) are much smaller than the trends in anthropogenic emissions and can be disregarded.

4. The impact of interannual changes in the NO_2 columns and the NO_2 lifetime due to meteorological variability on the NO_x emission estimates defined by Eq. (6) is small (in particular, in comparison with the impact of uncertainties in the measured and modeled NO_2 columns) and can also be disregarded.

These assumptions, which, in our opinion, are sufficiently reasonable and justifiable (see Sect. 5) in view of results of numerous previous studies employing satellite NO_2 measurements, allowed for us to get sufficiently accurate multiannual estimates of the NO_x emissions in a computationally efficient way. In particular, we have avoided multiannual simulations, which would be computationally too expensive in our case. Taking these assumptions into account, $\mathbf{C}_m$ and $\mathbf{C}_m^b$ could be calculated in one year-round model run. To avoid specific assumptions regarding the seasonal cycle of anthropogenic NO_x emissions, the simulations are performed without seasonal variation of the anthropogenic NO_x emissions (that is, with $E_{mi} = \text{const}$ for a given year). A possible seasonal variation of the real anthropogenic NO_x emissions in the given region is, however, not disregarded, and is taken into account in the satellite measurement inversions defined by Eq. (6).

Accordingly, after setting $E_{mi} = \text{const}$ in Eq. (6), the ratio of NO_x emissions in two different years can be estimated as follows:

$$\frac{E_t^{(NO_x)j}}{E_t^{(NO_x)k}} \approx \frac{\sum_{i=1}^{12}\left[\frac{(C_i^j - C_{mi}^{bk})}{(C_{mi}^k - C_{mi}^{bk})}\zeta_{ij}^k\right]}{\sum_{i=1}^{12}\left(\frac{C_i^k - C_{mi}^{bk}}{C_{mi}^k - C_{mi}^{bk}}\right)}, \; j,k \in \{1996..2008\}. \quad (7)$$

By fixing a value of k, we define a reference year and obtain time series of NO_x emissions related to those in the reference year. If the NO_2 lifetime values for different years were the same, the values of ζ defined by Eq. (5) could be assumed to be equal unity ($\zeta = 1$), and Eqs. (4), (6), and (7) could be accordingly simplified. In reality, however, even when the above assumption (4) are correct, some changes in the NO_2 lifetime can be caused by chemical nonlinearities as a result of seasonal and annual changes in anthropogenic emissions. Taking into account that the effects of chemical nonlinearities in our case were found to be rather small (see the corresponding discussion in Sect. 5), we approximated monthly values of ζ for different years by linearly interpolating between unity and its values calculated (employing Eq. 3 to express α_m) with $j = 2008$ and $k = 1996$ by using the EDGAR v4.2 emission data for the respective years. An impact of seasonal variability of emissions on the NO_2 lifetime is disregarded. Note that in most of previous studies employing satellite NO_2 measurements to infer NO_x emissions and their changes (e.g., Martin et al., 2003; Konovalov et al., 2006; Wang et al., 2007; Ghude et al., 2008; Lin et al., 2010; Lin, 2012), the effects of chemical nonlinearities were neglected entirely.

To estimate annual combustion CO_2 emissions, $\boldsymbol{E}_t^{(CO_2)}$, from the NO_x emissions, we define the average NO_x-to-CO_2 emission conversion factor (for a given year j) as

$$F^j = \frac{E_t^{(CO_2)j}}{E_t^{(NO_x)j}}, \; j \in \{1996..2008\}. \quad (8)$$

The ratio of CO_2 emissions in two different years can then be estimated as follows:

$$\frac{E_t^{(CO_2)j}}{E_t^{(CO_2)k}} \approx \frac{F^j E_t^{(NO_x)j}}{F^k E_t^{(NO_x)k}}, \; j,k \in \{1996..2008\}, \quad (9)$$

where the ratio of the NO_x emissions is defined by Eq. (7). Our final goal is the independent evaluation of combustion CO_2 emission trends by means of different approximations of the relative annual CO_2 emission estimates specified by Eq. (9). The simulations and corresponding estimations were made independently with $k = 1996$ and $k = 2008$. The corresponding different estimates of the emission trends were found to be almost indistinguishable; for definiteness, the results reported below are obtained with $k = 1996$.

In this study, estimates of the conversion factors are prescribed from the EDGAR v4.2 inventory for the baseline estimates and, additionally, from the REAS (v1.11 and v2.1) inventory (see Sect. 2.2). The behavior of the emissions factors along with the evolution of the corresponding NO_x and CO_2 emissions in China in the period from 1996 to 2008 are presented in Fig. 2. It is noteworthy that according to both EDGAR and REAS, the long-term changes in the NO_x and CO_2 emissions are rather similar (especially before 2003). This fact illustrates the usefulness of considering NO_x emissions as a proxy for CO_2 emissions. Some deviation in the NO_x and CO_2 emission changes after 2003 (and a corresponding increase of the conversion factor) is mainly due to the introduction of NO_x emission abatement measures (catalytic reduction or combustion modification), as reported e.g., by Zhang et al. (2007).

Note that in addition to the assumption listed above, our simulations do not take into account NO_x emissions from lightning and wildfires. The potential impact of these emissions on our estimates is also discussed in Sect. 5. Note also that, in contrast to emissions from wildfires, NO_x and CO_2 emissions from agricultural biomass burning are taken into

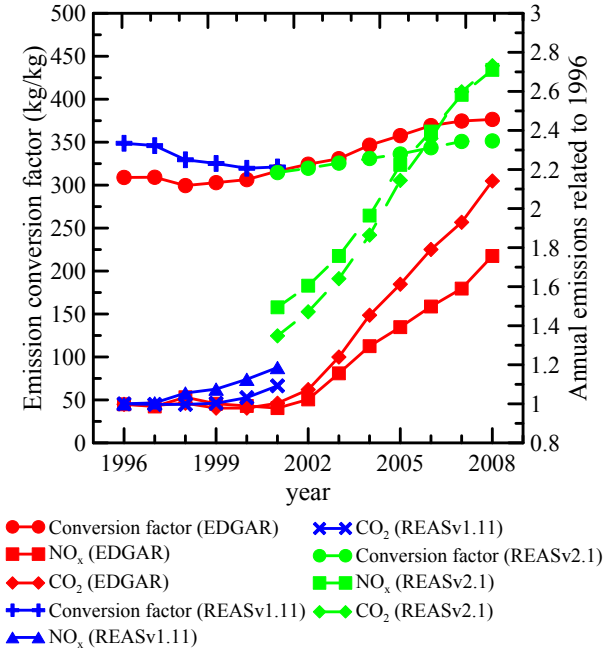


Fig. 2. Multiannual evolution of the CO_2 and NO_x anthropogenic emissions in China and the behavior of the corresponding NO_x-to-CO_2 emission conversion factor F (see Eq. 8) according to the EDGAR v4.2 and REAS (v1.11 and v2.1) emission inventories. The emissions are normalized to their values in 1996. Note that the v1.11 of the REAS inventory was used for the first (1996–2001) period analyzed in this study because the REAS v2.1 inventory was not available for this period.

account in the EDGAR inventory (as the sector 4F) and in our simulations.

The Eqs. (7) and (9) employed in the context of the assumptions listed above and with the NO_x-to-CO_2 conversion factors evaluated with the EDGAR v4.2 inventory constitute the baseline case (case I) of our estimation method. The emission estimates inferred in this way depend on the assumed seasonal variations of the NO_x lifetime that are implicitly evaluated by the chemistry–transport model. However, as it is discussed below in Sect. 4.1, our evaluation yields stronger seasonal variation of anthropogenic emissions than suggested by bottom-up emission inventories, and the possibility that this difference is due to some model uncertainties cannot be definitely ruled out. Accordingly, to take into account possible effects of these uncertainties on our emission trend estimates we consider additional cases (cases II and III) of our estimation method.

Specifically, to assess the impact of the background NO_2 columns on our estimates we consider a hypothetical case (case II) where $\mathbf{C}_{\mathrm{m}}^{\mathrm{b}}$ in Eq. (7) is set to be zero. Effectively, this case also addresses a situation where the relative changes in the background part of the measured NO_2 columns are the same as the relative changes in their anthropogenic part (including both seasonal and multiannual changes). Indeed, if any relative variations in the background and anthropogenic part of NO_2 columns were identical, then there would be no need to differentiate between them or to subtract the background from the total columns in order to evaluate trends in the anthropogenic emissions.

Case III is specified in the same way as case I except that the seasonal variation in the measured NO_2 column amounts in a reference year is assumed to be entirely due to a corresponding variation in the NO_x lifetime (or in other words, it is assumed that the seasonal variation of anthropogenic NO_x emissions is absent). To satisfy this assumption, $\mathbf{C}_{\mathrm{m}}$ in the formulations discussed above is replaced with $\mathbf{C}$ for the reference year. Accordingly, after modifying Eq. (7), we obtain in such a case

$$\frac{E_{\mathrm{t}}^{(\mathrm{CO_2})j}}{E_{\mathrm{t}}^{(\mathrm{CO_2})k}} \approx \frac{F^j}{12F^k}\sum_{i=1}^{12}\left[\frac{(C_i^j - C_{\mathrm{m}i}^{\mathrm{b}k})}{(C_i^k - C_{\mathrm{m}i}^{\mathrm{b}k})}\zeta_{ij}^k\right], \quad j,k \in \{1996..2008\}. \tag{10}$$

Similar to the estimates provided by Eqs. (7) and (9), the sensitivity of the trends derived from Eq. (10) to the choice of the reference year is found to be rather small (in comparison with statistical uncertainties of the trends). For definiteness, we choose $k = 1996$, and evaluate ζ as described above.

Finally, to address the possible uncertainties in the NO_x-to-CO_2 conversion factors, we consider case IV, where the conversion factor is calculated with the REAS inventory. We believe that these such defined four variants of our estimation procedure characterize the range of possible systematic uncertainties in our estimates, although the exact bounds for systematic uncertainties (see also the discussion in Sects. 4.1 and 5) can hardly be determined.

To evaluate emission trends, we built exponential approximations ($A\exp(\kappa t)$, with A and κ being the optimized parameters of the fits, and t being the time in years) of the annual estimates provided by Eqs. (7) and (9) independently for the two periods 1996–2001 and 2001–2008. The approximations were not constrained with any a priori information. The two periods were chosen taking into account a sharp bend in the time series of the EDGAR v4.2 emissions for China (see Fig. 2) between 2000 and 2002. The uncertainty of the exponential trends was estimated with the 95 % confidence level under the typical assumption that deviations from the approximations satisfy the normal distribution. Note that while using an exponential approximation, we assume that a relative rate of actual emission changes is nearly constant. This rate (in units of percent per year) is estimated as a value of the coefficient κ. Such estimation is valid in our case since the obtained values of κ are much smaller than unity.

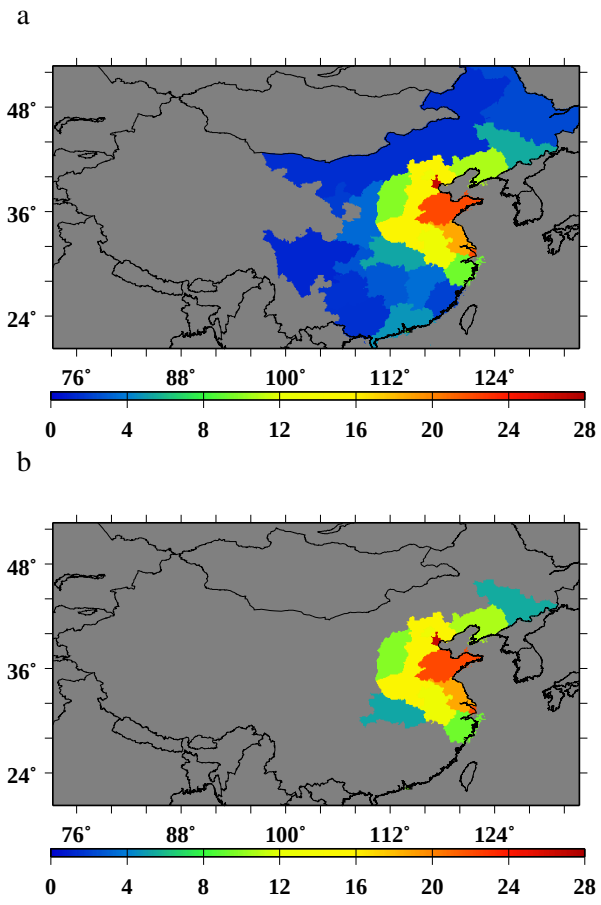
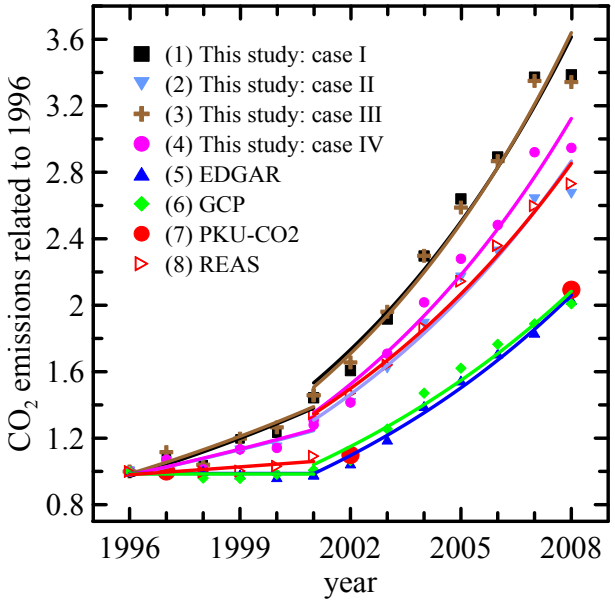


Fig. 3. The provinces of China taken into account in the analysis of the spatial distribution of emission changes for different threshold levels, $[NO_2]_c$, of the (depicted in the plots) annual mean measured tropospheric NO_2 column amounts for year 2008: **(a)** $[NO_2]_c = 1 \times 10^{15}$ molecules cm^{-2}, and **(b)** $[NO_2]_c = 5 \times 10^{15}$ molecules cm^{-2}.



trends (%/year)

1996-2001	2001-2008	1996-2001	2001-2008
(1) 7.0±1.3	12.2±1.0	(5) -0.6±0.3	10.5±0.4
(2) 4.8±1.1	11.2±0.8	(6) -0.1±0.6	9.9±0.7
(3) 7.2±1.4	12.2±0.9	(8) 1.6±0.6	10.4±0.5
(4) 4.9±1.2	11.9±0.9		

Fig. 4. Multiannual exponential total anthropogenic CO_2 emission trends in China derived from satellite measurements and calculated with the EDGAR v4.2, GCP, and REAS (v1.11 and v2.1) data for the periods from 1996 to 2001 and from 2001 to 2008. Note that the REAS v1.11 and v2.1 data are shown for the abovementioned periods, respectively. Additionally, the PKU-CO2 data are shown for three years (1997, 2002, and 2008). All the data are normalized to the values from 1996. Note that only relative changes (not absolute values) of emissions are evaluated in this study; thus the differences between the top-down and bottom-up emission estimates cannot be unambiguously attributed to certain years.

4 Results

4.1 Quantitative analysis of multiannual emission trends

Our estimates presented in this subsection address a large region (see Fig. 3a) covering the eastern industrial provinces of China and providing (according to EDGAR v4.2) more than 98 % of the total anthropogenic CO_2 emissions in China. The definition of the region was based on the administrative provincial map of China so that it includes 27 out of 32 provinces. We selected all provinces where the annual mean and spatially averaged tropospheric NO_2 columns retrieved from the SCIAMACHY measurements in 2008 exceed a threshold value ($[NO_2]_c$) of 10^{15} molecules cm^{-2}. Such a selection allowed for us to disregard the provinces where the anthropogenic contribution to the NO_2 columns is too small compared to contributions of natural factors and where, due to that, our emission estimates would be too uncertain.

The indirect top-down (hybrid) estimates of the multiannual trends in CO_2 emissions (see Eq. 9) are presented in Fig. 4 in comparison with corresponding bottom-up CO_2 emission data of the EDGAR v4.2, GCP, REAS and PKU-CO2 emission inventories. It can be seen that while the EDGAR v4.2 and GCP emission inventories demonstrate zero trends (insignificant negative tendencies) in the period from 1996 to 2001, our estimates in all the cases considered show a strong and significant positive trend (specifically, 7.0 ± 1.3 % per year in baseline case (case I). The difference of our baseline estimate for this period with the CO_2 emission trend calculated with the REAS v1.11 data is smaller but still considerable. This difference cannot be accounted for by the difference between the conversion factors from

the EDGAR v4.2 and REAS v1.11 emission inventories (cf. cases I and IV in Fig. 4).

In the second period (from 2001 to 2008), all the bottom-up and hybrid estimates show large positive trends. Our baseline estimates indicate that the increase of CO_2 emissions in China (12.2 ± 1.0 % per year in case I) during this period was almost twice as large as that in the previous period. The trends in the bottom-up emission inventories fall into the range of divergence and uncertainties of the different hybrid estimates. It may be noteworthy that the trends estimated for case III are only insignificantly different from those estimated for case I. This means that our baseline estimates of the CO_2 emission trends are insensitive to possible uncertainties in the simulated seasonal variation of NO_2 columns.

Note that qualitatively similar results were found with linear approximation of the trends (not shown). Moreover, we found that linear and exponential trend lines are almost indistinguishable. It should be kept in mind that only relative changes rather than absolute values of emissions are evaluated in this study. Although the differences between relative changes of emissions are inevitably associated with a deviation in absolute values (as it is evident in Fig. 4), the deviation in absolute values cannot be unambiguously attributed to certain years. For example, according to our results, the bottom-up CO_2 emissions may, in principle, be quite accurate in 2008 but overestimated in earlier years. It is equally possible that they may be accurate in 1996 and underestimated in later years.

The important result of our analysis is that satellite measurements confirm the accelerating and nonlinear CO_2 emission trend in China, which is manifested by emission inventory data. However, our analysis also reveals strong quantitative differences between the hybrid and bottom-up emission estimates for the first time period evaluated. These differences may be indicative of important uncertainties in the emission inventories, such as major uncertainty in activity data (annual fuel statistics, industrial production statistics, etc.), uncertainty in emission factors, and uncertainty in control abatement.

To clarify major factors contributing to our estimates of CO_2 emission trends, we have performed a similar analysis but in the case of NO_x emissions. The measurement-based estimates of NO_x emissions are obtained using the same formulations as our estimates of CO_2 emissions, but with $F = 1$ (see Fig. 5a). In parallel, we have evaluated trends directly in the measured NO_2 columns (see Fig. 5b) separately in the cold (1 November–31 March) and warm (1 May–30 September) seasons (these seasons are referred to below, for brevity, as winter and summer, respectively). It can be seen that the behavior of the top-down and bottom-up estimates of NO_x emissions is qualitatively and even quantitatively (taking into account the uncertainty of the trends) similar to the behavior of the respective hybrid estimates of CO_2 emissions presented in Fig. 4. This is an expected result, taking into account the assumed behavior of the con-

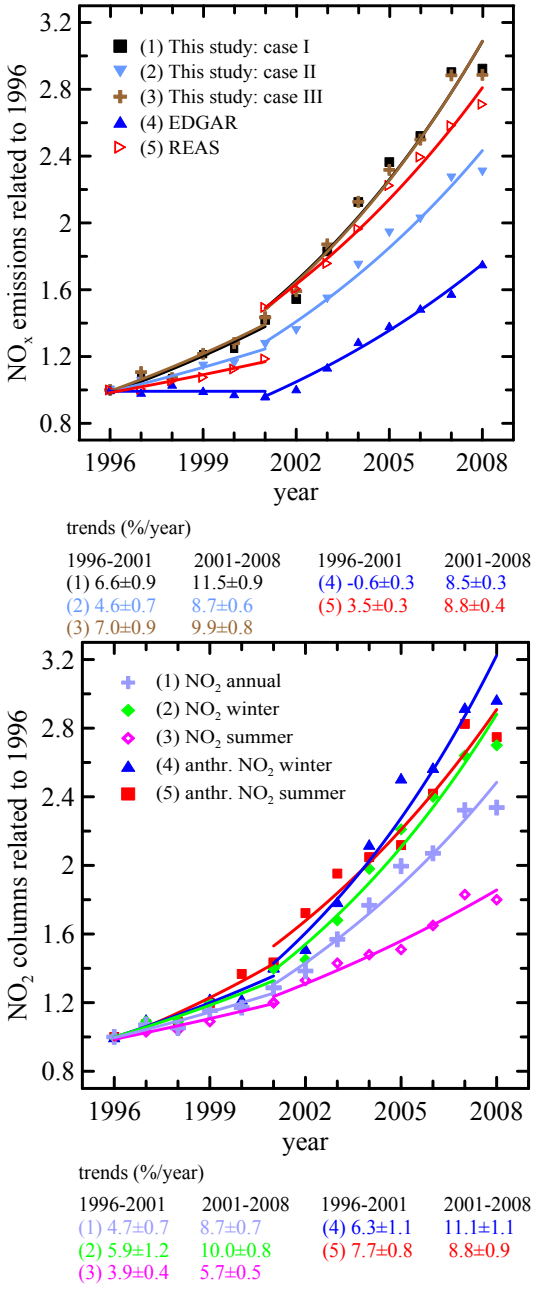


Fig. 5. (a) Multiannual exponential trends in the anthropogenic NO_x emission in China (1–3) derived from satellite measurements in cases I–III of the estimation procedure, respectively, and (4, 5) evaluated using the EDGAR v4.2 and and REAS (v1.11 and v2.1) inventories along with **(b)** multiannual exponential trends in the measured tropospheric NO_2 columns averaged over East China (see Fig 3a) annually (1) and seasonally for the cold (2) and warm (3) seasons. Additionally, the plot **(b)** demonstrates the trends in the estimated anthropogenic part of the measured tropospheric NO_2 columns for the cold (4) and warm (5) seasons. Note that the REAS v1.11 and v2.1 data are shown for the periods from 1996 to 2001 and from 2001 to 2008, respectively. All the data are normalized to the values from 1996.

version factor (see Fig. 2) discussed above. It is noteworthy that although the difference between the trends estimated with and without taking into account the background level of NO_2 columns (that is, the difference between cases I and II) is rather considerable, the trends constrained by the satellite data are statistically different from the corresponding trends of the EDGAR v4.2 NO_x emissions in the period from 1996 to 2001 in the both cases. The difference between the trends derived from satellite data for case II and calculated with the REAS v1.11 data is almost insignificant. Similar to the estimates of the CO_2 emission changes, all of the considered estimates of the NO_x emission changes show large positive trends in the second period. The trends in the bottom-up emission inventories (EDGAR v4.2 and REAS v2.1) fall into the range of divergence of different top-down estimates.

Our results concerning multiannual changes in NO_x emissions in China are qualitatively consistent with the results of earlier studies involving analysis of multiannual satellite measurements over China. In particular, an accelerating and rapid growth of NO_2 column amounts over China in the late 1990s–early 2000s was found by Richter et al. (2005), He et al. (2007), and Zhang et al. (2007). Quantitative comparison of results of the different studies is, however, difficult because of differences in areas and periods considered, as well as in methods of trend evaluation. It is noteworthy that Zhang et al. (2007) compared satellite measurements of tropospheric NO_2 columns with data of an original emission inventory that is based on results of a reanalysis of combustion sources and an improved "dynamic" methodology allowing for changes in both activities and emission factors. Their inventory data (which unfortunately were not available for this study in a numeric format) manifest larger changes in an east-central China region over the period from 1996 to 2001 ($\sim 12\,\%$) than the EDGAR v4.2 and GCP data considered here, but those changes are still much smaller than the corresponding changes in the NO_2 columns derived from GOME measurements ($\sim 31\,\%$). Based on the SCIAMACHY measurements, Lamsal et al. (2011) estimated the annual growth rate in the Chinese NO_x emissions in the period from 2003 to 2006 to be 8 % per year. This estimate is rather close to our estimate for case II ($8.7 \pm 0.6\,\%$ per year), but is significantly smaller than the trend estimated for case I ($11.5 \pm 0.9\,\%$ per year). Although such a comparison can hardly be conclusive because of the difference in the periods considered by Lamsal et al. (2011) and in this study, these results are qualitatively consistent with the fact that a contribution of the background NO_2 (independent on the anthropogenic NO_x emissions) to the measured NO_2 columns was not explicitly taken into account in Lamsal et al. (2011), similar to our case II.

As it is noted in Sect. 3, the different cases of our estimation procedure were specified in order to characterize possible systematic uncertainties in our estimates. In this sense, it is especially noteworthy that the difference between our estimates for cases I and III is almost negligible. This result means that our baseline estimates are almost insensitive to the estimated or assumed seasonal variation of anthropogenic NO_x emissions. The reason for this insensitivity is illustrated in Fig. 5b. It is seen that although the trends in the NO_2 columns for the cold season are much larger when compared to those for the warm season, the trends in the anthropogenic part of NO_2 columns (obtained after subtraction of the background NO_2 columns from the total columns) are almost the same both in summer and winter. Accordingly, the trend in the annual anthropogenic emissions turns out to be insensitive to the assumed contribution of NO_x emissions changes in different seasons. Note that, taking into account the seasonal variations in the measured and simulated NO_2 columns (see Fig. 6a), our results indicate that the most plausible explanation for the slower growth of the (total) tropospheric NO_2 columns in summer is a significant contribution of biogenic emissions (which probably do not experience any significant trends; see also Sect. 5) to the tropospheric NO_2 columns during the warm season. Some additional explanations of the seasonal difference between the trends in (total) tropospheric NO_2 columns derived from satellite measurements are discussed in Zhang et al. (2007).

The seasonal cycles of NO_x emissions for the years 1996 and 2008 estimated in the base case (case I) and in the additional test case (case II) of our method are shown in Fig. 6b. The anthropogenic NO_x emissions are estimated in both cases to be considerably larger in winter than in summer. Specifically, the ratio of the maximum and minimum monthly NO_x emissions is found to be of 3.2 and 1.7 in 1996 and 4.2 and 3.5 in 2008 in cases I and II, respectively. The considerable increase in the amplitude of the estimated seasonal variation of NO_x emissions from 1996 to 2008 in case II is consistent with the strong seasonal differences in multiannual changes of NO_2 columns. That is, if the background NO_2 is not explicitly taken into account (as in case II), the strong increase in NO_2 columns in winter is "converted" into the increase of the NO_x emission seasonal variation.

Our baseline estimates of the ratios of the maximum and minimum monthly anthropogenic emissions in China are much larger than available estimates of the similar ratios based on the bottom-up inventories. Specifically, the ratio of total anthropogenic emissions in China in December to emissions in July is of 1.3 according to Zhang et al. (2007) and of 1.2 according to an earlier study (based on similar statistical data) by Streets et al. (2003). Somewhat larger maximum monthly ratios are derived for fuel combustion emissions in East Asia (~ 1.5) by Jaeglé et al. (2005, Fig. 6c) and for total emissions (including biogenic soil emissions) for a northern China region (~ 2.1) by Wang et al. (2007, Fig. 7a) from the GOME NO_2 data using the GEOS-CHEM global chemistry–transport model for estimation of the NO_x lifetime. Note that our case II estimate for the year 1996 is much more similar to the estimate made by Wang et al. (2007, Fig. 7a) for the years 1997–2000 than our case I estimates.

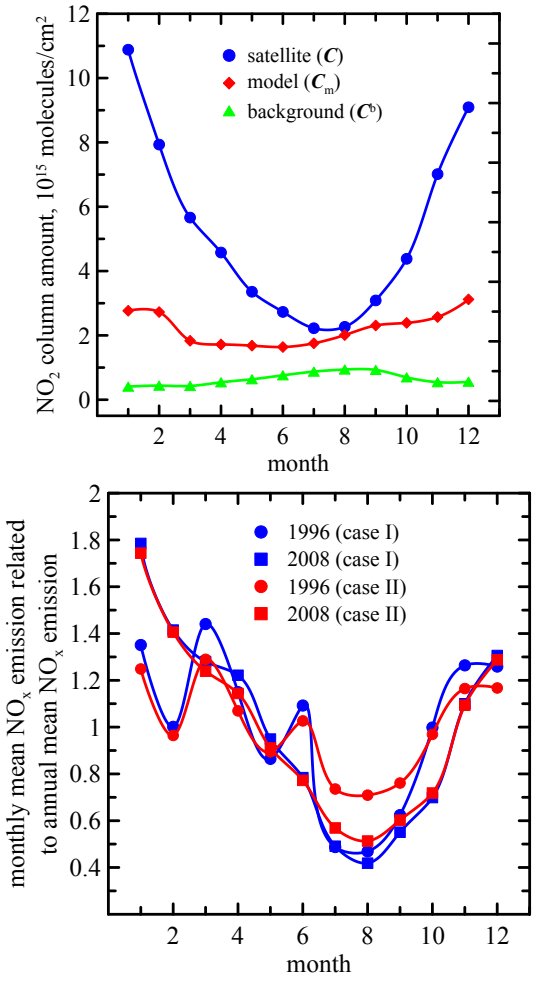


Fig. 6. The seasonal variation in NO_2 column amounts derived from satellite measurements (C) and simulated with CHIMERE with (C_m) and without anthropogenic NO_x emissions (C^b) in China **(a)** and the estimated seasonal cycle of anthropogenic NO_x emissions in 1996 and 2008 for cases I and II of the estimation method **(b)**. The NO_2 columns and NO_x emissions are averaged over the eastern China region shown in Fig. 3a. Note that C_m are calculated without any seasonal variation in anthropogenic emissions. Note also that oscillations in the plot **(b)** are most probably due to uncertainties in the monthly emission estimates.

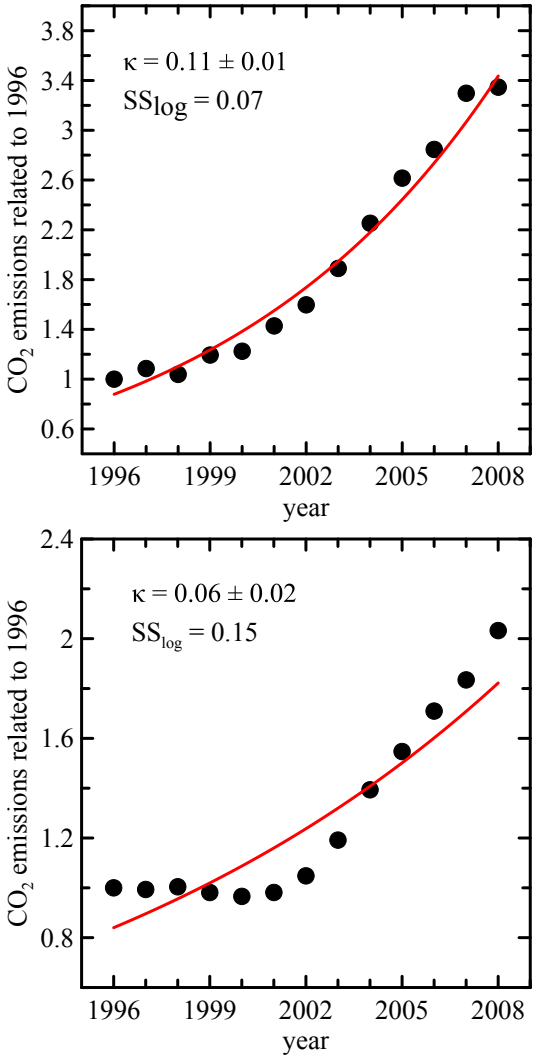


Fig. 7. Multiannual time series of the normalized CO_2 total anthropogenic emissions in China obtained from **(a)** satellite measurements for case I of the estimation procedure and **(b)** the EDGAR v4.2 emission inventory, along with corresponding exponential approximations over the whole period from 1996 to 2008.

This could be expected because case II effectively addresses the estimates of the seasonal cycle in total (anthropogenic plus biogenic) NO_x emissions that were evaluated by Wang et al. (2007). The wintertime maximum in the total NO_x emissions should indeed be less pronounced than that in anthropogenic emissions because biogenic (microbial) emissions in China have a distinctive maximum a summer.

We cannot exclude, however, that the seasonal variation of the anthropogenic NO_x emissions in our case I estimation may be overestimated due to some unidentified systematic overestimation of the background NO_2 columns (e.g., due to overestimated biogenic NO_x emissions) in the model or underestimation of the tropospheric NO_2 columns in the measurements. As explained above, we try to ensure that the main results of this study are sufficiently robust by considering simultaneously several different variants of our estimation procedure. The results presented above indicate that the background NO_2 columns and corresponding trends for cases I and III are more likely to be overestimated than underestimated. Indeed, further significant increase of the background NO_2 columns in our estimations would lead to a much stronger trend in the anthropogenic NO_2 emissions in summer than in winter. However, such a considerable positive difference between the trends in summer and winter could not be easily explained (e.g., by considering the dynamics of emission from major economic sectors; see Sect. 4.2) and

would be in contradiction with the available bottom-up NO_x emission estimates for China (Zhang et al., 2007), which demonstrate very similar changes both in winter and summer. On the contrary, smaller (than in case I) or increasing in time background NO_2 columns would enable diminishing of the difference between our estimates of the seasonal cycles in NO_x emissions and the abovementioned estimates based on the bottom-up information (as in case II). An increase in the background NO_2 in time could be due to an increase in soil NO_x emissions (see Sect. 5), which is probably much smaller than that in anthropogenic emissions but still is not entirely negligible. As has already been mentioned above, a positive difference between the trends in the estimated anthropogenic part of the measured NO_2 columns in winter and in summer could partly be explained in such a case by the factors discussed by Zhang et al. (2007). Therefore, based on the analysis described in this section, and also taking into account possible contributions of some other kinds of systematic uncertainties (see Sect. 5) to our estimates, we believe that actual trends in the anthropogenic NO_x emissions in China are most probably bounded by our estimates obtained for cases I and II.

4.2 Analysis of nonlinear features of the multiannual emission trends

A prominent feature of the time series of the bottom-up CO_2 emission estimates is the abovementioned sharp bend in the period from 2000 to 2002. Visually, this feature underlines the existence of the two different regimes of CO_2 emission evolution in China: the first regime is associated with a neutral/slightly decreasing CO_2 emission trend, while the second regime corresponds to a strongly positive and almost linear trend. Such different regimes should likely be associated with some fundamental changes in economic activities. Although our results confirm that the CO_2 emission trend was accelerating in China during the considered period, the difference between the trends in the periods 1996–2001 and 2001–2008 in the case of the top-down estimates is much smaller than that in the case of the bottom-up estimates.

The considerable difference in the behaviors of the hybrid vs. the bottom-up CO_2 emission estimates is further demonstrated in Fig. 7. Specifically, we have again fitted our estimates (for case I) and the EDGAR v4.2 data with exponential curves, but this time without splitting the whole period 1996–2008 into two stages. We considered as criteria for goodness of the fit (i) the uncertainty of the coefficient k (see Sect. 3) and (ii) the sum of squares of deviations of logarithms of the fitted data from the corresponding fits, SS_{log}. The obtained exponential fits are shown in Fig. 7. Both the criteria indicate that the dynamics of our hybrid emission estimates is much more consistent with a smooth exponential trend than the behavior of the EDGAR data. In other words, these results show that, in agreement with a simple visual inspection of the time series of our estimates, the relative rate of emission changes is accelerating much more smoothly in the case of the satellite-based emission estimates than in the case of the emission inventory data between 2000 and 2002. Therefore, we can conclude that the existence of the discussed sharp bend in emission time series is not corroborated by satellite measurements.

The origin of the strong nonlinearity in the CO_2 and NO_x emission trends found in the emission inventory data can be better understood after considering the dynamics of individual sectors of the EDGAR v4.2 emissions for China. The anthropogenic CO_2 and NO_x emissions from different sectors and their fractions in the total anthropogenic emissions are presented in Fig. 8 as a function of time. In addition, Fig. 8 also shows the ratio of CO_2 emissions to NO_x emissions in different sectors; these ratios are analogous to the NO_x-to-CO_2 emission conversion factor shown in Fig. 2. It is evident that according to EDGAR v4.2 the dominant sectors (at least since 2003) providing up to about 70 % (80 %) of the total CO_2 (NO_x) emissions in China are the sectors "public electricity and heat production" (PEHP) and "manufacturing industries and construction" (MIC), although the "residential and other" sectors also provide an important contribution to the CO_2 emissions. Considering the dynamics of the normalized emissions in the most important sectors, we find that the quasi-neutral CO_2 and NO_x emission trends in the period from 1996 to 2001 are due to compensation of the growth of emissions in the PEHP sector by the decrease of emissions in the MIC sector. After 2001, there is an acceleration of the growth in the PEHP sector, and an upward tendency appears also in the MIC sector. The combined result of these changes is the appearance of the sharp bend of the total CO_2 and NO_x emissions in the period from 2001 to 2002.

Importantly, the dynamics of both CO_2 and NO_x emissions in the major sectors looks very similar. This fact explains the similarity of the evolution of the total CO_2 and NO_x emissions in the considered period (see Fig. 2), and justifies using measurement-based information on NO_x emissions for evaluating changes in CO_2 emissions. There are, however, some quantitative differences in the evolutions of the CO_2 and NO_x emissions in the same sectors. In particular, it may be noteworthy that CO_2 emissions grow considerably faster than the NO_x emissions in the PEHP and the road transport sectors. These differences are reflected in the behaviors of both the CO_2-to-NO_x emission ratios shown in Fig. 8e and the (to a smaller degree) NO_x-to-CO_2 emission conversion factor shown in Fig. 2.

Unlike CO_2 emissions from the major sectors (PEHP and MIC), CO_2 emissions from the "other sectors" do not exhibit any strong nonlinearity, and therefore that cannot contribute to the strong nonlinearity of the total CO_2 emissions. The behavior of NO_x emissions from the "other sectors" is similar to that of CO_2 emissions except for a deviation in 2003. This deviation has no impact on the discussed behavior of the total NO_x emissions in the period from 2000 to 2002.

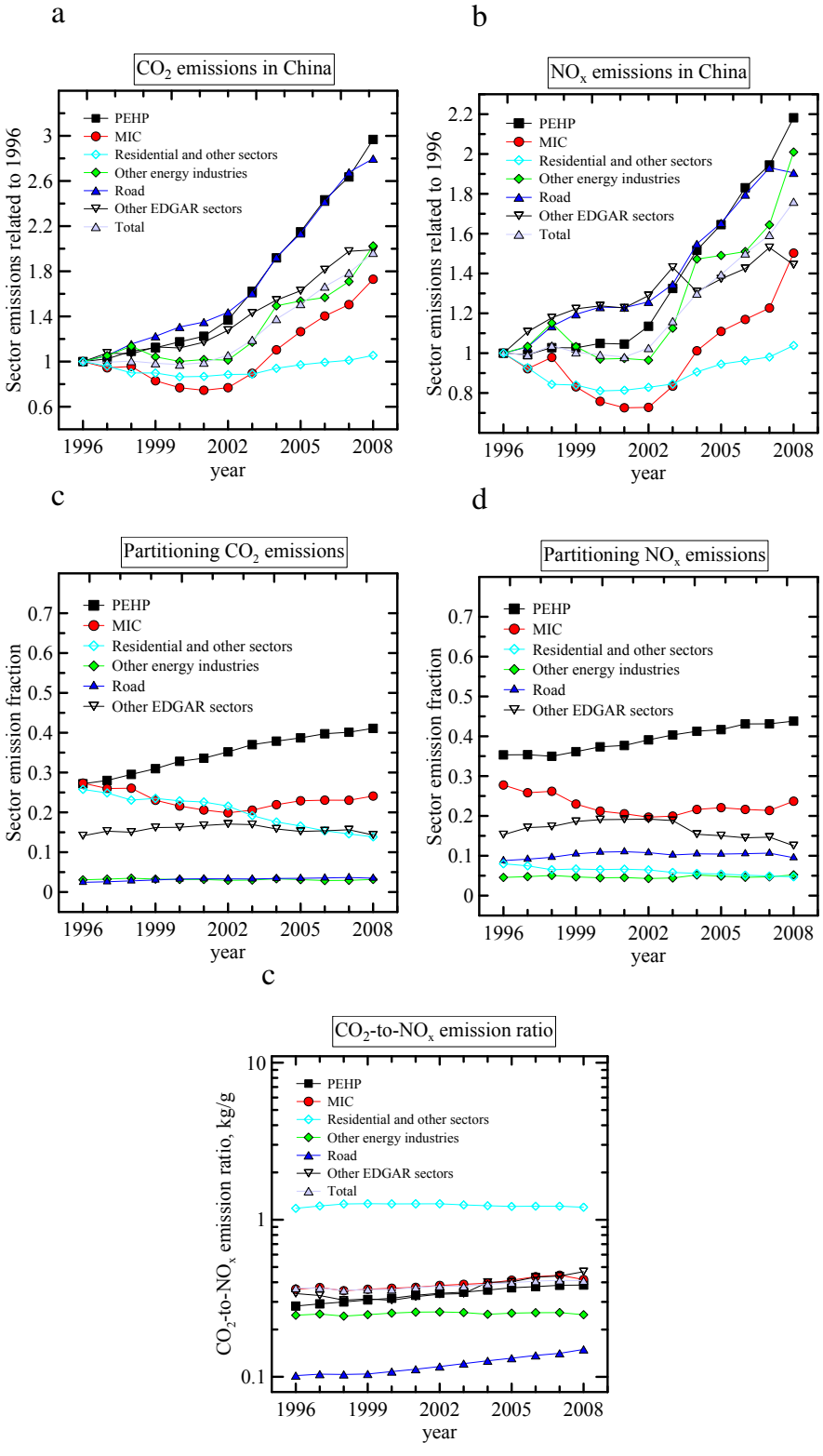


Fig. 8. The annual **(a)** CO_2 and **(b)** NO_x emissions from different anthropogenic emission sectors in China, **(c, d)** their partitioning among the emissions sectors according to the EDGAR v4.2 inventory, and **(e)** the CO_2-to-NO_x emission ratio (analogous to the NO_x-to-CO_2 emission conversion factor shown in Fig. 2) according to the EDGAR v4.2 inventory. The emission data are normalized to the values from 1996. PEHP: public electricity and heat production; MIC: manufacturing industries and construction.

We can conclude that if the contributions of the PEHP and MIC sectors to the total emissions from China are indeed predominant, then our estimates presented above can be considered as evidence that the acceleration of emission changes in these sectors in the period from 2000 to 2002 was much smaller than suggested by the emission inventories. One reason can be found in the activity data, which are for NO_x and CO_2 mainly coming from international energy statistics. It is commonly seen that fuel statistics do not operatively follow up rapid changes, but suffer a short changeable time lag, in particular in the case of fuel shifts, which have happened with residual fuel oil and bituminous coal in the Chinese cement production sector. Due to this inconstant time lag, the rapid changes that appeared in emission inventory data in the period from 2000 to 2002 might actually occur earlier and be smoother. At the same time, our results presented in the previous section confirm that the rates of emission changes in periods from 1996 to 2001 and from 2001 to 2008 were indeed significantly different, as is indicated by all of the considered emission inventories.

4.3 Analysis of the spatial structure of the long-term emission changes

The results presented in the previous section address changes of emissions in China on the national scale. Taking into account previous studies it seems reasonable to expect that, due to the short atmospheric lifetime of NO_2, satellite measurements of tropospheric NO_2 columns can reflect the changes of NO_2 (and, correspondingly, CO_2) emissions on even finer regional scales. Specifically, we performed estimation of the CO_2 and NO_x emissions changes by applying Eq. (9) to the data averaged separately over different Chinese provinces. Effects associated with chemical nonlinearities were disregarded (that is, ζ was set to be equal unity). Spatial aggregation of our emission estimates on the provincial level gives us an opportunity to use for comparison the province-averaged data of the PKU-CO2 inventory. Rather than evaluating emission trends (which is impossible to do with the PKU-CO2 data available only for three years for this study), we have estimated the ratios of emission changes for the pairs of years 2008/2002 and 2002/1997. The NO_x-to-CO_2 emission conversion factors involved in such estimation were evaluated in accordance to Eq. (8) separately for the years 1997, 2002 and 2008 with the EDGAR v4.2 inventory, as in case I of our estimation method. To reduce the random uncertainties, the top-down estimates were preliminary averaged over three consecutive years around the indicated years. That is, to get a more reliable estimate for, e.g., the year 2008, we have averaged the annual emission estimates over the years 2007, 2008, and 2009.

The magnitudes of the ratios of the top-down CO_2 emission estimates in the year 2008 to those in 2002 are presented in Fig. 9, and also plotted in Fig. 10 versus the similar ratios calculated with the EDGAR v4.2 and PKU-CO2 inventories. The scatterplots are presented separately for the two subsets of provinces depicted in Fig. 3. The first subset of provinces (see Fig. 3a) has been considered above. The provinces in the second subset feature considerably larger threshold magnitudes of mean NO_2 columns ($[NO_2]_c = 5 \times 10^{15}$ molecules $\cdot$ cm^{-2}) than the first subset ($[NO_2]_c = 1 \times 10^{15}$ molecules $\cdot$ cm^{-2}).

It is evident that any positive correlation between our estimates of the emission ratios for the years 2008/2002, and the EDGAR v4.2 data ratio is absent. Negligible or negative correlation takes place also between the EDGAR and PKU-CO2 data. These results reflect the fact that according to EDGAR v4.2, the spatial distribution of the emission changes is almost constant over these six years, while both of our hybrid estimates and the PKU-CO2 bottom-up data for different provinces are rather inhomogeneous (see Fig. 9). Note that the distribution of the EDGAR v4.2 emission data is performed with sector-specific spatial proxy datasets that are kept constant over time. As such, Fig. 9b visualizes only the impact of relative change in fraction of the different sectors, but not yet any urbanization process that might explain the spatial inhomogeneity of the emission changes. The agreement of our estimates with the PKU-CO2 data is noticeably better (although yet far from perfect). Specifically, the correlation coefficient (r) is of 0.23 and 0.46 for the first and second subsets of provinces (with the number of provinces included being 28 and 14), respectively. A likely explanation of the larger correlation coefficient for the smaller subset of highly polluted provinces is that smaller magnitudes of NO_2 columns are associated with larger relative uncertainties caused by absolute errors in the retrievals (e.g., due to a bias in estimation of a stratospheric part of the measured total NO_2 columns) and in simulations (e.g., due to uncertainties in biogenic emissions).

The fact that our estimates demonstrate much better agreement with the data of the PKU-CO2 inventory than with the EDGAR v4.2 inventory may be considered as indication that our estimates are sufficiently reasonable. Indeed, if our estimates were completely noisy, they would not be able to manifest a considerable correlation with any independent data. On the other hand, the same fact can be considered as evidence that the spatial structure of real emission changes in the period from 2002 to 2008 is considerably better reproduced in the PKU-CO2 inventory than in the EDGAR v4.2 inventory. This result could indeed be expected because the PKU-CO2 inventory is based on detailed statistical information for individual counties of China (thus even finer than the provincial level), while the EDGAR v4.2 inventory operates with the national fuel statistics, which are not all distributed to the point sources, but which uses proxy data for, e.g., the residential sector. Interestingly, the correlation coefficient between our estimates of CO_2 emission ratios and the PKU-CO2 data increases from 0.46 to 0.52 when the conversion factors are assumed to be constant both in time and space (this case is not shown). That is, applying

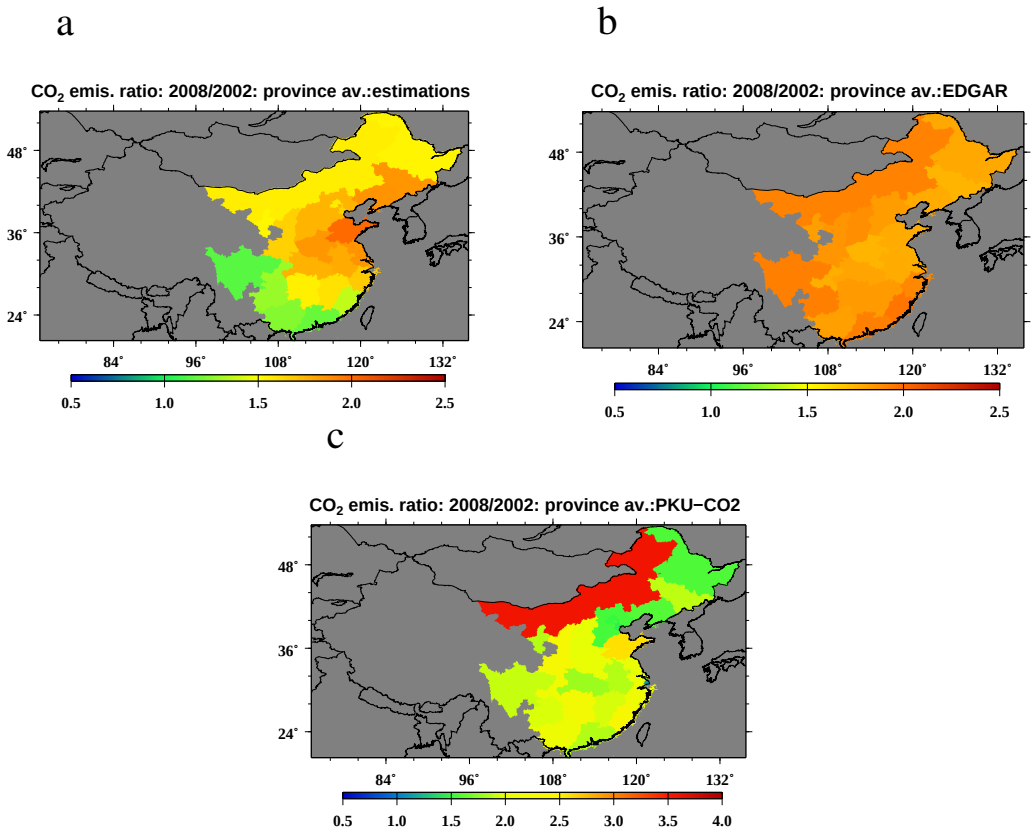


Fig. 9. Magnitudes of the ratio of the top-down satellite-based CO_2 emission estimates in the year 2008 to those in 2002 **(a)** along with the similar ratios calculated with the EDGAR v4.2 **(b)** and PKU-CO2 inventories **(c)**.

the conversion factors from EDGAR v4.2 reduces the correlation rather than increases it. This fact might be considered as indication of uncertainties in the conversion factors, but the difference is rather insignificant to allow for us to make any firm conclusions.

A similar analysis was performed for the ratio between the years 1997/2002. Results of this analysis that are not presented here do not contain any evidence that one of the emission inventories is considerably more accurate than the other over that period. Specifically, correlations with our top-down estimates were found to be almost negligible both with the PKU-CO2 and EDGAR v4.2 data. It seems unlikely that this result is due to a significantly different level of uncertainties in our top-down estimates. A more likely explanation is that both the PKU-CO2 and EDGAR v4.2 data for this period are very uncertain because of serious potential flaws in available statistical information about economic activities. Possible major errors in the statistical information used as input data for inventories of emissions in China have already been discussed in previous publications (Akimoto et al., 2006; Zhang et al., 2007; Gregg et al., 2008; Guan et al., 2012).

5 Discussion

In this section, we discuss possible reasons for the disagreement between our estimates of CO_2 and NO_x emission changes and the data of the emission inventories. Obviously, such disagreement may be either due to (1) biases in our top-down emission estimates, (2) errors in the bottom-up estimates from emission inventories, or (3) uncertainties in the both kinds of estimates. Since the focus of this paper is on the measurement-based emission estimates, possible uncertainties in emission inventory data are discussed only very briefly.

Uncertainties in our top-down estimates of CO_2 emission trends may contain both random and systematic parts. The random part is evaluated above (see Fig. 4) in a standard way as the uncertainty of an exponential fit to the time series of measurement-based estimates. It includes random uncertainties in satellite data as well as cumulative uncertainties associated with any kind of random interannual fluctuations of α and $\mathbf{C}^b$ (see Eq. 2) in the real atmosphere that are not reproduced by our model. Systematic uncertainties may be due to biases and errors in the input-satellite and model data, and may also be associated with the assumptions mentioned

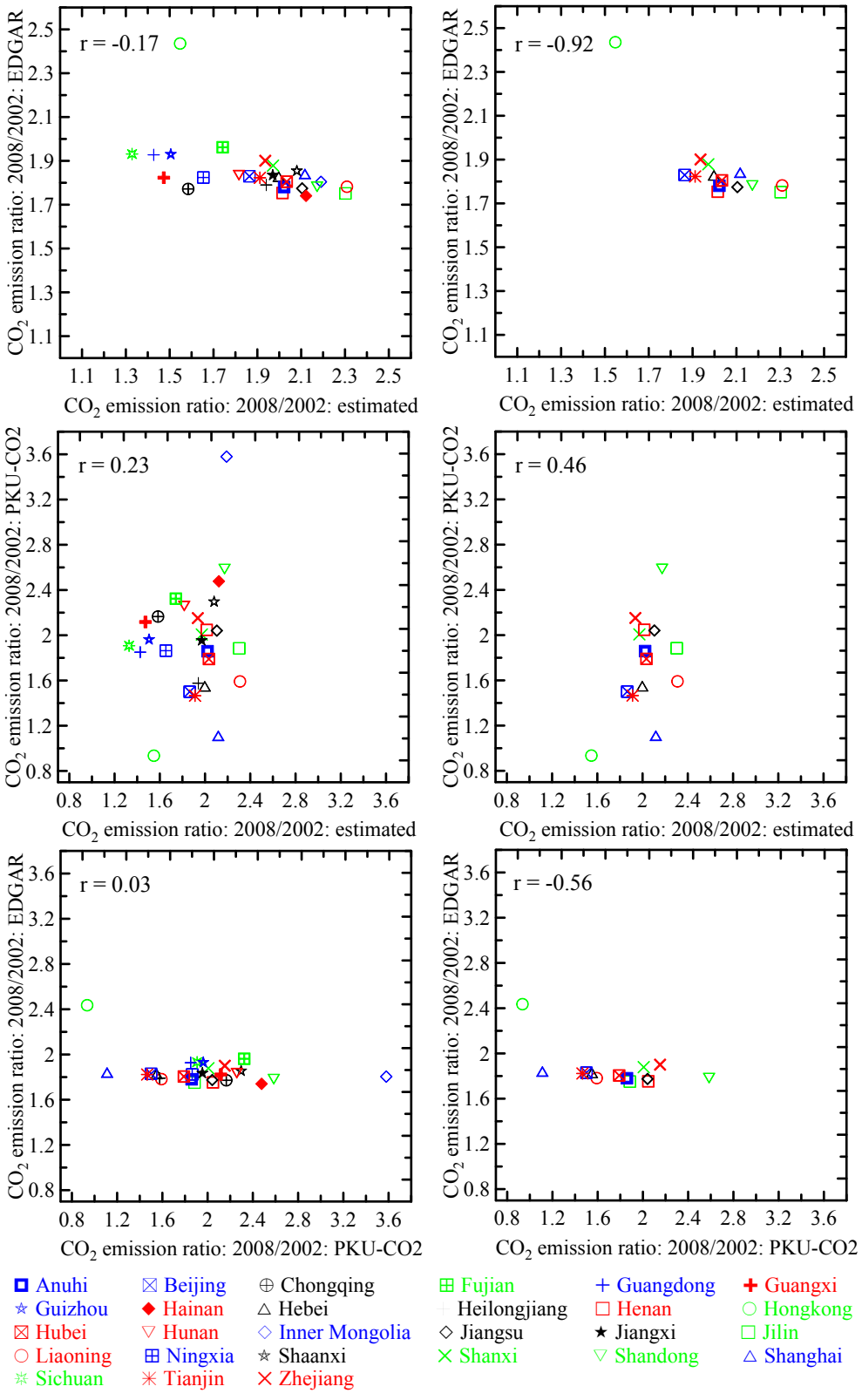


Fig. 10. Scatterplots of the ratios of different CO_2 emission estimates for the years 2008 and 2002: (left) for the eastern provinces depicted in Fig. 3a and (right) for the selected highly polluted provinces indicated in Fig. 3b.

in the previous section. In particular, our estimates may also be affected by systematic uncertainties in the conversion factor $\boldsymbol{F}$.

Systematic uncertainties associated with inverse modeling are usually difficult or even impossible to quantify in a regular way due to their complicated character and the limited knowledge about possible contributing factors. This remark fully applies to the given inverse modeling study as well. Below we discuss possible systematic uncertainties, and examine their potential impact on results of our analysis mainly in a qualitative way; any quantitative estimates provided below are rough and should be considered with caution.

Systematic uncertainties in satellite data include (1) retrieval errors due to neglected changes in optical properties of the atmosphere and the surface and (2) any other unidentified drifts in the data records (e.g., due to aging of satellite instruments). These kinds of uncertainties in the multiannual satellite data for tropospheric NO_2 columns over China were earlier addressed by Richter et al. (2005), who argued that a possible bias introduced by these factors to the trends in NO_2 columns retrieved from the GOME measurements over China is insignificant in comparison to the magnitude of the trend in the period from 1996 to 2002. There is no evidence that this conclusion may be invalid in the case of a longer time series considered in the given study.

The assumptions involved in our simple inverse modeling procedure (see Sect. 3) are based, in particular, on the experience of earlier studies exploiting satellite measurements of tropospheric NO_2 columns. This experience makes us believe that uncertainties associated with these assumptions cannot explain the strong disagreement between our estimates of CO_2 emission trends and corresponding data of emission inventories. Specifically, some potential uncertainties in our estimates can be associated with nonlinearity of the relationship between the NO_x emissions and the tropospheric NO_2 columns. Similar to virtually all earlier studies employing satellite measurements for estimation of NO_x emissions (e.g., Martin et al., 2003; Boersma et al., 2008; Konovalov et al., 2008; Lin et al., 2010; Kim et al., 2006, 2011), we assume that nonlinearity of this relationship is small (even though we do not disregard it completely as explained in Sect. 3). Figure 11 shows values of the NO_2 lifetimes (α) evaluated in CHIMERE for different months with the 1996 and 2008 data of the EDGAR v4.2 emission inventory. It can be seen that indeed, according to the model calculations, the lifetime changes over the considered period are rather small with a magnitude of the maximum difference (in February) being less than 15 %. Note that these lifetime changes reflect not only changes in NO_x emission but also in emissions of volatile organic compounds (VOC). To assess potential nonlinear effects associated with changes of only NO_x emissions, we additionally performed simulations of NO_2 columns with the CHIMERE model under different levels of anthropogenic NO_x emissions. Specifically, the EDGAR v4.2 anthropogenic NO_x emissions for the year

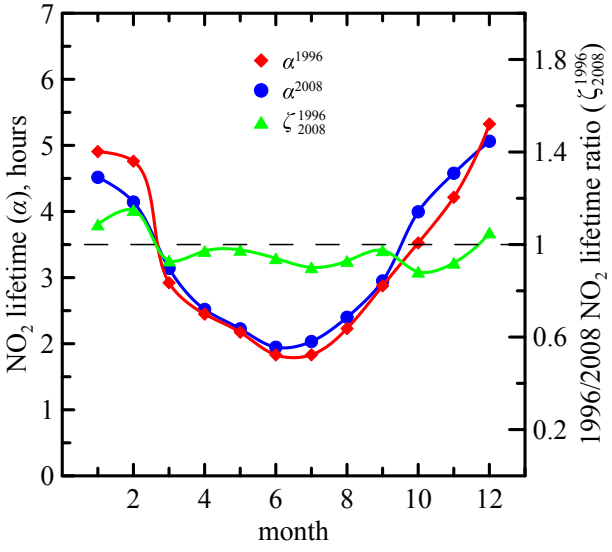


Fig. 11. Monthly values of the lifetime (α; see Eq. 3) of the tropospheric NO_2 columns over the East China as evaluated with the CHIMERE model using the EDGAR v4.2 emission data for the years 1996 and 2008, along with the corresponding values of ζ^{1996}_{2008} (see Eq. 5) for different months. Note that some irregular variations in ζ^{1996}_{2008} are mainly due to meteorological variability.

2008 were scaled with the scale factor ranging from 0.3 to 1. Such scaling represents the broad range of estimated NO_x emission changes in the period from 1996 to 2008 (see Fig. 5a). It is found that although the response of the annual mean NO_2 columns over East China to the NO_x emission changes is not exactly linear, the deviation of the relative changes in the anthropogenic part of the calculated NO_2 columns from the relative changes in the anthropogenic NO_x emissions was rather small (less than 11 %). This result indicates that a probable positive relative bias that could be introduced by chemical nonlinearities in the magnitudes of emission trends presented in Figs. 4 and 5a if the VOC emissions were constant would be unlikely to exceed 11 %, and cannot explain the differences with the bottom-up inventory data.

Note that in principle the nonlinearity of the relationship between NO_x emissions and NO_2 columns may be due to changes in the NO_x lifetime and the NO_2 / NO_x partitioning, which may to some extent compensate each other. In our case, it is found that the increase of NO_x emissions is associated with faster than linear increase of NO_2 columns averaged annually due to the increase of the NO_x lifetime. The changes of the NO_2 / NO_x ratio are found to be quite insignificant (less than 3 %). These results suggest that the atmospheric photochemistry in China is, on the whole, predominantly in the so-called high-NO_x regime (Sillmann, 1999).

Additional simulations were made to examine a possible effect of changes in emissions of volatile organic compounds (VOC) on our estimates. Specifically, along with the baseline simulations performed with the 1996 and 2008 EDGAR v4.2 emissions, we performed two model runs where the VOC emissions were swapped between the 1996 and 2008 datasets. The results of these runs indicate that a small nonlinearity in the response of the annual NO_2 columns to the NO_x emission changes between 1996 and 2008 is almost completely compensated by the corresponding changes in the VOC emissions, since an increase in VOC emissions decreases the NO_2 lifetime, and vice versa. A possible interpretation of these findings, which are in agreement with simulations performed earlier in Konovalov et al. (2010), is that the increase in VOC emissions can lead to an increase of OH concentration and the decrease of α in Eq. (2) due to the increased rate of NO_2 conversion into HNO_3.

It should be noted as a caveat that effects of chemical nonlinearities in the NO_2–NO_x relationship may be sensitive to the model grid resolution (Valin et al., 2011). For example, a considerably larger (than in this study) nonlinearity of the relationship between the NO_2 column and the NO_x emissions in China was reported by Stavrakou et al. (2008): based on global model simulations performed with a $5^\circ \times 5^\circ$ resolution (which is 25 times lower than the spatial resolution of the simulations in this study), they found that the lifetime of the NO_2 columns increases in January (by more than 10 %) and decreases in July (by more than 25 %) due to anthropogenic emissions changes between 1997 and 2006. Note that, in principle, our test results and the results by Stavrakou et al. (2008) may be different due to the differences in the treatment of the background NO_2 columns (specifically, they are excluded in our analysis but apparently not excluded in Stavrakou et al., 2008). Nonetheless, even taking the simulations by Stavrakou et al. (2008) into account, it appears that a decrease of the NO_x lifetime in winter and its increase in summer tend to compensate each other, such that the remaining bias in the derived trend in annual NO_x emissions is unlikely to exceed 15 %.

The impact of NO_x emitted in the previous month on the NO_2 columns in the given month should be quite negligible because of the short lifetime of NO_2 columns over China (see Fig. 11). Similarly, the short lifetime of NO_x prevents it from being transported over large distances. Figure 1 clearly indicates that the NO_2 content over China is indeed determined mainly by national sources, which are strongest in its eastern regions.

The omitted contribution of the NO_x emissions from lightning and wildfires can influence our estimates mainly through underestimation of the background level of the NO_2 content over China ($\mathbf{C}_m^b$). However, our analysis of seasonal differences in the NO_x emission trends (see Sect. 4.1) reveals no indications that $\mathbf{C}_m^b$ is really underestimated. Accordingly, the inclusion of NO_x emissions from lightning (which were recently estimated by Lin (2012) to be about two times smaller than soil emissions in China in 2006) and from wildfires (which, taking into account GFED3.1 (van der Werf et al., 2010) data and estimates from Lin (2012), contribute, on average, less than 10 % to the total natural NO_x emissions) could hardly result in more accurate estimates of the NO_x and CO_2 emission trends in our case. Note that possible uncertainties in $\mathbf{C}_m^b$ may not only be due to uncertainties in the natural emissions but also due to uncertainties associated with other factors and processes (such as boundary conditions, vertical transport scheme, chemistry and deposition). Note also that the magnitude of the annual soil NO_x emissions in East China in CHIMERE (0.37 $\mathrm{Tg\,N\,yr^{-1}}$) is in very good agreement with the recent top-down estimate ($0.38 \pm 65\ \%\ \mathrm{Tg\,N\,yr^{-1}}$) reported by Lin (2012) for the same region, and is also in agreement (taking into account the uncertainty range) with the corresponding bottom-up estimate ($0.50 \pm 25\ \%\ \mathrm{Tg\,N\,yr^{-1}}$) calculated in Lin (2012) by using a new parameterization of soil NO_x emissions developed by Hudman et al. (2012). An impact of possible uncertainties in $\mathbf{C}_m^b$ on the results of our analysis has already been discussed in Sect. 4.1.

Any direct evidence of changes in biogenic NO_x emissions in China during the considered period is not yet available. Nonetheless, the changes in biogenic NO_x emissions are unlikely to exceed the changes in fertilizer consumption, which, according to the World Databank (http://databank.worldbank.org/ddp/home.do), has increased in China in the period from 2002 to 2008 (similar data for an earlier period have not been available) by about 25 % (from 373 up to 468 kg per hectare of arable land). This is three times less than the supposed increase in anthropogenic NO_x emissions (see Fig. 2). Taking into account that the total annual soil NO_x emissions specified in our model for China are from 8 to 15 times smaller than the total Chinese anthropogenic emissions NO_x emissions in the 1996–2008 period, the impact of a possible trend in soil NO_x emissions on our estimates of changes in anthropogenic NO_x and CO_2 emissions is indeed likely to be rather negligible (less than 10 %). An upper limit for the uncertainty associated with a possible trend in soil emissions is assessed above by means of a special case (case II) of our estimation procedure (see Figs. 4 and 5a).

To make sure that our estimates of NO_x and CO_2 emission changes are indeed insensitive to the interannual meteorological variability (which was disregarded in our estimation procedure), we have performed test simulations with meteorological conditions for the years 1997 and 2007. These simulations were performed at an early stage of this study by using the EDGAR v4.1 data for 2005. The years 1997 and 2007 were chosen taking into account the ENSO precipitation index (ESPI) (Curtis and Adler, 2000) values that were predominantly positive during 1997 but was mostly negative during 2007. The ESPI index characterizes the long-term temporal variability of precipitation in the eastern Asia region; it seems reasonable to assume that

fluctuations of precipitation patterns are related to variability of atmospheric circulation driving air pollution transport processes. We found that replacing $\mathbf{C}_m$ and $\mathbf{C}_m^b$ calculated with the meteorology of year 2007 with those calculated for year 1997 in Eq. (7) resulted in very insignificant changes (less than 1 %). This test therefore confirms that the effect of interannual meteorological variability on our estimates of NO_x and CO_2 emission changes can be disregarded.

Finally, large uncertainties (biases), which are, however, difficult to evaluate, may appear in our estimates of the CO_2 emission trend due to errors in the conversion factor $\boldsymbol{F}$. The comparison of the results obtained with the NO_x-to-CO_2 emission conversion factors calculated with the data of the EDGAR and REAS inventories (see results for cases I and IV in Fig. 4) gives some idea about such uncertainty: specifically, the absolute differences between the CO_2 emission trends estimated with the different conversion factors are 2.1 and 0.3 % in the periods from 1996 to 2001 and from 2001 to 2008, respectively. It is important that using different estimates of conversion factors cannot "reconcile" our CO_2 and emission trend estimates with the corresponding EDGAR v4.2 and GCP data for the first period. Moreover, possible uncertainties in the NO_x-to-CO_2 emission conversion factors can hardly explain the sharp bend in the evolution of both CO_2 and NO_x emissions between 2001 and 2003, whose existence is not supported by observational evidence. It is noteworthy that the agreement of both our hybrid estimates of CO_2 emission trends and top-down estimates of NO_x emissions trends is considerably better with the corresponding data of REAS inventory than it is with the data of the EDGAR inventory. This is an expected result taking into account that the regional REAS inventory is based on more detailed information about emission processes in China than the global EDGAR inventory.

Taking into account the arguments outlined above, we can conclude that uncertainties associated with our hybrid method in inferring CO_2 emissions using satellite NO_x measurements and the input data (including the CO_2-to-NO_x emission ratio) used in our analysis can hardly be responsible for all of the considerable differences between the top-down and bottom-up estimates of NO_x and CO_2 emission changes. In our opinion, the most plausible interpretation of at least a part of the differences between our estimates and the global emission inventory data is that the evolution of the activity data for China in the global emission inventories in the first half of the considered period is strongly and negatively biased and shows a time lag in the reporting of fuel consumption growths (which is common in the case of a fuel shift). Specifically, as it was pointed out earlier (Akimoto et al., 200; Gregg et al., 2008; Guan et al., 2012), the available national energy consumption data used in emission inventories to calculate emissions from the major sectors (that is, the "public electricity and heat production" and "manufacturing industries and construction") may be negatively biased due to underestimation of coal production and consumption. A likely reason for this underestimation (as argued in the aforementioned papers) is that activities of an increasing number of small private enterprises dealing with coal mining and electricity generation are not properly reflected in national statistics. In particular, while a large number of small coal mines were "officially" shut down in the late 1990s as a result of a political campaign promoted by the Chinese government, some of them actually continued operating "illegally" (Horii and Gu, 2001). Such a situation can explain a possible underestimation of the rate of CO_2 and NO_x emission change in emission inventories in the period from 1996 to 2001. Interestingly, the highest CO_2 emission estimate provided by Guan et al. (2012) for 2008 is only insignificantly larger than the corresponding estimate provided by EDGAR v4.2. This fact can be regarded as an indication that biases in the EDGAR data with respect to our estimates (see Fig. 4) should probably be mainly attributed to the years before 2008.

A comparison of the emission inventory data with corresponding measurement-based estimates, which is performed in this study, cannot identify any specific problems in the bottom-up emission inventory data (for example, the differences between available bottom-up and our top-down estimates of CO_2 emissions may be, in principle, either due to biases in activity data or in emission factors). Nonetheless, if the emission inventory data were perfectly accurate, then the "bottom-up" and "top-down" estimates of CO_2 emission changes would be in agreement with each other (assuming that the "top-down" estimates are also sufficiently accurate). Since this is not the case in the considered situation, our results indicate that the current multiannual data of CO_2 and/or NO_x emission inventories for China may contain major uncertainties.

6 Summary and conclusions

This study explores the idea of verification of CO_2 emissions data by means of satellite measurements of tropospheric NO_2 column amounts. Indirect estimates of CO_2 emissions are obtained from the measurement-based estimates of NO_x emissions by applying the emission factors from the EDGAR v4.2 and REAS (v1.11 or v2.1) emission inventories. Using a simple inverse modeling method, we evaluate the multiannual changes (trends) in both CO_2 and NO_x anthropogenic emissions in China in the period from 1996 to 2008. The method is based on a linear relationship between NO_x emissions and the NO_2 columns, and it involves the simulations of the "background" NO_2 content in the troposphere and of a part of the seasonal cycle of NO_2 columns, which are not related to the anthropogenic NO_x emissions in China and their seasonal variations, respectively. The simulations are performed with the CHIMERE mesoscale chemistry–transport model. The multiannual trends in our indirect "top-down" estimates of CO_2 emissions are compared

with the corresponding trends calculated with the data of the "bottom-up" EDGAR v4.2 and GCP global emission inventories. The trends are evaluated by means of exponential approximations of the normalized time series of annual emissions separately in the periods from 1996 to 2001 and from 2001 to 2008. Additionally, our estimates of the multiannual CO_2 emission changes are compared with the data of the regional (REAS) and national (PKU-CO2) emission inventories.

Both the indirect top-down and bottom-up emission estimates indicate that the rate of CO_2 emission changes in China was significantly different during the two considered periods (1996–2001 and 2001–2008). Specifically, all the CO_2 emission estimates demonstrate large positive trends for the second period, while smaller trends (slightly negative or positive) are found for the first period.

However, results of our analysis also suggest that nonlinearity of CO_2 emission changes over China is strongly exaggerated in the global emission inventories. Specifically, we found no observational evidence supporting the existence of a sharp bend in the emission time series in the period from 2000 to 2002. There are also significant quantitative differences between the different kinds of the CO_2 emission estimates. In particular, while the top-down estimates exhibit a positive and statistically significant trend in the period from 1996 to 2001, the global emission inventories imply that the trend was slightly negative and statistically insignificant. The performed analysis involving several variants of our estimation procedure indicates that these differences cannot be explained by systematic uncertainties in our baseline estimates. In contrast, some quantitative differences between the strong positive trends in the bottom-up and indirect top-down estimates for the period from 2001 to 2008 are found to be in the range of possible systematic uncertainties in our estimates. It is noteworthy that the differences of our estimates with the REAS (v1.11 and v2.1) inventory data are considerably smaller than those with the EDGAR v4.2 data (for both the periods considered).

Along with the estimates of changes in total national CO_2 emissions in China, we considered the spatial structure of these changes. Specifically, using satellite measurements of tropospheric NO_2 we have estimated the ratios of emission changes for the years 2008 and 2002 in individual provinces of China, and compared them with corresponding ratios obtained with the EDGAR v4.2 and PKU-CO2 inventory data. The comparison has revealed that our estimates agree better with the PKU-CO2 inventory than with the EDGAR inventory. This result can be considered as evidence that the accuracy of our measurement-based estimates of emission changes is at least comparable with the accuracy of the corresponding data of emission inventories. Indeed, since the PKU-CO2 inventory was elaborated using the regional (provincial) energy production and consumption statistics, it is expected to more accurately reflect the spatial structure of emissions than the EDGAR inventory, which is based on total national statistical information.

Although we have attempted to "bracket" our estimates with regard to possible systematic uncertainties, and have concluded that the major discrepancies between our estimates and the data of the global emission inventories are unlikely due to possible uncertainties associated with our method and measurement data, it should be kept in mind that some possible systematic biases are difficult or even impossible to evaluate accurately. Therefore, we recommend considering our estimates with sufficient caution in the same way as the data of "traditional" emission inventories, which may also be subject to potential significant uncertainties. On the other hand, our results clearly indicate that the current knowledge about CO_2 emissions in China is so far insufficiently accurate. Taking into account that possible inaccuracies in emissions from China, which has grown to become recently the world's largest emitter of CO_2, have major implications for climate research and prediction, the results of our study call for further research aimed at verification of emission inventories with various satellite and ground-based measurement data as well as at improving available GHG emission estimates.

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References

Akimoto, H., Ohara, T., Kurokawa J., and Horii N.: Verification of energy consumption in China during 1996–2003 by using satellite observational data, Atmos. Environ., 40, 7663–7667, 2006.

Arellano, A. F., Kasibhatla, P. S., Giglio, L., van der Werf, G. R., and Randerson, J. T.: Top-down estimates of global CO sources using MOPITT measurements, Geophys. Res. Lett., 31, L01104, doi:10.1029/2003GL018609, 2004.

Bergamaschi, P., Frankenberg, C., Meirink, J. F., Krol, M., Dentener, F., Wagner, T., Platt, U., Kaplan, J. O., Körner, S., Heimann, M., Dlugokencky, E. J., and Goede, A.: Satellite chartography of atmospheric methane from SCIAMACHY onboard ENVISAT: (II) evaluation based on inverse model simulations, J. Geophys. Res., 112, D02304, doi:10.1029/2006JD007268, 2007.

Bergamaschi, P., Frankenberg, C., Meirink, J. F., Krol, M., Villani, M. G., Houweling, S., Dentener, F., Dlugokencky, E. J., Miller, J. B., Gatti, L. V., Engel, A., and Levin, I.: Inverse modeling of global and regional CH_4 emissions using SCIAMACHY satellite retrievals, J. Geophys. Res., 114, D22301, doi:10.1029/2009JD012287, 2009.

Bessagnet B., Hodzic, A., Vautard, R., Beekmann, M., Cheinet, S., Honorè, C., Liousse, C., and Rouil, L.: Aerosol modeling with CHIMERE – preliminary evaluation at the continental scale, Atmos. Environ., 38, 2803–2817, 2004.

Boden, T. A., Marland, G., and Andres, R. J.: Global, Regional, and National Fossil-Fuel CO_2 Emissions, Carbon Dioxide Information Analysis Center, Oak Ridge National Laboratory, US Department of Energy, Oak Ridge, Tenn., USA, doi:10.3334/CDIAC/00001_V2012, available at: http://cdiac.ornl.gov/trends/emis/overview_2009, last access: 4 January 2013, 2011.

Boersma, K. F., Jacob, D. J., Bucsela, E. J., Perring, A. E., Dirksen, R., van der A, R. J., Yantosca, R. M., Park, R. J., Wenig, M. O., Bertram, T. H., and Cohen, R. C.: Validation of OMI tropospheric NO_2 observations during INTEX-B and application to constrain NO_x emissions over the eastern United States and Mexico, Atmos. Environ., 42, 4480–4497, doi:10.1016/j.atmosenv.2008.02.004, 2008.

Bovensmann, H., Burrows, J. P., Buchwitz, M., Frerick, J., Nël, S., and Rozanov, V. V.: SCIAMACHY – mission objectives and measurement modes, J. Atmos. Sci., 56, 127–150, 1999.

Brioude, J., Petron, G., Frost, G. J., Ahmadov, R., Angevine, W. M., Hsie, E.-Y., Kim, S.-W., Lee, S.-H., McKeen, S. A., Trainer, M., Fehsenfeld, F. C., Holloway, J. S., Peischl, J., Ryerson, T. B., and Gurney, K. R.: A new inversion method to calculate emission inventories without a prior at mesoscale: application to the anthropogenic CO_2 emission from Houston, Texas, J. Geophys. Res., 117, D05312, doi:10.1029/2011JD016918, 2012.

Burrows, J. P., Weber, M., Buchwitz, M., Rozanov, V., Ladstätter-Weißenmayer, A., Richter, A., DeBeek, R., Hoogen, R., Bramstedt, K., Eichmann, K.-U., Eisinger, M., and Perner, D.: the Global Ozone Monitoring Experiment (GOME): mission concept and first scientific results, J. Atmos. Sci., 56, 151–175, 1999.

Burrows, J. P., Platt, U., and Borrell, P. (Eds.): the remote sensing of tropospheric composition from space: Series Physics of Earth and Space Environments, Springer-Verlag, Berlin-Heidelberg, Germany, 2011.

Chevallier, F., Breon, F.-M., and Rayner, P. J.: Contributing of the orbiting carbon observatory to the estimation of CO_2 sources and sinks: theoretical study in a variational data assimilation framework, J. Geophys. Res., 112, D09307, doi:10.1029/2006JD007375, 2007.

Chin, M., Rood, R. B., Lin, S.-J., Muller, J. F., and Thompson, A. M.: Atmospheric sulfur cycle simulated in the global model GOCART: model description and global properties, J. Geophys. Res., 105, 24671–24687, doi:10.1029/2000JD900384, 2000.

Chin, M., Ginoux, S., Kinne, P., Holben, B. N., Duncan, B. N., Martin, R. V., Logan, J. A., Higurashi, A., and Nakajima, T.: Tropospheric aerosol optical thickness from the GOCART model and comparisons with satellite and sunphotometer measurements, J. Atmos. Sci., 59, 461–483, 2002.

Ciais, P., Dolman, A. J., Dargaville, R., Barrie, L., Bombelli, A., Butler, J., Canadell, P., and Moriyama, T.: Geo Carbon Strategy, Geo Secretariat Geneva, FAO, Rome, 48 pp., 2010.

Curci, G., Palmer, P. I., Kurosu, T. P., Chance, K., and Visconti, G.: Estimating European volatile organic compound emissions using satellite observations of formaldehyde from the Ozone Monitoring Instrument, Atmos. Chem. Phys., 10, 11501–11517, doi:10.5194/acp-10-11501-2010, 2010.

Curtis, S. and Adler, R.: ENSO indexes based on patterns of satellite-derived precipitation, J. Climate, 13, 2786–2793, 2000.

Dufour, G., Wittrock, F., Camredon, M., Beekmann, M., Richter, A., Aumont, B., and Burrows, J. P.: SCIAMACHY formaldehyde observations: constraint for isoprene emission estimates over Europe?, Atmos. Chem. Phys., 9, 1647–1664, doi:10.5194/acp-9-1647-2009, 2009.

EC-JRC/PBL (European Commission, Joint Research Centre/Netherlands Environmental Assessment Agency): Emission Database for Global Atmospheric Research (EDGAR), release version 4.2: available at: http://edgar.jrc.ec.europa.eu (last access: January 2013), 2011.

Enting, I. G.: Inverse Problems in Atmospheric Constituents Transport, Cambridge University Press, 2002.

GCP: Ten Years of Advancing Knowledge on the Global Carbon Cycle and its Management (Authors: Lavinia Poruschi, Shobhakar Dhakal and Josep Canadell), Report No. 7, Tsukuba, 2010; available at: http://www.globalcarbonproject.org/, last access: 3 February, 2012.

Ghude, S. D., Fadnavis, S., Beig, G., Polade, S. D., and van der A, R. J.: Detection of surface emission hot spots, trends, and seasonal cycle from satellite-retrieved NO_2 over India, J. Geophys. Res., 113, D20305, doi:10.1029/2007JD009615, 2008.

Ginoux, P., Chin, M., Tegen, I., Prospero, J., Holben, B., Dubovik, O., and Lin, S.-J.: Sources and global distributions of dust aerosols simulated with the GOCART model, J. Geophys. Res., 106, 20255–20273, doi:10.1029/2000JD000053, 2001.

Gregg, J. S., Andres, R. J., and Marland, G.: China: emissions pattern of the world leader in CO_2 emissions from fossil fuel consumption and cement production, Geophys. Res. Lett., 35, L08806, doi:10.1029/2007GL032887, 2008.

Guan, D., Liu, Z., Geng, Y., Lindner, S., and Hubacek, K.: the gigatonne gap in China's carbon dioxide inventories, Nat. Clim. Change, 2, 672–675, doi:10.1038/nclimate1560, 2012.

Guenther, A. B., Jiang, X., Heald, C. L., Sakulyanontvittaya, T., Duhl, T., Emmons, L. K., and Wang, X.: The Model of Emissions of Gases and Aerosols from Nature version 2.1 (MEGAN2.1): an extended and updated framework for modeling biogenic emissions, Geosci. Model Dev., 5, 1471–1492, doi:10.5194/gmd-5-1471-2012, 2012.

He, Y., Uno, I., Wang, Z., Ohara, T., Sugimoto, N., Shimizu, A., Richter, A., and Burrows, J. P.: Variations of the increasing trend of tropospheric NO_2 over central east China during the past decade, Atmos. Environ., 41, 4865–4876, 2007.

Heald, C. L., Jacob, D. J., Jones, D. B. A., Palmer, P. I., Logan, J. A., Streets, D. G., Sachse, G. W., Gille, J. C., Hoffman, R. N., and Nehrkorn, T.: Comparative inverse analysis of satellite (MOPITT) and aircraft (TRACE-P) observations to estimate Asian sources of carbon monoxide, J. Geophys. Res., 109, D15S04, doi:10.1029/2004JD005185, 2004.

Hodzic, A. and Jimenez, J. L.: Modeling anthropogenically controlled secondary organic aerosols in a megacity: a simplified framework for global and climate models, Geosci. Model Dev., 4, 901–917, doi:10.5194/gmd-4-901-2011, 2011.

Horii, N. and Gu, S. H. (Eds.): Coal industry: development of small coal mines in market transition and its externality, in: Transformation of China's Energy Industries in Market Transition and

its Prospects, Chiba Institute of Developing Economics, 23–62, 2001.

Hourdin, F., Musat, I., Bony, S., Braconnot, P., Codron, F., Dufresne, J.-L., Fairhead, L., Filiberti, M.-A., Friedlingstein, P., Grandpeix, J.-Y., Krinner, G., Levan, P., Li, Z.-X., and Lott, F.: The LMDZ4 general circulation model: climate performance and sensitivity to parametrized physics with emphasis on tropical convection, Clim. Dynam., 27, 787–813, doi:10.1007/s00382-006-0158-0, 2006.

Houweling, S., Breon, F.-M., Aben, I., Rödenbeck, C., Gloor, M., Heimann, M., and Ciais, P.: Inverse modeling of CO_2 sources and sinks using satellite data: a synthetic inter-comparison of measurement techniques and their performance as a function of space and time, Atmos. Chem. Phys., 4, 523–538, doi:10.5194/acp-4-523-2004, 2004.

Houweling, S., Aben, I., Breon, F.-M., Chevallier, F., Deutscher, N., Engelen, R., Gerbig, C., Griffith, D., Hungershoefer, K., Macatangay, R., Marshall, J., Notholt, J., Peters, W., and Serrar, S.: The importance of transport model uncertainties for the estimation of CO_2 sources and sinks using satellite measurements, Atmos. Chem. Phys., 10, 9981–9992, doi:10.5194/acp-10-9981-2010, 2010.

Huang, C., Chen, C. H., Li, L., Cheng, Z., Wang, H. L., Huang, H. Y., Streets, D. G., Wang, Y. J., Zhang, G. F., and Chen, Y. R.: Emission inventory of anthropogenic air pollutants and VOC species in the Yangtze River Delta region, China, Atmos. Chem. Phys., 11, 4105–4120, doi:10.5194/acp-11-4105-2011, 2011.

Hudman, R. C., Moore, N. E., Mebust, A. K., Martin, R. V., Russell, A. R., Valin, L. C., and Cohen, R. C.: Steps towards a mechanistic model of global soil nitric oxide emissions: implementation and space based-constraints, Atmos. Chem. Phys., 12, 7779–7795, doi:10.5194/acp-12-7779-2012, 2012.

Hungershoefer, K., Breon, F.-M., Peylin, P., Chevallier, F., Rayner, P., Klonecki, A., Houweling, S., and Marshall, J.: Evaluation of various observing systems for the global monitoring of CO_2 surface fluxes, Atmos. Chem. Phys., 10, 10503–10520, doi:10.5194/acp-10-10503-2010, 2010.

IPCC: Climate change 2007: synthesis report, Contribution of Working Groups I, II and III to the Fourth Assessment Report of the Intergovernmental Panel on Climate Change, Core Writing Team, edited by: Pachauri, R. K. and Reisinger, A., IPPC, Geneva, Switzerland, 104 pp., 2007.

Jaeglé, L., Steinberger, L., Martin, R. V., and Chancec, K.: Global partitioning of NO_x sources using satellite observations: relative roles of fossil fuel combustion, biomass burning and soil emissions, Faraday Discuss., 130, 407–423, 2005.

Janssens-Maenhout, G., Dentener, F., Van Aardenne, J., Monni, S., Pagliari, V., Orlandini, L., Klimont, Z., Kurokawa, J., Akimoto, H., Ohara, T., Wankmueller, R., Battye, B., Grano, D., Zuber, A., and Keating, T.: EDGAR-HTAP: a Harmonized Gridded Air Pollution Emission Dataset Based on National Inventories, European Commission Publications Office, Ispra, Italy, EUR report No EUR 25229, 40 pp., 2012.

Kadygrov, N., Maksyutov, S., Eguchi, N., Aoki, T., Nakazawa, T., Yokota, T., and Inoue, G.: Role of simulated GOSAT total column CO_2 observations in surface CO_2 flux uncertainty reduction, J. Geophys. Res., 114, D21208, doi:10.1029/2008JD011597, 2009.

Kim, S.-W., Heckel, A., McKeen, S. A., Frost, G. J., Hsie, E.-Y., Trainer, M. K., Richter, A., Burrows, J. P., Peckham, S. E., and Grell, G. A.: Satellite-observed US power plant NO_x emission reductions and their impact on air quality, Geophys. Res. Lett., 33, L22812, doi:10.1029/2006GL027749, 2006.

Kim, S.-W., Heckel, A., Frost, G. J., Richter, A., Gleason, J., Burrows, J. P., McKeen, S., Hsie, E.-Y., Granier, C., and Trainer, M.: NO_2 columns in the western United States observed from space and simulated by a regional chemistry model and their implications for NO_x emissions, J. Geophys. Res., 114, D11301, doi:10.1029/2008JD011343, 2009.

Kim, S.-W., McKeen, S. A., Frost, G. J., Lee, S.-H., Trainer, M., Richter, A., Angevine, W. M., Atlas, E., Bianco, L., Boersma, K. F., Brioude, J., Burrows, J. P., de Gouw, J., Fried, A., Gleason, J., Hilboll, A., Mellqvist, J., Peischl, J., Richter, D., Rivera, C., Ryerson, T., te Lintel Hekkert, S., Walega, J., Warneke, C., Weibring, P., and Williams, E.: Evaluations of NOx and highly reactive VOC emission inventories in Texas and their implications for ozone plume simulations during the Texas Air Quality Study 2006, Atmos. Chem. Phys., 11, 11361–11386, doi:10.5194/acp-11-11361-2011, 2011.

Konovalov, I. B., Beekmann, M., Vautard, R., Burrows, J. P., Richter, A., Nüß, H., and Elansky, N.: Comparison and evaluation of modelled and GOME measurement derived tropospheric NO_2 columns over Western and Eastern Europe, Atmos. Chem. Phys., 5, 169–190, doi:10.5194/acp-5-169-2005, 2005.

Konovalov, I. B., Beekmann, M., Richter, A., and Burrows, J. P.: Inverse modelling of the spatial distribution of NO_x emissions on a continental scale using satellite data, Atmos. Chem. Phys., 6, 1747–1770, doi:10.5194/acp-6-1747-2006, 2006.

Konovalov, I. B., Beekmann, M., Burrows, J. P., and Richter, A.: Satellite measurement based estimates of decadal changes in European nitrogen oxides emissions, Atmos. Chem. Phys., 8, 2623–2641, doi:10.5194/acp-8-2623-2008, 2008.

Konovalov, I. B., Beekmann, M., Richter, A., Burrows, J. P., and Hilboll, A.: Multi-annual changes of NO_x emissions in megacity regions: nonlinear trend analysis of satellite measurement based estimates, Atmos. Chem. Phys., 10, 8481–8498, doi:10.5194/acp-10-8481-2010, 2010.

Kopacz, M., Jacob, D. J., Fisher, J. A., Logan, J. A., Zhang, L., Megretskaia, I. A., Yantosca, R. M., Singh, K., Henze, D. K., Burrows, J. P., Buchwitz, M., Khlystova, I., McMillan, W. W., Gille, J. C., Edwards, D. P., Eldering, A., Thouret, V., and Nedelec, P.: Global estimates of CO sources with high resolution by adjoint inversion of multiple satellite datasets (MOPITT, AIRS, SCIAMACHY, TES), Atmos. Chem. Phys., 10, 855–876, doi:10.5194/acp-10-855-2010, 2010.

Kurokawa, J., Yumimoto, K., Uno, I., and Ohara, T.: Adjoint inverse modeling of NO_x emissions over eastern China using satellite observations of NO_2 vertical column densities, Atmos. Environ., 43, 1878–1887, 2009.

Kurokawa, J., Ohara, T., Morikawa, T., Hanayama, S., Greet, J.-M., Fukui, T., Kawashima, K., and Akimoto, H.: Emissions of air pollutants and greenhouse gases over Asian regions during 2000–2008: Regional Emission inventory in ASia (REAS) version 2, Atmos. Chem. Phys. Discuss., 13, 10049–10123, doi:10.5194/acpd-13-10049-2013, 2013.

Lamsal, L. N., Martin R. V.,Padmanabhan A., van Donkelaar A., Zhang Q., Sioris C. E., Chance K., Kurosu T. P., and

Newchurch M. J.: Application of satellite observations for timely updates to global anthropogenic NO_x emission inventories, Geophys. Res. Lett., 38, L05810, doi:10.1029/2010GL046476, 2011.

Lee, C., Martin, R. V., van Donkelaar, A., Lee, H., Dickerson, R. R., Hains, J. C., Krotkov, N., Richter, A., Vinnikov, K., and Schwab, J. J.: SO_2 emissions and lifetimes: Estimates from inverse modeling using in situ and global, space-based (SCIAMACHY and OMI) observations, J. Geophys. Res., 116, D06304, doi:10.1029/2010JD014758, 2011.

Lin, J.-T.: Satellite constraint for emissions of nitrogen oxides from anthropogenic, lightning and soil sources over East China on a high-resolution grid, Atmos. Chem. Phys., 12, 2881–2898, doi:10.5194/acp-12-2881-2012, 2012.

Lin, J.-T., McElroy, M. B., and Boersma, K. F.: Constraint of anthropogenic NO_x emissions in China from different sectors: a new methodology using multiple satellite retrievals, Atmos. Chem. Phys., 10, 63–78, doi:10.5194/acp-10-63-2010, 2010.

Lott, F., Fairhead, L., Hourdin, F., and Levan, P.: the stratospheric version of LMDz: dynamical climatologies, arctic oscillation, and impact on the surface climate, Clim. Dynam., 25, 851–868, 2005.

Ma, J., Richter, A., Burrows, J. P., Nüß, H., and van Aardenne, J. A.: Comparison of model-simulated tropospheric NO_2 over China with GOME-satellite data, Atmos. Environ., 40, 593–604, 2006.

Martin, R. V., Jacob, D. J., Chance, K., Kurosu, T., Palmer, P. I., and Evans, M. J.: Global inventory of nitrogen oxide emissions constrained by space-based observations of NO_2 columns, J. Geophys. Res., 108, 4537, doi:10.1029/2003JD003453, 2003.

Millet, D. B., Jacob, D. J., Boersma, K. F., Fu, T., Kurosu, T. P., Chance, K., Heald, C. L., and Guenther, A.: Spatial distribution of isoprene emissions from North America derived from formaldehyde column measurements by the OMI satellite sensor, J. Geophys. Res., 113, D02307, doi:10.1029/2007JD008950, 2008.

Miyazaki, K., Eskes, H. J., and Sudo, K.: Global NO_x emission estimates derived from an assimilation of OMI tropospheric NO_2 columns, Atmos. Chem. Phys., 12, 2263–2288, doi:10.5194/acp-12-2263-2012, 2012.

Monteil, G., Houweling, S., Dlugockenky, E. J., Maenhout, G., Vaughn, B. H., White, J. W. C., and Rockmann, T.: Interpreting methane variations in the past two decades using measurements of CH_4 mixing ratio and isotopic composition, Atmos. Chem. Phys., 11, 9141–9153, doi:10.5194/acp-11-9141-2011, 2011.

Müller, J.-F. and Stavrakou, T.: Inversion of CO and NO_x emissions using the adjoint of the IMAGES model, Atmos. Chem. Phys., 5, 1157–1186, doi:10.5194/acp-5-1157-2005, 2005.

Napelenok, S. L., Pinder, R. W., Gilliland, A. B., and Martin, R. V.: A method for evaluating spatially-resolved NO_x emissions using Kalman filter inversion, direct sensitivities, and space-based NO_2 observations, Atmos. Chem. Phys., 8, 5603–5614, doi:10.5194/acp-8-5603-2008, 2008.

Nassar, R., Jones, D. B. A., Kulawik, S. S., Worden, J. R., Bowman, K. W., Andres, R. J., Suntharalingam, P., Chen, J. M., Brenninkmeijer, C. A. M., Schuck, T. J., Conway, T. J., and Worthy, D. E.: Inverse modeling of CO_2 sources and sinks using satellite observations of CO_2 from TES and surface flask measurements, Atmos. Chem. Phys., 11, 6029–6047, doi:10.5194/acp-11-6029-2011, 2011.

Nüß, J. H.: an improved tropospheric NO_2 retrieval for GOME and SCIAMACHY, PhD Thesis, University of Bremen, Germany, 2005.

Ohara, T., Akimoto, H., Kurokawa, J., Horii, N., Yamaji, K., Yan, X., and Hayasaka, T.: An Asian emission inventory of anthropogenic emission sources for the period 1980–2020, Atmos. Chem. Phys., 7, 4419–4444, doi:10.5194/acp-7-4419-2007, 2007.

Olivier, J. G. J., Berdowski, J. J. M., Peters, J. A. H. W., Bakker, J., Visschedijk, A. J. H., and en Bloos, J.-P. J.: Applications of EDGA R. Including a description of EDGAR 3.0: reference database with trend data for 1970–1995, RIVM, Bilthoven, RIVM report No 773301 001/ NOP report No 410200 051, 142 pp., 2001.

Olivier, J. G. J., Van Aardenne, J. A., Dentener, F., Pagliari, V., Ganzeveld, L. N., and Peters, J. A. H. W.: Recent trends in global greenhouse gas emissions: regional trends 1970–2000 and spatial distribution of key sources in 2000, Environ. Sci., 2, 81–99, doi:10.1080/15693430500400345, 2005.

Pak, B. C. and Prather, M. J.: CO_2 source inversion using satellite observations in the upper troposphere, Geophys. Res. Lett., 28, 4571–4574, 2001.

Palmer, P. I., Jacob, D. J., Fiore, A. M., and Martin, R. V.: Mapping isoprene emissions over North America using formaldehyde column observations from space, J. Geophys. Res., 108, 4180, doi:10.1029/2002JD002153, 2003.

Palmer, P. I., Suntharalingam, P., Jones, D. B. A., Jacob, D. J., Streets, D. G., Fu, Q. Y., Vay, S. A., and Sachse, G. W.: Using CO_2:CO correlations to improve inverse analyses of carbon fluxes, J. Geophys. Res., 111, D12318, doi:10.1029/2005JD006697, 2006.

Pétron, G., Granier, C., Khattatov, B., Yudin, V., Lamarque J. F., Emmons, L., Gille, J., and Edwards, D. P.: Monthly CO surface sources inventory based on the 2000–2001 MOPITT satellite data, Geophys. Res. Lett., 31, L21107, doi:10.1029/2004GL020560, 2004.

Rayner, P. J. and O'Brien, D. M.: the utility of remotely sensed CO_2 concentration data in surface source inversions, Geophys. Res. Lett., 28, 175–178, 2001.

Richter, A., Burrows, J. P., Nüß, H., Granier, C., and Niemeier, U.: Increase in tropospheric nitrogen dioxide over China observed from space, Nature, 437, 129–132, doi:10.1038/nature04092, 2005.

Rivier, L., Ciais, P., Hauglustaine, D. A., Bakwin, P., Bousquet, P., Peylin, P., and Klonecki, A.: Evaluation of SF_6, C_2Cl_2, and CO to approximate fossil fuel CO_2 in the Northern Hemisphere using a chemistry transport model, J. Geophys. Res., 111, D16311, doi:10.1029/2005JD006725, 2006.

Schmidt, H., Derognat, C., Vautard, R., and Beekmann, M.: A comparison of simulated and observed ozone mixing ratios for the summer of 1998 in Western Europe, Atmos. Environ., 36, 6277–6297, 2001.

Schulze, E.-D., Luyssaert, S., Ciais, P., Freibauer, A., Janssens, I. A., Soussana, J. F., Smith, P., Grace, J., Levin, I., Thiruchittampalam, B., Heimann, M., Dolman, A. J., Valentini, R., Bousquet, P., Peylin, P., Peters, W., Rödenbeck, C., Etiope, G., Vuichard, N., Wattenbach, M., Nabuurs, G. J., Poussi, Z., Nieschulze, J., Gash, J. H., and the CarboEurope Team: Importance of methane and nitrous oxide for Europe's

terrestrial greenhouse gas balance, Nat. Geosci., 2, 842–850, 2009.

Sillmann, S.: the relation between ozone, NO_x and hydrocarbons in urban and polluted rural environments, Atmos. Environ., 33, 1821–1845, 1999.

Skamarock, W. C., Klemp, J. B., Dudhia, J., Gill, D. O., Barker, D. M., Wang, W., and Powers, J. G.: A description of the Advanced Research WRF Version 2, NCAR Tech Notes-468+STR, 2005.

Stavrakou, T., Müller, J.-F., Boersma, K. F., De Smedt, I., and van der A, R. J.: Assessing the distribution and growth rates of NO_x emission sources by inverting a 10-year record of NO_2 satellite columns, Geophys. Res. Lett., 35, L10801, doi:10.1029/2008GL033521, 2008.

Stavrakou, T., Müller, J.-F., De Smedt, I., Van Roozendael, M., Kanakidou, M., Vrekoussis, M., Wittrock, F., Richter, A., and Burrows, J. P.: The continental source of glyoxal estimated by the synergistic use of spaceborne measurements and inverse modelling, Atmos. Chem. Phys., 9, 8431–8446, doi:10.5194/acp-9-8431-2009, 2009.

Streets, D. G., Bond, T. C., Carmichael, G. R., Fernandes, S. D., Fu, Q., He, D., Klimont, Z., Nelson, S. M., Tsai, N. Y., Wang, M. Q., Woo, J.-H., and Yarber, K. F.: an inventory of gaseous and primary aerosol emissions in Asia in the year 2000, J. Geophys. Res., 108, 8809, doi:10.1029/2002JD003093, 2003.

Suntharalingam, P., Jacob, D. J., Palmer, P. I., Logan, J. A., Yantosca, R. M., Xiao, Y., Evans, M. J., Streets, D. G., Vay, S. L., and Sachse, G. W.: Improved quantification of Chinese carbon fluxes using CO_2/CO correlations in Asian outflow, J. Geophys. Res., 109, D18S18, doi:10.1029/2003JD004362, 2004.

van der Werf, G. R., Randerson, J. T., Giglio, L., Collatz, G. J., Mu, M., Kasibhatla, P. S., Morton, D. C., DeFries, R. S., Jin, Y., and van Leeuwen, T. T.: Global fire emissions and the contribution of deforestation, savanna, forest, agricultural, and peat fires (1997–2009), Atmos. Chem. Phys., 10, 11707–11735, doi:10.5194/acp-10-11707-2010, 2010.

Valin, L. C., Russell, A. R., Hudman, R. C., and Cohen, R. C.: Effects of model resolution on the interpretation of satellite NO_2 observations, Atmos. Chem. Phys., 11, 11647–11655, doi:10.5194/acp-11-11647-2011, 2011.

Wang, R., Tao, S., Ciais, P., Shen, H. Z., Huang, Y., Chen, H., Shen, G. F., Wang, B., Li, W., Zhang, Y. Y., Lu, Y., Zhu, D., Chen, Y. C., Liu, X. P., Wang, W. T., Wang, X. L., Liu, W. X., Li, B. G., and Piao, S. L.: High-resolution mapping of combustion processes and implications for CO_2 emissions, Atmos. Chem. Phys., 13, 5189–5203, doi:10.5194/acp-13-5189-2013, 2013.

Wang, S. W., Streets, D. G., Zhang, Q., He, K. B., Chen, D., Kang, S. C., Lu, Z. F., and Wang, Y. X.: Satellite detection and model verification of NO_x emissions from power plants in Northern China, Environ. Res. Lett., 5, 044007, doi:10.1088/1748-9326/5/4/044007, 2010.

Wang, S. W., Zhang, Q., Streets, D. G., He, K. B., Martin, R. V., Lamsal, L. N., Chen, D., Lei, Y., and Lu, Z.: Growth in NO_x emissions from power plants in China: bottom-up estimates and satellite observations, Atmos. Chem. Phys., 12, 4429–4447, doi:10.5194/acp-12-4429-2012, 2012.

Wang, Y., McElroy, M. B., Martin, R. V., Streets, D. G., Zhang, Q., and Fu, T.-M.: Seasonal variability of NO_x emissions over east China constrained by satellite observations: Implications for combustion and microbial sources, J. Geophys. Res., 112, D06301, doi:10.1029/2006JD007538, 2007.

Wicker, L. J. and Skamarock, W. C.: Time splitting methods for elastic models using forward time schemes, Mon. Weather Rev., 130, 2088–2097, 2002.

Zhang, Q., Streets, D. G., He, K., Wang, Y., Richter, A., Burrows, J. P., Uno, I., Jang, C. J., Chen, D., Yao, Z., and Lei, Y.: NO_x emission trends for China, 1995–2004: the view from the ground and the view from space, J. Geophys. Res., 112, D22306, doi:10.1029/2007JD008684, 2007.

Zhao, B., Wang, P., Ma, J. Z., Zhu, S., Pozzer, A., and Li, W.: A high-resolution emission inventory of primary pollutants for the Huabei region, China, Atmos. Chem. Phys., 12, 481–501, doi:10.5194/acp-12-481-2012, 2012.