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NO_x lifetimes and emissions of hotspots in polluted background estimated by satellite observations

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We present a new method to quantify NO_x emissions and corresponding atmospheric lifetimes from OMI NO_2 observations together with ECMWF wind fields without further model input for sources located in polluted background. NO_2 patterns under calm wind conditions are used as proxy for the spatial patterns of NO_x emissions, and the effective atmospheric NO_x lifetime is determined from the change of spatial patterns measured at larger wind speeds. Emissions are subsequently derived from the NO_2 mass above background integrated around the source of interest.

Lifetimes and emissions are estimated for 17 power plants and 53 cities located in non-mountainous regions across China and the US. The derived lifetimes for non-mountainous sites are 3.8 ± 1.0 h on average with ranges of 1.8 to 7.5 h. The derived NO_x emissions show generally good agreement with bottom-up inventories for power plants and cities. Global inventory significantly underestimated NO_x emissions in Chinese cities, most likely due to uncertainties associated with downscaling approaches.

1 Introduction

Nitrogen oxides (NO_x) are toxic air pollutants and play an important role in tropospheric chemistry as precursors of tropospheric ozone and secondary aerosols (Jacob et al., 1996; Seinfeld and Pandis, 2006). Power plants and cities with large vehicle populations and intense industrial activities are significant anthropogenic emitting sources of NO_x . Accurate knowledge of NO_x emissions on urban scales is thus a critical factor for accurate bottom-up emission inventories, which are important inputs for chemical transport models (CTMs) and for the development of mitigation strategies.

Bottom-up emission inventories depend on information of fuel consumptions and emission factors, which are subject to substantial uncertainties (Butler et al., 2008; Zhao et al., 2011). A significant improvement in accuracy of emission inventories for power plants has been achieved by the installation of continuous emissions monitoring

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systems (CEMS). For example, in the US, under the 1990 Clean Air Act, power plant operators are required to install an automated data acquisition and handling system for measuring and recording pollutant concentrations from plant exhaust stacks and follow the monitoring regulations to ensure that the reported emission data is consistent and of high quality (Kim et al., 2009). For countries where reliable CEMS data is not available (like China), activity rates and emission factors can be adopted at plant-level to improve the accuracy of power plant emissions (e.g. Zhao et al., 2008; Liu et al., 2015). But developing emission inventories for individual cities with high accuracy faces enormous challenges, considering the lack of a complete and reliable database including fuel consumptions and emission factors at city level. Emissions at city level are often forced to downscale regional emissions based on surrogates, like population density or industrial productivity, which however often just roughly reflect the magnitude and spatial distribution of urban emissions. Thus, independent emission estimates would be a desirable complement to validate and improve existing emission inventories.

The NO_2 tropospheric vertical column densities (TVCD, the vertically integrated concentration in the troposphere) retrieved from satellite measurements provide valuable global information on the spatio-temporal patterns of NO_x , including trends (e.g., Richter et al., 2005; Schneider and van der A, 2012; Hilboll et al., 2013), responses of NO_2 level changes to air quality control as well as economical and political factors (e.g., Duncan et al., 2013; Lelieveld et al., 2015), and temporal variations like weekly cycles in NO_2 TVCDs (Beirle et al., 2003; Russell et al., 2010; Valin et al., 2014). In addition, the satellite NO_2 measurements allow quantifying NO_x emissions. In a pioneering study (Leue et al., 2001), the downwind decay of NO_2 TVCDs in continental outflow regions was used to estimate a (constant) NO_x lifetime, which was then applied to project global NO_x emissions from the measured mean NO_2 TVCDs. Later on, CTMs were employed to exploit satellite observations as a constraint towards improving NO_x emission inventories (e.g., Martin et al., 2003; Konovalov et al., 2006; Kim et al., 2009; Lamsal et al., 2011). The derived top-down inventories show pronounced differences relative to bottom-up estimates and their accuracy has been validated by

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the improved performance of model simulations with respect to in-situ measurements (e.g., Martin et al., 2006). However, the top-down inventories are usually determined at regional/global scale related to the spatial resolution of CTMs, while the spatial scales relevant for individual emission hotspots (power plants or cities) are not resolved. In addition, modelled lifetimes have large uncertainties (Lin et al., 2012) due to the highly non-linear small-scale chemistry in urban areas, and are thus probably not appropriate for relating NO_x TVCDs to NO_x emission rates at city level.

With the launch of the Ozone Monitoring Instrument (OMI) (Levelt et al., 2006) with high spatial resolution, individual large sources like Megacities and power plants can be resolved. In a recent study, Beirle et al. (2011) averaged OMI NO_2 measurements separately for different wind directions, thereby constructing clear downwind plumes which allow a simultaneous fit of the effective NO_x lifetimes and emissions, without the need of a chemical model. Similar lifetime/emission estimates have been performed recently by e.g. Valin et al. (2013), Lu et al. (2015) and de Foy et al. (2015). However, so far all studies assume that the source of interest can be considered as a “point source”, which works well for isolated sources like e.g. the city of Riyadh, showing a high contrast against clean background with small and smooth TVCDs. However, for sources located in a heterogeneously polluted background, a modification of these methods is needed in order to account for the effect of interfering sources within small distances.

In this work, we present a new method for the quantification of NO_x lifetimes and emissions for power plants and cities located in polluted background. The mean OMI NO_2 distribution for 2005–2013 is calculated separately for calm conditions as well as for different wind direction sectors according to ECMWF (European Center for Medium-range Weather Forecast) wind fields. The mean lifetime is derived from the change of the observed NO_2 patterns under windy vs. calm conditions. NO_x emissions of power plants and cities over China and the US are subsequently quantified from the integrated TVCDs and the derived lifetimes, and compared to bottom-up emission inventories.

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2 Methodology

2.1 Satellite NO₂ data

We base this study on NO₂ TVCDs from the OMI tropospheric NO₂ (DOMINO) v2.0 product (Boersma et al., 2011), which is provided by the Tropospheric Emissions Monitoring Internet Service (TEMIS, <http://www.temis.nl>). OMI is a UV-VIS nadir-viewing satellite spectrometer (Levelt et al., 2006) on board the Aura satellite (Celarier et al., 2008), launched in 2004. NO₂ columns are derived from radiance measurements, using the Differential Optical Absorption Spectroscopy (DOAS) algorithm (Platt, 1994). OMI provides daily global coverage with a local equator crossing time of approximately 13:45 p.m. It detects radiance spectra by 60 across-track pixels with ground pixel sizes ranging from 13 km × 24 km at nadir to about 13 km × 150 km at the outermost swath angle (57°). The 10 outermost pixels on both sides of the swath are excluded in this study to limit the across-track pixel width < 40 km. From June 2007, OMI has shown severe spurious stripes, known as row anomalies that are likely caused by an obstruction in part of OMI's aperture (<http://www.knmi.nl/omi/research/product/rowanomaly-background.php>). The affected pixels are also excluded from the analysis. Only mostly cloud free observations (effective cloud fraction < 30 %) are considered in this study.

Mean NO₂ TVCDs over the US and China during “ozone season” (May–September) for 2005 to 2013 are calculated separately for calm (wind speed below 2 m s⁻¹) and 8 different wind direction sectors following the approach in Beirle et al. (2011). We focus on the ozone season to include the photochemically relevant months for ozone production (USEPA, 2014) and to exclude the winter data with larger uncertainties due to larger solar zenith angles, variable surface albedo (snow), and longer NO_x lifetime. Wind fields at a lat/long grid of 0.36° width are taken from the ECMWF ERA interim reanalysis (Dee et al., 2011), and the horizontal wind components of the lowermost 500 m are averaged. Individual clear-sky observations of NO₂ TVCDs are assigned to a 2 × finer grid (0.18°, comparable to the extent of OMI ground pixels) according to the

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pixel center coordinates, and associated with the corresponding ECMWF wind fields interpolated in time.

2.2 Fitting procedure

In this section, we present a modified method compared to Beirle et al. (2011) for the determination of lifetimes and emissions for complex source distributions. The basic idea is to use the measured NO₂ spatial pattern under calm wind conditions as proxy for the distribution of NO_x sources, instead of assuming a single point source.

Below, we (a) recap the fitting procedure of Beirle et al. (2011) and demonstrate that this method cannot be applied for multiple sources (Sect. 2.2.1), (b) describe the model function for the modified lifetime fit (Sect. 2.2.2), and (c) eventually explain how emission rates are determined (Sect. 2.2.3).

We select Harbin (45.8° N, 126.7° E), the capital of Heilongjiang province in China, with a population of about 6 million (city) to 10 million (greater area) inhabitants, to demonstrate our approach. Harbin is a typical city located in polluted background, surrounded by three other large NO_x sources (i.e. the cities of Daqing, Jilin and Changchun) within ~ 200 km radius. Figure 1 displays mean NO₂ TVCDs around Harbin for calm conditions (a), southerly wind (b) and their difference (c). The outflow plume of NO₂ from Harbin is not as clear as that from isolated sources (e.g. Riyadh in Beirle et al. 2011), due to the interferences from surrounding sources. But the spatial pattern of their difference (Fig. 1c) still clearly reveals outflow patterns, consistent with ECMWF wind fields.

In order to investigate the downwind plume evolution, 1-dimensional NO₂ “line densities”, i.e. NO₂ per cm, are calculated as function of distance for each wind direction sector separately by integration of the mean NO₂ TVCDs (i.e. NO₂ per cm²) perpendicular to the wind direction, as in Beirle et al. (2011).

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2.2.1 The original fitting procedure

In Beirle et al. (2011), a simple model function $M(x)$ (Eq. 1) was used to fit the observed line densities, which is composed of an exponential function $e(x)$ (Eq. 2) describing the transport pattern and chemical decay, and a Gaussian function $G(x)$ (Eq. 3) accounting for different effects causing spatial smoothing (e.g., the spatial extent of the source, the OM⁵ ground pixel size, or wind fluctuations).

$$M(x) = E \times (e \otimes G)(x) + B \quad (1)$$

$$e(x) = \exp\left(-\frac{x - X}{x_0}\right) \quad \text{for } x \geq X, 0 \quad \text{otherwise} \quad (2)$$

$$G(x) = \frac{1}{\sqrt{2\pi}\sigma} \exp\left(-\frac{x^2}{2\sigma^2}\right) \quad (3)$$

- ¹⁰ E represents total emissions, B represents a constant background; X is the location of the source (relative to the a priori co-ordinates of the site under investigation), x_0 is the e-folding distance downwind; and σ is the SD of $G(x)$. The mean lifetime τ is derived from the e-folding distance x_0 by division by w , the mean projected wind speed.

¹⁵ By this approach, emissions and lifetimes of NO₂ are fitted simultaneously. In Beirle et al. (2011) it is applied for nine isolated hot spots exhibiting high NO₂ TVCDs over a clean background within about 200 km. But it cannot be applied to hot spots surrounded by additional significant sources, like Harbin (Fig. 1), as by definition, the method can only represent a single “point source” convolved with a Gaussian function. For instance, an additional source at 100 km with only 10 % of the emissions of ²⁰ the source under investigation causes a lifetime bias of ~20 %, as the fit tries to “explain” increased downwind values by a longer lifetime (see Fig. S1 and explanations in the Supplement). For an interfering source of the same order as the source of interest, the method fails completely.

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2.2.2 New lifetime fitting procedure

We develop an alternative method accounting for emissions from multiple sources. The basic idea is to use the 1-dimensional NO₂ patterns observed under calm conditions as proxy of emission patterns instead of assuming a single point source. Note that the 1-D pattern of line densities under calm conditions has to be determined along the same (wind) direction, for which the line densities under windy conditions are determined. That means that in total eight 1-D line densities under calm conditions are determined for the eight wind directions. However, only directions with reasonable reliability are considered where mean NO₂ line densities for both calm and windy conditions are well defined (i.e., gaps due to missing data are less than 10 % in the across-wind integration interval i and less than 20 % in the fit interval in wind direction f). With $C(x)$ being the line density under calm wind conditions, we define the new model function $N(x)$ as:

$$N(x) = a \times [e \otimes C](x) + b \quad (4)$$

where $e(x)$ is again a truncated exponential function (Eq. 2 with $X = 0$). The scaling factor a and offset b are included to account for possible systemic differences between windy and calm wind conditions (e.g. cloud conditions, vertical profiles, or lifetimes), which will be discussed in Sect. 3.1 in detail.

We perform a non-linear least-squares fit of $N(x)$ to the observed line densities with a , b , and x_0 as fitting parameters. We set the fit interval in wind direction f to 600 km (300 km in downwind direction, which corresponds to 3 times of the e-folding distance for a lifetime of 5 h and a mean wind speed of 6 m s⁻¹). The across-wind integration interval i is set to be half (300 km). f and i are indicated in Fig. 1a and b. The intervals are larger than those in Beirle et al. (2011), since not only the source under investigation, but also interfering sources have to be appropriately accounted for when comparing line densities of calm and windy conditions. We also perform fits with different intervals (± 100 km, see Table S1) and find only small changes ($\sim 10\%$) for the resulting lifetimes.

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Figure 2a displays the observed line densities for calm (blue) and southerly winds (red) around Harbin, and the fitted model function $N(x)$ (grey). Generally, $N(x)$ describes the observed downwind patterns well: the coefficients of determination (R^2) between observation and fit are 0.96–0.99 for different wind directions, as shown in Fig. 2a–e.

Like in Beirle et al. (2011), the lifetime τ is derived by the ratio of the fitted e-folding distance and the mean wind speed¹: $\tau = x_0/w$. For Harbin, τ is computed to be 3.9 h with a typical 95 % confidence interval (CI) of ± 0.6 h for southerly winds. Averaging the fit results for all wind direction sectors with a good fit performance (i.e. $R > 0.9$, lower bound of $CI > 0$, and CI width < 10 h,) yields $\tau = 3.5$ h with a SD of 0.6 h (Fig. 2), using the fit residues as well as the CI of τ as inverse weights, as in Beirle et al. (2011).

2.2.3 New emission fitting procedure

The modified fitting function $N(x)$ proved to be capable of gaining lifetime information even for complex source distributions. The interferences from multiple neighboring sources, which cannot be represented by a single-source Gaussian distribution, are successfully described by the new model function using $C(x)$ as proxy for the spatial distribution of NO_x sources. However, in contrast to the previous fitting function $M(x)$ in Beirle et al. (2011), $N(x)$ does not contain the magnitude of NO_x emissions directly, but only the emission pattern represented by NO_2 under calm conditions. Thus, total NO_x emissions have to be estimated separately.

According to mass balance, the total mass of NO_x equals the emission rate times lifetime. Emissions can thus be derived in a three-step approach by (a) integrating observed TVCDs originating from the source of interest to calculate the total mass of

¹Note that we subtracted the residual mean wind speed under calm wind conditions from w in order to account for the subtle movement of $C(x)$ compared to the emission pattern; this is, however, a small effect (the relative change between τ determined by wind speeds with and without subtracting calm wind speeds is within $-2 \sim 3\%$).

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NO₂, (b) scaling NO₂ to NO_x, and (c) division by the lifetime τ , which was derived as described in the previous section.

a. Total NO₂ mass

In order to quantify the total NO₂ mass of the target source, the observed TVCDs have to be integrated around the source, in which (1) interferences with neighboring sources have to be avoided and (2) a polluted background has to be appropriately accounted for. Thus, we base the estimation of the total NO₂ mass on the mean TVCDs under calm conditions, to minimize interferences by advection. Again, we calculate line densities by integrating the NO₂ TVCDs in “across-wind” direction², but for a smaller interval v representing the spatial extent of megacities or urban centers, but exclude neighboring sources. Here we define $v = 40$ km. We then perform a non-linear least-squares fit of a modified Gaussian function $g(x)$ to these line densities under calm wind condition, as illustrated in Fig. 3. The line densities integrated perpendicular to the different wind direction sectors are used to constrain the fitted A in $g(x)$:

$$g_i(x) = A \times \frac{1}{\sqrt{2\pi}\sigma_i} \exp\left(-\frac{x^2}{2\sigma_i^2}\right) + \varepsilon_i + \beta_i x \quad (5)$$

i represents the wind direction sector, i.e., Southeast–Northwest, South–North, Southwest–Northeast and East–West. σ_i is the SD of the Gaussian $g_i(x)$, and ε_i and β_i represent an offset and a possible linear gradient in the background field respectively. The NO₂ amount A (in molecules) around the source on top of the (wind sector dependent) background is determined by fitting the functions $g_i(x)$ simultaneously for all available wind directions.

²Though focussing on calm conditions, we calculate the projections for different wind direction sectors analogue to the lifetime fit procedure.

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The fit interval h has to be chosen to be larger than v in order to allow for a meaningful fit of $g(x)$. We set h to 200 km for cities (see Fig. S2) and 100 km for power plants respectively. The fit interval thus potentially includes interfering sources. However, these interferences are in first order accounted for by the linear variation of the background fitted in function $g_i(x)$. Note that the fit $g(x)$ is less sensitive to interfering sources compared to the original fit of $M(x)$ in Beirle et al. (2011), as lifetime is not involved here.

The small interval v (40 km) excludes neighboring sources, but does not capture the full plume in across wind direction due to dilution. This effect is corrected for by scaling A afterwards by a factor $f(\sigma_i)$ based on the fitted plume width σ_i :

$$f(\sigma_i) = \frac{\int_{-20\text{ km}}^{20\text{ km}} \frac{1}{\sqrt{2\pi}\sigma_i} \exp\left(-\frac{x^2}{2\sigma_i^2}\right) / \int_{-\infty}^{\infty} \frac{1}{\sqrt{2\pi}\sigma_i} \exp\left(-\frac{x^2}{2\sigma_i^2}\right)}{ } \quad (6)$$

Note that we consider a larger interval (60 km for v and 300 km for h) for Pearl River Delta, which is a megalopolis covering nine prefectures over an area of about 56 000 km².

The resulting emissions are rather insensitive with respect to modified settings for v and h (see Supplement, Sect. 3). Again, fit results with poor performance ($R < 0.9$, lower bound of CI < 0, CI width > $0.8 \times A$) are discarded.

b. Scaling NO₂ to NO_x

According to the typical [NO]/[NO₂] ratio of 0.32 under urban conditions at noon (Seinfeld and Pandis, 2006), the total NO₂ mass is scaled by a factor of 1.32 in order to derive total NO_x mass following Beirle et al. (2011).

c. Emission rates (NO_x amount per time unit) are derived by division of the total NO_x mass by the lifetime derived for the respective location as described in Sect. 2.2.2.



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For Harbin, the total mass (in terms of NO₂) is computed to be 33.2×10^{28} molec with a CI of 2.4×10^{28} molec. The total NO_x emissions derived for Harbin are 58.1 mols⁻¹.

2.3 Uncertainties

We define total uncertainties of the fitted lifetimes and emissions based on (a) the fit confidence intervals (CIs) and (b) the dependencies on the a priori settings as investigated in sensitivity studies, analogue to the procedure described in Beirle et al. (2011).

The CIs resulting from the least-squares fits of Eqs. (4) and (5) directly reflect the uncertainties of the derived lifetimes and emissions. In addition, the SDs of the fitted lifetimes for different wind direction sectors provide information on the consistency of the method. Beyond this, the results are also affected by the a priori choices of wind fields, integration intervals etc. In particular, the calculation of line densities (a), wind fields (b), potential dependence of lifetimes on wind conditions (c) and fit errors (d) contribute to the uncertainties for both τ and emissions, and uncertainties in the total NO₂ mass fit (e), tropospheric NO₂ TVCDs and the NO₂/NO_x ratio (f) affect the derived emissions. We define total uncertainties as the root of the quadratic sum of the above mentioned contributions, which are assumed to be independent. We investigate the different sources of uncertainties contributing to the overall uncertainties of the derived lifetimes and emissions. Detailed sensitivity studies on the dependency of the fit results on the a priori settings are provided in Sect. 2 of the Supplement.

2.4 Bottom-up emission inventories

We use bottom-up emission inventories to pre-select promising sites and for a comparison to the derived top-down estimates. We select inventories that provide up-to date, multi-year NO_x emissions at high spatial resolution and are widely used in the community. The following inventories are considered:

For power plants, we use the China coal-fired Power plant Emissions Database (CPED) developed by Liu et al. (2015) based on unit-level fuel consumptions and

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emission factors derived from various sources, and the US Emissions and Generation Resource Integrated Database (eGRID) using emissions derived from continuous emissions monitoring systems (available at <http://www.epa.gov/cleanenergy/energy-resources/egrid/>) (USEPA, 2014). For cities, we use the Multi-resolution Emission Inventory for China (MEIC: <http://www.meicmodel.org>) compiled by Tsinghua University and the global inventory of the Emissions Database for Global Atmospheric Research (EDGAR) v4.2 (EC-JRC/PBL, 2011) for the US.

For the comparison to the derived top-down estimates, a 8 year (2005–2012) average from CPED and a 4 year (2005, 2007, 2009, and 2010) average from eGRID for the ozone season are used for power plants, of which the uncertainties are about 30 % (Liu et al., 2015) for CPED and 10 % for eGRID (5 % arise from continuous emissions monitoring systems (Gluck et al., 2003) and another 5 % arise from yearly variations in emissions after 2010), respectively. In addition, the mean emissions for the ozone season of the years 2005–2012 in MEIC and the mean annual emissions for the years 2005–2008 in EDGAR are used for cities, of which the uncertainty is estimated to be within a factor of 1/2 and 2 according to the MEIC and EDGAR expert judgment of “medium magnitude of uncertainty” (Olivier et al., 2002). The bottom-up urban emissions derived from regional/global inventories have larger uncertainties compared to power plant emissions, primarily arising from the low-resolution activity rates/emission factors at regional level, and the spatial allocation technique using surrogates to break regional-based emission data down to cities. Furthermore, temporal coverage of bottom-up emissions is limited, inducing additional uncertainties. For instance, a decline in NO₂ TVCDs from the years 2005–2008 to 2009–2013 with an average total reduction of $14 \pm 9\%$ (mean \pm standard variation) is detected for investigated US cities (Fig. S3). However, the most recent year available in EDGAR v4.2 is 2008, which cannot reflect the recent decline in NO_x emissions, thus overestimate the average emissions.

For the comparison of bottom-up and top-down emissions for individual sites, the power plant inventories directly represent the stack emissions of individual facilities. For total city emissions, the gridded emission inventories have to be integrated over the

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metropolitan area for which the proposed top-down method is sensitive. Here, we define this area as $40\text{ km} \times 40\text{ km}$, consistent with the considered interval v in Sect. 2.2.3. For PRD, we consider a larger interval of $120\text{ km} \times 120\text{ km}$.

2.5 Selection of investigated sources

- 5 For this study, we choose large power plants and cities across China and the US as the pre-selected candidates, of which bottom-up emission information is available from inventories described above. Power plants with NO_x emission rates greater than 10 Ggyr^{-1} (CPED/eGRID) are investigated. Power plants located in urban areas (100 km around city centers) are excluded by inspecting satellite imagery from Google
- 10 Earth. The top 150 largest cities (rank in GDP/GDP per capita in 2013) in China and the 47 large US cities selected for analyses in Russell et al. (2012) were also examined. To assure a good fit performance, the following criteria have been defined: (1) the signal of the source is strong, i.e., the mean NO_2 TVCD in a circle of 100 km around the location center is larger than $1 \times 10^{15}\text{ molec cm}^{-2}$; and (2) fit results with poor performance
- 15 are discarded (see Sects. 2.2.2 and 2.2.3 for details). Table S2 of the Supplement provides a list of all sources under investigation which passed the criteria, including 24 power plants and 69 cities across China and the US.

2.6 Impact of topography

- 20 The accuracy of fitted lifetimes is highly dependent on the accuracy of the a priori wind directions (used for “sorting” the satellite NO_2 observations) and velocities (used for converting x_0 into τ). However, accurate modelling of wind fields on small scales is challenging for large-scale models like ECMWF, which do not resolve urban scales. Consequently, wind fields might be biased in particular over complex mountainous terrain, related to the difficulties in resolving the characterization of small-scale orography
- 25 in models (Beljaars et al., 2004).

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We investigate the impact of topography by comparing ECMWF wind fields to 2005–2013 sounding measurements assembled by University of Wyoming (<http://weather.uwyo.edu/upperair/sounding.html>), and illustrate it for the cities of Harbin (plain terrain) and Taiyuan (mountainous city in Shanxi, China) in Fig. 4. In the top panels, topography used by ECMWF is compared to the topographic data from the 30 arcsec global land topography “GTOPO30” archived by the US Geological Survey (available at <https://lta.cr.usgs.gov/GTOPO30>, rescaled to 0.05°). Topographic variations are smeared out significantly by the topographic model used in ECMWF, due to its coarser spatial resolution of 0.36°. The bottom panels show statistics for wind vectors below 500 m during daytime (12:00) and nighttime (00:00) from both ECMWF and the sounding measurements. The frequency distribution of wind directions (in 45° bins) shows a very good agreement in Harbin, but not in Taiyuan: here southerly flows dominate according to sounding measurements, while easterly winds dominate in ECMWF.

We compared wind fields for cities where the fits work properly (Table S2) and the sounding measurements are available simultaneously, as presented in Table S3. For a mountainous city where the elevation in ECMWF contrasted sharply with that in GTOPO, Denver for instance, the correlation in wind speeds between ECMWF and sounding measurements is found to be much lower than for a non-mountainous city like Harbin.

Note that an error in a priori wind direction generally leads to a misclassification during the sorting of the satellite data. In such a case, the assumed wind component in direction of the sector is higher than the actual projection; if, for instance, the true wind would be 5 m s⁻¹ from north, but the model wind is 5 m s⁻¹ from east, the case is classified as easterly, while the actual easterly wind is 0. This leads to a systematic high biased projected wind speed in Eq. (4), and thus a low biased lifetime. Thus, mountainous sites often yield very low lifetimes (Table S2).

As the fitted lifetimes, and thus also emissions, rely on appropriate wind fields, we exclude mountainous sites from the following analysis. We simply define a site as mountainous where the absolute difference in elevation between ECMWF and GTOPO is

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larger than 250 m. A total of seven power plants and 16 cities are rejected based on the criteria, as listed in Table S4. Seven sites in Table S3 fulfill this criteria and 6 of them present low correlation ($r^2 < 0.5$) in wind speeds between ECMWF and sounding measurements.

5 3 Results and discussions

We applied our modified method for determining NO_x lifetimes and emissions to 17 power plants and 53 cities across China and the US (see Fig. 5), which passed the criteria defined in Sects. 2.5 and 2.6. Some strong hotspots are not included as they are mountainous, e.g. Denver or Salt Lake City.

10 3.1 Lifetimes

Figure 6 illustrates the fitted NO_x lifetimes for power plants and cities across China and the US, which demonstrates the wide applicability of the modified method developed in this study. The derived lifetimes in “ozone season” (May–September) are 3.8 ± 1.0 h on average with ranges of 1.8 to 7.5 h. These values are in agreement to previously reported NO_x lifetimes (e.g., Beirle et al., 2004, 2011; Schaub et al., 2007; Valin et al., 15 2013) and correspond to a mean OH concentration of the order of $10^7 \text{ molec cm}^{-3}$ (Valin et al., 2013), which is a realistic number for a polluted urban plume around noon (e.g., Kramp and Volz-Thomas, 1997; Dillon et al., 2002; Hofzumahaus et al., 2009). For the investigated sites, average lifetime for Power Plants (3.5 h) was found to be 20 slightly shorter than for cities (3.9 h). Individual lifetimes have uncertainties of about 60 %. But, still, Fig. 6 indicates that lifetimes are not completely random, but show systematic spatial patterns. We could not unambiguously relate the variability of NO_x lifetime to a driving parameter, like surface elevation, mean wind characteristics, or latitude. But there is a tendency that NO_x lifetime is longer in heavily polluted regions with higher NO_2 TVCDs, e.g., eastern China and eastern US: the mean NO_2 TVCD for the 25

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ozone season in a circle with a radius of 100 km around sources with lifetimes over 5 h is 6.3×10^{15} molec cm $^{-2}$, while it is only 1.3×10^{15} molec cm $^{-2}$ for sources with lifetime less than 2 h. This finding might be related to nonlinear NO $_x$ chemistry, resulting in a positive correlation between NO $_x$ lifetimes and NO $_2$ TVCDs when the concentration of NO $_x$ is high (Valin et al., 2013). However, we also find that a high NO $_x$ concentration does not necessarily correspond to a long lifetime, and the correlation between NO $_2$ TVCDs and NO $_x$ lifetime is rather low ($r^2 = 0.22$), probably due to the complex NO $_x$ chemistry, which is as well affected by meteorological and chemical variability, like variations in UV flux, water vapor and VOC levels.

The proposed method estimates the mean lifetime basically from the change of NO $_2$ patterns for windy vs. calm conditions. Valin et al. (2013) report on a dependency of the NO $_x$ lifetime on wind speed, with generally shorter lifetimes for higher wind speed. In addition, other factors, like the satellite's sensitivity (affected by e.g. cloud properties or the vertical NO $_x$ profile) and the NO $_2$ background might change systematically between calm and windy conditions. In the fitted model function $N(x)$, a scaling factor a and an offset b are required in order to achieve a good fit performance for the individual fits, which probably compensate for these effects. But on average, the derived values for a and b are close to 1 and 0, respectively: a is 0.9 ± 0.1 (mean \pm SD) and b is $0.0 \pm 0.1 \times 10^{23}$ molec cm $^{-1}$ (mean \pm SD).

Thus, possible systematic effects due to all kind of changes between calm and windy conditions are small, and they are considered with a 10 % of contribution in the total uncertainty for NO $_x$ lifetimes (see Supplement).

We also performed an additional analysis of seasonal mean lifetimes (see Supplement, Fig. S4). Wintertime is excluded in the seasonal analysis, because in winter satellite data exhibits larger uncertainties and line densities under calm wind condition are often unrepresentative of the emission pattern due to longer NO $_x$ lifetimes. The seasonal lifetimes reveal higher uncertainties due to a smaller number of available satellite observations compared to the ozone season, but a systematic seasonal vari-

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ability is still observed for most non-mountainous cases: mean lifetimes are found to be shorter in summer (3.2 h) compared to spring (4.2 h) and autumn (4.5 h), as expected.

3.2 Emissions

Figure 7 compares the derived NO_x emissions to bottom-up emission inventories (Sect. 2.4) for all 17 power plants and 53 cities. For power plants, the comparison (Fig. 7a) shows excellent agreement with a high correlation coefficient ($r^2 = 0.93$). Average emissions are 29 mol s^{-1} in bottom-up inventories and 31 mol s^{-1} in top-down estimates. The relative difference (defined as $(E_{\text{top down}} - E_{\text{bottom-up}})/E_{\text{bottom-up}}$) is within 30 % for most sites, and $5\% \pm 27\%$ (mean \pm SD) on average. For China and the US, the relative differences are $4\% \pm 18\%$ and $5\% \pm 31\%$ respectively, confirming the accuracy of CPED and eGRID bottom-up emission inventories.

For the investigated cities, good agreement (Fig. 7b) between the derived emissions and the bottom-up emissions is reassuring and the r^2 reaches 0.84 (0.87 and 0.74 for China and the US respectively). The relative difference between derived NO_x emissions and bottom-up emissions for cities is larger than that for power plants, reaching $9\% \pm 49\%$ ($1\% \pm 46\%$ and $20\% \pm 51\%$ for China and the US respectively) on average. This is probably related to the higher uncertainties of the bottom-up inventories for cities compared to those for power plants. We further compared the representations of China's urban emissions between MEIC and EDGAR, as shown in Fig. 8. Huge discrepancies are found between EDGAR and top-down estimates (relative difference: $311\% \pm 412\%$) with large negative bias in the bottom-up. Considering the deviation in national total NO_x emissions is far less (20.7 and 24.9 Tg- NO_2 for year 2008 in EDGAR and MEIC respectively), the large bias could be primarily explained by the spatial distributions in the two inventories.

Both MEIC and EDGAR calculate emissions as province/country totals and distribute them to grids using spatial proxies. By comparing spatial proxies used in the two inventories, we identified the major differences in spatial allocation methods between them: (1) MEIC used an in-house high-resolution database (CPED) to represent power plant

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emissions in China while EDGAR used CARMA (Wheeler and Ummel, 2008). The coordinates of power plants in CARMA are highly uncertain for China (Liu et al., 2015); (2) for industrial emissions, MEIC first downscaled provincial totals to counties using industrial GDP, and then allocate county emissions to grids with population density.
5 EDGAR directly distributed provincial emissions by population density (EC-JRC/PBL, 2012); and (3) MEIC allocated on-road emissions by vehicle and road type using the China Digital Road-network Map (Zheng et al., 2014), while EDGAR used the product of population density (Gridded Population of the World (GPW) version 3, CIESIN et al., 2005) and road network (the Global Roads Inventory Project (GRIP), PBL, 2008). All
10 above factors may contribute to the better representations of urban emissions in MEIC than in EDGAR over China.

It is interesting that EDGAR represents urban emissions much better in the US than in China, even though EDGAR shared the same spatial allocation approach across different countries. One plausible explanation is that spatial proxies work better in the
15 US, implying the linear relationships between emissions and proxies, e.g., vehicle emissions and road densities, industrial/residential emissions and population densities. Different accuracy of spatial proxies among regions may also contribute to the discrepancy of performance in the two inventories. For instance, the GRIP database (<http://geoservice.pbl.nl/website/GRIP/>) missed too many roads for China (Fig. S6).
20 By comparing with a high-resolution emission inventory, the Database of Road Transportation Emissions (DARTE), Gately et al. (2015) argued that EDGAR overestimated on-road emissions in city centers while underestimate at the suburban and exurban fringes, resulting from mismatches between road density and the actual spatial patterns of vehicle activity at urban scales. To better understand the uncertainties associated with the performance of spatial proxies, further source-by-source comparison
25 is required between downscaled regional inventories and high-resolution inventories independent to spatial proxies (e.g., DARTE).

The emissions are derived based on the individual fitted lifetimes for each site. If, instead, the mean lifetime of all sites (3.7 h) would be considered for the calculation of

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emissions, the correlations to bottom-up emissions are worse compared to the individual fitted NO_x lifetime (Fig. 9). This holds for both, power plants and cities. We conclude that variation of the fitted lifetime is not just the result of statistical noise, but actually carries information on local variability of the oxidizing capacity of urban plumes. The individual lifetimes are thus well suited for the determination of emissions by a mass balance approach.

3.3 Uncertainties

Based on the approaches presented in Sect. 3 of the Supplement, we estimated that total uncertainties of NO_x lifetime and emissions are within 47–88 and 61–97 % respectively for all the investigated sites (see Sect. 2.5). For Harbin, relative uncertainties for mean lifetime and emissions are 52 and 64 %, respectively. However, it is worth noting that our uncertainty estimate is rather conservative. For power plants, relative differences between bottom-up and top-down estimates are all within 50 % (Fig. 7a). As bottom-up emission inventories for power plants are well developed with low uncertainties, the good consistency increases our confidence that the fitted emissions well represent the real-word emission characteristic. Thus, bottom-up inventories may have large biases for cities where emission estimates differ significantly from top-down constraints (i.e., the relative difference far exceeds 50 %).

From the quantitative analysis approach described in Sect. 2.3, we identify the uncertainties induced by individual factors. Detailed discussions are presented in the Supplement. In summary, we conclude that

- the uncertainty due to wind data is ~20 % (affecting both τ and emissions),
- effects of a possible systematic change of NO_2 TVCDs from calm (used for fit of E) to windy (used for fit of τ) conditions are small (< 10 %),
- the derived emissions (but not the lifetimes) are affected by the uncertainty of the NO_2 TVCDs (~30 %) and the NO_x/NO_2 scaling factor (~10 %),

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- the dependency on the definition of integration and fit intervals is about 20 %.

All involved uncertainties contain both statistical fluctuations as well as systematic effects. By ongoing satellite measurements (e.g. TROPOMI), i.e. longer available time periods, and the much better temporal sampling of upcoming geostationary satellite missions such as GEMS (Kim et al., 2012), TEMPO (Chance et al., 2012), or Sentinel-4 (Ingmann et al., 2012), statistical uncertainties will decrease. In addition, we expect further improvement of the presented lifetime fit method by using regional meteorological models that are more capable of representing wind fields in the planetary boundary layer especially for mountainous region. Also the uncertainties of TVCDs from satellite retrievals, which is still the largest single component of total uncertainty in top-down emission estimates, is expected to decrease in the coming years: input data such as surface albedo or a priori profiles will improve, and the current intensive validation efforts (e.g., DISCOVER-AQ (<http://discover-aq.larc.nasa.gov/>) and AROMAT (<http://uv-vis.aeronomie.be/aromat/>) will help to identify and remove systematic errors. It can thus be expected that total uncertainties of the proposed method will decrease significantly within the next decade.

4 Conclusion

We developed a new method to estimate NO_x lifetimes and emissions of power plants and cities in polluted background from satellite NO_2 observations. The method improves upon that of Beirle et al. (2011) by explicitly accounting for interferences with neighboring strong NO_x sources by using NO_2 spatial patterns under calm wind conditions as proxy of the patterns of emission sources. Lifetimes are derived from the change of NO_2 distributions under windy compared to calm conditions. NO_x emissions are derived by mass balance: the total mass of NO_2 originating from the source of interest is divided by the lifetime derived for the corresponding source.

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The new method for determining NO_x lifetimes and emissions was applicable for 24 power plants and 69 cities over China and the US, including 23 mountainous sites. We exclude the derived results for 23 mountainous sites from the analysis, which are expected to have larger uncertainties owing to the inaccurate wind data. The derived 5 lifetimes for 70 non-mountainous sites are 3.8 ± 1.0 h on average with ranges of 1.8 to 7.5 h. We observed systematic spatial patterns for the derived lifetimes, which however could not be simply explained by a specific driving parameter. Generally, higher lifetimes were found in heavily polluted regions, but the overall correlation between NO₂ TVCDs and NO_x lifetime is quite low ($r^2 = 0.22$).

10 The derived top-down NO_x emissions are generally in very good agreement with bottom-up emission inventories, in particular for power plants, while correlations for cities were lower, probably due to the higher uncertainty of the bottom-up inventories for cities. Compared to MEIC, the EDGAR global inventory significantly underestimated 15 NO_x emissions for Chinese cities, because spatial proxies used in EDGAR may misrepresent emission spatial patterns for China.

Owing to the global continuous monitoring of satellite measurements, this method can be applied to quantify the emissions from various hotspots even in polluted background around the world. This capability will further be enhanced with future satellite instrument like TROPOMI (Veefkind et al., 2012) featuring higher spatial resolution. In 20 addition, upcoming geostationary satellite instruments will enable studies on the diurnal cycle of the NO_x lifetime. More accurate estimates for emission rates, trends and seasonality can be expected, which will serve as an independent data source to validate bottom-up emission estimates in the future.

25 **The Supplement related to this article is available online at
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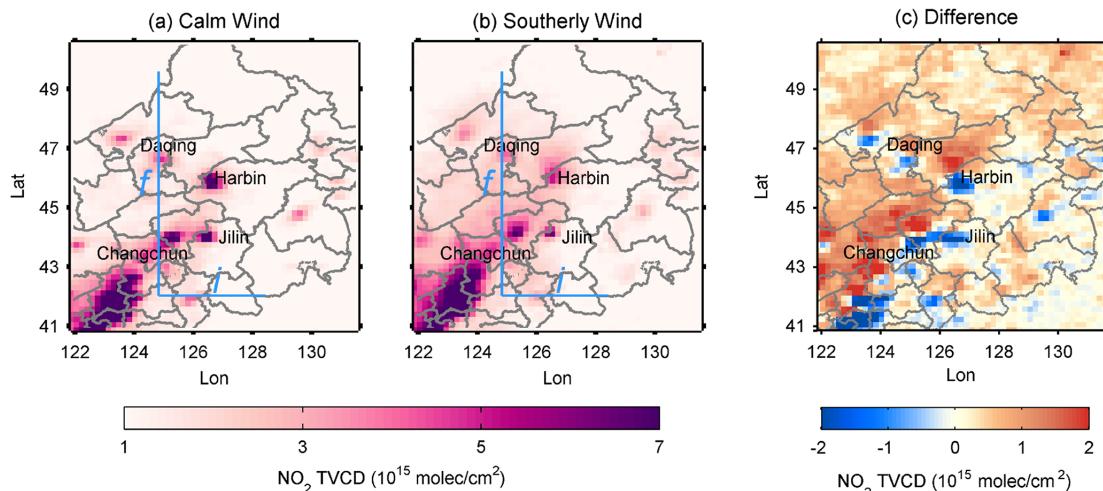


Figure 1. Mean NO_2 TVCDs around Harbin for **(a)** calm, **(b)** southerly wind conditions and **(c)** their difference (southerly – calm). For the fit of lifetimes, the mean NO_2 TVCDs are integrated over interval i in across-wind direction to calculate line densities and the fit is performed over the fit interval f (blue lines in **(a)** and **(b)**; see Sect. 2.2.2 for details).

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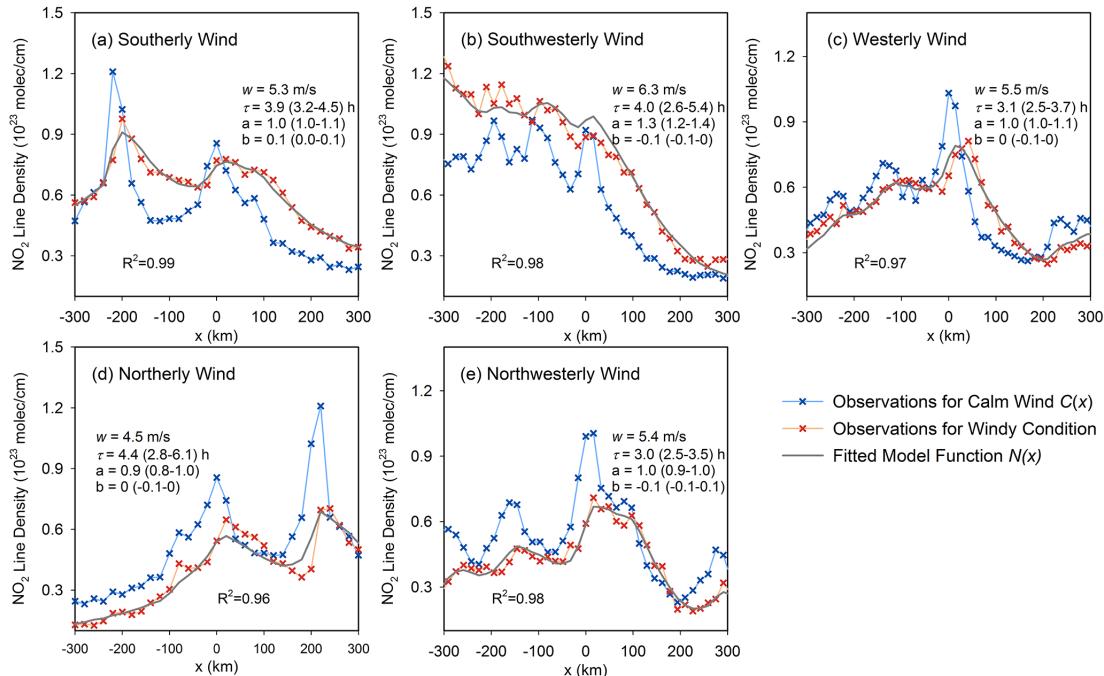


Figure 2. NO_2 line densities around Harbin for different wind direction sectors. Crosses: NO_2 line densities for calm (blue) and (a) southerly, (b) southwesterly, (c) westerly, (d) northerly and (e) northwesterly (red) winds as function of the distance x to Harbin center. Grey line: the fit result $N(x)$. The numbers indicate the net mean wind velocities (windy – calm) from ECMWF (w), the lifetime τ , the factor a and offset b resulting from the least-squares fit with 95 % confidence interval. NO_2 line densities for the remaining wind direction sectors are dismissed due to missing data (see the criteria of “reliability” defined in Sect. 2.2.2).

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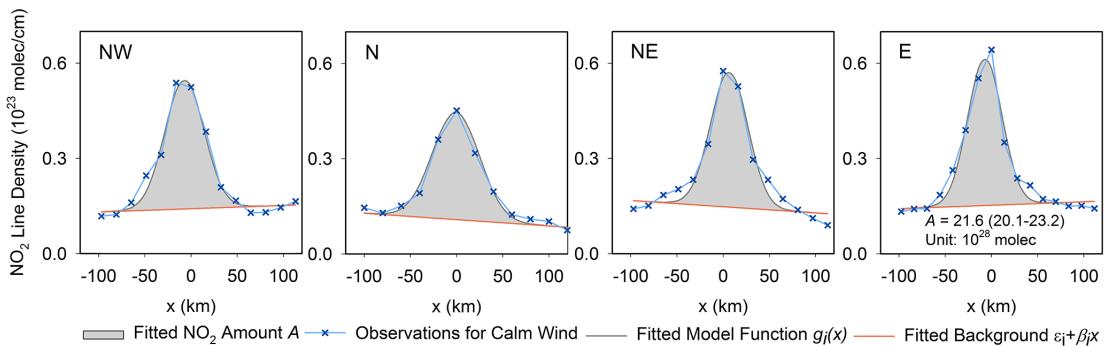


Figure 3. NO₂ line densities in Harbin for northwest, north, northeast and east directions (from left to right). Crosses: NO₂ line densities for calm winds as function of the distance to Harbin center x . Grey line: the fit result $g_i(x)$. Pink line: the fitted background $\varepsilon_i + \beta_j x$. Grey shade: the magnitude of the fitted NO₂ amount A . The number indicates A resulting from the least-squares fit with 95 % CI.



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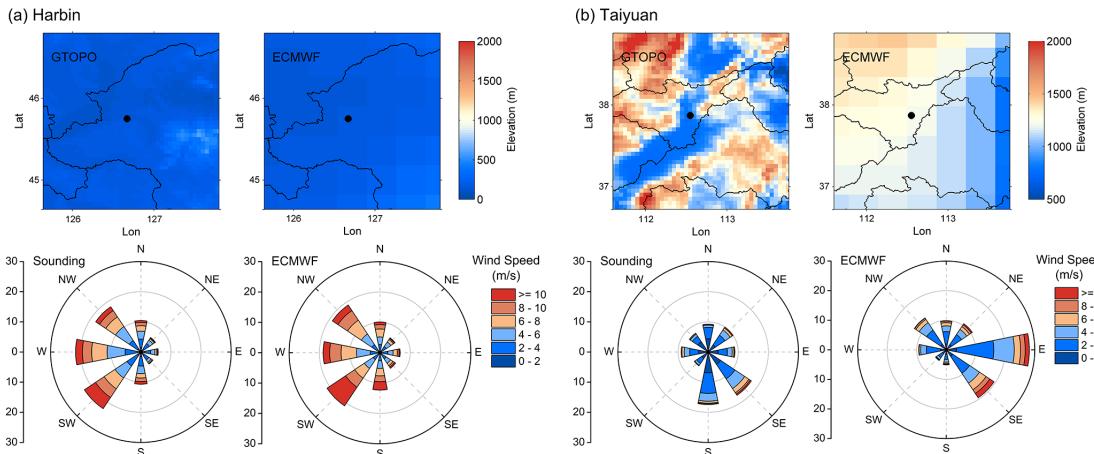


Figure 4. Comparison of the topography (top panel) and wind roses (bottom panel) from ECMWF (right panel) and higher resolution data sets (left panel) around **(a)** Harbin and **(b)** Taiyuan. The land surface elevation on the left panel is derived from GTOPO30. The wind roses on the left panel are generated from sounding measurements assembled by University of Wyoming. Radial units are percent per 45° direction band.

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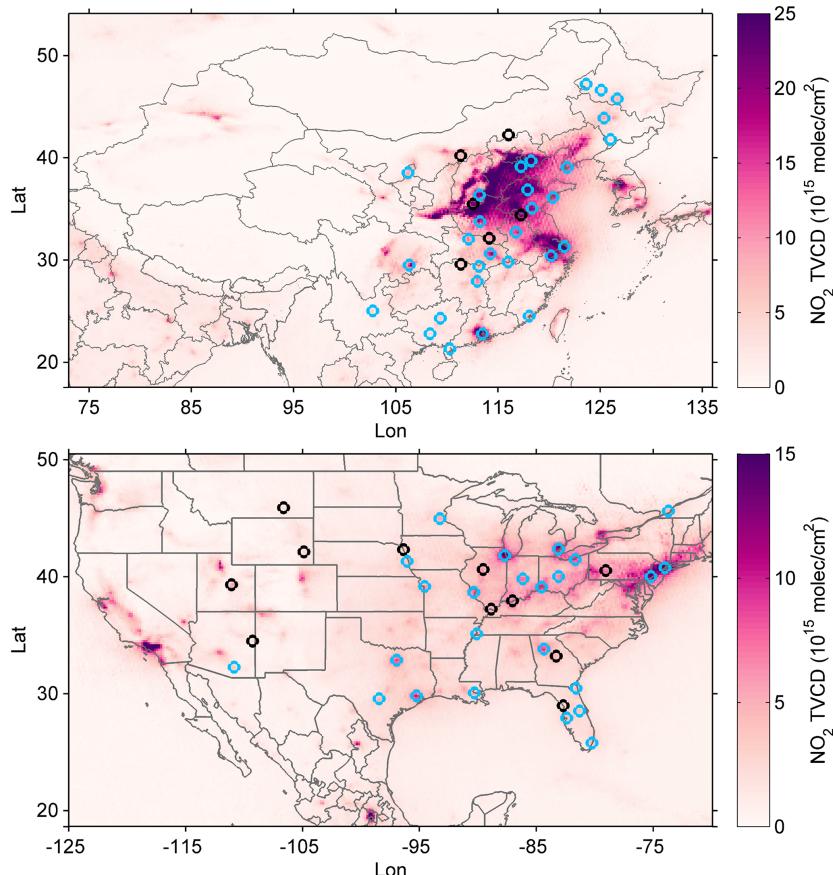


Figure 5. Average OMI NO₂ TVCDs during ozone season (i.e., May to September) over China and the US for the period 2005–2013. Black and blue symbols indicate the 17 power plants and 53 cities investigated in this work, respectively.

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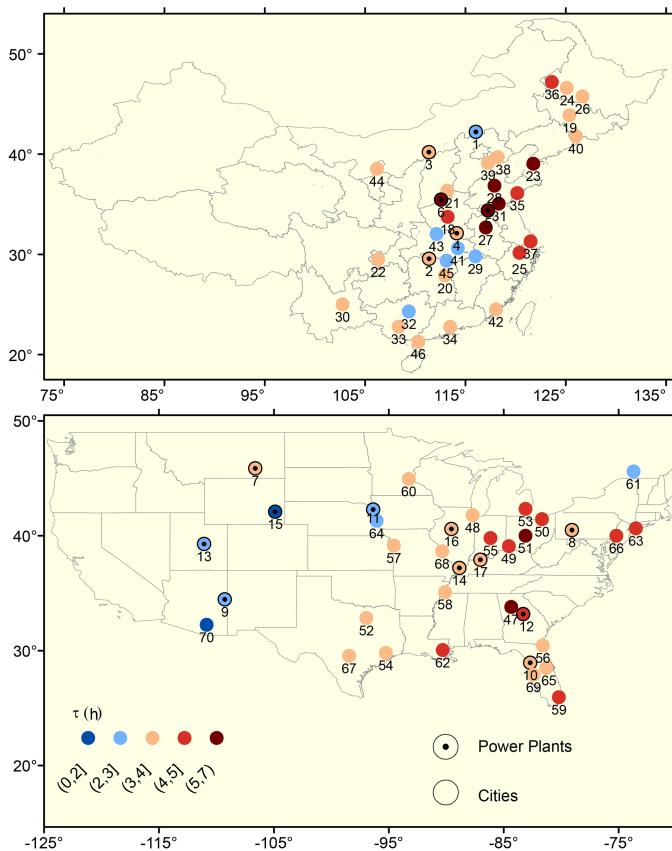


Figure 6. Fitted NO_x lifetimes (color coded) for investigated emission sources over China and the US. Locations of power plants are indicated by dots. Power plants and cities are labelled by their IDs (see Table S2).

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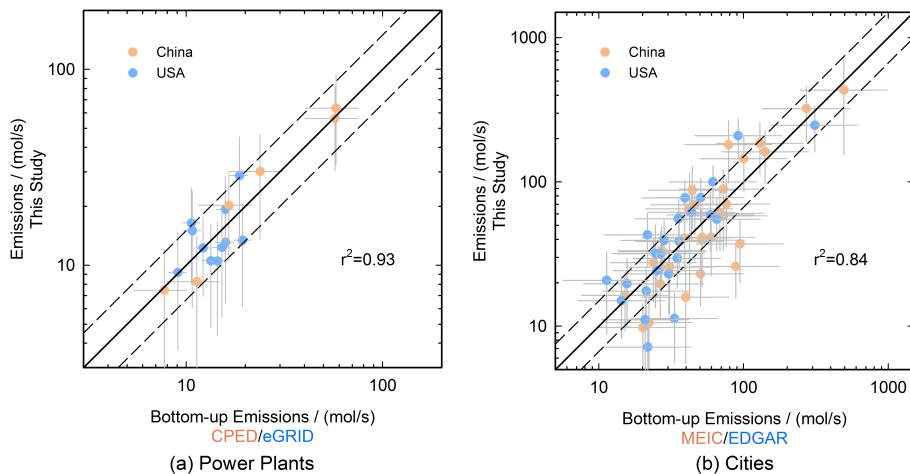


Figure 7. Scatterplots of the derived NO_x emissions for investigated **(a)** power plants and **(b)** cities vs. bottom-up emission inventories. Emissions are given in mol s^{-1} calculated assuming a constant emission rate. Urban emissions from bottom-up inventories are integrated over $40\text{ km} \times 40\text{ km}$ (see text). Error bars show the uncertainties for emissions by this method (see Sect. 2.3) and bottom-up inventories (see Sect. 2.4). The straight and dashed lines represent the ratio of 1 : 1 and 1.5 : 1 / 1 : 1.5, respectively.

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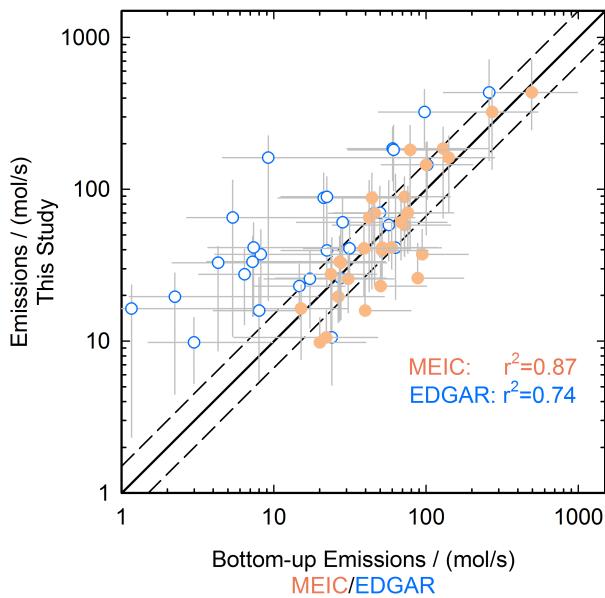


Figure 8. Same as Fig. 7 but scatterplots of the derived NO_x emissions for investigated cities vs. MEIC and EDGAR estimates over China.

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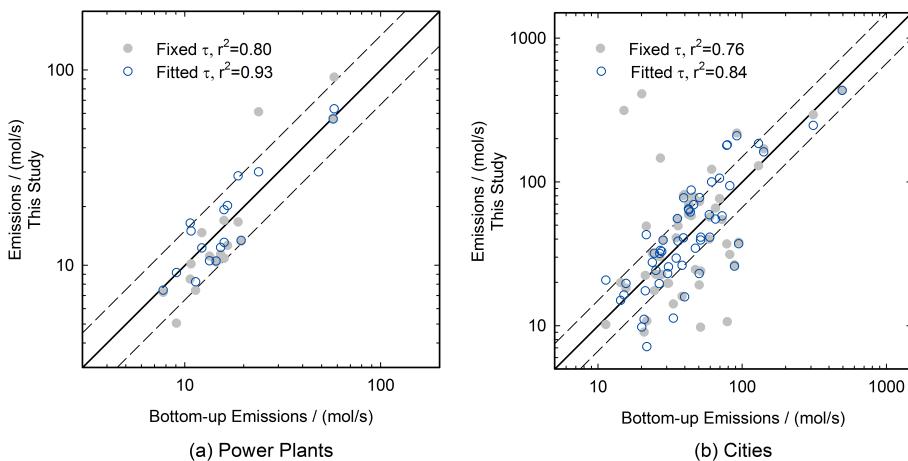


Figure 9. Scatterplots of the resulting NO_x emissions for the investigated power plants and cities using fitted lifetimes (open circles) and fixed lifetimes (3.7 h) (filled circles) vs. the respective estimates from bottom-up emission inventories. Emissions are given in mol s^{-1} calculated assuming a constant emission rate. The straight and dashed lines represent the ratio of 1 : 1 and 1.5 : 1/1 : 1.5 respectively.

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