

1 **NO_x lifetimes and emissions of cities and power plants**
2 **in polluted background estimated by satellite**
3 **observations**

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1 **Abstract**

2 We present a new method to quantify NO_x emissions and corresponding atmospheric
3 lifetimes from OMI NO₂ observations together with ECMWF wind fields without
4 further model input for sources located in polluted background. NO₂ patterns under
5 calm wind conditions are used as proxy for the spatial patterns of NO_x emissions, and
6 the effective atmospheric NO_x lifetime is determined from the change of spatial
7 patterns measured at larger wind speeds. Emissions are subsequently derived from the
8 NO₂ mass above background integrated around the source of interest.

9 Lifetimes and emissions are estimated for 17 power plants and 53 cities located in
10 non-mountainous regions across China and the US. The derived lifetimes are 3.8 ± 1.0
11 hours (mean \pm standard deviation) on average with ranges of 1.8 to 7.5 hours. The
12 derived NO_x emissions show generally good agreement with bottom-up inventories
13 for power plants and cities. Regional inventory shows better agreement with top-down
14 estimates for Chinese cities compared to global inventory, most likely due to different
15 downscaling approaches adopted in the two inventories.

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1 **1 Introduction**

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

10 Bottom-up emission inventories depend on information of fuel consumptions and
11 emission factors, which are subject to substantial uncertainties (Butler et al., 2008;
12 Zhao et al., 2011). A significant improvement in accuracy of emission inventories for
13 power plants has been achieved by the installation of continuous emissions
14 monitoring systems (CEMS). For example, in the US, under the 1990 Clean Air Act,
15 power plant operators are required to install an automated data acquisition and
16 handling system for measuring and recording pollutant concentrations from plant
17 exhaust stacks and follow the monitoring regulations to ensure that the reported
18 emission data is consistent and of high quality (Kim et al., 2009). For countries where
19 reliable CEMS data is not available (like China), activity rates and emission factors
20 can be adopted at plant-level to improve the accuracy of power plant emissions (e.g.
21 Zhao et al., 2008; Liu et al., 2015). But developing emission inventories for individual
22 cities with high accuracy faces enormous challenges, considering the lack of a
23 complete and reliable database including fuel consumptions and emission factors at
24 city level. Emissions at city level are often downscaled from regional emission
25 estimates, based on surrogates (e.g. population density, industrial productivity, and
26 etc.), which however often just roughly reflect the magnitude and spatial distribution
27 of urban emissions. Thus, independent emission estimates would be a desirable
28 complement to validate and improve existing emission inventories.

1 The NO₂ tropospheric vertical column densities (TVCD, the vertically integrated
2 concentration in the troposphere) retrieved from satellite measurements provide
3 valuable global information on the spatio-temporal patterns of NO_x, including trends
4 (e.g., Richter et al., 2005; Schneider and van der A, 2012; Hilboll et al., 2013),
5 responses of NO₂ level changes to air quality control as well as economic and political
6 factors (e.g., Duncan et al., 2013; Lelieveld et al., 2015), and temporal variations like
7 weekly cycles in NO₂ TVCDs (Beirle et al., 2003; Russell et al., 2010; Valin et al.,
8 2014). In addition, the satellite NO₂ measurements have been applied to quantify NO_x
9 emissions. In a pioneering study (Leue et al., 2001), the downwind decay of NO₂
10 TVCDs in continental outflow regions was used to estimate a (constant) NO_x lifetime,
11 which was then applied to project global NO_x emissions from the measured mean
12 NO₂ TVCDs. Later on, CTMs were employed to exploit satellite observations as a
13 constraint towards improving NO_x emission inventories (e.g., Martin et al., 2003;
14 Konovalov et al., 2006; Kim et al., 2009; Lamsal et al., 2011). The derived top-down
15 inventories show pronounced differences relative to bottom-up estimates and their
16 accuracy has been validated by the improved performance of model simulations with
17 respect to in-situ measurements (e.g., Martin et al., 2006). However, the top-down
18 inventories are usually determined at regional/global scale related to the spatial
19 resolution of CTMs, while the spatial scales relevant for individual emission hotspots
20 (power plants or cities) are not resolved. In addition, modelled lifetimes have large
21 uncertainties (Lin et al., 2012) due to the highly non-linear small-scale chemistry in
22 urban areas, and are thus probably not appropriate for relating NO₂ TVCDs to NO_x
23 emission rates at city level.

24 With the launch of the Ozone Monitoring Instrument (OMI) (Levelt et al., 2006) with
25 high spatial resolution (13×24 km² at nadir), individual large sources like Megacities
26 and power plants can be resolved. In a recent study, Beirle et al. (2011) averaged OMI
27 NO₂ measurements separately for different wind directions, thereby constructing clear
28 downwind plumes which allow a simultaneous fit of the effective NO_x lifetimes and
29 emissions, without the need of a chemical model. Valin et al. (2013) adopted this

1 approach, but rotated satellite NO₂ observations according to wind directions such
2 that all the NO₂ columns are aligned in one direction (from upwind to downwind).
3 The rotation procedure accumulated a statistically significant data set to examine the
4 dependence of NO_x lifetime on the wind speed. Following studies e.g. de Foy et al.
5 (2015) and Lu et al. (2015) adopted this plume rotation technique and quantified NO_x
6 emissions from isolated power plants and cities over the US respectively, which
7 showed that the method can give reliable estimates over multi-annual averages and
8 even provide estimates of emission trends with reasonable accuracy. de Foy et al.
9 (2014) also analyzed the performance of the method using model simulations with
10 fixed *a priori* lifetimes and realistic wind data, which proved that the fitted results
11 were accurate in general and showed best performance for strong wind cases.
12 Alternative approaches based on model functions with multiple dimensions, e.g. a two
13 dimensional Gaussian functions (Fioletov et al., 2011) and a three dimensional
14 function (Fioletov et al., 2015), were also proposed to estimate lifetimes and
15 emissions.

16 However, so far all studies assume that the source of interest can be considered as a
17 “point source”, which works well for isolated sources like e.g. the city of Riyadh,
18 showing a high contrast against clean background with small and smooth TVCDs.
19 However, for sources located in a heterogeneously polluted background, a
20 modification of these methods is needed in order to account for the effect of
21 interfering sources within small distances.

22 In this work, we present a new method for the quantification of NO_x lifetimes and
23 emissions for power plants and cities located in polluted background. The mean OMI
24 NO₂ distribution for 2005–2013 is calculated separately for calm conditions as well as
25 for different wind direction sectors according to ECMWF (European Center for
26 Medium-range Weather Forecast) wind fields. The mean lifetime is derived from the
27 change of the observed NO₂ patterns under windy versus calm conditions. NO_x
28 emissions of power plants and cities over China and the US are subsequently

1 quantified from the integrated TVCDs and the derived lifetimes, and compared to
2 bottom-up emission inventories.

3 **2 Methodology**

4 **2.1 Satellite NO₂ data**

5 We base this study on NO₂ TVCDs from the OMI tropospheric NO₂ (DOMINO)
6 v2.0 product (Boersma et al., 2011), which is provided by the Tropospheric Emissions
7 Monitoring Internet Service (TEMIS, <http://www.temis.nl>). OMI is a UV-VIS
8 nadir-viewing satellite spectrometer (Levelt et al., 2006) on board the Aura satellite
9 (Celarier et al., 2008), launched in 2004. NO₂ columns are derived from radiance
10 measurements, using the Differential Optical Absorption Spectroscopy (DOAS)
11 algorithm (Platt, 1994). OMI provides daily global coverage with a local equator
12 crossing time of approximately 13:45 pm. It detects radiance spectra from 60
13 across-track pixels with ground pixel sizes ranging from 13×24 km² at nadir to about
14 13×150 km² at the outermost swath angle (57°).

15 The 10 outermost pixels on both sides of the swath are excluded in this study to limit
16 the across-track pixel width <40 km. From June 2007, OMI has shown severe
17 spurious stripes, known as row anomalies that are likely caused by an obstruction in
18 part of OMI's aperture
19 (<http://www.knmi.nl/omi/research/product/rowanomaly-background.php>). The
20 affected pixels are also excluded from the analysis. Only mostly cloud free
21 observations (effective cloud fraction <30%) are considered in this study.

22 Mean NO₂ TVCDs over the US and China during “ozone season” (May-September)
23 for 2005 to 2013 are calculated separately for calm (wind speed below 2 m/s) and 8
24 different wind direction sectors following the approach in Beirle et al. (2011). We
25 focus on the ozone season to include the photochemically relevant months for ozone
26 production (USEPA, 2014) and to exclude the winter data with larger uncertainties
27 due to larger solar zenith angles, variable surface albedo (snow), and longer NO_x
28 lifetime. Wind fields at a lat/long grid of 0.36° width are taken from the ECMWF

1 ERA interim reanalysis (Dee et al., 2011), and the horizontal wind components of the
2 lowermost 500 m are averaged. Individual clear-sky observations of NO₂ TVCDs are
3 assigned to a 2×finer grid (0.18°, comparable to the extent of OMI ground pixels)
4 according to the pixel center coordinates, and associated with the corresponding
5 ECMWF wind fields interpolated in time.

6 **2.2 NO₂ outflow models and lifetime/emission fits**

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

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

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

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

1 2.2.1 Isolated point source outflow model: Lifetime and Emissions

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

$$7 \quad M(x) = E \times (e \otimes G)(x) + B \quad (1)$$

$$8 \quad e(x) = \exp\left(-\frac{x-X}{x_0}\right) \quad \text{for } x \geq X, 0 \text{ otherwise} \quad (2)$$

$$9 \quad G(x) = \frac{1}{\sqrt{2\pi}\sigma} \exp\left(-\frac{x^2}{2\sigma^2}\right) \quad (3)$$

10 E represents total emissions, B represents a constant background; X is the location of
11 the source (relative to the a priori co-ordinates of the site under investigation), x_0 is
12 the e-folding distance downwind; and σ is the standard deviation of $G(x)$. The mean
13 lifetime τ is derived from the e-folding distance x_0 by division by w , the mean
14 projected wind speed. By this approach, emissions and lifetimes of NO_2 are fitted
15 simultaneously.

16 Uncertainties are estimated from the confidence intervals of individual fits, the
17 variability of fit results of the same location for different wind directions, and the
18 dependency of *a priori* assumptions like fit intervals and the detailed choice for the
19 applied wind data, as inferred from sensitivity studies (see the supplementary online
20 material of Beirle et al., 2011, for details). In addition, the uncertainty of NO_2 VCDs
21 of about 30% is transmitted to the final emission estimate. Final errors are of the order
22 of 50% for lifetimes, and 60% for emissions, with (i) the fit uncertainty, (ii) the
23 uncertainties introduced by the applied wind data, and (iii) uncertainties for VCDs
24 (affecting only the emission estimate) being the most important contributions.

25 In Beirle et al. (2011) lifetime and emissions are derived for nine isolated hot spots
26 exhibiting high NO_2 TVCDs over a clean background within about 200 kilometers.

1 But this method cannot be applied to hot spots surrounded by additional significant
2 sources, like Harbin (Fig. 1), as by definition, the method can only represent a single
3 “point source” convolved with a Gaussian function. For instance, an additional source
4 at 100 km with only 10% of the emissions of the source under investigation causes a
5 lifetime bias of ~20 %, as the fit tries to “explain” increased downwind values by a
6 longer lifetime (see Fig. S1 and explanations in the supplement). For an interfering
7 source of the same order as the source of interest, the method fails completely.

8 **2.2.2 Mixed source outflow model: Lifetime**

9 We develop an alternative method accounting for emissions from multiple sources.
10 The patterns of line densities under windy conditions result from the transport,
11 chemical decay and spatial smoothing of emission patterns. The basic idea is to use
12 the NO₂ patterns observed under calm conditions, $C(x)$, as proxy of emission patterns
13 instead of assuming a single point source as in previous studies. Lifetime information
14 is then obtained based on the observed change of the NO₂ patterns under windy versus
15 calm conditions. Note that the 1-D pattern of line densities under calm conditions has
16 to be determined along the same (wind) direction, for which the line densities under
17 windy conditions are determined. That means that in total eight 1-D line densities
18 under calm conditions are determined for the eight wind directions. However, only
19 directions with reasonable reliability are considered where mean NO₂ line densities
20 for both calm and windy conditions are well defined (i.e., gaps due to missing data are
21 less than 10% in the across-wind integration interval i and less than 20% in the fit
22 interval in wind direction f). We define the new model function $N(x)$ as:

$$23 \quad N(x) = a \times [e \otimes C](x) + b \quad (4)$$

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

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

11 Figure 2a displays the observed line densities for calm (blue) and southerly winds (red)
12 around Harbin, and the fitted model function $N(x)$ (grey). Generally, $N(x)$ describes
13 the observed downwind patterns well: the coefficients of determination (R^2) between
14 observation and fit are 0.96–0.99 with the range of 3.0–4.4 hours for different wind
15 directions, as shown in Fig. 2a-e.

16 Like in Beirle et al. (2011), the lifetime τ is derived by the ratio of the fitted e-folding
17 distance and the mean wind speed¹: $\tau = x_0/w$. For Harbin, τ is computed to be 3.9
18 hours with a typical 95% confidence interval (CI) of ± 0.6 hours for southerly winds.
19 Averaging the fit results for all wind direction sectors with a good fit performance (i.e.
20 $R > 0.9$, lower bound of CI > 0 , and CI width < 10 h,) yields $\tau = 3.5$ hours with a
21 standard deviation of 0.6 hours (Fig. 2), using the fit residues as well as the CI of τ as
22 inverse weights, as in Beirle et al. (2011).

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 $-2\% \sim 3\%$). But the effect could be larger for persistent winds and for larger thresholds for calm.

1 Here we assumed that the removal of NO_2 can be simply described by a first order
2 loss, and thus the chemical decay of NO_2 follows an exponential decay function $e(x)$
3 (Eq. (2)) with an e-folding distance x_0 , which yields an overall, effective lifetime τ . In
4 Beirle et al. (2014), it was investigated how far the estimated lifetime by this approach
5 might be biased in case of temporal fluctuations of both emissions and instantaneous
6 lifetimes. The impact of such fluctuations was found to be rather small.

7 **2.2.3 Mixed source outflow model: Emissions**

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

16 According to mass balance, the total mass of NO_x equals the emission rate times
17 lifetime. Emissions can thus be derived in a three-step approach by (a) integrating
18 observed TVCDs originating from the source of interest to calculate the total mass of
19 NO_2 , (b) scaling NO_2 to NO_x , and (c) division by the lifetime τ , which was derived as
20 described in the previous section.

21 (a) Total NO_2 mass

22 In order to quantify the total NO_2 mass of the target source, the observed TVCDs have
23 to be integrated around the source, in which (1) interferences with neighboring
24 sources have to be avoided and (2) a polluted background has to be appropriately
25 accounted for. Thus, we base the estimation of the total NO_2 mass on the mean
26 TVCDs under calm conditions, to minimize interferences by advection. Again, we

1 calculate line densities by integrating the NO₂ TVCDs in “across-wind” direction²,
2 but for a smaller interval v representing the spatial extent of megacities or urban
3 centers, but exclude neighboring sources. Here we define $v=40$ km.

4 We then perform a non-linear least-squares fit of a modified Gaussian function $g(x)$ to
5 these line densities under calm wind condition, as illustrated in Fig. 3. The line
6 densities integrated perpendicular to the different wind direction sectors are used to
7 constrain the fitted A in $g(x)$:

$$8 \quad g_i(x) = A \times \frac{1}{\sqrt{2\pi}\sigma_i} \exp\left(-\frac{(x-X)^2}{2\sigma_i^2}\right) + \varepsilon_i + \beta_i x \quad (5)$$

9 i represents the wind direction sector. Note that the projections of line densities under
10 calm wind conditions for opposite wind direction sectors, e.g., north and south, are
11 just mirrored. Thus, we combined the projections for opposite wind direction sectors.
12 That is, i represents Southeast-Northwest, South-North, Southwest-Northeast and
13 East-West respectively. X is the location of the source (relative to the a priori
14 co-ordinates of the site under investigation). σ_i is the standard deviation of the
15 Gaussian $g_i(x)$, and ε_i and β_i represent an offset and a possible linear gradient in the
16 background field respectively. While the e-folding distance is fitted for each wind
17 direction separately (and mean lifetimes might actually be different for each wind
18 direction), the emissions are not expected to depend on wind direction. We thus use
19 all available wind directions to perform one fit of all functions $g_i(x)$ simultaneously
20 with wind sector dependent backgrounds, but one overall parameter A .

21 The NO₂ amount A (in molecules) around the source on top of the (wind sector
22 dependent) background is determined by fitting the functions $g_i(x)$ simultaneously for
23 all available wind directions.

24 The fit of total NO₂ mass is performed over the interval h in wind direction (see Fig.
25 S2). The fit interval h has to be chosen to be larger than v in order to allow for a
26 meaningful fit of $g(x)$. We set h to 200 km for cities (see Fig. S2) and 100 km for

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

1 power plants respectively. The fit interval thus potentially includes interfering sources.
2 However, these interferences are in first order accounted for by the linear variation of
3 the background fitted in function $g_i(x)$. Note that the fit $g(x)$ is less sensitive to
4 interfering sources compared to the original fit of $M(x)$ in Beirle et al. (2011), as
5 lifetime is not involved here.

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

$$9 \quad f(\sigma_i) = \int_{-20km}^{20km} \frac{1}{\sqrt{2\pi}\sigma_i} \exp\left(-\frac{(x-X)^2}{2\sigma_i^2}\right) / \int_{-\infty}^{\infty} \frac{1}{\sqrt{2\pi}\sigma_i} \exp\left(-\frac{(x-X)^2}{2\sigma_i^2}\right)$$

10 (6)

11 Note that we consider a larger interval (60 km for ν and 300 km for h) for Pearl River
12 Delta, which is a megalopolis covering nine prefectures over an area of about 56,000
13 km². We tabulated the intervals chosen for fits for different cases in Table 1.

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

17 (b) Scaling NO₂ to NO_x

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

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

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

1 **2.3 Uncertainties**

2 We define total uncertainties of the fitted lifetimes and emissions analogue to the
3 procedure described in Beirle et al. (2011), basically based on the fit performance and
4 the dependencies on the a priori settings as investigated in sensitivity studies. Here we
5 shortly list the main sources of uncertainties and how they are quantified. Further
6 details are provided in Sect.3 of the Supplement. The resulting quantitative error
7 estimates are given and discussed below along with the derived lifetime and emission
8 estimates.

9 The confidence intervals (CIs) resulting from the least-squares fits of Eq. (4) and Eq.
10 (5) directly reflect the uncertainties of the derived lifetimes and emissions. In addition,
11 the standard deviations of the fitted lifetimes for different wind direction sectors
12 provide information on the consistency of the method. Both effects can be quantified
13 straightforward and are included in the total uncertainty, contributing about 30% for
14 lifetimes and 20% for emissions arising from CI and less than 40% for both arising
15 from standard mean error (see Sect.3 of supplement), respectively. The dependency
16 on the a priori choices of integration and fit intervals are quantified based on
17 sensitivity studies and found to be of the order of 10%.

18 Accurate wind fields are required for the sorting procedure as well as for the
19 conversion of the downwind decay from a function of distance into a function of time.
20 Again, the impact of the *a priori* settings (horizontal ECMWF wind fields vertically
21 integrated over the lowest 500m) are estimated based on sensitivity studies. In
22 addition, ECMWF wind fields have been checked by comparison to in-situ sonde
23 measurements, which generally agree well, except over complex terrain (see Sect.
24 2.6). The comparison of projected wind speeds of from ECMWF and sonde
25 measurements allows to estimate the uncertainty of the lifetime fit caused by
26 uncertainties of both ECMWF wind speeds and direction. Overall, the uncertainty
27 related to wind fields is about 30%.

28 In addition, the derived emissions (but not the lifetime) are affected by the uncertainty
29 of tropospheric NO₂ TVCDs (30%) and the NO₂/NO_x ratio (10%).

1 In the supplement, we also discuss sophisticated effects such as the potential
2 dependence of lifetimes on wind conditions, the assumption of a constant NO_2/NO_x
3 ratio, and the concept of a single lifetime describing the downwind evolution of NO_2
4 over several hours. These effects have been found to be rather small.

5 We define total uncertainties of the resulting lifetimes and emissions as the root of the
6 quadratic sum of the above mentioned contributions, which are assumed to be
7 independent.

8 **2.4 Bottom-up emission inventories**

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

13 For power plants, we use the China coal-fired Power plant Emissions Database
14 (CPED) developed by Liu et al. (2015) based on unit-level fuel consumptions and
15 emission factors derived from various sources, and the US Emissions & Generation
16 Resource Integrated Database (eGRID) using emissions derived from continuous
17 emissions monitoring systems (available at
18 <http://www.epa.gov/cleanenergy/energy-resources/egrid/>) (USEPA, 2014). For cities,
19 we use the Multi-resolution Emission Inventory for China (MEIC:
20 <http://www.meicmodel.org>) compiled by Tsinghua University, the accuracy of which
21 has been validated by extant researches (e.g., Ding et al., 2015), and the global
22 inventory of the Emissions Database for Global Atmospheric Research (EDGAR)
23 v4.2 (EC-JRC/PBL, 2011) for the US.

24 For the comparison to the derived top-down estimates, a 8-year (2005–2012) average
25 from CPED and a 4-year (2005, 2007, 2009 and 2010) average from eGRID for the
26 ozone season are used for power plants, of which the uncertainties are about 30% (Liu
27 et al., 2015) for CPED and 10% for eGRID (5% arise from continuous emissions
28 monitoring systems (Gluck et al., 2003) and another 5% arise from yearly variations

1 in emissions after 2010), respectively. In addition, the mean emissions for the ozone
2 season of the years 2005–2012 in MEIC and the mean annual emissions for the years
3 2005–2008 in EDGAR are used for cities, of which the uncertainty is estimated to be
4 within a factor of 1/2 and 2 according to the MEIC and EDGAR expert judgment of
5 “medium magnitude of uncertainty” (Olivier et al., 2002). The bottom-up urban
6 emissions derived from regional/global inventories have larger uncertainties
7 compared to power plant emissions, primarily arising from the low-resolution activity
8 rates/emission factors at regional level, and the spatial allocation technique using
9 surrogates to break regional-based emission data down to cities. Furthermore,
10 temporal coverage of bottom-up emissions is limited, inducing additional
11 uncertainties. For instance, a decline in NO₂ TVCDs from the years 2005–2008 to
12 2009–2013 with an average total reduction of $14 \pm 9\%$ (mean \pm standard variation) is
13 detected for investigated US cities (Fig. S3). However, the most recent year available
14 in EDGAR v4.2 is 2008, which cannot reflect the recent decline in NO_x emissions,
15 thus overestimate the average emissions.

16 For the comparison of bottom-up and top-down emissions for individual sites, the
17 power plant inventories directly represent the stack emissions of individual facilities.
18 For total city emissions, the gridded emission inventories have to be integrated over
19 the metropolitan area for which the proposed top-down method is sensitive. Here, we
20 define this area as $40 \times 40 \text{ km}^2$, consistent with the considered interval v in Sect. 2.2.3.
21 For PRD, we consider a larger interval of $120 \times 120 \text{ km}^2$.

22 **2.5 Selection of investigated sources**

23 For this study, we choose large power plants and cities across China and the US as the
24 pre-selected candidates, of which bottom-up emission information is available from
25 inventories described above. Power plants with NO_x emission rates greater than 10
26 Gg/yr (CPED/eGRID) are investigated. Power plants located in urban areas (100 km
27 around city centers) are excluded by visual inspection satellite imagery from Google
28 Earth. The top 150 largest cities (rank in GDP/GDP per capita in 2013) in China and

1 the 47 large US cities selected for analyses in Russell et al. (2012) were also
2 examined. To assure a good fit performance, the following criteria have been defined:
3 (1) The signal of the source is strong, i.e., the mean NO₂ TVCD in a circle of 100 km
4 around the location center is larger than 1×10^{15} molec/cm²; and (2) Fit results with
5 poor performance are discarded (see sections 2.2.2 and 2.2.3 for details). The number
6 of wind direction sectors with a good lifetime fit performance is 4 on average. Table
7 S2 of the supplementary material provides a list of all sources under investigation
8 which passed the criteria, including 24 power plants and 69 cities across China and
9 the US.

10 **2.6 Impact of topography**

11 The accuracy of fitted lifetimes is highly dependent on the accuracy of the a priori
12 wind directions (used for “sorting” the satellite NO₂ observations) and velocities
13 (used for converting x_0 into τ). However, accurate modelling of wind fields on small
14 scales is challenging for large-scale models like ECMWF, which do not resolve urban
15 scales. Consequently, wind fields might be biased in particular over complex
16 mountainous terrain, related to the difficulties in resolving the characterization of
17 small-scale orography in models (Beljaars et al., 2004).

18 We investigate the impact of topography by comparing ECMWF wind fields to
19 2005–2013 sounding measurements assembled by University of Wyoming
20 (<http://weather.uwyo.edu/upperair/sounding.html>), and illustrate it for the cities of
21 Harbin (plain terrain) and Taiyuan (mountainous city in Shanxi, China) in Fig. 4. In
22 the top panels, topography used by ECMWF is compared to the topographic data from
23 the 30-arc-sec global land topography “GTOPO30” archived by the U.S. Geological
24 Survey (available at <https://lta.cr.usgs.gov/GTOPO30>, rescaled to 0.05°). Topographic
25 variations are smeared out significantly by the topographic model used in ECMWF,
26 due to its coarser spatial resolution of 0.36°. The bottom panels show statistics for
27 wind vectors below 500 m during daytime (12:00) and nighttime (0:00) from both
28 ECMWF and the sounding measurements. The frequency distribution of wind

1 directions (in 45 degree bins) shows a very good agreement in Harbin, but not in
2 Taiyuan: here southerly flows dominate according to sounding measurements, while
3 easterly winds dominate in ECMWF.

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

10 Note that an error in a priori wind direction generally leads to a misclassification
11 during the sorting of the satellite data (see also Sect. 3 of the supplement). In such a
12 case, the assumed wind component in direction of the sector is higher than the actual
13 projection; if, for instance, the true wind would be 5 m/s from north, but the model
14 wind is 5 m/s from east, the case is classified as easterly, while the actual easterly
15 wind is 0. This leads to a systematic high biased projected wind speed in Eq. (4), and
16 thus a low biased lifetime. Thus, mountainous sites often yield very low lifetimes
17 (Table S2).

18 As the fitted lifetimes, and thus also emissions, rely on appropriate wind fields, we
19 exclude mountainous sites from the following analysis. We simply define a site as
20 mountainous where the absolute difference in elevation between ECMWF and
21 GTOPO is larger than 250 m. A total of seven power plants and 16 cities are rejected
22 based on the criteria, as listed in Table S4. Seven sites in Table S3 fulfill this criteria
23 and 6 of them present low correlation ($r^2 < 0.5$) in wind speeds between ECMWF and
24 sounding measurements.

25 **3 Results and Discussions**

26 We applied our modified method for determining NO_x lifetimes and emissions to 17
27 power plants and 53 cities across China and the US (see Fig. 5), which passed the

1 criteria defined in Sect. 2.5 and Sect.2.6. Some strong cities and power plants are not
2 included as they are mountainous, e.g. Denver or Salt Lake City.

3 **3.1 Lifetimes**

4 Figure 6 illustrates the fitted NO_x lifetimes for power plants and cities across China
5 and the US, which demonstrates the wide applicability of the modified method
6 developed in this study. The derived lifetimes in “ozone season” (May-September) are
7 3.8 ± 1.0 hours (mean \pm standard deviation) on average with ranges of 1.8 to 7.5 hours.
8 These values are in agreement to previously reported NO_x lifetimes (e.g., Beirle et al.,
9 2004; Schaub et al., 2007; Beirle et al., 2011; Valin et al., 2013) and correspond to a
10 mean OH concentration of the order of 10^7 molecules/cm³ (Valin et al., 2013), which
11 is a realistic number for a polluted urban plume around noon (e.g., Kramp and
12 Volz-Thomas, 1997; Dillon et al., 2002; Hofzumahaus et al., 2009). For the
13 investigated sites, average lifetime for Power Plants (3.5 hours) was found to be
14 slightly shorter than for cities (3.9 hours). Individual lifetimes have uncertainties of
15 about 60%. But, still, Fig. 6 indicates that lifetimes are not completely random, but
16 show systematic spatial patterns. We could not unambiguously relate the variability of
17 NO_x lifetime to a driving parameter, like surface elevation, mean wind characteristics,
18 or latitude. But there is a tendency that NO_x lifetime is longer in heavily polluted
19 regions with higher NO₂ TVCDs, e.g., eastern China and eastern US: The mean NO₂
20 TVCD for the ozone season in a circle with a radius of 100 km around sources with
21 lifetimes over 5 hours is 6.3×10^{15} molec/cm², while it is only 1.3×10^{15} molec/cm² for
22 sources with lifetime less than 2 hours. This finding might be related to nonlinear NO_x
23 chemistry, resulting in a positive correlation between NO_x lifetimes and NO₂ TVCDs
24 when the concentration of NO_x is high (Valin et al., 2013). However, we also find that
25 a high NO_x concentration does not necessarily correspond to a long lifetime, and the
26 correlation between NO_x lifetime and NO₂ TVCDs is rather low ($r^2=0.22$), probably
27 due to the complex NO_x chemistry, which is as well affected by meteorological and
28 chemical variability, like variations in UV flux, water vapor and VOC levels. In
29 addition, we used tropospheric HCHO columns from OMI (provided by BIRA, De

1 Smedt et al., 2015) to investigate a potential link between VOCs and the estimated
2 NO_x lifetimes. We averaged the HCHO columns for the ozone season during
3 2005–2013, and explore their relationship with NO_x lifetime. We observed systematic
4 spatial patterns for the HCHO columns, e.g., the concentration of HCHO is higher in
5 the eastern US than the western US, which is similar to the spatial distribution of NO_x
6 lifetime. However, the overall correlation between HCHO TVCDs and NO_x lifetime
7 is still rather low ($r^2 = 0.13$). Thus, we see no indication that VOCs are the main
8 drivers for the spatial variability of NO_x lifetime.

9 The proposed method estimates the mean lifetime basically from the change of NO₂
10 patterns for windy vs. calm conditions. Valin et al. (2013) report on a dependency of
11 the NO_x lifetime on wind speed, with generally shorter lifetimes for higher wind
12 speed. In addition, other factors, like the satellite's sensitivity (affected by e.g. cloud
13 properties or the vertical NO_x profile) and the NO₂ background might change
14 systematically between calm and windy conditions. In the fitted model function $N(x)$,
15 a scaling factor a and an offset b are required in order to achieve a good fit
16 performance for the individual fits, which probably compensate for these effects. But
17 on average, the derived values for a and b are close to 1 and 0, respectively: a is $0.9 \pm$
18 0.1 (mean \pm standard deviation) and b is $0.0 \pm 0.1 \times 10^{23}$ molec/cm (mean \pm standard
19 deviation).

20 Thus, possible systematic effects due to all kind of changes between calm and windy
21 conditions are small, and they are considered with a 10% of contribution in the total
22 uncertainty for NO_x lifetimes (see supplement).

23 We also performed an additional analysis of seasonal mean lifetimes (see supplement,
24 Fig. S4). Wintertime is excluded in the seasonal analysis, because in winter satellite
25 data exhibits larger uncertainties and line densities under calm wind condition are
26 often unrepresentative of the emission pattern due to longer NO_x lifetimes. The
27 seasonal lifetimes reveal higher uncertainties due to a smaller number of available
28 satellite observations compared to the ozone season and thus reduced number of wind
29 direction sectors that yielding a valid fit. The uncertainty is sometimes too large to get

1 reasonable seasonal patterns for a specific location. But still a systematic seasonal
2 variability can be observed for most non-mountainous cases: mean lifetimes are found
3 to be shorter in summer (3.2 hours) compared to spring (4.2 hours) and autumn (4.5
4 hours), as expected.

5 For some locations, the resulting emissions vary considerably over season, which
6 again can be attributed to the poor statistics; in particular spatial gaps can cause high
7 uncertainties of the determined total NO₂ mass based on Eq. (5).

8 **3.2 Emissions**

9 Figure 7 compares the derived NO_x emissions to bottom-up emission inventories
10 (Sect. 2.4) for all 17 power plants and 53 cities. For power plants, the comparison (Fig.
11 7a) shows excellent agreement with a high correlation coefficient ($r^2=0.93$). Average
12 emissions are 29 mol/s in bottom-up inventories and 31 mol/s in top-down estimates.
13 The relative difference (defined as $(E_{\text{top down}} - E_{\text{bottom-up}})/E_{\text{bottom-up}}$) is within 30% for
14 most sites, and $5\% \pm 27\%$ (mean \pm standard deviation) on average. For China and the
15 US, the relative differences are $4\% \pm 18\%$ and $5\% \pm 31\%$ respectively, confirming the
16 rather good agreement between CPED/eGRID bottom-up emission inventories and
17 top-down estimates.

18 For the investigated cities, good agreement (Fig. 7b) between the derived emissions
19 and the bottom-up emissions is reassuring and the r^2 reaches 0.84 (0.87 and 0.74 for
20 China and the US respectively). The relative difference between derived NO_x
21 emissions and bottom-up emissions for cities is larger than that for power plants,
22 reaching $9\% \pm 49\%$ ($1\% \pm 46\%$ and $20\% \pm 51\%$ for China and the US respectively)
23 on average. This is probably related to the higher uncertainties of the bottom-up
24 inventories for cities compared to those for power plants. Bottom-up emission
25 inventories, developed by different researchers, often differ significantly from each
26 other, due to the application of various assumptions and extrapolations associated
27 with their knowledge of activity data and emission factors. We further compared the
28 representations of China's urban emissions between MEIC and EDGAR, as shown in

1 Fig. 8. Huge discrepancies are found between EDGAR and top-down estimates
2 (relative difference: $311\% \pm 412\%$) with large negative bias in the bottom-up.
3 Considering the deviation in national total NO_x emissions is far less (20.7 and 24.9
4 Tg- NO_2 for year 2008 in EDGAR and MEIC respectively), the large bias could be
5 primarily explained by the spatial distributions in the two inventories.

6 Both MEIC and EDGAR calculate emissions as province/country totals and distribute
7 them to grids using spatial proxies. By comparing spatial proxies used in the two
8 inventories, we identified the major differences in spatial allocation methods between
9 them: (1) MEIC used an in-house high-resolution database (CPED) to represent
10 power plant emissions in China while EDGAR used CARMA (Wheeler and Ummel,
11 2008). The coordinates of power plants in CARMA are highly uncertain for China
12 (Liu et al., 2015); (2) for industrial emissions, MEIC first downscaled provincial
13 totals to counties using industrial GDP, and then allocate county emissions to grids
14 with population density. EDGAR directly distributed provincial emissions by
15 population density (EC-JRC/PBL, 2012); and (3) MEIC allocated on-road emissions
16 by vehicle and road type using the China Digital Road-network Map (Zheng et al.,
17 2014), while EDGAR used the product of population density (Gridded Population of
18 the World (GPW) version 3, (CIESIN et al., 2005)) and road network (the Global
19 Roads Inventory Project (GRIP), (PBL, 2008)). All above factors are expected to
20 contribute to the better representations of urban emissions in MEIC than in EDGAR
21 over China, and thus gain better agreement with top-down estimates.

22 It is interesting that EDGAR represents urban emissions much better in the US than in
23 China, even though EDGAR shared the same spatial allocation approach across
24 different countries. One plausible explanation is that spatial proxies work better in the
25 US, implying the linear relationships between emissions and proxies, e.g., vehicle
26 emissions and road densities, industrial/residential emissions and population densities.
27 Different accuracy of spatial proxies among regions may also contribute to the
28 discrepancy of performance in the two inventories. For instance, the GRIP database
29 (<http://geoservice.pbl.nl/website/GRIP/>) missed too many roads for China (Fig. S6).

1 By comparing with a high-resolution emission inventory, the Database of Road
2 Transportation Emissions (DARTE), Gately et al. (2015) argued that EDGAR
3 overestimated on-road emissions in city centers while underestimate at the suburban
4 and exurban fringes, resulting from mismatches between road density and the actual
5 spatial patterns of vehicle activity at urban scales. To better understand the
6 uncertainties associated with the performance of spatial proxies, further
7 source-by-source comparison is required between downscaled regional inventories
8 and high-resolution inventories independent to spatial proxies (e.g., DARTE).

9 The emissions are derived based on the individual fitted lifetimes for each site. If,
10 instead, the mean lifetime of all sites (3.7 hours) would be considered for the
11 calculation of emissions, the correlations to bottom-up emissions are worse compared
12 to the individual fitted NO_x lifetime (Fig. 9). This holds for both, power plants and
13 cities. We conclude that variation of the fitted lifetime is not just the result of
14 statistical noise, but actually carries information on local variability of the oxidizing
15 capacity of urban plumes. The individual lifetimes are thus well suited for the
16 determination of emissions by a mass balance approach.

17 Satellite observations also enable the study of spatial and temporal distributions of
18 SO_2 emissions (e.g., Fioletov et al. (2011)) and even to obtain estimates of SO_2
19 lifetimes and emissions under special circumstances (e.g., Beirle et al. (2014)).
20 However, if the method developed in this study would be applied to SO_2 directly,
21 higher uncertainties have to be expected due to the longer lifetime of SO_2 (see Sect. 5
22 of the supplement for a detailed discussion).

23 **3.3 Uncertainties**

24 Based on the approaches presented in Sect. 3 of the supplement, we estimated that
25 total uncertainties of NO_x lifetime and emissions are within 39%–80% and 55%–91%
26 respectively for all the investigated sites (see Sect. 2.5). For Harbin, relative
27 uncertainties for mean lifetime and emissions are 43% and 58%, respectively.
28 However, it is worth noting that our uncertainty estimate is rather conservative. For

1 power plants, relative differences between bottom-up and top-down estimates are all
2 within 50% (Fig. 7a). As bottom-up emission inventories for power plants are well
3 developed with low uncertainties, the good consistency increases our confidence that
4 the fitted emissions well represent the real-world emission characteristic. Thus,
5 bottom-up inventories may have large biases for cities where emission estimates
6 differ significantly from top-down constraints (i.e., the relative difference far exceeds
7 50%).

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

- 11 • the uncertainty due to wind data is ~30% (affecting both τ and emissions),
- 12 • effects of a possible systematic change of NO₂ TVCDs from calm (used for fit
13 of E) to windy (used for fit of τ) conditions are small (<10%),
- 14 • the derived emissions (but not the lifetimes) are affected by the uncertainty of
15 the NO₂ TVCDs (~30%) and the NO_x/NO₂ scaling factor (~10%),
- 16 • the dependency on the definition of integration and fit intervals is about 10%,
- 17 • the CI of fitted lifetimes and total NO₂ mass is about 30% and 20%,
18 respectively; the standard mean error of fitted lifetimes for different wind
19 directions is less than 40% (see Sect.3 of supplement).

20 All involved uncertainties contain both statistical fluctuations as well as systematic
21 effects. By ongoing satellite measurements (e.g. TROPOMI), i.e. longer available
22 time periods, and the much better temporal sampling of upcoming geostationary
23 satellite missions such as GEMS (Kim et al., 2012), TEMPO (Chance et al., 2012), or
24 Sentinel-4 (Ingmann et al., 2012), statistical uncertainties will decrease. In addition,
25 we expect further improvement of the presented lifetime fit method by using regional
26 meteorological models that are more capable of representing wind fields in the
27 planetary boundary layer especially for mountainous region. Also the uncertainties of
28 TVCDs from satellite retrievals, which is still the largest single component of total

1 uncertainty in top-down emission estimates, is expected to decrease in the coming
2 years: input data such as surface albedo or a priori profiles will improve, and the
3 current intensive validation efforts (e.g., DISCOVER-AQ
4 (<http://discover-aq.larc.nasa.gov/>) and AROMAT (<http://uv-vis.aeronomie.be/aromat/>))
5 will help to identify and remove systematic errors. It can thus be expected that total
6 uncertainties of the proposed method will decrease significantly within the next
7 decade.

8 **4 Conclusion**

9 We developed a new method to estimate NO_x lifetimes and emissions of power plants
10 and cities in polluted background from satellite NO₂ observations. The method
11 improves upon that of Beirle et al. (2011) by explicitly accounting for interferences
12 with neighboring strong NO_x sources by using NO₂ spatial patterns under calm wind
13 conditions as proxy of the patterns of emission sources. Lifetimes are derived from
14 the change of NO₂ distributions under windy compared to calm conditions. NO_x
15 emissions are derived by mass balance: the total mass of NO₂ originating from the
16 source of interest is divided by the lifetime derived for the corresponding source.

17 The new method for determining NO_x lifetimes and emissions was applicable for 24
18 power plants and 69 cities over China and the US, including 23 mountainous sites.
19 We exclude the derived results for 23 mountainous sites from the analysis, which are
20 expected to have larger uncertainties owing to the inaccurate wind data. The derived
21 lifetimes for 70 non-mountainous sites are 3.8 ± 1.0 hours (mean \pm standard deviation)
22 on average with ranges of 1.8 to 7.5 hours. We observed systematic spatial patterns
23 for the derived lifetimes, which however could not be simply explained by a specific
24 driving parameter. Generally, higher lifetimes were found in heavily polluted regions,
25 but the overall correlation between NO₂ TVCDs and NO_x lifetime is quite low ($r^2 =$
26 0.22).

27 The derived top-down NO_x emissions are generally in very good agreement with
28 bottom-up emission inventories, in particular for power plants, while correlations for
29 cities were lower, probably due to the higher uncertainty of the bottom-up inventories

1 for cities. Compared to MEIC, the EDGAR global inventory significantly
2 underestimated NO_x emissions for Chinese cities, because spatial proxies used in
3 EDGAR may misrepresent emission spatial patterns for China.

4 Owing to the global continuous monitoring of satellite measurements, this method can
5 be applied to quantify the emissions from various cities and power plants even in
6 polluted background around the world. For this study, we choose large sources across
7 China and the US as the pre-selected candidates, of which the good-quality bottom-up
8 emission information, particularly for power plants, is available. Further investigation
9 on sources located in other regions, in particular, Europe, will be performed in the
10 near future, with collating the corresponding bottom-up emission inventories. This
11 capability will further be enhanced with future satellite instrument like TROPOMI
12 (Veefkind et al., 2012) featuring higher spatial resolution. In addition, upcoming
13 geostationary satellite instruments will enable studies on the diurnal cycle of the NO_x
14 lifetime. More accurate estimates for emission rates, trends and seasonality can be
15 expected, which will serve as an independent data source to validate bottom-up
16 emission estimates in the future.

17

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12

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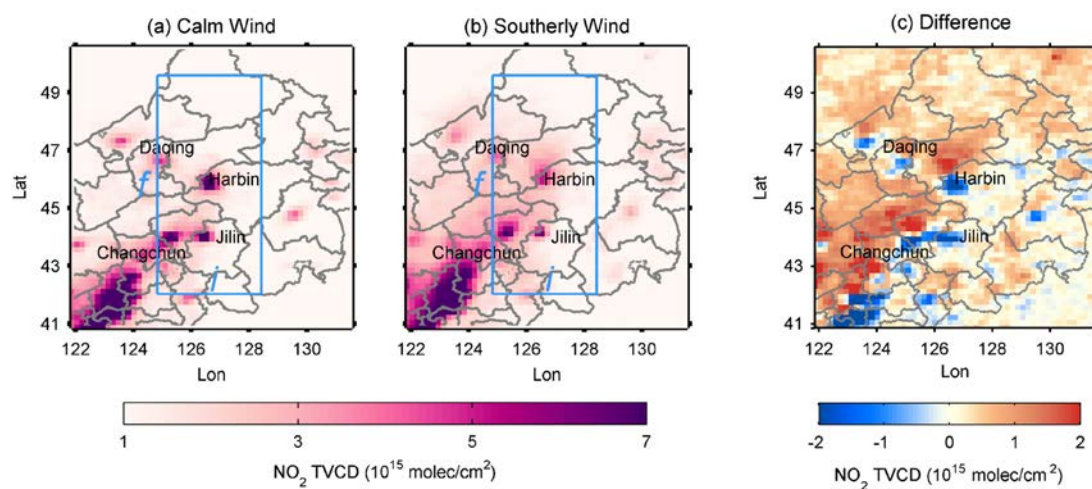
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1 **Tables**

2 Table 1. Intervals chosen for the fit of the NO_x lifetime and total mass.

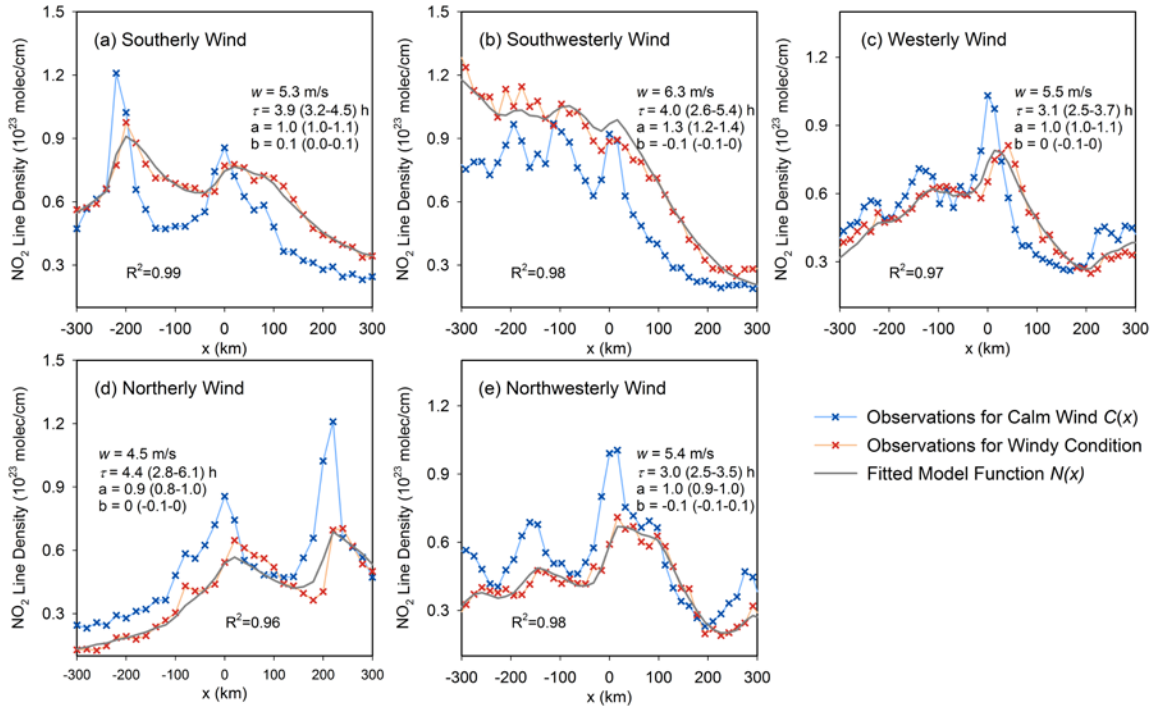
Category	Interval (km)	
	Wind direction (fit)	Across-wind direction (integration)
Fit lifetime	f 600	i 300
Fit total mass	h	v
Power plant	100	40
City	200	40
Pearl River Delta	300	60

1 Figures



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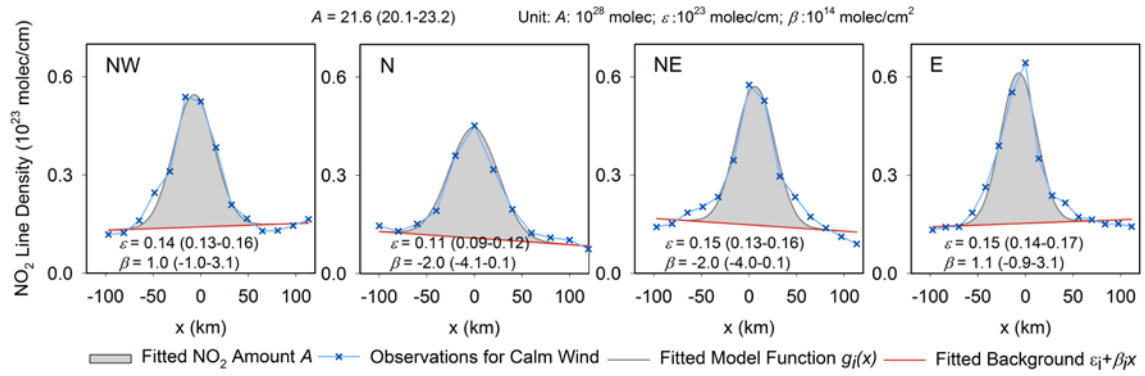
4 Figure 1. Mean NO₂ TVCDs around Harbin for (a) calm, (b) southerly wind conditions and (c) their
5 difference (southerly – calm). For the fit of lifetimes, the mean NO₂ TVCDs are integrated over
6 interval i in across-wind direction to calculate line densities and the fit is performed over the fit interval
7 f (blue lines in (a) and (b); see Sect. 2.2.2 for details).



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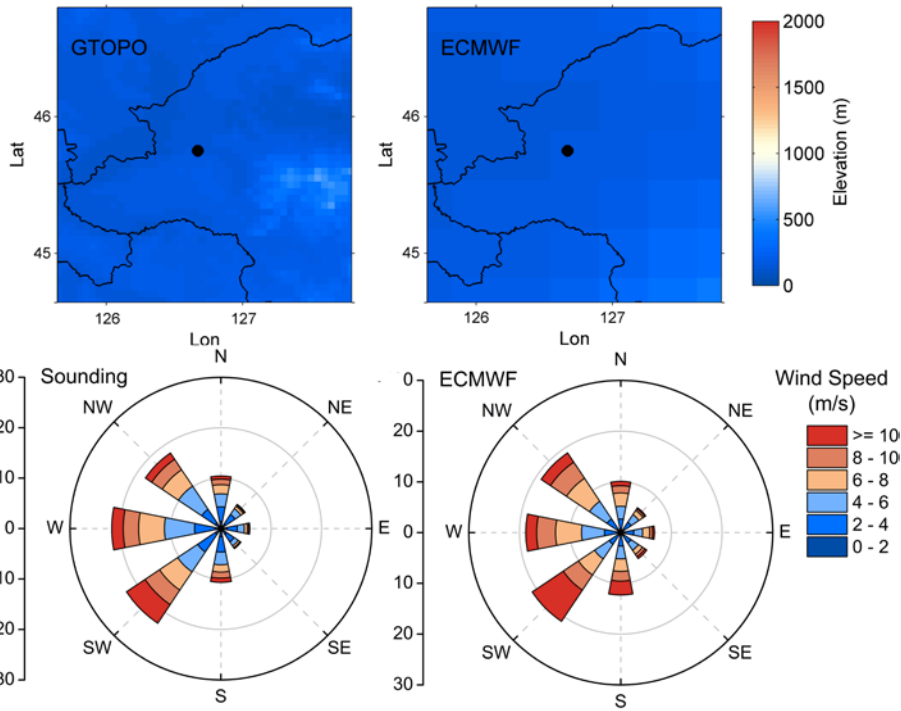
3 Figure 2. NO₂ line densities around Harbin for different wind direction sectors. Crosses: NO₂ line
 4 densities for calm (blue) and (a) southerly, (b) southwesterly, (c) westerly, (d) northerly and (e)
 5 northwesterly (red) winds as function of the distance x to Harbin center. Grey line: the fit result $N(x)$.
 6 The numbers indicate the net mean wind velocities (windy – calm) from ECMWF (w), the lifetime τ ,
 7 the factor a and offset b resulting from the least-squares fit with 95% confidence interval. NO₂ line
 8 densities for the remaining wind direction sectors are dismissed due to missing data (see the criteria of
 9 “reliability” defined in Sect. 2.2.2).



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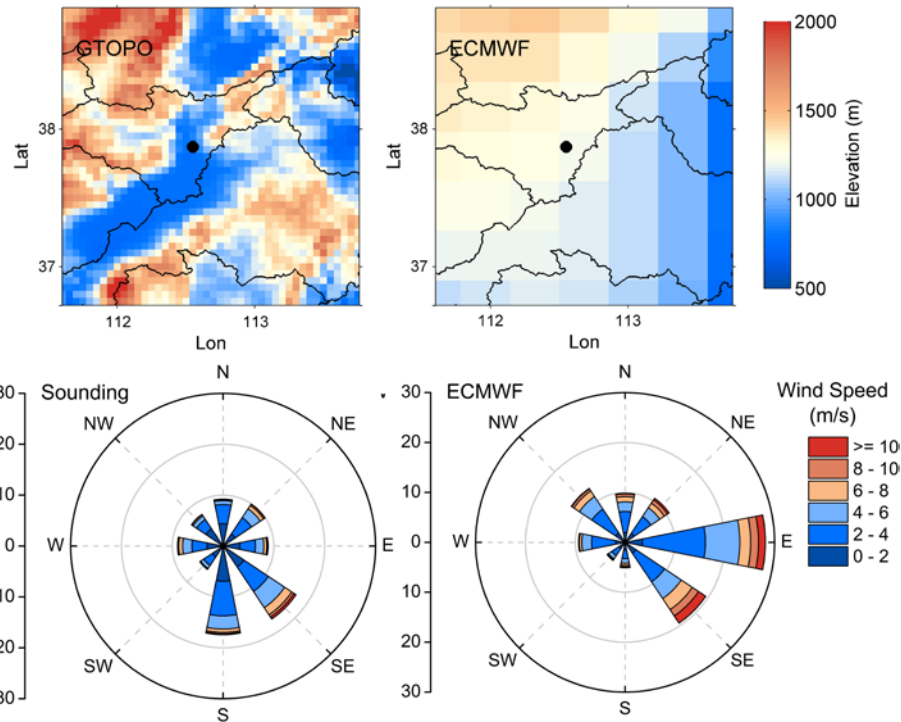
3 Figure 3. NO₂ line densities in Harbin for northwest, north, northeast and east directions (from left to
 4 right). Crosses: NO₂ line densities for calm winds as function of the distance to Harbin center x . Grey
 5 line: the fit result $g_i(x)$. Pink line: the fitted background $\varepsilon_i + \beta_i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

1 (a) Harbin



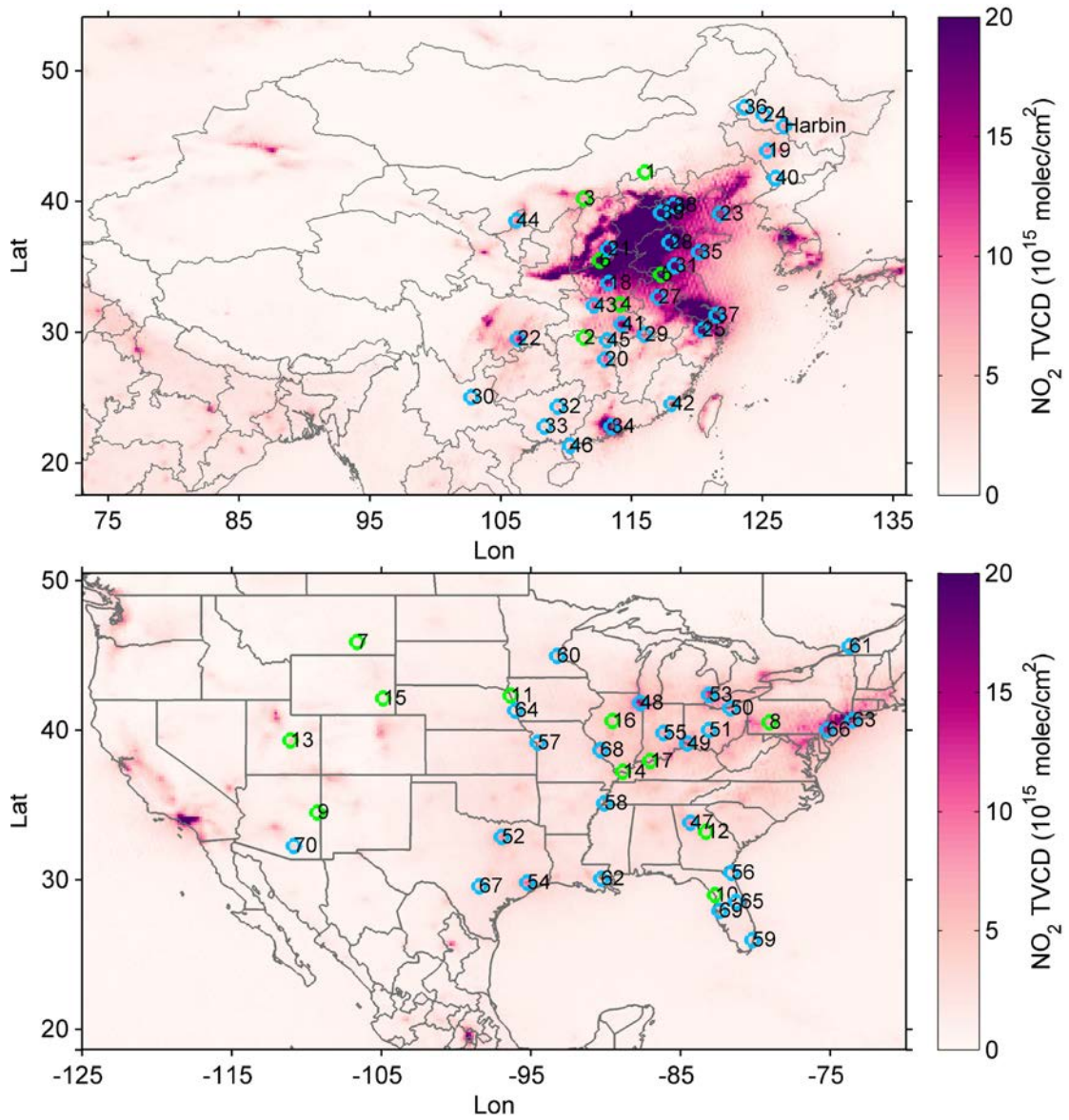
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3 (b) Taiyuan



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



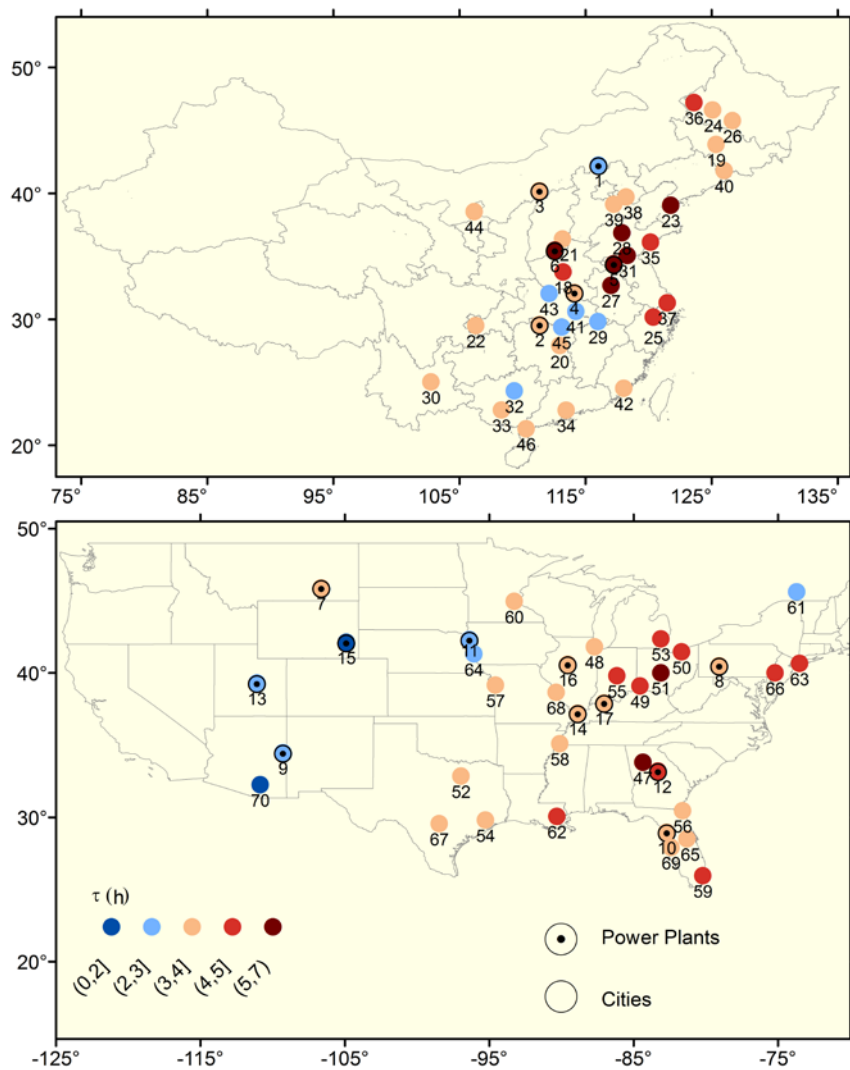
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3 Figure 5. Average OMI NO₂ TVCDs during ozone season (i.e., May to September) over China and the

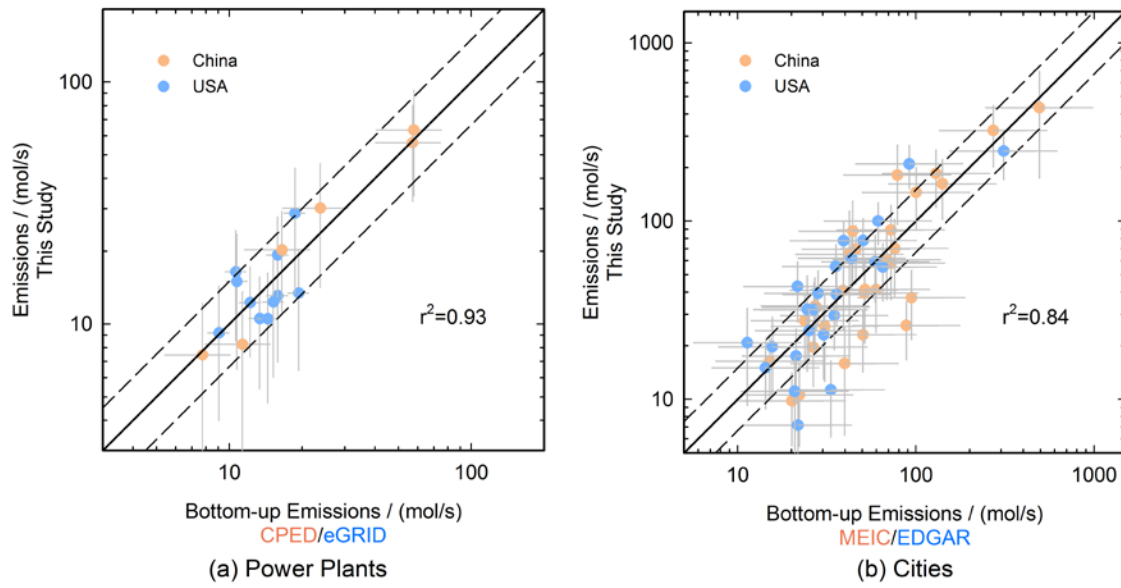
4 US for the period 2005–2013. Green and blue symbols indicate the 17 power plants and 53 cities

5 investigated in this work, respectively. Power plants and cities are labelled by their IDs (see Table S2).



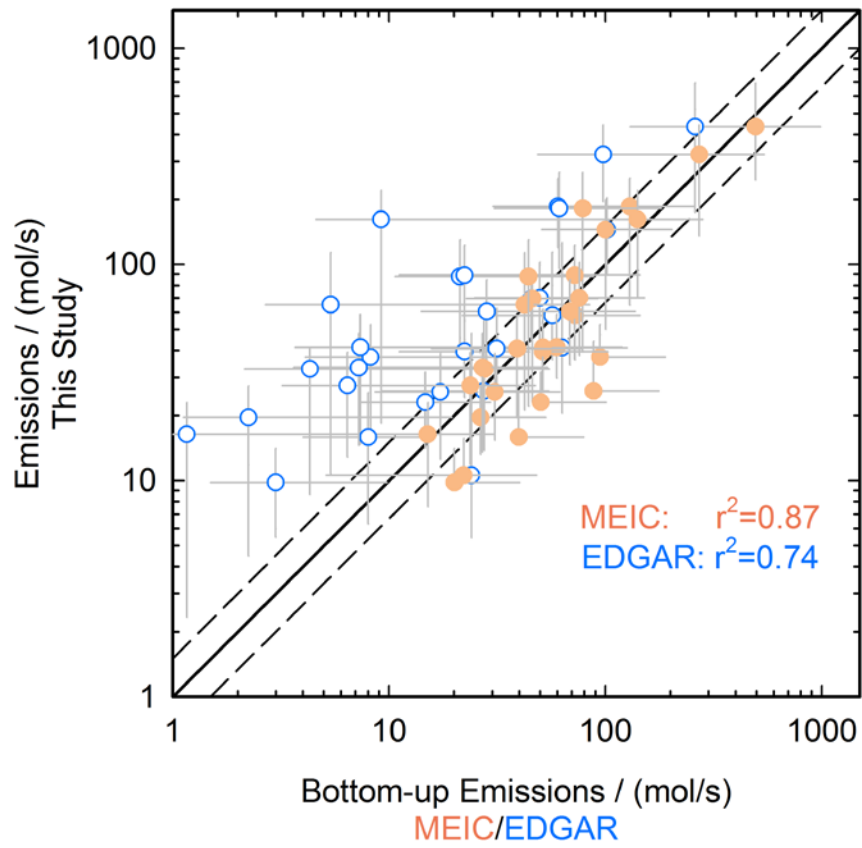
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Figure 6. Fitted NO_x lifetimes (color coded) for investigated emission sources over China and the US. Locations of power plants are indicated by dots. Power plants and cities are labelled by their IDs (see Table S2).



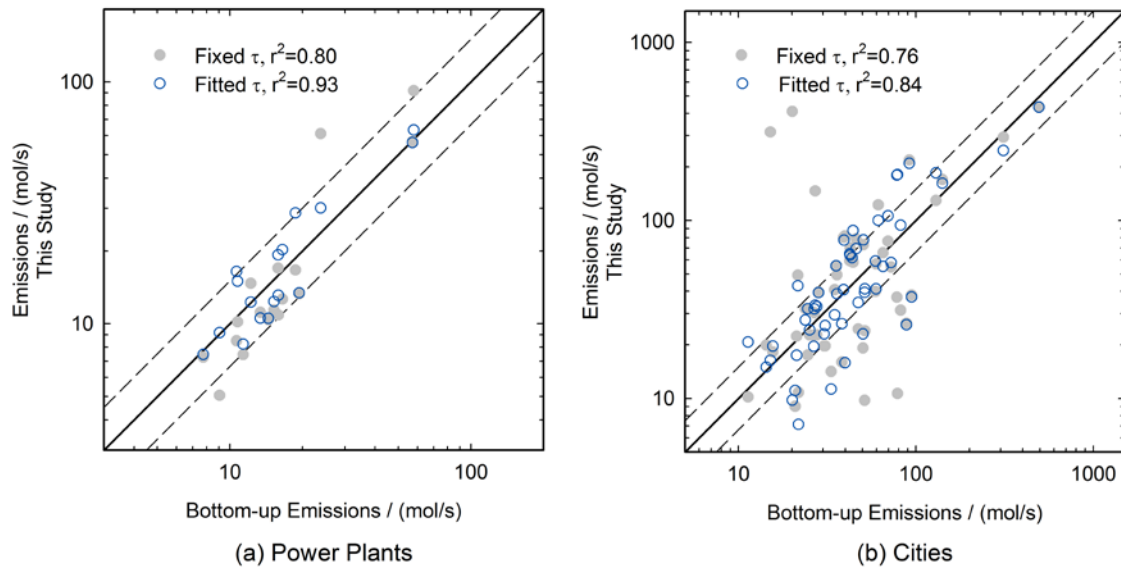
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Figure 7. Scatterplots of the derived NO_x emissions for investigated (a) power plants and (b) cities versus bottom-up emission inventories. Emissions are given in mol/s calculated assuming a constant emission rate. Urban emissions from bottom-up inventories are integrated over 40 km ×40 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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3 Figure 8. Same as Figure 7 but Scatterplots of the derived NO_x emissions for investigated cities versus
4 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 hours) (filled circles) versus the respective estimates from bottom-up emission inventories. Emissions are given in molec/s 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.