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Recent reduction in NO_x emissions over China: synthesis of satellite observations and emission inventories

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Abstract

Tropospheric nitrogen dioxide (NO_2) column densities detected from space are widely used to infer trends in terrestrial nitrogen oxide (NO_x) emissions. We study changes in NO_2 column densities using the Ozone Monitoring Instrument (OMI) over China from 2005 to 2015 and compare them with the bottom-up inventory to examine NO_x emission trends and their driving forces. From OMI measurements we detect the peak of NO_2 column densities at a national level in the year 2011, with average NO_2 column densities deceasing by 32% from 2011 to 2015 and corresponding to a simultaneous decline of 21% in bottom-up emission estimates. A significant variation in the peak year of NO_2 column densities over regions is observed. Because of the reasonable agreement between the peak year of NO_2 columns and the start of deployment of denitration devices, we conclude that power plants are the primary contributor to the NO_2 decline, which is further supported by the emission reduction of 56% from the power sector in the bottom-up emission inventory associated with the penetration of selective catalytic reduction (SCR) increasing from 18% to 86% during 2011–2015. Meanwhile, regulations for vehicles also make a significant contribution to NO_x emission reductions, in particular for a few urbanized regions (e.g., Beijing and Shanghai), where they implemented strict regulations for vehicle emissions years before the national schedule for SCR installations and thus reached their NO_2 peak 2–3 years ahead of the deployment of denitration devices for power plants.

1. Introduction

Nitrogen oxides (NO_x) play a key role in tropospheric chemistry as a precursor of tropospheric ozone and secondary aerosols, both of which impact human health and climate significantly (Jacob *et al* 1996, Seinfeld and Pandis 2006). China is the largest NO_x emitter, which is thought to contribute 18% of global NO_x emissions (EDGAR 4.2, European Commission (EC): Joint Research Centre (JRC)/Netherlands Environmental Assessment Agency (PBL) 2011). Power plants and vehicles are two of the largest sources of NO_x emissions in China, which contributed to 28.4% and 25.4% of the total anthropogenic emissions in 2010 respectively according to the study of Zhao *et al* (2013). As a consequence of the rapid growing economy, China's NO_x emissions have increased by a factor of three during the last two decades (Zhang *et al* 2007, Kurokawa *et al* 2013). The rapid increase in emissions has caused serious environmental problems, particularly poor air quality.

To reduce the severe air pollution, the Chinese government has announced emission control actions to reduce national NO_x emissions by 10% during 2011–2015 (The State Council of the People's Republic of China 2011). To meet this plan, new emission regulations have been implemented, such as installation of selective catalytic reduction (SCR) equipment at power plants and stricter emission standards for vehicles. These regulations have been accelerated since 2012 after the first national environmental standard for limiting the amount of fine particles in the air was approved by China's State Council (Zhang *et al* 2012), in particular for 'key regions' including the Greater Beijing region (including Beijing, Tianjin and Hebei), the Yangtze River delta and the Pearl River delta (Zhao *et al* 2013). The upward trend of NO_x emissions in China has been slowed down or reversed and the turning point is expected to be different for various regions due to the disparities in the implementation of the regulations.

Satellite observations have been widely used to quantify NO_x emissions (e.g., Martin *et al* 2003, Beirle *et al* 2011) and evaluate emission changes (e.g., Richter *et al* 2005, van der A *et al* 2008, Russell *et al* 2012) by providing up-to-date and continuous time series of tropospheric NO_2 columns with global coverage. For China, satellite data have been successfully used to estimate surface NO_x emissions (Wang *et al* 2012, Mijling *et al* 2013, Liu *et al* 2016), detect the long-term increasing trend in NO_x emissions (van der A *et al* 2006, Zhang *et al* 2007) and the short-term emission decline during specific events such as the Beijing Summer Olympic Games (Mijling *et al* 2009) and the economic recession (Lin and McElroy 2011). In addition, measurements from multiple sensors of both aerosols and trace gases have also been used simultaneously to provide better constraints on the emission inversion (Xu *et al* 2013). Recent studies have observed a decreasing trend in NO_2 columns for more economically developed regions, e.g., the Pearl River Delta, Shanghai and Beijing (Gu *et al* 2013, Jin and Holloway 2015, Duncan *et al* 2016). Krotkov *et al* (2016) and Miyazaki *et al* (2016) further reported a more widespread decline in NO_2 levels over China from 2012. However, most existing studies point out the recent decline of NO_2 over China only at a national level or for a few specific cities. Few researchers have yet attempted to explore the regional diversity of the NO_2 trend and give in depth interpretations of the cause of the changes.

The main objective of this work is to identify the major reason for the recent change in NO_2 columns over China using both satellite observations and bottom-up emission inventories. We investigated the changes in tropospheric NO_2 column densities from 2005 to 2015 at the provincial level to figure out the spatial distribution of the timeline of the NO_2 decline. Only NO_2 columns dominated by anthropogenic sources were considered by subtracting background NO_2. We also compared satellite data with timely bottom-up information, in particular unit-based power plant emissions, to better understand the observed change in NO_x column densities and diversity over regions.

2. Data and method

2.1. Satellite data and bottom-up inventory

The Ozone Monitoring Instrument (OMI) is a UV–vis nadir-viewing satellite spectrometer (Levelt *et al* 2006) on board the Aura satellite (Celarier *et al* 2008) that was launched in 2004. It detects radiance spectra by 60 across-track pixels with ground pixel sizes from $13 \times 24\,\mathrm{km}^2$ at nadir to about $13 \times 150\,\mathrm{km}^2$ at the outermost swath angle (57°). It provides daily global coverage with a local equator crossing time of approximately 13:40 h. The OMI radiance measurements are fitted to obtain slant NO_2 columns using the differential optical absorption spectroscopy algorithm (Platt 1994). We use retrieved NO_2 column data from the Dutch OMI tropospheric NO_2 (DOMINO) v2.0 product (Boersma *et al* 2011) in this study, which are available from the tropospheric emissions monitoring internet service (TEMIS, http://www.temis.nl).

The individual OMI measurements are sampled at $0.125° \times 0.125°$ resolution by averaging the original satellite observations weighted by the size of the overlapping surface area. Only cloud-free observations (cloud fraction <30%) with surface albedo values less than 0.3 are used. From June 2007, OMI has shown severe spurious stripes, known as row anomalies that are likely caused by an obstruction in part of the OMI's aperture (http://www.knmi.nl/omi/research/product/rowanomaly-background.php). Thus, the observations affected by row anomalies are filtered out.

We use the Multi-resolution Emission Inventory for China (MEIC: http://www.meicmodel.org) compiled by Tsinghua University as bottom-up emission information. The MEIC model provides anthropogenic NO_x and other pollutant emissions from the year 1990 to the present, using a technology-based methodology (Li *et al* 2015). It can represent emission characteristics, in particular changes, from multiple sources by considering the influence of technology renewal and regulations on emissions. It improved the accuracy of bottom-up NO_x emissions developed by the same group (Zhang *et al* 2007, 2009a) by including a unit-based China coal-fired power plant emissions database (CPED, Liu *et al* 2015) and a high-resolution vehicle emission modelling approach (Zheng *et al* 2014).

2.2. Method

We extracted yearly and monthly OMI tropospheric NO_2 column densities for each province of China during 2005–2015. We defined background regions in this study as regions with average annual NO_2 column densities less than $1 \times 10^{15}\,\mathrm{molec\,cm^{-2}}$ (Cui *et al* 2016) or with average NO_2 column densities for summer exceeding those for winter (Van der A *et al* 2006). These regions are dominated by natural NO_x sources and excluded from the analysis. Figure 1

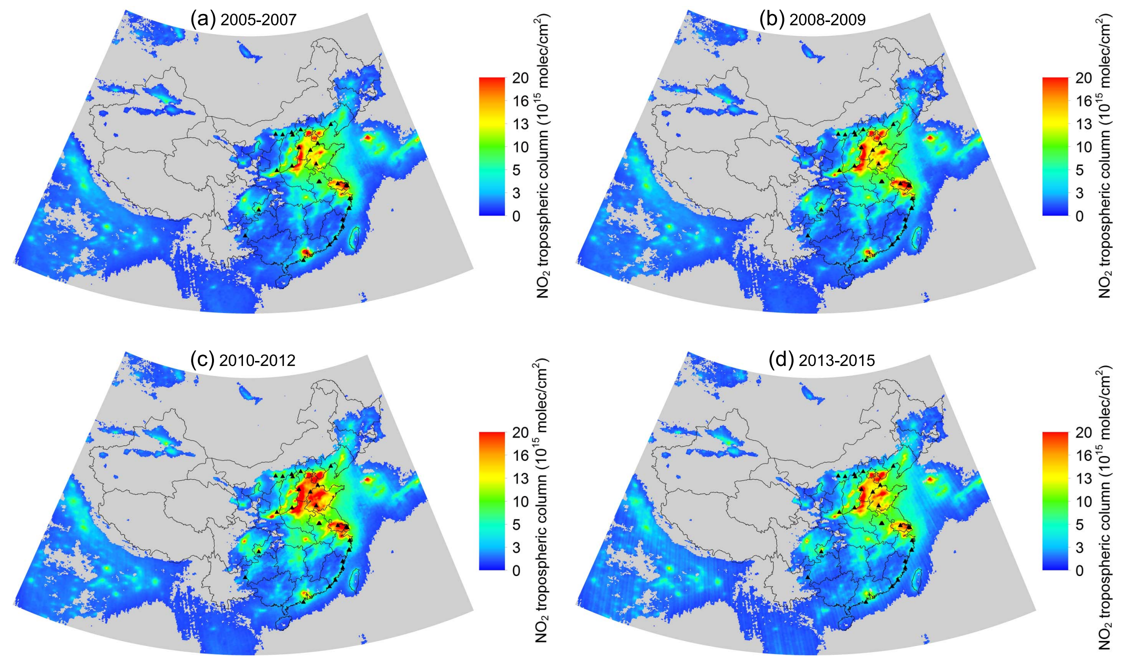


Figure 1. Average OMI NO_2 column densities over China for (a) 2005–2007, (b) 2008–2009, (c) 2010–2012 and (d) 2013–2015. The background regions are coloured by grey. The locations of selected power plants are indicated by triangles.

shows an average map of NO_2 columns used for analysis of the period of 2005–2015 over China.

We further calculate monthly mean NO_2 column densities of grid cells where power plants are located to infer NO_x emissions from the power sector. Because of the fact that the current OMI NO_2 products only capture strong power plant plumes (Street *et al* 2013), only power plants with a capacity larger than 2500 MW were selected. The locations of 27 large power plants used in this analysis are shown in figure 1. The overall unit capacity for the selected power plants reached 89 GW by 2014, which is equivalent to 11% of the total national capacity.

We computed a running average for a time window of 12 months to smooth monthly fluctuations in NO_2 column densities, thereby removing seasonal variations to indicate the turning point of NO_2 TVCDs for each province, defined as the year when NO_2 TVCDs reached the maximum of the 12 month moving averages for the whole time series (hereafter mentioned as the peak year of NO_2). We compared the peak year of NO_2 with the process for denitration in power plants indicated by the CPED database and other bottom-up emission information from the MEIC model to interpret the reasons for NO_2 changes.

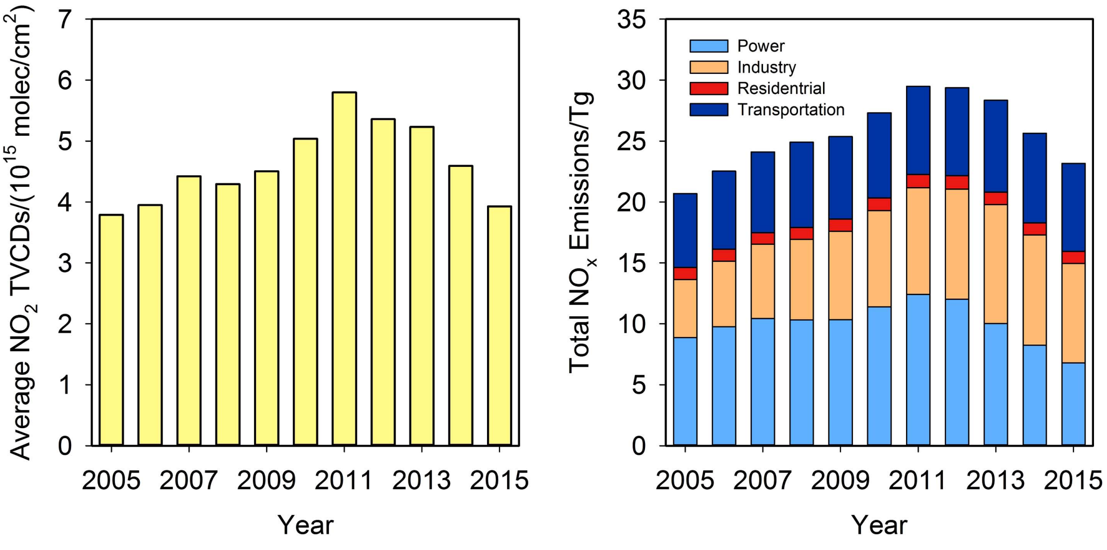
3. Results and discussion

3.1. Observed changes in OMI NO_2 column densities

Figure 2(a) shows the annual OMI NO_2 column densities for 2005–2015. A NO_2 growth of up to 53% is observed from 2005 to 2011, and in contrast, a NO_2 reduction of 32% is observed from 2011 to 2015. A good agreement is detected in trends between OMI observations and NO_x emissions in the bottom-up inventory (figure 2(b)), which displays a large increase of 42% between 2005 and 2011 and a sharp decline of 21% afterwards.

Figure 3 shows the time series (blue circles) of the 12-month moving average of monthly mean OMI NO_2 values for China (only non-background regions in figure 1). In general, a significant and relatively linear growth is observed prior to the year 2012, except the period of the global economic slowdown (2008–2009), which is in good agreement with the findings in Lin and McElroy (2011). The turning point of temporal variation is clearly visible: a continual decline in OMI NO_2 values after the year 2012 is evident.

The dramatic growth in NO_2 prior to 2012 is driven by the increasing fuel consumption (grey circles), in particular of coal which is the dominant fuel type in China. However, coal consumption continued to rise until 2014, increasing with 2% from 2012 to 2013 (National Bureau of Statistics 2014) for instance, which could not explain the simultaneous decline in NO_2 column densities but suggests the effectiveness of emission control measures. One of the major measures to regulate NO_x emissions is the rapid deployment of denitration devices at power plants. The average NO_x emission factors of coal-fired power plants (red squares) decreased from 6.2 to 2.6 g kg^{-1} from 2011 to 2015 with the penetration of denitration devices (defined as the percentage of unit capacity of power plants installing SCR in the total capacity of all the power plants, red triangles)



(a) (b)

Figure 2. (a) Average OMI NO_2 column densities for China during 2005–2015. The background regions coloured in grey in figure 1 are removed from calculation. (b) Total anthropogenic NO_x emissions by sector for China during 2005–2015. The emission data are derived from the MEIC model.

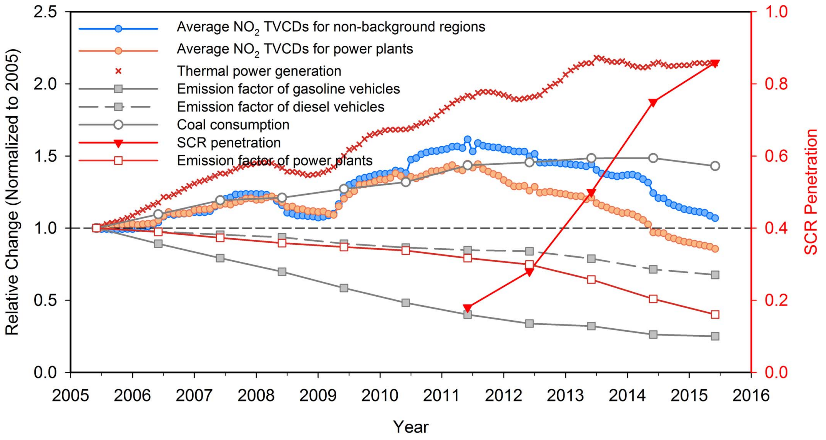


Figure 3. Monthly variations in average NO_2 TVCDs for non-background regions in China and selected power powers, and thermal power generations after a 12-month moving average. Note that the first point of the 12-month moving average represents the data for the period from January to December of 2005. Annual coal consumption (grey circles), average emission factor of gasoline vehicles (grey squares with solid lines), diesel vehicles (grey squares with dash lines) and coal-fired power plants (red squares) , and SCR penetrations (red triangles) for China are also shown.

increasing from 18% to 86%. In addition, significant progress has been made in controlling vehicle emissions in China: gasoline (grey squares with solid lines) and diesel vehicles (grey squares with dash lines) showed a continual decline in average NO_x emission factors, decreasing by respectively 75% and 32% during 2005–2015 based on the estimates in the MEIC model. After the year 2014, a decline of 3.7% in coal consumption contributed to the NO_2 reduction as well.

Figure 3 also displays the moving averages of monthly mean OMI NO_2 values for selected power plants (pink circles) and Chinese thermal power generation (red crosses). Thermal power generation is a good proxy for fuel consumed by power plants, and thus NO_x emissions when no new control equipment is put into operation. Generally, observed changes in NO_2 column densities are consistent with changes in power generation before early 2012, but after that year, NO_2 columns tend to show different trends compared to power generation. The thermal power generation kept stable from 2013 to 2015, indicating that the reduction with a corresponding decrease of 26% in NO_2 column densities was not associated with the reduced electricity production, but with emission control measures.

3.2. Comparison with deployment of denitration devices

New emission standards for thermal power plants (Ministry of Environmental Protection of China (MEP) 2011) were carried out in 2012; these required power plants to install flue-gas denitration devices like SCR. Afterwards, multiple policies were being put into place to guarantee proper operations of denitration devices. Power plants are required to install continuous monitoring systems and transfer real-time data to the government and the electricity produced with high SCR operation rate can be sold at premium price (Ministry of Environmental Protection of China (MEP) 2013). In this way, the total capacity of denitration devices rose sharply with a share growing from 18% to 86% during 2011–2015 (China Electricity Council (CEC) 2012–2016) overtaking the rate of construction of new coal power plants.

The deployment procedure of denitration devices for power plants is found to be in reasonable agreement with the peak year of NO_2 (see the definition in section 2.2), which suggests that the observed reduction in NO_2 was presumably the result of the installation of denitration devices for power plants. Figure 4(a) shows how the peak year of NO_2 is distributed over the provinces. The results for the province of Heilongjiang, Liaoning and Tibet are dismissed due to a lack of observations (i.e., minimum 30 observations for every month). Most provinces (18 out of 29) reached the NO_2 peak in the year 2012. However, the five most urbanized regions (i.e., Beijing, Tianjin, Shanghai, Guangdong and Taiwan) were far ahead with a NO_2 maximum prior to 2010 and five other provinces (i.e., Hebei, Sichuan, Chongqing, Liaoning and Xinjiang) fell behind with a peak year in 2013/2014. It is interesting to note that the decrease in NO_2 TVCDs accelerated after 2013, with an average reduction of 3%, 8% and 14% for the period of 2012–2013, 2013–2014 and 2014–2015, respectively.

Figure 4(b) displays the SCR use for each province during 2011–2014 and the year when SCRs were initially deployed for power plants with a share of 10% (hereafter defined as the year of SCR installations). Over 70% of provinces have their peak of NO_2 level in the year of (12 provinces) or one year after (8 provinces) the year of SCR installations. Provinces installing SCR for power plants at slower speed, e.g., Chongqing, Liaoning and Xinjiang, tended to reach the peak of NO_2 columns later. However, the difference between the year of SCR installations in figure 4(b) and the peak year of NO_2 in figure 4(a) is striking for provinces with peaking NO_2 levels prior to 2010 (e.g., Beijing, Shanghai and Guangdong), in which there has been a greater and earlier effort to regulate emissions from other sources (e.g., on-road vehicles) other than power plants.

3.3. Comparison with bottom-up emission inventory

The consistent temporal patterns of NO_2 columns and bottom-up NO_x emissions compared in figure 2 shed light on the driving force behind NO_2 changes. Figure 5 displays changes of NO_x emissions from 2005–2010 by sector and infers how changes in source categories have influenced trends in NO_2 columns. The observed sharp growth in NO_2 in the early years was driven by emissions emitted from multiple sources. Industrial activities are the most notable source, representing 45% of total emission growth during 2005–2010 in China, as a result of the drastic industrial development. Power plants contributed significantly to the growth as well, which was estimated to account for another 36% of total growth. For a certain province, the contribution from power plants was striking, e.g., reaching 65% for Inner Mongolia, due to the rapid construction of new power plants (Zhang *et al* 2009b).

The decline in vehicle emissions for provinces with peaking NO_2 level prior to 2010 shown in figure 5 explains the earlier reductions in NO_2 column densities observed for those urbanized regions. Their NO_x emissions from on-road vehicles decreased by 18% on average when the national average experienced a growth of 15%, as a result of stricter emission standards, control of the stock and shorter turnover times of vehicles. These urbanized regions including Beijing, Shanghai and Guangzhou (the capital of Guangdong) have been required to meet more stringent vehicle emission standards ahead of the national schedule. For example, the Euro III emission standard for gasoline vehicles was implemented in Beijing in 2006, two years earlier than the national requirement, of which the NO_x emission factor is only 12% of aged vehicles with Euro 0 standard (Huo *et al* 2012). Additionally, the speed of vehicle population expansion for urbanized regions slow down as a result of the general trend of developed areas that tend to have slower vehicle population growth (Zheng *et al* 2014), as well as some local policies for controlling vehicle populations. For example, Shanghai restricted vehicle sales by implementing a license plate auction policy in 1994. From then on, only a limited number of new license plates became available to the public each year. As a result of the notable success of emission controls induced by stricter emission standards, contributions to overall emissions from high-emitting aged vehicles (Euro 0 in most cases) are becoming increasingly significant. It has been reported that Euro 0 vehicles contributed to more than 50% of the total vehicle emissions in China in 2009 (Ministry of Environmental Protection of China (MEP) 2010). Therefore, these regions also carried out vehicle retirement programmes to scrap aged vehicles. Other factors including improving urban public transportation system like expanding underground road networks and promotion of alternative fuel technologies also contributed to the emission reduction.

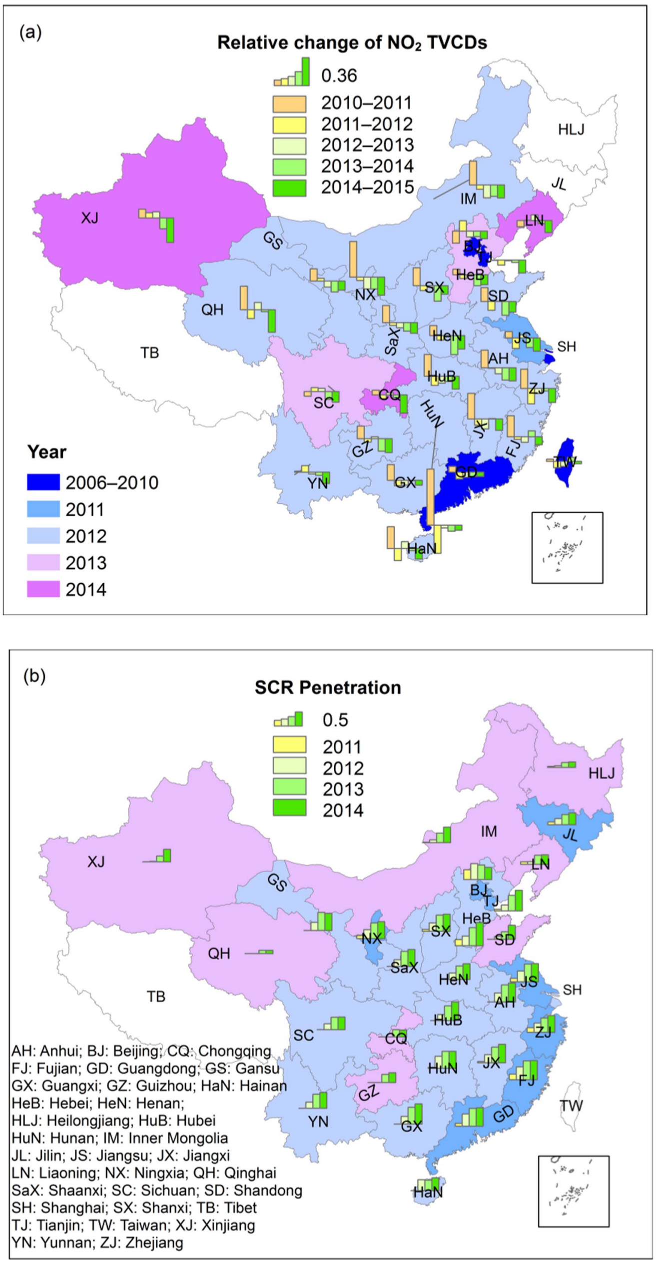


Figure 4. (a) Spatial distribution of the peak year of NO_2. The bar charts denote the average annual change rate of NO_2 TVCDs for the period of 2010–2011, 2011–2012, 2012–2013, 2013–2014 and 2014–2015, respectively. (b) Spatial distribution of the year of SCR installations. The bar charts denote the temporal variations in average SCR penetration during 2011–2014. The bars on the top of the figure are the chart symbol of the legend. The value of 0.36/0.5 next to the bar chart relates to the attribute value that the longest bar in the chart represents.

Figure 5 tells a different story for the period of 2010–2015. In general, emissions were estimated to stop rising, with all sectors experiencing a synchronously slow growth and even decline to meet the cap on NO_x emissions (The State Council of the People's Republic of China 2011). The national total emissions decreased by 15% during 2010–2015, while it increased by 32% for the period of 2005–2010. Power plants have made increasingly significant contributions to emission regulations with a growing use of SCR equipment, 98% of reduction in NO_x emissions was a result of installing SCR during 2010–2015. Meanwhile, due to the implementation of the Euro IV standard for on-road vehicles and the retirement of aged vehicle nationwide, the general increase in vehicle emissions was modest with an annual growth rate of 1% on average for the period of 2010–2015. Not surprisingly, the industrial sector still experienced a growth in emissions, since abatement measures focusing on this sector are not as effective as those for the power sector. It is critical that appropriate development strategies are designed for industry; otherwise rising NO_x emissions associated with highly polluted industry will compensate the reductions earlier achieved by power plants. The improvements in energy efficiency, the phasing out of small and

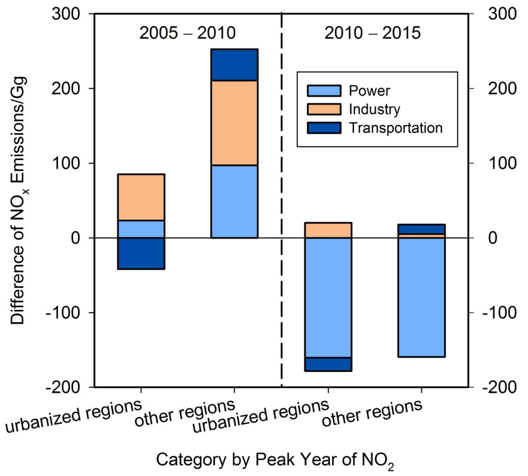


Figure 5. Changes of NO_x emissions between 2005–2010 (left) and 2010–2015 (right) by sector (units: Gg). Urbanized regions are defined as provinces with the peak year of NO_2 prior to 2010 in figure 4. The changes of NO_x emissions are determined by the average emission of provinces in a certain year minus that in the prior 5 years. The emission data are derived from the MEIC model.

inefficient factories, and a wider deployment of denitration devices for polluting industries (i.e. cement and iron industries), should be pursued in the coming years to continue decreasing the NO_2 levels.

3.4. Uncertainty analysis

The variations in NO_2 column concentrations do not necessarily correlate linearly with NO_x emissions (e.g., Turner *et al* 2012, Miyazaki *et al* 2016), due to changes of meteorology, NO_x chemistry and transport. Averaging NO_2 columns over large areas (the size of a province in this study) for a long-term period (12 months in this study) contributes to reducing those influences. We estimate the uncertainty associated with meteorological fluctuations as 5% based on the model simulations which yielded variations in the annual average NO_2 concentrations of about 5% at the comparable spatial scale due to meteorological variability (Andersson *et al* 2007, Velders and Matthijsen 2009). The NO_2/NO_x ratio might differ when NO_x emissions change, due to the feedback of NO_x emissions on NO_x chemistry (Valin *et al* 2013). But the influence is not dramatic and assigned to an assumed uncertainty of 5% based on the finding that the relationship (β) between relative changes in surface NO_x emissions and changes in NO_2 columns has minor variations (<3%) when the perturbation of emissions used for establishing β is doubled (Lamsal *et al* 2011). In addition, the horizontal NO_x transport from neighbouring provinces over the NO_x lifetime contributes to the density of local NO_2 columns, which smears the local relationship between NO_2 columns and NO_x emissions. However, the zonal mean lifetime of NO_x is 3–10 h (Martin *et al* 2003) and the corresponding smearing length scale is around 100 km (Palmer *et al* 2003), far less than the size of a typical province of China; therefore we neglect the influence of transport in our analysis for most provinces. But for the provinces with a relative small size, i.e., the municipality of Beijing, Tianjin and Shanghai, the contribution of NO_2 columns from neighbouring provinces is not negligible. We roughly estimate the uncertainty associated with transport as 5%, based on the assumption that nonlocal sources contribute to nearly half of the local NO_2 over the spatial scale of Beijing (Mijling *et al* 2013) and the interannual variations (standard deviation for the period of 2005 −2015) in NO_2 columns is 10% on average. We define the total uncertainty as the root of the quadratic sum of the above mentioned contributions, which are assumed to be independent, and calculate the uncertainty to be about 10%.

We further quantify the uncertainty of using the peak year of NO_2 columns to indicate the emission peak as the lower and upper bounds of years with NO_2 column concentrations falling between the peak NO_2 ± uncertainty (see table S1 in the supplement). The determination of the peak year is generally robust, with an uncertainty of ±1 year on average. The conclusion about the regional diversity in the peak year has not been fundamentally changed. Beijing, Shanghai and Guangdong are still found to reach their NO_2 peak ahead of the deployment of denitration devices for power plants, taking this uncertainty into account.

Note that the main purpose of this study is to explore emission trends and their driving forces by an intercomparison of two independent databases (bottom-up inventory and top-down observations), rather than to quantify the relationship between variations in NO_2 columns and that in NO_x emissions, which requires chemical transport model simulations. Bottom-up emission estimates for a specific year are expected to have large uncertainties associated with highly uncertain emission factors. However, the bottom-up emission trends are much more reliable, because their driving factors including activities and technology penetrations are derived from reliable statistical sources and assign low uncertainties. Changes in OMI NO_2 columns have been widely used to represent NO_x emission trends in previous studies (e.g., Richter *et al* 2005, Russell *et al* 2012, Krotkov *et al* 2016). The representative of the peak year of NO_2 columns for emission peaks have been confirmed by a top-down inventory (van der A *et al* 2016) using DECSO (daily emission estimates constrained by satellite observation) algorithm (Mijling and van der A 2012), which takes both meteorology and NO_x transport into account. The similar temporal pattern from the two independent databases provides solid evidence for the recent decline in NO_x emissions over China.

4. Conclusion

In this study, we provided a detail description of changes in NO_2 column densities at the provincial level over China from 2005 to 2015 and explored the driving forces behind changes based on the bottom-up emission inventory. The average NO_2 column densities of China peaked at the year 2011, and decreased by 32% from 2011 to 2015. This is in good agreement with the bottom-up emission inventory with a simultaneous decline of 21% in NO_x emissions. The peak year of NO_2 showed a strong diversity over the regions. The NO_2 columns of urbanized regions like Beijing, Shanghai and Guangdong peaked prior to 2010, which was expected as a result of control of vehicle emissions, for example, through new emission standards, shorter vehicle turnover, and limiting the number of vehicles. For other regions, the peak year of NO_2 is closely related to the year of SCR installations, which suggested that the observed reduction in NO_2 was primarily the result of installing denitration devices for power plants. The finding is further supported by the bottom-up estimates that power plants reduced emissions by 56% due to the growth of SCR penetration from 18% to 86% during 2011–2015. Relatively rapid growth in emissions was detected for the industrial sector, which is without effective controls; to further curb air pollution, a stricter control of industrial emissions is required. Otherwise the achieved emission reductions in power plants will be compensated by growing industrial emissions. Nevertheless, we conclude that emission control measures are capable of reducing China's NO_x emissions to a large extent, as is shown by our results, in particular after the year 2012. This strong decrease in NO_x emissions suggests that measures can be taken in developing countries that will reduce emissions alongside with rapid economic development.

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