Comparison of satellite observed tropospheric NO$_2$ over India with model simulations

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ABSTRACT

Nitrogen dioxide (NO$_2$) plays a key role in the chemistry of the atmosphere and is emitted mainly by combustion processes. These emissions have been increasing over India over the past few years due to rapid economic growth and yet there are very few systematic ground based observations of NO$_2$ over this region. We thus take recourse to satellite data and compare tropospheric NO$_2$ column abundances simulated by a chemical transport model, MOZART, with data from the Global Ozone Monitoring Experiment (GOME) for a few locations in India that have seen a rapid economic growth in the last decade. The model generally simulates higher columnar abundances of NO$_2$ compared to GOME observations and does not reproduce the features of the observed seasonal behaviour. The combined uncertainties of the emission inventory and retrieval of the satellite data could be contributing factors to the discrepancies. It may be thus worthwhile to develop emission inventories for India at a higher resolution to include local level activity data. The ten year data (1996–2006) from GOME and SCIAMACHY (SCanning Imaging Absorption spectroMeter for Atmospheric CHartographY) show increasing trends for Indian cities where rapid industrial and vehicular traffic growth has been observed during this period.

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1. Introduction

Nitrogen dioxide (NO$_2$) plays an important role in the production of tropospheric ozone. Its sources, emitted by all combustion processes, lie close to the Earth's surface with the exception of emissions from aircraft and production from lightning. Therefore, an accurate estimate of NO$_x$ emissions is important for understanding ozone chemistry. Rapid economic development has the potential to increase the emissions of nitrogen oxides, especially due to the growing fossil fuel consumption in East Asia (Akimoto, 2003). The global anthropogenic emission for NO$_x$ (NO + NO$_2$), for the year 1990 was about 31 million tons N year^{-1}, with major anthropogenic source identified as fossil fuel combustion (70%, of which the major sources are road transport and power plants) (Olivier et al., 1998).

Studies based on bottom up approaches indicate that the NO$_x$ emissions have been increasing and will continue to increase over India in the coming years. According to one scenario, vehicle population in India increased by about 1.5 times from year 2000 to 2005 (Rao et al., 2004). Energy consumption in India has gone up from 7 million tons of gasoline in 2001 to 8.6 million tons in 2005 (Ministry of Petroleum and Natural Gas, India; Baidya, 2007). A study carried out by the Asian Development Bank has estimated that for India, the total on-road vehicle fuel consumption in the year 2005 was 58 million tons of oil equivalent (Mtoe) (which includes gasoline and petrol), which could rise by about six times to 371 Mtoe in the year 2035 (Lohani, 2006). Correspondingly, they estimate the total NO$_x$ emissions from on-road vehicles at 2.2 million tons in 2005 which could touch a figure of 3.6 million tons in 2035.

Tropospheric NO$_2$ column data retrieved from satellite instruments GOME (Global Ozone Monitoring Experiment) and SCIA-MACHY (SCanning Imaging Absorption spectroMeter for Atmospheric CHartographY) have been used earlier as a proxy for NO$_x$ emissions (Irie, 2005). Due to large uncertainty in emission estimates for India, it is crucial to make a validation comparison for emission inventories used in chemical transport models against satellite data for the Indian region (Kunhikrishnan et al., 2004). In this work, we compare model simulated tropospheric NO$_2$ column abundances with GOME observations for a few selected Indian locations to assess the performance of emission inventories.

It is also important to understand the long term temporal changes in NO$_2$ over India associated with changes in local emissions resulting from rapid urbanisation and increasing demand of energy consumption. However, there are only a few sites in India providing

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limited measurements of NO_2. For example, four years of NO_2 column measurements are available for Ahmedabad (Chakrabarty et al., 2006). The Central Pollution Control Board (CPCB) of India also measures surface NO_2 in a few cities of India, but the uncertainties are too large to make the data useful for long term trend studies. Satellite data can fulfil this requirement by covering a large spatial domain for long periods of consistent observations. A significant increase in tropospheric columnar NO_2 has been found over an industrial area of China (Richter et al., 2005). Increasing trends of NO_2 column have also been detected by Ghude et al. (2008) over polluted regions of India using data from GOME and SCIA-MACHY for the period 1996–2006. Whereas Ghude et al. (2008) report trends and seasonal variation of NO_2 over broad regions of India, we look at the same time series for four Indian cities: Delhi (28.4°N, 77.1°E), Ahmedabad (23°N, 72.5°E), Pune (18.5°N, 73.8°E), and Bangalore (12.9°N, 77.6°E), where vehicular traffic and economic growth have increased rapidly during the period 1996–2000 (www.siamindia.com and Dalvi et al., 2006) and are still on the rise.

2. Data used in the work

2.1. Model description

MOZART is a comprehensive global chemical transport model of atmospheric composition designed to simulate tropospheric chemical and transport processes. MOZART-2 includes detailed NO_x–CH_4–NMHC chemistry for the troposphere, with stratospheric chemistry constrained to climatologies (Horowitz et al., 2003). The horizontal resolution used in this run is 2.8° × 2.8° (equivalent to T42). Thus, there are 128 longitude and 64 latitude grid points. There are 28 vertical sigma levels (dictated by the meteorological input data), from surface to about 40 km altitude.

The model incorporates approximately 63 chemical constituents including O_x, NO_x, HO_x, CH_4, C_2H_6, C_3H_8, C_2H_4, C_3H_6, C_4H_{10}, isoprene, terpenes. The chemical mechanism includes 133 gas-phase, 2 heterogeneous, and 33 photolytic reactions. Source gas emissions are taken at the surface for CO, NO, CH_4, C_2H_6, C_3H_8, C_4H_{10}, C_2H_4, C_3H_6, isoprene, terpenes, CH_3COCH_3 and CH_3OH, from lightning for NO_x (~4 Tg N yr^{-1}) (Price et al., 1997a,b; Pickering et al., 1998) and from aircraft for CO, CH_4, NO_x (0.44 Tg N yr^{-1}). The emissions are taken from the POET (Precursors of Ozone and their Effect on the Troposphere) emission inventory.

The meteorological fields (winds and temperatures) required by MOZART are taken from NCEP reanalysis in this study. The dynamical processes in the model include: (1) *Advection*: this is based on a flux-form semi-Lagrangian scheme (Lin and Rood, 1996) (2) *Convection*: the convection scheme is based on the work of Hack (1994) for mid-level convection and Zhang and McFarlane (1995) scheme for deep convection (3) *Boundary layer exchange*: it is parameterized based on the scheme by Holtslag and Boville (1993) (4) *Surface deposition*: The wet deposition is represented as a first-order loss process within the chemistry operator, using large scale and convective precipitation rates diagnosed by MATCH (Model of Atmospheric Transport and CHemistry), soluble species removed by in-cloud scavenging (Giorgi and Chameides, 1985), highly soluble species are also removed by below cloud washout (Brasseur et al., 1998).

2.2. Satellite measurements of NO_2

The GOME instrument was launched on the ERS-2 in April 1995 and provided global observations of trace gases till June 2003 by observing scattered sunlight in nadir viewing geometry. The SCIAMACHY was launched in March 2002 onboard the ENVISAT satellite and has provided nadir and limb data since then.

Measurements by the above two instruments have been used to retrieve tropospheric columns of NO_2 (Leue et al., 2001; Richter and Burrows, 2002; Martin et al., 2002; Beirle et al., 2003; Boersma et al., 2004). Here we use the KNMI (Royal Netherlands Meteorological Institute) tropospheric columns, which are obtained in three steps (Boersma et al., 2004). First, NO_2 columns integrated along the light path (slant columns) are determined by means of the Differential Optical Absorption Spectroscopy (DOAS) method. Second, the tropospheric slant columns are isolated by subtracting from the retrieved slant columns the stratospheric slant column calculated from the TM4 chemistry-transport model. In the last step, the effect of light path is accounted for by application of air-mass factors (van der A et al., 2008). Only data having less than 20% cloud cover are used in the analysis. The final monthly mean NO_2 column data are publicly available on the TEMIS project website (www.temis.nl) on a global grid of 0.25° × 0.25°. Details on the differences between GOME and SCIAMACHY data can be found in Richter et al. (2005). The city of Delhi has the largest number of missing satellite data.

3. Results and discussion

Daily NO_2 mixing ratio vertical profiles are taken from MOZART runs for the local time of satellite pass (around 10.30 h local time over the equator). The daily tropospheric NO_2 column is calculated from these data according to the method discussed below and then averaged over each month. The monthly mean NO_2 column values thus obtained are used for comparison with satellite data. The satellite data have been averaged over the model grid cell nearest to the city of interest for comparison with the model to study the seasonal variation of NO_2 column in Section 3.1 (Table 6). For the analysis of long term trends (Section 3.2), from the gridded satellite data, we select the grid that is nearest to the location of the city.

The NO_2 column T_c is calculated from the model output mixing ratio as follows.

$$T_c = \int_{z1}^{z2} \chi(z) N_{air}(z) dz = \int_{z1}^{z2} \chi(z) \frac{A\rho_{air}(z)}{M_{air}} dz \tag{1}$$

where $N_{air}(z)$, $\rho_{air}(z)$ and M_{air} are the number density profile, mass density profile and molecular mass of air (in kg $mole^{-1}$). A is the Avogadro's number. From the hydrostatic equation for air, we substitute for the mass density in the above equation, $\rho_{air}(z)dz = dp/g$ where p is the atmospheric pressure and g is the acceleration due to gravity. The above equation therefore reduces to

$$T_c = \frac{A}{(M_{air}g)} \int_{p1}^{p2} \chi(p) dp \tag{2}$$

The NO_2 mixing ratios from the model at different pressure levels can, thus, be integrated to obtain the desired NO_2 column. The tropospheric column is calculated from Equation (2) by integrating from the surface to the thermal tropopause in the model.

Fig. 1(a) and (b) show the spatial distribution of the tropospheric NO_2 column as observed by the GOME instrument and those simulated by MOZART respectively, over the Indian region for the month of December 2000. Overall, MOZART simulated NO_2 columnar amounts are more than those observed by GOME. Large values can be seen over the northern region 25–30°N and 76–80°E (close to the city of Delhi), which we have included in our trend analysis along with the four Indian cities. MOZART simulates higher NO_2 columns than the GOME data for the southern region of India also.
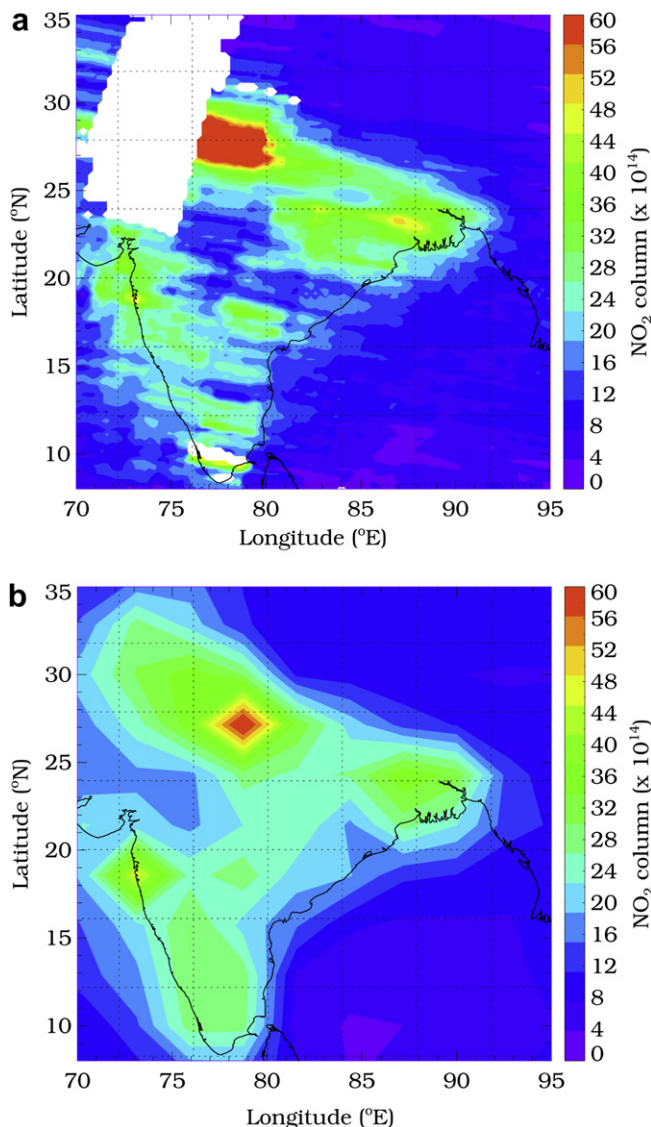
Fig. 1. The spatial distribution of the tropospheric NO_2 column for the month of December 2000 as observed by GOME (a) and as simulated by the model MOZART (b).

3.1. Comparison of model simulation with satellite data: seasonal variation

Lifetime of NO_2 is of the order of 1 day depending on meteorological conditions, its photolysis rate and the concentration of OH radicals. The combination of the variability in meteorological conditions, chemistry and emissions leads to a seasonally dependent NO_2 concentration.

There have been limited observations of NO_2 over the Indian region. Total column density of NO_2 has been observed at Ahmedabad by a ground based spectroscopic technique (Chakrabarty et al., 2006) and are found to be of the order of $\sim 70 \times 10^{14}$ molecules cm^{-2} for the period 1996–2000 which are higher by a factor of 2 compared to total NO_2 columns derived from GOME Level-1 spectra during the same period (Andreas Richter, University of Bremen Scientific Product). However both data sets show a maximum during the summer season (May–June). Meena et al. (2003) have observed the total vertical column densities for NO_2 of the order of $10–100 \times 10^{14}$ molecules cm^{-2} over Pune based on visible spectroscopy, which also show a summer maxima. As tropospheric NO_2 column is a proxy for the local emissions, in this section we look at its seasonal variation and compare MOZART simulated columns with those observed by GOME.

The seasonal variation of tropospheric NO_2 column for the selected cities for the year 2000 is shown in Fig. 2, for both MOZART simulated NO_2 column (Fig. 2(b)) and that observed by the GOME instrument (Fig. 2(a)). Only for the city of Delhi, GOME observations show a prominent peak during winter (November–December–January), while a less dominant summer (May–June) peak can be seen for Ahmedabad, Bangalore and Pune. The monthly mean temperature and precipitation of the four cities for the year 2000 are shown in Tables 1 and 2 respectively (source: Indian Meteorological Department, New Delhi, India, http://www.imd.gov.in/). During winter, the temperatures are low (Table 1) leading to a decrease in loss rate of NO_2 through photolysis. Also low precipitation during winter (Table 2) makes less OH radicals available, again increasing the lifetime of NO_2. Tropospheric NO_2 column values during monsoon season (July–August) are low for all the cities.

The winter maxima are well reproduced by the model for Delhi while the low values during monsoon can be seen for Delhi, Pune and Bangalore. MOZART also shows a winter high for Pune compared to GOME observations. The wintertime maxima of NO_2 are also seen in the east of China, which is due to strong
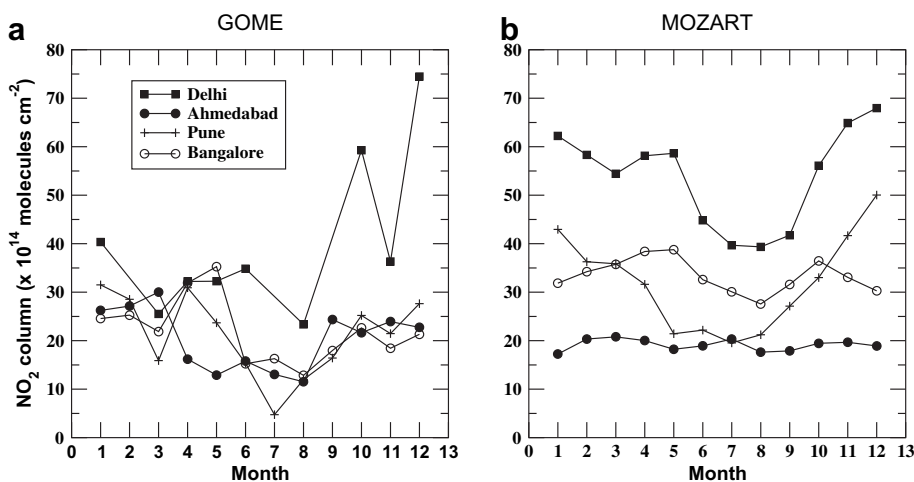

Fig. 2. The tropospheric NO_2 column for four major Indian cities. The left panel is based on GOME measurements for the year 2000. The data on the right panel has been simulated by MOZART for the year 2000.

Table 1
Observed monthly mean temperature (in °C) for the four cities of India, for the year 2000 (source: Indian Meteorological Department, New Delhi, India).

	Jan	Feb	Mar	Apr	May	Jun	Jul	Aug	Sep	Oct	Nov	Dec
Delhi	13.9	15.4	22.1	30.9	35.1	32.6	29.9	30.6	29.9	27.8	21.8	15.6
Ahmedabad	20.6	21	26.6	32.8	33.4	33.7	29.4	29.4	29.8	29.7	25.4	21.4
Pune	20.9	22.5	26.1	29.0	29.8	27.4	25.1	24.5	24.9	25.3	22.7	20.6
Bangalore	22.3	24.4	26.2	27.9	27.1	24.3	23.9	23.6	24.4	23.7	22.9	20.9

anthropogenic emissions and longer lifetime (Zhang et al., 2007). MOZART does not show a significant seasonal variation for the region of Ahmedabad. This may be due to inadequate OH radicals available in the model grid representing Ahmedabad, as the water vapour is prescribed in MOZART. Also the maxima and minima are broader in the case of model simulations compared to observations.

The model simulated tropospheric NO_2 column amounts show a small peak during summer for the cities of Delhi and Bangalore. The summer high values could be due to more natural emissions of NO_x from soil. Higher temperatures during summer (Table 1) lead to more soil emissions (Yienger and Levy, 1995). Another reason for higher column values during summer may be higher biomass burning which peaks during March–May (ATSR, World fire atlas).

The monsoon low is due to transport of cleaner air from the Arabian Sea and Indian Ocean and also due to the loss of NO_2 through reaction with OH radicals (which have higher concentration in the monsoon due to higher moisture content (Table 2)). Due to higher moisture content during monsoon (Table 2), the wet deposition rate of NO_2 may also be faster, leading to lower NO_2 column values.

Also, observations and model show that for all the months in the year 2000, Delhi has the highest total as well as tropospheric NO_2 columns. According to model simulations, Ahmedabad has the lowest tropospheric NO_2 columnar abundances.

The discrepancies between MOZART and GOME values could be due to either the uncertainties in the emission inventory taken as input in MOZART or due to errors in retrieval from the satellite data (resulting from increased aerosol loading in Indian cities). The emissions are those from the emission inventory POET (http://www.aero.jussieu.fr/projet/ACCENT/POET.php), which is in turn based on GEIA (Global Emission Inventory Activity) and EDGAR (Emissions Database for Global Atmospheric Research) data and include new developments on biogenic VOC emissions (Olivier et al., 2003). GEIA is a consortium of about 100 partners worldwide in the framework of IGBP and IGAC and version 1 provides comprehensive global emissions on a $1° \times 1°$ grid for the years 1985 and 1990. Apart from black carbon, biomass burning data are obtained from the inventory by Hao and Liu (1994). The data are aggregated in annual, seasonal or monthly averages. The recommended anthropogenic emissions are from the EDGAR data set, which is a global anthropogenic emissions inventory (Olivier et al., 2005), comprising data from 1990 to 1995 on a $1° \times 1°$ grid.

The POET data set consists of $1° \times 1°$ gridded anthropogenic, biomass burning, and natural emission data. Anthropogenic emissions are annual data, while biomass burning and natural emissions are monthly data. The total emissions for each month and the percentage contribution from natural and anthropogenic sources

are shown in Table 3 for the year 2000. The contribution of biomass burning to the total emissions is very small and hence not shown. Natural emissions for all the cities exhibit the maximum seasonal variation (Table 3). Thus, one can see that the variation in the model simulated columns is largely arising from the seasonality of the natural emission.

A Japanese group has developed a new emission inventory of anthropogenic emissions for Asia (Regional Emission inventory in ASia (REAS) Version 1.1) for the period 1980–2020 (Ohara et al., 2007). Similar to Table 3, the total emissions and the contribution from each source as estimated in the REAS inventory for the year 2000 are shown in Table 4. Here only the anthropogenic emissions have been taken from REAS, whereas emissions from the other two sources (natural and biomass burning) are the same as in the POET inventory.

For the year 2000, the total emissions of NO_2 from the POET inventory (used in MOZART) for Ahmedabad is 157.2 kilo tons, while that from REAS is 101.5 kilo tons. The corresponding figures for Delhi are 414 kilo tons (POET) and 123.5 kilo tons (REAS) respectively. Amongst the four cities, the highest estimates are for Bangalore − 556.5 kilo tons from the POET inventory and 218.9 kilo tons from the REAS inventory. In fact the effect of these high emissions on MOZART simulated tropospheric NO_2 columns can be seen in the comparison of Fig. 1(a) and (b).

Thus, the model has been run with a higher surface emission for NO_2 and correspondingly the column amount simulated is higher than the satellite measurements (Fig. 2 (a) and (b)).

3.2. Long term trends

In urban cities of both developing and developed countries, it is predominantly mobile or vehicular pollution that contributes to overall air quality problems. In Delhi, for example, close to two-thirds of the total pollution (66%) is from vehicles. Increase in vehicular population seen in major Indian cities over the last decade indicates that there should have been a corresponding increase in the NO_x emissions as well. In this section, we look at the long term trend for the monthly mean tropospheric NO_2 column observed by GOME from 1996 to 2003 and SCIAMACHY from 2003 to 2006. We fit the time series with a non-linear regression model given by (van der A et al., 2008)

$$Y = A + \frac{B}{12}t + C \, Sin\left(\frac{\pi}{6}t + \phi\right) + R \tag{3}$$

Where Y is the monthly mean NO_2 column at t months, with January 1996 taken as $t = 1$. The first two terms in the above

Table 2
Observed monthly mean precipitation (in mm) for the four cities of India, for the year 2000 (source: Indian Meteorological Department, New Delhi, India).

	Jan	Feb	Mar	Apr	May	Jun	Jul	Aug	Sep	Oct	Nov	Dec
Delhi	30.9	60.3	21.4	10	12.6	129.4	257.5	130.8	27.2	2.2	1.4	0
Ahmedabad	0	0	0	0	72.5	74.2	476.6	113.7	4.4	0	0	1.2
Pune	1.6	1.1	2.7	13.6	33.3	120.4	179	106.4	129.1	78.8	28.6	5.3
Bangalore	0	57.7	0	76.2	56.6	91.8	115.9	276.7	208.8	286.2	8.9	15.1

Table 3
POET emission inventory of NO_2, for the four cities of India, for the year 2000 taken as input to MOZART.

Month	Delhi			Ahmedabad			Pune			Bangalore		
	Total (kilo tons)	Anthro (% of total)	Soil (% of total)	Total (kilo tons)	Anthro (% of total)	Soil (% of total)	Total (kilo tons)	Anthro (% of total)	Soil (% of total)	Total (kilo tons)	Anthro (% of total)	Soil (% of total)
Jan	32.8	99.7	0.3	9.6	96.4	3.5	27.8	96.7	3.3	44.8	92.7	7.3
Feb	30.0	98.3	1.7	10.3	81.1	18.8	25.1	96.6	2.8	40.5	92.6	7.1
Mar	33.9	96.3	3.7	12.5	73.7	25.4	29.0	92.6	4.2	47.0	88.4	10.8
Apr	34.6	91.4	7.2	14.4	62.3	36.3	28.5	91.1	4.6	47.6	84.6	15.0
May	37.0	88.4	11.6	16.8	55.0	45.0	29.1	92.3	5.3	47.6	87.4	12.5
Jun	36.1	87.6	12.4	14.2	63.2	36.8	27.5	94.5	5.5	49.6	81.1	18.9
Jul	38.3	85.4	14.6	15.4	59.9	40.1	28.2	95.2	4.8	50.2	82.7	17.3
Aug	37.7	86.7	13.3	14.5	64.0	36.0	28.3	95.0	5.0	46.4	89.5	10.5
Sep	35.0	90.5	9.5	16.3	54.8	45.2	27.6	94.3	5.7	45.6	88.2	11.8
Oct	33.6	97.2	2.8	12.3	75.2	24.8	28.4	94.7	5.3	45.7	91.0	9.0
Nov	32.1	98.5	1.5	10.2	87.5	12.5	26.8	97.1	2.3	45.9	87.7	12.3
Dec	32.9	99.3	0.7	10.7	86.5	13.5	28.0	96.0	1.9	45.5	91.4	8.6
Total	414.0			157.2			334.3			556.5		

equation represent a linear trend, with B representing the annual trend in the NO_2 tropospheric column. The third term is the seasonal component for the annual cycle of NO_2, with amplitude C and a phase shift ϕ. The residual unexplained by the fit function is represented by R. The non-linear fitting is done by the least squares method. A linear trend B is taken as statistically significant with 95% confidence level if $|B/\sigma_B| > 2$, where σ_B is the error in B.

Fig. 3 shows the time series of the GOME and SCIAMACHY data for the four cities in India and an industrial region of north India. The observed trends and its error from the non-linear regression model (Equation (3)) are shown in Table 5. The annual growth rate with respect to the mean NO_2 column of 1996 is given in the last column of Tables 5 and 6.

As can be seen from Fig. 3 and Table 5, the tropospheric NO_2 column over the capital city of Delhi shows the largest significant increase from 1996 to 2006 of 11.29 ± 3.01% per annum. Such a growth rate is commensurate with the economic development and vehicular traffic increase during the same period. According to the records from Regional Transport Office (RTO), Delhi, the vehicular population in Delhi rose from 1.9 million vehicles in 1990 to nearly 3.6 million in the year 2001 (an increase of nearly 8.7% per annum). Earlier, Gurjar et al. (2004) have shown that transport emissions are responsible for most of the NO_x in Delhi. They showed that NO_x emissions have increased from 94 Gg of NO_2 in 1990 to 161 Gg in 2000. NO_x emissions for the first half of the 1990 decade, vary within the range of 11–131 Gg of NO_2 year^{-1} according to different literature sources (Gurjar et al., 2004). Even after the introduction of Compressed Natural Gas (CNG) as fuel in 2001 for

the public transport system in Delhi, NO_x concentrations in Delhi's ambient air have shown an increase (Sharma and Pundir, 2008). It has been estimated that in Delhi, the NO_x has increased from 3.5 Gg to 4.5 Gg for the period 1990–1996 from gasoline consumption alone, while it has increased from 8 Gg to 12.8 Gg from diesel consumption (Sharma, 2003). With the same GOME and SCIAMACHY time series, van der A et al. (2008) report an increasing trend of 7.4 ± 1% per year for the city of Delhi, whereas Ghude et al. (2008) estimate an increase of 2.4 ± 1.2% per year for a larger region of north India which includes Delhi.

The tropospheric NO_2 columnar abundances over Ahmedabad have clearly increased from 1996 to 2006 (Fig. 3 and Table 5). The annual growth rate of 9.97 ± 2.26% per annum is comparable to the 10% per annum growth rate in the vehicular population in Ahmedabad (Regional Transport Office, Ahmedabad). The linear trend is also statistically significant and is much higher than the growth rate (2.1 ± 1.1%) reported by Ghude et al. (2008) in a western region of India which includes Ahmedabad and Mumbai. The satellite data also show a clear increase in the tropospheric NO_2 column for Bangalore with a growth rate of 3.54 ± 1.39% per annum with respect to 1996, as the vehicular traffic has increased substantially in Bangalore also during the period from 1996 to 2006.

However the linear trend for Pune is statistically not significant ($|B/\sigma_B| = 1.5$). The 1.54 ± 1.04% per annum growth rate is the lowest amongst the four cities, though according to an estimate by the Society of Indian Automobile Manufacturers, vehicles in Pune have increased by 7% from the year 2002 to 2007. According to another study, in 2005 there were more than 1.3 million vehicles in Pune of

Table 4
REAS emission inventory of NO_2, for the four cities of India, for the year 2000.

Month	Delhi			Ahmedabad			Pune			Bangalore		
	Total (kilo tons)	Anthro (% of total)	Soil (% of total)	Total (kilo tons)	Anthro (% of total)	Soil (% of total)	Total (kilo tons)	Anthro (% of total)	Soil (% of total)	Total (kilo tons)	Anthro (% of total)	Soil (% of total)
Jan	8.1	98.8	1.2	4.9	93.0	6.9	5.4	83.3	16.7	16.2	79.8	20.2
Feb	7.7	93.5	6.5	6.0	67.7	32.1	4.9	82.8	14.2	14.6	79.6	19.7
Mar	9.3	86.4	13.6	7.8	57.8	40.8	6.7	67.6	18.3	18.3	70.3	27.6
Apr	10.7	72.3	23.3	9.8	44.6	53.2	6.9	63.2	19.0	19.8	63.0	35.9
May	12.3	65.2	34.8	12.1	37.3	62.7	6.8	66.8	23.0	18.9	68.3	31.5
Jun	12.2	63.3	36.7	9.6	45.6	54.4	5.9	74.1	25.9	21.8	57.2	42.8
Jul	13.6	58.9	41.1	10.7	42.2	57.8	5.9	77.0	23.0	21.6	59.8	40.2
Aug	13.0	61.5	38.5	9.7	46.5	53.5	5.9	76.1	23.9	17.8	72.7	27.3
Sep	11.1	69.9	30.1	11.7	37.2	62.8	6.0	73.4	26.6	17.9	69.9	30.1
Oct	9.0	89.5	10.5	7.6	59.7	40.3	6.0	74.9	25.1	17.0	75.8	24.2
Nov	8.2	94.3	5.7	5.6	77.4	22.6	5.1	85.1	11.8	18.1	68.8	31.2
Dec	8.2	97.3	2.7	6.0	75.8	24.2	5.6	79.9	9.4	16.8	76.8	23.2
Total	123.5			101.5			71.1			218.9		
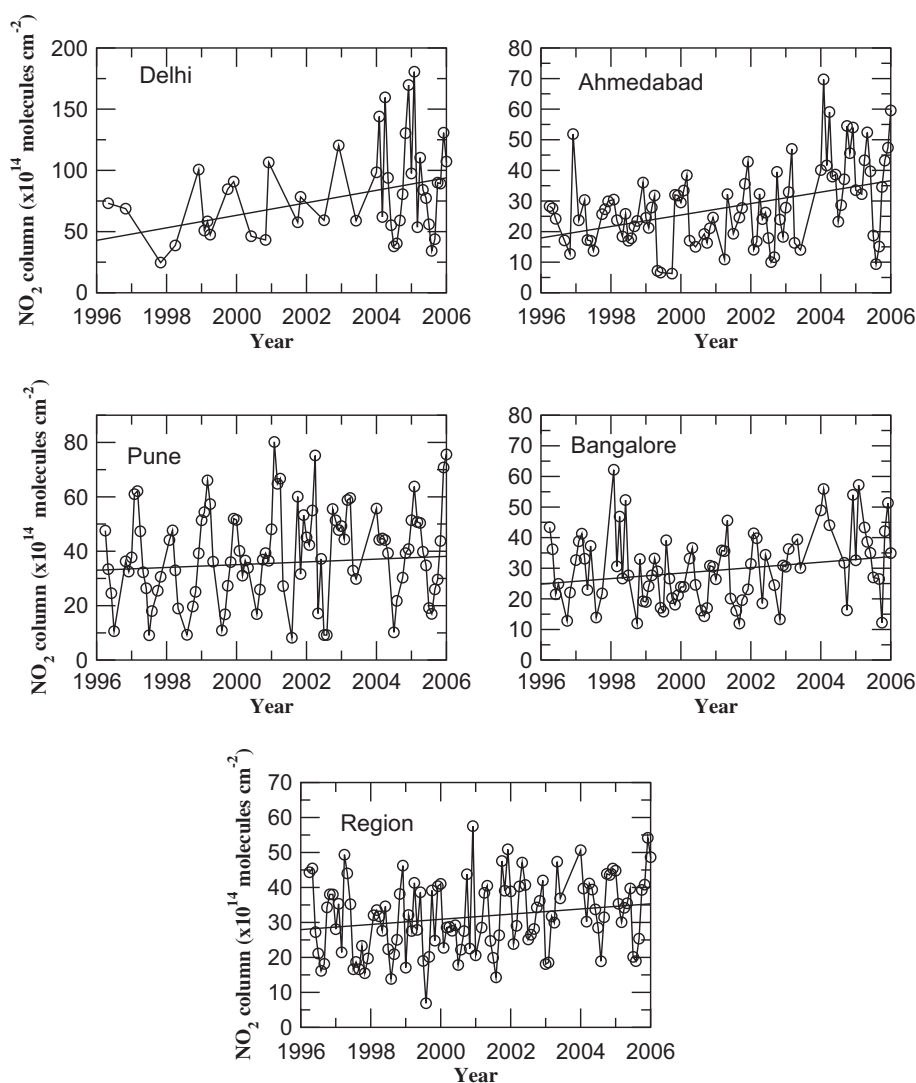
Fig. 3. The tropospheric NO_2 column for the four cities and an industrial region of India, from 1996 to 2006 as measured by GOME and SCIAMACHY. The solid line represents the linear fit as obtained by Equation (3).

which, about 1 million were two-wheelers with an addition of 10,000–13,000 new vehicles each month, which corresponds to about 10% increase per year (Safai et al., 2005, Pune Municipal Corporation).

A recent study shows that the emission from megacities is not well represented in global emission inventories (Butler et al., 2008). Three global emission inventories were used in this study, viz. EDGAR, IPCC-AR4 (Dentener et al., 2005) and RETRO (Pulles et al., 2007). Emissions of NO_x, globally and from megacities, are always higher in the EDGAR inventory compared to the other two. For example, for the city of Delhi, the EDGAR emissions are 60 Gg (of N) year^{-1}, the IPCC-AR4 emissions are 40 Gg (of N) year^{-1} and RETRO emissions are 20 Gg (of N) year^{-1}. The total emissions from

the POET inventory (used in MOZART) for Delhi is 126 Gg (of N) for the year 2000.

For the megacities of Shanghai and Beijing in China, a significant increase in NO_2 column amounts of 20% and 18% per year respectively, has been reported for the period 1997–2006 from GOME and SCIAMACHY data (Zhang et al., 2007). A significant trend of $29 \pm 5\%$ per annum has been reported by van der A et al. (2008) for Shanghai. The increase in NO_2 column in Beijing is fairly well correlated with the traffic increase during the above period (Zhang et al., 2007). In fact, the traffic restrictions implemented during the Sino-African Summit in Beijing, were remarkably successful in reducing emissions of NO_x by about 40% (Wang et al., 2007).

Table 5
Observed trends for the four cities (and a region) of India from 1996 to 2006.

City	Coordinates (latitude, longitude)	1996 mean column (10^{14} mol cm^{-2})	Linear trend in NO_2 (10^{14} mol cm^{-2} year^{-1})	Annual growth rate (% with respect to 1996)
Delhi	28.5N, 77.2E	45.19	5.1 ± 1.36	11.29 ± 3.01
Ahmedabad	23N, 72.5E	18.77	1.87 ± 0.42	9.97 ± 2.26
Bangalore	12.9N, 77.6E	25.24	0.89 ± 0.35	3.54 ± 1.39
Pune	18.5N, 73.8E	33.25	0.51 ± 0.34	1.54 ± 1.04
Region	25–30N, 76–80E	28.27	0.74 ± 0.29	2.61 ± 1.01

Table 6
The model grid cells representing the Indian cities, over which satellite data have been averaged.

City	Coordinates	Model grid cell
Ahmedabad	23°N	21.43°N–24.28°N
	72.5°E	70.31°E–73.12°E
Delhi	28.4°N	27.14°N–30°N
	77.1°E	75.94°E–78.75°E
Pune	18.5°N	15.71°N–18.57°N
	73.8°E	73.12°E–75.94°E
Bangalore	12.9°N	12.86°N–15.71°N
	77.6°E	75.94°E–78.75°E

This was estimated by comparing the observations from the Ozone Monitoring Instrument (OMI) aboard the Aura satellite and simulations with the GEOS-Chem global 3-D chemical transport model with resolution of $2° \times 2.5°$ (Wang et al., 2007). On the other hand compared to China, we do not see such a high growth rate for the Indian region, with a maximum of 11.3% per annum for Delhi.

We have also looked at the tropospheric NO_2 column abundances for a small industrial region over north India (25–30°N, 76–80°E). The data from GOME and SCIAMACHY (Fig. 3 and Table 5) show a slightly increasing trend of $2.6 \pm 1\%$ per year for this region. This can be compared with the growth rate of $2.4 \pm 1.2\%$ estimated by Ghude et al. (2008) for a geographically similar (but larger) region. However, when we compare this with an industrial region of East China studied by Richter et al. (2005), the NO_2 values over India are about a factor of 5 or more less. Also a clear increasing trend, as seen for China, is not so evident for India. This may have to do with more fossil fuel burning in China. Zhang et al. (2007) report a rapid increase in tropospheric NO_2 over the industrial region of East China with about 8.2×10^{14} molecules cm^{-2} per annum, during the period 1997–2006. They attribute this to the increasing emissions from industry and traffic. On the other hand, the industrial region of India that we have considered, the increase is only at the rate of about 0.74×10^{14} molecules cm^{-2} per annum (Table 5).

4. Conclusions

We have used GOME and SCIAMACHY data to evaluate the capability of a global chemical transport model in simulating the tropospheric NO_2 columnar abundances over a few locations of India that have witnessed increasing NO_x emissions over the past few years. Tropospheric columnar abundances of NO_2 are generally overestimated by the MOZART model compared to satellite data. This could be due to higher emission estimates in the POET inventory used in MOZART. The model also does not reproduce the general features of the observed seasonal behaviour and the quantitative comparison at the local crests and troughs is also not very good. This may be attributed to the lack of seasonal variation in anthropogenic emission estimates of the POET inventory. The discrepancies between MOZART and GOME values could be due to either the uncertainties in the emission inventory or due to errors in retrieval from the satellite data (as a result of increased aerosol loading in Indian cities).

According to Velders et al. (2001), the measured tropospheric NO_2 column densities derived from GOME data are a factor of 2–3 higher than those calculated by MOZART for Asia. In a similar comparative study, Ma et al. (2006) observed that the regional chemical transport model used for their study, generally reproduced higher tropospheric NO_2 column densities compared to those observed by GOME, in polluted areas of China. Another comparison of observed SCIAMACHY NO_2 columns with a simulation from a 3D Eulerian air quality model showed a large bias of 60% due to poor emission data used in the model (Shi et al., 2008). There

is, therefore, a need to refine the global emission inventories with respect to megacities and other regions of Asia, so that NO_2 column concentrations simulated by global chemical transport models agree more closely with satellite data.

We have also examined trends over the period 1996–2006 in tropospheric NO_2 columns over a few Indian regions where high economic growth in the present decade is likely to see increased NO_x emissions. Except for Pune, the observed NO_2 columnar abundances show a fast increase commensurate with that seen in the growth of anthropogenic activities in those regions of high economic growth (Table 5). However, even the highest growth rate of NO_2 seen in the present study, is less when compared with similar polluted regions of China, where a much more rapid increase in NO_2 column values have been observed from the GOME and SCIAMACHY data (van der A et al., 2008; Richter et al., 2005).

References

Akimoto, H., 2003. Global air quality and pollution. Science 302, 1716–1719.
Baidya, S., 2007. Trace Gas and Particulate Matter Emissions from Road Transportation in India: Quantification of Current and Future Levels, Uncertainties, and Sensitivity Analysis. Universität Stuttgart, Germany. (PhD dissertation, unpublished).
Beirle, S., Platt, U., Wenig, M., Wagner, T., 2003. Weekly cycle of NO_2 by GOME measurements: a signature of anthropogenic sources. Atmospheric Chemistry and Physics 3, 2225–2232.
Boersma, K.F., Eskes, H.J., Brinksma, E.J., 2004. Error analysis for tropospheric NO_2 retrieval from space. Journal of Geophysical Research 109, D04311. doi:10.1029/2003JD003962.
Brasseur, G.P., Hauglustaine, D.A., Walters, S., Rasch, P.J., Müller, J.-F., Granier, C., Tie, X.X., 1998. MOZART, a global chemical transport model for ozone and related chemical tracers, 1, model description. Journal of Geophysical Research 103, 28265–28289.
Butler, T.M., Lawrence, M.G., Gurjar, B.R., van Aardenne, J., Schultz, M., Lelieveld, J., 2008. The representation of emissions from megacities in global emission inventories. Atmospheric Environment 42, 703–719.
Chakrabarty, D.K., Patel, Shailesh, Vala, Prakash, 2006. Seasonal and solar activity variation of NO_2 over Ahmedabad. Indian Journal of Radio & Space Physics 35, 344–348.
Dalvi, M., Beig, G., Patil, U., Kaginalkar, A., Sharma, C., Mitra, A.P., 2006. A GIS based methodology for gridding large scale emission inventories: application to carbon-monoxide emissions over Indian region. Atmospheric Environment 40, 2995–3007. doi:10.1016/j.atmosenv.2006.01.013.
Dentener, F., Stevenson, D., Cofala, J., Mechler, R., Amann, M., Bergamaschi, P., Raes, F., Derwent, R., 2005. The impact of air pollutant and methane emission controls on tropospheric ozone and radiative forcing: CTM calculations for the period 1990–2030. Atmospheric Chemistry and Physics, 1731–1755.
Ghude, Sachin D., Fadnavis, S., Beig, G., Polade, S.D., van der A, R.J., 2008. Detection of surface emission hot spots, trends, and seasonal cycle from satellite-retrieved NO_2 over India. Journal of Geophysical Research 113, D20305.
Giorgi, F., Chameides, W.L., 1985. The rainout parameterization in a photochemical model. Journal of Geophysical Research 90, 7872–7880.
Gurjar, B.R., van Aardenne, J.A., Lelieveld, J., Mohan, M., 2004. Emission estimates and trends (1990–2000) for megacity Delhi and implications. Atmospheric Environment 38, 5663–5681.
Hack, J.J., 1994. Parameterization of moist convection in the NCAR community climate model (CCM2). Journal of Geophysical Research 99, 5551–5568.
Hao, W., Liu, M.H., 1994. Spatial and temporal distribution of tropical biomass burning. Global Biogeochemical Cycle 8, 495–504.
Holtslag, A., Boville, B., 1993. Local versus nonlocal boundary-layer diffusion in a global climate model. Journal of Climate 6, 1825–1842.

Horowitz, L., et al., 2003. A global simulation of tropospheric ozone and related tracers: description and evaluation of MOZART, version 2. Journal of Geophysical Research 108, 4784. doi:10.1029/2002JD002853.

Irie, H., 2005. Evaluation of long term tropospheric NO$_2$ data obtained by GOME over East Asia in 1996–2002. Geophysical Research Letters 32, L11810.

Kunhikrishnan, T., Lawrence, Mark G., von Kuhlmann, Rolf, Richter, Andreas, Ladstätter-Weißenmayer, Annette, Burrows, John P., 2004. Analysis of tropospheric NO$_x$ over Asia using the model of atmospheric transport and chemistry (MATCH-MPIC) and GOME-satellite observations. Atmospheric Environment 38, 581–596.

Leue, C., Wenig, M., Wagner, T., Platt, U., Jähne, B., 2001. Quantitative analysis of NO$_x$ emissions from GOME satellite image sequences. Journal of Geophysical Research 106, 5493–5505.

Lin, S.-J., Rood, R.B., 1996. Multidimensional flux-form semi-Lagrangian transport schemes. Monthly Weather Review 124, 2046–2070.

Lohani, B.N., 2006. Energy Efficiency and Climate Change Considerations for On-road Transport in Asia (Report). Asian Development Bank (ADB).

Ma, Jianzhong, Richter, Andreas, Burrows, John P., Nüß, Hendrik, van Aardenne, John A., 2006. Comparison of model-simulated tropospheric NO$_2$ over China with GOME-satellite data. Atmospheric Environment 40, 593–604.

Meena, G.S., Jadhav, D.B., Bhosale, C.S., 2003. Total column density variations of NO$_2$ and O$_3$ by automatic visible spectroscopy over Pune, India. Current Science (India) 85, 171.

Martin, R.V., et al., 2002. An improved retrieval of tropospheric nitrogen dioxide from GOME. Journal of Geophysical Research 107, 4437–4456.

Ohara, T., Akimoto, H., Kurokawa, J., Horii, N., Yamaji, K., Yan, X., Hayasaka, T., 2007. An Asian emission inventory of anthropogenic emission sources for the period 1980–2020. Atmospheric Chemistry and Physics 7, 4419–4444.

Olivier, J.G.J., Bouwman, A.F., Van der Hoek, K.W., Berdowski, J.J.M., 1998. Global air emission inventories for anthropogenic sources of NO$_x$, NH$_3$ and N$_2$O in 1990. Environmental Pollution 102 (Issue 1, Suppl. 1), 135–148.

Olivier, J.G.J., et al., 2003. Present and Future Surface Emissions of Atmospheric Compounds. POET Rep 2, EU Prof. EVK2-1999-00011. European Union, Brussels.

Olivier, J.G.J., van Aardenne, J.A., Dentener, F., Ganzeveld, L., Peters, J.A.H.W., 2005. Recent trends in greenhouse gas emissions: regional trends and spatial distribution of key sources in 2000. Environmental Sciences 2.

Pickering, K.E., Wang, Y., Tao, W.-K., Price, C., Mueller, J.F., 1998. Vertical distributions of lightning NO$_x$ for use in regional and global chemical transport models. Journal of Geophysical Research 103, 31203–31216.

Price, C., Penner, J., Parther, M., 1997a. NO$_x$ from lightning. Part 1: Global distribution based on lightning physics. Journal of Geophysical Research 102, 5929–5941.

Price, C., Penner, J., Prather, M., 1997b. NO$_x$ from lightning. Part 2: Constraints from the global atmospheric electric circuit. Journal of Geophysical Research 102, 5943–5951.

Pulles, T., van het Bolscher, M., Brand, R., Visschedijk, A., 2007. Assessment of Global Emissions from Fuel Combustion in the Final Decades of the 20th Century, Application of the Emission Inventory Model TEAM. Technical Report A-R0132B. Netherlands Organization for Applied Research (TNO), Apeldoorn, The Netherlands.

Rao, P.S., Pandit, V.I., Devotta, S. Emerging fuel specification – Indian Scenario, Energy Summit 2004 – Oil and Gas Meet, 30 October 2004, Chennai, India, Organized by Confederation of Indian Industries (CII), New Delhi, 2004.

Richter, A., Burrows, J.P., 2002. Retrieval of tropospheric NO$_2$ from GOME measurements. Advances in Space Research 29, 1673–1683.

Richter, Andreas, Burrows, John P., Nüß, Hendrik, Granier, Claire, Niemeier, Ulrike, 2005. Increase in tropospheric nitrogen dioxide over China observed from space. Nature 437, 129–132.

Safai, P.D., Rao, P.S.P., Momin, G.A., Ali, Kaushar, Chate, D.M., Praveen, P.S., Devara, P.C.S., 2005. Variation in the chemistry of aerosols in two different winter seasons at Pune and Sinhagad, India. Aerosol and Air Quality Research 5 (1), 115–126.

Sharma, C., Pundir, R., 2008. Inventory of green house gases and other pollutants from the transport sector: Delhi. Iranian Journal of Environmental Health Science and Engineering 5 (2), 117–124.

Sharma, C., 2003. Urban Policy Integration of Energy Related Environmental Issues in Selected Asian Mega-cities: Final Activity Report Submitted to APN, Feb 2003, Institute for Global Environmental Strategies.

Shi, Chune, Fernando, H.J.S., Wang, Zifia, An, Xingqin, Wu, Qizhong, 2008. Tropospheric NO$_2$ columns over East Central China: comparisons between SCIAMACHY measurements and nested CMAQ simulations. Atmospheric Environment 42, 7165–7173.

van der A, R.J., Eskes, H.J., Boersma, K.F., van Noije, T.P.C., Van Roozendael, M., De Smedt, I., . Peters, D.H.M.U., Meijer, E.W., 2008. Trends, seasonal variability and dominant NO$_x$ source derived from a ten year record of NO$_2$ measured from space. Journal of Geophysical Research 113, D04302. doi:10.1029/2007JD009021.

Velders, G.J.M., Granier, C., Portmann, R.W., Pfeilsticker, K., Wenig, M., Wagner, T., Platt, U., Richter, A., Burrows, J.P., 2001. Global tropospheric NO$_2$ column distributions: comparing three-dimensional model calculations with GOME measurements. Journal of Geophysical Research 106, 12643–12660.

Wang, Y., McElroy, M.B., Boersma, K.F., et al., 2007. Traffic restrictions associated with the Sino-African summit: reductions of NO$_x$ detected from space. Geophysical Research Letters 34, L08814. doi:10.1029/2007GL029326.

Yienger, J.J., Levy II, H., 1995. Empirical model of global soil biogenic NO$_x$ emissions. Journal of Geophysical Research 100, 11447–11464.

Zhang, X., Zhang, P., Zhang, Y., Li, X., Qiu, H., 2007. The trend, seasonal cycle, and sources of tropospheric NO$_2$ over China during 1997–2006 based on satellite measurement. Science in China Series D: Earth Sciences 50, 1877–1884.

Zhang, G.J., McFarlane, N.A., 1995. Sensitivity of climate simulations to the parameterization of cumulus convection in the Canadian Climate Centre general circulation model. Atmosphere-Ocean 33, 407–446.