Multi-year satellite observations of tropospheric NO₂ concentrations over the Indian region

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An assessment of satellite-derived long-term tropospheric nitrogen dioxide (NO₂) data is performed over the Indian region and their implications on the regional air quality are discussed. The Indo-Gangetic plain (IGP) shows an increasing trend in NO₂ of the order of 3×10^{13} mol/cm²/yr. The pixel-wise (0.25 km) trend for the period 2005–2014 reveals various regions having increased rates of pollution over the study period. Further, the mean seasonal concentrations of NO₂ are segregated for different parts of the country including oceanic regions and the trends are brought out. The highest rate of increase of tropospheric NO₂ (2×10^{14} mol/cm²/yr) is seen around coal mining areas and certain industrial areas such as ports and thermal power stations. Using the data spanning 10 years, the wavelet analysis is carried out to study the influence of semi-annual oscillations (SAO) on trace gas concentrations in different parts of the country. The study reveals that the SAO are stronger in the northern parts of India, including IGP and western India, whereas South India and oceanic regions are having very low SAO component and strong annual oscillation component.

Keywords. Tropospheric NO₂; satellite concentrations; long-term trend; AO; SAO.

1. Introduction

The ability to measure air pollution and other chemically reactive trace gases in the lower atmosphere from satellites has a heritage dating back nearly three decades when the first measurements of carbon monoxide were made from the space shuttle Challenger in November 1981 (Reichle *et al.* 1986). Continuous measurements of atmospheric trace gases are now available, which can be utilised to monitor long-term changes in concentrations. Of late, satellite remote sensing has helped to address the challenges associated with air-quality monitoring to a great extent although there are some

uncertainties associated with the measurements. With the launch of a series of satellites ERS-2, ENVISAT, AURA and METOP since 1995, information on the global distribution of total and tropospheric nitrogen dioxide (NO₂) columns has become available from the payloads named Global Ozone Monitoring Experiment (GOME), Scanning Imaging Absorption spectrometer for Atmospheric CHartographY (SCIAMACHY), Ozone Monitoring Instrument (OMI) and GOME-2, respectively (e.g., Bovensmann *et al.* 1999; Burrows *et al.* 1999; Levelt *et al.* 2006; Valks *et al.* 2011). The retrievals of the NO₂ columns from these ultraviolet–visible measurements are based

on the technique of differential optical absorption spectroscopy (Platt 1994; Platt and Stutz 2008; Wagner *et al.* 2008).

NO₂ is an important trace gas in the troposphere. It has a significant impact on O₃ as well as on the strongest atmospheric oxidising agent, OH. It also acts as a gaseous precursor of aerosols and acid rain and contributes to radiative forcing of climate (Seinfeld and Pandis 1998; Solomon *et al.* 1999). Since the atmospheric pollutant NO₂ has a relatively low lifetime in the atmosphere, the anthropogenic emissions of the compound are concentrated, or, higher, at the source region of emissions and dissipate as they get transported away from the source. Natural sources of NO₂ in the atmosphere are soil emissions and lightning. NO₂ concentrations are also known to be affected by planetary phenomena such as the El Nino Southern Oscillation (ENSO). Reduction in tropospheric column observations of NO₂ was seen to coincide with ENSO and resulting suppression of convection and lightning.

The major anthropogenic sources of NO₂ are industrial and vehicular emissions, soil emissions (natural) and emissions from biomass burning. Through remote sensing, it is now possible to identify the areas with high emissions by monitoring the tropospheric NO₂ concentrations. NO₂ is known to cause respiratory problems for humans and has been listed as one of the primary pollutants with national ambient air-quality standards in many countries. Increasing traffic, power generation and industrial activities, especially in megacities (>10 million population), are immense sources of nitrogen oxides (NO_x ≡ NO + NO₂) and other pollutants, degrading air quality and adversely affecting human health (Molina and Molina 2004).

Earlier studies on satellite concentrations around coal mining areas using limited database showed positive correlations with thermal power plant output and NO₂ concentrations (Prasad *et al.* 2012). The short-term linear trends were carried out for years 2005–2008 which showed an increase over error levels in Indo-Gangetic Plain (IGP). Another study undertaken using data up to 2009 showed positive correlations of NO₂ with O₃ in certain cities, mainly in the IGP (Streets *et al.* 2013).

The present investigation utilises the level-3 data product of tropospheric NO₂ over the Indian region from 2005 to 2014 and reveals interesting results on the evolution of hotspots, quantification concentrations over different geographical zones over

the country and also the general trends on air quality. The study has been conducted to quantify the increasing/decreasing trends of troposphere column NO₂ and to see how the species vary with latitude and season over the Indian subcontinent and surrounding oceanic environments. In addition, the long-term trend in troposphere NO₂ for each pixel of 25 km resolution over the Indian subcontinent is quantified using 10 years of data. Further, the influence of semi-annual oscillations (SAO) on trace gas concentrations is also investigated.

2. Data and methodology

Total column of tropospheric NO₂ concentrations is obtained from the Ozone Monitoring Instrument (OMI) on-board the Aura satellite (Schoeberl *et al.* 2004). Daytime global NO₂ data sets are produced daily using OMI aboard polar-orbiting, sun-synchronous Earth Observing System Aura satellite with the equator crossing time of 13:45 LT. OMI makes nadir measurements of the Earth's backscattered radiation at a spectral range of 264–504 nm (Streets *et al.* 2013). The attenuation is utilised to obtain the slant column densities which are converted to vertical column densities using appropriate air mass factors to obtain the tropospheric column concentrations of NO₂.

The retrieval errors are reported to be up to 30% in NO₂ in high emission areas (Streets *et al.* 2013). The retrieval of tropospheric column NO₂ takes into account the clear and cloudy conditions in the air mass factor derived for the simulated NO₂ profiles (Bucsela *et al.* 2006). Tropospheric NO₂ has an uncertainty of 0.1×10^{15} mol/cm² and is underestimated by 15–30% (Celarier *et al.* 2008).

3. Observations

Figure 1 depicts the mean seasonal variations in NO₂ concentrations spanning 2005–2014 as obtained from the satellite. The four seasons are: March–April–May (MAM) comprising the pre-monsoon, June–July–August–September (JJAS) which is the monsoon months, October–November (ON), the post-monsoon season and the winter comprising of December–January–February (DJF) months. The highest concentrations of NO₂ are seen over the thermal power stations, and the regions of Singrauli and Talcher, in particular, have

Figure 1. Mean tropospheric NO_2 concentrations over the Indian region from 2005 to 2014.

the highest levels of NO_2 concentrations. Also, Delhi city figures among cities with increased emissions with values above $4.5 \times 10^{15} \text{ mol/cm}^2$. Overall, it can be seen that the central Arabian Sea and the central Bay of Bengal have low values of concentrations around $5 \times 10^{14} \text{ mol/cm}^2$. Over mainland, the least emissions are seen over the northeast as well as the densely forest covered areas of Chhattisgarh state. Emissions are also low over desert regions of Rajasthan. Rainfall has a significant diminishing effect on the NO_2 concentrations and values are less over southern Indian coastal regions near Karnataka. NO_2 lifetime and spatial extent in the atmosphere is determined by a variety of meteorological factors including rainfall, wind direction and the planetary boundary layer height which vary from season to season. It is important to look at the changes in concentrations owing to seasonal changes. Least concentrations of NO_2 are during the JJAS months owing to the overall reduction during monsoon through direct wet deposition as well as reduced soil emissions due to lower temperatures. It is seen that, overall, the highest concentration over the mainland

is observed during the MAM months followed by DJF. NO_2 concentration over the northeast part of India is high during MAM, the season where biomass burning significantly contributes to the overall emissions. Concentration over Rajasthan is high during JJAS and low during rest of the year. This may be due to the differences in soil temperatures contributing to the emissions of NO_2 , since there is no other known source in this region. Continental outflow from the mainland into the surrounding oceanic regions is evident; higher values are seen around eastern Arabian Sea except during the monsoon season. There is a significant increase seen over northern and parts of eastern Bay of Bengal during MAM and DJF, and with lesser spatial extent during ON.

The NO_2 concentrations over the Indian region have been quantified next, by obtaining the mean of the following zones shown in figure 2.

The zones over mainland have been chosen to be representative of the different levels of NO_2 spatial extent. The entire mainland is divided into six zones; northeast (NE) having relatively less emissions and predominantly affected by biomass

Figure 2. Selection of zones across the country where NO_2 concentrations are studied.

burning, the highly industrialised zone extending over northern India along the IGP, parts of the central India (CI) where the influences of increase in spatial extent of pollutants from IGP can be inferred, western India (WI) where the few source regions are located owing to the presence of the Thar desert and further, southern India (SI) is classified into two regions, SI1 and SI2, based on the levels of urbanisation and the presence of high source of emissions such as thermal power plants.

Figure 3 shows the yearly mean concentrations of different regions over the Indian mainland for

different seasons. The IGP has the highest values of mean NO_2 concentrations across all seasons. A slightly increasing trend in emissions is seen from 2005 to 2010 after which there is a tapering of the rate of increase for ON, DJF while there is a negative trend after 2011 during pre-monsoon season. But the fact remains that there is around 25% increase in concentrations along the IGP compared to CI. This is a significant rise when compared to the differences in concentrations between other regions and shows the urgent need to tackle the growing NO_2 emissions with improved industrial practices. The next zone having high concentration is the CI, adjoining the IGP. A consistently increasing trend in CI concentrations is seen during the ON as well as DJF season, an increasing trend is also seen in MAM, although not clear during JJAS months. This indicates a possible spread in the spatial extent of pollution from IGP as well as the increased levels of urbanisation along these areas. More detailed studies are warranted as to how the release of pollutants and their regional spatial extent can be curbed and controlled.

WI has the least value of NO_2 concentrations over the entire country during the pre-monsoon. The NE region is having the least values of NO_2 concentrations during all seasons except MAM. Increased levels of biomass burning during this season may be a factor contributing to the observations of increased levels of NO_2 . There have been no significant changes in the trends over the

Figure 3. Mean yearly variations of NO_2 concentrations over different geographic locations over the Indian mainland.

observation period for NE. Coming to southern India, SI1 has highest concentrations during MAM and least values during ON and JJAS. SI2 has lower levels of NO_2 pollution compared to SI1 owing to the absence of any significant sources of NO_2 emissions other than relatively less urbanised cities. No particular trends in this region have been identified from the data.

Figure 4 shows the yearly mean variations of NO_2 concentrations over oceanic regions. The regions have been identified based on the proximity to the mainland and also deep-sea regions. NO_2 concentrations over oceans are low owing to lack of strong emission sources. The main component of NO_2 presence is from the continental outflow and certain amount of emissions from ship exhaust. The emissions from ship exhaust are limited to a very narrow lane along southern Bay of Bengal along the international shipping lanes. The concentrations from ship emissions are not tabulated in the present work. Among oceanic regions, the highest level of concentrations is recorded along the NBoB. This is the region where there is a significant outflow of pollutants from the IGP as well as NE, particularly during MAM. There are some studies that also link the influence of mineral dust on NO_2 concentrations, although the contribution may be small (Wang *et al.* 2003). During MAM and JJAS, concentrations over WBoB and NBoB are high. During ON months, highest values are observed over

North Arabian Sea (NAS). The wind directions during the different seasons play an important role in transporting the NO_2 away from the source regions. Since there are no significant sources of NO_2 over oceans, the prevailing meteorological conditions have to be studied in order to effectively monitor trace gas constituents over oceanic regions.

The value of the slope of the fit and the corresponding standard error in fitting the lines are given in table 1. The value of slope and intercept is obtained by combining the data set for each region for all the four seasons (figure 5).

The trend analysis has allowed us to identify and monitor the areas where high rates of increase in concentrations are found. It is seen that, in general, the increase in NO_2 is only marginal in most of the subcontinents. The average rate of increase is around $3.5 \times 10^{13} \text{ mol/cm}^2/\text{yr}$. However, some of the regions around the coal mining areas in Chhattisgarh, Orissa, show a significant rate of increase, of the order of $2 \times 10^{14} \text{ mol/cm}^2/\text{yr}$. Also, the port in Jamnagar and the thermal power plants located near Bellary, Karnataka, are also showing increased rates. Upon close observation, the shipping lanes starting from the gulf towards Sri Lanka and the shipping route from Sri Lanka to East Asia show a slightly declining trend, although the value is very small, of the order of $-1.5 \times 10^{13} \text{ mol/cm}^2/\text{yr}$.

Figure 4. Mean yearly variations of NO_2 concentrations over different geographic locations over oceanic regions.

Table 1. The slope, intercept and R values for the linear trends in troposphere NO_2 concentrations obtained for different parts of the country.

Region		Value	Std. error	R value
WI	Intercept	$-2.97\text{E}+16$	$1.02\text{E}+16$	0.44033
	Slope	$1.54\text{E}+13$	$5.09\text{E}+12$	
IGP	Intercept	$-7.94\text{E}+16$	$2.59\text{E}+16$	0.45582
	Slope	$4.06\text{E}+13$	$1.29\text{E}+13$	
CI	Intercept	$-6.54\text{E}+16$	$1.77\text{E}+16$	0.52552
	Slope	$3.35\text{E}+13$	$8.79\text{E}+12$	
NE	Intercept	$-2.44\text{E}+15$	$3.96\text{E}+16$	0.0149
	Slope	$1.81\text{E}+12$	$1.97\text{E}+13$	
SI1	Intercept	$-3.48\text{E}+16$	$2.95\text{E}+16$	0.1964
	Slope	$1.81\text{E}+13$	$1.47\text{E}+13$	
SI2	Intercept	$-3.03\text{E}+16$	$1.53\text{E}+16$	0.31734
	Slope	$1.57\text{E}+13$	$7.63\text{E}+12$	
NAS	Intercept	$-3.87\text{E}+16$	$1.91\text{E}+16$	0.32005
	Slope	$1.98\text{E}+13$	$9.49\text{E}+12$	
CAS	Intercept	$-2.81\text{E}+15$	$7.61\text{E}+15$	0.0686
	Slope	$1.60\text{E}+12$	$3.78\text{E}+12$	
EAS	Intercept	$-1.23\text{E}+16$	$2.47\text{E}+16$	0.08614
	Slope	$6.55\text{E}+12$	$1.23\text{E}+13$	
WBoB	Intercept	$-2.25\text{E}+16$	$1.60\text{E}+16$	0.23209
	Slope	$1.17\text{E}+13$	$7.97\text{E}+12$	
NBoB	Intercept	$-2.62\text{E}+16$	$3.16\text{E}+16$	0.13929
	Slope	$1.36\text{E}+13$	$1.57\text{E}+13$	
EBoB	Intercept	$-1.70\text{E}+15$	$1.47\text{E}+16$	0.02596
	Slope	$1.17\text{E}+12$	$7.31\text{E}+12$	
CBoB	Intercept	$-3.02\text{E}+15$	$6.47\text{E}+15$	0.08661
	Slope	$1.73\text{E}+12$	$3.22\text{E}+12$	

4. Frequency distribution of NO_2 concentrations

A frequency distribution of the number of days, which are having varying levels of NO_2 concentrations over the year, reveals important information apart from the general yearly trends of absolute values. The OMI data is consistently available over the region of observation throughout the year. Using this data, it is possible to look at the progression of days into levels of higher concentrations. The frequency distribution is performed for all the regions earmarked earlier. Figure 6 shows the range of concentrations as coloured bars. The y -axis shows the percentage of days in a year where the range of concentrations is indicated by the colour bar. A slightly different colour bar is used for oceans where the concentrations are much below that on land.

Over IGP, the number of days above $4 \times 10^{15} \text{ mol/cm}^2$ has drastically increased from 2007 to 2010 and has been remaining more or less constant at 10% till 2014. A steady increasing trend of increased percentage of days with values greater than $2 \times 10^{15} \text{ mol/cm}^2$ is seen over CI. This is indicative of the fact that the air quality of the region is getting affected by the increased level of the emissions from the neighbouring IGP. For SI, there is an increasing trend of days

Figure 5. Slope of linear trends obtained for NO_2 for each pixel over the study region.

Figure 6. Frequency distribution of NO_2 concentrations for different parts of the country.

Figure 7. (a) Wavelet periodograms of trace gas concentrations over IGP and NAS. (b) Wavelet amplitudes of AO and SAO for different regions.

progressing towards 2×10^{15} mol/cm² from 2005 to 2011 and a negative trend ensued. The latest figures for 2014 indicate that there is a reduction of 10% of the number of days above 2×10^{15} mol/cm². Both in NAS and NBoB, there is a clear indication of the progression in a number of days in a year with recorded values of NO₂ above 1.2×10^{15} mol/cm².

The availability of long-term data facilitates the use of wavelet analysis technique to delineate periodicities present in the data set. Here the data for different regions over the span of 10 yr is subject to wavelet analysis after smoothing, to remove the harmonics of the data length. Figure 7(a) shows the wavelet periodograms of IGP and NAS. The *x*-axis shows the number of days since 1 January 2005. The *y*-axis shows the periodicity (in days). The band around 365 is the yearly oscillation and the band around 180 days is the semi-annual oscillation. It can be seen that while the NAS is having strong annual oscillation component only (*y*-axis indicates periodicity), there is a distinct signal pertaining to SAO at a periodicity centred around 180 days also, for IGP. Similar study is conducted for all regions and the periodicities corresponding to AO and SAO are extracted. The regions showing significant SAO strengths are WI, IGP and CI. The results are tabulated in figure 7(b). It is to be noted that while interpreting the magnitude of SAO/AO, there is no normalisation done

for all the regions. The strength of SAO can only be inferred with respect to the strength of AO for the same region. The regions IGP, CI and WI are showing relatively higher magnitudes of SAO compared to respective AO component. This study reveals the potential influence of SAO prevalent in middle latitudes on the tropospheric trace gas concentrations.

5. Conclusion

A comprehensive multi-year analysis of tropospheric NO₂ observations over the Indian region is undertaken using the OMI satellite data product. The investigations using a large database of 10 yr have helped to track and monitor the emissions from major sources of NO₂ emissions spread across the country. Areas with persistent and increasing trends during the period of observations have been identified and the mean yearly values have been obtained. IGP shows the highest level of concentrations while SI shows the least concentrations throughout the year. Influence of local emissions during biomass burning season is clearly seen in NE (MAM) and Punjab/Haryana (ON). Influence of continental outflow is seen in concentrations in some seasons, owing to prevailing metrological conditions. The long-term trends are estimated for the concentrations at each pixel using 10 years of data. The high rate of increase in NO₂ concentrations is identified around coal mines and some other sites such as ports and thermal power plants. The rate of change of NO₂ is seen to vary from 9.5×10^{13} mol/cm²/yr in certain areas in IGP to 1×10^{12} mol/cm²/yr over ocean. Using the wavelet technique, the SAO in trace gas concentrations are estimated, and it is found that the signatures of SAO are stronger in northern India. The impact of alternate sources of emissions such as biomass burning as well as meteorological impact over the NO₂ concentrations has to be investigated in order to fully delineate the observed differences in NO₂ concentrations and forms part of the future work.

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