Continuation of Satellite Observations of NO_2 over South Asia using Ozone Monitoring Instrument during the time period of 2004-2016

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(2019)

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A thesis submitted in partial fulfillment of the requirements for the degree of

Master of Science in Environmental Science

By

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THESIS ACCEPTANCE CERTIFICATE

It is certified that the contents and form of the thesis entitled "Continuation of Satellite Observations of NO_2 over South Asia using Ozone Monitoring Instrument during the time period of 2004-2016" submitted by **Ms. Maryam Sarfraz (Reg # 00000118899)** has been found satisfactory for the requirements of the degree of Master of Science in Environmental Science.

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I dedicate this thesis to my Mother for her unconditional support, Love & encouragement

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List of Abbreviations

NO2	Nitrogen dioxide
OMI	Ozone Monitoring Instrument
TEMIS	Tropospheric Emission Monitoring Internet Service
OH	Hydroxyl
ORP	Oil Refining Process
AS	Agriculture Soil
RT	Road Transport
NRT	Non-Road Transport
IP	Industrial Process
EI	Energy Industry
AWB	Agriculture Waste Burning
BE	Biogenic Emission
CM	Combustion in Manufacturing
WHO	World Health Organization
GOME 2	Global Ozone Monitoring Experiment-2
VCDs	Vertical Column Densities
SCD	Slant Column Density

Abstract

NO_2 is one often criterion air pollutant considered as an indicator for air quality index. More the concentrations, more the toxic environment for living beings. Different sources impart in NO_2 atmospheric concentrations, for instance biofuel consumption, soils emissions, aircrafts and fossil fuels burning. Elevated concentrations of NO_2 are fallen in serious category for health risk assessment. South Asia is one of the most affected regions as its economic potential is engrossing interests from all over the world. In this study, TEMIS NO_2 data sets were used for the time period of 2004-2016. NO_2 from Satellite Observations is retrieved using a DOAS-base algorithm. Spatio temporal analysis and multi-year mean maps clearly depicts different concentrations in different seasons and regions. For instance, relatively large column densities have been reported over hyper arid regions during Monsoon time spans due to augmented bacterial activities in their soils. In addition to it, lightening is also prime source for enhanced atmospheric NO_2 levels the emissions. Temporal mean maps have indicated enhanced column densities in India, Pakistan and Bangladesh region. On the contrary, Bhutan and Nepal are contributing relatively in smaller amounts. This propense pertains in Industrial emissions and transport sector. Additionally, recent economic boom in south Asian countries is major driver behind enhanced atmospheric NO_2 levels across the region.

Key Words: *OMI Observations, Emission inventories, Tropospheric NO_2, South Asian Pollution, NO_2 Seasonal Trends*

CHAPTER 1

INTRODUCTION

1.1 South Asia

South Asian region comprises of eight countries named as Pakistan, India, Bangladesh, Sri Lanka, Afghanistan, Bhutan, Maldives and Nepal. It holds the area of approximately 4.5 million sq. km. The region hosts more than one fifth of the global population. Geographical features are diverse as it holds Lofty Mountains of Himalayas, Hindu kush and Karakoram, river systems with canals and tributaries, verdurous plains, mighty deserts, lush green meadows, and approximately coastline of 10,000 km long. Winter and summer spells of monsoon winds are key drivers for sustaining tropical monsoon climate of the region. These spells bring rain which feed the rivers and aquifers mainly accommodating human needs and agricultural activities.

South Asian region has been prone to air pollution from the last two decades. Every passing year urbanization, especially metropolis areas, and demand for socioeconomic growth has triggered continuous pressure on resource consumption. More the populace, more the demands, and eventually more the resources retrenchment. Population shift has also been observed as one of the key factor that contributes towards enhanced NO_2 emissions in the region.

1.2 Back Ground

Activities going on in the biosphere are severely affecting the composition of both lower and upper atmosphere. Emission sources may include motor vehicles, industries, agriculture, power generators and other anthropogenic activities. Resultant emissions make their way to the atmosphere and the permissible limits are either touched or has been frequently exceeded during the previous 2 decades. Urban and indoor air quality indexes are declared as World's toxic polluted region by "Blacksmith Institute world's worst polluted places report (2008)".

For the assessment of air pollution and its control in South Asia, it is most important to trace out the root causes of its sources and removal pathways. South Asian Countries (Pakistan, Bangladesh, Afghanistan, Bhutan, Nepal, India and Sri Lanka,) are still ranked as developing economies. This region is number one on world population and is a home for half of the world' underprivileged masses (World Bank, 2019). Agriculture is the main occupation and almost 60% of the population is earning its livelihood through different agricultural sectors

1.3 Air Pollution

Air pollution affects living beings on 24/7 cycle as we breathe air every day. During breathing, we encounter polluted gases, and consequences are health effects. Most of the air pollutants are poisonous and their daily encounter with living beings results in lung diseases, respiratory problems, eye irritation, nasal infections, skin redness, chronic obstructive pulmonary disease, emphysema and muscle fatigue (Bender, 2014). Urban areas are hub of air pollution sources for instance heavy traffic on the cities roads, industrial zones in the metropolitan areas. According to Global Livability Index, south Asian cities are losing their safe livability positions every year. For instance Kathmandu felled from 127th position to 129th in 2018. Same down ranking happened with Dhaka which dropped down from 137th to 139th (koh, 2018).

Main characteristics of NO_2 can be summarized as following,

- NO_2 is part of gaseous air pollutants produced as a result of fossil fuel combustion processes and road traffic.

- According to IPCS 2007 report, it has high oxidizing capacity and corrosive in nature (IPCS: International Program on Chemical Safety).
- It is one of the precursor of Ozone formation and plays its role in Acid rain.
- Nitrogen dioxide has significant impacts on ecosystem and human health as it is one of the contributor for the formation of photochemical smog and acid rain.

1.4 Socio Economic Scenario of the Area

In Figure 1.4 (a) per capita GDP of south Asian countries have been depicted. Sri Lanka is on the top with the GDP of 3835.39 USD per capita. While the Afghanistan ranked last with the GDP of 561.78 USD per capita for the year of 2016. Figure 1.4 (b) exhibits the population of South Asian countries in millions. Indian is most populace following by Pakistan, Nepal, Sri-Lanka, Bangladesh, Afghanistan and Bhutan.

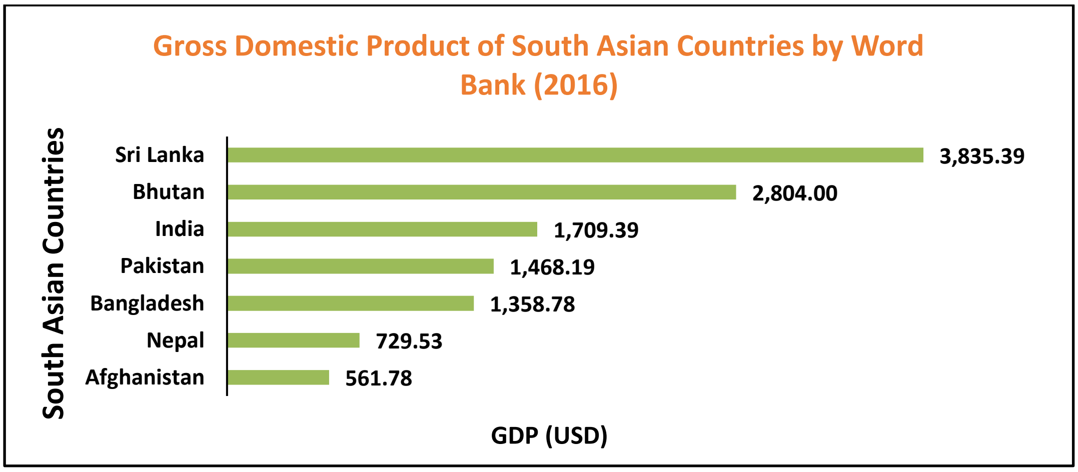


Figure 1.4.1: GDP of South Asian countries (World Bank)

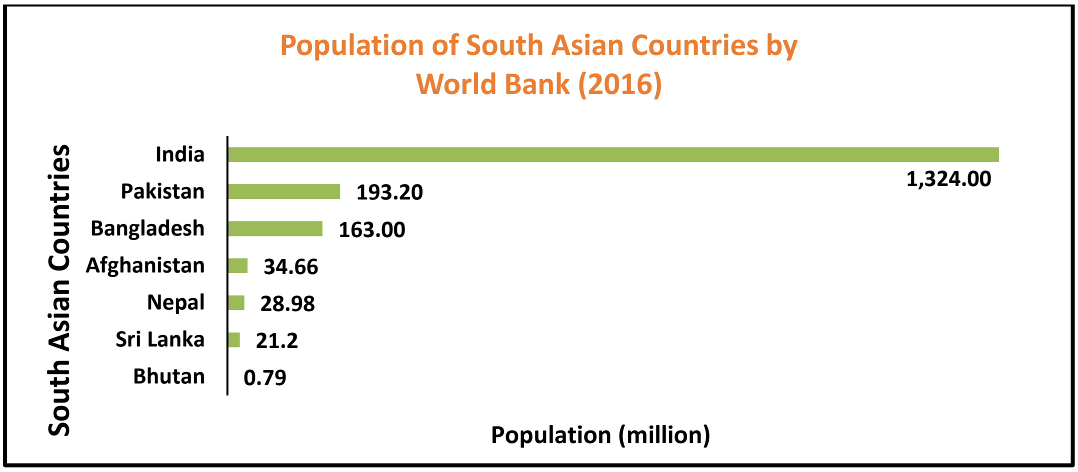
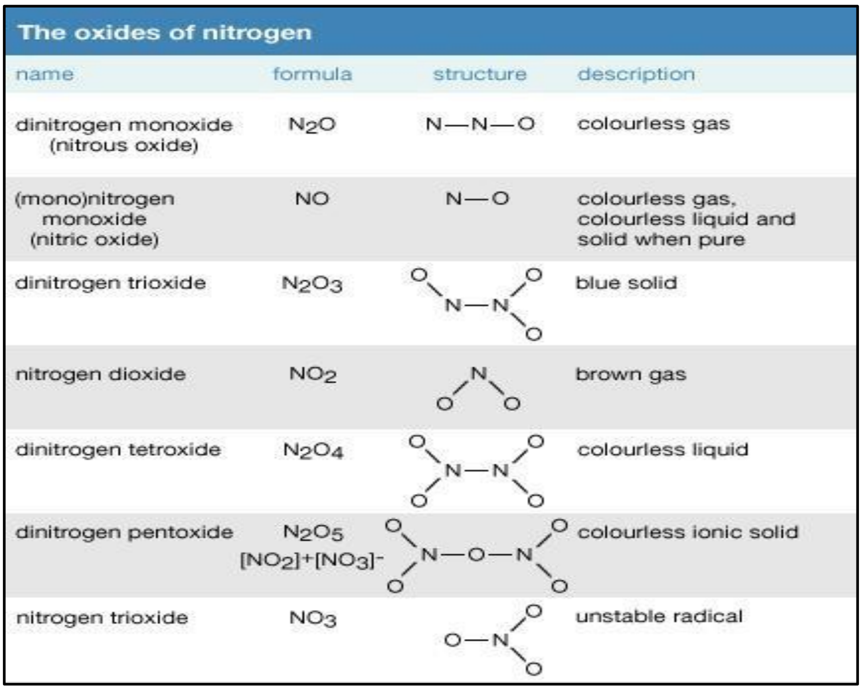


Figure 1.4.2: Population of South Asian countries (World Bank)

1.5 Introduction to Nitrogen Oxides

There are approximately 7 oxides of nitrogen (NO, NO_2, NO_3, N_2O, N_2O_3, N_2O_4, N_2O_5) prevailing time to time depending on the temperature zones in the atmosphere. Scientific communities and Regulatory authorities are mostly concerned about the presence of NO_2 due to its persistent nature. Rest of the oxides are made temporarily in the atmosphere under specific conditions (Mijling, 2012).

NO_2 is gas with reddish brown color, bitter odour (O'Neil, 2006; IPCS, 1997) and has pungent smell, at temperatures below 21.15ºC, It is brown liquid and exists as a colorless solid below -11ºC (Lewis ; 2002). It has high oxidizing capacity and is corrosive (IPCS, 1997); also noncombustible (Lewis, 2002).



The oxides of nitrogen

name	formula	structure	description
dinitrogen monoxide (nitrous oxide)	N_2O	N—N—O	colourless gas
(mono)nitrogen monoxide (nitric oxide)	NO	N—O	colourless gas, colourless liquid and solid when pure
dinitrogen trioxide	N_2O_3		blue solid
nitrogen dioxide	NO_2		brown gas
dinitrogen tetroxide	N_2O_4		colourless liquid
dinitrogen pentoxide	N_2O_5 $[NO_2]^+[NO_3]^-$		colourless ionic solid
nitrogen trioxide	NO_3		unstable radical

Figure 1.5: Oxides of Nitrogen (Source: www.npi.gov.au)

1.6 Air Quality in South Asia

- 30 top most polluted cities in the world with the poorest air quality in 2016 and 2017 are located in South Asia. In Asia, Air pollution was ranked sixth most dangerous killer in a Global burden of disease assessment by World Bank .Noxious gases and matter cause climate anomalies and subsequently global warming. Air pollution/particle haze results in minimum 10-15% reduction in the amount of sunlight reaching the earth surface (DeWitt *et al.*, 2009). Particles change the winter and monsoon pattern significantly reducing the rainfall over Afghanistan, Pakistan and other north western Asia.
- South Asian mega cities are notorious for poor air quality. According to Unicef report, about 70% of infants living in south Asia are at the risk of brain diseases.

1.7 Physical and Chemical properties

No.	Property	Value	Reference
1	**Molecular weight**	46.01 $gmol^{-1}$	Lide, 2007: O'Neil, 2006
2	**Physical State**	Clear colorless volatile liquid	
3	**Melting Point**	-9.3°C	
4	**Boiling Point**	21.15° C	
5	**Density (Liquid)**	1.448 (at 20°C)	O'Neil, 2006
6	**Density (Gas)**	1.58	
7	**Vapor Pressure**	58.66 kPa (at 10°C) 121 kPa (at 25°C)	RSC, 2007 HSDB, 2005
8	**Solubility in Water**	Reacts with Water, decomposes forming Nitric acid and Nitric oxide	Lide, 2007: O'Neil, 2006
9	**Solubility**	Soluble in concentrated sulphuric acid, nitric acid, carbon disulphide, chloroform	RCS, 2007 : Lewis, 2000
10	**Conversion factors for vapours (at 25°C and 101.3 kPa)**	1mg.m^{-3} = 0.532ppm 1ppm= 1.88mg.m^{-3}	HSDB, 2005

1.8 NOx Sources

75% of the total global emissions are from anthropogenic sources. Most of the Human induced NOx emission occurs in combustion processes. On Northern Hemisphere, the fossil fuel combustion supplies the majority of the NOx emissions. 70% of the global anthropogenic NOx emissions are from fossil fuel combustion (Olivier *et al.*, 1998).

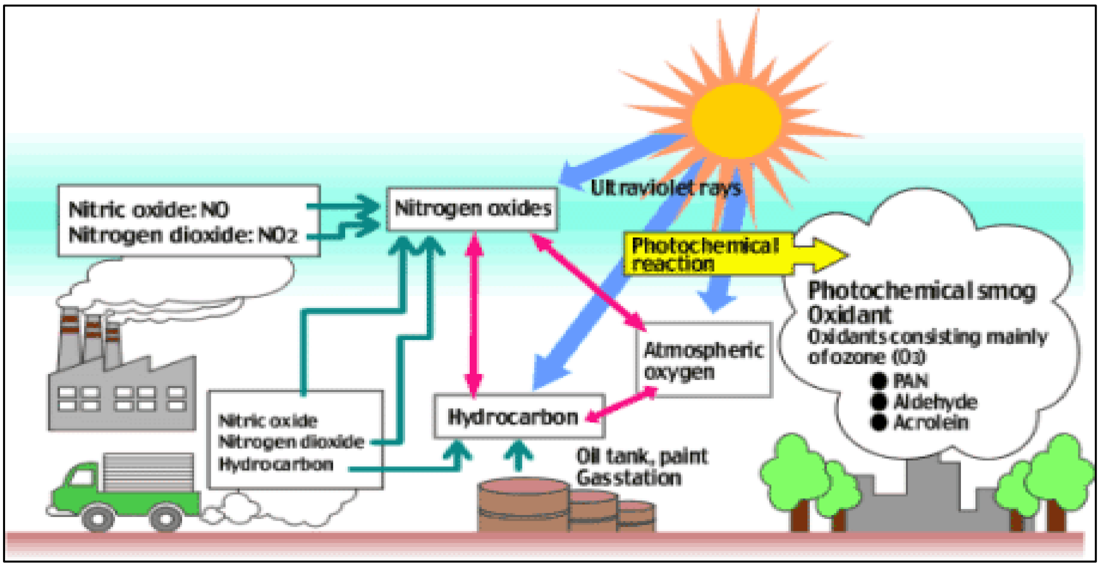


Figure 1.8: NOx Sources

1.8.1 Natural Sources

Soil emissions (oxidation of nitrogenous compounds present in the soil by different microorganisms), forest fires and lightning discharges (HSDB, 2005; Lee *et al.*, 1997). Lightning and soil emissions account 10% and 15% of the total NO_2 emissions respectively (Lee *et al.*, 1997).

1.8.2 Anthropogenic Sources

Combustion of fuels (oil, coal and natural gas) in vehicles or industrial processes (HSDB, 2007). Vehicles and industries are predicted to produce about 50% of NO_2, while biomass burning contributes about 20% (Lee *et al.*, 1997). These sources mostly emit NOx in the form of NO with less than 10% NO_2 (IPCS, 1997).

1.9 Sinks of Nitrogen Dioxide

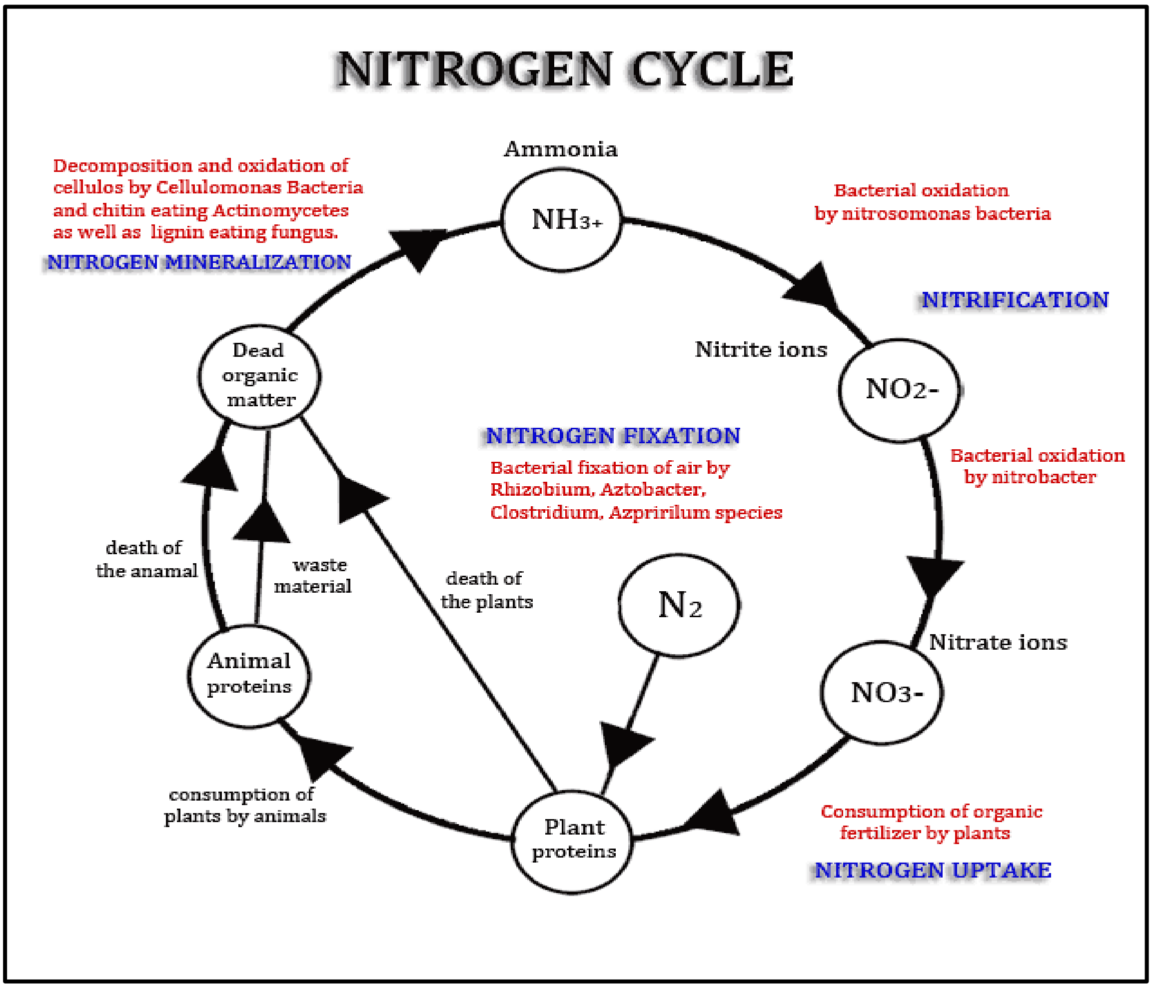


Figure 1.9: Nitrogen Cycle in Nature (www.yourarticlelibrary.com)

1.10 Standards

- According to WHO Guidelines for annual mean limits is 40µg/m^3 whereas for 1-hour mean limit is 200 µg/m^3.
- The Pakistan Environmental Protection Agency has drafted the NEQS for Ambient Air.

Pollutant	**Time Weighted Average**	**Concentration in Ambient Air**		**Method of Measurement**
		In 2009	In 2012	
NO_2	Annual Average	40µg/m^3	40µg/m^3	Gas-phase Chemiluminescence
	24 hour	80µg/m^3	80µg/m^3	

1.11 NOX Chemistry

NOx emission sources are both natural and anthropogenic. Natural Sources includes lightning and biogenic emissions from soils, whereas anthropogenic encompasses transportation, Oil refineries, energy sector, fossil fuel combustion both from house hold usage and industrial zones, and most importantly biomass burning. With every passing decade, human capital demand is increasing many folds (Oiamo *et al.*, 2015). Although, sustainable technologies are making their in the market but still resource consumption has doubled up. NOx plays significant role in tropospheric chemistry. We see different but almost set trends in four categorized seasons (Pre-monsoon, Monsoon, Post-Monsoon and winter). Land cover change and human activities are thriving sources for the chemical cycle of NO_X. Increased tropospheric NO_X returns to land and marine water through the processes of wet and dry depositions. Ecosystems are primarily dependent on the concentration of Nitrogen. Plants (Nitrogenous) need nitrogen for their productivity and growth. Sharp change in NO_X concentration can influence the marine and terrestrial ecosystems services. Acid rain and dry deposition are boons for the soil facing nitrogen deficiencies. But the situation can be alarming when exceeding nitrogen content get mixed with run off, thus results in eutrophication.

Rapid cycling between NO and NO_2

$NO + O_3 \rightarrow NO_2 + O_2$

$NO_2 + h\nu\ (+O_2) \rightarrow NO + O$

Sink by formation of HNO_3

Daytime Reactions:

$NO_2 + OH + M \rightarrow HNO_3 + M$

Nighttime Reactions:

$NO_2 + O_3 \rightarrow NO_3 + O_2$

$NO_3 + NO_2 + M \rightarrow N_2O_5 + M$

$N_2O_5 + \text{aerosol}\ (+H_2O) \rightarrow 2HNO_3$

CHAPTER 2

LITERATURE REVIEW

2.1. Background Concentration

Modelling studies have been using to analysis the back-ground concentration level and predicting the future trend of trace gases. For instance, in Ottawa, Ontario, Canada LUR (Land Use Regression) modelling is used as an effective method to figure out the fine scale measurements of pollutants in the ambient environment. A study was conducted for low urban populated areas to estimate pollutants in the overall environment. The objective of the study was to provide exposure assessments and health effects on low level industrial zones. Targeted pollutants were NO_2, VOCs benzene, m- and p- xylene. Samplers were places at 50 locations throughout the Ottawa for two 14 days. LUR models predicted 96% spatial variability in NO_2 and almost 80% change in VOCs. Pollution pools were expected on all point and line sources. Final calculations of the model exhibited significant differences on proximity to green spaces, highways, residential and industrial hubs. All pollutants were reported high on two sources Industries and road networks (Oiamo *et al.*, 2015).

2.2. Contribution to Atmosphere

NO_2 contributing major role in the atmospheric environment and chemistry. On one hand, NOx emissions are an indirect cause of climate change. Besides, it also dole out the reduction of methane lifetime so enhancing negative radiative forcing. These are the reasons that make it need of the hour to investigate the emission sources. All over the world, studies are being conducted such as ground base surveillance, Satellite observations and aircraft measurements for the approximate identification and estimation of NO_2. Modelling parameters are also used for assessment but all these methods have their constraints (Geddes *et al.,* 2012).

2.3. Developed Countries Scenarios

Developed countries from Europe and USA are actively using satellites to inspect the NO_2 concentrations and then comparing them with the real time emissions. A study conducted in china for the time period of 2005-2010 also inferred the highest concentrations in the most populated cities (Beijing & Shanghai) in the whole region. Region was divided into 6 geographical zones and the division was based on socio-economic levels. Ozone Monitoring Instrument measurement was used for annual concentrations estimations (Levelt *at al.,* 2006). In the study period, the year 2008 exhibited exception as that was Beijing Olympic Period and pollution reduction strategies of South Korea and Japan triggered down the average concentrations of NO_2 over time. The seasonal trend indicated apparent increase in winter season and no specified change in summer. OMI instrument measurements are often validated with ground based measurements e.g., Toronto (r=0.86) & California (r=0.93) showed significant correlation (Kim *et al.,* 2015).

2.4. Satellite Data Sets

Satellite data helps in the overall estimation of average concentrations over a large area whereas the ground based assessments are site-specific. Measuring instrument are placed at points where maximum emissions are expected to be high and it does not calculate background concentrations for comparison. Furthermore, data for validation is relatively short and, therefore, it makes difficult to analyze data statistically. But on the other hand, satellite (OMI) estimations with ground based observations are generally in good correlation (Grajales *et al.,* 2014).

2.5. Role of NO_2 in Atmosphere

NO_2 plays crucial role in both tropospheric and stratospheric chemistry. In the stratosphere, it is involved in the destruction of ozone (O_3). Whereas in troposphere, its role is reverted back to the formation of ozone as major precursor in the chemical reactions. Both ozone and NO_2 are toxic for biota in the atmosphere zone. Human health can be prone to risk, for instance longer exposure to ozone and NO_2 is concorded with deteriorated lung function and other respiratory disorders. Tracking down the pollutants from both the primary sources and secondary sources can help to figure out the relationship between natural and anthropogenic influences (Sauvage;2007).

2.6. Air Quality Index

Air Quality conditions may be different in different parts of one city. A study conducted on hourly basis for the time period of 1 year (May 2014- April 2015) in Beijing. National Ambient Air Quality Standards set in 2012 imposed hourly air quality monitoring and real time concentrations of 6 pollutants. It was made compulsory to publicize the index (AQI). It was found that northern and southern Beijing have different concentrations of pollutants of concern. Reasons for that change was the wind velocity and wind direction. As far the temporal trends of the city were concerned, early morning and late night were most polluted time zones when compared with late afternoon time with least recorded concentrations. One exception was recorded for Ozone that showed maximum concentration during afternoon time in the city. Seasonal trends depicted more pollutants in winters as compared to summers (Chen *et al.*, 2015).

2.7. Sources Contribution

Individual source contribution such as isolated power plants can be used as test methods to figure out emissions accurately. Quantitative analysis of sources contribution helps the observers and policy makers to categorize the root causes and their potential implications. Comparison with regional numerical models with analysis of column retrieval have shown many constraints. For example, Kalman filter paper methodwasused by Mijling (2012) to estimate emissions over China. In addition to it, Schaap and his coworkers in 2013 used a chemical transport model to calculate changes in emissions over Europe. Chemical life time of species has also been studied with the help of space borne instrument. An interesting result was reported about life span of NOx specie in the urban areas of china. Summer showed 3 hours difference while winters season exhibited up to 13 hours difference. So regional model combined with OMI data can be approximate best fit to estimate NO_X emissions and chemical decay in the area (de Foy;2015).

2.8. MAX-DOAS Studies

A study was conducted in Mexico for the time period of 2006-2011. The objective was to compare the DOAS (Differential Optical Absorption Spectroscopy) measurements with space borne Ozone Monitoring Instrument (OMI). DOAS results showed maximum measurements and were approximately three times larger than OMI derived averages over observation site. This discrepancy is mainly due to higher horizontal inhomogeneity of lower troposphere NO_2 Columns. Another major reason for this variation is the sensitivity of the instrument. OMI has reduced sensitiveness near the surface which can be approved hot spots by real time measurements by Ground based measurements. OMI data sets were used to generate NO_2 distribution maps including three main areas expecting higher concentrations. These areas include Metropolitan area of Mexico City, Industrial zone of Tula and Cuernavaca valley in the south. Seasonality of the NO_2 over three zones was also checked and analysis depicted higher NO_2 during cold and dry seasons and relatively lower concentration was detected (Rivera;2013).

2.9. **Comparison of Different Instruments**

Apart from OMI, other space borne instruments, SCIAMACHY and GOME have been used for the measurements of NOx emissions. A study carried out in China for the time period of 10 years (1995-2004) exhibited unceasing increase over east central china. Although both satellites and ground inventories pointed out the increase, however ground based emission inventory was undermined by Satellite measurements. On an average, inventories result by all sector represented 61% increase over the region while Satellites observed 95% rise for the same time period. As far the seasonality in the area was concerned, summertime trends were consistent all over the years. Whereas, winter season showed up with little discrepancies. Most probable reasons for this divergence are meteorological conditions, injection height of NOx and elevated levels of sulfate aerosols (Zhang;2007).

2.10. Model Studies

Chemistry Transport Models (CMTs) have been used to analyze the global distribution of NO_X (NO_2+NO) because of the limitations. Air-borne instruments measurements were spatially limited and temporary (Zhang *et al.*, 2007). Similarly ground based or real time measurements are spatially inclusive. Satellite measurements have overcome the issue as the instrument carrying by satellite can measure trace gases with global coverage and with adjustable temporal resolutions for a longer time period. GOME (Global Ozone Monitoring Experiment) has been recording measurements both in UV and visible Spectral ranges, so it was quite copacetic for the trace gases column retrievals including NO_2 (Toenges *et al.*, 2006).

CHAPTER 3

Methodology

3.1. Area of Study

South Asian (25.0376° N, 76.4563° E) region is gaining attention for increasing urbanization and consequently pollution every passing year. Countries are under developing and 70% of the poor population lies in the region. Region has diverse climate zones and physical ranges due to which the region depicts noticeable changes from one zone to another.

1) The high Himalayan and Karakoram mountains in the north;
2) Balochistan Plateau, the driest area of the study region that covers the Suleman and Kirthar mountains in southern boundary of Afghanistan and Pakistan;
3) the southern lowlands (Indus-Ganges-Brahmaputra) that expand from Pakistan to the delta lands of Bangladesh making up the core and densely populated areas;
4) the peninsular India, dominated by the Deccan and fertile coastal plains receded by north-south mountain ranges called western Ghats and eastern Ghats;
5) and the island realm that includes Sri Lanka and Maldives

3.2. Objectives

The objectives of this study are;

1) Source identification of nitrogen dioxide in South Asia
2) Seasonal cycles analysis and their prominent effects

3) To observe the spatio-temporal trend of nitrogen dioxide in South Asia from 2004-2016

3.3. Software and Tools

- Notepad++ was used to filter out the error values in the Data
- Envi 5.0 was used for the formation of raster images of data
- Arc map version 10.3.1 was used for the maps formation
- R-studio helped to extract the values over all hotspots
- And finally, MS Excel was used for time series and graphical representation of data.

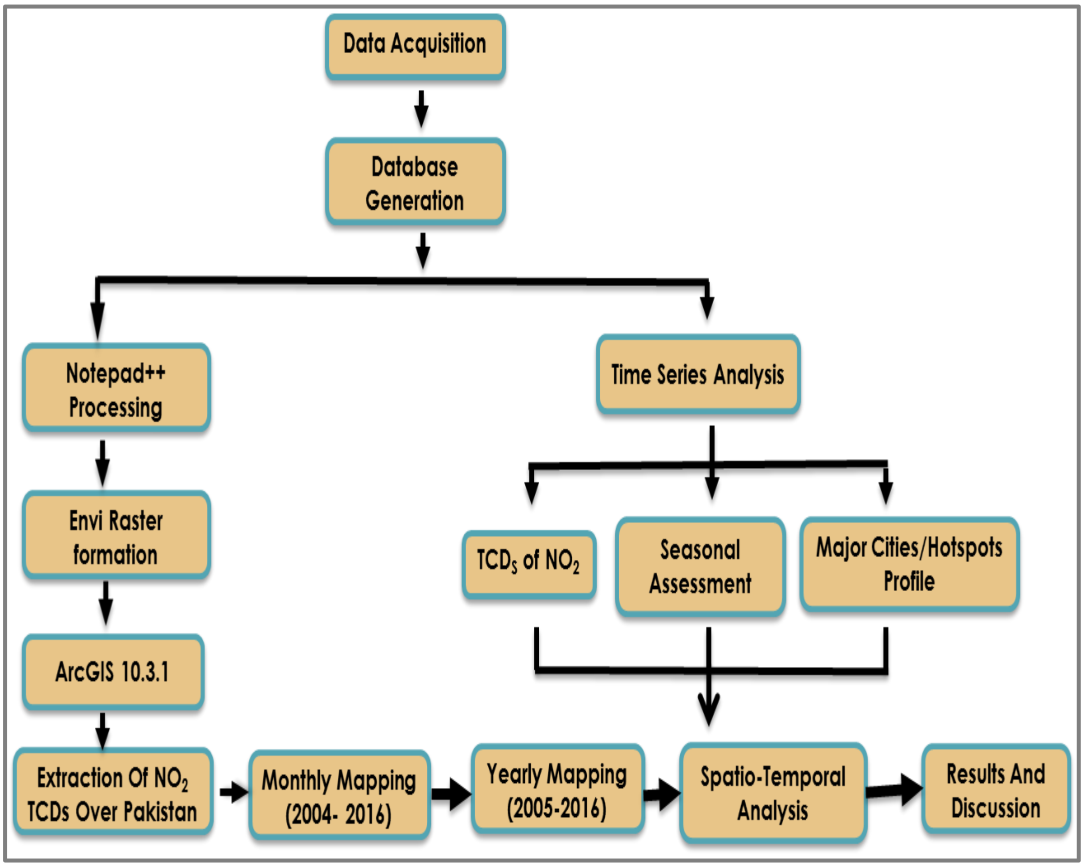


Figure 3.3: Methodology Layout of Data Processing

3.4. Data Processing

Monthly data sets for the time period of October 2004-December 2016 was downloaded into ASCII Grid format. Text files were converted into raster images in ENVI 5.0. These raster images were georeferenced and WGS 1984 spatial reference was assigned in ArcGis 10.3.1. A georeferenced shapefile of all countries of south Asian region was added to extract regional NO_2 data from the global one. Monthly and yearly averages of NO_2 over all countries were extracted. At the end spatial maps of NO_2 were created to have an idea about the major hotspot or polluted areas. Seasonality trends and time series were generated over all hotspots in order to show that how the NO_2 column densities differ over time.

3.5. Main features of OMI are summarized in Table

Instruments	**OMI**
Stand	Aura
Estimation Period	2004-till now
Crossing Equator at	1:45 pm
Spatial decree (km^2)	24×13
Spectral decree (nm)	0.5
Spectral Region	UV-Vis
Global reporting	1 day
Grid Size	0.25×0.25

Table 3.5: Main components and Features of OMI

3.6. No_2 Column Densities Retrieval From Omi/Aura Observations

The NASA's Aura satellite team carries OMI, HIRDLS, TES and MLS. The satellite retrievals are usually in good arrangement with bottom-up emission inventories and in situ evaluations. Boersma *et al.*, 2004 found a good association between OMI-NO_2 datasets and aircraft based NO_2. OMINO_2 Level-2 data product is used to observe the temporal evolution of tropospheric NO_2 over Pakistan. The data set was acquired from the "Tropospheric Emission Monitoring Internet Service" called TEMIS.

3.7. Ozone Monitoring Instrument

A spectrograph using nadir view estimates direct as well as backscattered sunlight in the UV-Vis region from 270 – 500 (nm). The data set is available from October 2004 till now. It has a high spatial resolution of 13 × 24 km^2 (wide swath of measurement) with global coverage of one day. It has spectral range of 0.5 nm (Levelt *et al.*, 2006). OMINO_2 level 2 data is based upon a better correction for across-track stripes and air mass factors (AMFs) obtained from calibration errors in the OMI backscattered reflectance. The previous studies indicated good data quality but also high biases upto 40%, because of faults and errors in the air mass factor calculations. DOAS method, was used to estimate tropospheric slant column densities (SCDs) by spectral fit to back scatter solar irradiance (Platt, 1994; Boersma *et al.*, 2005; Bucsela *et al.*, 2008) and by integrating slant columns into Chemistry transport Model (CTM) with wind fields and stratospheric chemistry. Tropospheric AMF was applied to convert slant column into vertical column. Surface albedo, cloud fraction, geometry and trace absorbers are the major constraints that affect the retrieval and contribute error (Grajales *et al.*, 2014).

3.8. Emission Inventories Data Layout

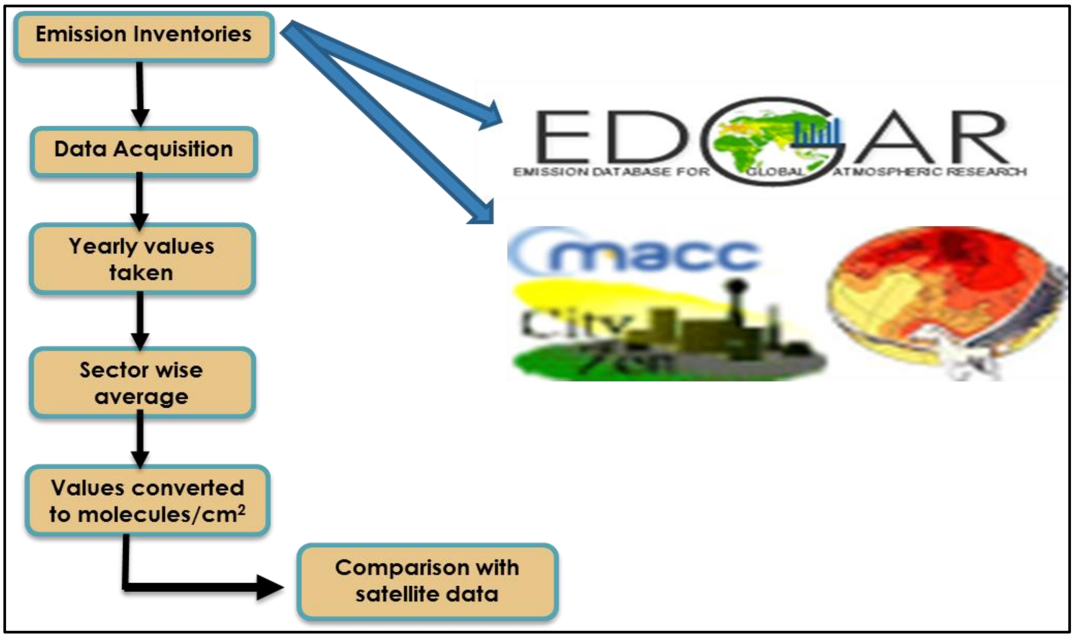


Figure 3.8: Methodology for inventories data sets

3.9. Emission Inventory

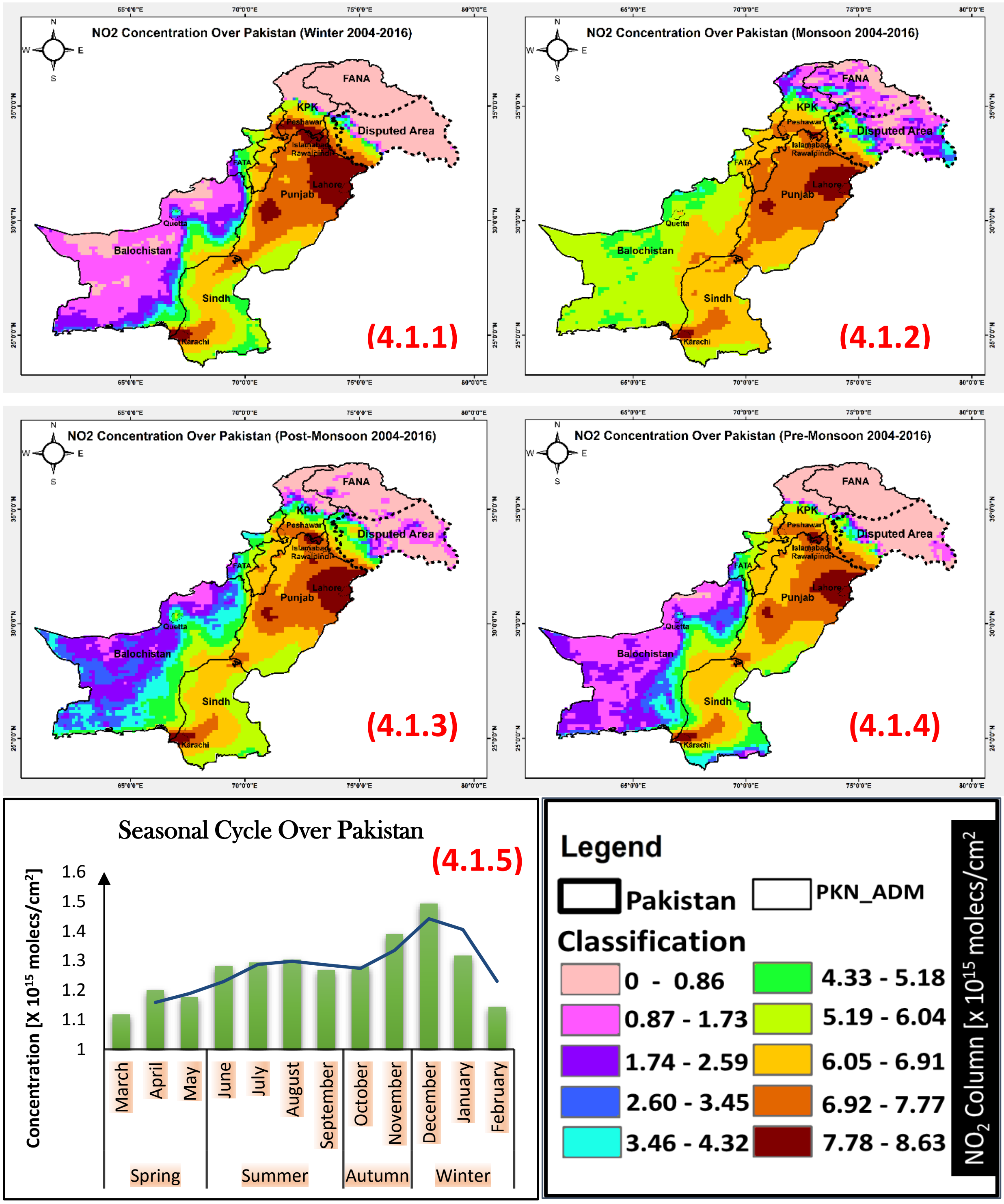
EDGARv4.2 (Emission Database for Global Atmospheric Research) emissions inventory has been used to estimate the trace gases concentration from different sectors. The main objective of this inventory is to keep an account of economic bustle for all countries in the world. The next step is the transformation of that data to emissions by using regional emission factors for each gas individually. Next to it includes mapping of these emissions to geographical grid of the resolution of 1 ×1 degree by using data maps of gridded activity. Sectors wise emissions data was categorized differently according to sources. For instance, for fuel combustion studies, excluding power plants and industries, population density maps were used. Whereas for steel and cement, point source maps were used globally. The uncertainty of global source strength of NOX emission is estimated to about 50% with 95% of confidence interval. The reason for this ambivalence is the iffiness of road transport and extensive biomass burning emissions.

CHAPTER 4

RESULTS and DISCUSSIONS

In this study, we analyzed the spatio temporal variability of OMI (Ozone Monitoring Instrument) observations. Monthly averaged tropospheric column densities of NO_2 over whole south Asian region for 12 years (2004-2016) were used in this study. The results have shown distinct spatio temporal trends for the study period. In addition, bottom up emission inventories (EDGARv4.2 & MACCity) were used to compare with satellite recorded tropospheric concentrations. These inventories manifest record from NOx contributing sources and after applying global and regional models, sources are segregated. Comparison of the satellite driven values with the emission data, we can say that the inventories are highly underestimating the OMI observed values (Leue *et al.*, 2001). The most probable reason for this underrating could be incapability to gather data from all contributing sources on regular basis. Though inventories data sets are playing their role to estimate real time emissions and their contemporary sources, still much needed to comprehend. Seasonal (Winter, Summer, Autumn & Spring) and temporal (Oct 2004 – Dec 2016) trends over all countries of south asia were studied and discussed in detail in this study. All South Asian region (Afghanistan, India, Nepal, Bhutan, Pakistan, Bangladesh and Sri Lanka) were mapped to analyze the seasonal and overall variation in the countries. With respect to NO_2 emissions, India is on the top followed by Bangladesh, Pakistan, Afghanistan, Sri Lanka, Nepal and Bhutan respectively. In this study, we assessed concentrations of NO_2 not only over the countries but cities too. In India alone, 21 cities were identified as NO_2 hotspots. Moreover, 11 cities in Pakistan, 4 cities in Afghanistan, 3 zones in Bangladesh, 4 cities in Nepal and 3 cities in Sri Lanka were identified as NO_2 hotspots. No evident hotspot was identified over Bhutan. Source apportionment for the NO_2 concentrations were also calculated by using inventories data sets. In the region, both natural and anthropogenic sources have contributed the enhanced NO_2 emissions. But anthropogenic sources uphold larger part in this as economic activities are increasing every passing day to fulfill needs and accomplish development goals in the region. For NO_2 particularly, Oil refining, road traffic and industrial processes are main sources. While energy industry, combustion process in factories, non-road traffic and agriculture waste burning also trigger the NO_2 concentrations in urban hubs and their surrounding areas (Chong *et al.*, 2016).

4.1. Seasonal and Temporal Trends over Pakistan



Figures (4.1.1, 4.1.2, 4.1.3, 4.1.4 & 4.1.5): Show NO_2 Concentration maps of Average Winter (4.1.1), Average Summer (4.1.2), Average Autumn (4.1.3), Average Spring (4.1.4), and Seasonal Cycle (4.1.5) over Pakistan from 2004-2016

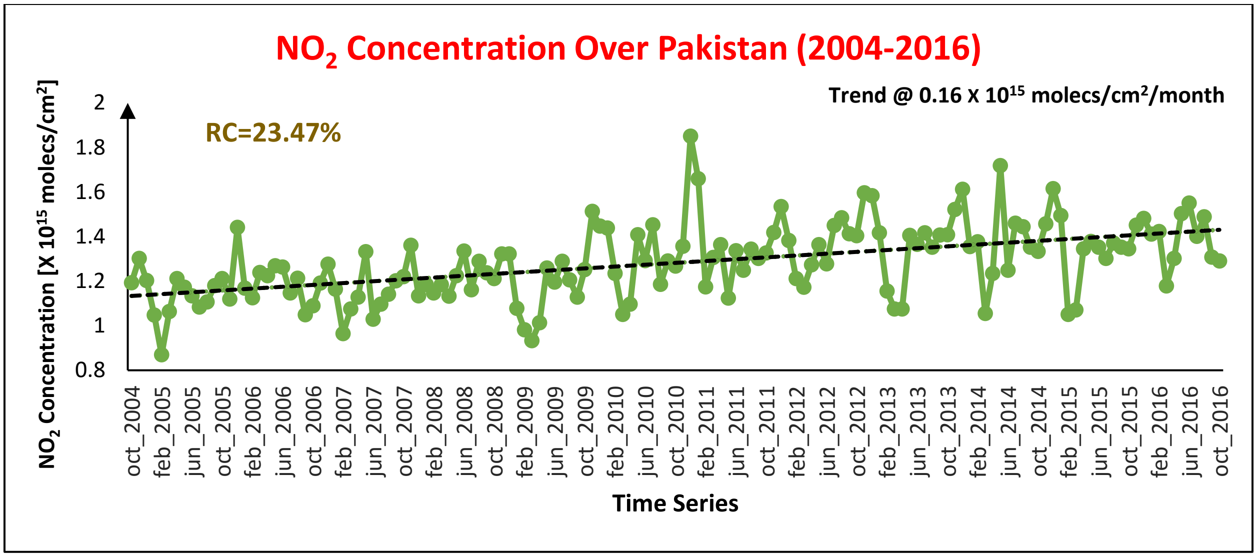


Figure 4.1.6: NO_2 time series over Pakistan from Oct-2004 to Oct-2016

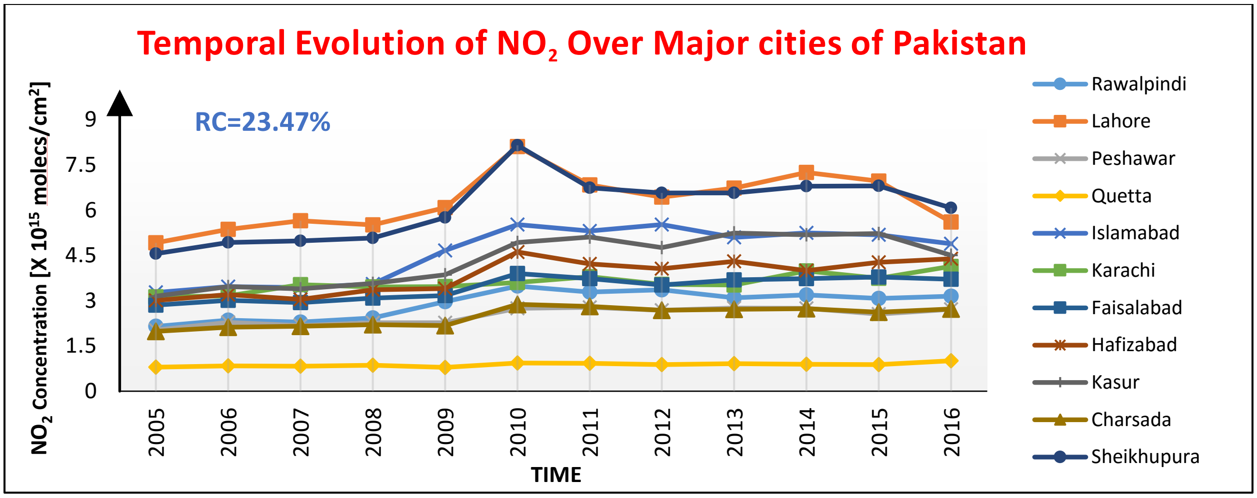
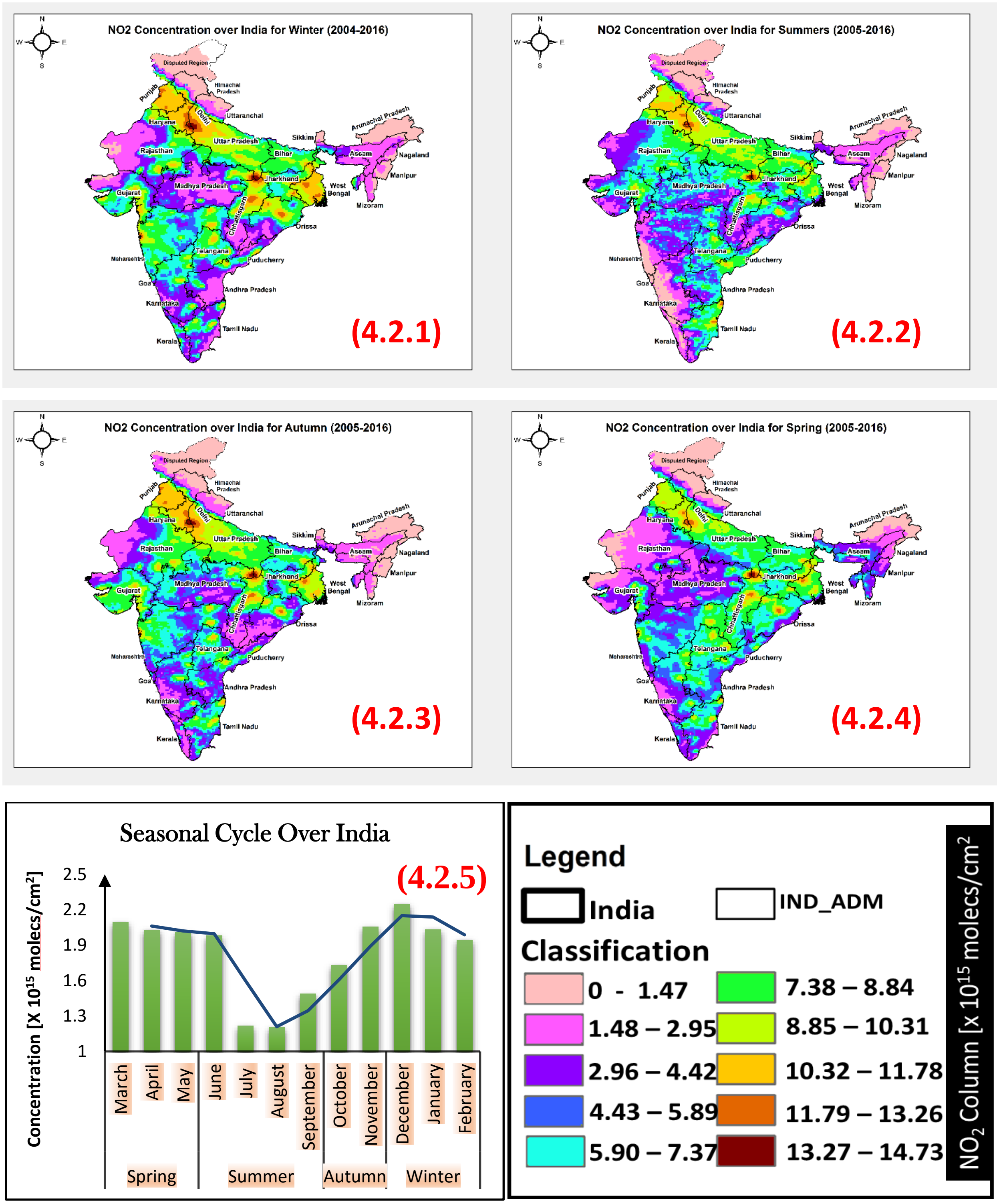


Figure 4.1.7: Temporal analysis of NO_2 Hotspots over Pakistan from year 2005 to 2016

Time Series of NO_2 column in Pakistan from Oct-2004 to Oct-2016 was calculated and presented in figure (4.1.6). A gradual increase was observed over Pakistan (Khokhar *et al.*, 2015) with an instant peak over 2010. Mainly ten cities were identified as NO_2 hotspots all over the country. Sheikhupura and Lahore are on the top followed by Islamabad, Kasur, Hafizabad, Karachi, Faisalabad, Rawalpindi, Peshawar, Charsada and Quetta (See figure 4.1.7). Relatively change of 23.47% has been observed for twelve years (2004-2016)

NO_2 is removed from atmosphere in reaction with OH to form nitric acid. NO_2 concentration decreases in summers (Seinfeld & Pandis, 2016) and increases in winters. Enhanced NO_2 levels during winter might be attributed to increased anthropogenic practices like space heating and accumulation mode of atmosphere due to temperature inversion. Moreover it is also contributed by agricultural fires, biomass burning and forest fires. This maxima might be attributed to cloud cover which decreases the removal of NO_2 through phenomenon of photolysis. There is enhanced usage of fossil fuels in winter season which might be another reason for high NO_2 concentration.

4.2. India Seasonal and Temporal Trends



Figures (4.2.1, 4.2.2, 4.2.3, 4.2.4 & 4.2.5): Show NO_2 Concentration maps of Average Winter (4.2.1), Average Summer (4.2.2), Average Autumn (4.2.3), Average Spring (4.2.4) and Seasonal Cycle (4.2.5) over India from 2004-2016

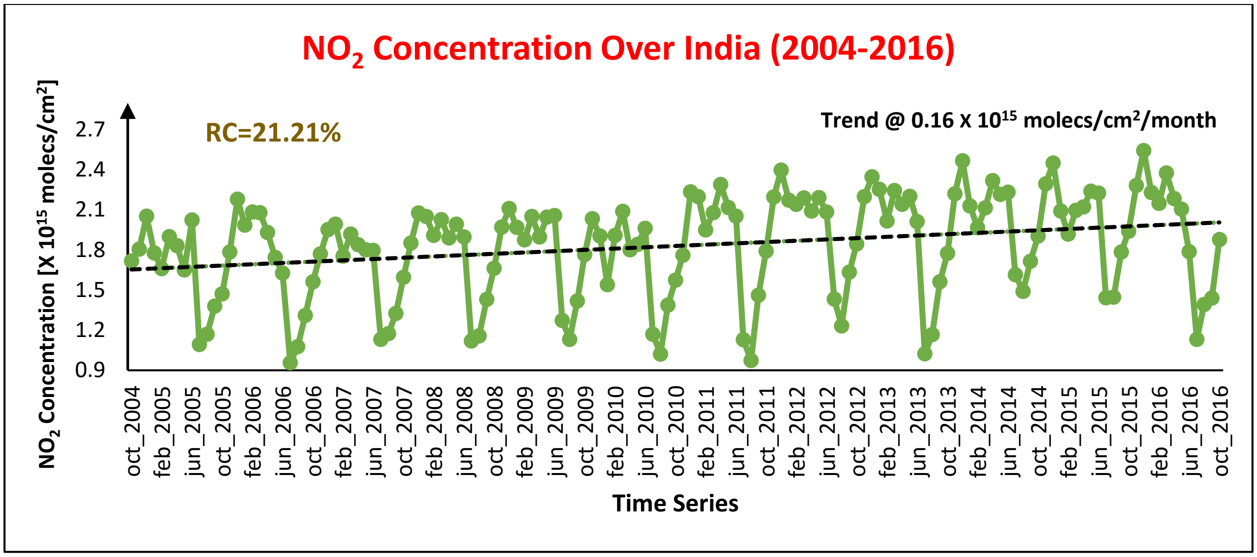


Figure 4.2.6: NO_2 Time series over India from Oct-2004 to Oct-2016

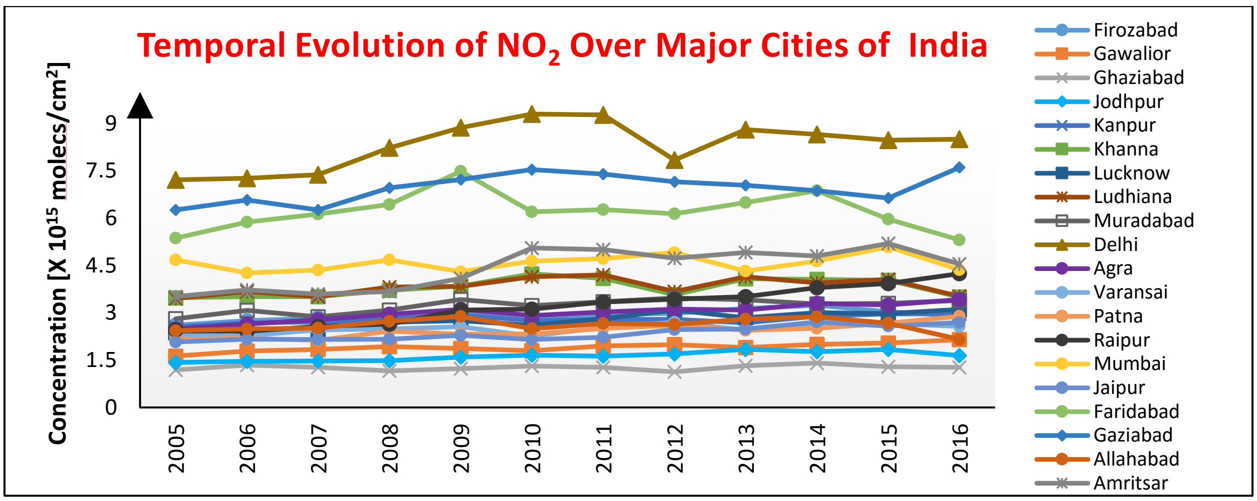


Figure 4.2.7: NO_2 Hotspots over India from 2005 to 2016

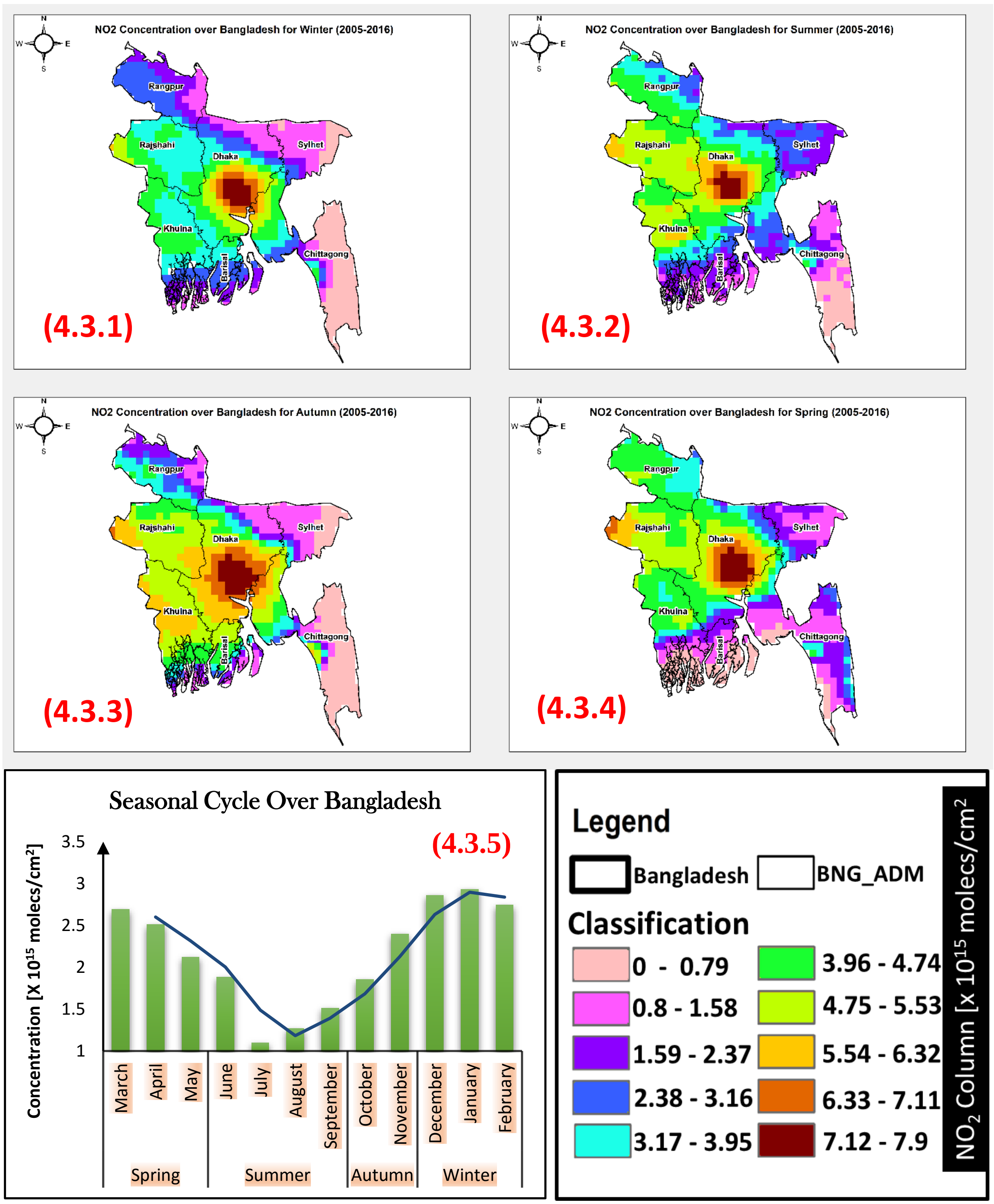
Subcontinent region has 4 dominant seasons spring, summer, autumn and winter. As far the NO_2 levels are concerned, summer season has minima concentration as compared to other seasons (Boersma, 2005). One reason is the availability of OH Radical. In summer, due to monsoon period plus maximum amount of sunlight entering the biosphere thus enhancing the process of evaporation and availability of OH radicals. While on the other hand, winter maxima is due to less availability of OH radical plus increased consumption of fossil fuels & wood burning for heating purposes.

Relatively high values in March–May and October-November are also associated with emissions of NO_2 from large scale open field crop residue burning (nearly 7–10 tons of crop waste per hector) in the study area during wheat-rice rotation periods (Jain *et al.,* 2014) see figures (4.2.1, 4.2.2, 4.2.3, 4.2.4). A winter season maxima with a peak in the month of December, is due to meteorology (dry weather weak winds and conditions), using biomass fuel for winter season house heating specifically in the northern areas, and less UV radiations available for the initialization of photolysis reactions that break down NO_2 (Cantrell *et al.,* 2015). In winter, shallower boundary layer results in lower vertical dispersion which decreases the dilution and removal rates of NO_2 (Sitnov, 2009).

Rapid growth and industrialization over the last few years has led to some major changes in India's environment. Everyday cities are suffering from harmful levels of air pollution as a direct consequence of rapid urbanization and growth in population. A continuous gain was observed over india with the Relative change of 21.21% see figure (4.2.6).

Twenty most populated cities were identified in Indian Territory named as Firozabad, Gawalior, Ghaziabad, Jodhpur, Kanpur, Khanna, lucknow, Ludhiana, Muradabad, agra, Varansai, patna, Raipur, Mumbai, Jaipur, Faridabad, Gaziabad, Allahabad, Amritsar and on the top Delhi as mentioned in figure (4.2.7). Overall India has an increasing trend over every passing year.

4.3. Bangladesh Seasonal and Temporal Trends



Figures (4.3.1, 4.3.2, 4.3.3, 4.3.4 & 4.3.5): Show NO_2 Concentration maps of Average Winter (4.3.1), Average Summer (4.3.2), Average Autumn (4.3.3), Average Spring (4.3.4) and Seasonal Cycle (4.3.5) over Bangladesh from 2004-2016

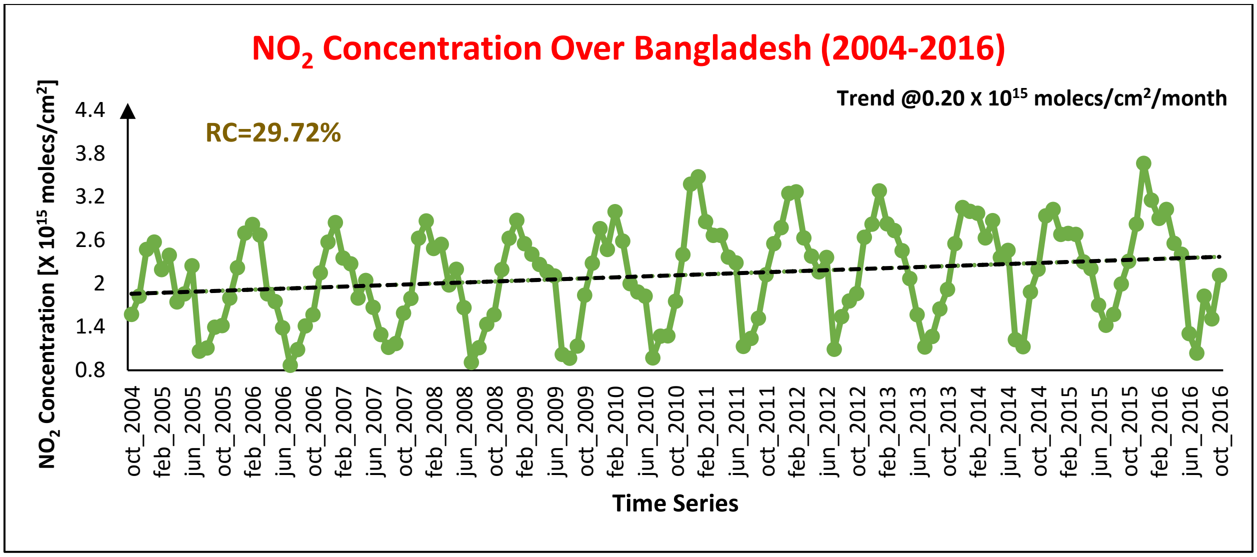


Figure 4.3.6: NO_2 Time series over Bangladesh from Oct-2004 to Oct-2016

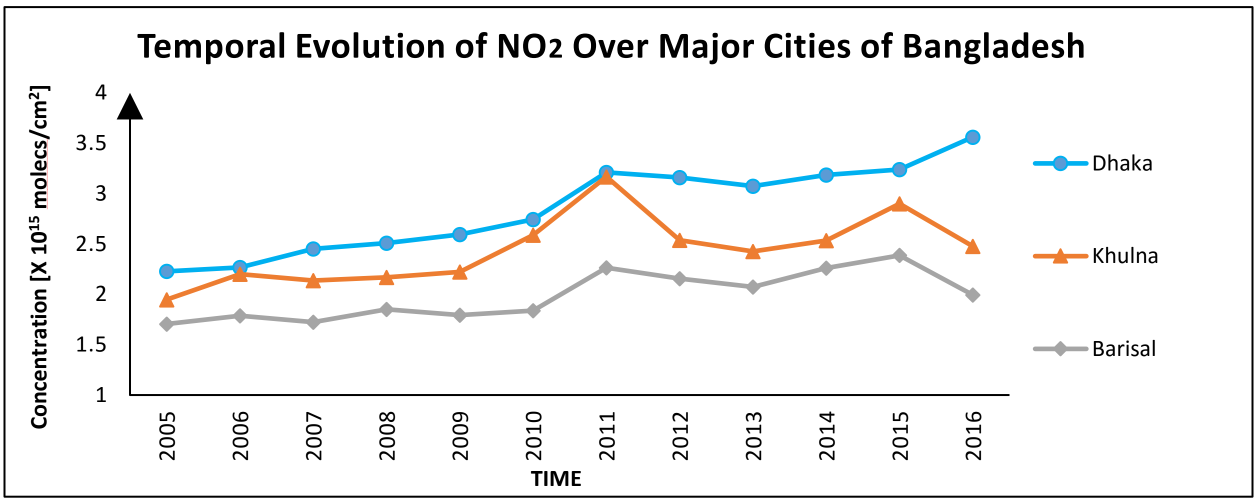
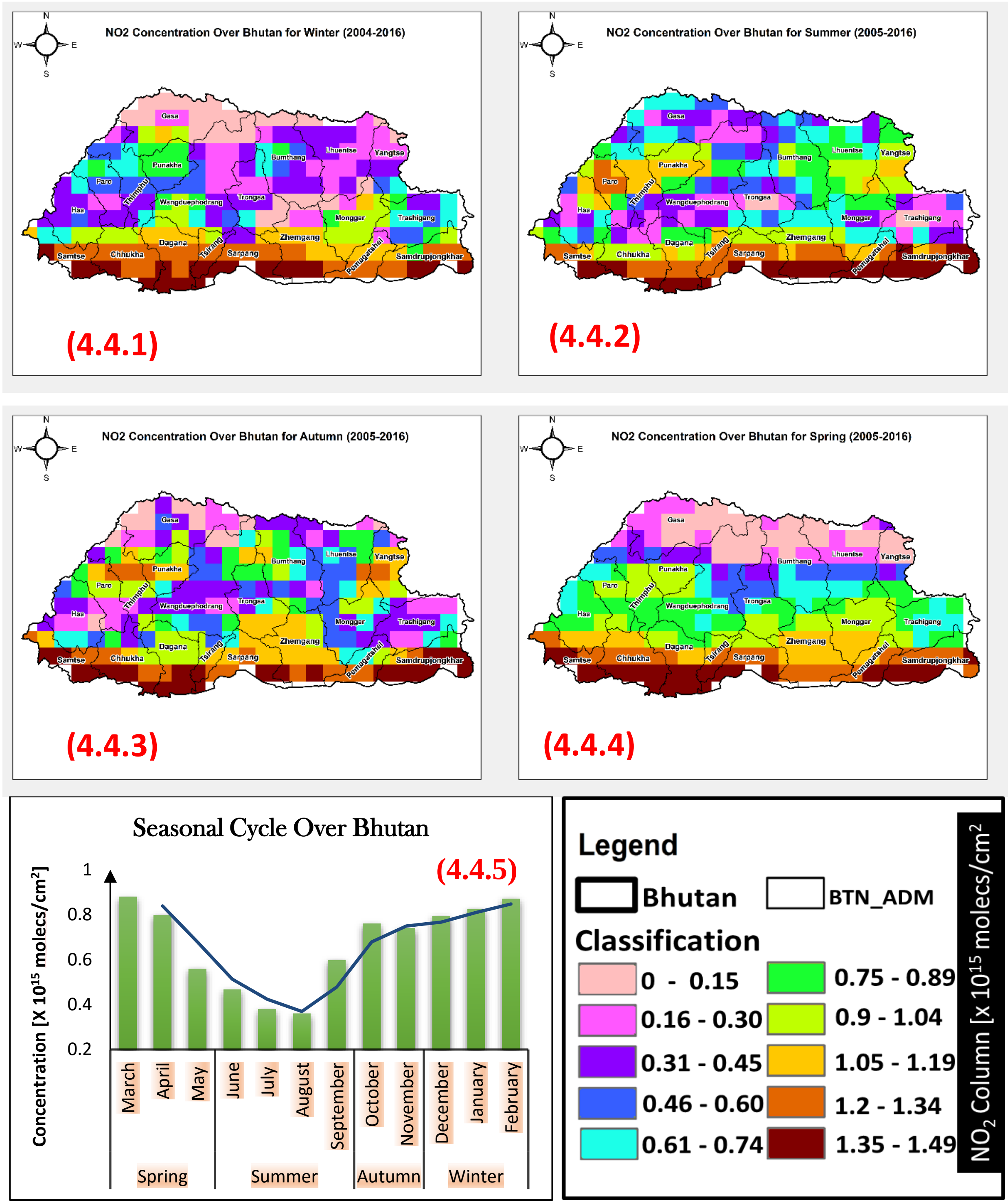


Figure 4.3.7: NO_2 Hotspots over Bangladesh from 2005 to 2016

Seasonal analysis of Bangladesh demonstrate the strong seasonality over the country. From March till may, pre-monsoon hot season extends. While rainy monsoon season which lasts from June through October and filter out the NO_X emissions and other pollutants (Beirle *et al.*, 2003). Autumn is of short duration and lastly from November through February cool dry winter season pervails which promotes the concentration of NO_2, see figures (4.3.1, 4.3.2, 4.3.3, 4.3.4 & 4.3.5). Among all the south Asian countries, Concentrations over Bangladesh were recorded highest as Relative change of 29.72% was recorded for the study period, see figure (4.3.6). Dhaka, the capital of Bangladesh was the top most hotspot following by Khulna and Barisal, see figure (4.3.7).

4.4. Bhutan Seasonal and Temporal Trends



Figures (4.4.1, 4.4.2, 4.4.3, 4.4.4 & 4.4.5): Show NO_2 Concentration maps of Average Winter (4.4.1), Average Summer (4.4.2), Average Autumn (4.4.3), Average Spring (4.4.4) and Seasonal Cycle (4.4.5) over Bhutan from 2004-2016

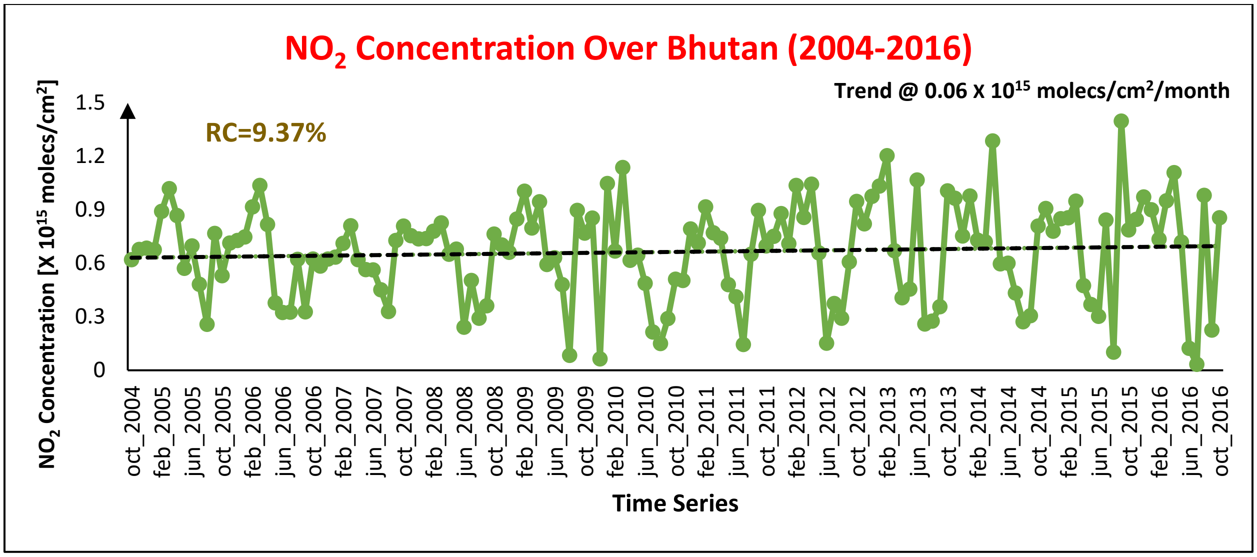


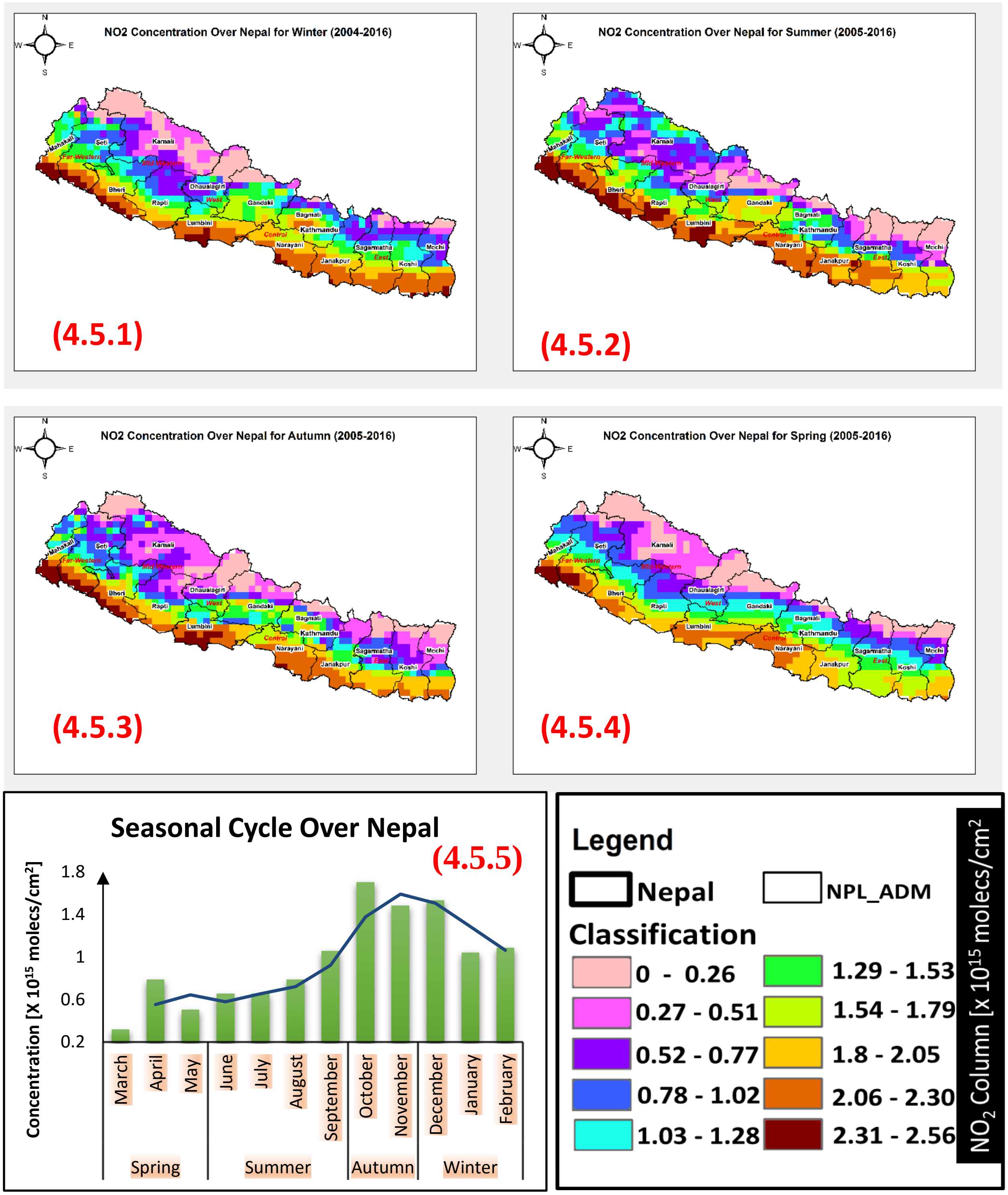
Figure 4.4.6: NO_2 Time series over Bhutan from Oct-2004 to Oct-2016

Indian monsoon is main driving force behind seasonality in Bhutan. The Indian summer monsoon starts from June to late-September and is mostly restrained to the southern region of Bhutan. These rains bring between 60 and 90 percent of the western region's rainfall. Annual precipitation varies widely in various parts of the country (kim *et al.*, 2015). In the Northern border towards Tibet, about forty millimeters of precipitation a year is recorded which primarily is in the form of snow.

Beginning of July marks the advent of south east monsoon till the earlier September. From late November till March, winter sets in with frosting all over the country and snowfall is common above elevations of 3,000 meters. So, maximum concentration is recorded in autumn and winter as exhibited in figures 4.4.1 & 4.4.3.

No evident hotspot was identified in Bhutan. Reason is the 60% forest cover which covers the area and reduce the pollution with no distinct industrial activities in the country.

4.5. Nepal Seasonal and Temporal Trends



Figures (4.5.1, 4.5.2, 4.5.3, 4.5.4 & 4.5.5): Show NO_2 Concentration maps of Average Winter (4.5.1), Average Summer (4.5.2), Average Autumn (4.5.3), Average Spring (4.5.4) and Seasonal Cycle (4.5.5) over Nepal from 2004-2016

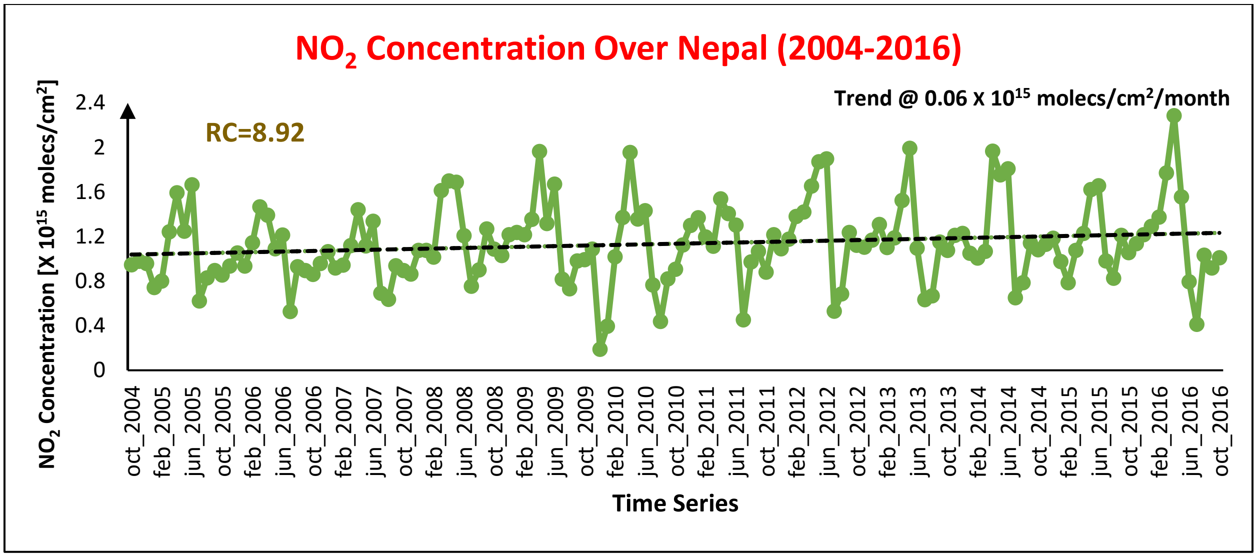


Figure 4.5.6: NO_2 Time series over Bangladesh from Oct-2004 to Oct-2016

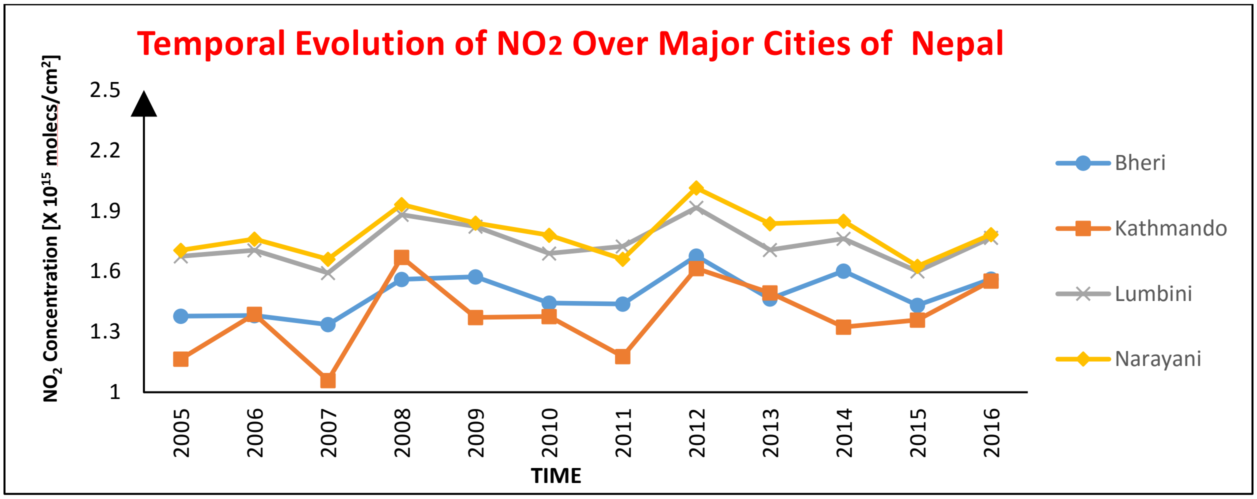


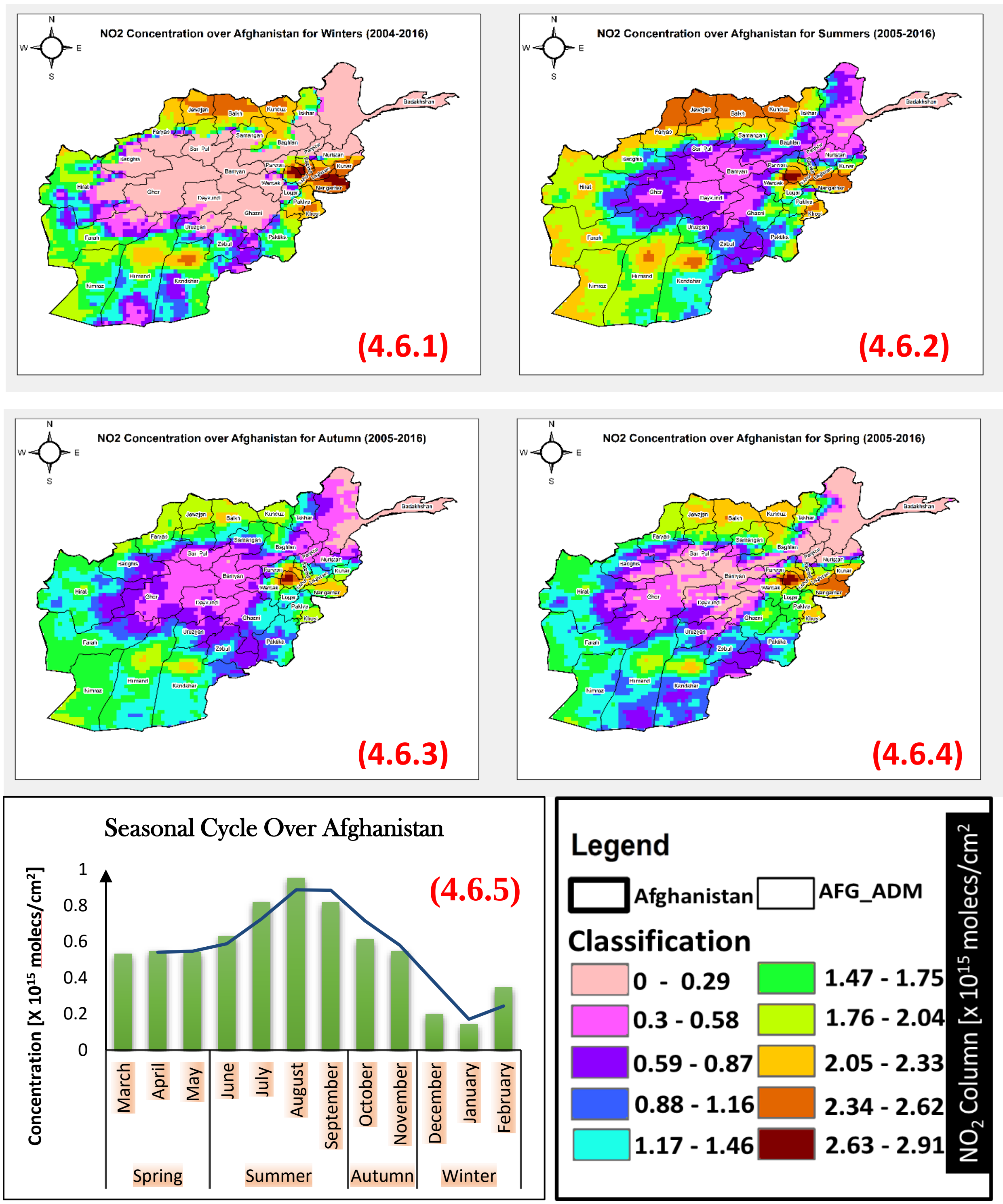
Figure 4.5.7: NO_2 Hotspots over Nepal from 2005 to 2016

Nepal depicted seasonal increase in both autumn and winter season, see figures 4.5.1 &4.5.4. The monsoon of Nepal has two distinct seasons, first wet and second dry season. Wet days of summer are Monsoon days. Most of the rainfall in Nepal is due to Monsoon in summer. The rest are dry days. Generally the monsoon commences from mid-June and ends during beginning of September. Also, september begins with the end of Monsoon and ends with beginning of winter in november. It is also a festival season. Best season for outdoor goings-on like rafting, trekking and wild life jungle safari.

Winter season is from November till February. Winter days or cold day are also dry days with a few rain. Typical day in winter season is as warm as 20◦C in a sunny days but night temperature falls below freezing in Kathmandu valley

Unceasing population growth of years and lack of a rigorous pollution regulation and management systems has left a deep stamp on the environment in Nepal (Saud & Paudel, 2018). Air quality in both urban and rural areas is worsening in the country especially in Kathmandu being at very high levels of risk. The air movement are restricted due to bowl like topography of the Kathmandu valley, in so doing accumulating high levels of pollutants.

4.6. Afghanistan Seasonal and Temporal Trends



Figures (4.6.1, 4.6.2, 4.6.3, 4.6.4 & 4.6.5): Show NO_2 Concentration maps of Average Winter (4.6.1), Average Summer (4.6.2), Average Autumn (4.6.3), Average Spring (4.6.4) and Seasonal Cycle (4.6.5) over Nepal from 2004-2016

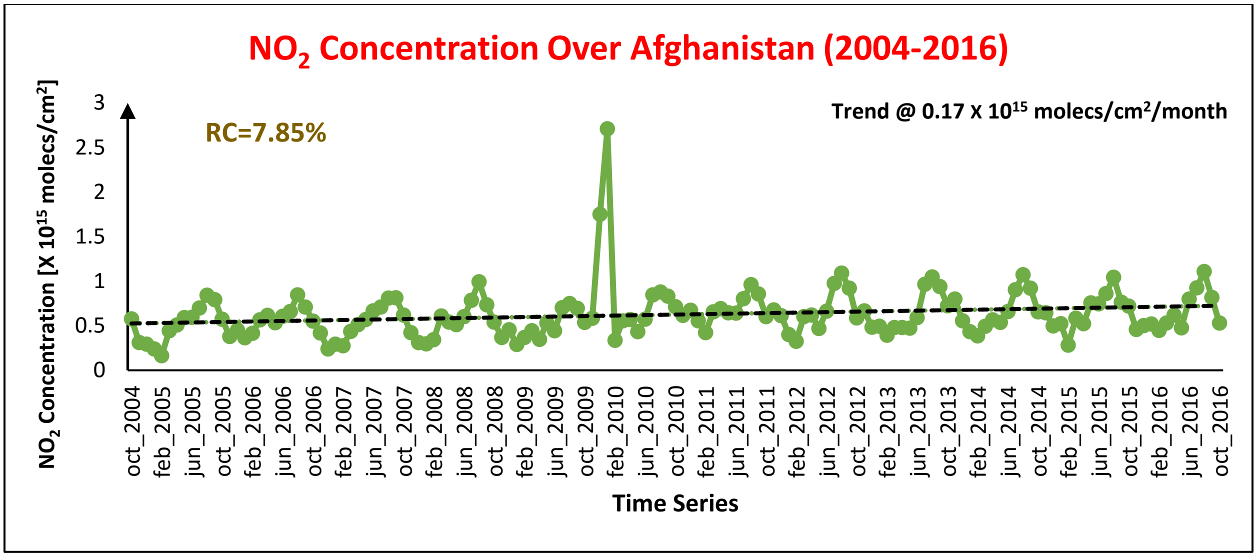


Figure 4.6.6: NO_2 Time series over Afghanistan from Oct-2004 to Oct-2016

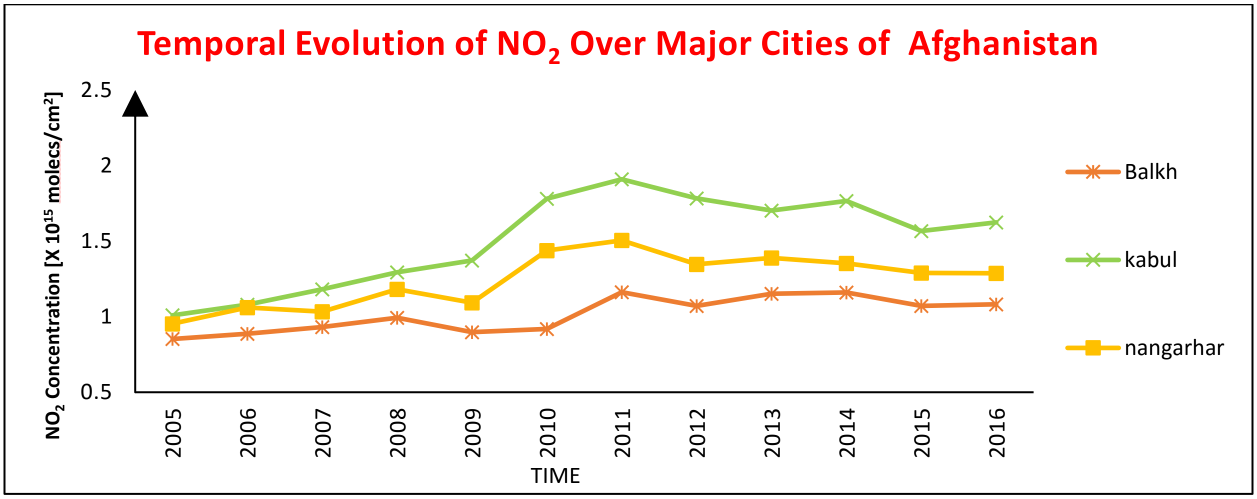


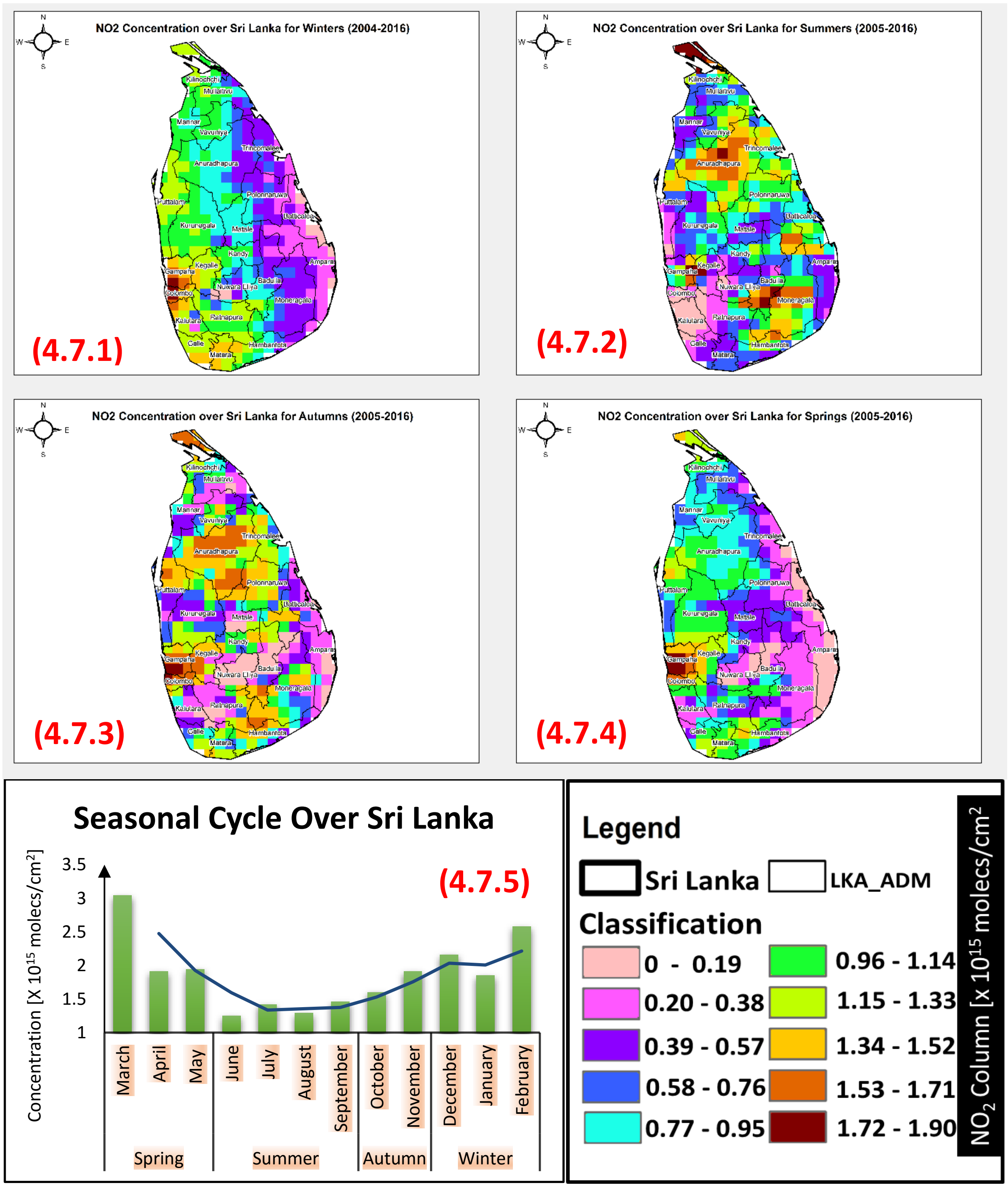
Figure 4.6.7: NO_2 Hotspots over Afghanistan from 2005 to 2016

Afghanistan holds a subarctic climate with dry and cold winter, which have arid and semiarid climates. In the mountains and few of the valleys sharing bordering with Pakistan, a fringe effect of the Indian monsoon, brings moist tropical air in summer. Afghanistan has defined seasons; summers are scorching and winters can be intensely cold, so commercial activities are terminated due to harsh climate. Summer temperatures as high as 49 °C (120 °F) have been recorded in the northern valleys.

The weakening of security was most apparent as increase in roadside bomb explosions was recorded for that time period, which rose 82% over the same period in 2009 (Rubin, 2010).

There has been an increase in the number of civilians killed in the war in Afghanistan for the second year in a row, according to a UN report. More than 2,700 civilians were killed in 2010.The excessive use of Local explosive materials and Weapons used by NATO contributed towards the peak in the year 2009.

4.7. Sri Lanka Seasonal and Temporal Trends



Figures (4.7.1, 4.7.2, 4.7.3, 4.7.4 & 4.7.5): Show NO_2 Concentration maps of Average Winter (4.7.1), Average Summer (4.7.2), Average Autumn (4.7.3), Average Spring (4.7.4) and Seasonal Cycle (4.7.5) over Sri Lanka from 2004-2016

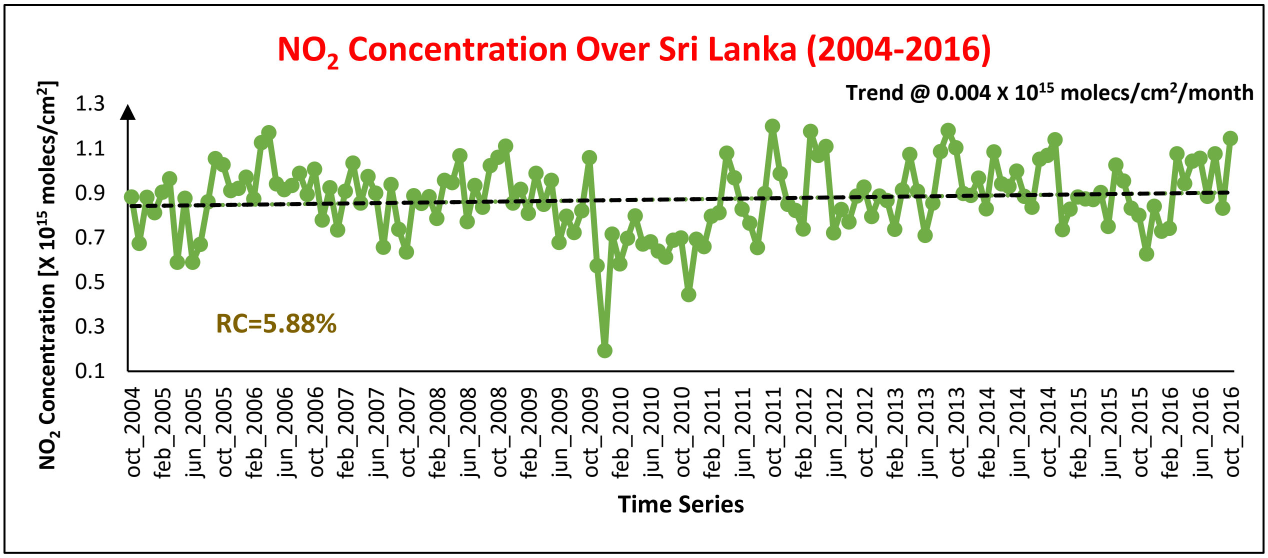


Figure 4.7.6: NO_2 Time series over Sri-Lanka from Oct-2004 to Oct-2016

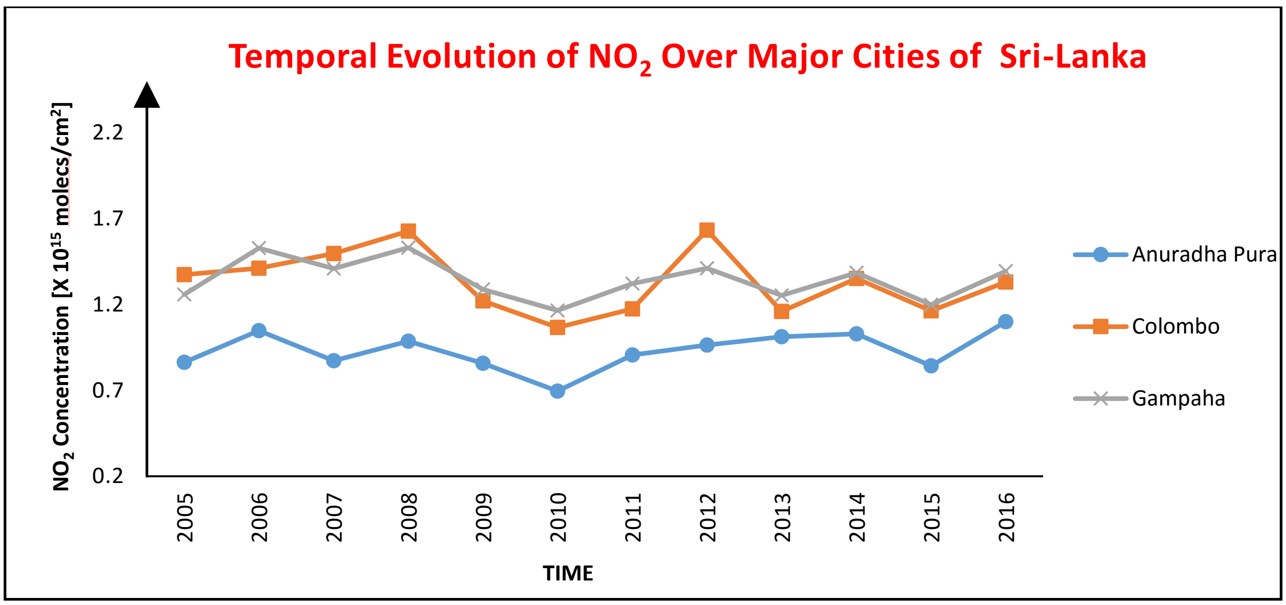
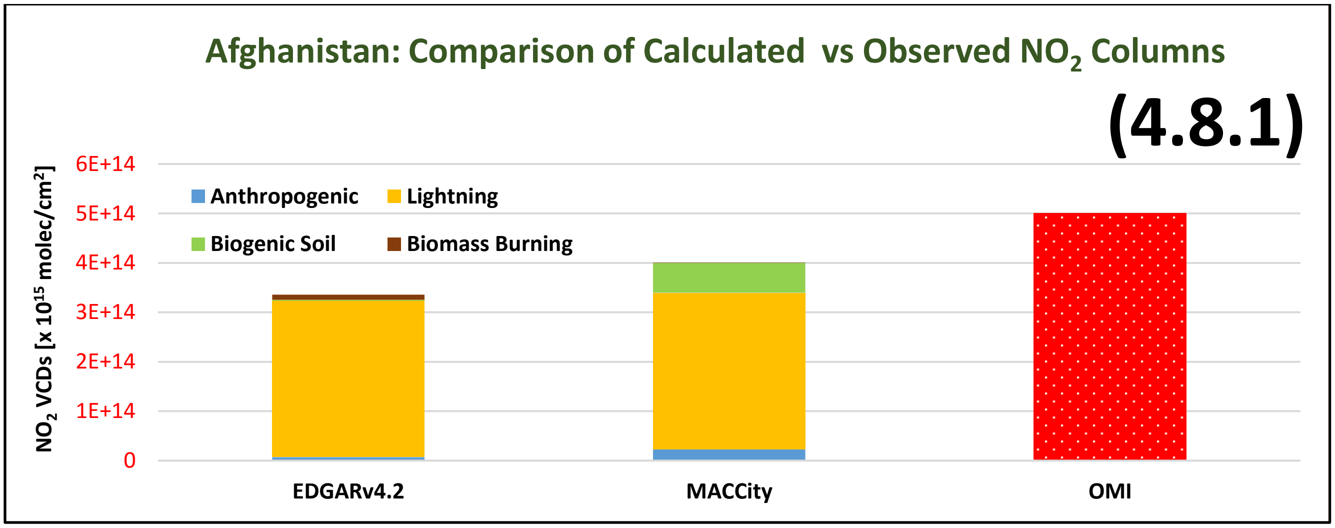


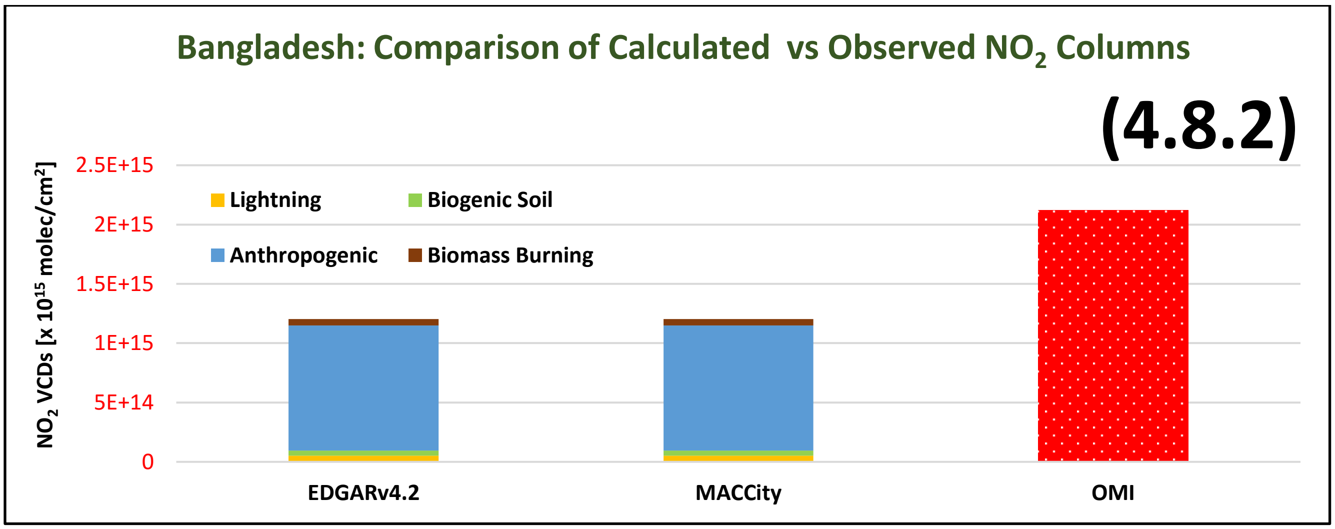
Figure 4.7.7: NO_2 Hotspots over Sri-Lanka from 2005 to 2016

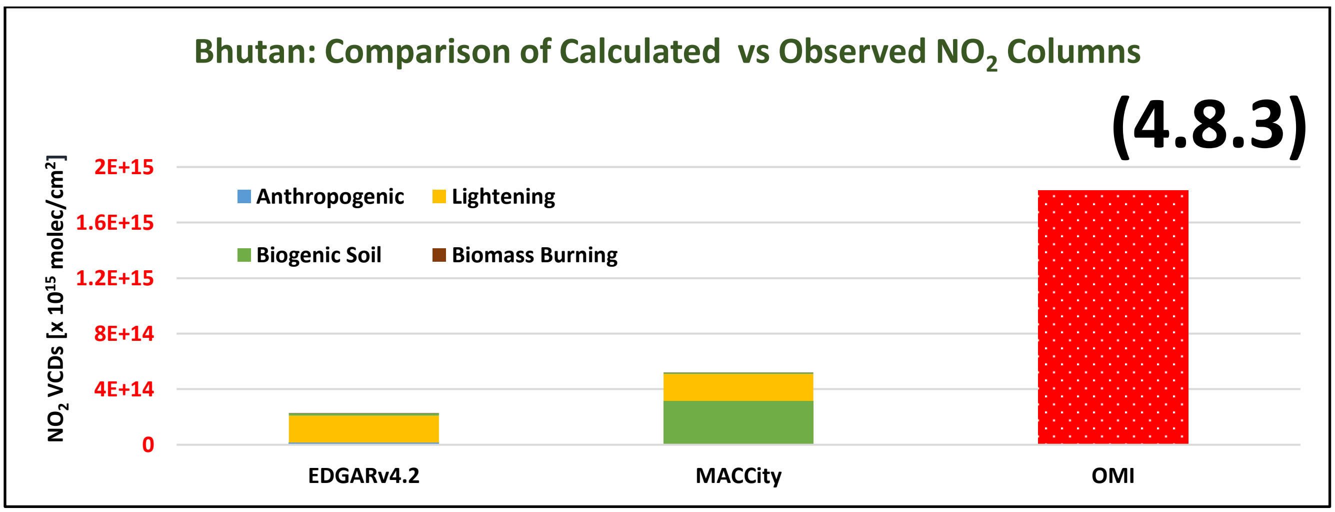
Sri Lanka has 2 monsoonal cycles. First one is southwest monsoon season or the summers (May-September) Monsoon and 2nd one is Northeast Monsoon season or winters (November to February). We can see the dip in the trend line due to these rainy seasons filtering out the NO_2 in the country (Figure 4.7e). Autumn, the period of October – November is the period with the most evenly balanced distribution of rainfall over Sri Lanka. Almost the entire island receives in excess of 400 mm of rain during this season. Consequently, the whole country experiences strong winds with wide spread rain, sometimes leading to floods and landslides. The mean annual temperature fluctuates from 27°C to 16°C in the coastal lowlands.

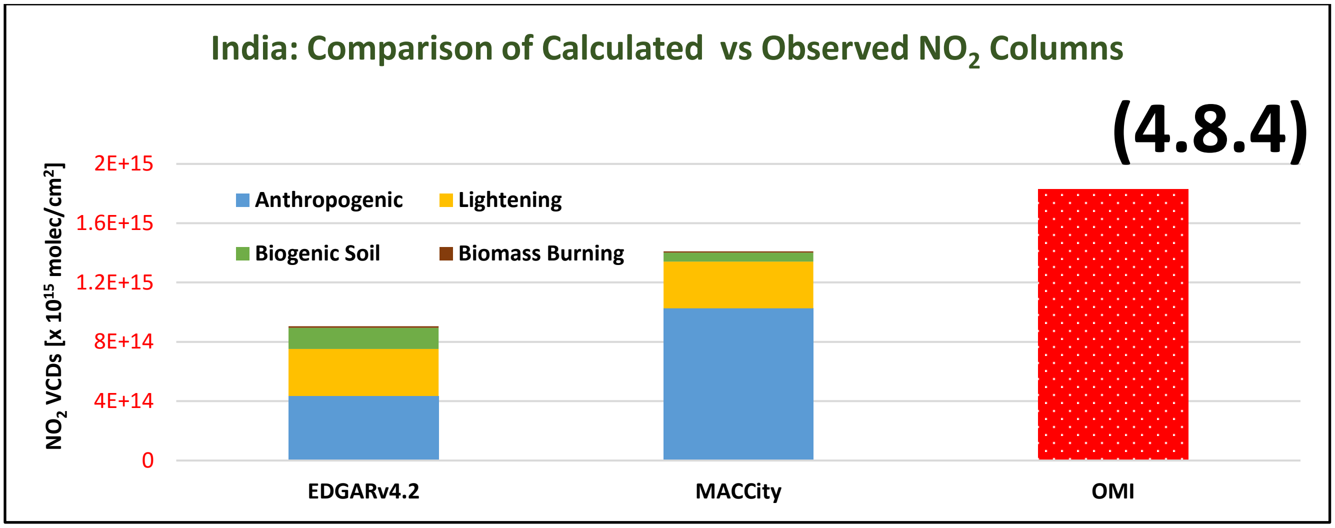
A steady increase on the overall time series can be observed over Sri Lanka (Figure 4.7.6). However, 2009 and 2010 depicted a certain decrease in the trend. Year 2009 was the most important year in the history of Sri Lanka as in this year civil war between Sri Lankan government and Tamil tigers was on peak so economic activities were ceased in most part of the country. In May 2010, pre-monsoon rains strengthened by Cyclone Laila saw thunder storms and high winds, accompanied by floods and landslides. Reports say it swept through 14 of Sri Lanka's 25 districts. The extreme weather caused unadorned distress to 0.5 million people and billions of rupees in damages to public and private possessions. This report is a record of this unanticipated natural disaster and is also a guide to future post disaster assessments and highlights the importance of investment in disaster risk reduction. 3 Hotspots names as Colombo, the capital city, Amerada Pura Economic hub, and Gampaha were identified (Figure 4.7.7).

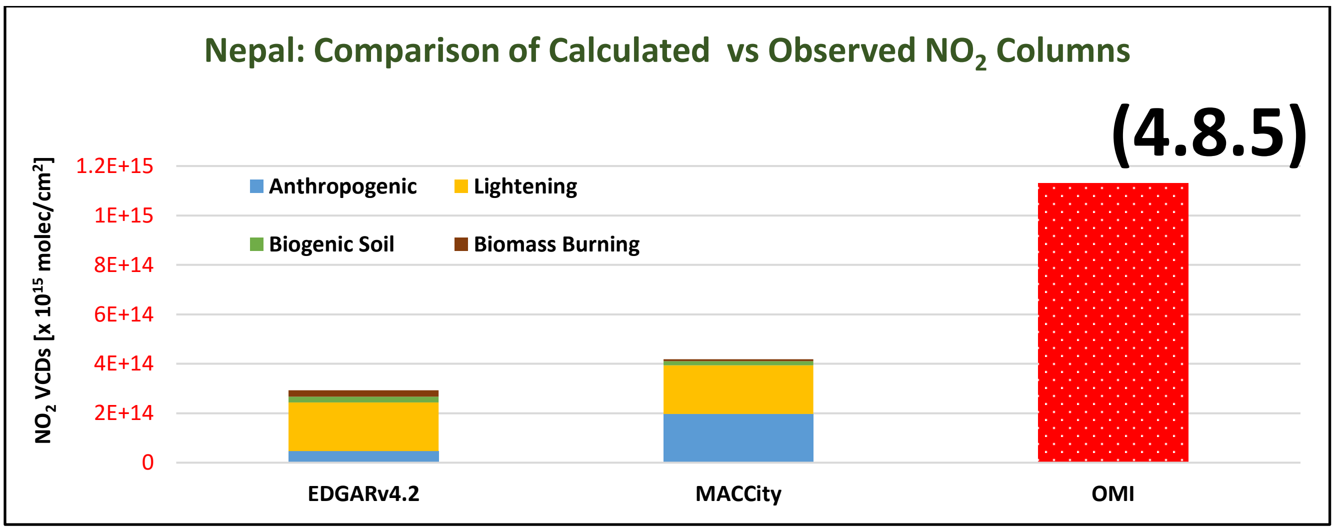
4.8. Comparison with Satellite

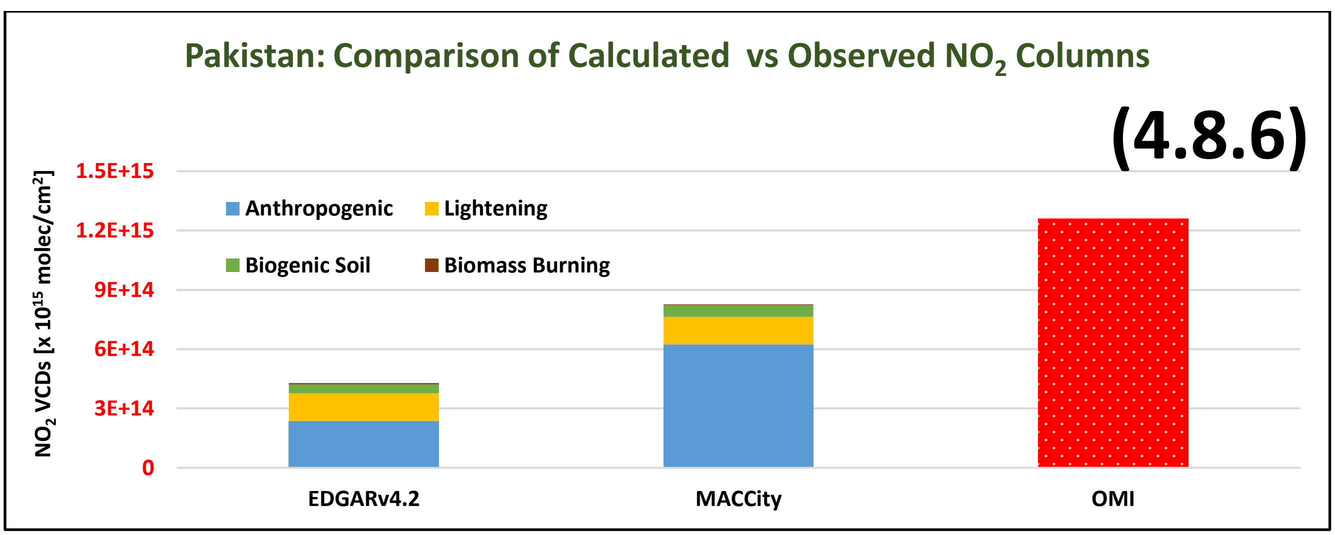


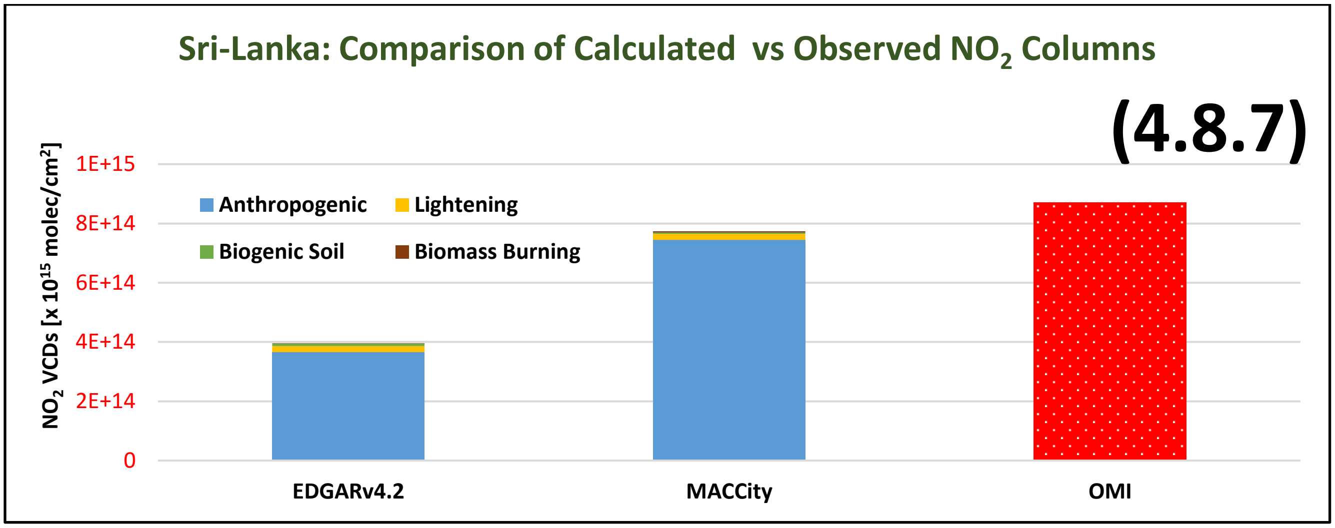





India: Comparison of Calculated vs Observed NO2 Columns
(4.8.4)
NO2 VCDs [x 10^15 molec/cm2]
2E+15
1.6E+15
1.2E+15
8E+14
4E+14
0
Anthropogenic
Lightening
Biogenic Soil
Biomass Burning
EDGARv4.2
MACCity
OMI


Nepal: Comparison of Calculated vs Observed NO2 Columns
(4.8.5)
NO2 VCDs [x 10^15 molec/cm2]
1.2E+15
1E+15
8E+14
6E+14
4E+14
2E+14
0
Anthropogenic
Lightening
Biogenic Soil
Biomass Burning
EDGARv4.2
MACCity
OMI


Pakistan: Comparison of Calculated vs Observed NO2 Columns
(4.8.6)
NO2 VCDs [x 10^15 molec/cm2]
1.5E+15
1.2E+15
9E+14
6E+14
3E+14
0
Anthropogenic
Lightening
Biogenic Soil
Biomass Burning
EDGARv4.2
MACCity
OMI



Figures (4.8.1, 4.8.2, 4.8.3, 4.8.4, 4.8.5, 4.8.6,& 4.8.7): Graphs (4.8.1: Afghanistan), (4.8.2: Bangladesh), (4.8.3: Bhutan), (4.8.4: India), (4.8.5: Nepal), (4.8.6: Pakistan) and (4.8.7: Sri-Lanka) showing the comparison of calculated NO_2 VCDs for respective NOx emission from anthropogenic emissions, biomass burning, lightning, and soil emissions.

In figures (4.8.1, 4.8.2, 4.8.3, 4.8.4, 4.8.5, 4.8.6 & 4.8.7) comparison of the global emission inventories and satellite data depicts that the EDGARv4.2 and MACCity data sets are underestimating the NO_2 levels as compared to OMI observations and we need much more detailed inventories so the comparison will be well relatable. Edgar detailed emission inventory has categorized in the table.

- In Afghanistan (Figure: 4.8.1), usage of electricity generators, biomass burning, and vehicular traffic are important sources of NO_2. The use of convenient generators during power outages is a major source of NO_2 in the country. In Kabul alone, there are about 173,759 gasoline and diesel power generators in which 99.5% are used by households. Animal manure is also used in 85-90% of rural homes and in about 15-17% of urban homes for heating and cooking. The transport sector faces challenges of unlawful import of used vehicles, continued use of very old and poorly maintained vehicles, poor quality of transport fuel, and limited road capacity leading to high emissions of air pollutants. After more than 3 decades of battle, Afghanistan is striving for reconstruction and establishing new systems itself. A lot of prior services, facilities, and capacities have either been wrecked by war or no longer meet the growing demands of the population. As the economy grows, there are increasing demands for energy and mobility. Homes and industries are forced to resort to using dirty fuels and even garbage as fuels for heating and cooking. Second-hand vehicles using poor-quality fuel and with high rates of emissions are increasingly being imported into Afghanistan.

- In Bangladesh (Figure: 4.8.2), NO_2 is mainly emitted from energy transformation industries, motor vehicles, biomass burning for industrial processing and home cooking, burning of agricultural residues, and iron and steel industries.
- In Bhutan (Figure: 4.8.3), the sector-wise emissions estimate of NO_2 indicate that Oil refining process by 3 private companies and energy sectors are responsible for NO_2 emissions. Bhutan is one of the few countries in the world where the environment is still protected largely due to its vast forest cover and widespread use of hydropower and biomass energy. Forest fires are the biggest sources of air pollution in Bhutan. Agriculture Soil and combustion are the main sources for NOx in Bhutan.
- In India (Figure: 4.8.4), anthropogenic activities are major contributors of increasing NO_2 in the country followed by lightning, biogenic soil and biomass burning. Industrial zones, commercial hubs and large-scale biomass burning every year increases the NO_2 emissions. In India, Oil refining process and the road transport are the dominant source of NO_2 emissions as compared to industry and power sector. The number of vehicles, registered in India, was 55 million in 2001 which has grown to around 173.5 million by

2015. Another important source of NO_2 emissions is the industrial process, especially the production of nitric acid, used in fertilizer manufacturing. India is an agrarian country and generates a large quantity of agricultural wastes. Noteworthy amount of NO_2 is released from coal fired power plants. India is the largest energy user, followed by Pakistan and Iran. Coal is India's most copious source of energy and presently almost 60% of its commercial energy requirements are fulfilled by it. At last, widespread use of traditional sources of energy such as fuel wood and animal dung has also been contributing to NO_2 emissions.

- In Nepal (Figure: 4.8.5), inventory data is highly underestimating the Satellite observations.
- In Pakistan (Figure: 4.8.6), Oil refining process, Biogenic emissions and Road transport are to most sources for NO_X emissions. Air pollution is one of the most pressing concerns for environmental protection agencies in Pakistan. Despite having very low energy consumption in comparison to international standards, air pollution in Pakistani cities is soaring (Khwaja & Khan, 2005). Not able to afford gas or electric stoves and heaters, poor people across the country use firewood to cook food and provide heat during the coldest months of winter. Biomass burning is problematic because of the high level of particulate matter produced as well as carbon monoxide and other harmful gases.
- In case of Sri Lanka (Figure: 4.8.7) Oil refining process, Agriculture Waste burning and Biogenic emissions are top most sources. OMI observations are over estimated as compared to inventory sources.

Edgar Emission data sets(Agricultural Soils,Combustion in Manufacturing,Oil Refining Process,Agricultural Waste Burning, Road Transport, Energy Industry, Lightning, Industrial Process, Biogenic Emissions, Non Road Transport, Manure for South Asian Countries											
Countries	Manure	CM	BE	Lightning	AWB	EI	IP	NT	RT	AS	**OP**
India	2.42	10.38	9.52	17.83	4.32	3.65	2.29	4.39	14.78	3.95	**18.14**
Pakistan	1.88	7.74	5.5	5.98	6.26	7.52	9.47	10.68	14.44	14.55	**15.71**
Bhutan	4.32	5.34	3.29	13.45	5.04	14.86	3.11	11.94	10.22	5.37	**15.32**
Bangladesh	5.53	4.46	4	3.39	8.02	20.36	5.1	7.09	16.33	13.31	**3.64**
Nepal	2.18	18.02	3.54	10.48	3.87	6.82	3.49	9.5	3.74	19.08	**2.5**
Afghanistan	3.28	11.56	4.95	4.98	7.82	5.82	20.92	4.21	5.97	4.98	**7.93**
Sri Lanka	6.96	5.36	7.54	12.93	18.07	3.11	3.89	10.37	3.25	17.97	**25.04**

Table 4.8.1: Edgar Data (%Contribution of Sectors emitting NO_2)

CHAPTER 5

CONCLUSION & RECOMMENDATIONS

Tropospheric NO_2 shows high spatiotemporal variability mainly modulated by local emission changes, seasonal cycles, and meteorological conditions. South Asia is experiencing severe air quality degradation due to high population growth rate, burgeoning urbanization and industrialization, expanding demand of agricultural products, and exponentially increasing energy consumption rate. Therefore, in order to develop the effective strategies to reduce its emissions, it is necessary to assess spatiotemporal distribution of NO_2 and identify its emission sources over the study region.

5.1. Recommendations

1. Use data from some other satellite to create and updated database
2. A model should be developed to have a clear picture of the impacts of current emissions and future scenarios
3. Systems must be designed and installed for the ground-based monitoring of NO_2 in all megacities
4. New low-NOx burners are effective in reducing NOx emissions from both new power plants and existing plants that are being retrofitted. Low NOx burners limit the formation of nitrogen oxides by controlling the mixing of fuel and air, in effect automating low-excess-air firing or staged combustion. Compared with older conventional burners, low-NOx burners reduce emissions of NOx by 40–60%
5. NO_2 is converted to acids secondarily. The extent and amount of these acids formation must be monitored and linked to health issues in hotspot areas

6. Policy and Research institute must cooperate to formulate pollution reduction strategies and goals.

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