An Assessment of the NAME III Model Capability in Reproducing Seasonal Variation of SO_2 and O_3 Pollutant Concentrations: A Focus on the Mpumalanga Highveld Area

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By

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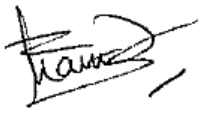
Submitted in fulfilment of the academic
requirements for the degree of
Master of Science in the
School of Agriculture, Earth and Environmental Sciences,
University of KwaZulu-Natal,
Durban.

As the candidate's supervisor, I have/~~have not~~ approved this dissertation for submission.

Signed:  **Name: ----------**

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ABSTRACT

The South African Weather Service Air Quality Modelling programme initiative has a long term goal to develop a system capable of generating atmospheric air quality forecasts for a range of primary and secondary pollutants in order to advise and warn the public on possible high levels pollutant concentration in the air and also provide support in relevant policy development. In this study a pilot NAME III air quality modelling system was developed and tested for its performance in simulation of sulphur dioxide (SO_2) and ozone (O_3) concentrations over the Mpumalanga Highveld. The agreement of model predictions generated in this study with observations was evaluated using the statistical analysis of the monthly averages of SO_2 and O_3 concentrations based on the Bias, NMB, RMSE, NRMSE statistical measures. In addition, the seasonal distribution and variation of the modelled SO_2 and O_3 concentrations over the South African domain were assessed. The results demonstrate that the modelling system under-predicts SO_2 and O_3 concentrations. However, in most cases the modelled concentrations are in the same order of magnitudes with the measured data except for two incidences of very low modelled SO_2 in Middelburg during April and May months, which may be attributed to the poor initialisation of the model. For each season, the model was initialised for the first five days to allow for the pre calculation of the initial pollutant concentrations. This was not possible for the autumn season as no Numerical Weather Prediction (NWP) data were available for initialisation during this period. In general the overall results indicate that the NAME III modelling system is a promising and cost-effective tool for providing real time air quality forecasts, in particular, the ground level O_3 concentration in South Africa. The NAME III modelling system therefore has the potential to be used operationally as a national air quality forecasting system and, as a tool to conduct air quality modelling studies. Specifically the modelling system could assist in the amendment and development of relevant air quality policies that have a direct impact on the environment, health and other related sectors. However, it is suggested that while more evaluation exercises must be undertaken, advancements in term of a comprehensive emissions inventory and improved representation of meteorological information are needed.

PREFACE

The experimental work described in this dissertation was carried out in the School of Agriculture, Earth and Environmental Sciences, University of KwaZulu-Natal, Westville, Durban, from August 2011 to June 2015, under the supervision of Doctor Tirusha Thambiran.

These studies represent original work by the author and have not otherwise been submitted in any form for any degree or diploma to any tertiary institution. Where use has been made of the work of others it is duly acknowledged in the text.

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Signed: ……

This thesis is dedicated to:

My Son: Njabulo Sibiya

&

My Daughter: Bandile Sibiya

Acknowledgements

First and foremost, I would like to thank God for providing me with the life opportunity to live and grow my knowledge.

I would wish to convey my earnest gratitude to my supervisor Dr Tirusha Thambiran for her contribution towards the culmination of this dissertation. Her invaluable help, guidance and patience throughout this project is extremely valued.

To my late parents, Simon P Sibiya and Chrestinah T Shongwe, I am grateful to be your son and you will always have a special place in my heart.

The South African Weather Service is acknowledged for financial assistance and for providing a workplace environment that is conducive for one to be able to pursue a higher degree while working.

The colleagues, friends and family are most thanked for their support and the belief they had in me.

The South African Air Quality Information System team acknowledged for providing me with the air quality data.

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List of Abbreviations

ADMS	Atmospheric Dispersion Modelling System
AERMOD	Environmental Protection Agency Regulatory Model
AERSCREEN	Screening Version of AERMOD
AMFIC	Air Quality Monitoring and Forecasting In China
AQMFS	Air Quality Modelling and Forecasting System
AQMPs	Air Quality Management Plans
AR5	Fifth Assessment Report, IPCC
CMIP	Coupled Model Inter-comparison Project
CO	Carbon Monoxide
COMPLEX1	EPA's Screening Model
DEAT	Department of Environment Affairs and Tourism, South Africa
DEA	Department of Environmental Affairs, South Africa
DEBITs	Deposition of Biogeochemical Important Trace species
ESA	European Space Agency
FB	Fractional Bias
GEIA	Global Emission Inventory Activity
GOME-2	Global Ozone Monitoring Experiment, Second Generation
GURME	GAW Urban Research Meteorology and Environment
g/m^3	gram per Cubic meter
HPA	Highveld Priority Area
IGAC	International Global Atmospheric Chemistry
IPCC	Intergovernmental Panel on Climate Change
$\mu g/m^3$	micro gram per Cubic meter
MG	Geometric Mean Bias
NAEIS	National atmospheric Emission Information System
NAME III	Numerical Atmospheric Modelling Environment, third generation
NMB	Normalised Mean Bias
NO_x	Oxide of nitrogen

NRMSE	Normalised Root Mean Square Error
NWP	Numerical Weather Prediction
O_3	Ozone
PM	Particulate Matter
Ppb	Part Per Billion
R	Correlation coefficient
RDTM3.2	Reduced Differential Transform Method
RMSE	Root Mean Square Error
SAAQIS	South African Air Quality Information System
SAIEA	Southern African Institute for Environmental Assessment
SAFARI	Southern African Regional Science Initiative
SAWS	South African Weather Service
SO_2	Sulphur Dioxide
UKMO	United Kingdom Meteorological Office
UM	Unified Model
UNEP	United Nations Environmental Programme
US EPA	Environmental Protection Agency, US
VG	Geometric Variance
VTAPA	Vaal Triangle Airshed Priority Area
WHO	World Health Organisation

CHAPTER 1: INTRODUCTION

1.1 Background

The deterioration of air quality has been a major concern since the industrial revolution in Europe since the 18th century during which smoke and ash were the predominant air pollution problems, especially in the northern European countries. Great Britain took the lead in developing an air quality management system to address the air pollution problem as early as 1848 (Stern *et al*., 1984). The development of automobiles as well as an increase in population size and growth of industrial activities during the 19th century exacerbated urban air quality problems as a result of anthropogenic emissions due to the increased consumption rate of non-renewable resources for energy generation and transportation.

Several studies (Scorgie (2012), Jayachandran (2009)) have shown a strong relationship between poor air quality and health problems that may result in loss of life. At present, many countries around the world have implemented air pollution regulations and there has further been significant growth in air quality research capacities and the development of pollution control technologies. As a result, ambient air quality has improved considerably in the last few decades in many developed countries (WHO, 2003).

Contrary to this in many developing countries, air pollution still has significant impacts on human health, with South Africa being no exception. South Africa's economy has been growing rapidly ever since it became a democratic country, it is indicated that between 2002 to 2012, the country's gross domestic product grew by about US $270-billion (ESA, 2013). The largest portion of this growth has been and still is boosted by industrial activities. Freiman and Piketh (2003) confirms that South Africa has one of the largest industrialized economies in the southern hemisphere and is the only industrialised regional energy producer on the African continent. Unfortunately, such economic growth is often associated with various environmental problems, one critical problem is poor air quality related to industrial activities that often lead to high levels of atmospheric pollutants which can have a negative impact on people's health. The rapid increase in vehicle density in the urban areas and high dependency on fossil fuel burning for domestic purposes has also exacerbated air pollution in the region (Forbes and Rohwer, 2008).

It is estimated that about 950 000 households in South Africa use coal as a household energy source (Balmer, 2007). Consequently many people, particularly the lower socio-economic status, are exposed to an atmosphere that is contaminated with high levels of pollutants on a daily basis. This has triggered threats to the health and well-being of the people living and working in the country. As a result, much effort has been invested in studies related to air pollution (Tyson *et al.*, (1997); Piketh *et al*., (2004); Zunckel *et al.*, (2004)) and its impact (Zwi *et al*., 1990; Engelbrecht *et al.*, 2014; Wright *et al.*, 2014) in the region.

It has been reported that approximately 4000 to 5000 South African population die each year as a result of air pollution (Jairam, 2008) and about 2000 of the total mortality are among children (Scorgie *et al*., 2003). Norman *et al*. (2007) estimate that in urban areas, ambient air pollution is responsible for 3.7% of the total mortality from cardiopulmonary disease in adults aged 30 years older, results in 5.1% of cancer of the trachea and bronchus in adults, and 1.1% of total mortality of children under the age of five due to acute respiratory disease. The Trade and Industry Chamber (2004) estimates that illness related to air pollution costs the South African government around 800 million Rand per annum in 2004.

The development of the national ambient air quality standards (DEAT, 2008a) and air quality monitoring networks are seen as a means to address the problem. However, there is still a gap in preventing people's exposure to the contaminated ambient air. The air quality monitoring instruments are very sparse in South Africa, monitoring networks are often concentrated in the urban areas and mostly in industrial areas (e.g. Vaal Triangle Airshed Priority Area (VTAPA), Highveld Priority Area (HPA)) and in metropolitan municipalities. As a result, little information is known on the state of air quality in remote areas. A more promising approach in addressing air pollution problems must therefore also include mathematical tools that would support the prediction of air pollution at any location on a daily basis. This is the key to equipping the public, especially those more vulnerable to air pollution with information that will allow them to plan their daily activities with the state of air quality in mind.

The provision of air quality forecasts may thus minimize the risk of human exposure to outdoor contaminated air and thus help to alleviate pollution related illnesses and deaths.

1.2 Motivation for the Research

The South African Weather Service (SAWS) is developing an operational state of the art air quality modelling and forecasting System (AQMFS) with a primary goal of providing the national air quality forecasts to the general public of South Africa. It is also expected that the system will facilitate air quality research projects that may provide support to policy makers in amending current laws, and developing and implementing new laws for addressing air quality problems locally and nationally.

At this stage a pilot AQMFS has been developed and is currently running under research mode. The current AQMFS is physically composed of three separate modules including the air dispersion tool, emissions as well as Numerical Weather Prediction (NWP) modules which communicate with one another as indicated in Figure 1.1 below.

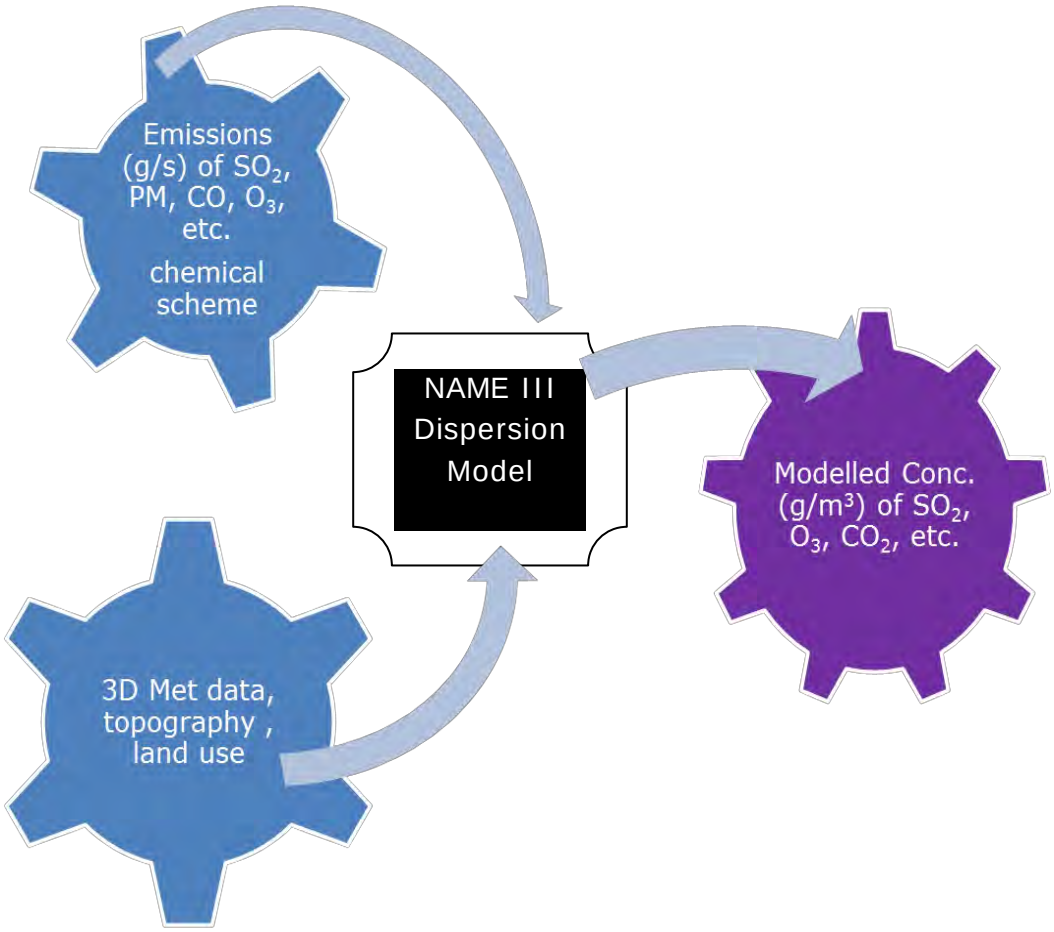


Figure 1.1: Diagrammatic Representation of the SAWS AQMFS Flow.

The third generation of Numerical Atmospheric-dispersion Modelling Environment (NAME III) dispersion model accounts for the system's dispersion module and is the central part of this AQMF system. The NAME III was developed by the United Kingdom Meteorological Office (UKMO) and is used for various purposes such as volcanic plume tracking, response to emergencies, modelling the deposition of radioactive material as well as daily forecasting. According to Jones *et al.* (2004), NAME III has been applied in various European countries, including the UKMO and has performed well in predicting concentrations of atmospheric pollutants.

However models are developed theoretically under certain assumptions and conditions. For this reason, there is a need to evaluate the model performance under different observed conditions and where necessary to modify the model to best account for local climatic condition before it is used for operational

purposes. Chang and Hanna (2004) acknowledge the importance of this exercise as it may demonstrate the model's fidelity in representing the phenomena of interest.

With the above information in mind, thus it is of great importance that the NAME III modelling system is tested and evaluated using statistical methods before it is calibrated and validated for use as an operational air quality forecasting tool by SAWS. Consequently, the aim of this study was to investigate the performance of the NAME III model in calculating the concentration of different trace gases in the atmosphere over South Africa. This was achieved through comparing the model results with the actual observation from the available monitoring networks within the Highveld.

It is important to note that it is not only the dispersion model that should undergo the process of evaluation as it is also important to ensure the credibility of the dispersion model input information. This is required as the complete system is an integration of the three components as shown in Figure 1.1. However, for this study the archived UKMO global NWP and the IPCC (Intergovernmental Panel on Climate Change) global emissions data which had a longer time scale, were assumed to be appropriate for use in this study. The archived NWP data are taken to be of good quality as the source of this information has been tested and found to be useful for regional climate studies (Boutle and Abel, 2012). The Unified Model (UM) is being used operationally to produce national weather forecasts in South Africa and the global UM output is used to initialise the SAWS nested model (Landman *et al*, 2012). On the other hand the IPCC emission data are also recognised globally and used for various international researches that add value on decision making at international level. One example of the importance of the IPCC data is in Coupled Model Inter-comparison Project (CMIP), the CMIP facilitate the inter-comparison and improvement of global climate models (Meehl *et al*., 2000). Hence it is assumed that the data are of high calibre.

1.3 Statement of Purpose

Aim: The aim of this study is to investigate the capability of the NAME III model in Reproducing Seasonal Variation of sulphur dioxide (SO_2) and ozone (O_3) pollutants concentrations in South Africa using the eastern Highveld region as a case study.
The objective of this project is to determine

1) How the monthly average SO_2 and O_3 concentrations generated by the NAME III model compare against the concentrations measured by the monitoring stations in the eastern Highveld.
2) Which of the trace gases investigated in this study (SO_2 or O_3) is better simulated by NAME III.
3) If the NAME III model provide a realistic/accurate prediction of the ambient air quality over the South Africa.
4) The temporal and spatial distribution of the two pollutants over South Africa.

1.4 Structure of Thesis

The body of the thesis has been constructed as a series of coherent sub-studies which build into hierarchical chapters. Each of these chapters follows the general structure of an academic report. Chapter 2 contains a detailed literature review of $O_3$and SO_2 outlining the sources and sinks, chemical reactions in the atmosphere as well as health impacts. Chapter 3 discusses the methods and the data sources used in the research study. The results and analysis are presented in Chapter 4. Chapter 5 summarises the findings of this research and presents conclusion and recommendations.

CHAPTER 2: LITERATURE REVIEW

2.1 Introduction

Air pollution can be defined as the atmospheric condition in which substances are present at concentrations higher than their normal ambient levels to produce significant effects on living organism and the environment (Arya, 1999). A polluted atmosphere can contain particulate matter (PM), sulphur-containing compounds, organic compounds, nitrogen-containing compounds, carbon monoxide (CO), halogen compounds, and radioactive compounds that may be in gaseous, liquid or solid state injected from a variety of sources (Steyn, 2005).

Air pollution has increased over time due to anthropogenic activities including industrial activities, vehicular emission and increased biomass burning and other economic activities which has led to global and localized deterioration in air quality. The current state of air quality has become a major threat to the health and wellbeing of people as well as the environment at localities around the world. Health impacts of air pollution can increase morbidity and mortality in both developed and developing countries, with the rate of incidences of premature mortality being seen to be more severe in the African continent and particularly in southern Africa (SAEIA, 2003). The World Health Organisation (WHO, 2014) estimated that urban air pollution causes the premature death of more than 3.7 million people in low and middle countries per year, emphasising the need to better understand the levels of these pollutants.

In South Africa, O_3 and SO_2 are considered to be priority pollutants of concern within the country. SO_2 is a primary pollutant as it is directly emitted from a source and has a well-defined emission inventory as its main sources are well understood, particularly in the Highveld. O_3 is secondary pollutant thereby making it a pollutant that allows for testing capability of the model's chemistry. There is further a volume of studies concerning these pollutants have been conducted in South Africa {e.g. Josipovic, (2009), Zunckel *et al*. (2000), Igbafe, (2008)} making it easier for this study results to be compared/validated against the findings from previous studies. Thus, even though there a number of pollutants are of importance in South Africa (as listed in sub-section 2.7.1) only the O_3 and SO_2 pollutants will be discussed here. Specifically, a review of the sources, sinks and impacts of these pollutants as well as the approaches to inventorying and modelling their pollution levels is relevant to this study.

2.2 Characterisation of the sources, sinks and impacts of SO_2 and O_3

2.2.1 Sulphur dioxide

2.2.1.1 Characteristics and sources

Sulphur Dioxide is a colourless gas that is soluble in water and can be oxidised to form sulphur trioxide resulting in the formation of sulphuric acid in the presence of water. The SO_2 can be directly produced from both natural and anthropogenic sources. Natural sources of this pollutant include volcanic activity and the oceans. The concentration level of SO_2 from volcanic plumes can be in the range of several tens of parts per million (ppm) (EPA, 2008). The most significant anthropogenic source of SO_2 emissions is notably from point sources, particularly from power generation and industrial processes. Atmospheric SO_2 is formed as a by-product of the combustion of fossil fuels and smelting of sulfidic ore (Nikolić *et al.*, 2010), with fossil fuel combustion at electric utilities contributing a major portion of total global emissions (Klimont *et al*., 2013).

2.2.1.2 Sinks and impacts

Once released into the atmosphere SO_2 may be converted to other compounds or removed from the atmosphere through various atmospheric processes such as oxidation, wet and dry deposition, absorption by vegetation and by soil. As such the accumulation, reaction and dispersal rate of SO_2 in the atmosphere is highly dependent on the state of the meteorology (Zarate, 2007). Specifically, it has been shown that the occurrence of weak surface wind speed and low surface temperatures in the morning may induce a surface inversion layer. The inversion layer may trap pollutants, resulting in an accumulation of pollutant concentrations near the surface. Higher temperatures in the afternoon can break the inversion layer, allowing for better dispersion and mixing of pollutants (Abiodun, 2013). The precipitation process may allow for the washout of SO_2 pollutants in the atmosphere. The residence time of the SO_2 in the atmosphere is estimated to range between one to seven days (Hakkarinen, n.d.).

Exposure to SO_2 may result in an irritant effect on the lining of the nose, throat and lungs which can cause tightness in the chest and constriction of the airways, reducing air flow (Angle *et al.*, 2006). Asthmatics are particularly sensitive, and high SO_2 concentrations may trigger asthma attacks (COMEAP 2010,

2011). It is further understood that SO_2 has a negative impact on ecosystems. When emitted into the atmosphere, SO_2 may react with water and other compounds to form various acidic compounds and fine particles resulting in the acidification of lakes and streams (US-EPA 2002). It can also cause harm to sensitive forest and coastal ecosystems as well as causing accelerated decay of materials, paints and structures (US-EPA 2002).

2.2.2 Ozone (O_3)

2.2.2.1 Characteristics and sources

The O_3 molecule, also referred to as triatomic oxygen, consist of three oxygen atoms bound together. O_3 occurs at various levels in the atmosphere, however there are only two regions where O_3 is of major concern to human well-being. These include the stratospheric and the tropospheric regions, which are the two regions which have the largest sum O_3 concentrations of the atmosphere (Figure 2.1).

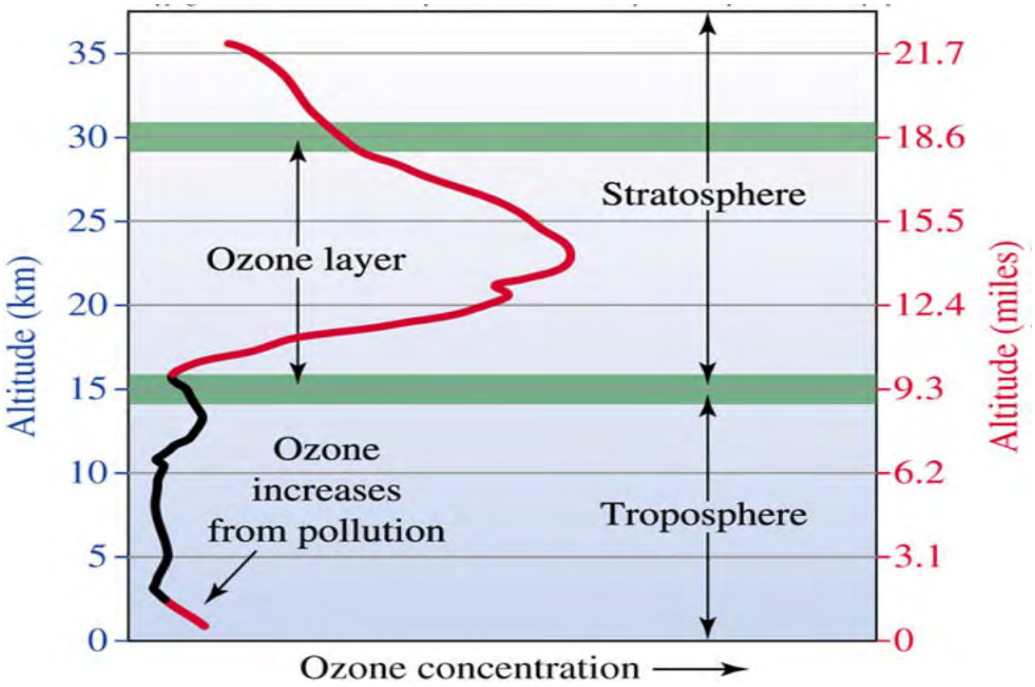


Figure 2.1 Diagrammatic representation of the vertical distribution of O_3 in the atmosphere (http://www.theozonehole.com/twenty.htm)

The mechanisms of O_3 formation and its effects in these regions are quite different and are discussed in more detail below.

2.2.2.2 Stratospheric O_3: sources and impacts

The stratosphere is the region where the major (about 90 %) portion of atmospheric O_3 concentration is located. Stratospheric O_3 is found at about 16-35 km (stratospheric region) above the earth. Ozone at this region occurs naturally in a three step process involving the oxygen (O_2) molecule and ultraviolet (UV) radiation as shown in the following reactions below (2.1-2.3) (Middlebrook *et al*, 2000):

$$O_2 + light \rightarrow O + O \quad \textbf{2.1}$$

$$O + O_2 + M \rightarrow O_3 + M \quad \textbf{2.2}$$

$$Net: 2O_2 + uv \rightarrow O + O_3 \quad \textbf{2.3}$$

The O_3 molecule is produced when UV radiation breaks the bonds of oxygen molecules containing two O_2 atoms in the stratosphere. The single O_2 atom is highly reactive and bonds with another oxygen molecule creating O_3. The stratospheric O_3 plays a crucial role ("good ozone") to the earth life as it is a primary absorber of the solar UV and thus shielding life at the surface from the damaging effects of UV radiation (Gopalapillai 2012).

2.2.2.3 Tropospheric O_3: sources and impact

The tropospheric O_3 is found from the ground and extending up to 15 km vertically. In this region, the O_3 is mainly contributed by two major sources; the first involves the movement of stratospheric O_3 into the atmosphere (stratospheric-tropospheric exchange). The second source is the photochemical production of O_3 due to reactions involving sunlight and O_3 precursor gases including nitrogen oxides (NO_x) and non-methane volatile compounds such as methane (CH_4) or carbon monoxide (CO) (Cooper *et al*., 2014) originating from natural and anthropogenic sources.

The anthropogenic activities are acknowledged to have significant contribution in the total tropospheric O_3 production. (Piketh *et al*., 2006). The transport sector is at the top of the list of the important sources of

O_3 precursors (World Bank Group, 1998) especially in the urban environment and other sources may include industries, domestic as well as biomass burning activities. The chemical reactions (equations 2.4-2.8) below represent the production of O_3 in the troposphere.

$$CO(or \quad VOC) + OH\bullet \rightarrow CO_2 + H\bullet \qquad \textbf{2.4}$$

$$H\bullet + O_2 \rightarrow HO_2\bullet \qquad \textbf{2.5}$$

$$HO_2\bullet + NO \rightarrow NO_2 + OH\bullet \qquad \textbf{2.6}$$

$$NO_2 + h\nu \rightarrow NO + O\bullet \qquad \textbf{2.7}$$

$$O\bullet + O_2 + M \rightarrow O_3 \qquad \textbf{2.8}$$

Where hν is a photon of wavelength, $\lambda \leq$ nm and M is any atmospheric molecule to which the excess produced is transferred. **O●** and **OH●** are the oxygen and hydroxyl radicals.

In reaction (2.4), CO is oxidised by OH● radical (produced from atmospheric oxidation of water vapour) to form carbon dioxide (CO_2) and a free hydrogen atom (H). In (.2.5), H and O_2 combine to form a peroxy radical (HO_2). In reaction (2.6), NO-to-NO_2 conversion occurs as HO_2 is transformed into OH. In (2.7) to (2.8) NO_2 absorbs the energy of a photon light (hν) and dissociate into nitric oxide (NO) and an oxygen radical (O●) and the O● will react with a diatomic O_2 to form the O_3 (2.8)

Tropospheric O_3 plays an important role as it can oxidise some of the greenhouse gases reducing their lifetimes in the atmosphere, during photolysis process, O_3 is broken down resulting in the production of hydroxyl (OH●) (the detergent of the atmosphere) radical. The OH● radical through its oxidizing potential will react with many trace components in the atmosphere (Jöckel *et al*., 2002) transforming them into water-soluble forms which then get removed from the atmosphere through rain. However, on the other hand the tropospheric O_3 is a pollutant (bad ozone) that is harmful to human health and can cause damage to plants and crops (US-EPA 2002). It is also recognised as a powerful greenhouse gas (Wang *et al*., 2008). The residence time of tropospheric O_3 in the atmosphere ranges from a week to a few months depending on the season, altitude as well as the region (Wild *et al*., 2004).

The removal of the tropospheric O_3 primarily depends on the meteorological factors. O_3 can be horizontally advected by wind from one region to another. The main sink of the tropospheric O_3 is

photolysis in the presence of water vapour (Jacob and Winner, 2009). The dry deposition process also plays an important role in the removal of tropospheric O_3. The tropospheric O_3 may be dry deposited into the ground and absorbed by plants. It can also be uplifted by processes associated with convection or frontal system higher up into the free troposphere where its lifetime may be extended (Wild 2004).

2.2.3 Air Quality Modelling and Health Implications

Despite the significant progress in understanding emissions and fate of air pollutants as well as implementing strategies to reduce emissions, air pollution continues to present human health challenges. A substantial body of published scientific literature (e.g. Brown *et al.* (2003); Basham (2001); Bridgman (2000)) has highlighted that exposure to high ambient concentrations of pollutants have the potential for serious adverse human health effects.

SO_2 is among the first pollutants to be considered for causing adverse health effects (Clench-Aas *et al.*, 1995). Studies have revealed that exposure to high level of SO_2 may result into decreased lung functioning for children and increased respiratory symptoms in adults, variability in heart rate (WHO, 2013), causes inflammation of the respiratory tract resulting to coughing and aggravate asthma (COMEAP 2010, 2011). High rates of hospital admission and mortality has been observed during high episodes of SO_2 in some other areas (Brown et al. 2003).

Tropospheric O_3 is considered the second most important criteria pollutant after particulate matter (PM) with respect to health and the third most important greenhouse gas after carbon dioxide and CH_4. Tropospheric O_3 acts as an irritant in the respiratory tract, at high O_3 concentrations, coughing and chest discomfort while breathing may occur, followed by inflammation and narrowing of the airways and reduction in physical performance (World Bank Group, 1998). It is estimated that 0.7 million respiratory deaths globally each year can be attributed to exposure to tropospheric ozone (UNEP, 2012).

As mentioned earlier, there still present challenges to preventing people's exposure to air pollution. Given the types of impacts known to occur as a result of O_3 and SO_2, there is a need to supplement the limited coverage of air quality monitoring networks with mathematical tools that would support the prediction of air pollution at any location on a daily basis. The following section discusses the overview of air quality modelling application and processes.

2.3 Air quality modelling and forecasting

2.3.1 Overview

Air quality modelling and forecasting involves the application of mathematical tools with an attempt to predict or reproduce pollutant concentrations in the atmosphere (Collett and Oduyemi, 1997). This scientific method began early in the 19th century following World War I when scientists tried to estimate the chemical plume concentrations from poison-gas attacks under various wind conditions (Wannberg *et al.*, 2010). Since then, modelling in the air quality field has been evolving with the advancing trend in computing technology in that the simple Gaussian and box models have evolved into more advanced statistical models, Eulerian-grid and Langragian-grid models (Arasa *et al*., 2010). Numerical models have been identified as of critical importance in gaining understanding about the behaviour of various chemical elements and compounds in the atmosphere.

Consequently, models have become useful tools in air quality assessments of source-receptor relationships so that responsibility for specific impacts can be apportioned. Models can also be used as a guide in assessing the existing and future air quality impacts, in understanding the transportation and the variation of air pollutants at various temporal and spatial scales. Air quality modelling also plays a crucial role in determining the location of monitoring stations during the development of a monitoring network. The use of models could further reduce the often costly activity of air quality monitoring (Arasa *et al*., 2010).

Currently a variety of different models exist that range from a simple model to more advanced models (MFE, 2004). Three broad types of models can be categorized based on the purpose of application namely screening models, refined models and advanced models. The first two categories of models are mainly used for regulatory purposes. Screening models are primarily applied to worst case scenarios and have an advantage of using very little input data and have a built-in set of meteorological conditions which may require very little resources and computation time (BC MOE 2008). Most common simple models include the screening version of AERMOD (AESCREEN) (EPA 2011), Toxics Screening Model (TSCREEN) (EPA 1994), Rough Terrain Diffusion Model Version 3.20 (RDTM3.2) (Paine and Egan, 1987), and a screen version of Industrial Source Complex (ISC3) dispersion Model (SCREEN3) (EPA 1995).

Refined models include a more detailed treatment of the atmospheric processes and require more detailed input data and are thus more expertise to run and to interpret the results. These models provide concentration distributions both spatially and temporally. The most commonly known models of this class include AMS/EPA Regulatory Model-AERMOD (Quality and Division 2009) and Atmospheric Dispersion Modelling System-ADMS (CERC 2012).

The third category involves advanced models which allow for the comprehensive treatment of meteorology, emissions and chemistry (BC MOE, 2008). These models employ three dimensional (3D) meteorological fields to drive pollutants in the atmosphere. Advanced models are generally applied for research and forecasting purposes and are grouped into particles, puff and grid-point models. These can be complex Gaussian models, Eulerian or Langragian models, or a combination of these. Figure 2.2 below represents the distribution of the models and their applicability.

The Numerical Atmospheric-dispersion Modelling Environment (NAME III) and the CALPUFF models are examples of advanced models as shown in Figure 2.2.

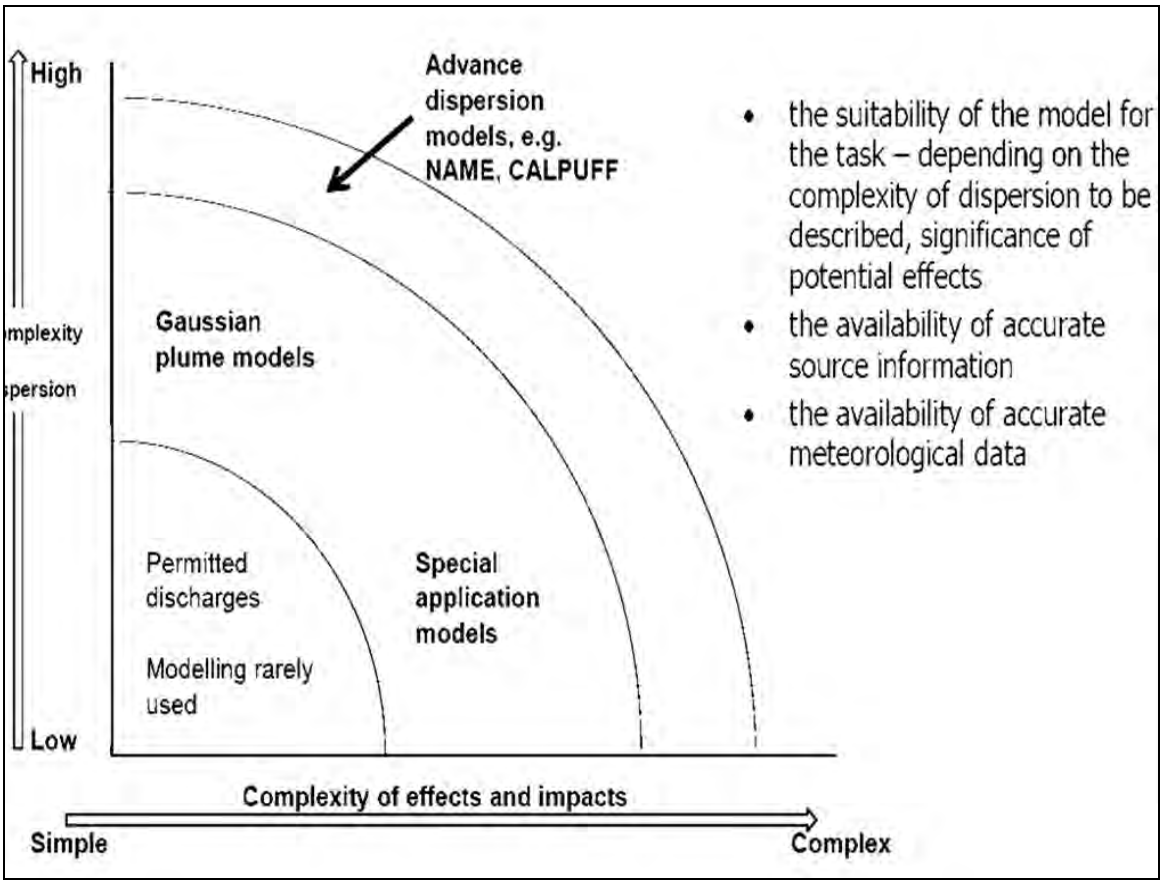


Figure 2.2: Types of models typically applied according to the complexity of the problem (Source: MFE, 2004).

2.4 Key requirements for air quality modelling and forecasting

To predict pollutants concentrations in the atmosphere with air quality model, several processes need to be followed. These include the processing and compilation of emission inventory, prediction of meteorological factors and lastly the verification of model results. The air quality modelling system links the meteorological, emissions and air quality models (see Figure 1.1) to make air quality forecasts. The subsections below provide a description of each of these components.

2.4.1 Emission Inventories

Azkar *et al*. (2012) acknowledge that the emission inventories have long been the primary requirement for air quality modelling effort. The key element for obtaining better results in air quality modelling/forecasting approach therefore lies in the availability of accurate and reliable information on the characteristics of emissions sources.

In order to properly simulate the concentrations of pollutants in the atmosphere the dispersion model requires comprehensive information on the sources responsible for emitting pollutants. The emission inventories play an important role in the accurate prediction of the atmospheric pollutant concentration as they contain detailed information on the emissions profile of the study area. Emission inventories provide comprehensive information on emission sources and fluxes in the area of interest (Zarate, 2007). The inventories are designed in a generic format such that it can be used by various stakeholders for different purposes in the area of atmospheric studies. Two different methods are used in the estimation of emission information, namely the bottom-up approach and top-down approach. In a bottom-up approach, the information is derived from relating the source information (source location and activity rates) to the emission factor of a particular pollutant as proposed by Breu *et al*. (2008) in equation (2.9) below.

$$E = A \times EF\left(\frac{1-ER}{100}\right) \qquad 2.9$$

Where:

E = emissions unit per time

A = activity rate

EF = emission factor, and

ER = overall emission reduction efficiency

The top-down methodology involves inverting measurements in combination with additional information, such as the results of atmospheric transport and transformation models (Benkovitz and Akimoto, 2004). The data are normally stored in a uniform format as is required by most models for chemical simulation exercises.

2.4.2 Meteorological Factors

The fate of gaseous and solid particle pollutants in the atmosphere is a function of meteorology. The meteorological factors (temperature, relative humidity, solar intensity, etc.) determine the transport, and mixing and influence chemistry and deposition of atmospheric pollutants (EPA, 2009a). Though the practice of air quality modelling can be accomplished by the use of limited meteorological variables from monitoring instruments, in most cases the NWP models are the preferred source of meteorological information. The NWP models provide more sophisticated 3D gridded meteorological fields which are normally required by advanced air quality models for better estimation of dispersion and transformation of pollutants. The most important advantage of the NWP models in air quality modelling is the production of meteorological fields (gridded) on a wide variety of time and distance scales and can provide both the surface and upper air meteorological data needed for air quality studies including atmospheric stability and boundary layer parameters.

2.4.3 Validation of modelled results

As much as the numerical (air quality) modelling tools provide a significant contribution in our understanding of complex scientific problems, these are only the representation of the land and atmospheric processes (involving assumptions) and limitations in their performance are expected. Model performance may vary across different regions mostly due to regional differences such as weather, topography and more importantly the emission data. BC MOE (2006) acknowledges that there is no single model that is suitable to handle all geophysical, atmospheric and source situation.

As such, it is required that models are verified and validated for a particular application. Chang and Hanna (2004) acknowledged the importance of this exercise and pointed out the link between air quality models and its importance in decision making that have impacts on public health and economy. The authors also highlighted that models that underestimate pollutant concentration pose a threat to human health and the environment, while models that overestimate the concentration may have serious economic consequences.

In emphasising the value of model validation, the US EPA has developed guidelines that outline the process of model evaluation (EPA, 2009b). The benefit of evaluation exercise includes identification of an individual model's limitations and allowing for continuous improvement of the model. (Sawyer

(2007), Dolwick (2005)) indicate that meteorological input to Air Quality Models (AQMs) may have their own weakness hence need not to be accepted as appropriate for air quality modelling but it is important to understand how their bias and error affect the AQM results.

2.4.4 Air quality modelling outputs and uses

The air quality modelling exercise can help to assess the spatial and temporal distribution of pollutants concentration across domains and also assess the impact of different emission reduction scenarios (San José *et al*., 2013). As mentioned above modelling of air quality is performed at various institutions for air quality and climate research, policy and regulation purpose as well as in air forecasting. The focus of the latter application is mainly to protect the human health from exposure to poor air quality. The air quality forecast is issued at a short term scale where the model output is compared with the air quality guidelines on daily basis and therefore used to create warning systems and plan mitigation actions to severe episodic situations (Baklanov *et al*., 2007). The air quality forecasts are issued and communicated to the public through various media, including television, radio and internet. Figure 2.3 below is an example of the air quality forecast (NO_2) presented to the United Kingdom (UK) community via internet.

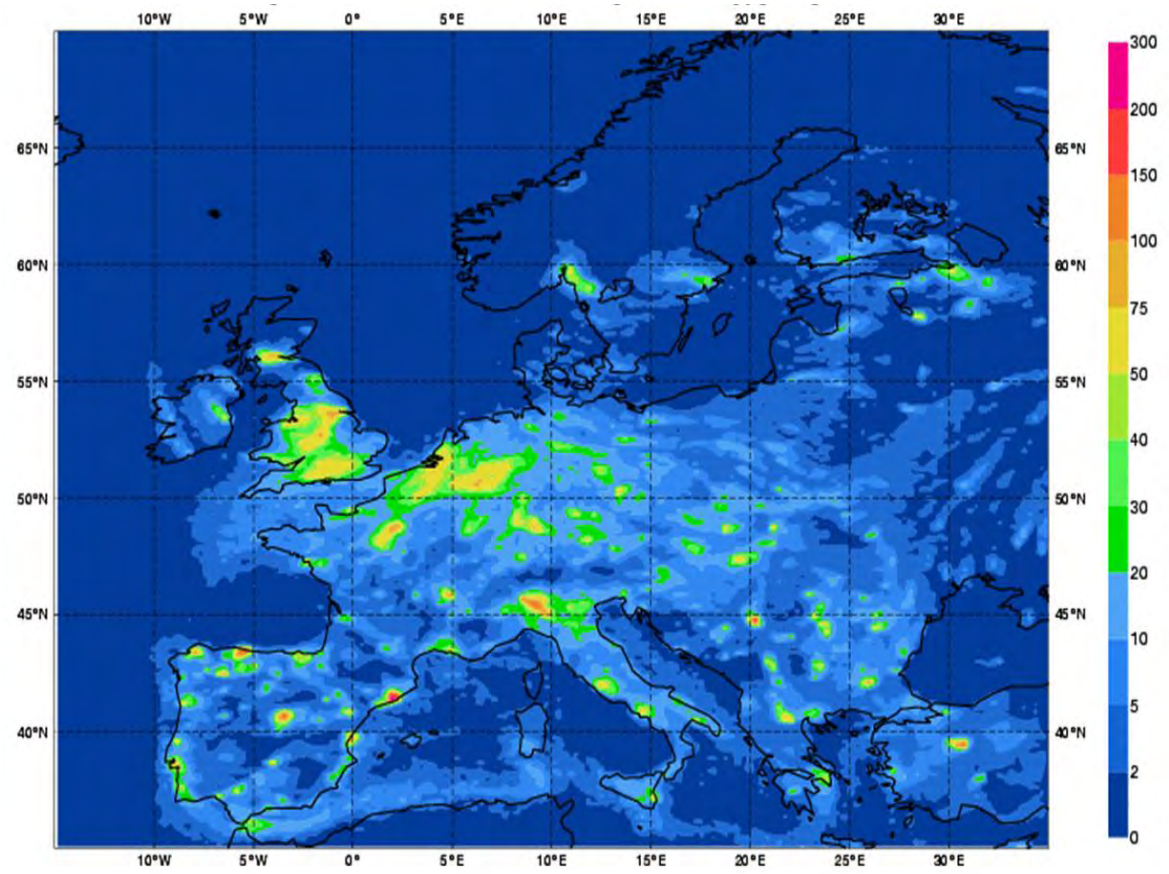


Figure 2.3 Graphical representation of the NO_2 concentration over UK on the 9th October 2009 (Defra, 2014).

This information is presented together with a scale that measure the severity of pollution in the area and a table that describe the relative impact of each band (Table 2.1).

Table 2.1: Information on the recommended action and advices (DEFRA, 2014).

Air Pollution Banding	Value	Accompanying health messages for at-risk individuals*	Accompanying health messages for the general population
Low	1-3	Enjoy your usual outdoor activities.	Enjoy your usual outdoor activities.
Moderate	4-6	Adults and children with lung problems, and adults with heart problems, who experience symptoms, should consider reducing strenuous physical activity, particularly outdoors.	Enjoy your usual outdoor activities.
High	7-9	Adults and children with lung problems, and adults with heart problems, should reduce strenuous physical exertion, particularly outdoors, and particularly if they experience symptoms. People with asthma may find they need to use their reliever inhaler more often. Older people should also reduce physical exertion.	Anyone experiencing discomfort such as sore eyes, cough or sore throat should consider reducing activity, particularly outdoors.
Very High	10	Adults and children with lung problems, adults with heart problems, and older people, should avoid strenuous physical activity. People with asthma may find they need to use their reliever inhaler more often.	Reduce physical exertion, particularly outdoors, especially if you experience symptoms such as cough or sore throat.

The importance of air quality service is also recognized by the World Meteorological Organisation (WMO). The WMO has initiated a project called the GAW Urban Research Meteorology and Environment (GURME) aimed at supporting the development of air quality service programme and the provision of air quality forecasts of urban environment in various countries around the world. This project has resulted in many countries offering air quality forecast services through their respective national programmes, especially in the developed countries. The following section discusses some of the countries with well-established AQMFS.

2.5 Air Quality Services Internationally and in South Africa

Leading national meteorological services (NMS) around the world provide not only weather and climate services to their respective countries, but also air quality services. These services include the provision of meteorological data for dispersion modelling studies, forecasting pollutant concentrations, environmental impact assessments, environmental policy support and emergency planning support in response to environmental incidents. Air quality modelling research is conducted to better describe the dispersion and transportation of air pollutants for both environmental and health impact assessments; to understand trans-boundary pollution contributions to air quality; and for atmospheric chemistry research. Operational air quality forecasting systems predict daily levels of photochemical smog, atmospheric particles and pollutants. These air quality services provide advice that can be used for making scientifically based decisions on environmental questions. Most NMSs provide these air quality services through joint initiatives with government/environmental institutes. A brief review on some of the well-established NMS is provided in the subsections below.

2.5.1 AQMF programs in an international context

2.5.1.1 The Australian Bureau of Meteorology Air Quality Modelling Forecasting System

The Australian Bureau of Meteorology (BOM) in joint partnership with the Commonwealth Scientific Industrial Research Organisation (CSIRO) and Environmental Protection Agency (EPA), developed the Air Quality Forecasting System through the clear air project sponsored by the Australian government (Manni et al., 2012). The Australian Air Quality Forecasting System (AAQFS) generates air quality forecasts twice (09h00 and 15h00) a day for a forecast period of 24 to 36 hours. Two forecast products are provided by the system one for suburb level at a horizontal resolution of a 1 km and for the regional level

at 5 km horizontal resolution. The predicted pollutants include NO_x, O_3, SO_2, CO and particulate matter (both PM_{10} and $PM_{2.5}$), as well as air toxics (e.g. benzene) (Manni et al., 2012). The AAQFS is an offline system and consists of five major components namely; the BOM's NWP model called the Limited Area Prediction System (LAPS), the Emission Inventory Module which provide calculation of emissions of a range of pollutants including CO, NO_x, PM_{10}, SO_2, VOC, etc. The emissions module takes into account different sources such as point sources, area sources, motor vehicles, biogenic sources, prescribed burning and wildfires, sea salt aerosol and wind-blown dust. The third component of the system is the Chemical transport Model (CTM), a multiscale Eulerian regional model allows for calculating the chemical reaction of different pollutants in the atmosphere, the last two modules involve evaluation and data archiving and display (Cope *et al*., 2004). The design of the complete system is given in the diagram (2.3) below:

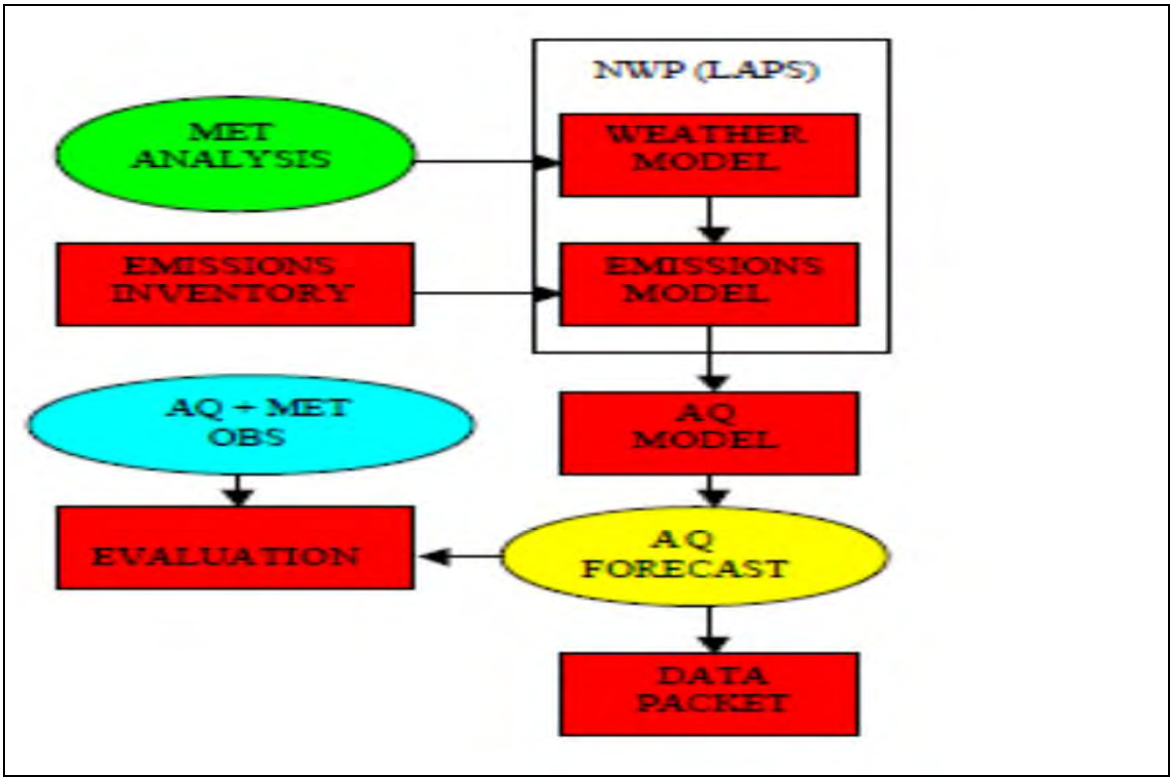


Figure 2.4 Schematic diagram of the Australian Air Quality Forecasting System (Manni et al., 2012).

2.5.1.2 The Norwegian Meteorological Institute: Urban Air Quality Information and Forecasting System (UAQIFS)

The Norwegian Meteorological Institute in partnership with the Norwegian Road Authority and the Norwegian Institute for air Research-NILU use the AirQUIS (Air Quality Information System) dispersion model to provide an air quality forecast for the Norway cities. The air quality forecasts are produced during high air pollution episode in winter from the October to April. The pollutants of concern include particulate matter (both PM_{10} and $PM_{2.5}$) and NO_2 (Gjerstad et al. 2005). The forecast products are presented on the Web as text and graphs as shown in Figure 2.5 below.

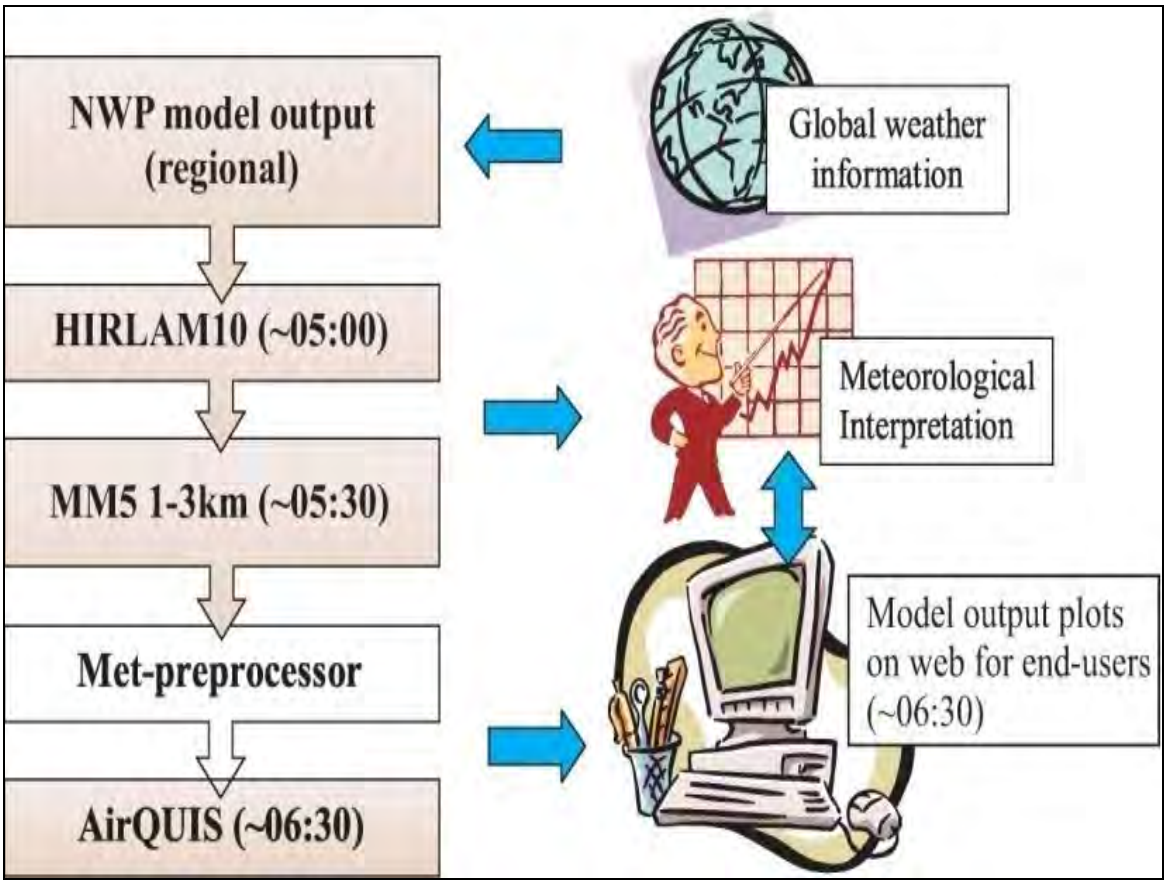


Figure 2.5 Schematic diagram of the of the Norwegian air quality forecasting System (Gjerstad *et al.*, 2005).

2.5.1.3 China: Air Quality Monitoring and Forecasting (AMFIC) System

In addressing the air quality challenges in China, the government of China collaborated with a number of different stakeholders within and outside the China to develop an integrated information system for monitoring and forecasting tropospheric pollutants over China. The collaboration is composed of various meteorological institutions including KNMI in Netherlands, BIRA-IASB and VITO in Belgium, DUTH, NOA LAP-AUTH all from Greece, the FMI in Finland, IFE at the University of Bremen in Germany and as well as the National Satellite Meteorological Center and the Institute of Atmospheric Physics (IAP-CAS) both from China. The system is known as the Air Quality Monitoring and Forecasting In China (AMFIC) (Van der A, 2008). AMFIC uses satellite and in situ air quality measurements and modelling to generate consistent air quality information over China.

The forecasting system is composed of the Chimere chemical transport model, the NWP data from the European Center for Medium-Range Weather Forecasts (ECMWF) and the emission data from the Intercontinental Chemical Transport Experiment-Phase B (INTEX-B) and it provides 2 day air quality forecast for the eastern region of china (van der A, 2008).

2.5.2 Air Quality modelling/forecasting in the southern African region

In the southern African region activities related to air quality services, especially air quality modelling and forecasting are lacking (Zunckel *et al*., 2007). The southern African region is composed of seven countries i.e Botswana, Malawi, Tanzania, Swaziland, South Africa, Zambia and Zimbabwe. However a scoping report from the Air Pollution Network for Africa (APINA) reveals that in the whole region, South Africa and Tanzania are the only two countries that have a relatively strong body of researchers with an interest in air quality modelling (Zunckel *et al*., 2007). Moreover, none of the countries in the region has a well-established air quality program that include capability on modelling and forecasting (as well as issuing of warnings) of air quality. Most of the NMS within this region have a strong focus on meteorological modelling.

The review of other forecasting systems and the lack thereof from a southern African context highlights the importance of this work beginning in this study for South Africa. As such the next section in this chapter focuses on the air pollution challenges in South Africa, as well as the legislative context for air quality modelling and forecasting with respect to ambient air quality standards.

2.6 Air pollution in South Africa

2.6.1 Background to air pollution in South Africa

As a developing country, South Africa has experienced rapid economic growth during last two decades. Unfortunately, strong economic growth often leads to environmental problems. South Africa has the most industrialized economies in southern Africa and is the sole industrial energy producer in the region (Josipovic, 2009). However, industrial development is accompanied by an extensive increase in air pollution. Consequently, in South Africa air pollution challenges are confined to industrial centres like the Vaal Triangle, the eastern Highveld region, Richard's Bay industrial zone, South Durban, Milnerton in Cape Town, Rustenburg/Bojanalo, and Waterberg. These areas are now known as air pollution hotspots (Nuwarinda, 2007) due to the geographical distributions of both industries and population. According to the most recent State of the Environment Report, the quality of ambient air is decreasing in South Africa due to continued increases in air pollution, particularly in urban areas and highly industrialised areas (DEAT, 2005).

2.6.1.1 Air pollution sources in South Africa

Studies of air pollution, its sources and sinks within the southern African boarders have been conducted and it has been suggested (e.g. Scorgie, 2012) that in South Africa fossil fuel burning activities are a significant source of emissions. These include the burning of fossil fuel for domestic, transport and industrial use and more importantly for generation of power (Scorgie, 2012). These sources are categorized as key due to the fact that they are active almost throughout the year. The Department of Environmental Affairs (DEA) acknowledges that energy generation and industrial emission accounts for a large amount of atmospheric emissions (DEA, 2011).

2.6.2 Climatological Controls on Air Pollution in the Region

Significant efforts have been made in studying the climatology of the South African region and the information is well documented (Tyson *et al.* (1996); Jury (2013); Archer *et al*. (2010); Kane (2009)). The study on air transport climatology for subtropical southern Africa by Tyson *et al*. (1996) revealed that in the southern African context, the general atmospheric circulation is dominated by four major synoptic

scale circulation types, occurring in different frequencies throughout the year (Figure 2.6). The most important of these is the semi-permanent, subtropical continental anticyclones, which dominate 70% of the time during winter and 20% of the time in summer. This leads to the establishment of extremely stable atmospheric conditions which can persist at various levels in the atmosphere for long periods. Other systems include the transient mid-latitude ridging anticyclones which show little annual variation and occur with a frequency of 10-15%, the westerly baroclinic disturbances made of rossby waves and cut-off lows with a maximum of 40% observed in spring and lastly the easterly tropical disturbances, these are more frequent during summer and their occurrence reaches maximum of 60% in January and a minimum of less than 5% in July (Tyson and Garstang, 1996).

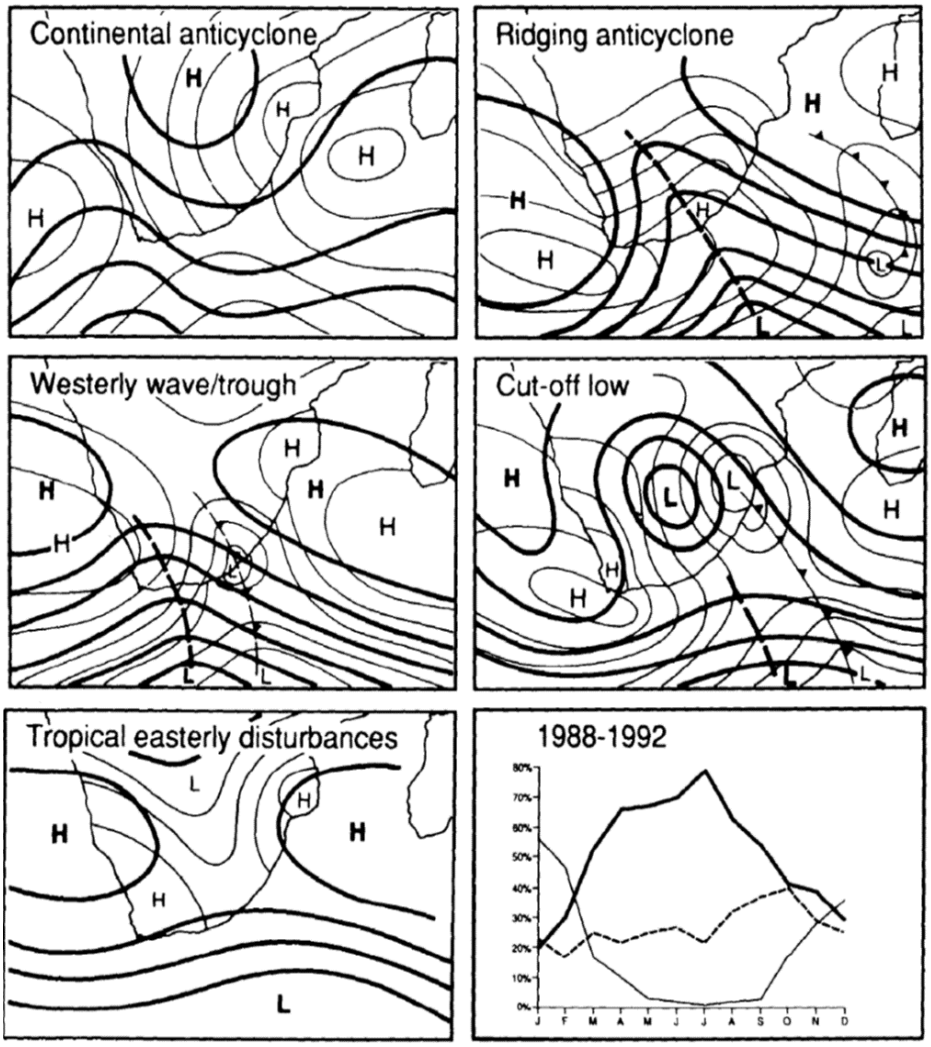


Figure 2.6 Major Synoptic circulation types affecting southern Africa and their monthly frequencies of occurrence over a five year period (after Preston-Whyte and Tyson 1988 and Garstang *et al*., 1996).

Over the Highveld itself, the region is generally dominated by dry conditions with scant annual rainfall, north-westerly winds in summer and westerly winds in winter. The general flow pattern is anti-cyclonic. This circulation is more intense during winter than in summer (Figure 2.6), resulting in frequent occurrence of atmospheric inversion layer which may vary between few hundred metres deep at night to about one or two thousand metres during the day (Tyson *et al*, 1988). During summer when the anti-cyclonic belt is shifted further south of the region, the moist, unstable conditions (conducive for rainfall) are experienced in this sub-region.

2.7 Regulatory Control: National Ambient Air Quality Standards

In striving to protect health and the environment from the air pollution challenges, the World Health Organisation (WHO) has issued guidelines for air quality management and a number of countries and states have since issued regulations. Since 1965 pollution control in South Africa was framed by the Atmospheric Pollution Prevention Act (No. 45 of 1965) (APPA). The APPA's approach was to control emissions at the source by imposing emission limits on large sources. There was no mention of air concentrations, standards, or guidelines, nor was there any explicit recognition in policy instruments of any principle of exposure to people or the environment.

Since 2006 there has been a significant shift in South Africa's approach in tackling air pollution problems following the implementation of the new National Environmental Management: Air Quality Act (No.39 of 2004) (AQA) in 2004 (Gurannah, 2011).

The AQA updated concepts of the South African approach to air quality control and follows international best practice, to include:

- The establishment of national norms and standards for the receiving environment.
- A framework for air quality management planning and reporting regime and numerous regulatory instruments for the control of air pollution and compliance and enforcement.
- It further clarifies the roles and responsibilities of all the different spheres of government in addressing air pollution problems.

The success towards implementation of various regulations within the AQA has been visible in many key areas in South Africa, these include but are not limited to: the publication of air quality governance guidelines by national government, declaration of air quality management priority areas, establishment of the South African Air Quality Information System (SAAQIS) and more importantly the development and publication of National Ambient Air Standards (NAAQS). The NAAQS provide a means to regulate pollutants identified as a priority for air quality management in the country. A brief discussion of criteria pollutants, with particular emphasis on the two criteria pollutants which are the main focus of the study is provided in following sub-section.

2.7.1 Criteria Pollutants

Currently there are eight (SO_2, PM_{10}, O_3, CO, NO_2, Pb, $PM_{2.5}$, C_6H_6) criteria pollutants that are the focus of air quality management in South Africa.

The NAAQS are the legally binding health risk threshold aiming to protect human health due to exposure to pollutants within the living environment. They comprise of a set of requirements namely; threshold level, margin of tolerance, permissible frequencies of exceedance and timeframe for achieving compliance with the threshold. Tables 2.1 and 2.3 below show the air quality Standards of SO_2 and O_3.

Table 2.2: National Ambient Air Quality Standards for Sulphur Dioxide (SO_2)

Averaging Period	**Concentration**	**Frequency of Exceedance**	**Compliance Date**
10 minutes	500 grams/m^3 (191 ppb)	526	Immediate
1 hour	350 grams/m^3 (134 ppb)	88	Immediate
24 hour	125 gram/m^3 (48 ppb)	4	Immediate
1 year	50 micrograms/m^3 (19 ppb)	0	Immediate

Table 2.3: National Ambient Air quality Standards for Ozone (O_3)

Averaging Period	Concentration	Frequency of Exceedance	Compliance Date
8 hours	120 gram/m^3 (61 ppb)	11	Immediate

2.7.2 Characterisation of emissions sources in the country

The quantification of emission source contributions shows that power generation contributes the largest amount of the SO_2 and NO_2 total emissions and industrial activity contribute largest amount of the total PM_{10} emissions (DEA 2009). Other secondary sources include agricultural and wildfires due to their seasonal nature (Scorgie 2012). These sources are responsible for air pollution in many parts of South Africa. Even though this information is known there is only sparse information on a national emissions inventory in South Africa as historically little effort has been made in developing a comprehensive, transparent and accurate national inventory of criteria non-greenhouse gases.

Consequently, regional long range transport models rely mostly on Global Emission Inventory Activity (GEIA) databases (e.g. Zunckel *et al*., 2006). The major challenge with these types of databases is that emissions are of low resolution and they are estimated using little information available. On the other hand it is important to acknowledge that some of the governmental and non-profit organizations have been able to develop their own local high resolution emissions inventories, especially for municipalities like Cape Town and eThekwini, Richard's Bay Industrial Area, and DEA database for certificates for scheduled processes. The major shortcoming with these emissions inventories is that they are not accessible and are developed based mostly on overseas emission factors e.g. from United States Environmental Protection Agency (US EPA).

It has been proposed that in future the South African Air Quality Information System (SAAQIS) will extend its capacity to include the NAEIS as mentioned above. The NAEIS database will provide information of emissions on a national scale. The system will serve as a national emission inventory and is expected to be implemented early in 2015. It is proposed that system will currently provide annual emissions data from the listed industries. Emissions from other (non-listed) sources such as Energy,

Agriculture, Forestry and other Land Use and Waste will be estimated in or outside NAEIS depending on the emission source type (DEA, 2013).

The emissions of nitrogen oxides (NO_x) from the Highveld region account for 91% of South Africa's total emissions. More than 60% of these NO_x emissions originate from eight large coal fired power plants and a synthetic fuel industry. The NO_x may increase tropospheric O_3 concentrations (Scholes and Villiers, 2007) in the presence of VOC and CO, with CO as an important O_3 precursor in a South African context (Lourens *et al*., 2011). As such the levels of high O_3 concentration are observed in the Highveld region (e.g. Martins *et al*., 2007) and are often accompanied by the high levels of CO that are associated with the occurrence of veld fires events during winter and spring seasons (Lourens et al. 2011). Furthermore, a country wide study revealed that approximately 20% of national particulate emissions results from biomass burning activities (Wagner *et al*., 2005). The recent quantification of regional air pollution by satellite (GOME-2 instrument) observations showed that elevated concentrations of particulates and trace gases are dominant over the Highveld region (ESA, 2013). Figure 2.4 below represents the tropospheric NO_2 concentration over South Africa as monitored by the GOME-2 instrument.

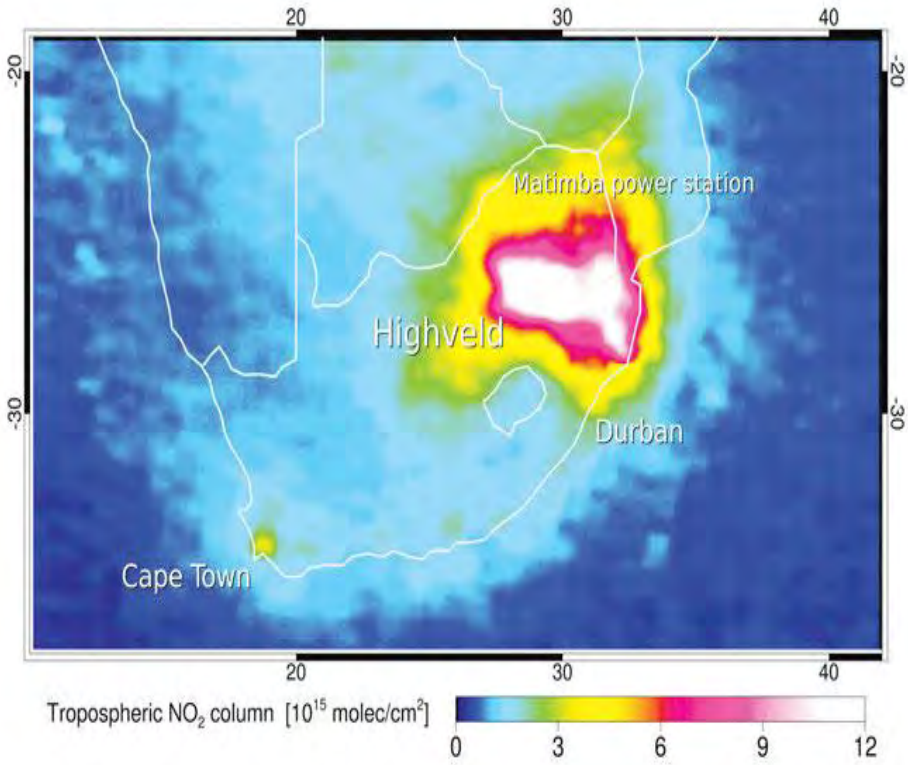


Figure 2.7: Graphical representation of tropospheric NO_2 over South Africa, (ESA 2013)

2.7.3 Air quality monitoring and forecasting capacity in South Africa

In support of the national ambient air quality standards in partnership with the DEA, SAWS is hosting SAAQIS. The objectives of SAAQIS are to provide relevant air quality information for informed decision-making and efficient air quality management. SAAQIS will gather emission databases of trace gases (NO_x, SO_2, CO, etc.), GHG and persistent organic pollutants (POPs). The information on the system will play a crucial part in climate change adaptation measures and compliance with international treaties such as the United Nations Forum on Climate Change Conventions (UNFCCC) and the Montreal protocol. Furthermore, research aircraft campaigns by SAWS are used to investigate atmospheric pollutants, in particular over air quality hot spots of South Africa, like the Cape Town, Johannesburg and eThekwini metropolis and the Highveld. A much better understanding of the air quality over these hotspots has been gained through the use of the research aircraft in recent years.

Notwithstanding the air quality services SAWS is currently providing in support of the AQA, there is a recognised need to establish an air quality modelling and forecasting system for public good services. There are no comprehensive air quality dispersion modelling and forecasting systems for trace atmospheric species important for air quality and climate in South Africa. Statutory functions of SAWS place the organisation in a position to sustain and extend such air quality services country-wide. To date, air quality modelling is conducted mainly by private consultants and a handful of municipalities like Cape Town, EThekwini and Johannesburg. The modelling is also limited in spatial extent and confined to specific time frames like the winter seasons when strong air pollution episodes are usually anticipated.

2.8 Summary

From the above discussion, it can be concluded that ozone is generally found between two atmospheric regions, the stratosphere and the troposphere. The stratospheric ozone occurs naturally in the stratosphere. The tropospheric O_3results from the transportation of the stratospheric ozone to the troposphere through various atmospheric processes and also produced in the troposphere when the mixture of nitrogen oxides and other gases are exposed to sunlight. In the latter, the sunlight causes NO_2 to release oxygen radical that combines with O_2, to produce O_3. The colourless SO_2 is primarily the result of anthropogenic sources. The anthropogenic activities results in high level concentrations of the of air pollutants which poses risk

to human health. The O_3 and SO_2 are associated with high rates of hospital visits and death. The air quality modelling tools are the integral part of addressing the air pollution problem, such tools can provide guidance in the development of effective air quality management policies. Moreover, there is a growing interest of using air quality models in reducing human exposure to poor air quality, which is especially relevant in countries such as South Africa that have limited air quality monitoring.

CHAPTER 3: DATA AND METHODS

3.1 Introduction

The model performance testing exercise requires a comparison between model predicted trace gas concentrations and actual measured values. The most important requirements for an air quality modelling task are meteorological, topography and emissions data as well as the chemistry scheme for allowing calculation of chemical reactions. For this study, the UM global meteorological and topography data as well as the IPCC emission information were used to drive the NAME III model. The STOCHEM chemistry model is linked with the NAME model. As the input data used in this study were of a coarse resolution, it was set to produce monthly averages of pollutant concentrations and the model results were compared with the corresponding monthly averaged monitored data. The output of this study may not be used for comparison with the NAAQS, however the information may help in understanding the temporal and spatial distribution of the modelled pollutants over South Africa, which in turn can provide a picture of the location and contribution of the emission sources. This chapter provides a description of the NAME III modelling tool, detailed information on the meteorological and emissions data used, as well as the description on the monitoring network used for comparison with modelling results.

3.2 The Third Generation of Numerical Atmospheric Dispersion Modelling Environment (NAME III)

The earlier version of NAME III was designed to simulate the transport of airborne pollutants following the Chernobyl accident in 1986. Since then the model has been improving and expanding its role from one version to another until the present version called NAME III version 6.2.

The NAME III model is a complete Lagrangian system, designed to predict simple trajectories to complex dispersion and deposition of gases and particulates by releasing a large number of particles into a three dimensional atmosphere (Redington and Wbebster, 2000). The model employs 3D meteorological fields provided by a grid-point atmospheric general circulation (Unified Model). The UKMO NWP model, Unified Model (UM) is in daily operation at SAWS. The modelling environment that is provided by the

NAME III model is spatially and temporally flexible as it can be configured to model spatial scales from the kilometre (km) range to the global scale and in a temporal range from minutes to years.

The chemistry component of the NAME III model is based on the STOCHEM scheme where more than 30 predominant species are represented (Jones *et al*., 2007). Both gaseous and aqueous phase chemistry and sulphate and nitrate chemistry can be modelled. The OH, HO_2 (hydroperoxyl radical) H_2O_2 (hydrogen peroxide) and O_3 background concentrations are initialised using monthly mean 3D background fields. The primary pollutants (SO_2, NO and NH_4) that are emitted on particles and secondary species that form are added to the total. Removal of particles from the model atmosphere is achieved through gravitational fallout, surface impaction, washout by falling precipitation, and rainout, where the pollutant is absorbed directly into cloud droplets as they form (Jones *et al*., 2007b). Figure (3.1) below represents the main processes involved in the NAME III model.

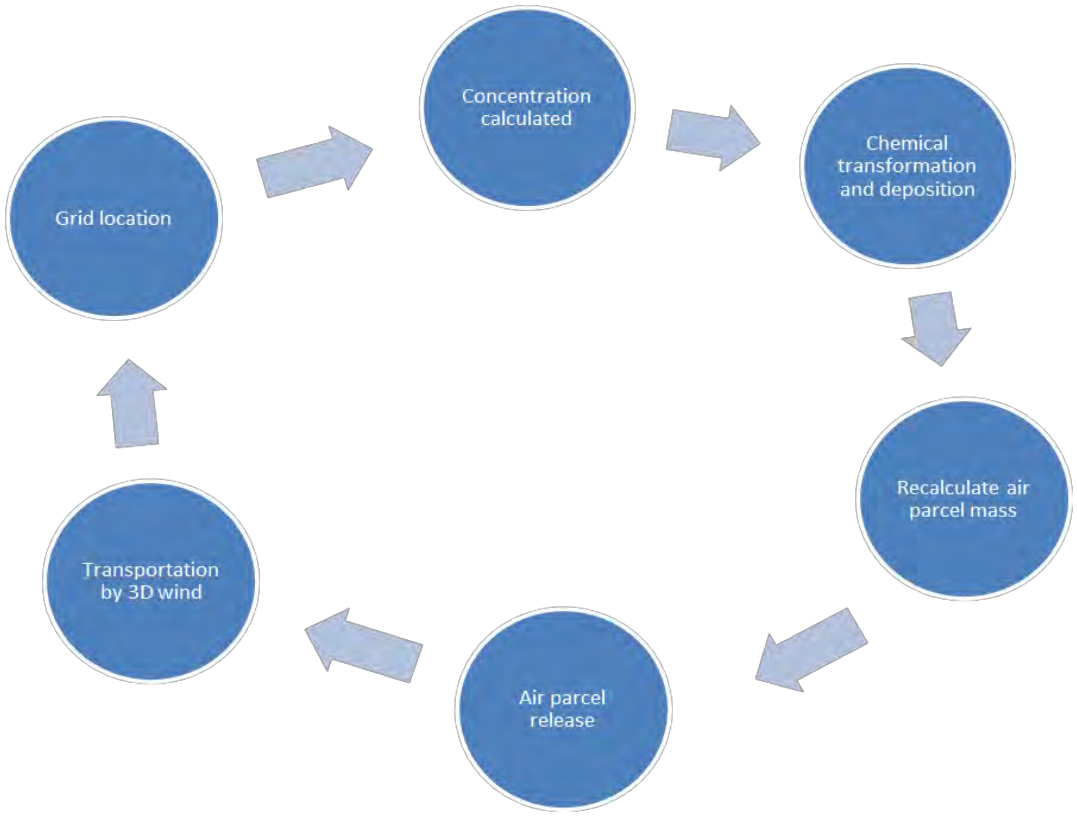


Figure 3.1 Diagram summarising the NAME III Main Process

The NAME III model has been tried and tested in many cases. In a campaign to evaluate the model performance, Jones *et al*. (2005) has validated the NAME III model output against the experimental data

set and it was found that the model compare satisfactory with the observations. The NAME III has also been used in research studies. Ryall *et al*. (2002) applied NAME III to reveal long-range dispersion of Sahara dust over the UK, the model results were found to be correlated to ground base measurements and to the satellite images. Redington and Derwent (2002) tested the skill of the NAME III model in estimating annual average concentrations of secondary pollutants in the atmosphere and the authors found that the results obtained compared favourably with maps derived from both observed data and that of the European Monitoring and Evaluation Program (EMEP).

Moreover a large volume of published work has also been conducted with the NAME III application these include and not limited to: The origin of high particulate concentrations over the United Kingdom (Ryall et al. 2002), here authors found that on some days, long range transport of primary PM_{10} from mainland Europe impacted air quality over London. In studying the impacts of Russian biomass burning on UK air quality this study revealed that episodes of biomass burning occurring in Russia can affect UK air quality (Witham *et al*., 2007). The model was also used to study emission distributions and country contributions of ozone depleting substances across Europe (Manning *et al*., 2003).

Until recently and before the establishment of the Air Quality in the Unified Model (AQUM), the NAME III was previously used at the UKMO for operational air pollution forecasting and related air quality services. The air quality forecast was issued twice daily to the UK community (Jones, 2004). However with the introduction of the online AQUM (Savage et al., 2012), NAME III is now used to provide advice in all areas linked to the atmospheric dispersion (e.g emergency response on accidental release of radioactive material from nuclear industries) to various stakeholders in the UK.

3.3 Meteorological and Topography inputs

A reliable and easily accessible meteorological input data must be available for any modelling exercise. Arciszewska and McClatchey (2001) highlight that the meteorological input is the critical component of air quality modelling. Arya (1999) also emphasises that the quality of the dispersion model strongly depends on the quality of meteorological input. The meteorological factors such as wind, temperature, atmospheric pressure, humidity, precipitation as well as observing cloud cover can be measured using meteorological instruments.

A number of the meteorological stations are available in South Africa and a large portion (about 228) of them are owned and managed by SAWS and the rest by other government departments and other stakeholders. These are distributed nation-wide to measure temperature, humidity, rainfall, sunshine and pressure. The data are reported every 5 minutes to the SAWS climate database. However, data from these stations are very sparse and limited to certain parameters. The dispersion models require more meteorological information than those provided by the stations, as an example, sophisticated dispersion models like NAME III requires the information about boundary layer height as a function of time but this information cannot be obtained from the Automatic Weather Stations (AWS). Other model requirements include the surface stress, soil moisture and all this information may not be recorded at the AWS stations but can be estimated by using appropriate scientific equations. However such calculations cannot be simply done by hand, the alternative, more timeous approach of calculating and obtain this information is through the NWP models. These tools solve complex mathematical equations to predict the possible present and future state of atmospheric weather. The NWP models are often used to provide meteorological information where there is a lack of monitoring systems. As a result the NWP models are applied in many air quality studies to provide information on the general state of the atmosphere in the region of interest.

In this study, the meteorological data utilized for inputs in the NAME III model were obtained directly from the global (40^0 X 40^0 km) NWP UM. It is acknowledged (Davis and Dacre, 2009) that higher resolution NWP data normally produce results of good quality in air quality dispersion. The high resolution (12 km) regional UM is used at SAWS for operational weather forecasting, however this model is currently not configured to produce meteorological parameters that are required in NAME III, hence the lower resolution global data from the UKMO were used. The section below provides a brief discussion on the UM.

3.3.1 Unified Model

The UM is a well-established and internationally recognized prognostic model. It is a weather and climate modelling tool which is used across a wide range of spatial and temporal scales, from a short-range weather forecasting at 1.5 km resolution to multi-decadal simulation in a global configuration. The UM dynamical core is non-hydrostatic and fully compressible and does not apply the shallow atmosphere approximation approach (Savage 2013). A semi-implicit, semi-langragian predictor scheme is used to

solve the non-hydrostatic atmospheric equations on a staggered Arakawa C-grid (Savage et al. 2012). The boundary layer processes are represented by the non-local, first order closure (Lock 2007).

The topography effect is represented by the use of height based hybrid coordinate system, Charney Phillips grid staggering, the coordinate is a terrain following sigma near the surface. In this study, the archived UM NWP data for the period of 2010 to 2011 was used as input to the NAME III model. The frequency of the data is three hourly with a spatial horizontal resolution of approximately 40 by 40 km. The basic meteorological (input) data required for NAME III include wind (speed and direction), air temperature (10 m temperature was used) and cloud cover, the temporal scale of the input data can range between seconds to multi years. The complete meteorological input data set used in this study by NAME III is listed in Table 3.1 below. The topographical data are also provided from the UM, so as to ensure that the topographical with land cover information and meteorological values correspond.

Table 3.1: List of meteorological input parameters for the NAME III model (Witham et al. 2012)

Parameter	**Parameter**	**Parameter**
Wind (u-component)	Temperature (K)	Roughness length (m)
Wind (v-component),	Convective cloud amount (0-1)	Boundary layer depth (m)
Wind (w-component),	Convective cloud base height (m)	Roughness length (m)
Temperature (K),	Convective cloud top height (m)	Soil moisture in layer (kg/m^2)
Specific humidity (kg/kg),	Dynamic rain rate ($kg/(m^2\ s)$)	Canopy height (m)
Dynamic cloud liquid water (kg/kg),	Convective rain rate ($kg/(m^2\ s)$)	Canopy water (kg/m^2)
Dynamic cloud ice (kg/kg),	Dynamic snow rate ($kg/(m^2\ s)$)	Stomatal conductance (m/s)
Pressure (Pa),	Convective snow rate ($kg/(m^2\ s)$)	Surface stress (v-component) (N/m^2),
Surface stress (u-component) (N/m^2),	Dynamic high cloud amount (0-1)	Surface sensible heat flux (w/m^2),
Sea level pressure (Pa)	Dynamic medium cloud amount (0-1)	
Pressure (Pa)	Dynamic low cloud mount	

3.3.2 Emissions inventory

For this study NAME III requires a detailed information on the existing emissions sources for it to successfully predict the concentration of tracers in the air. The required information includes the geographical location of a pollution source, the temperature, the height and more importantly the rate at which the polluting material is released.

In South Africa there is no comprehensive national emission inventories, a number of groups and research studies (e.g. SAFARI 2000, Richards Bay Clean Air Association (RBCAA), Bellville South, EThekwini Municipality, Cape Town and the Department of Environmental Affairs database for certificates of scheduled processes) have compiled local emissions inventories for various studies with specific purposes (Zunckel *et al*., 2007). However obtaining such emissions information is often a challenging task as there is currently a lack of transparency making sharing of this information limited to certain parties. In particular the industrial emissions data are treated as highly confidential information and is difficult to obtain in South Africa. The new NAEIS being developed for SAAQIS is aimed at enhancing the development of a transparent, accurate and comprehensive emission inventory for South Africa.

Given those circumstances, the available global (0.5^0 X 0.5^0) IPCC database of the Fifth Assessment Report (AR5) was used to derive South African emissions. The IPCC AR5 database is available to the public via the internet (ftp://ftp-ipcc.fz-juelich.de/pub/emissions/gridded_netcdf).

The IPCC emissions dataset has been used in several regional to global studies, (Bouarar et al. 2011) for example used the IPCC AR5 to examine the influence of different emissions on the distribution of surface ozone over Equatorial Africa during the summer mansoon. Together with the DEA industrial emission database, Zunckel *et al*. (2000) applied the IPCC global emission inventory to study the transportation and deposition of sulphur over the southern Africa region.

3.3.3 IPCC AR5 inventory

The IPCC AR5 inventory is constructed from monthly mean estimates for various sectors and species covering the period 1850 to 2000. In this study, the latest (year 2000) emission scenarios were used. The data are composite for various existing global inventories and harmonised according to sector and

regional temporal trends. The horizontal resolution is 0.5º X 0.5º degrees. The emissions inventory includes pollutants such as SO_2, NO, NO_2, NH_3, CO, O_3, Sulphate, OH, particulate matter (PM_{10} and $PM_{2.5}$) and a number of Volatile Organic Compounds (VOC) e.g. toluene, xylene, benzene, formaldehyde, etc. The sources of emissions include anthropogenic sources (mobile and stationary sources) and biomass burning as well as natural sources such biogenic sources, volcanic emissions, oceanic and dust emission (IPCC, 2009). Figure 3.2 is the representation of the regional (southern Africa) CO emission for the month of July in 2000 extracted from the IPCC global emission database.

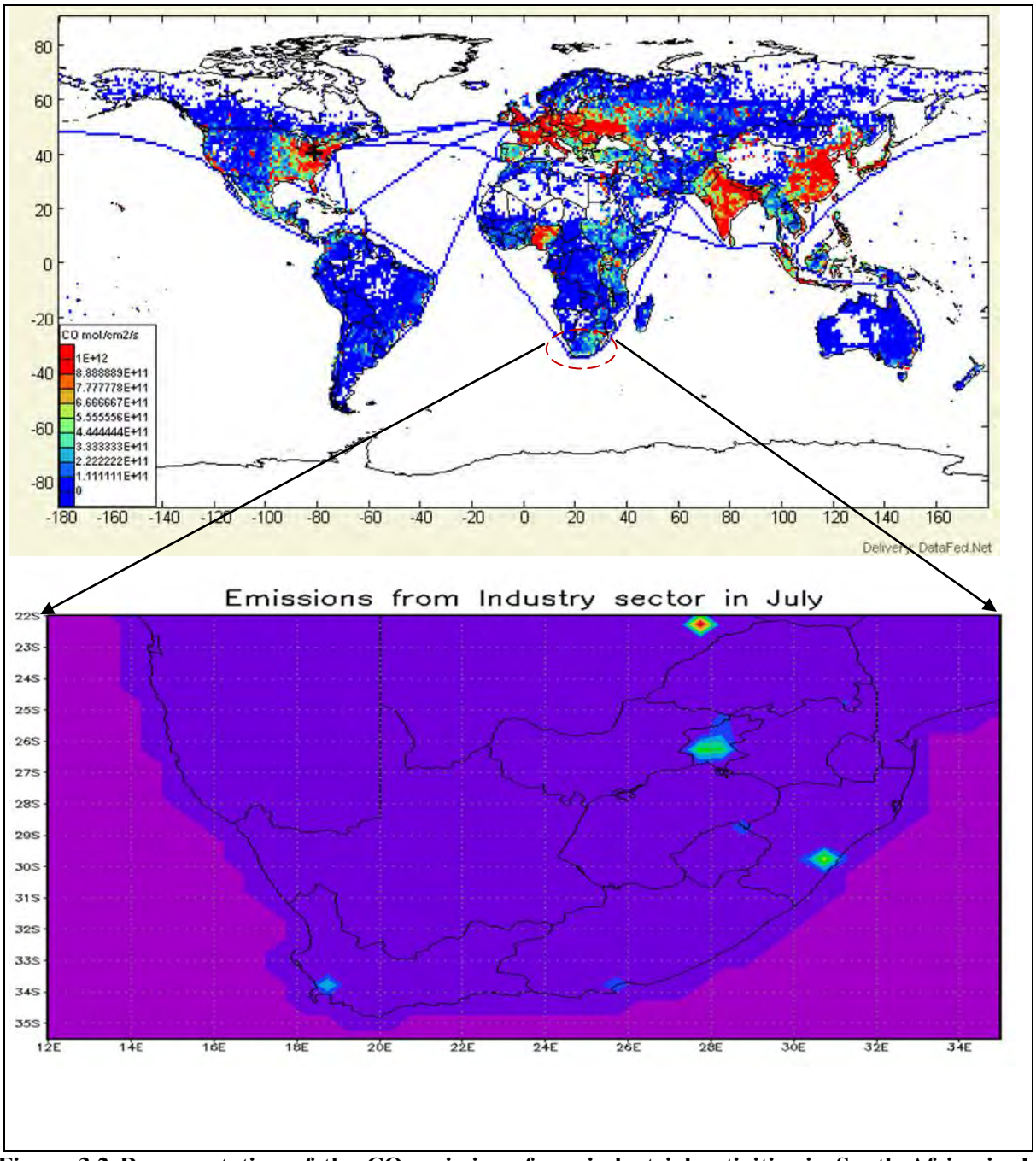


Figure 3.2 Representation of the CO emissions from industrial activities in South Africa in July 2000 (IPCC).

Some drawbacks were involved in using the IPCC emission inventory for this study that included the low spatial resolution of emission data compared to that of the NAME III. The spatial resolution of the IPCC emission inventory is about 0.5^0 x 0.5^0, whereas the NAME III domain was set up with 0.11^0 X 10.11^0. To minimize this mismatch of spatial resolution, the original data were interpolated to a 0.11^0 X 0.11^0

resolution to be at common spatial resolution with the model. The IPCC emissions data are monthly averages that are not adjusted to daily or hourly variations, however, this was adjusted to a finer temporal scale as outlined below.

In this study the original IPCC emission data were gridded to a 0.11^0 X 0.11^0 horizontal grid from the 0.5^0 X 0.5^0 using the Pingo interpolation method (Schulzweida *et al*., 2007) and the monthly emission rates in each grid cell were reduced to a gram/seconds by simple division, assuming that no changes in emission rate occur over a month. Lightning and ship emissions were not included in this study as it was assumed that the contribution from these sources will have negligible impacts on the results (Aghedo *et al*., 2007). It was also assumed that no changes occurred in the emission rate since the last two decades. It is important to mention that a number of sources (e.g. area, line, and point sources) can be defined in NAME III (Maryon *et al*.,1999) but in this study, the final data set was represented as a non-stop stationary point sources (operating 24 hours 7 days a week) emissions and given geographical location in latitudes and longitudes. This was chosen because the case study area of interest, which is the Highveld is concentrated with industries (major contributors) which operate non-stop throughout the year so it is expected that large volume of emissions is from industrial operations. However it is acknowledged that this assumption may not be valid in a real environment especially for transport and domestic emission sources which have diurnal and seasonal variations.

3.4 Study area description

The South African Highveld region is associated with poor air quality, and was declared by the South African government as an air pollution priority area in 2007. The HPA (latitude 25.2^0 to 27.2^0 S and 27.2^0 to 29.2^0 E) covers 31 106 km^2, and includes parts of the Gauteng and Mpumalanga Provinces (Figure 3.3). It is composed of a metropolitan municipality, three district municipalities and nine local municipalities and incorporates a population of approximately 3 596 891 with the largest portion (71%) concentrated in Gauteng (Zunckel *et al.* 2010). The economy within the eastern Highveld area is based primarily on agriculture, mining industry and forestry. Although domestic fossil fuel burning activities are the practical means of energy source for space heating and cooking within this area, the major contribution to the wider atmospheric pollution in the HPA is by industrial activities, specifically coal fired power plants, open-cast coal mines, fuel oil refining and petrochemical plants (Igbafe, 2008).

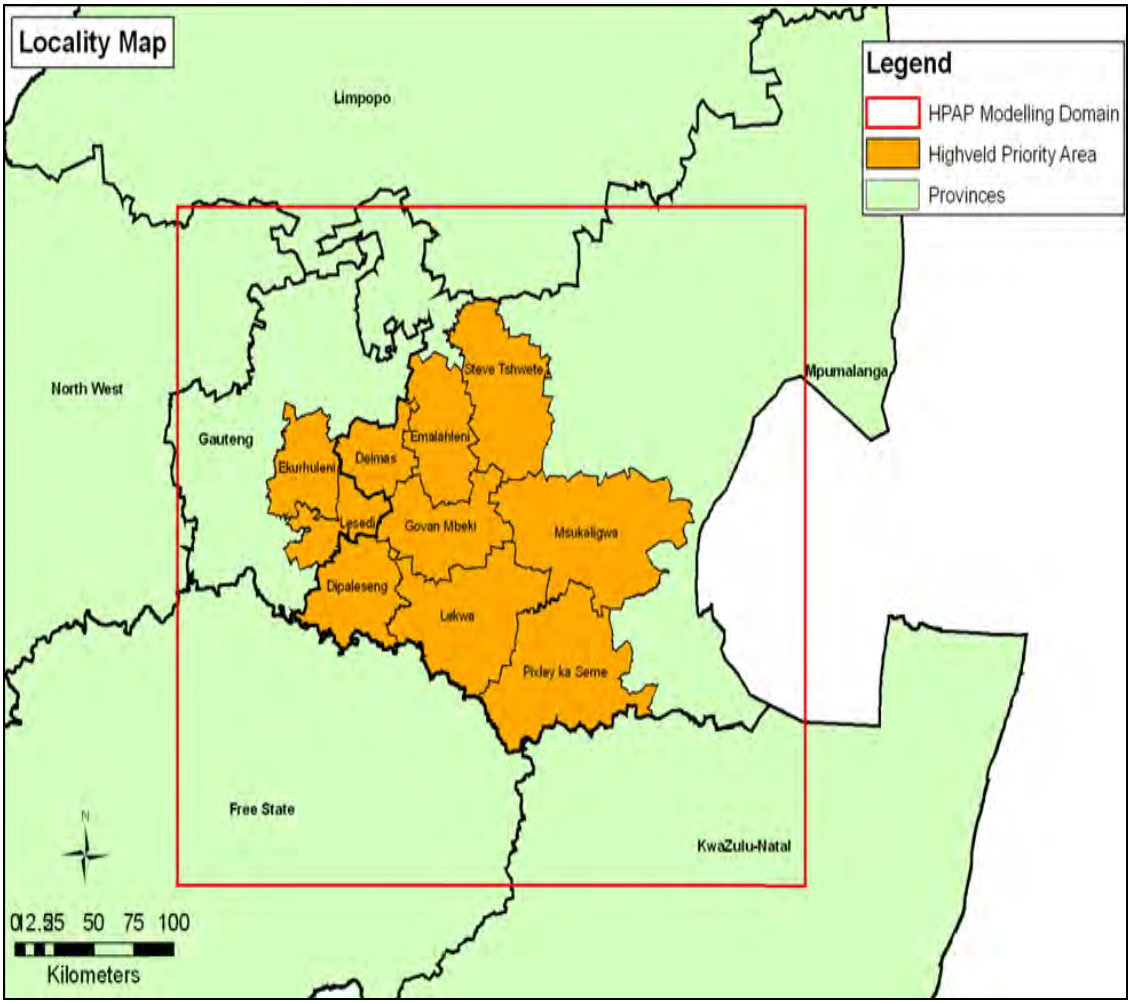


Figure 3.3 Map showing relative location of the Highveld Priority Area (Zunckel *et al*, 2010).

The meteorology of the HPA possesses similar characteristics as that of the southern African region. The Highveld of South Africa experiences a frequent anti-cyclonic climate condition, especially in winter, this inhibits the vertical air masses exchange and stratifies the troposphere into persistent layers in which the residence times of pollutants are prolonged from several days to weeks over this area (Fourie *et al*., 1983). On the other hand the baroclinic westerly disturbances are active during the winter months, hence relative improvement in air quality is also experienced owing to the passage of the occasional occurrence of wintertime westerly flows which brings clean and mostly maritime air over this area (Zunckel *et al*., 2010). While the summer months are associated with strong vertical mixing and development of thundershowers which result in the washout of atmospheric pollutants.

3.4.1 Air quality Monitoring Network

Different types of ambient air monitoring methods are available, and these may include passive samplers which can be used in more remote areas because they do not require power supply. Passive samplers are cost effective, light and reusable. In South Africa, a passive sampling network was introduced during an initiative of the IDAF-International Global Atmospheric Chemistry, Deposition of Biogeochemical Trace Species (IGAC DEBITS Africa) program to determine atmospheric composition deposition of key gaseous and aerosol species over the African continent (Adon et al. 2010). An alternative sampling method involves automated stationary instruments which continuously measure air quality at a specific site. The latter is a preferred technique as it provides real time data and the system is capable of monitoring more than one pollutant concurrently (LIBERTI, 1975). Figure 3.4 below is a photographic image showing an example of the automated monitoring station (picture taken during site visit).

Figure 3.4: Photographic image showing an automated air quality monitoring station at Witbank in the HPA.

However, this technique is expensive as designing of the monitoring network is usually determined by the available budget (Hardy *et al*., 2001). In South Africa there is a significant number of automated monitoring networks distributed around the country (over 100 government owned stations).

Zunckel *et al*. (2006) developed a comprehensive overview on the current air quality monitoring stations in South Africa. Specifically it was reported that monitoring services are conducted mainly by metropolitan councils and industry hence the monitoring stations are mostly located in or close to the major urban areas and industrial developments. SO_2, NO_x, O_3 and PM_{10} are the most commonly monitored air pollutants. Figure 3.4 shows the location of SO_2 monitoring sites countrywide. Although this figure only represents SO_2 monitoring sites, the distribution of stations is representative for other criteria pollutants like PM_{10}, O_3 and NO_x (Zunckel *et al*., 2006).

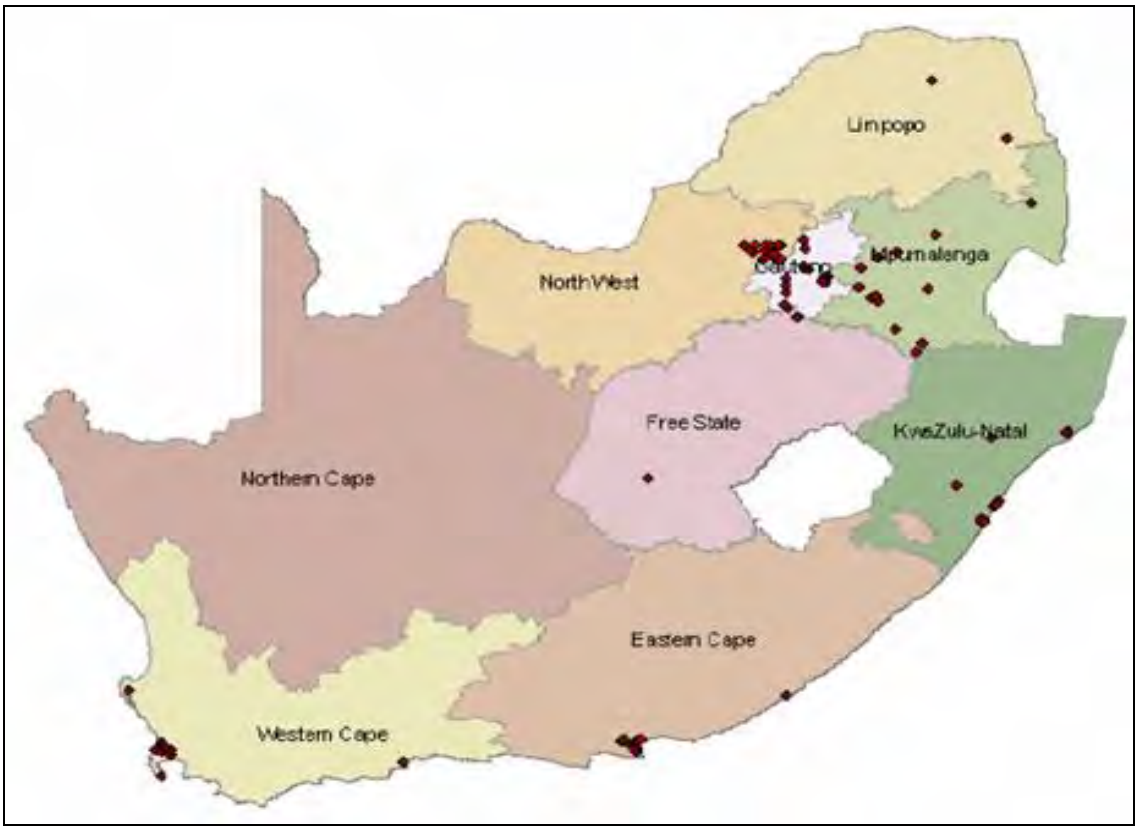


Figure 3.4: Overview of relative location of air quality monitoring in South Africa (Zunckel *et al*., 2006).

The HPA network (Figure 3.5) is one of the monitoring networks that have been in existence for some time (since 2007) and is thought to provide efficient data as the measured data are quality controlled and managed by the SAWS through the SAAQIS. The data from this network are available for public use. This study employs the historical air quality data from the HPA monitoring network for a comparison of model output with these measurements. The Highveld monitoring network consists of 5 sites that are strategically placed in Witbank, Middelburg, Ermelo, Hendrina and Secunda. For all these stations, chemical species including PM_{10}, $PM_{2.5}$, SO_2, NO, NO_2, O_3, SO_2, CO and BTEX are monitored.

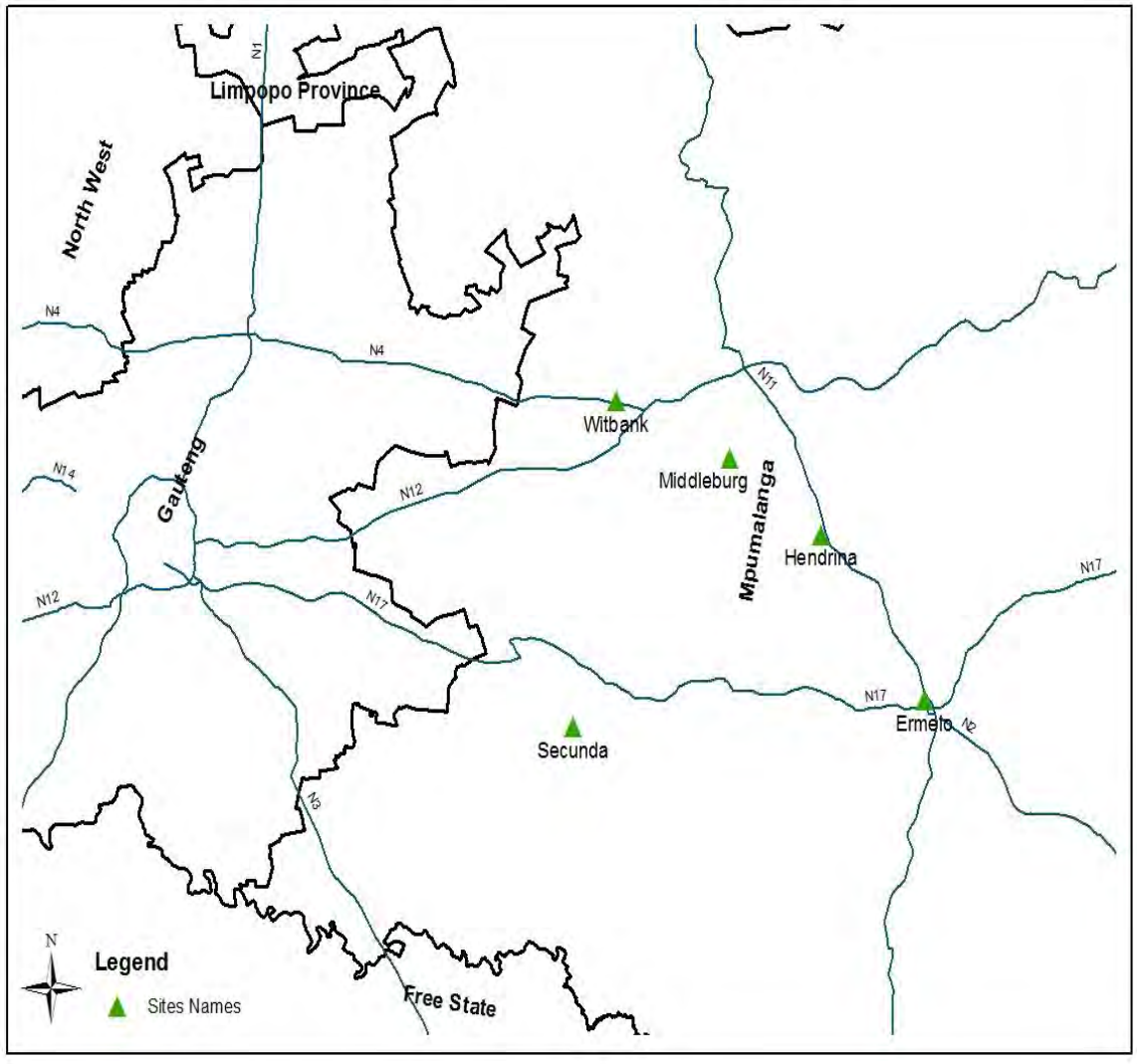


Figure 3.5: Graphical representation of the location of air quality monitoring stations in the Mpumalanga Highveld.

Brief descriptions of each of these monitoring sites is provided below.

3.4.1.1 Witbank

The SAWS Witbank site is located at the Elukhanyisweni Secondary school in Lynnville (25.877812^0 S 29.188664^0 E). The site was selected by the DEA to monitor ambient pollutants resulting from industrial and mining activity located to the north and south within a 5km radius. Other potential sources of atmospheric pollutants include informal settlements 5km to the west, industrial activities within 5km to the north east, and two highways, the N4 500m to the south east and the N12 6km to the south east. It is expected that the major sources of ambient air pollution in the region are mining and industry, while domestic fuel burning and transportation play an important role at certain times (DEAT, 2009).

3.4.1.2 Middelburg

This SAWS monitoring station is located at Middelburg Christian School (25.9944^0 S 29.4733^0 E) at an altitude of 1515m. It is in the proximity of large industrial sources such as Columbus Steel and Middelburg Ferrochrome. Pollution concerns in the region include elevated PM concentrations from industrial sources and mine dumps to the north-west and south. It is not expected that domestic burning is a major local contributing source in this region although there are some low income residential areas in the vicinity of the monitoring site (DEAT, 2009).

3.4.1.3 Secunda

Secunda is a town in the HPA. The main industry in Secunda is the SASOL plant, which converts coal to liquid fuel through the Fischer-Tropsch process. It is therefore expected that industrial emissions will have a significant impact on the air quality in the town. The DEA Monitoring station is located in the Embalenhle Township (26.548578^0 S 29.080055^0 E). The SASOL plant is situated 6.5km to the south east of the monitoring station, the Winkelhoek ash dump is located 3.5km to the north east while other ash dumps are located 2.5km to the south-east. There are informal settlements to the east and low income communities to the west and south of the monitoring station (all within a distance of 2km) and these are likely to be sources of PM_{10} and SO_2 in the region (DEAT 2009).

3.4.1.4 Hendrina

The monitoring station at Hendrina is located at a school in the suburb of Kwazamokuhle (26.1522^0 S 29.7025^0 E) There are formal houses to the east and south of the monitoring station and coal mines approximately 15km to the north and north-west (DEAT 2009).

3.4.1.5 Ermelo

The monitoring site at Ermelo is located at 26.49348^0 S and 29.96002^0 E in the suburb of Wesselton which is in the northern part of the town. There are low cost formal houses to the south of the monitoring station and informal settlements to the north-west and west. The airport is situated to the east while the closest point on the national highway N11 is located 200m to the north-east. There is coal mining activity 2.5km to the north and north-east (DEAT 2009).

3.5 Comparison method

Chang and Hanna (2004) propose that atmospheric dispersion model can be evaluated scientifically, statistically or operationally. For scientific analysis technique the accuracy of mathematical formulas, the physics assumptions and chemistry is examined. Statistical analysis is based on direct comparison of model predictions against actual observations and operational approach concerns issues related to the user-friendliness of the model, output format and features such as error checking. For this study, the statistical evaluation approach was used. This approach includes a set of statistical parameters, namely the Bias, normalised mean bias (NMB), the geometric mean bias (MG), Root Mean Square Error (RMSE), normalized mean square error (NMSE), the geometric variance (VG) the correlation coefficient (R) and the fraction of predictions within a factor of two of the corresponding observations. However in this study only four (4) of this parameters were used namely Bias, NMB, and RMSE as well as NRMSE and detailed information of each of these parameters is provided below.

The statistical parameters recommended by Chang *et al.* (2004) are described below.

3.5.1 Bias and normalised bias

The bias represents a consistent difference between the modelled and observed values. The bias between the modelled concentrations and the observed is a testing parameter used by (Malcom and Manning, 2001; Kang *et al.*, 2005; Kang *et al.,* 2007; Denby *et al*., 2010) and the formula is (which is equivalent to the sum of the error):

$$Bias = C_p - C_o \qquad 3.1$$

Where,

C_p is the concentration of the predicted value

C_o is the concentration of the observed values

This difference is dependent on the magnitude of the concentrations. This is sometimes problematic since the concentration magnitude is likely to change between measurement periods or locations; therefore a number of normalised versions of the bias value have been used, including the Normalised Mean Bias (NMB) (Kang *et al.,* 2005; Denby *et al.,* 2010). The NMB is expressed as:

$$NMB = \frac{1}{n}\sum_{i=1}^{n}\left(\frac{P_i - O_i}{O_i}\right) \qquad 3.2$$

Where n is the size of the sample

P is the predicted value

O is the observed value

A value of zero indicates perfect agreement between the model and the observations.

3.5.2 Root Mean Squared Error (RMSE) and Normalised RMSE

The RMSE is a statistical test that reveals the precision of the model, in other words how close the model projection is to the observed concentrations. This technique has been used in (Malcom and Manning, 2001; Kang *et al.*, 2005; Kang *et al.,* 2007; Denby *et al*., 2010). The RMSE can be calculated for a time series or array of data points as

$$\boldsymbol{RMSE} = \sqrt{\frac{\sum_{i-1}^{n}(\boldsymbol{C_P} - \boldsymbol{C_O})^2}{\boldsymbol{n}}} \qquad 3.3$$

And is equivalent to

$$\boldsymbol{RMSE} = \sqrt{\boldsymbol{BIAS^2} + \boldsymbol{STDDEV^2}} \qquad 3.4$$

Where:

n is the number of observations

STDDEV is the standard deviation

A problem with using the RMSE is that the value is influenced by the magnitude of the concentrations. This makes comparison between different compounds, seasons or regions difficult. A way to circumvent this problem is to normalise the RMSE, to the range of the observed and modelled data to produce the Normalised Root Mean Squared Error Metric (NRMSE) (Denby *et al*., 2010).

$$NRMSE = \frac{(RMSE^2)}{(\overline{C_O} * \overline{C_P})} \quad 3.5$$

Where,

$$\overline{C_P}$$

is the average of the predicted values and

$$\bar{C}_o$$

is the average of the observed values

The ideal value would be a zero which would indicate perfect agreement between the model and the observations.

CHAPTER 4: RESULTS AND DISCUSSION

The NAME III-global (NWP and emission) data air quality modelling system predictions of SO_2 and O_3 concentrations were compared against the ambient air quality data from the Mpumalanga Highveld monitoring network. The model was run for twelve months from the 1st of April 2010 to the 31st of March 2011 at a temporal scale of three months per run. As such, a separate model run was performed for each season, Autumn (March to May), Winter (June to August), Spring (September to November) and Summer (December to February). For each run, the first five days were used to initialise the model except for the autumn season where there was a lack of NWP data in March. Monthly averaged concentrations of 24 pollutants (SO_2, CO, NO, NO_2, O_3, PM_{10}, sulphate, NH_4SO_2, NH_4NO_3, NH_4, NO_3, N_2O_5, HNO_3, HCHO, C_2H_4, C_3H_6, C_5H_8, oxyl, toluene , butadiene, CH_3CHO, peroxy acetyl nitrate , HONO) were produced for each season. However only SO_2 and O_3 concentrations were considered for further evaluation in this study. The level of agreement of model predictions with observations was evaluated using statistical analysis of the monthly average SO_2 and O_3 concentrations and is presented below.

4.1 Monthly variation of measured and predicted concentrations

A large volume of literature (eg. K. S. Collett, Piketh *et al*., 2010, Igbafe, 2008) have discussed the variation and transportation of air pollution in the Mpumalanga Highveld domain. These studies have shown that the Highveld area normally serves as a source of pollution exported to other regions and sometimes serve as a sink of its own polluted air through the recirculation process that is linked to the semi-permanent high pressure system on the eastern part of the country. The regular occurrence of surface inversion enhances the accumulation of air pollutants in this region especially in winter. Figure 4.1 – 4.8 shows the seasonal variation of the predicted and measured monthly averages of the O_3 and SO_2 concentrations at the five monitoring sites during 2010 to 2011.

4.1.1 Overview of observations

The data analysis of the observed monthly averages of SO_2 concentrations shows a seasonal dependence with higher SO_2 concentration levels occurring in the winter months and low concentrations in summer. The seasonal pattern might be attributed to the frequent inversion layer which is often shallower in winter

resulting in the accumulation of pollutants concentrations and generally higher in summer allowing for efficient mixing and better dispersal of pollutants. In addition to this, the use of fossil fuel for domestic purposes (space heating) in this region may also contribute to the high concentration levels during winter. Among all the monitoring stations, the highest monthly average SO_2 concentration appears in Ermelo, especially in winter months. The monthly SO_2 average concentrations vary between 2.93 - 27.73 $\mu g/m^3$ and 3.02 - 34.13 $\mu g/m^3$ in summer and winter. The high concentration levels in winter may be attributed to domestic coal combustion. The station is closely situated in the low income communities and during winter, the local people use coal as their source of energy for space heating and cooking. The Middelburg monitoring station measured the lowest monthly average concentration levels of SO_2 of all the stations (Figures 4.1(e), 4.3(e), 4.5(e), 4.7(e)). The low concentrations in this area may result from the fact that little domestic biomass burning activities are experienced within this area as discussed in the literature above.

On the other hand, the measured O_3 concentrations show relatively high peaks during the spring and summer seasons, particularly in September (46.17 $\mu g/m^3$) and February (53.96 $\mu g/m^3$) months at Ermelo (Figure. 4.5(c)) and Secunda (Figure. 4.8(a)) stations respectively. These O_3 values are above the threshold value of 40 $\mu g/m^3$ for which plants exposed to such concentration levels could be damaged (Zunckel *et al*., 2006). This appears to be associated with the warmer temperatures (in the presence of O_3 precursors) during these seasons favouring for the production of O_3. As was the case for SO_2, the Middelburg station measured the lowest concentrations of O_3 of all the stations in almost all seasons.

The observed data therefore shows that typically high concentration values of SO_2 occur during winter seasons and low SO_2 concentration values were recorded in summer whereas the O_3 concentrations were high during spring and summer months. These findings are in good agreement with previous studies found in the literature for O_3 (Zunckel *et al*,. 2004) and SO_2 (Scott and Diab, 2000) climatology over South Africa. The following section focuses on the comparison of the model data to that of observations.

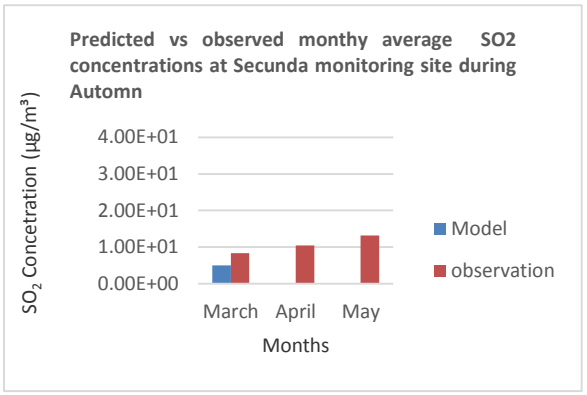
4.1.2 Model-observation data comparison

The highest peak of measured SO_2 concentrations was observed in Witbank during the first month (June) of winter and the interesting part of this analysis is that the model captured almost the same peak at the same place but a month later (July). A similar behavior was observed in Redington and Webster (2000) when the NAME III was used to investigate an SO_2 episode in the UK. The study found that the NAME

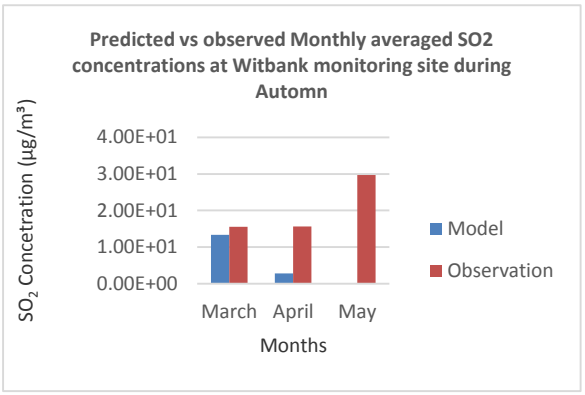
III model captured a peak that was similar to observation concentrations but at a delayed time. The delayed model peak was associated with the model settings where the model was set to account for the buoyancy effect (plume rise scheme) hence allowing the vertical transportation and dilution of the plume. However, in this study the model plume rise scheme was not considered.

For O_3, the highest peak measured was observed in Secunda during the last month (February) of the summer season and another significant high concentration peak was observed in Ermelo during spring (Figure 4.8(a) and left of second row in Figure 4.6 (c) respectively). In contrast to this, observed O_3 concentrations in winter were slightly lower (below 35 $\mu g/m^3$ on average) at all monitoring sites except for Secunda in July. On the other hand the predicted O_3concentrations are lower than the observed concentrations in almost all cases except for the autumn months (specifically May). Overall the modelling system under-predicts the concentrations of SO_2 and O_3 though the modelling results are in the same order of magnitude with the observations at all the stations. A number of scenarios were observed where the model values are in close agreement with observation these include concentrations of O_3; at Secunda during spring (Figure 4.6(a)), at Ermelo and Hendrina both during summer (Figures 4.8(c) and 4.8(d)), concentrations of SO_2; at Witbank during summer (Figure 4.7(b)).

It should be noted that the uncertainty may have arisen from the model input data including the low resolution meteorological information and the emission inventory. The emission inventory used may be incomplete and is out dated as it does not account for the local incidences such as domestic combustion, new developments and land-use changes, the phasing out of high octane fuel as well as changes in industrial operations (e.g changes in technology). It is also possible that the model setting and assumptions inherit therein may have impacted the results. For example the use of a reduced number of model particles to represent the pollutants mass in the model domain could have resulted in the under-estimated concentration values. The reduced number of particles was used to allow the model to run smoothly since the initial model testing with a recommended (large) number of particles resulted in computer failure due to limited available space.



(a)



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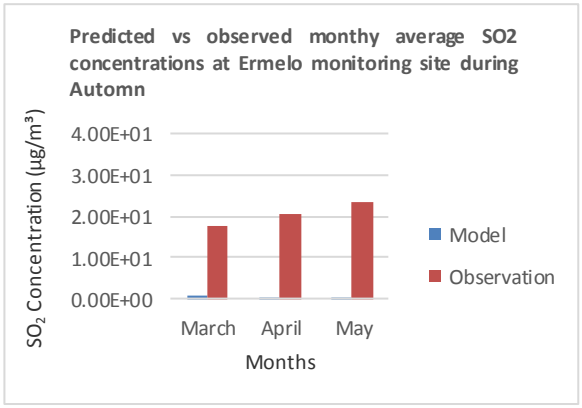
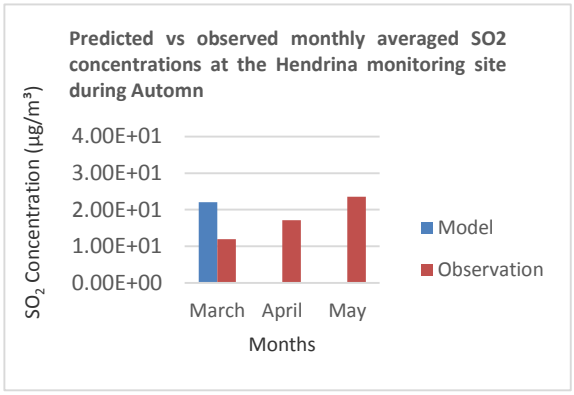
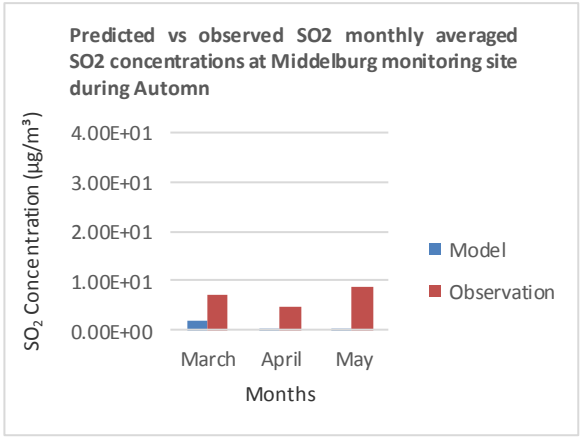


(c)

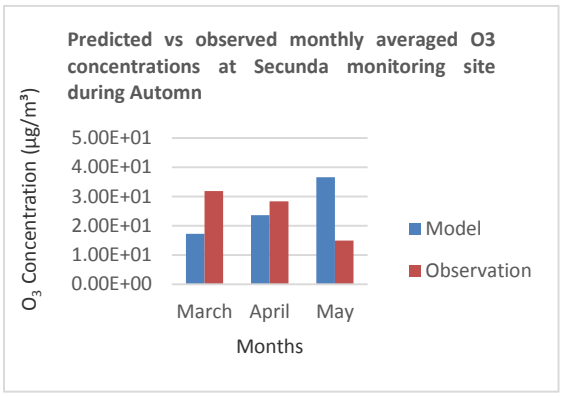


(d)

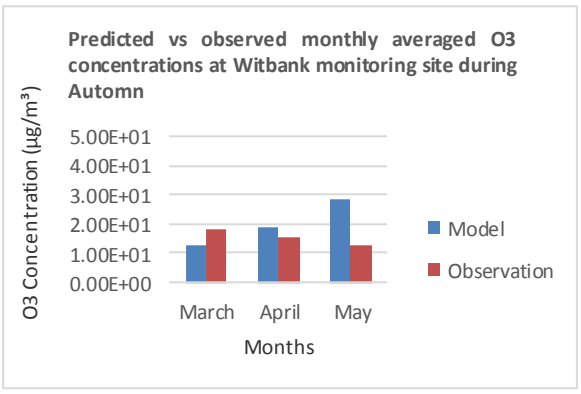


(e)

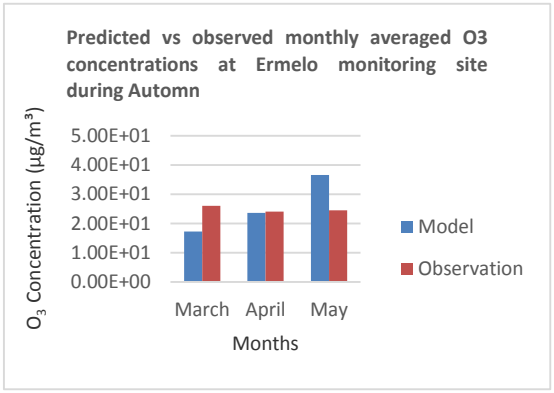
Figure 4.1: Bar graphs representing modelled vs measured monthly averaged SO_2 concentrations at five monitoring sites in the Mpumalanga Highveld during March-May 2010.



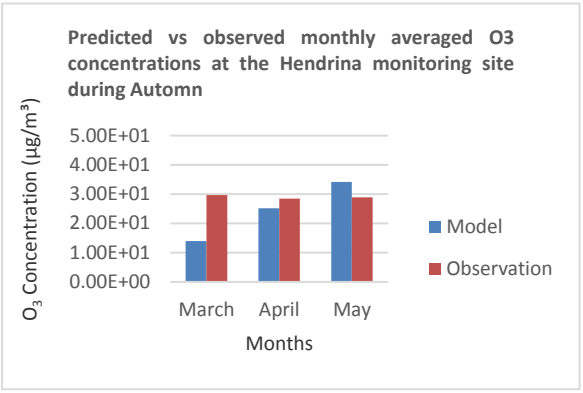
(a)



(b)



(c)



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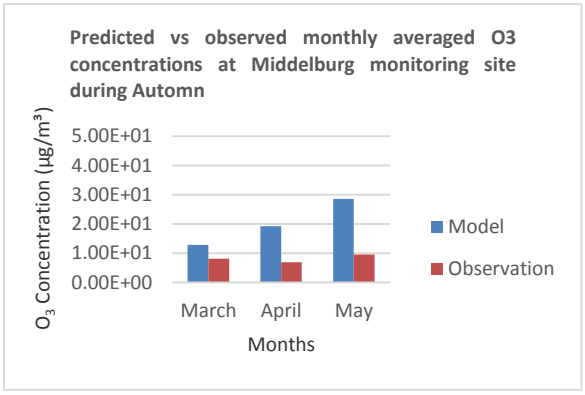
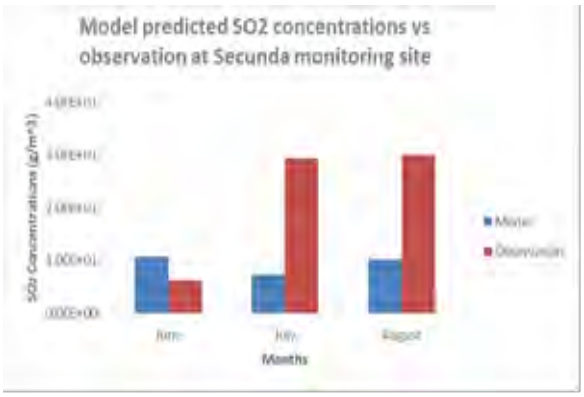
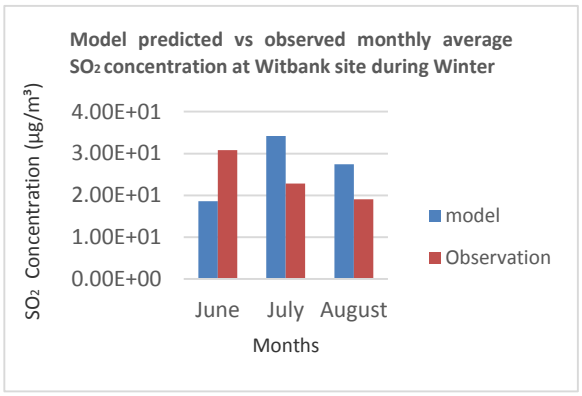


(e)

Figure 4.2: Bar graphs representing modelled vs measured O_3 concentrations at five monitoring sites in the Mpumalanga Highveld during March-May2010



(a)



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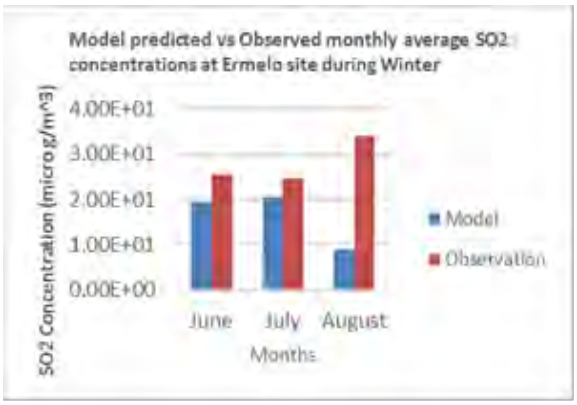
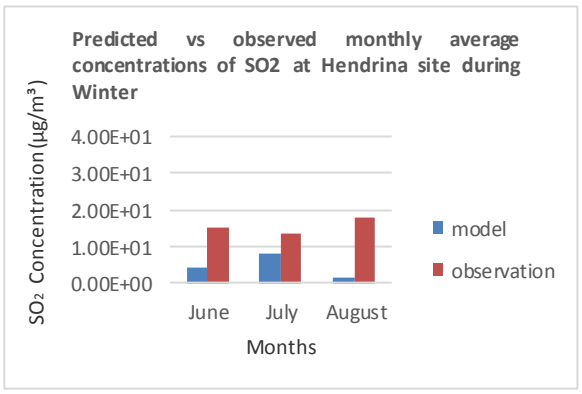


(c)



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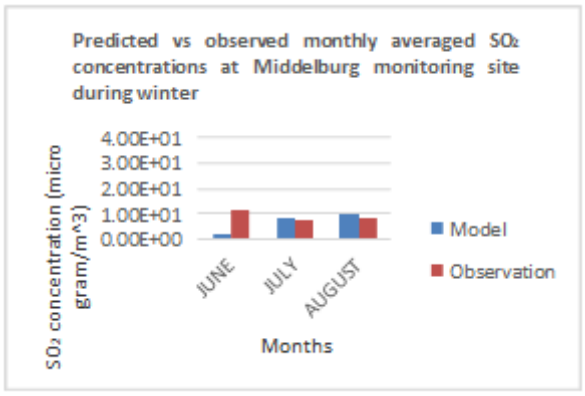
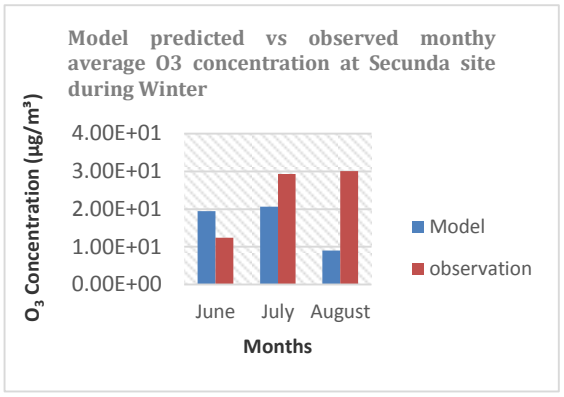
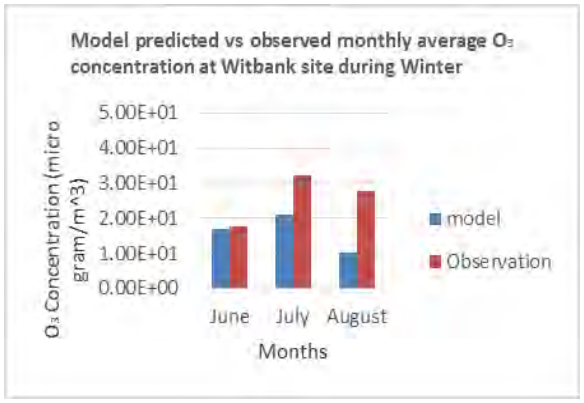


(e)

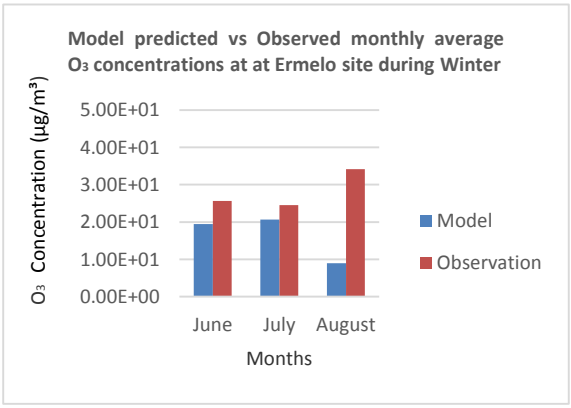
Figure 4.3: Bar graphs representing modelled vs measured monthly averaged SO_2 concentrations at five monitoring sites in the Mpumalanga Highveld during Jun- August 2010



(a)



(b)



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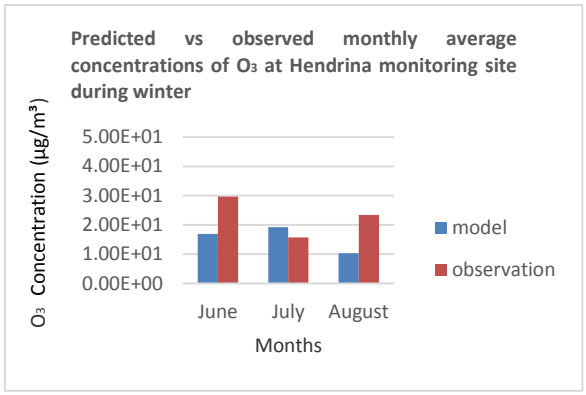

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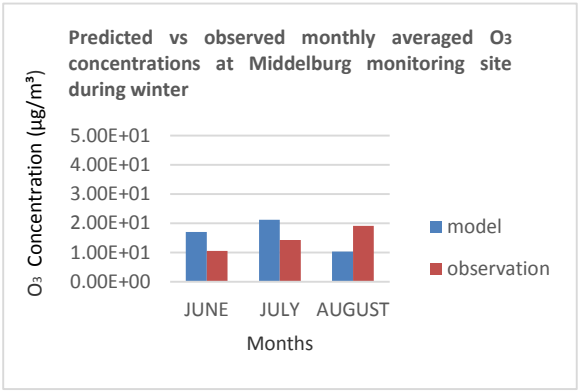
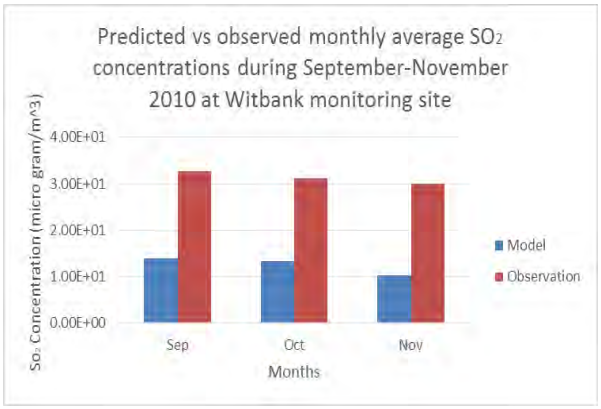


(e)

Figure 4.4: Bar graphs representing modelled vs measured monthly averaged O_3 concentrations at five monitoring sites in the Mpumalanga Highveld during Jun- August 2010

(a)



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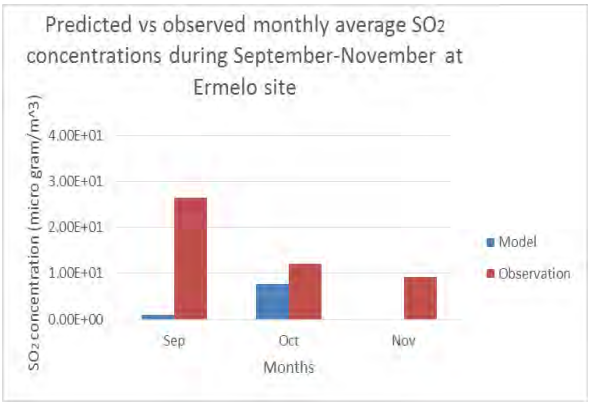

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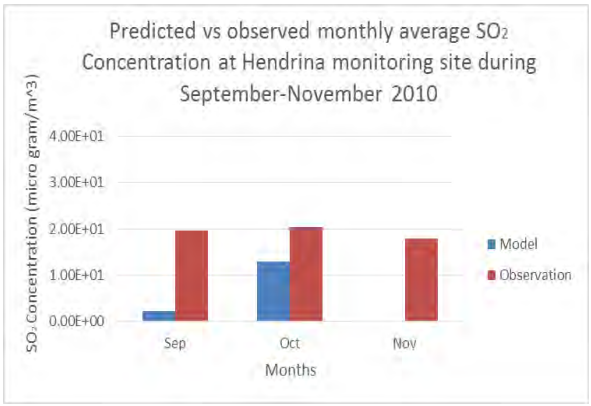

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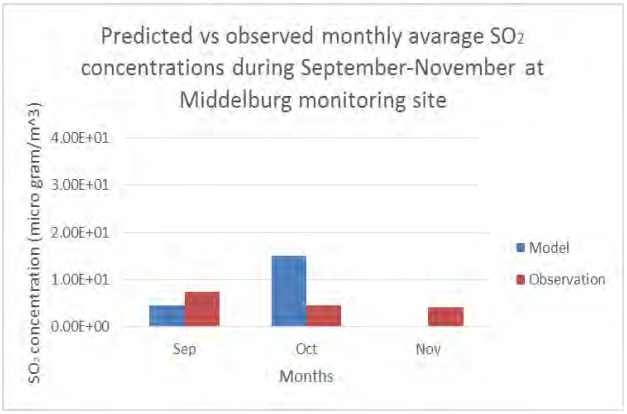
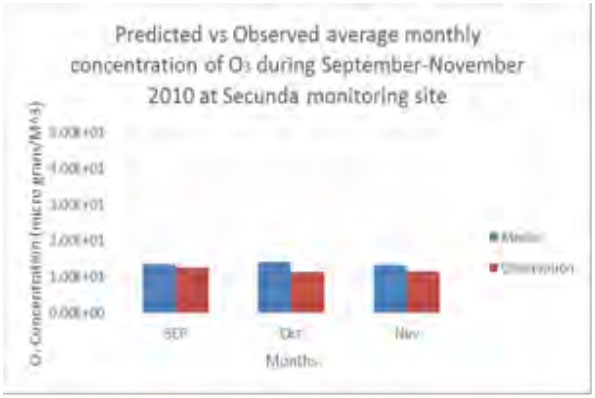
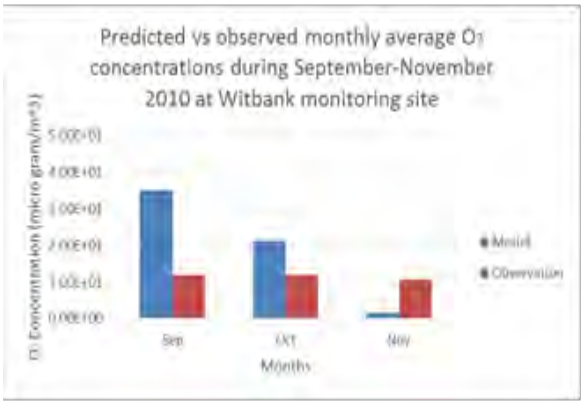


(e)

Figure 4.5: Bar graphs representing modelled vs measured monthly averaged SO_2 concentrations at five monitoring sites in the Mpumalanga Highveld during September-November 2010



(a)



(b)

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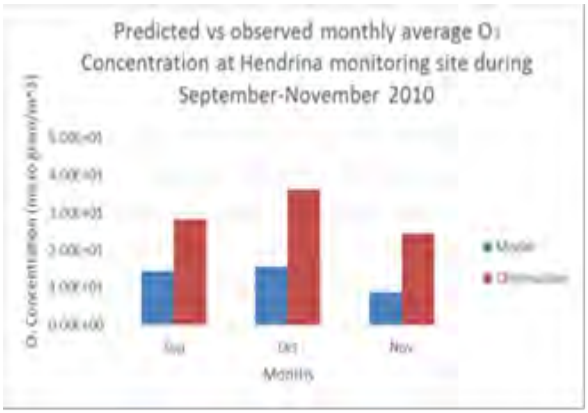

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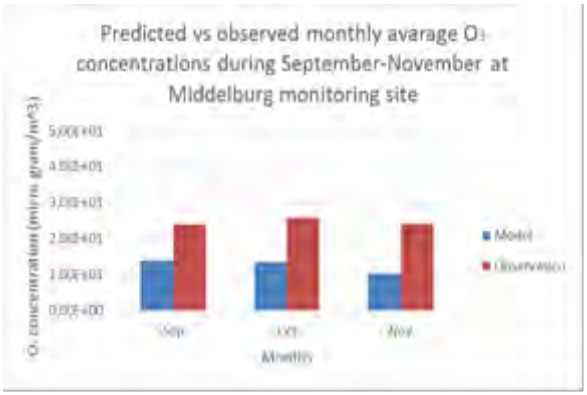
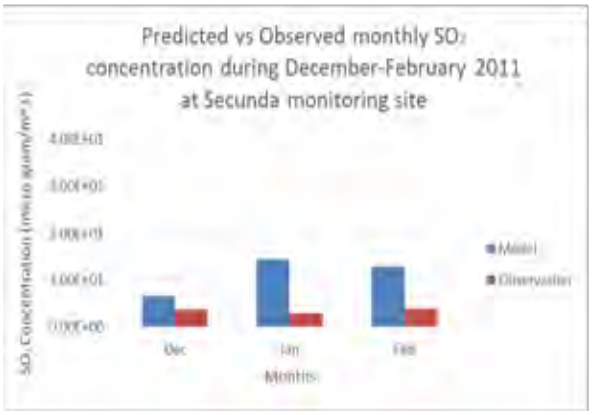
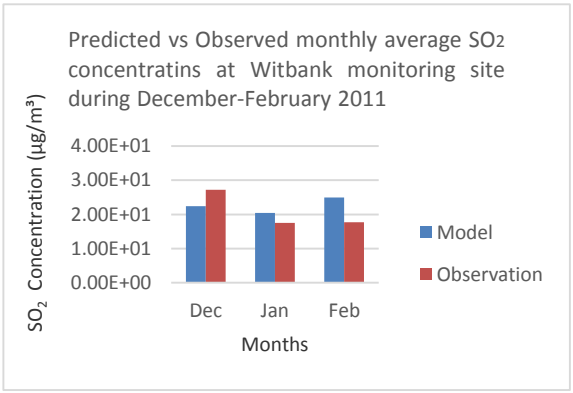


(e)

Figure 4.6: Bar graphs representing modelled vs measured monthly averaged O_3 concentrations at five monitoring sites in the Mpumalanga Highveld during September-November 2010



(a)



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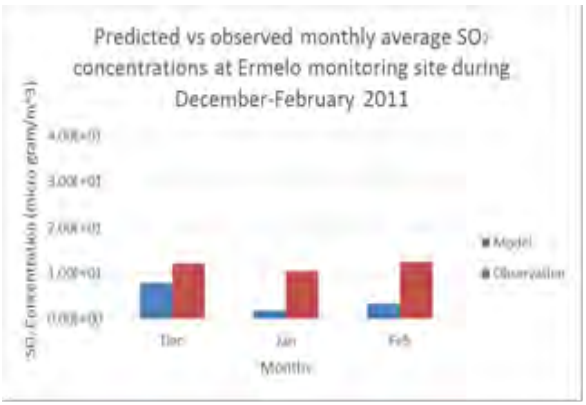

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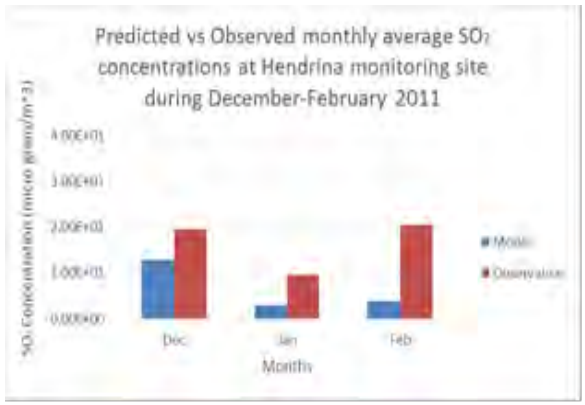

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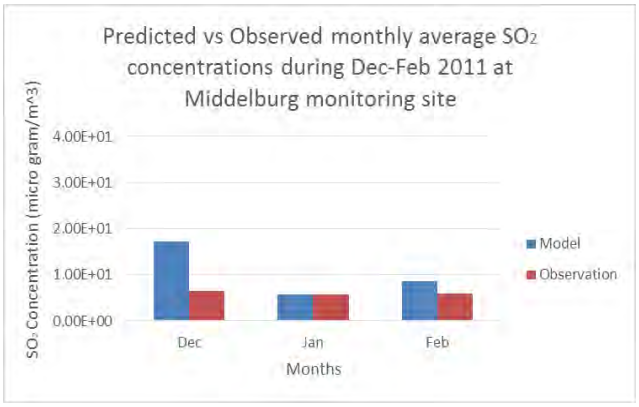
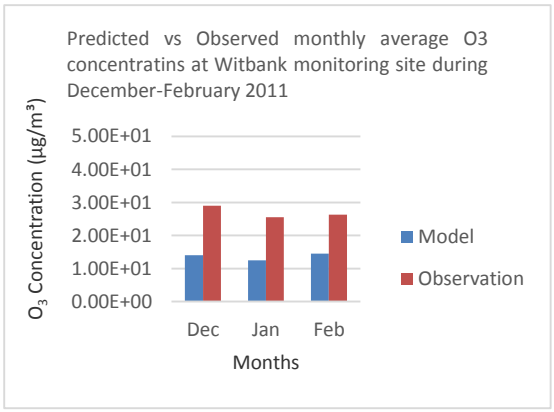


(e)

Figure 4.7: Bar graphs representing modelled vs measured monthly averaged SO_2 concentrations at five monitoring sites in the Mpumalanga Highveld during December-February 2010/2011

(a)



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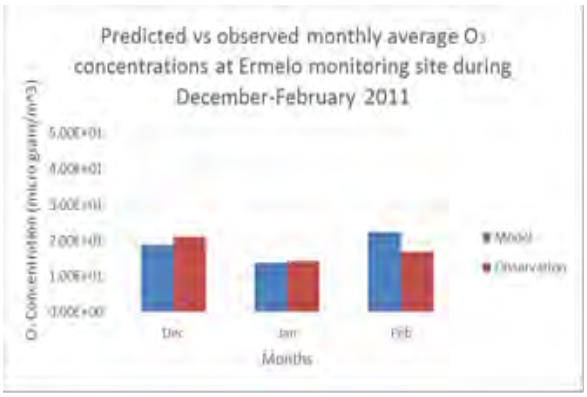

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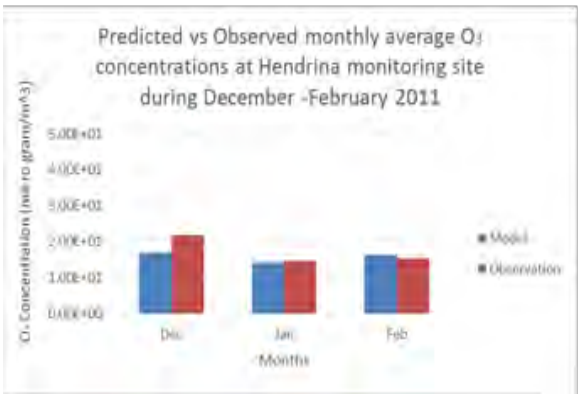


(d)

(e)

Figure 4.8: Bar graphs representing modelled vs measured monthly averaged O_3 concentrations at five monitoring sites in the Mpumalanga Highveld during December-February 2010/2011

4.2 Statistical analysis of the measured and predicted concentration

The NAME III-global (NWP and emission data) air quality modelling system was tested against monthly averaged concentrations of SO_2 and O_3 from the Mpumalanga Highveld air quality monitoring network. The statistical analysis technique was applied here and it includes calculation of Bias, NMB, RMSE and NRMSE. Tables 4.1 to 4.4 present the level of comparison for the measured and observed monthly average concentrations of SO_2 and O_3.

4.2.1 Sulphur Dioxide

The difference between the predicted and measured concentrations of SO_2 varies from a bias value of 1.02 to 20.5 $\mu g/m^3$ between the stations and the seasons, with the highest bias occurring in Ermelo station during autumn (Figure. 4.1c) at approximately 33% (100*NMB) and the lowest bias (about 19% difference) is at Hendrina. However, for the same season the predicted concentrations are closest (1.02 $\mu g/m^3$ average difference) to the observation at the Hendrina station. The overall performance of the model was good with a NRMSE varying between 0.14 - 9.4. In most cases the NRMSE is below 5 which indicate that the model is within the $50^{\%}$ range of the observed SO_2 concentration. The far outlying values of NRMSE occur at Middelburg station in April and May, on these months the predicted concentrations were very low (about 10^{-3} $\mu g/m^3$).

4.2.1 Ozone

The statistical difference in predicted and measured concentrations of O_3 varies with a bias value ranging from 0.78 at the Secunda station to 27.7 in Ermelo. Again, as in the case of SO_2 above, the difference between modelled and observed O_3 concentrations is high in Ermelo station but this time it occurs in winter. The interesting part is on the frequent occurrence of NRMSE that are below 1 with some even closer to zero, about 70 % of the cases where the NRMSE is close to zero.

Table 4.1: Statistical analysis of the predicted and observed monthly averaged concentrations of O_3 and SO_2 in autumn season 2011.

	Ozone				Sulphur Dioxide			
Site	Bias	NMB	RMSE	NRMSE	Bias	NMB	RMSE	NRMSE
Secunda	0.78	0.010	16.35	0.414	-8.98	-0.28	9.884	0.559
Witbank	4.59	0.09	9.83	0.31	-10.49	-0.25	18.73	3.20
Ermelo	0.97	0.01	8.58	0.12	-20.5	-0.33	20.66	4.70
Middelburg	12.0	0.48	13.37	1.08	-6.13	-0.30	6.41	9.35
Hendrina	-4.57	-0.05	9.71	0.014	-1.02	-0.19	17.77	0.14

Table 4.2: Statistical analysis of the predicted and observed monthly averaged concentrations of O_3 and SO_2 in spring 2010.

	Ozone				Sulphur Dioxide			
Site	Bias	NMB	RMSE	NRMSE	Bias	NMB	RMSE	NRMSE
Secunda	1.81	0.00	1.95	0.024	-6.75	-0.16	9.95	1.03
Witbank	-10.88	-0.19	18.78	0.90	7.80	0.22	15.35	1.06
Ermelo	-25.53	-0.21	26.10	1.29	-13.0	-0.27	15.84	5.47
Middelburg	-12.10	-0.16	12.23	0.48	1.18	0.07	6.71	1.30
Hendrina	-16.68	-0.18	17.00	0.75	-10.43	-0.24	15.08	2.34

Table 4.3: Statistical analysis of the predicted and observed monthly averaged concentrations of O_3 and SO_2 in Summer 2010/11.

	Ozone				Sulphur Dioxide			
Site	Bias	NMB	RMSE	NRMSE	Bias	NMB	RMSE	NRMSE
Secunda	-12.26	-0.13	19.75	0.68	7.76	0.72	8.56	1.40
Witbank	-13.3	-0.16	13.32	0.48	1.77	0.03	5.29	0.059
Ermelo	0.96	0.11	3.32	3.46	-7.27	-0.20	7.59	1.127
Middelburg	-11.10	-0.14	16.73	0.83	4.44	0.24	6.43	0.48
Hendrina	-1.38	-0.02	2.82	0.09	-10.1	-0.20	11.07	1.14

Table 4.4: Statistical analysis of the predicted and observed monthly averaged concentrations of O_3 and SO_2 in winter 2010.

	Ozone				Sulphur Dioxide			
Site	Bias	NMB	RMSE	NRMSE	Bias	NMB	RMSE	NRMSE
Secunda	-12.4	-0.18	17.26	1.43	-7.55	-0.10	13.78	0.48
Witbank	-2.47	-0.09	17.64	0.59	-9.85	-0.13	12.04	0.35
Ermelo	-27.7	-0.31	28.05	10.75	-11.7	0.06	15.09	0.49
Middelburg	-17.21	-0.24	19.16	2.37	-9.20	-0.12	11.30	0.31
Hendrina	-2.38	-0.03	2.82	0.16	-10.1	-0.20	11.07	1.14

Following the above outcome of the seasonal analysis, this work was extended further to investigate the model capability in quantifying temporal and spatial distribution of the two (SO_2 and O_3) pollutants over South Africa, as discussed below.

4.3 Distribution of modelled SO_2 and O_3 over South Africa

4.3.1 Sulphur Dioxide concentrations

The model results (Figures 4.9 to 4.14) indicate that there are two distinct areas of concern in South Africa with regards to SO_2 concentrations, these include the north eastern part of the country, the Mpumalanga Highveld where a large number of industrial activities take place. The model produces high level of SO_2 concentrations in this region, especially during late summer and in early winter. The other region with second highest levels of SO_2 concentration after Highveld region is the south western part of the country with strong signatures observed in the Western Cape during winter months. The autumn season possesses lowest concentrations of SO_2 throughout the country. These results show a similar trend with other regional modelling studies of SO_2 performed within the southern Africa, e.g. Zunckel et al. (2000) used the Multi-scale Atmospheric Transport and Chemistry (MATCH) to model the transportation and deposition of SO_2 over the region and it was found that the Highveld region experiences the highest levels of SO_2 of the entire South Africa. Again an investigation of Highveld emission transport pathways revealed that Cape Town may act as a sink to SO_2 transport from the Highveld (Abiodun et al., 2014).

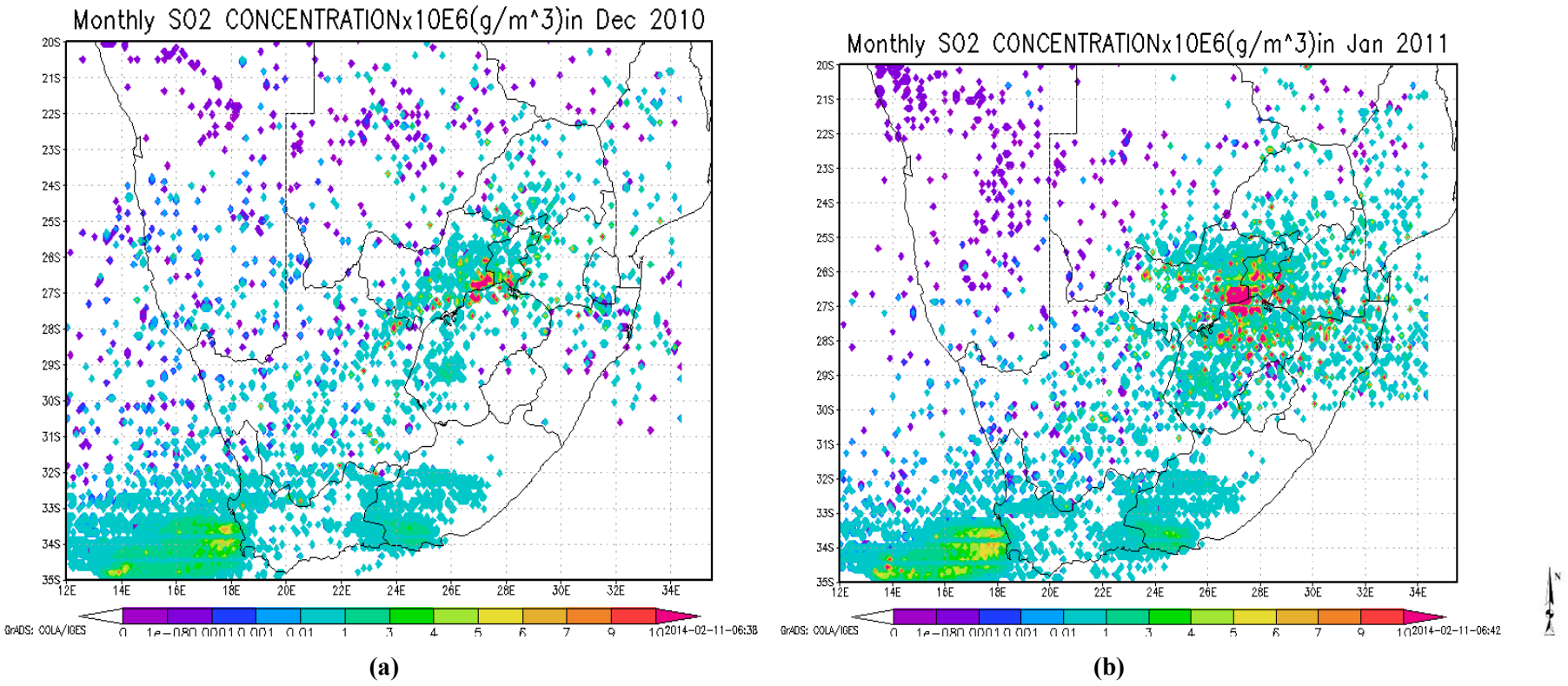
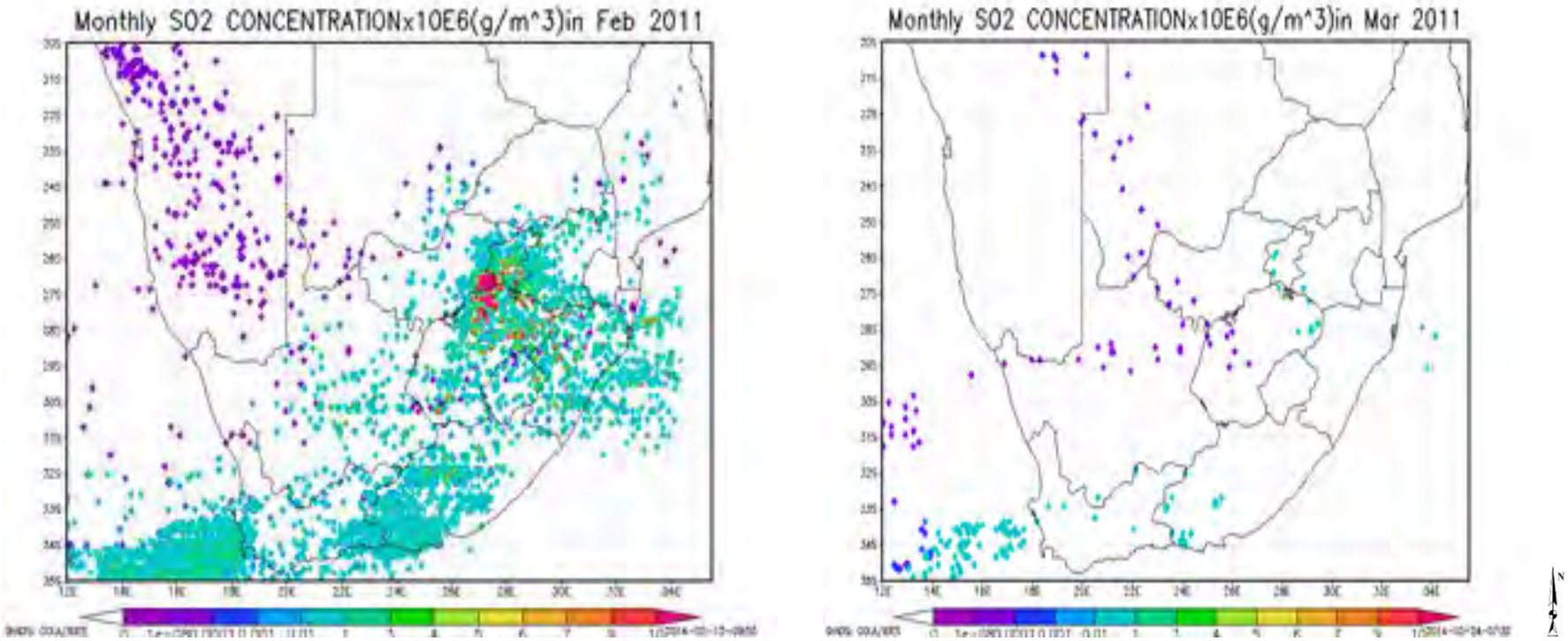


Figure 4.9: Plots representing the distribution of SO_2 over South Africa during December-Jan 2011.



(a) **(b)**

Figure 4.10: Plots representing the distribution of SO_2 over South Africa during February-March 2011.

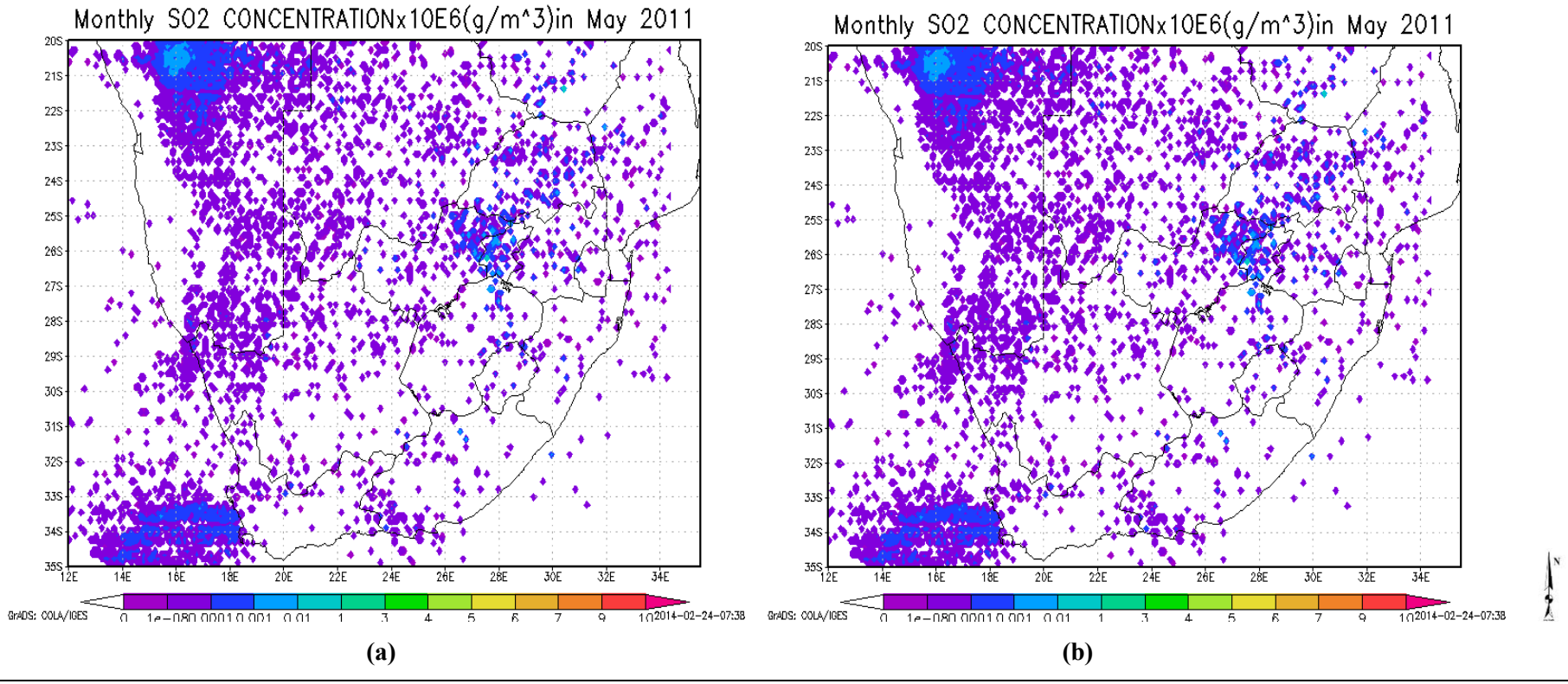


Figure 4.11: Plots representing the distribution of SO_2 over South Africa during April-May 2010.

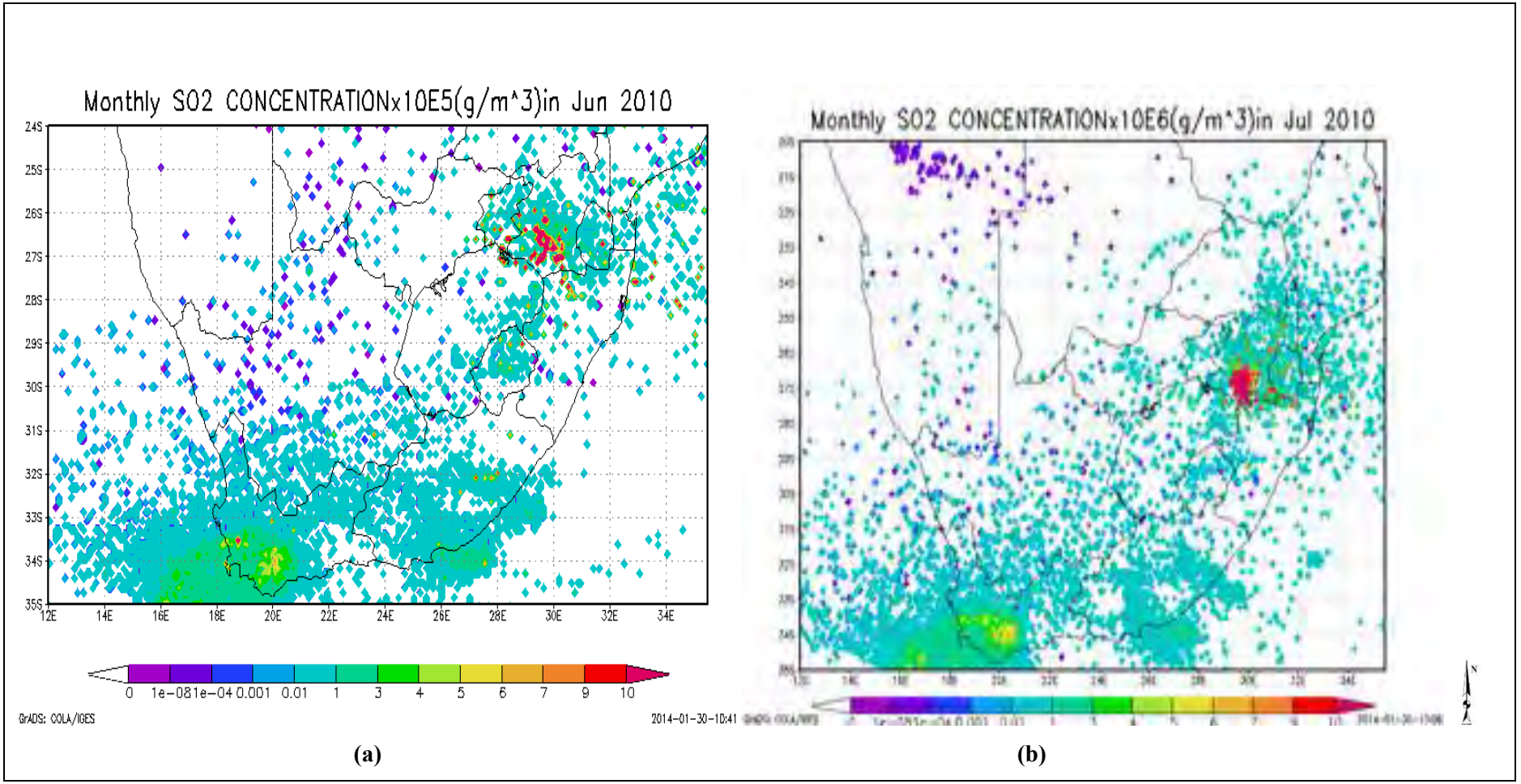


Figure 4.12: Plots representing the distribution of SO_2 over South Africa during June-July 2010.

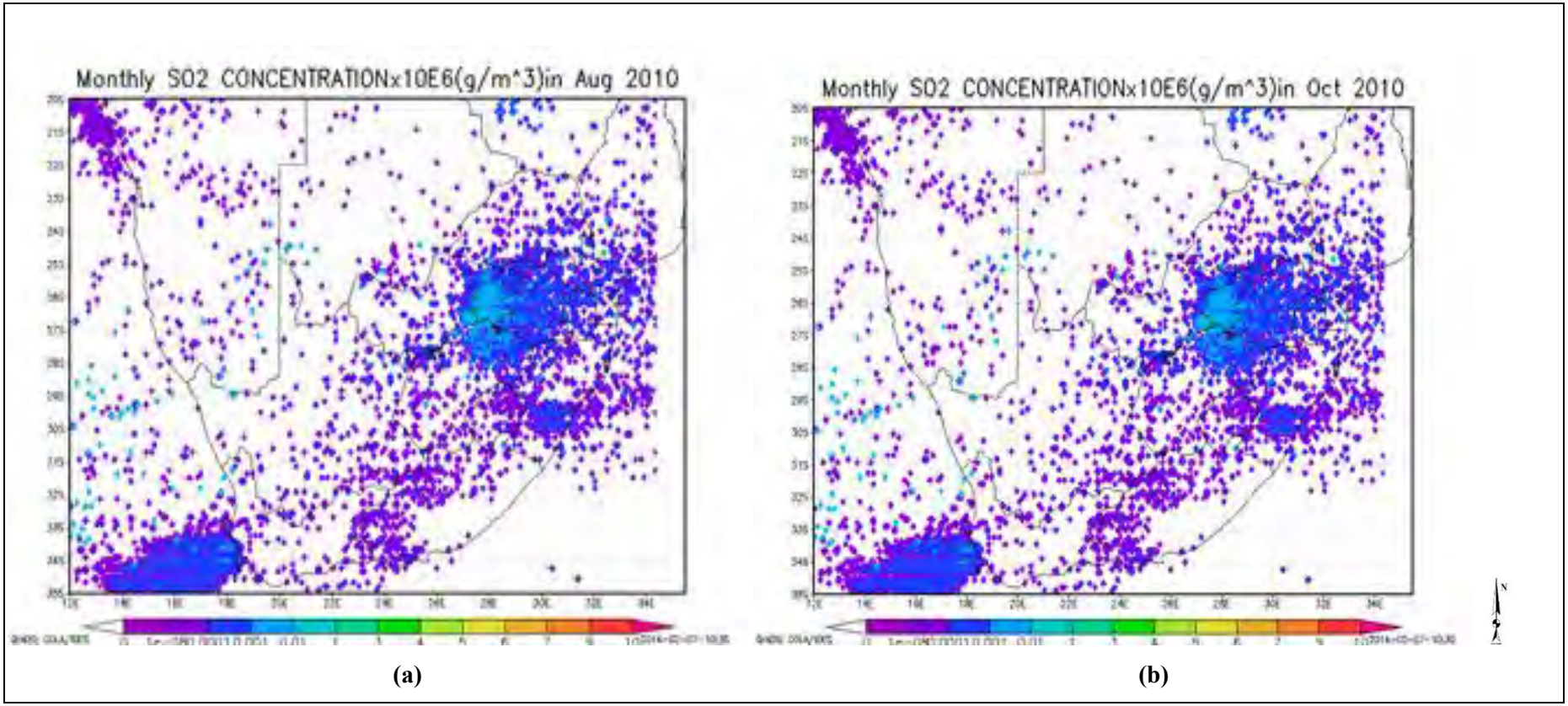


Figure 4.13: Plots representing the distribution of SO_2 over South Africa during August-September 2010.

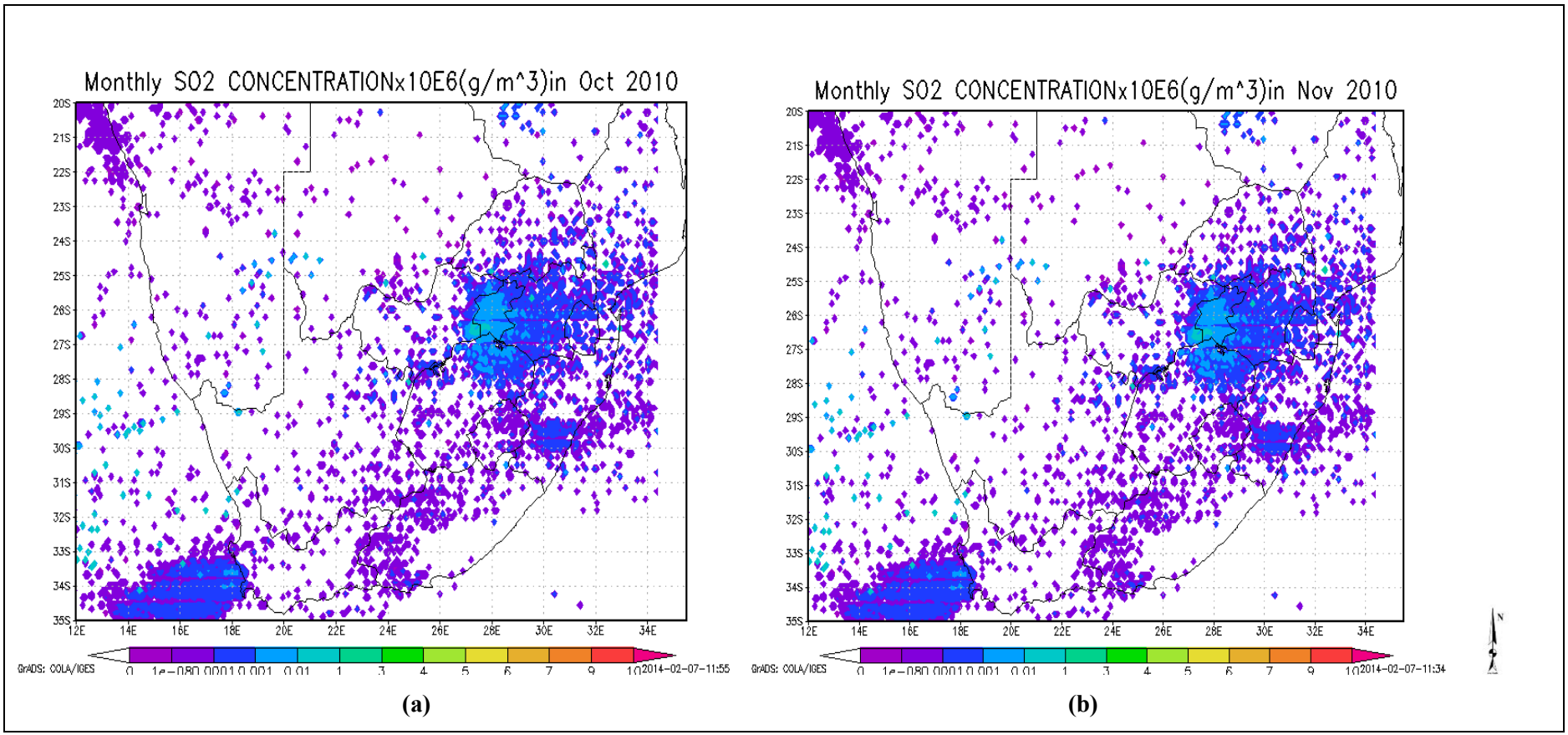


Figure 4.14: Plots representing the distribution of SO_2 over South Africa during October-November 2010.

4.3.2 Ozone concentrations

Unlike the SO_2 pollutant which is directly emitted from the source, the presence of O_3 in the troposphere is mainly through two distinct processes namely; stratospheric ozone ejection that causes the transport of stratospheric air, rich in ozone, into the troposphere and as a product of the photochemical process in the polluted boundary layer as discussed in Chapter 2. Figures 4.15 to 4.20 below present the distribution of O_3 over South Africa as predicted by the model. The visual analysis of these figures shows that maximum O_3 concentrations are located in the northern part of South Africa with strong concentrations occurring in the eastern Highveld region almost throughout the year. The existence of these high load concentrations corresponds to the industrial activities within this area which are in operation 365 days a year. On the west, a band of relatively higher O_3 concentrations stretches from the Atlantic Ocean towards the southern central region of Namibia and sometimes extended across towards east of the southern region. This event is noticed to begin in July (Figure 4.17(a)) and become more intense during September (Figure 4.18(a)) where a band of high O_3 move across the sub-region between 28^0 and 25^0 south. Such occurance can be attributed to the biomass burning activities that take place within the region during this period of the year (Silva, 2003). A country spread high ozone concentration is observed early in summer during December (Figure 4.19 (b)) but decrease in the late summer months (January to March), with the latter month showing well-defined concentration plumes over the Highveld and the southern Namibia with relatively clean air over the rest of the sub-region. The decrease in O_3 during this season may be associated to atmospheric conditions during this period. As discussed from the literature in Chapter 2, summer is associated with elevated inversion height allowing for efficient mixing of and dispersal of gases in the atmosphere. Furthermore, the wet deposition processes may also have an impact in the reduction of O_3 as rain producing systems are more apparent during this period of the year in the southern South African region. The rainfall may washout the existing pollutant gases from the atmosphere into the ground.

In general the results of this present study have shown similar trends of O_3 distribution with the results obtained from previous studies for example Zunckel *et al*. (2006) used the Comprehensive Air quality Model with extensions (CAMx) model to examine the monthly distribution of O_3 over the southern African region. The results of this study indicated that there is a bulge of high concentration of O_3 extending from the north western and north eastern coast moving along the northern part of the country. This forms a persistent belt crossing South Africa along the northern part of the country and extending outward the boundary to the northern countries throughout the year. In further a similar study by Babatunde *et al*. (2011) revealed that positive seasonal O_3 anomalies covering parts of Namibia,

Botswana and Zimbabwe are observed, specifically between August and October. These anomalies were attributed to biomass burning that prevail in the southern African region during spring time.

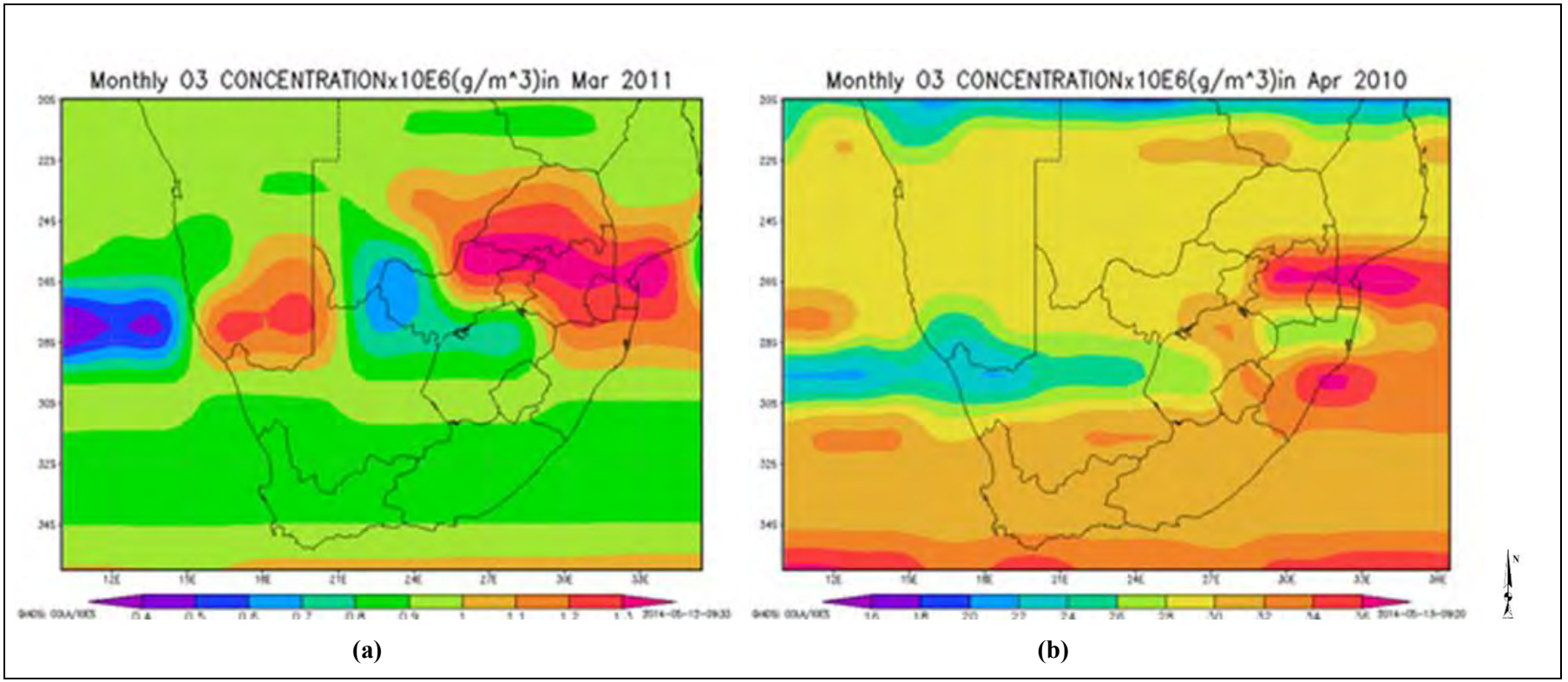
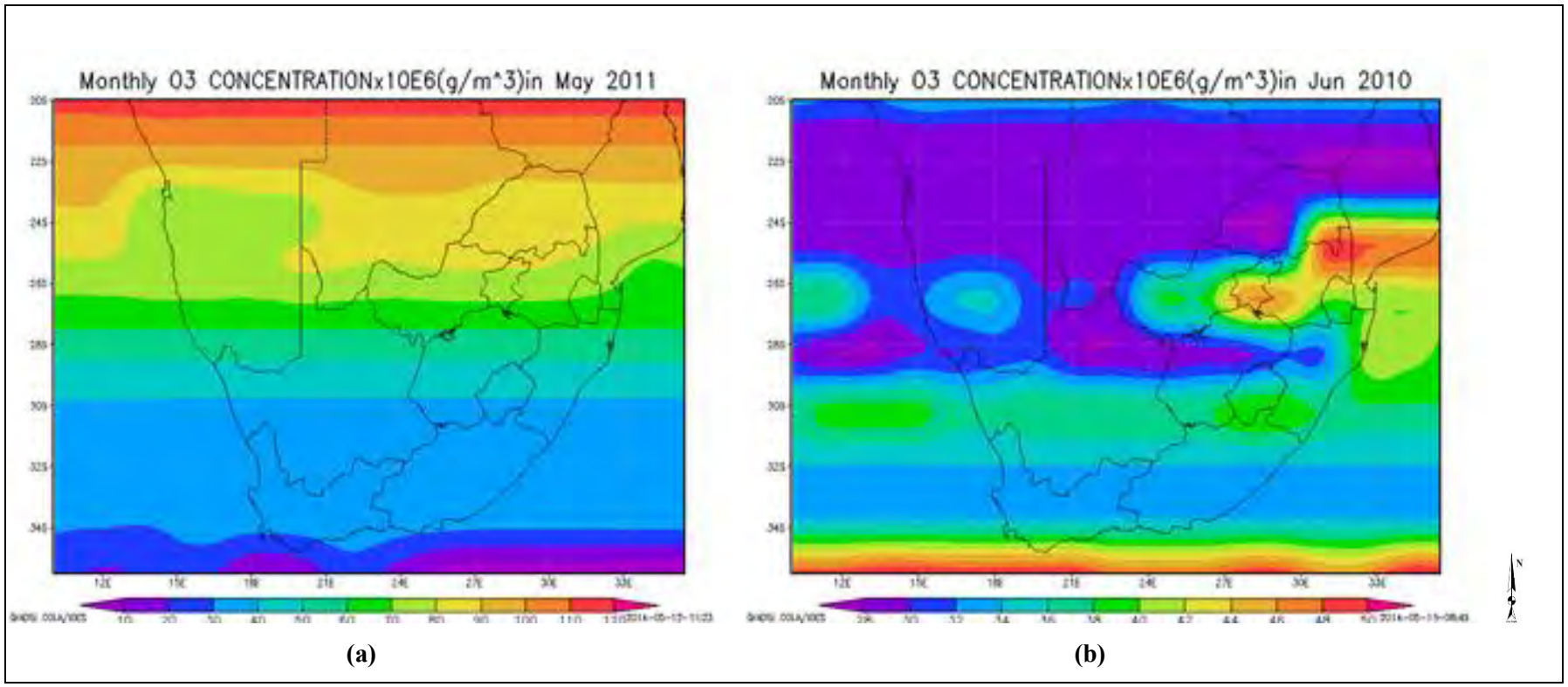


Figure 4.15: Plots representing the distribution of O_3 over South Africa during March-April 2010.



(a) **(b)**

Figure 4.16: Plots representing the distribution of O_3 over South Africa during May-June 2010.

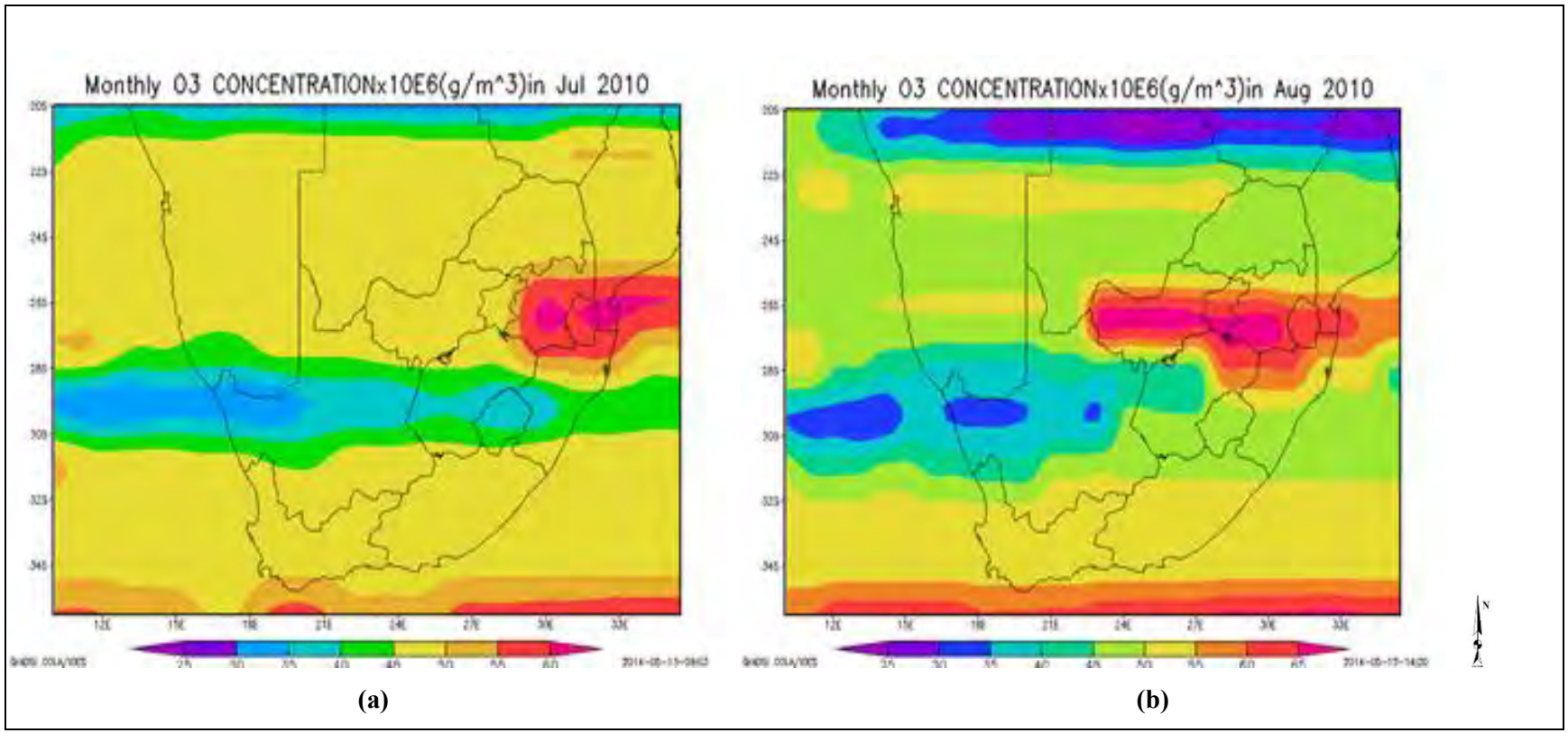


Figure 4.17: Plots representing the distribution of O_3 over South Africa during July-August 2010.

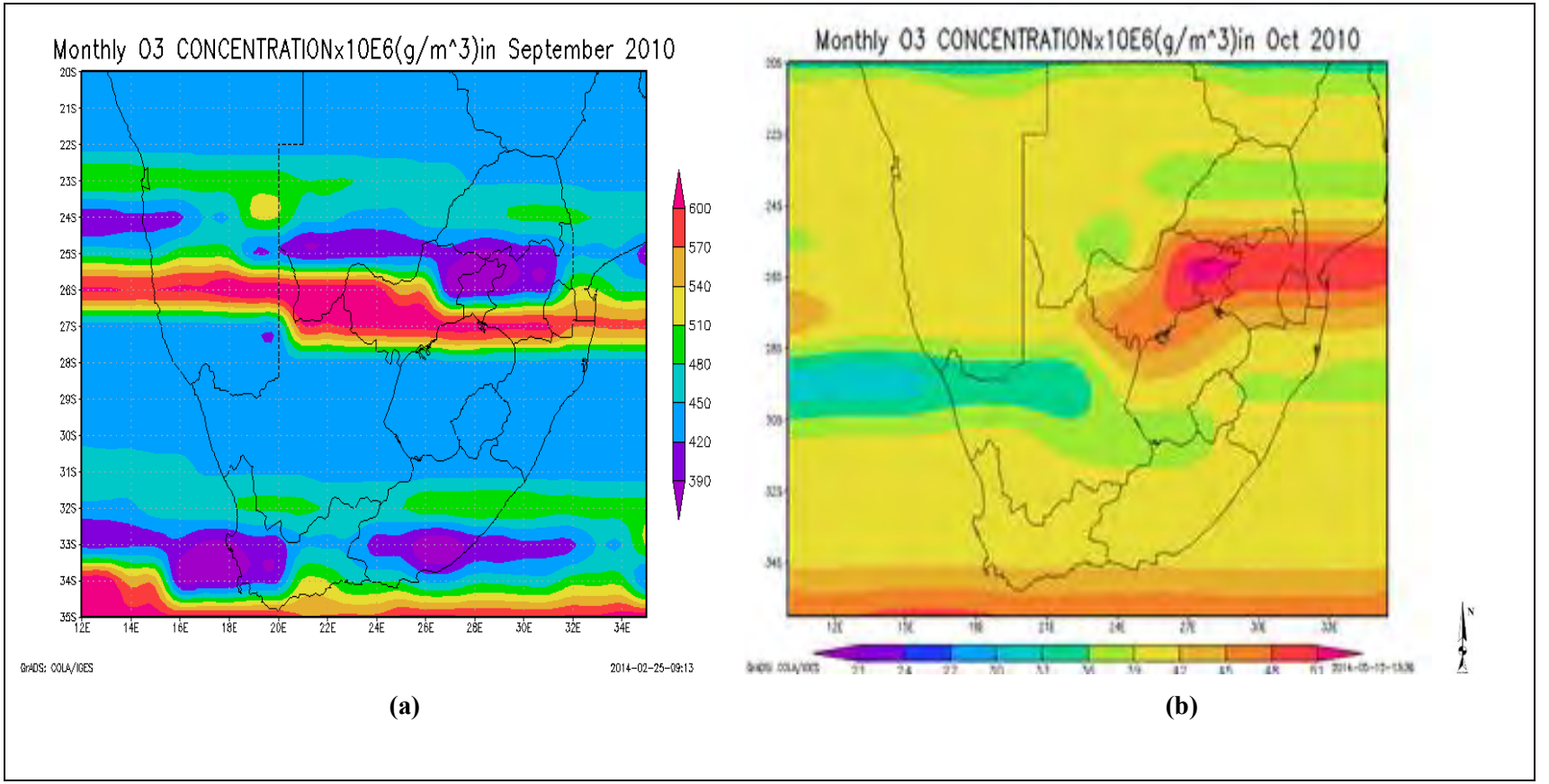


Figure 4.18: Plots representing the distribution of O_3 over South Africa during September-October 2010.

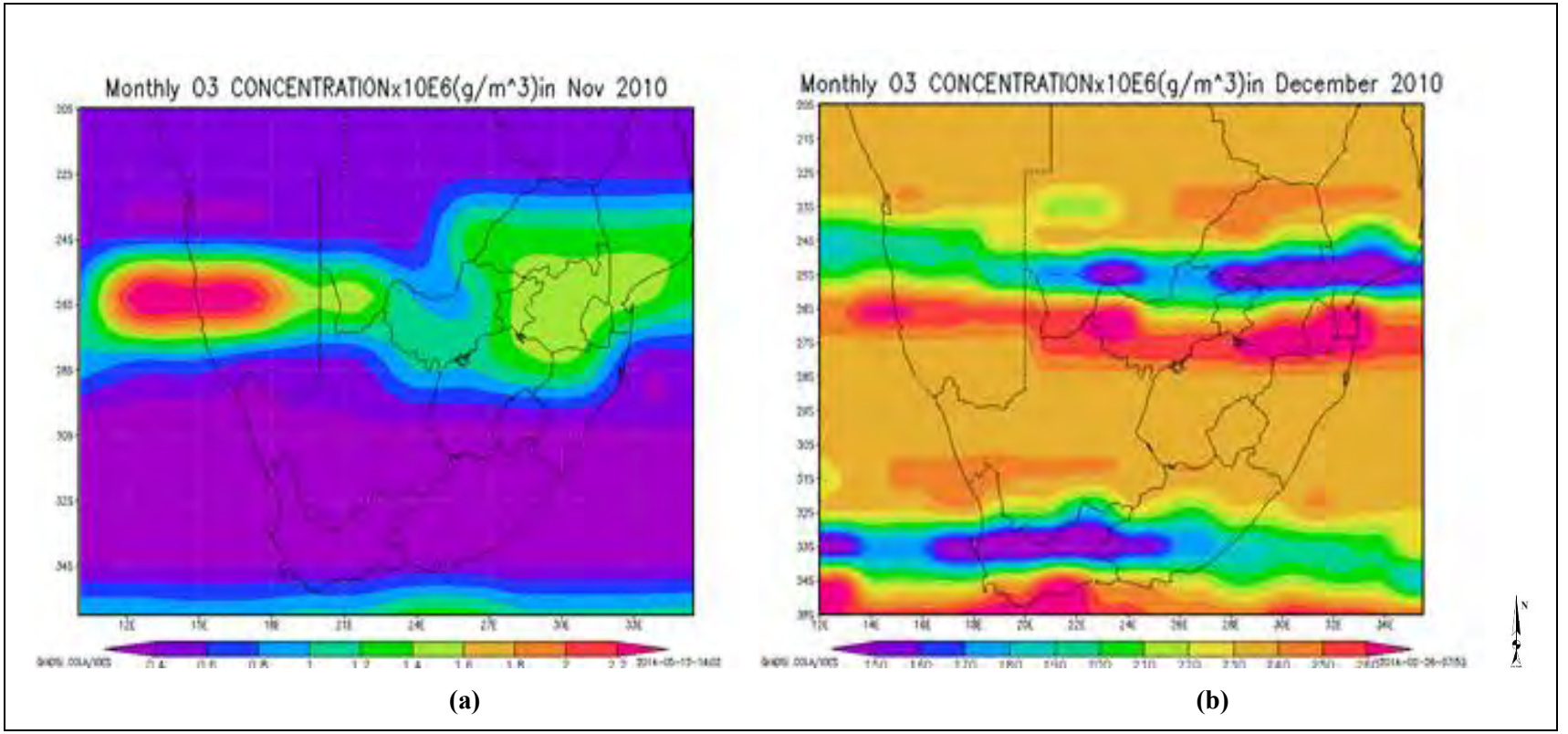


Figure 4.19: Plots representing the distribution of O_3 over South Africa during November-December 2010.

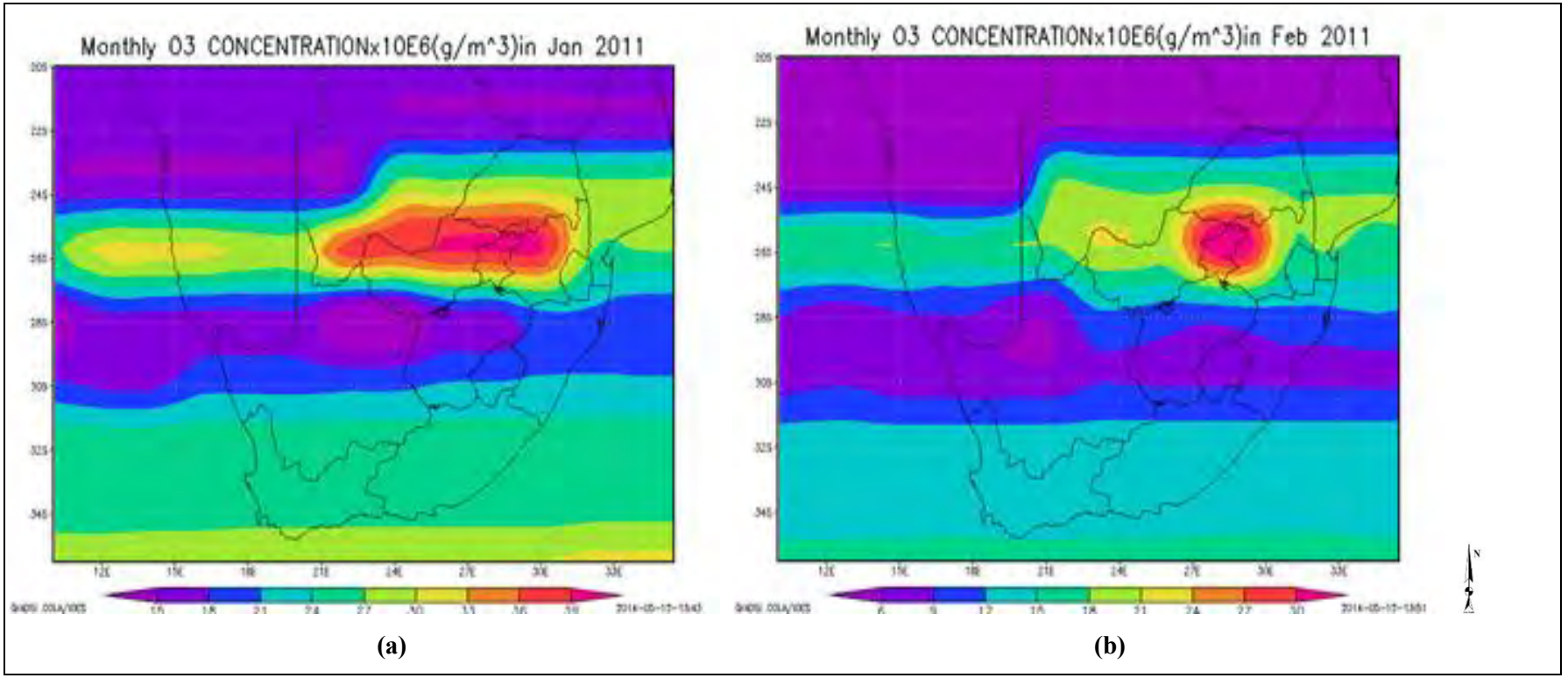


Figure 4.20: Plots representing the distribution of O_3 over South Africa during January-February 2011.

4.4 Summary

The seasonal trends for SO_2 and O_3 concentrations were apparent in observation, winter having the highest concentrations of SO_2 and the minimum peaks are observed in summer. Elevated O_3 peaks are observed in the spring and summer months and lower concentration peaks occur during winter. The model-observation data comparison indicates that the model under-estimates the concentrations of SO_2 and O_3, but the model values are in the same order of magnitude with the observed values. The model predicted the spatial distribution of the SO_2 concentrations are located in two distinct areas namely the eastern Highveld where a large number of industries are located and the south western part of the country, in the Western Cape. Relatively high O_3 concentrations are located in the northern part of South Africa with strong concentrations occurring in the eastern Highveld region almost throughout the year.

CHAPTER 5: CONLUSION

5.1 Summary

This study focused on investigating the capability of NAME III photochemical model for use in air quality modelling and forecasting in South Africa using two pollutants (SO_2 and O_3) as indicators. The model was used to predict the monthly average SO_2 and O_3 pollutants concentrations over the HPA. The seasonal distribution of the two pollutants over South Africa was also investigated. The global IPCC emissions data and the UKMO global model data were used to drive the NAME III model. The results of the model were compared with the corresponding monthly averaged concentrations of the two pollutants measured from five monitoring stations (Ermelo, Hendrina, Secunda, Middelburg and Witbank) within the HPA.

The statistical parameters (Bias, NMB, RMSE, and NRMSE) were used to investigate the level of agreement between model results and observation. Further the model was used to predict the seasonal distribution of the two criteria pollutants over South Africa and the results were compared to other similar modelling studies conducted locally. This is the first attempt to apply the (UKMO) NAME III / UM modelling system to simulate the concentration of different pollutants (SO_2 and O_3) in South Africa. It is important to mention that the study focussed more on the variation of air pollutants concentration on an extended (seasonal) time scale. This was driven by the availability of the data required in the model. The low resolution global NWP and emission data were the only available information for this study. Consequently, the model used global IPCC emission data and the global UM NWP data as the NAME III model input and which is of critical importance in air quality modelling.

Comparison of observed and simulated concentrations of SO_2 and O_3 in HPA showed that concentrations are under-predicted by NAME III. However, the modelled results were in the same order of magnitude as the measured data in overall except for two incidences in SO_2 concentration in Middelburg during April and May months, which may be attributed to the poor initialization of the model. For each season, the model was initialised for the first five days to allow for the pre calculation of the initial pollutant concentrations. This was not possible for the autumn season as no NWP data were available for initialisation during this period.

O_3 has been well captured in the summer months at all stations and was poorly represented in spring with only Secunda station showing better comparability. The autumn and winter concentrations were under-predicted but the model managed to pick the trend of increasing O_3 concentrations from March to August as observed at the stations. The SO_2 concentrations were the most under-predicted when compared with O_3 concentrations. The existing modelling system has been found to be very promising tool for air quality modelling studies in the Mpumalanga Highveld. Though it has shown to underestimate the concentrations of the modelled pollutants, especially the SO_2 but it has shown to be a potential valuable tool as it captures most of the observed information especially in the prediction of ground O_3 concentration.

Clearly, the underestimated concentrations are associated with the poor or low resolution emissions information of the global emission inventories. The emission inventory used in this study is of a low resolution and outdated. It does not take into account the local incidences, furthermore the changes in emissions as brought by the changes in land use, new developments and the introduction of the new air quality management policies were not taken into consideration. The other source of errors is associated with the course resolution NWP data used to drive the model, this information was not the true representation of the local meteorological conditions of South Africa. Furthermore, the modelling setup could not be appropriately handled, for example a relatively smaller (maximum of 500 000 particles) number of model particles were released to represent pollutants. This was due to the limited computational capacity which could not handle the calculations if a large number of particles were used. Webster and Thomson (2011) acknowledge that an increased number of particles may reduce the computational noise in NAME III and improve the model results.

The analysis of the distribution of pollutants over South Africa showed that the Mpumalanga Highveld region and the Western Cape are the two areas that are of distinct importance in South Africa, with the strongest signatures of SO_2 occurring over the Highveld region, an area of intense industrial activity. Also high level O_3 concentrations persist almost the whole year over this area and extending eastwards towards the Indian Ocean.

Furthermore the concentration of these pollutants also exhibit a seasonal variability, in which higher concentrations are predicted in winter and the opposite was true during summer months. The high concentration levels in winter may be attributed to the occurrence of shallow inversion layers that dominate the country during this season trapping pollutant near the ground. During the winter season, biomass burning and the industrial activities maybe the main sources of emissions, particularly in the

Highveld region. The summer months are associated with elevated inversion layers and rain producing systems which may result to the reduction in the atmospheric pollutants. The results of the present study were in good agreement with those reported in the literature, particularly the high level O_3 concentrations that prevail along the northern sub-region and extending east towards .

Overall, the method of using the NAME III modelling system for simulating pollutant concentrations in South Africa has shown encouraging results. However, a more detailed and updated national emission inventory, improved and localised NWP inputs and a detailed model evaluation are necessary to demonstrate the reliability and applicability of the UM-NAME III modelling system in the South African region.

5.2 Recommendations

Even though NWP meteorological input and IPCC emissions data were used, there was some agreement with the modelled and observed data. It is only when a detailed emissions inventory is developed (which is beyond the scope of this study) for use in such a model will there be sufficient evidence to support its use as a tool for forecasting air pollution in the country. However, whilst this study does offer promise for the use of tools it also highlights core issues that need to be resolved if South Africa is to develop an operational air quality forecasting model. These issues relate to the inventories, NWP/meteorological data and computational capacity which are discussed below.

Specifically, it is recommended that there is a need for more studies of this nature to be undertaken using different models as this will allow a better model inter-comparison and thus lead to the identification of suitable models that can be used for air quality prediction in South Africa. Critical to the testing of these models is the availability of a full emissions inventory for the country. A more comprehensive and accessible South African emission inventory needs to be developed and must be continually updated to allow for the inclusion of new sources and local incidences of pollution. The accessible detailed local emissions inventory will undoubtedly attract more air quality modelling studies that may be effective in the development and amendment of air quality management policies in South Africa.

The SAWS as a meteorological institution needs to expand its focus and provide NWP data specifically for air quality modelling at various resolutions (both temporally and spatially). Additional computational capacity will be required at SAWS for efficient air quality modelling activities. If SAWS were to

implement the air quality forecasting system, it is necessary that the existing computational resources are upgraded to allow for efficient running of the operational modelling systems since the current resources have limited space that is currently prioritised for operational weather predictions

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