- MASTERS DISSERTATION -

MODELLING THE EFFECTS OF BIOGENIC NO$_x$ AND INDUSTRIAL H$_2$S EMISSIONS ON THE SOUTH AFRICAN HIGHVELD AND WATERBERG REGIONS

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MODELLING THE EFFECTS OF BIOGENIC NO\textsubscript{X} AND INDUSTRIAL H\textsubscript{2}S EMISSIONS ON THE SOUTH AFRICAN HIGHVELD AND WATERBERG REGIONS

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SUMMARY

A comprehensive deposition and dispersion model was built for the South African Highveld and Waterberg areas using CALPUFF with the aim of studying the effects of biogenic NO\textsubscript{X} emissions on sulphur and nitrogen deposition. The effect of industrial H\textsubscript{2}S emission on sulphur deposition was also investigated for the Highveld.

Emission sources inventoried or the Highveld and Waterberg area included industrial sources, vehicle exhaust emissions, household fuel burning emissions and emissions from power stations. The Highveld model was the most extensive. Three scenarios were modelled: average rainfall year (2001), below average rainfall year (2003) and above average rainfall year (2010). The modelling domain was 350 km × 350 km. The Waterberg priority area was only modelled for 2006 and the domain size was 130 km × 100 km.

To quantify biogenic soil NO\textsubscript{X} emissions, models was constructed for both areas using land use data from CALMET, rainfall data and atmospheric ground level temperatures covering each modelling domain. Use was made of work done by Yienger and Levy (1995). To accommodate CALPUFF each area was divided into smaller area sources, each with a specific hourly NO\textsubscript{X} emission rate.

The biogenic NO\textsubscript{X} emitted made up 3.96 %, 4.14 % and 3.34 % of total released NO\textsubscript{X} for 2001, 2003 and 2010 respectively. This is significantly more than is released by household fuel burning, small industrial sources and biomass burning. Dry nitrogen deposition rates were affected most, adding between 1.69 - 6.19 % at various
receptor locations. Wet deposition rates were affected very little (0.13 % to 0.75 %). Effect on total nitrogen deposition rates ranged from 0.32 % to 1.77 %.

CALPUFF was unable to account for H₂S conversion to SO₂ in its reaction scheme model, therefore conversion rates had to be approximated from observations made on the Highveld by Igbafe (2007). Assuming different conversion percentages for each season, and inputting the converted emissions rates as SO₂ emissions sources into CALPUFF, it was predicted that H₂S contributes an average of 4.85 %, an average of 5.95 %, and an average of 5.15 % for wet S, dry S and total S deposition respectively.

Highveld dispersion and deposition predictions are reported on for the three modelling periods of 2001, 2003 and 2010. The modelled biogenic emissions were included in the model. Spatial plots for wet, dry and total S and N deposition were produced. Wet, dry and total S and N deposition rates at specific receptor locations are reported on.

Waterberg biogenic emission are only 2.3 % of total NOₓ emissions for the Waterberg area and would affect nitrogen deposition values very little compared to the nitrogen deposition produced by the emissions from Matimba and Medupi power stations. Because of this it was decided not to run a CALPUFF dispersion and deposition model for the Waterberg area.

**Keywords:** South African Highveld, South African Waterberg, biogenic NOₓ, dispersion modelling, deposition modelling, H₂S.
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1. INTRODUCTION

1.1 Background

Nitrogen and Sulphur deposition on the South African Highveld has been studied for at least two decades. This is because most of the country’s coal power generation facilities and related petrochemical industries are located within this area and are responsible for producing high concentrations of atmospheric sulphur and nitrogen.

A study was done by Scorgie and Kornelius in 2009 to study the following:

- How total sulphate and nitrogen deposition rates over the Highveld vary spatially and temporally;
- How wet and dry sulphate and nitrogen rates varies over the defined area;
- Relative anthropogenic source contributions and their spatial relationship to each other;
- How much these deposition rates vary during high, average and low rainfall years.

To this end S and N deposition was modelled using the US Environmental Protection Agency’s CALPUFF Lagrangian puff modelling system. This system uses various inputs such as:

- Terrain topography and specific land use;
- Spatial and temporal variations in meteorology;
- Wide variety of emission sources such as point, line and area sources.

Other capabilities of CALPUFF include:

- The ability to include first order atmospheric chemical transformation calculations to predict secondary pollutants formed from the primary pollutants;
• Using a resistance deposition model to predict spatial, temporal varying gas and dry deposition rates;

• Using scavenging coefficients to predict wet deposition.

This study builds on the work done by Scorgie and Kornelius. By adding biogenic NO\textsubscript{x} sources and specific H\textsubscript{2}S point sources, it is hoped that a more complete model of nitrogen and sulphur deposition can be achieved. Biogenic NO\textsubscript{x} emissions will also be estimated for the Waterberg region with the intent of constructing a nitrogen and sulphur dispersion model similar to the Highveld model.

1.2 Constructing a comprehensive dispersion model of the South African Highveld

The original emission inventories were obtained from Yvonne Scorgie. By the time this model was set up, significant changes had been made to the current version of the CALPUFF program and considerable effort was required to update the old input files to the newer formats. Remodelling the previous Scorgie and Kornelius model included the following tasks:

• The original meteorological data was sourced from local weather stations, while in this case MM5 (Fifth-Generation Penn State/NCAR Mesoscale Model) data (which consist of pre-rendered wind fields and weather data) will be used. One advantage of using MM5 is that one doesn’t waste time in sourcing and composing meteorological data from scratch;

• The emission inventories had to be reformatted to be used in conjunction with the new MM5 data. This included changing the UTM (Universal Transverse Mercator) system used in the previous build of the model to Lambert Conic Conformal (LCC) system. The advantage with LCC is that one can project a single Cartesian grid that distorts very little. When UTM was used the map distortion was excessive and the modelling domain fell outside the 350 × 350 km boundary;

• Sourcing and importing of new terrain and land use data;
• Three scenarios had been chosen to model: an average rainfall year (2001), an above average rainfall year (2010) and a below average rainfall year (2003);

• Compiling and inputting the various emission sources into CALPUFF. For the Highveld, these included 120 area sources from household burning activities, 158 small industrial point sources, 148 point sources and 66 volume sources from major industries, 16 area sources and 291 line sources representing vehicle emissions.

• Each scenario period (see above) consists of 22 separate simulation runs that must be completed. These are:
  
  o A CALMET (CALPUFF Meteorological input) file must be generated, which extracts the meteorology from the MM5 files and combines it with terrain and land use data into one file for CALPUFF to use;
  
  o Household burning area sources;
  
  o Vehicle tailpipe emissions area sources;
  
  o Vehicle tailpipe emission line sources, which was split into 13 separate runs, because CALPUFF only allowed for input of 24 line sources at a time;
  
  o Small industrial point sources;
  
  o Major Industrial point and area sources. Because the quantities these sources emitted were much higher compared to the other emission sources, the deposition due to these sources will be affected by seasonal variation in deposition velocities. Instead of running for the entire yearly period, 5 run times were selected for these sources: 2 summer periods (at start and end of each modelling period), 1 autumn, 1 winter and 1 spring.

• After generating 22 output files, these files are combined into one final file. CALPUFF comes with specific tools to extract both spatial and discrete receptor data.
After generating the spatial and receptor data the current build must be compared to the older modelled results.

Cross checking various dispersion settings and adjusting them for the current version of CALPUFF.

1.3 Building a comprehensive biogenic NOx emission model for both the South African Highveld and Waterberg areas

Modelling biogenic NOx emissions for the Highveld will consist of:

- Sourcing temperature and rainfall data from CALMET data file each of the three years (2001, 2003 and 2010).
- Composing a biogenic emission simulation spreadsheet.
- Sourcing land use for each area source for input into the biogenic emission spreadsheet. Land use basically defines what biome is encountered within each area source and affects which emission coefficient must be used for calculating the NO\textsubscript{x} emission rate.
- Calculating biogenic emission for each of the 196 area sources, for each hour per year, for each of the three modelling scenarios.
- Composing the calculated NO\textsubscript{x} emissions into a BAERMARB (Buoyant Area Source Emissions File with Arbitrarily Varying Emissions) input file.

Modelling biogenic NOx emissions for the Waterberg area:

- Sourcing temperature data from CALMET data file.
- Requesting rainfall data for Lephalale from the South African Weather Service and composing it into an 8760 hours (365 days × 24 hours) input file.
- Calculating emission coefficients for South African savannah from published figures
- Composing biogenic emission simulation spreadsheet.
o Sourcing land use for each area source for input into the biogenic emission spreadsheet.

o Calculating biogenic emission for each of the 25 area sources for an entire year.

o Composing the calculated NO\(_x\) emissions into a BEARMARB input file.

- Running deposition models with biogenic emission and add the output files to the previous modelling results for the Highveld.

- Generating the spatial and receptor data that shows how the biogenic NO\(_x\) emission influences deposition for the Highveld.

### 1.4 H\(_2\)S Emissions

H\(_2\)S emissions were not quantified in the original study by Scorgie and Kornelius (2009), because CALPUFF modelling suite does not have H\(_2\)S as part of the RIVAD reaction scheme. With significant H\(_2\)S emissions occurring on the Highveld the H\(_2\)S emission sources should be compiled into an emission inventory, which must be then be converted into a suitable CALPUFF input file.

### 1.5 Waterberg Emission Inventory and Nitrogen Dispersion

The same methodology used above for the Highveld will then be used to determine the effect biogenic emissions have on the Waterberg area. A comprehensive emission inventory will be compiled to study the impact biogenic NO\(_x\) will have on nitrogen emissions for the area. It should be noted that in view of findings in section 4.3 it was decided not to do the additional dispersion modelling.

### 1.6 Key Research Questions

The following are main topics that will be researched:
• Constructing a comprehensive emission inventory and dispersion model for both the South African Highveld and Waterberg area.

• Building a comprehensive biogenic NO\textsubscript{x} model for both the South African Highveld and Waterberg areas to study the effect of biogenic NO\textsubscript{x} emissions on the nitrogen deposition in these regions.

• Quantifying H\textsubscript{2}S sources on the Highveld and finding a way to include the H\textsubscript{2}S emissions in the deposition model.
2. LITERATURE

2.1 Introduction

Global nitrogen and sulphur natural cycles are first investigated. The major pathways these elements follow as they enter and leave the global environmental system are discussed. Anthropological activities have a major influence on the release and conversion of these elements into chemically active species such $\text{SO}_x$ and $\text{NO}_x$. Tapping into the natural occurring reservoirs has been hugely beneficial for human technological progress. The effects human activities have on the overall environment are shown.

From here the focus shifts to atmospheric chemistry. The atmosphere acts as a large sun powered reactor. It also disperses the released chemicals over large regions at a distance from their sources. Major reaction pathways and residence times of the major chemical species that are important for this study are shown.

Atmospheric deposition deals with the formation and transport of the final chemical products formed in the atmosphere. Both wet and dry deposition processes are described mathematically. These processes are then further studied in the context of the South African Highveld. Various quantification and simulation methods are described, while a summary of deposition rates as they occur spatially and temporally is included.

Finally, the mechanics of biological generated nitrogen emissions are looked at. This is once again done within a South African context. Under this section the effect of fertilizer addition is also described, since it affects the emission of nitrogen species through evaporation processes and by stimulating biological activities of bacteria found in the soil.

2.2 Global Nutrient Cycles
An overview of the deposition of a number of atmospheric pollutants, including sulphur and nitrogen, was recently provided by Vet et al (2014). This shows that measurements of deposition of S and N species over South Africa are widely spaced and that no continuous measurement record exists. The species of major concern in South Africa are discussed below.

2.2.1 Sulphur

Sulphur-containing species are primarily found in solid form on earth. The largest reservoirs of sulphur are found in the Lithosphere ($2.4 \times 10^{10}$ Tg S) (Reeburgh, 1997) and sediments on land and in the ocean ($7.8 \times 10^9$ Tg S) (Galloway, 1996). About $1.3 \times 10^6$ Tg S is dissolved in the ocean, while very little (1.6 Tg S) sulphur (compared to the other reservoirs) can be found in the atmosphere (Reeburgh, 1997; Galloway 1996).

Natural processes that convert sulphur minerals to reactive sulphur include volcanic eruptions ($20$ Tg S yr$^{-1}$) (Galloway, 1996) where the gases are composed mostly of SO$_2$, H$_2$S and SO$_3$ (Kellogg et al., 1972) and are ejected directly into the atmosphere. Another major source is mineral weathering through aeolian processes ($20$ Tg S yr$^{-1}$) (Galloway, 1996). Finally, mineral weathering happens through aerobic biogeochemical processes in the soil. Products thereof can either be emitted directly into the atmosphere or in the form of soluble sulphate compounds that are absorbed by ground water (about 60 Tg S yr$^{-1}$) (Galloway, 1996).

The released sulphur which easily dissolves in water is then transported by rivers to the ocean at a rate estimated to be $104$ Tg S yr$^{-1}$ (Galloway, 1996; Reeburgh, 1997). During transport downstream or in the ocean, the sulphates can be reduced to sulphides, which will either be released into the atmosphere as a gas or be deposited as sediment (Galloway, 1996).

The residence time of sulphur in the atmosphere is short, usually days (Galloway, 1996). The sulphur is removed from the atmosphere either as SO$_2$ or as sulphate aerosols (Reeburgh, 1997).
Anthropogenic activities mobilize about 150 Tg S yr\(^{-1}\) from the lithosphere (Galloway, 1996; Kellogg et al., 1972; Reeburgh, 1997). About 93 Tg S yr\(^{-1}\) of the reactive S produced through human activities are emitted into the atmosphere and the rest is released as waste water (29 Tg S yr\(^{-1}\)) or used in fertilization (28 Tg S yr\(^{-1}\)) (Galloway, 1996).

Sulphur deposition rates over continental and oceanic areas have increased since preindustrial times. In 1997 Reeburgh made an estimation from various sources that in preindustrial times the global deposition rates over continental areas was 43 Tg S yr\(^{-1}\) and over oceanic areas 159 Tg S yr\(^{-1}\). For current industrial times (mid-1980s), the global deposition rates for continental and oceanic areas were estimated at 65 Tg S yr\(^{-1}\) and 231 Tg S yr\(^{-1}\) respectively.

### 2.2.2 Nitrogen

Unlike sulphur, which is most stable as a solid and for which the biggest reservoir is found in the lithosphere, nitrogen’s most stable form is N\(_2\) gas and this comprises the largest fraction of the gases composing earth’s atmosphere (3.9 – 4 × 10\(^9\) Tg N). In this gaseous form, it takes a large amount of energy to separate the two triple bonded N atoms to make reactive Nitrogen (N\(_r\)). Only a few natural processes on Earth produce N\(_r\) that can become available for use by organisms and geophysical processes. The reactive forms of N are a major source of nutrients for all organisms on earth and are often the limiting nutrient in soil ecosystems (Vitousek et al., 1997; Galloway, 1996; Reeburgh, 1997).

#### 2.2.2.1 Nitrogen Fixation

The two main N fixation processes in nature are lightning (< 10 Tg N yr\(^{-1}\)) and biological fixation through microorganisms in mutual relationships with higher plants like legumes (90 – 140 Tg N yr\(^{-1}\)) on land and algae (30 – 300 Tg N yr\(^{-1}\)) in the ocean (Vitousek et al., 1997).
Before the 1900’s the primary N\textsubscript{r} source was either recycled biomatter or through cultivating crops that promote nitrogen fixation such as rice or leguminous crops like soybeans. It was common practice to mine guano and nitrate deposits, which were used as fertilizer for food production in Europe. One big breakthrough was the discovery of the Haber-Bosch process through which atmospheric N\textsubscript{2} was converted to NH\textsubscript{3} in very large amounts. Today it’s estimated that about 85 Tg N\textsubscript{r} yr\textsuperscript{-1} is created through this process and is required for a big part of the food production to support the growing human population on earth (Galloway and Cowling, 2002).

Nitrogen fixation through the cultivation of leguminous crops is still one of the primary methods used by farmers to enrich their crop fields. Just before 1900 leguminous crops were estimated to fixate about 15 Tg N\textsubscript{r} yr\textsuperscript{-1}. Today this is estimated to be about 33 – 40 Tg N\textsubscript{r} yr\textsuperscript{-1}(Galloway and Cowling, 2002).

\subsection{2.2.2.2 Anthropogenic N\textsubscript{r} mobilization}

Up to now only processes that converts unreactive N\textsubscript{2} to N\textsubscript{r} have been discussed. There are numerous other ways in which humans produce N\textsubscript{r}, whether by accident or on purpose. There is also a lot of uncertainty in determining the emission rates for some of these processes due to complex nature in which these processes interact with each other.

\subsection{2.2.2.3 Fossil fuel and biomass burning}

Along with developments in technology that gave us access to the atmospheric pool of N\textsubscript{2} to produce enough fertilizer for food production, advances have been made in power generation since the 20\textsuperscript{th} century. The combustion of fossil fuel allows us access to cheap plentiful energy. Fossil fuel is composed of remains of extinct life forms and nitrogen is a key component in proteins and nucleic acids of animal and plant life. When burned, it releases mostly NO and NO\textsubscript{2}, but if the reaction temperature is high enough to allow the oxygen to react with the atmospheric N\textsubscript{2}
gas, N₂O gas which is a greenhouse gas is also released. Combustion of fossil fuels has become an integral part of our economy since the early 1900’s. Currently it is a major source of anthropogenic Nᵣ, which releases about 20 Tg Nᵣ yr⁻¹ directly into the atmosphere (Vitousek et al., 1997; Galloway, 1996; Reeburgh, 1997).

In countries where there is no access to fossil fuels biomass burning is a common source of Nᵣ. It is estimated that biomass burning is responsible for about 12 Tg Nᵣ yr⁻¹ (Reeburgh, 1997).

### 2.2.2.4 Agricultural sources of Nᵣ

It was estimated that in the 1990’s about 170 Tg Nᵣ yr⁻¹ was introduced into agricultural ecosystems for food production. About 70% of the Nᵣ came from fertilizer and biological nitrogen fixation (BNF) through cultivation. The remaining 30% came from already existing Nᵣ, such as animal manure and atmospheric deposition. Very little of that Nr is stored in the soil itself (about 5%), while about 25% is emitted to the atmosphere in various forms (N₂, N₂O, NOₓ and NH₃). About 50% is used by the crop and about 20% is lost due to Nᵣ that dissolves in water effluent (Galloway et al., 2004).

The Nᵣ that is lost, either through runoff water or by being emitted to the atmosphere usually ends up in other unmanaged ecosystems. The addition of more N from agricultural sources increases the microbial activity responsible for processing fixed N from the atmosphere, which leads to increased amounts of Nᵣ emitted to the atmosphere by soil and water bodies. The additional N can also lead to eutrophication in water bodies and loss of biodiversity on land (Vitousek et al., 1997).

### 2.2.2.5 Denitrification

Denitrification is an important sink for Nᵣ. This happens mostly in the anaerobic conditions for example in waterlogged soil found in wetlands or coastal areas. This process converts dissolved forms of NOₓ to NO, N₂O and N₂ (Galloway and Cowling,
This then completes the nitrogen cycle which converts N, back into the atmospheric N₂, although the NO and N₂O produced through denitrification is still active and can still be transported into the atmosphere where it will take part in atmospheric chemistry. When areas like wetland are lost the dissolved N, in streams and rivers is not removed, spreading N, to more unmanaged ecosystems (Vitousek et al., 1997; Galloway and Cowling, 2002).

2.2.2.6 Role of N, in atmospheric chemistry

The most prominent N, chemicals in the atmosphere are N₂O, NOₓ and NH₃. All three of these are produced directly or indirectly by multiple anthropogenic sources and since the 1900’s their emissions has increased significantly (Galloway et al., 2004; Vitousek et al., 1997).

Nitrous oxide (N₂O) gas is produced by multiple anthropogenic sources such as burning of fossil fuel, biomass burning, land clearing and fertilized soils in agricultural areas. The main concern about N₂O is that it is a greenhouse gas and reacts differently at different altitudes. In the stratosphere, it reacts with other chemicals in the presence of sunlight to destroy O₃, but in the troposphere it is inert and can remain there for years (Vitousek et al., 1997). Currently anthropogenic emissions are responsible for about 30 to 40 percent of all N₂O emissions with the balance produced by natural sources (Galloway et al., 2004; Vitousek et al., 1997). Galloway et al. (2004) estimated that in total, about 15 Tg N yr⁻¹ are produced globally.

Roughly 46 Tg N yr⁻¹ of NOₓ is being emitted to the atmosphere. Anthropogenic sources account for about 80 percent of the all NOₓ emitted globally. The most prominent sources are from fossil fuel burning (60%), food production (20%) and natural sources (20%) (Galloway et al., 2004; Vitousek et al., 1997).

The concentration of NOₓ in the atmosphere plays a major role on the formation of tropospheric O₃, which is also a greenhouse gas. If the NOₓ concentration is high enough it acts as a catalyst in the oxidation reaction of CO and VOCs (Volatile Organic Compounds) in the atmosphere which produces O₃. NOₓ therefore has both direct and indirect environmental effects. It also reacts with hydroxyl radicals in the atmosphere to produce HNO₃, which is deposited as acid rain or nitrates (Steinkamp
et al., 2008; Vitousek et al., 1997); global NO$_x$ deposition rates were calculated by Galloway et al. (2004) to be 45.8 Tg N yr$^{-1}$ which mean that most of the NO$_x$ emitted to the atmosphere is removed by deposition processes. Indirectly, the formation of ozone has serious effects on humans and on plant life especially in industrial areas where NO$_x$ concentrations tend to be high.

NH$_3$ has a short atmospheric lifetime due to the high reactivity of the gas in the atmosphere. The residence time in the atmosphere for NH$_3$ is approximately 10 days. Sources include: biomass burning, natural soils, fertilized soils and industrial sources. Anthropogenic sources account for between 70 and 80 percent of emissions. NH$_3$ acts as an acid neutralizing agent in the atmosphere and reacts with acids to produce ammonium ($\text{NH}_4^+$) which is then removed from the atmosphere by either wet deposition (dissolved in rain) or as dry particles on areas not very far from where it was formed (Galloway et al., 2004; Schlesinger & Hartley, 1992; Vitousek et al., 1997). As with NO$_x$, most of the NH$_3$ emitted to the atmosphere (58 Tg N yr$^{-1}$) is removed via deposition processes (57 Tg N yr$^{-1}$)(Galloway et al., 2004).

### 2.3 Atmospheric transformations

The main chemicals that are important to the present study are: NO$_x$, NH$_3$, SO$_x$ and H$_2$S. Several of them participate in gas and aqueous phase reactions taking place within the troposphere and all of them are removed from the atmosphere via deposition processes. With several thousand chemical reactions taking place simultaneously in the troposphere, the chemistry can become quite complex (Pienaar & Helas, 1996).

#### 2.3.1 Formation of atmospheric oxidants

Most reactions that take place within in the troposphere are oxidation reactions. The chemicals responsible for this are oxidants like the hydroxyl radical (HO$^-$), hydroperoxyl radical (HO$_2^-$) and the methylperoxy radical (CH$_3$O$_2^-$). The formation of these chemicals is important, because their atmospheric concentrations have a major influence on the oxidation rates of nitrogen and sulphur species. These radicals are produced through radical chain oxidation reactions. The photolysis of
ozone is considered the reaction that starts of the entire process. In the presence of sunlight, the ozone molecule is split into O₂ and reactive oxygen atoms. The reactive oxygen atoms react with water to produce hydroxyl radicals.

The hydroxyl radicals react mostly with carbon trace species such carbon monoxide and methane due to their higher concentrations relative to other hydrocarbons in the troposphere. Reactions with CO produce CO₂ and hydrogen radicals, while reactions with methane produce water and methyl peroxy radicals. These radicals in turn react with atmospheric oxygen producing hydroperoxyl and methyl peroxy radicals. Hydroxyl also plays an important role in the gas phase oxidation reactions of NO₂ and SO₂ in the troposphere and the formation of ozone.

### 2.3.2 Formation of ozone in the troposphere

Although ozone plays an important part in the formation of atmospheric oxidants, the formation of ozone is controlled by the atmospheric concentration of NO₂. NO₂ is produced by numerous natural, agricultural and industrial sources and generally has a short residence time (1 to 2 days), but it can be carried by reservoir species such as peroxyacetyl nitrate (PAN) much further away, affecting areas not close to the emission source.

NO₂ can absorb solar radiation and when it does it starts off a radical chain reaction that eventually produces O₃. The reaction involves hydroxyl and hydroperoxyl radicals (which are linked to photolysis of ozone) and atmospheric VOCs. A simplified reaction pathway is provided by Pienaar & Helas (1996) (equations 1 to 5):

\[
2(NO₂ + hv + O₂ → NO + O₃) \tag{1}
\]

\[
NMHC + HO^- + O₂ → RO_2^- \tag{2}
\]

\[
RO_2^- + NO + O₂ → NO₂ + HO_2^- + CARB \tag{3}
\]

\[
HO_2^- + NO → HO^- + NO₂ \tag{4}
\]
NHMC represents non-methane hydrocarbons of the VOCs present in the troposphere, and CARB represents carbonyl compounds. This is also the only known way ozone is formed in the troposphere.

### 2.3.3 Gas phase oxidation processes

The most important pollutants in power station flue gas are composed of $\text{SO}_x$ (mostly $\text{SO}_2$) and $\text{NO}_x$ ($\text{NO} + \text{NO}_2$) gases which are released directly into the atmosphere. $\text{NO}_x$ gases are composed mostly of $\text{NO}$ (approx. 95%) and $\text{NO}_2$ (approx. 5%). Once it is released the atmosphere the NO is oxidised to $\text{NO}_2$ by reacting with either $\text{O}_3$ or the peroxy radicals mentioned above. Overall $\text{NO}_2$ has three main reaction pathways to $\text{HNO}_3$. The first is by reacting with hydroxyl during the day to form $\text{HNO}_3$. The second pathway involves $\text{NO}_2$ reacting with $\text{O}_3$ to form $\text{NO}_3$ radical. $\text{NO}_3$ in turn can react with either with hydroxyl or with atmospheric organics to form $\text{HNO}_3$. (Hewit, 2001; Pienaar & Helas, 1996).

The oxidation of $\text{H}_2\text{S}$ in the atmosphere was initially very poorly understood. $\text{H}_2\text{S}$ does not absorb sunlight to undergo photo-oxidation and gas-phase reactions with $\text{O}_3$ happen too slowly to account for the short residence time of $\text{H}_2\text{S}$ in the troposphere (Hales et al., 1973; Cox & Sandalls, 1974; Eggleton & Cox, 1977).

The main oxidation pathway for $\text{H}_2\text{S}$ is by reacting with the hydroxyl radical to form $\text{HS}$ and $\text{H}_2\text{O}$. $\text{HS}$ reacts with either $\text{O}_3$ or $\text{NO}_2$ to form $\text{HSO}$. Both these reactions are very quick compared to first one, taking place within microseconds compared to at least 2 days for the first reaction. The next step is the oxidation of $\text{HSO}$ by the same atmospheric oxidants (30 s for $\text{O}_3$ and 4 s for $\text{NO}_2$) as in the previous step to form $\text{HSO}_2$, which is in turn very quickly oxidised by $\text{O}_2$ to form $\text{SO}_2$ and $\text{HO}_2$. From here on the reaction pathway is the same for any $\text{SO}_x$ molecule (Wang & Howard, 1990; Shooter, 1998).
The most prominent gas phase reaction path for SO\textsubscript{2} is being oxidised by hydroxyl to HOSO\textsubscript{2}, which takes about 13 days. From here on the reactions take place within microseconds. HOSO\textsubscript{2} reacts with O\textsubscript{2} to produce a hydroperoxyl radical and SO\textsubscript{3}. The SO\textsubscript{3} then reacts with water so form H\textsubscript{2}SO\textsubscript{4} (Hewit, 2001; Pienaar & Helas, 1996; Wang & Howard, 1990; Shooter, 1998). The sulphur reaction pathway is summarised in Figure 2.1.

### 2.3.4 Aqueous phase oxidation processes

Aqueous oxidation of NO\textsubscript{x} does not play as large a role as the gas phase oxidations reactions. The reaction constants at ambient conditions ensure that gas phase reaction will always dominate the conversion of NO\textsubscript{x} to HNO\textsubscript{3} in the troposphere.

For SO\textsubscript{x}, aqueous phase reactions cannot be ignored as in the case for NO\textsubscript{x}. In water droplets dissolved SO\textsubscript{2} exist as various species of the same oxidation state S(IV). The reaction paths for the three species are shown below.

![Figure 2.1 Atmospheric Sulphur Oxidation (Wang & Howard, 1990)](image-url)
All of them are in equilibrium with each other and the concentration of each species is dependent on the acidity of the solution shown below in equations 6 to 7:

\[
SO_2 + H_2O \rightleftharpoons SO_2 \cdot H_2O
\]  
(6)

\[
SO_2 \cdot H_2O \rightleftharpoons HSO_3^- + H^+
\]  
(7)

\[
HSO_3^- \rightleftharpoons SO_3^{2-} + H^+
\]  
(8)

In solution S(IV) is always oxidised to the oxidation state of S(VI). S(VI) exist for the most part as H_2SO_4 or SO_2. There are three reaction pathways which are relevant when considering aqueous oxidation of SO_2.

The first is the reaction of S(IV) with dissolved O_2. This reaction pathway is relatively slow compared to the reaction pathways discussed below, even when catalysed by metal catalyst such as Fe(III) or Mn(II).

The next pathway is the reaction with dissolved ozone. As gases SO_2 and ozone do not react with each other, but when dissolved in water they react sufficiently to be considered an important reaction pathway.

The last pathway is the reaction of S(IV) with dissolved hydrogen peroxide (H_2O_2). Both these reactions are pH dependant. At a pH less than 5 the reaction with H_2O_2 dominates, while at a pH higher than 5 the reaction with ozone will dominate the hydrogen peroxide reaction (Hewit, 2001; Pienaar & Helas, 1996).

2.4 Removal of nitrogen and sulphur species from the atmosphere

NH_3 acts as an acid neutralising agent in the troposphere and readily reacts with both acids, HNO_3 and H_2SO_4, formed through the oxidation of NO_x and SO_x. The salts produced by this neutralising reaction: NH_4NO_3 and (NH_4)_2SO_4) exist either as aerosol or solid particles in the troposphere. Ammonium sulphate tends to be formed first due to its lower vapour pressure compared to ammonium nitrate. These reactions are very fast and the residence times for the salts are very low, so not very much of the NH_3 is oxidised by the hydroxyl radical. The above sulphur and nitrogen
species are continually removed by wet and dry deposition processes (Hewit, 2001; ApSimon et al. 1987; Ferm, 1998).

2.4.1 Atmospheric Deposition

NO\textsubscript{x} and SO\textsubscript{x} trace gases and salts are deposited from the atmosphere on to surfaces ranging from plants to soils. The mechanisms by which this happens are called wet or dry deposition. Although wet deposition is very effective for removing trace gases and particles from the atmosphere, it happens sporadically and mostly in summer on the Highveld. In the winter season, due to lack of rain dry deposition plays a far more important role than wet deposition (Zunckel et al., 1996; Scorgie and Kornelius, 2009).

2.4.2 Dry deposition

A number of factors need to be taken into account when determining dry deposition rates and this can be a complicated process. Direct measurement techniques tend to be very expensive and can be difficult to implement. Historically the Inferential method has been used to estimate dry deposition on the Highveld indirectly (Turner et al., 1995; Zunckel et al., 1996; Scorgie and Kornelius, 2009; Ferguson, 2009).

This method quantifies the vertical pollutant deposition flux (\(F\)) (gm\textsuperscript{-2}s\textsuperscript{-1}) by using the deposition velocity (\(V_d\)) (m/s) and pollutant concentration (\(C\)) (gm\textsuperscript{-3}). \(C\) and \(V_d\) are functions of the height above ground level. By convention downward flux is negative. The equation relating \(F\) with \(V_d\) and \(C\) is shown below (equation 9) (Turner et al., 1995; Zunckel et al., 1996; Scorgie and Kornelius, 2009; Seinfeld & Pandis, 2006).

\[
F = -V_dC
\]  

(9)

The deposition velocity term \(V_d\) is used to quantify a range of processes affecting gases and particles as they travel from the atmosphere to the surface of an object. These processes can be represented as electrical resistances coupled in series. The main resistances important in gas and particle transport are defined below:
R_a = aerodynamic resistance: takes into account atmospheric processes affecting transport of gases and particles through the atmosphere from the point of emission.

R_b = boundary resistance: takes into account transport of particle and gases across a stagnant layer of air.

R_c = canopy resistance: all uptake processes controlling particles and gases adhering to different types of surfaces.

A total resistance (equation 10) is then calculated from the sum of all the resistance mentioned above. The total R is then inverted to calculate V_d (Turner et al., 1995; Zunckel et al., 1996; Scorgie and Kornelius, 2009; Ferguson, 2009; Seinfeld & Pandis, 2006):

\[ V_d = 1/(R_a + R_b + R_c) \] (10)

Typical V_d values are shown below in Table 2.1.

**Table 2.1 Dry deposition velocities (V_d) from literature (Scorgie and Kornelius, 2009).**

<table>
<thead>
<tr>
<th>Species</th>
<th>Surface</th>
<th>Vd (cms(^{-1}))</th>
</tr>
</thead>
<tbody>
<tr>
<td>SO(_2)</td>
<td>Grass</td>
<td>0.06 – 3.5</td>
</tr>
<tr>
<td></td>
<td>Alfalfa</td>
<td>0.7 – 2.8</td>
</tr>
<tr>
<td></td>
<td>rough grazing</td>
<td>0.7</td>
</tr>
<tr>
<td></td>
<td>Wheat</td>
<td>0.3 – 1.5</td>
</tr>
<tr>
<td></td>
<td>medium crop</td>
<td>0.7</td>
</tr>
<tr>
<td></td>
<td>barley leaves</td>
<td>1.5</td>
</tr>
<tr>
<td></td>
<td>rye grass</td>
<td>0.62</td>
</tr>
<tr>
<td></td>
<td>Vegetation</td>
<td>2.8</td>
</tr>
<tr>
<td></td>
<td>Soil</td>
<td>0.19 – 1.2</td>
</tr>
<tr>
<td></td>
<td>bare soil (dry)</td>
<td>0 – 1.5</td>
</tr>
<tr>
<td></td>
<td>base soil (wet)</td>
<td>0 – 1.5</td>
</tr>
<tr>
<td></td>
<td>acid soil (dry)</td>
<td>0.4</td>
</tr>
<tr>
<td></td>
<td>acid soil (wet)</td>
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</tr>
<tr>
<td></td>
<td>calcareous soil</td>
<td>0.8</td>
</tr>
<tr>
<td></td>
<td>Land</td>
<td>1.2 – 1.8</td>
</tr>
<tr>
<td></td>
<td>Forest</td>
<td>1.4</td>
</tr>
<tr>
<td></td>
<td>water (fresh)</td>
<td>0.5 – 2.2</td>
</tr>
<tr>
<td></td>
<td>Ocean</td>
<td>0.5</td>
</tr>
<tr>
<td></td>
<td>ocean (calm)</td>
<td>0.2</td>
</tr>
<tr>
<td>Species</td>
<td>Surface</td>
<td>Vd (cms(^{-1}))</td>
</tr>
<tr>
<td>------------------</td>
<td>----------------------------------</td>
<td>--------------------</td>
</tr>
<tr>
<td>ocean (turbulent)</td>
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</tr>
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<td>mixed forest</td>
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<td>1.8 – 3.7</td>
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<tr>
<td>Corn</td>
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<td>pine trees/forest</td>
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</tr>
<tr>
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</tr>
<tr>
<td></td>
<td>fresh water</td>
<td>0.07</td>
</tr>
<tr>
<td></td>
<td>Ocean</td>
<td>0.04 – 0.05</td>
</tr>
<tr>
<td></td>
<td>dry grass</td>
<td>0.13</td>
</tr>
<tr>
<td></td>
<td>bare soil (saturated)</td>
<td>0.05 – 0.24</td>
</tr>
<tr>
<td></td>
<td>base soil (dry)</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Lake</td>
<td>0.005 – 0.04</td>
</tr>
<tr>
<td></td>
<td>pine forest</td>
<td>0 – 1.16</td>
</tr>
<tr>
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<td>NO(_2)</td>
<td>Vegetation</td>
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</tr>
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<td>HNO(_3)</td>
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<td></td>
<td>Grass</td>
<td>0 – 4.9</td>
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<tr>
<td></td>
<td>Wheat</td>
<td>2.9 – 4.9</td>
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<td>coniferous forest</td>
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<tr>
<td></td>
<td>Grass</td>
<td>0.02 – 2.3</td>
</tr>
<tr>
<td>Particle diameter:</td>
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<td></td>
</tr>
<tr>
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<td>Grass</td>
<td>0.03</td>
</tr>
<tr>
<td>1</td>
<td>Grass</td>
<td>0.03</td>
</tr>
<tr>
<td>2</td>
<td>Grass</td>
<td>0.1</td>
</tr>
<tr>
<td>0.8</td>
<td>bean leaves</td>
<td>0.0035</td>
</tr>
<tr>
<td>0.2</td>
<td>short grass</td>
<td>0.02</td>
</tr>
</tbody>
</table>
2.4.3 Wet Deposition (Scire et al., 2000)

Wet deposition is an important pathway by which gaseous $\text{SO}_2$ and salt particles are removed from the atmosphere. Gases are scavenged by dissolving into droplets and precipitation, while particles are either removed within clouds (“rainout”) or can be washed out by precipitation. In CALPUFF wet deposition is estimated by an empirical scavenging method. In equation 11 below, $X$ represents the pollutant concentration at time $t$ and $t+\Delta t$ and $\Lambda$ represent the scavenging ratio:

$$X_{t+\Delta t} = X_t \exp[-\Lambda\Delta t] \quad (11)$$

The scavenging ratio is calculated from equation 12:

$$\Lambda = \lambda \left( \frac{R}{R_1} \right) \quad (12)$$

Where $\lambda$ is the scavenging coefficient ($\text{s}^{-1}$) and is determined through laboratory and field measurements, $R$ is the precipitation rate ($\text{mm hr}^{-1}$) and $R_1$ is a reference precipitation rate at 1 mm hr$^{-1}$. Some basic pollutants scavenging coefficient values provided by Scire et al (2000) are summarised in Table 2.2 below.
Table 2.2 Scavenging coefficients for common nitrogen and sulphur species (Scorgie and Kornelius, 2009).

<table>
<thead>
<tr>
<th>Species</th>
<th>Scavenging coefficient, $\lambda$ (s$^{-1}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Liquid Precipitation</td>
</tr>
<tr>
<td>SO$_2$</td>
<td>$3 \times 10^{-5}$</td>
</tr>
<tr>
<td>Sulphate</td>
<td>$1 \times 10^{-4}$</td>
</tr>
<tr>
<td>NO$_x$</td>
<td>0</td>
</tr>
<tr>
<td>HNO$_3$</td>
<td>$6 \times 10^{-5}$</td>
</tr>
<tr>
<td>Nitrate</td>
<td>$1 \times 10^{-4}$</td>
</tr>
</tbody>
</table>

2.4.4 Techniques employed on the South African Highveld to quantify deposition.

Various techniques have been employed over the years to study nitrogen and sulphur deposition on the Highveld. Since the middle 80’s, due to the presence of large power stations run by Eskom, monitoring stations network run by both the CSIR (Council for Scientific and Industrial Research) and Eskom were established (Scorgie and Kornelius, 2009). Located across several provinces downwind of the industrial Highveld at places like Ermelo, Amersfoort, Piet Retief, Warden, Vryheid, Ladysmith and also as far north as Louis Trichardt these stations forms a large network. These stations have been used extensively in the study of deposition on the Highveld and include studies done by Bluff et al. (1991), Turner et al. (1993), Held and Mphepya (2000) and Galpin and Turner (1999a; 1999b). This usually involved analysing rainfall chemistry, where nitrate and sulphate species concentrations within rain water were quantified from which deposition values are calculated. But this technique is mostly used in determining wet deposition rates.

Other rainfall studies include that of Snyman et al. (1991), Zunckel et al. (1994), van Tienhoven et al. (1995) where rainfall chemistry from 16 sites was recorded. All the major chemical species were expressed as volume weighted concentrations. Deposition rates were then calculated by multiplying mean rainfall coverage with the mean volume weighted concentrations.
In 1993 Wells did a study in which long term SO$_2$ concentrations for the Mpumalanga Highveld were used to estimate dry deposition rates. In this study, he recommends that the best method for estimating dry deposition rates is the inferential method. Several later studies made use of inferential techniques to calculate dry deposition rates. These include studies done by Piketh and Annegarn (1994), Mphepya and Held (1999, 2000), Zunckel et al. (1996), Zunckel (1998), Zunckel et al. (1999), Zunckel (1999) and Galy-Lacaux et al. (2003). This technique makes use of hourly concentrations of NO$_X$ and SO$_X$, O$_3$ and particulate concentrations. Meteorological parameters like temperature, wind speed, wind direction, sigma theta and total radiation are also required. Other parameters included are ground cover and leaf area index values, which varies as the seasons change.

Several attempts at modelling sulphur and nitrogen deposition have been made. In 2000 Zunckel et al. modelled ambient SO$_2$ and sulphur species deposition by using the MATCH modelling system. The system is a three-dimensional Eulerian multi-layered model that includes vertical and horizontal transport, vertical diffusion, atmospheric chemical transformations, dry deposition and wet scavenging. The model requires as input meteorological information, chemical transformations and emission sources. From these inputs the model estimates ambient SO$_2$ concentrations. These SO$_2$ concentrations together with deposition velocities and wet scavenging rates are used to determine dry and wet deposition fluxes of SO$_2$ and particulate sulphur.

Scorgie et al. (2002) used CALPUFF to model sulphur deposition over the Highveld. CALPUFF taking into account emission sources, chemical transformations, topography and meteorology to estimate deposition rates. Fourie et al. (2005) used the Lagrangian-Eulerian Diffusion (LED) model to model long range transport and chemical transformations of pollutants across southern Africa. The model could include boundary layer dynamics to account for turbulent diffusion processes as well as chemical transformations, dry and wet depositions.
2.4.5 Summary of measured Sulphur and Nitrogen deposition rates on the South African Highveld.

2.4.5.1 Sulphur

Sulphur deposition has been studied extensively on the South African Highveld. One of the earliest studies was done by Bluff et al. in (1991), who collected data from Eskom’s acid rain network from 1985 to 1990 to calculate annual average wet sulphur deposition within the Highveld. Values ranged from 5.8 kg S ha\(^{-1}\) yr\(^{-1}\) (central Highveld) to 4.7 kg S ha\(^{-1}\) yr\(^{-1}\) on the south-eastern periphery.

Turner et al. (1993) also made use of Eskom’s acid rain network. Six stations were located downwind of the industrial part of the Highveld (Ermelo, Amersfoort, Piet Retief, Warden, Vryheid and Ladysmith) and one far north at Louis Trichardt. This study was seven years long, from 1986 to 1992. They reported a maximum average SO\(_4\) wet deposition flux of 20 kg S ha\(^{-1}\) yr\(^{-1}\) located south of Middelburg/Witbank area, which decrease to less than 16 kg S ha\(^{-1}\) yr\(^{-1}\) in the south of Mpumalanga. They also concluded from the same study that rainfall acidity was mostly influenced by fossil fuel burning (50 to 60%), burning of biomass (19 to 28%) and other sources (10 to 31%).

Mphepya and Held (1999, 2000) used measured wet deposition values for Amersfoort for 1996, 1997 and 1998 (4.64, 6.22, 4.21 kg S ha\(^{-1}\) yr\(^{-1}\)) to estimate wet deposition for Elandsfontein. Comparing these values to estimated dry deposition calculated by using an inferential method for this site they concluded that dry deposition exceeds wet deposition by a factor of roughly 2. Scorgie et al. (2002) predicted wet sulphur deposition for the same site to be 2.6 kg S ha\(^{-1}\) yr\(^{-1}\).

Dry deposition flux rates are influenced not only by ambient atmospheric concentrations, but also by deposition velocities. Deposition velocities in turn are affected by a range of variables, which include solar radiation, photosynthetic activities of local plants, Leaf Area Index (LAI) and meteorological variables such as temperature. These properties are affected by the time of year and day, therefore deposition velocities exhibit diurnal and seasonal patterns. Two major studies done by Mphepya and Held (1999, 2000) and Zunckel (1999) both investigated dry deposition rates at the same locations of Elandsfontein (located near the centre of
the electricity generation activities on the Highveld) and Palmer (towards the north-eastern edge of these activities).

Both made use of the Inferential Method, where atmospheric concentration of SO$_2$, NO$_2$, NO$_x$, O$_3$, sulphate and meteorological parameters were recorded and inputted into a model from which dry deposition flux rates were determined.

Mphepya and Held (1999) found that average inferred SO$_2$ deposition velocities were larger during summer than winter. They attributed this to increased LAI values, solar radiation and photosynthesis rates during summer. Annual dry sulphur deposition at Elandsfontein was found to be in a range of 8.9 kg ha$^{-1}$ to 10 kg ha$^{-1}$ with dry SO$_2$ the most prominent component and is responsible for between up72 to 92 % of the total estimated dry sulphur deposition. At Palmer, annual dry sulphur deposition was found to be 1.6 to 3 kg ha$^{-1}$, with dry SO$_2$ composing 21 to 66 % of the total dry deposition. These rates were estimated over a three-month period.

Zunckel (1999) found similar trends for dry sulphur deposition, in that the inferred velocities were larger during the summer compared to those for the winter (Scorgie and Kornelius, 2009). The total dry sulphur deposition flux at Elandsfontein was estimated at 13.1 kg ha$^{-1}$yr$^{-1}$ with 83% due to SO$_2$ and the rest is sulphate related.

The seasonal variations in deposition velocities were attributed to there being greater turbulence and photosynthetic activity during the summer. During the summer, larger variations in day-to-day deposition velocities were also observed compared to deposition velocities recorded in winter. This was due to a more stable nocturnal boundary layer during winter. Other seasonal affects included higher ambient SO$_2$ concentrations during winter as compared to the summer.

Both gas and particulate deposition velocities were found to have similar diurnal variations, with minimum values during the night and maxima during the day. This can be seen below in Figure 2.2.
2.4.5.2 Nitrogen

Galy-Lacaux et al. (2003) used the same techniques discussed previously to estimate dry (inferential model using ambient concentrations of NH$_3$ and NO$_2$) and wet (precipitation concentrations of NO$_3^-$ and NH$_4^+$ with annual mean rainfalls) deposition rates at Amersfoort and Louis Trichardt. Amersfoort was classified as an industrial site and was predicted to receive 15 kg N ha$^{-1}$ yr$^{-1}$. 63% of this was due to wet deposition, while 37% was due to dry deposition. Due to Louis Trichardt being very far away from the Highveld industrial area it received only 9 kg N ha$^{-1}$ yr$^{-1}$. The composition of this was 59% dry deposition and 41% wet deposition.

In 2003 Lowman estimated that total wet deposition for the Highveld ranged from 6.5 to 10.6 kg N ha$^{-1}$ yr$^{-1}$, with an average of 7.84 kg N ha$^{-1}$ yr$^{-1}$. They found nitrate and ammonium to be the most important ions within wet deposition, with 53% nitrate and 47% ammonium. Galy-Lacaux et al. reported similar values for Amersfoort with ammonium forming 42% and nitrate 58% of wet N depositions.
Another study done by Collet et al. in 2010 at Elandsfontein, to assess the atmospheric nitrogen budget for the South African Highveld, reported total dry N deposition of 0.18 to 6.6 kg N ha\(^{-1}\) yr\(^{-1}\) and total N deposition of 2.4 to 8.8 kg N ha\(^{-1}\) yr\(^{-1}\), but these values only included NO, NO\(_2\) and NO\(_3\). By incorporating NH\(_3\) data from the earlier Lowman study mentioned above, the total dry N deposition increased to 4.5 to 10 kg N ha\(^{-1}\) yr\(^{-1}\) and total N deposition to 6.7 to 13.1 kg N ha\(^{-1}\) yr\(^{-1}\).

### 2.4.6 Combined acid deposition measurements

In 2011 Josipovic et al. calculated total acidic deposition rates as part of a study assessing of critical load exceedances over the Highveld area. Use was made of 37 passive sampling technology, as applied in the International Geosphere Atmosphere Trace Species (IGAC-DEBITS-Africa/IDAF) network. Dry deposition and wet deposition were calculated using inferential methods. Deposition for two time periods were reported: September 2005 to August 2006 and September 2006 to August 2007. They reported total acidic values of 23.5 kg ha\(^{-1}\) yr\(^{-1}\) and 15.8 at Elandsfontein, 15.2 kg ha\(^{-1}\) yr\(^{-1}\) and 11.3 kg ha\(^{-1}\) yr\(^{-1}\) at Standerton and 19 kg ha\(^{-1}\) yr\(^{-1}\) and 11.4 kg ha\(^{-1}\) yr\(^{-1}\) at Amersfoort.

### 2.5 Nitrogen Soil Emissions

On a global scale combustion of fossil fuel is the largest source of nitrogen oxides emissions, but agriculture and natural sources are also an important source of atmospheric NO\(_x\). There is some uncertainty as to the scale of global biogenic NO\(_x\) emissions. Numerous estimates have been attempted. Signor and Cerri (2013) provide a review. A study by Yienger and Levy (1995) estimated that globally, soil NO\(_x\) emissions are 5.5 (3.3 – 7.7) Tg N yr\(^{-1}\). 40% of their estimate was due to anthropogenic activities. The main biogenic emitters of NO\(_x\) were agriculture (41%), grasslands (35%) and rain forest (16%).
In 1997 Davidson & Kingerlee estimated that total global NO\textsubscript{x} soil emissions were 21 Tg N yr\textsuperscript{-1} with an uncertainty of at least 4 to 10 Tg yr\textsuperscript{-1}. They also state that agriculture was an important source and that biogenic emissions contribute between 10% and 40% of NO\textsubscript{x} globally. Galloway et al. (2004) estimated that global NO\textsubscript{x} emissions are between 42 and 47 Tg N yr\textsuperscript{-1}, with anthropogenic emissions (due to energy and food production) making up the majority (36 Tg N yr\textsuperscript{-1}). In 2014 Vinken et al. estimated global NO\textsubscript{x} emissions at 12.9 ± 3.9 Tg N yr\textsuperscript{-1}, which are inferred from OMI (Ozone Monitoring Instrument) tropospheric NO\textsubscript{2} columns.

In contrast to NO\textsubscript{x} the largest sources for N\textsubscript{2}O globally is the soil, which produces about 9 – 11.5 Tg yr\textsuperscript{-1} (about 57% of global atmospheric N\textsubscript{2}O emissions) with agricultural sources producing about 3.5 Tg N\textsubscript{2}O yr\textsuperscript{-1} (Skiba & Smith, 2000, IPCC 1997, Scholes et al., 1997).

Total biogenic NO\textsubscript{x} emissions from southern Africa are estimated to be about 1 Tg yr\textsuperscript{-1}. Due to long dry seasons and low moisture contents of the soil, NO is produced rather than N\textsubscript{2}O and due to NO’s short residence time in the atmosphere it is expected that biogenic emissions will play a more important role on a regional scale (Feig 2009, Scholes and Scholes, 1998).

As with the nitrogen oxide species emissions, food production also plays a major part in NH\textsubscript{3} emissions as it contributes more than 50% of the global emissions. Globally total NH\textsubscript{3} emissions are about 59 million tons N yr\textsuperscript{-1} (FAO, 2001).

### 2.5.1 Soil nitrogen cycle

For the most part the primary processes that affect nitrogen contained within the soils are microbial by nature. These processes include the oxidation or reduction of nitrogen compounds such as NH\textsubscript{3}, NH\textsubscript{4}\textsuperscript{+}, NO, NO\textsubscript{2}\textsuperscript{-} and organic nitrogen compounds contained within dead plant, animal and microbial matter. These nitrogen species are referred to as reactive Nitrogen (N\textsubscript{r}), since it is they that take part in the chemical reactions within the soil.
The main process pathways controlling N\textsubscript{r} content within soils are removal processes (denitrification and leaching), conversion processes (ammonification, nitrification, net mineralization and immobilization) and processes that allow new nitrogen compounds to enter the soil system (biological nitrogen fixation, deposition and the addition of fertilizers) (see Figure 2.3). Due to the importance of emissions of NO and N\textsubscript{2}O by soil, the two most important processes for this study are nitrification and denitrification. Both produce NO as a required intermediary and are responsible for the production of N\textsubscript{2}O under specific anoxic conditions. Nitrification is the biological oxidation of NH\textsubscript{4}\textsuperscript{+} to NO\textsubscript{2}\textsuperscript{-} and finally to NO\textsubscript{3}\textsuperscript{-}, while denitrification is the respiratory reduction of NO\textsubscript{3}\textsuperscript{-} and NO\textsubscript{2}\textsuperscript{-} to N\textsubscript{2}O, NO and N\textsubscript{2} in the absence of oxygen (Feig, 2009; Ludwig et al., 2001; Hutchinson, 2012).

2.5.2 Biogenic emissions mechanisms
The main environmental conditions that control the rate at which NO and N\(_2\)O are released to the atmosphere and the rates at which nitrification and denitrification take place are soil moisture, soil temperature and the soil nutrients status.

Figure 2.4 Production of nitrogen gas species as a function of WFPS (Kirkman, 2000)

### 2.5.2.1 Soil Moisture

Water plays an important role within the soil environment and is usually quantified as Water Filled Pore Space (WFPS). It creates micro sites within the soil, which are either aerobic or anaerobic which can exist very close to each other. This makes it possible for nitrification and denitrification to occur simultaneously within the soil and affects the rates at which NO and N\(_2\)O are produced. Water within soils also affects the diffusive transport properties of the soil, which in turn affect the rates at which reactants and products of denitrification and nitrification leave or enter the soil.

Figure 2.4 shows how WFPS has an effect on the production of nitrogen gaseous species and the rates at which they are produced as the percentage WFPS increases. As WFPS goes up less oxygen is available until finally anaerobic conditions exist and only N\(_2\)O and N\(_2\) are produced.

It has been observed in many studies that after the first rain of the rain season large pulses of NO and N\(_2\)O follows. This pulse can last anything from hours to a week.
Rain pulsing makes 1.3 Tg N yr$^{-1}$ globally (Delmas et al., 1997; Yienger & Levy, 1995; Scholes et al., 1997).

### 2.5.2.2 Soil Temperature

Nitrification and denitrification are biological processes and are connected to microbial enzymatic processes which are affected by temperature. Therefore, soil temperature affects the rates at which NO is produced and several studies (Ludwig et al., 2001, Feig, 2009) have shown that this is indeed the case. In Figure 2.5 the net flux rate of NO was measured as WFPS increased at three different temperatures. As the temperature increases, so does the net flux of NO.

![Figure 2.5 Net flux of NO as a function of WFPS at different temperatures (Ludwig et al., 2001)](image)

Because of this sensitivity to temperature NO emission also shows diurnal patterns as atmospheric temperatures, to which surface soil temperature is closely related, vary throughout a 24-hour period. In Figure 2.6 net flux rate of NO is shown to change with the time of day. The bulk of NO emissions will take therefore place during the day.
It should be noted that soil temperature plays a larger role in short term emissions, while the other factors play a larger role on the long term (Ludwig et al., 2001; Feig 2009).

![Figure 2.6 The net flux of NO over a 24 hour period. Also shown is the soil temperature (Ludwig et al., 2001)](image)

### 2.5.2.3 Soil nutrients

The amount of soil nutrients available to be converted by microorganisms into NO or N\textsubscript{2}O plays a significant role in biological emissions. In agriculture, large amounts of mineral nitrogen compounds are added either by the application of fertilizers or by manure and urine from grazing animals. This in turn stimulates large fluxes of NO and N\textsubscript{2}O from agricultural soils. Ammonia-based fertilizers and urea, after being hydrolysed to NH\textsubscript{3}, act as substrate for nitrifying bacteria. The final product of nitrification is NO\textsubscript{3}\textsuperscript{-}. Ammonia-based fertilizer can serve as an indirect source of substrate for denitrifying bacteria, while NO\textsubscript{3}\textsuperscript{-} based fertilizers can only be used by denitrification bacteria. (Skiba& Smith, 2000; IFA 2001; Ludwig et al., 2001)

It should be noted that the relationship between substrate pools and NO/N\textsubscript{2}O fluxes is complex. In wet soils, because of the anoxic environment, denitrification dominates and NO\textsubscript{3}\textsuperscript{-} pool size is an important indicator for NO/N\textsubscript{2}O fluxes. In dry soils nitrification likely dominates and NH\textsubscript{4}\textsuperscript{+} pool size is a stronger indicator for NO/N\textsubscript{2}O fluxes (Ludwig et al., 2001; Feig, 2009).
2.5.3 Measured emission rates

Biogenic soil emissions have been recognised as important sources of NO and N\textsubscript{2}O since the early 90’s. Two major ecosystems that are present within South Africa are the grasslands of the Highveld, situated east and south of Pretoria and semi-arid savanna north and west of Pretoria.

2.5.3.1 Semi-arid savannas

Savannah ecosystems cover about 11.5% of the global surface and they have been identified as large sources of NO (as much as 40% globally) (Feig, 2008; Scholes, 1997). The general characteristic of savannas ranges from grasslands with scattered woody plants to woodlands with heterogeneous grass cover (Bustamante, 2006). Davidson & Kingerlee (1997) calculated that savanna/woodland biomes have NO fluxes ranging of 0.1 – 10 kg N ha\textsuperscript{-1} yr\textsuperscript{-1} with a mean flux of 3.1 kg N ha\textsuperscript{-1} yr\textsuperscript{-1} and contribute 7.4 Tg N yr\textsuperscript{-1}. Globally semi-dry savannas produce N\textsubscript{2}O at a rate of 1 Tg N yr\textsuperscript{-1} (IPCC, 2001).

NO and N\textsubscript{2}O soil emissions in semi-dry savanna ecosystems have been intensively studied in southern Africa due to the uncertainty and the lack of knowledge pertaining to semi-dry savannas biomes. At Nylosvley Scholes et al. (1997) did a study to determine the flux rates of NO and N\textsubscript{2}O after the first seasonal rains. A ‘pulsing’ effect is usually observed after the occurrence of the first seasonal rain due to build-up of nitrogen substrates such as ammonium and nitrates in the soil during the dry winter periods. The dry soil produced a NO flux of 2 ng N-NO m\textsuperscript{-2} s\textsuperscript{-1} before it is wetted. Adding the water increased the NO flux to between 76± 54 ng N–NO m\textsuperscript{-2} s\textsuperscript{-1} for the nutrient poor soils and 65 ± 26 ng N–NO m\textsuperscript{-2} s\textsuperscript{-1} for the nutrient rich soils. After a period of up to 72 hours the emission rate dropped to between 10 and 30 ng N–NO m\textsuperscript{-2} s\textsuperscript{-1}. N\textsubscript{2}O emissions were very low from the dry soil and after wetting the soil the N\textsubscript{2}O peaked between 3.4 and 9.7 ng N N\textsubscript{2}O m\textsuperscript{-2} s\textsuperscript{-1}, although it dropped quickly to 0.1 – 0.9 ng N N\textsubscript{2}O m\textsuperscript{-2} s\textsuperscript{-1}. After estimating the annual NO emission using
average NO emissions the ‘pulsing’ effect only contributed about 6% of the annual emissions.

Feig et al. (2008) did a similar study in the Kruger National Park. NO emissions were measured across different landscape positions. The entire Skukuza land system was divided into four landscapes positions: Footslope, Downslope, Upslope and Crest. This was done because the landscape position affects the water content of the soil and how nutrients accumulate in these micro-environments (Feig, 2009). Once the net flux rate of each type of landscape position had been determined, it was scaled up to include the entire Skukuza land system, which is about 56000 ha. Rainfall patterns, WFPS measurements and soil temperature were tracked from 2003 to 2005 for each type of landscape position and were then used to calculate NO fluxes for each landscape for the years the measurements were made. The NO fluxes range between 0.2 and 0.8 kg N ha\(^{-1}\) yr\(^{-1}\). Total mass of nitrogen lost from the Skukuza land system for each year was 33.7 Mg, 20 Mg and 27.4 Mg for 2003, 2004 and 2005 respectively.

### 2.5.3.2 Temperate and Semi-arid grasslands emission studies

Although there has been no attempt at determining NO and N\(_2\)O flux rates on the Highveld grassland, internationally grasslands have been studied extensively. In their attempt to estimate global NO emissions Davidson & Kingerlee (1997) included 11 studies done on temperate grassland. They concluded that globally temperate grasslands account for 1.1 Tg N yr\(^{-1}\) with a mean flux of 1.2 kg N ha\(^{-1}\) yr\(^{-1}\) ranging from 0 – 1.8 kg N ha\(^{-1}\) yr\(^{-1}\). According to the Intergovernmental Panel on Climate Change (IPCC, 2001) globally grassland releases between 0.5 – 2.0 Tg N per year of N\(_2\)O.

The majority of South African coal-based power generation is located on the South African Highveld, which results in large amounts of acid deposition which in turn (Scorgie and Kornelius 2009) can lead to leaching of nutrients in sensitive soils. Several studies have been done recently to understand the effects of sulphur and nitrogen deposition. In an attempt to quantify the effects that acid deposition has on
the Highveld soil nutrient cycling processes, Hutchinson (2012) conducted a study in which monthly soil, vegetation and litter samples were collected for an eight-month period at the Sandspruit catchment in the southern Mpumalanga. This was used to construct a sulphur and nitrogen budget for the soil of the area. Three nutrient budget scenarios were calculated corresponding to the minimum, average and maximum deposition rates modelled on the Highveld. Due to lack of published literature on Highveld grasslands, a study on savannah was used to calculate nitrogen losses through gaseous emissions. It was assumed that nitrogen losses through denitrification was 5% of the measured mineralization rates for each scenario and that these are represented as a combined NO and N\(_2\)O flux value. The calculated emission rates for the minimum, average and maximum are 0 kg N ha\(^{-1}\) yr\(^{-1}\), 2.2 kg N ha\(^{-1}\) yr\(^{-1}\) and 13.7 kg N ha\(^{-1}\) yr\(^{-1}\) respectively. The range of these estimates was far larger than that reported by Davidson & Kingerlee, although the average deposition compares to the reported value.

### 2.5.4 Agricultural emissions studies

With more than 50% of the world’s food crops being fertilized to yield better growth, agriculture produces large amounts of NO and N\(_2\)O. About 41% of global soil NO\(_x\) emissions are due to agriculture (Skiba, 1997; Yienger & Levy, 1995). Only about 50% of the nitrogen introduced through fertilization is used by the plants. The rest is lost through numerous processes. One of the main mechanisms by which the excess nitrogen leaves these systems is by being emitted in some form into the atmosphere (FAO, 2001). According to Davidson & Kingerlee (1997), cultivated land in temperate climates releases about 1.8 Tg N yr\(^{-1}\) with a mean flux of 3.6 kg ha\(^{-1}\) yr\(^{-1}\) ranging from 0.2 to 23 kg N ha\(^{-1}\) yr\(^{-1}\). As with natural soils WFPS and soil temperature play an important role in controlling NO and N\(_2\)O emission rates. The rate of fertilizer application is important, as the largest emission rates are observed right after the fertilizer has been added to the soil. These can last from a couple of days to weeks. These emissions are also dependant on the type of fertilizer used. Urea and ammonium nitrate (NH\(_4\)NO\(_3\)) are the most used fertilizers globally. These two fertilizers account for 64% of global N application. Urea is the cheapest form of fertilizer available and is mostly used in Asia and South America, while NH\(_4\)NO\(_3\) is mostly used in Europe and anhydrous NH\(_3\) in North America (Skiba et al., 1997).
Cultivation methods, such as tillage has been shown to stimulate biogenic emission. Land use plays an important role. Maize crops produce over 40% of total biogenic NO in the USA, while only using 4% of the land. The Free State and Mpumalanga provinces are responsible for a large part of South Africa’s food production. The dominant crops being grown in either province is maize, with 95% of crops being fertilized (FAO, 2005). It is therefore expected to be an important source of NO and N₂O. Finally grassland that is used for grazing livestock receives extra N input through the excreted waste produced by the animals, which indirectly increases the nitrogen emissions of the land on which the grazing is taking place (Skiba et al., 1997). The Food and Agricultural organisation of the United Nations (FAO) estimated that in 1995 N fertilizer and manure application on crops globally produced 24 Gg of NO and 72 Gg of N₂O, while fields produced 14 Gg of NO and 13 Gg of N₂O emissions (FAO, 2001).

2.5.4.1 Ammonia volatilization

Common sources of ammonia in soil are ammonium yielding mineral and organic fertilizers as well ammonia formed through biological degradation. Ammonia exists as a gas and can volatilize to the atmosphere very easily. The most common reactions for ammonium are shown below. There are two end points, one where ammonium decomposes to ammonia gas, the other where it is nitrified by bacteria to NO₃⁻ (FAO, 2001):

\[
\begin{align*}
\text{NH}_4^+ \text{ (absorbed)} & \rightarrow \text{NH}_4^+ \text{ (solution)} \rightarrow \text{NH}_3 \text{ (solution)} \rightarrow \text{NH}_3 \text{ (atmosphere)} \quad (13) \\
\text{NH}_4^+ & \leftrightarrow \text{NH}_3 + H^+ \quad (14) \\
\text{NH}_4^+ + 1.5O_2 & \rightarrow 2H^+ + H_2O + NO_2 \quad (15) \\
NO_2^- + 0.5O_2 & \rightarrow NO_3^- \quad (16)
\end{align*}
\]

Environmental and soil conditions determine whether the ammonium decomposes to ammonia (equation 13) or is nitrified to NO₃⁻ (equations 14 - 16). The main environmental factor driving ammonia volatilization is the difference between the partial pressure of NH₃ in the soil or water within the soil, and that of the ambient atmosphere. The partial pressure of NH₃ is thus controlled by how quickly NH₃ is
removed in solution by a specific reaction, or by changing any of the equilibria. Increasing temperature decreases the solubility of NH$_3$ and increases the diffusion rate of NH$_3$ though soil. Nitrification has a twofold effect on NH$_3$ concentration within a watery solution. By converting NH$_4^+$ to NO$_3^-$ it removes any NH$_4^+$ that could be converted to NH$_3$. By producing H$^+$ it makes the solution more acidic, shifting the equilibrium reaction to produce more NH$_4^+$ (see reaction 14). Any compound added to the soil, which either absorbs the NH$_4^+$ or increases the acidity of the soil will therefore also affect the volatilization rate of NH$_3$.

In southern Africa NH$_3$ loss for 1995 has been calculated by the FAO (2001) at 20 000 t NH$_3$-N for fertilized grassland (about 25% N loss) and 23 000 t NH$_3$-N for upland crops (about 30% N loss). Volatilization loss of common fertilisers is shown below in Table 2.3.
Table 2.3 NH3 volatilization loss for mineral and animal fertilisers (adapted from IFA and FAO, 2001)

<table>
<thead>
<tr>
<th>Fertilizer Category</th>
<th>Volatilization Loss</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Mean (%)</td>
</tr>
<tr>
<td>Ammonium sulphate</td>
<td>16</td>
</tr>
<tr>
<td>Urea</td>
<td>21</td>
</tr>
<tr>
<td>Ammonium nitrate</td>
<td>6</td>
</tr>
<tr>
<td>Calcium ammonium nitrate</td>
<td>3</td>
</tr>
<tr>
<td>Ammonia, direct application</td>
<td>2</td>
</tr>
<tr>
<td>Nitrogen solutions</td>
<td>5</td>
</tr>
<tr>
<td>Other straight nitrogen</td>
<td>15</td>
</tr>
<tr>
<td>Ammonium phosphates</td>
<td>11</td>
</tr>
<tr>
<td>Other compounds NP-N</td>
<td>11</td>
</tr>
<tr>
<td>Compound NK-N</td>
<td>2</td>
</tr>
<tr>
<td>Compound NPK-N</td>
<td>9</td>
</tr>
<tr>
<td>Total mineral fertilizers</td>
<td>14</td>
</tr>
<tr>
<td>Animal manure</td>
<td>23</td>
</tr>
</tbody>
</table>

2.5.5 Modelling of Biogenic soil emission

There have been several attempts over the years to either model NO\textsubscript{x} emissions for a certain piece of land and/or to scale up those emissions to include even larger pieces of land i.e. to quantify emissions on a global scale. In his doctoral dissertation, Feig (2009) roughly divided them into five categories: empirical models, remote sensing, statistical models, process based models and local models based on soil measurements. The first category makes use of empirical methods due to large uncertainties in either knowing how the inherent biological processes function or in lack of experimental data. Different ecosystems are assigned an emission factor that can be affected by environmental parameters such as soil temperature and WFPS. Examples of empirical models are those done by Williams et al (1992), Yienger and Levy (1995), Davidson and Kingerlee (1997) and Steinkamp and Lawrence (2011).

Remote sensing models make use of the fact that NO\textsubscript{2} has a very good absorption band and due to the rapid conversion of NO to NO\textsubscript{2}, NO emissions can be inferred.
from NO₂ column densities. Jeagle et al. (2005) used this to map NOₓ emissions over Africa in 2000, Hudman et al. (2010) to study NOx soil emissions over the United States and Vinken et al. (2014) inferred global NOₓ emission from atmospheric NO₂.

Using relationships between measured NO fluxes and environmental variables such as WFPS, soil temperature and wind speed, statistical models must be trained to fit existing data sets to predict NOₓ emissions. Examples include models by Yan et al. (2005) and Delon et al (2007). In the case of Delon et al. a neural network was trained to find the best non-linear regression between NO fluxes and chosen environmental variables.

Process based models look at how nutrients move through an ecosystem. The different processes such as decomposition, nitrification and denitrification rates are determined through soil measurements, which are then added to the model. NO emissions are then modelled as a fraction of the nitrification rate. One example is the work done by Hutchinson (2012) already referenced above.

Local scaled models based on soil measurements are a cross between empirical and process based modelling approaches. They were originally proposed by Galbally and Johnson (1989) who assumed that by looking at NO production and consumption processes within the soil and the diffusional transport through the soil, they could calculate the NO flux for a specific soil type. This method requires extensive field and laboratory work because NO fluxes must be determined at a wide range of temperatures and WFPS. After sufficient data is collected the NO fluxes can be estimated as a function of soil temperature and WFPS. This can then be up scaled using land use distribution data. This model has been extensively used by Feig to determine NO flux rates for savannah grassland in the Kruger National Park and other dry ecosystems (See study references above).
3. EXPERIMENTAL AND METHOD

3.1 Dispersion modelling

Air dispersion models are used to compute ambient pollutants concentration and deposition rates at ground level. By taking into account various types of pollutant sources, emission rates and the meteorology of a specific time period they can predict the spatial and temporal patterns of the pollutant concentration at specified heights.

The modelling system used for this study was CALPUFF (Version 6.42), which has three main components and a number of pre- and post-processing units. CALPUFF is preferred by the US Environmental Protection Agency because it is ideal for modelling transport of pollutants across large areas Scorgie and Kornelius, 2009), and also because of the built-in atmospheric chemistry modules.

3.1.1 CALPUFF

CALPUFF is a regional Langrangian puff model and can accommodate various effects such as spatial and temporal variability of the meteorology over a region, dry deposition and dispersion over spatially varying land surfaces. The main reason CALPUFF was used for this study is because it builds on a work done by Scorgie and Kornelius in 2009, which not only reduced the resources required to quantify emission sources, but had also been shown to correlate reasonably well with measured \( \text{SO}_2 \), \( \text{SO}_4 \), \( \text{NO} \), \( \text{NO}_2 \) and \( \text{NO}_3 \) ambient concentrations as well as recorded and simulated wet, dry and total sulphur deposition rates for the Highveld (Scorgie and Kornelius, 2009).

To summarise, the CALPUFF modelling suite was selected because:

- Very large modelling domains can be modelled.
- It can characterise spatial and temporal variation in meteorology.
- CALMET can include complex terrain topography.
• CALPUFF can undertake first order chemical transformation calculations using two different calculation schemes and can predict the secondary pollutants formed due to primary pollutants emitted to the atmosphere.
• CALPUFF uses a resistance deposition model to predict spatially and temporally varying gas dry deposition rates.
• CALPUFF uses pollutant specific scavenging coefficients to predict wet deposition.
• Different type of emissions sources can be specified. These include point, volume, area and line sources.

The CALPUFF suite comprises three main program components: CALMET which is used to construct a three-dimensional wind field, CALPUFF which models the dispersion of the pollutants within a three-dimensional wind field and CALPOST to extract and process the results.

3.2 Highveld dispersion model and emission inventory

3.2.1 Domain

The modelling domain was chosen to be large enough to include important pollutant sources and relevant water catchments on the Highveld. Because this study relies on data produced for the previous study done by Scorgie and Kornelius (2009), this domain is very similar to the original domain set up for their model. The south west corner of the domain is at -27.398º longitude and -28.5644º latitude and covers an area of 350 km (east-west) by 350 km (north-south). A receptor grid consisting of 70 cells by 70 cells with a resolution of 5 km was set up to investigate the spatial and temporal pollutant dispersion and deposition rates at ground level. To enable model verification a separate set of discrete receptor points coinciding with various air quality monitoring stations was specified (see Table 3.1).
Table 3.1 Discrete Receptor Points (Scorgie and Kornelius, 2009)

<table>
<thead>
<tr>
<th>Longitude</th>
<th>Latitude</th>
<th>Receptor Type</th>
<th>Receptor name</th>
</tr>
</thead>
<tbody>
<tr>
<td>29.8896°</td>
<td>-27.3278°</td>
<td>AQ Station</td>
<td>Verkykkop(Eskom)</td>
</tr>
<tr>
<td>29.4214°</td>
<td>-26.2525°</td>
<td>AQ Station</td>
<td>Elandsfontein</td>
</tr>
<tr>
<td>28.9820°</td>
<td>-26.0945°</td>
<td>AQ Station</td>
<td>Kendal 2</td>
</tr>
<tr>
<td>28.9330°</td>
<td>-26.3670°</td>
<td>AQ Station</td>
<td>Leandra</td>
</tr>
<tr>
<td>29.8000°</td>
<td>-27.1128°</td>
<td>AQ Station</td>
<td>Majuba 1</td>
</tr>
<tr>
<td>29.5661°</td>
<td>-27.0154°</td>
<td>Miscellaneous</td>
<td>Misc</td>
</tr>
<tr>
<td>29.6786°</td>
<td>-27.0869°</td>
<td>AQ Station</td>
<td>Majuba 3</td>
</tr>
<tr>
<td>27.9030°</td>
<td>-26.8334°</td>
<td>AQ Station</td>
<td>Makalu</td>
</tr>
<tr>
<td>30.0774°</td>
<td>-25.5171°</td>
<td>AQ Station</td>
<td>Palmer</td>
</tr>
<tr>
<td>30.1090°</td>
<td>-26.6226°</td>
<td>AQ Station</td>
<td>Camden</td>
</tr>
<tr>
<td>29.8667°</td>
<td>-27.0167°</td>
<td>AQ Station</td>
<td>Amersfoort</td>
</tr>
<tr>
<td>29.9333°</td>
<td>-26.3167°</td>
<td>Catchment headwater</td>
<td>Komati catchment: subcatchment 1</td>
</tr>
<tr>
<td>29.4028°</td>
<td>-26.4597°</td>
<td>Catchment headwater</td>
<td>Olfants catchment: subcatchment 5</td>
</tr>
<tr>
<td>29.9731°</td>
<td>-27.3167°</td>
<td>Catchment headwater</td>
<td>Sandspruit catchment: subcatchment 8</td>
</tr>
</tbody>
</table>

3.2.2 Land use and topographical inputs

CALMET requires geophysical data to take the relevant geography effects into account when generating a three-dimensional wind field. Relevant inputs include land use types, elevation, various surface parameters (vegetation leaf area index, surface roughness length, soil heat flux) and anthropogenic heat flux. Land use and terrain data was obtained from www.WebGIS.com.

3.2.3 Meteorological input

CALMET contains a diagnostic wind field module that takes into account the terrain effects that may produce highly variable wind speeds such as slope flows, terrain channelling and terrain kinematics effects.

An initial guess wind field is set up by the user. This can either be done by collecting wind speed data at specified heights from upper air and surface weather stations, or by using MM5 data. MM5 is a prognostic wind field model that has been developed.
by Penn State University and the National Centre for Atmospheric Research (NCAR) to predict mesoscale and regional scale atmospheric circulation by using meteorological data from airport meteorological stations. In this case MM5 data was obtained from Lakes Environmental, due to the difficulty of converting the older meteorological data from Scorgie and Kornelius to be compatible to the current version of CALMET. A limitation of using MM5 data was that the largest tile size available was 350 km × 350 km, which is smaller than the original domain size of 380 km by 430 km used by Scorgie and Kornelius. Ten vertical layers were specified to characterise terrain effects within the model. These were a combination of both MM5 specified heights and simulation heights chosen by Scorgie and Kornelius for the original model. They are at ground level, 20m, 40m, 160m, 200m, 500m, 600m, 1000m, 1500m, 2200m and 3000m.

3.2.4 Emission Scenarios

Scorgie and Kornelius chose the 2001 hydrological year (1 October 2000 to 31 September 2001) as the base case scenario for primarily model verification purposes as the meteorology was typical for an “average rain” year. Another big limitation of using MM5 data was that no pre-2001 MM5 data could be obtained from Lakes Environmental for the chosen domain. This meant that a model run time from 1 January 2001 to 31 December was chosen instead. Other reasons for chosen runtime include:

- 2001 represented an average rainfall year.
- 2001 was originally chosen by Scorgie and Kornelius due to all emission and source data being available for that year.
- Published deposition monitoring data used for model verification was available for that time period (Scorgie and Kornelius, 2009).

To include the effects of rain, a below average rainfall year (2003) and an above average rainfall (2010) year were selected as additional scenarios. The years were selected after comparing annual total rainfall data recorded at towns located within the study area from 2001 to 2012. The rainfall data was supplied by the South African Weather Services.
3.2.5 Emission Sources

Various SO$_x$ and NO$_x$ emission sources were inventoried by Scorgie and Kornelius and included the following:

- All large coal fired power generation located on the Highveld.
- From industries holding licences under the Air Quality Act of 2004 or combustion and other chemical industrial emissions permits under the Atmospheric Pollution Prevention Act of 1965.
- Household coal, liquefied petroleum gas (LPG), paraffin and wood burning.
- Institutional and commercial (schools, hospitals and businesses) burning of coal, heavy fuel oil, wood and gas.
- Vehicle tailpipe emission from petrol and diesel vehicles.
- Biomass burning such as veld fires.

3.2.5.1 Large Coal-fired Power Stations

Power generation has increased significantly over the last hundred years on the Highveld and is the most significant source of NO$_x$ and SO$_x$ emissions for the region. In the 1950’s emissions were far below 100 000 tpa, but due to sharp increase in the country’s electricity demands over the next 50 years, emissions started to exceed 900 000 tpa by the end of the century. In Figure 3.1 SO$_x$ and NO$_x$ generated by Highveld power stations are summarized for the period 1927 to 2007. All sources and emission data in this category used by Scorgie and Kornelius and by this study was provided by Eskom Ltd.
Figure 3.1 SOx and NOx emissions for power stations located on the Highveld (Scorgie and Kornelius, 2009)

CALPUFF requires source input parameters such as stack height, stack diameter, gas exit velocity and gas exit temperature. In Table 3.2 emission rates for the base case scenario are summarised. In Table 7.1 Major industrial and Large Coal-fired Power Station point sources input parameters for 2001 in the Appendix A Industrial emission sources all 18 point sources with the required input parameters are shown.
### Table 3.2 Emissions for base case operations (Scorgie and Kornelius, 2009)

<table>
<thead>
<tr>
<th>Power Station</th>
<th>SO₂ (tpa) 2000/1</th>
<th>NO (tpa)(a) 2000/1</th>
<th>NO₂ (tpa)(a) 2000/1</th>
</tr>
</thead>
<tbody>
<tr>
<td>Arnot</td>
<td>65,360</td>
<td>21,648</td>
<td>677</td>
</tr>
<tr>
<td>Duvha</td>
<td>165,340</td>
<td>43,433</td>
<td>1,359</td>
</tr>
<tr>
<td>Hendrina</td>
<td>108,034</td>
<td>26,748</td>
<td>837</td>
</tr>
<tr>
<td>Kendal</td>
<td>241,250</td>
<td>56,985</td>
<td>1,783</td>
</tr>
<tr>
<td>Kriel</td>
<td>106,895</td>
<td>38,769</td>
<td>1,213</td>
</tr>
<tr>
<td>Lethabo</td>
<td>142,263</td>
<td>61,873</td>
<td>1,936</td>
</tr>
<tr>
<td>Majuba</td>
<td>48,253</td>
<td>12,145</td>
<td>380</td>
</tr>
<tr>
<td>Matla</td>
<td>177,909</td>
<td>52,937</td>
<td>1,657</td>
</tr>
<tr>
<td>Tutuka</td>
<td>71,104</td>
<td>19,106</td>
<td>598</td>
</tr>
<tr>
<td><strong>Total</strong></td>
<td><strong>1,126,409</strong></td>
<td><strong>333,646</strong></td>
<td><strong>10,441</strong></td>
</tr>
</tbody>
</table>

### 3.2.5.2 Major Industrial Sources

Major sources within the study area are the Natref Refinery, Sasol 2 & 3 (Secunda), Sasol 1 (Sasolburg), ArcelorMittal Vanderbijlpark, ArcelorMittal Vereeniging and Sappi Ngondwana. A major industrial source was defined as any industrial facility emitting greater than 10 ktpa of either NOₓ or SOₓ by Scorgie and Kornelius.

Several sources were used to quantify emissions from these industries. These were:

- Data supplied by the specific companies in question.
- An emissions inventory compiled for the Vaal Triangle Air Quality Management Plan (AQMP) development project (Liebenberg-Enslin *et al.*, 2008).

Data sets that were obtained by Scorgie and Kornelius from specific industrial facilities include:
• Natref Refinery - Vaal Triangle Air Quality Management Plan (AQMP) Emissions Inventory used for post 2000 period.
• Sasol 2 & 3 (Secunda) – current and historical emissions data received from Christi Brand of Sasol Secunda Operations.
• Sasol 1 (Sasolburg) - Vaal Triangle AQMP Emissions Inventory for post 2000.
• ArcelorMittal Vanderbijlpark - used Vaal Triangle AQMP data for post 2000.
• ArcelorMittal Vereeniging - used Vaal Triangle AQMP data.
• Sappi Ngondwana - based on DEAT 1994 emissions data and recent public data.

For the base case scenario summary of all the defined 128 point sources see Table 7.1 Major industrial and Large Coal-fired Power Station point sources input parameters for 2001 in Appendix A Industrial emission sources. Composed mostly (99%) of stack emissions they were assumed to remain constant due to the lack of seasonal and diurnal emission data. It was also assumed that all NO\textsubscript{x} emissions comprised of 10% NO\textsubscript{2} (EPA, 2005). Over 60 low level sources from ArcelorMittal Vanderbijlpark Works were included emitting roughly 30 tpa SO\textsubscript{2} and less than 0.5 tpa NO\textsubscript{x} (Table 7.2 Major Industrial Volume Source input parameters for 2001 in the Appendix A Industrial emission sources). Major industrial source emissions for the base case are summarised below in Table 3.3.

Table 3.3 Base case scenario major sources summary (Scorgie and Kornelius, 2009)

<table>
<thead>
<tr>
<th>Facility</th>
<th>SO\textsubscript{2} (tpa)</th>
<th>NO\textsubscript{x} (tpa)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Highveld Steel</td>
<td>35,207</td>
<td>1,575</td>
</tr>
<tr>
<td>ArcelorMittal Vanderbijlpark</td>
<td>13,649</td>
<td>14,867</td>
</tr>
<tr>
<td>ArcelorMittal Vereeniging</td>
<td>11</td>
<td>193</td>
</tr>
<tr>
<td>Vanchem eMalahleni</td>
<td>3,091</td>
<td>-</td>
</tr>
<tr>
<td>Sasol 1</td>
<td>18,521</td>
<td>18,274</td>
</tr>
<tr>
<td>Sasol 2&amp;3</td>
<td>202,535</td>
<td>144,875</td>
</tr>
<tr>
<td>Natref (NO\textsubscript{x} as NO\textsubscript{2})</td>
<td>11,605</td>
<td>598</td>
</tr>
</tbody>
</table>
3.2.5.3 Minor Industrial Sources

Industrial facilities emitting greater than 0.1 tpa of either NO\textsubscript{x} or SO\textsubscript{x} were classified as a minor industrial source. 159 point sources were identified by Scorgie and Kornelius for input into CALPUFF and the data was compiled from previous studies: the NEDLAC Dirty Fuels Study (Scorgie et al., 2004b) and the Eskom 2006 Study (Scorgie and Thomas, 2006). See Table 7.3 in the Appendix A Industrial emission sources for a complete list of point sources. Total SO\textsubscript{2} emissions for this category were estimated at 11,866 tpa and total NO\textsubscript{x} emissions at 4,100 tpa (Scorgie and Kornelius, 2009).

3.2.5.4 Household Burning

Household energy consumption was very difficult to compute. A range of factors had to be considered when trying to estimate emissions from household burning. Some of these are related to the type of house, the number of inhabitants, the season, available fuel types, the cost of the specific fuel types being used, income and whether or not the house is electrified (Scorgie and Kornelius, 2009).

To determine the number of households and which fuel types were being used Scorgie and Kornelius based their estimates on work previously done by Wicking-Baird (1997), Scorgie et al. (2003) and Irurah et al. (2000). Energy use statistics and house numbers were obtained from data published by the South African Institute of Race Relations. To calculate total quantities of each type of fuel (coal, paraffin, wood, LPG) consumed, typical monthly fuel use figures for various house types (Afrane-Okese, 1998) were used together with the numbers obtained above. Emission factors were then used to estimate the resultant emissions for each.
relevant pollutant. Seasonal and diurnal variations were also taken into account. During cold periods the hourly maximum emission rate was usually 10 times higher than the average yearly hourly emission rate (Scorgie and Kornelius, 2009). In Table 3.4 the total SO$_2$ and total NO$_x$ emission for each type of household fuel are shown for the original base case scenario.

A total of 120 area sources were identified for input into CALPUFF. For each area, an hourly aggregated emission for each species was specified. This was done for every hour in every day for the entire length of the scenario period. Due to the extent of the emission data no summary of the emission rates can be included in this document.

Table 3.4 Estimated total annual household fuel burning emissions for the Highveld (Scorgie and Kornelius, 2009)

<table>
<thead>
<tr>
<th>Fuel Combusted</th>
<th>Annual Emissions</th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>SO$_2$ (tpa)</td>
<td>NO$_x$ (tpa)</td>
</tr>
<tr>
<td>Coal</td>
<td>9,091.0</td>
<td>3,134.8</td>
</tr>
<tr>
<td>Wood</td>
<td>19.6</td>
<td>146.8</td>
</tr>
<tr>
<td>Paraffin</td>
<td>11.9</td>
<td>154.9</td>
</tr>
<tr>
<td>LPG</td>
<td>0.1</td>
<td>12.8</td>
</tr>
<tr>
<td><strong>Total</strong></td>
<td><strong>9,122.6</strong></td>
<td><strong>3,449.2</strong></td>
</tr>
</tbody>
</table>

3.2.5.5 Vehicle Exhaust Emissions

Vehicle exhaust emissions were calculated by Scorgie and Kornelius by looking at local fuel sales and vehicle fleet information instead of using traffic count data to calculate spatial and temporal emissions.

For petrol, vehicle fleet information was composed from a national vehicle population database and spatially resolved technology mix and energy capacity profiles published for Johannesburg, Pretoria and the Vaal Triangle (Scorgie and Kornelius, 2009). The South African Petroleum Industry Association (SAPIA) supplied annual unleaded petrol sale data that was used to calculate total vehicle kilometres travelled.
for each magisterial district. Emissions were then calculated by multiplying total vehicle kilometres travelled with emission factors developed locally by Wong (1999).

The national diesel vehicle fleet was subdivided into three types: light, medium and heavy commercial vehicles. Consumption rates for each type were obtained from Stone (2000) and Wong (1999). Annual fuel sales data was again obtained from SAPIA to estimate the total vehicle kilometres travelled for each magisterial district. In this case, locally developed emission factors were obtained from Stone (2000). Emission factors for both diesel and petrol vehicles are shown in Table 3.5.

### Table 3.5 Emission factors for petrol and diesel vehicles (Scorgie and Kornelius, 2009)

<table>
<thead>
<tr>
<th>Pollutant</th>
<th>Units</th>
<th>Petrol-driven Vehicles</th>
<th>Diesel-driven Vehicles</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>Catalytic</td>
<td>Non-catalytic</td>
</tr>
<tr>
<td>NO&lt;sub&gt;x&lt;/sub&gt;</td>
<td>g/km</td>
<td>0.93</td>
<td>2.15</td>
</tr>
<tr>
<td>SO&lt;sub&gt;2&lt;/sub&gt;</td>
<td>g/km</td>
<td>0.015</td>
<td>0.043</td>
</tr>
</tbody>
</table>

Total vehicle emissions for NO<sub>2</sub> and NO<sub>x</sub> were estimated by Scorgie and Kornelius to be 23221 tpa and 147577 tpa respectively. 292 line sources representing various roads across the Highveld and 16 area sources representing major metropolitan and municipalities were compiled for CALPUFF. Diurnal patterns for the following species were also included: SO<sub>2</sub>, NO, NO<sub>2</sub> and PM10 for each line source and area source.

### 3.3 Chemical transformation and deposition modelling

CALPUFF has several chemical transformation schemes which must be specified by the user to simulate the transformation processes. Scorgie and Kornelius used the
RIVAD/ARM3 scheme to model the gas phase reactions. The reasons this scheme were used are:

- The scheme allowed for separate modelling of NO and NO\textsubscript{2}, whereas the standard MESOPUFF II scheme only models nitrogen compounds as NO\textsubscript{x}.
- The conversion of NO and NO\textsubscript{2}, NO\textsubscript{2} and total NO\textsubscript{3}\textsuperscript{-} and SO\textsubscript{2} and SO\textsubscript{4}\textsuperscript{2-} are done in equilibrium with gaseous HNO\textsubscript{3} and ammonium nitrate aerosol.
- Daily OH\textsuperscript{-} concentration variations can be approximated with user specified daily ozone data.

The above mentioned hourly varying ozone data was composed from data received from Eskom’s Verkykkop, Elandsfontein, and Kendal 2 stations for the years of 2000/1.

Gas and particle dry deposition rates are predicted by a resistance deposition model within CALPUFF. To assist in dry deposition calculations site specific deposition velocities were input into CALPUFF by Scorgie and Kornelius. Hourly variations of seasonal averages were specified for each of the following species: SO\textsubscript{2}, NO, NO\textsubscript{2} and HNO\textsubscript{3}. Below in Figure 3.2 the velocities specified for SO\textsubscript{2} can be seen for each season. For wet deposition default values were assumed for the scavenging coefficients for SO\textsubscript{2}, SO\textsubscript{4}\textsuperscript{2-}, NO\textsubscript{x}, HNO\textsubscript{3} and NO\textsubscript{3}\textsuperscript{-}.

![Seasonal-average Deposition Velocities for SO2](image)

**Figure 3.2 Seasonal average dry deposition velocities for SO2 on the Highveld (Scorgie and Kornelius, 2009)**
Total sulphur deposition includes wet and dry deposition of gaseous SO$_2$ and particulate SO$_4^{2-}$, while total nitrogen deposition includes the dry deposition of NO, NO$_2$, HNO$_3$, NO$_3^-$ and ammonium sulphate and the wet deposition of HNO$_3$, NO$_3^-$ and ammonium sulphate.

Gaseous NH$_3$ and biogenic NO$_x$ emission were not included in the original model of Scorgie and Kornelius and their results therefore underpredict the total N deposited on the Highveld. To account for the contribution of ammonium sulphate and ammonium nitrate it was assumed by Scorgie and Kornelius that SO$_4^{2-}$ and NO$_3^-$ were completely neutralized by the NH$_4^+$. To calculate the amount of ammonium that is deposited due to the ammonium salts the following factors were applied to the SO$_4^{2-}$ and NO$_3^-$ fluxes predicted by CALPUFF. These factors are calculated by doing a basic mass balance for each salt ((NH$_4$)$_2$SO$_4$ and NH$_4$NO$_3$) forming reaction and is the fraction of the mass of ammonium per mass of salt:

\begin{align*}
0.292 \times \text{SO}_4^{2-} & \quad (17) \\
0.226 \times \text{NO}_3^- & \quad (18)
\end{align*}

### 3.4 Important differences between Scorgie & Kornelius model and the current model

The main differences between the current model and the model compiled by Scorgie and Kornelius are in the meteorological and in the emission source inputs.

For the original model, upper and surface air data had to be compiled from 19 surface and upper air monitoring stations located across the Highveld. Rainfall was composed from 92 stations (Scorgie and Kornelius, 2009).

Instead of composing meteorological data from scratch the current model made use of MM5 data from Lakes Environmental which included upper air, surface data and rainfall data. As a result, there will not only be differences in the wind speed and rainfall data, but also in the spatial distribution of this of data. The earliest MM5 data that
could be obtained was from the start of the year 2001 and the current models run time was chosen to start on 1 January 2001 instead of 1 October 2001 for the average rainfall year. Background data and emission data such as ozone and household burning data had to be edited to reflect this change.

All sources except biomass burning on the Highveld were included in the current model. In the original model biomass burning accounted only for 0.04% of SO\textsubscript{2} emissions and 0.5% of NO\textsubscript{x} emissions.

### 3.5 Model verification

To distinguish between the two models below, M1 (Model 1) refers to the older model by Scorgie and Kornelius and M2 (Model 2) refer to current model in this report.

To assess whether the model built for this study functions properly, the base case scenario of M2 was compared to measured ambient air concentrations recorded by Eskom and recorded deposition rates from literature. The range of uncertainty for dispersion models is given by the US-EPA (United States Environmental Protection Agency) as -50% to 200% (EPA, 1986). Model predictions falling outside this range will be labelled as unrepresentative and will be indicated as bold red.

A comparison between the previous model predictions by Scorgie and Kornelius and those of the current model is also made using the same uncertainty range to assess how the current and old models compare to each other. The following output values at the defined receptor locations (shown Table 3.1) were used for the comparison:

- Annual average ambient air concentrations of SO\textsubscript{2}, SO\textsubscript{4}^\textsuperscript{-}, NO, NO\textsubscript{2} and NO\textsubscript{3}^\textsuperscript{-}.
- Total annual S and N deposition rates at.
- Annual wet and dry deposition rates.
Spatial graphs were also plotted for the following output values for each of the scenarios above and compared to the older model runs done by Scorgie and Kornelius in 2009:

- Annual average ambient air concentrations of $\text{SO}_2$, $\text{SO}_4^-$, $\text{NO}$, $\text{NO}_2$ and $\text{NO}_3$.
- Total annual $\text{S}$ and $\text{N}$ deposition rates.
- Annual wet and dry deposition rates.

### 3.5.1 Model limitations

The following model limitations need to be considered when assessing the model outputs.

#### 3.5.1.1 Over prediction of rainfall by MM5 data

M2 generally has lower ambient air gaseous concentrations than M1, while deposition values (wet and total deposition) for M2 are higher than for M1. To assess why concentrations and depositions varied from those predicted by M1 the rainfall figures for both models were compared. Rainfall has a direct impact on wet deposition rates and will affect the overall mass transfer rates for the entire model. Rainfall for the M1 model was based of local weather station data and will reflect the rainfall patterns of the Highveld more accurately. As can be seen below in Table: 3.6 the MM5 data used to generate the meteorological inputs for M2 over estimated rainfall by almost a factor of two in some cases.

#### 3.5.1.2 Ammonia emissions

Ammonia emission were not quantified as part as the emission inventory nor were they simulated. Because of this the model (M2) will under predict dry $\text{N}$ deposition. Dry deposition of ammonia has been estimated by Galy-Lacaux et al (2003) to account for ~ 30% of total $\text{N}$ deposition at Amersfoort (see Table 3.6 below). Because dry $\text{NH}_3$ is responsible for most dry $\text{N}$ deposition, it is expected that predicted wet deposition will have much more significant impact on total $\text{N}$ deposition rising from ~ 60 % to ~ 90%.
Table 3.6 Nitrogen deposition composition for Amersfoort (Galy-Lacaux et al., 2003)

<table>
<thead>
<tr>
<th>Predicted Deposition (kg N/ha/annum)</th>
<th>Previously Measured Deposition (kg N/ha/annum)(a)</th>
<th>% of total N Deposition</th>
</tr>
</thead>
<tbody>
<tr>
<td>NO, NO₂, HNO₃ (gas)</td>
<td>dry</td>
<td>1.3</td>
</tr>
<tr>
<td>NO₃ (particle)</td>
<td>dry</td>
<td>0.12</td>
</tr>
<tr>
<td>NH₃ (gas)</td>
<td>dry</td>
<td>5.55</td>
</tr>
<tr>
<td>NO₃ (particle)</td>
<td>wet</td>
<td>5.5</td>
</tr>
<tr>
<td>HNO₃ (gas); (NH₄)₂SO₄ (particle)</td>
<td>wet</td>
<td>4</td>
</tr>
<tr>
<td>TOTAL (all constituents)</td>
<td></td>
<td>16.47</td>
</tr>
</tbody>
</table>
Table 3.7 Total annual rainfall for each receptor comparison

<table>
<thead>
<tr>
<th>Total Rainfall mmhr(^{-1})</th>
<th>Average Rainfall Period</th>
<th>Below Average Period</th>
<th>Above Average Period</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>2001 M2</td>
<td>2000/1 M1</td>
<td>M2/M1</td>
</tr>
<tr>
<td>Verkykkop(Eskom)</td>
<td>1139.72</td>
<td>600</td>
<td>1.90</td>
</tr>
<tr>
<td>Elandsfontein</td>
<td>904.54</td>
<td>700</td>
<td>1.29</td>
</tr>
<tr>
<td>Kendal 2</td>
<td>840.93</td>
<td>400</td>
<td>2.10</td>
</tr>
<tr>
<td>Leandra</td>
<td>903.35</td>
<td>500</td>
<td>1.81</td>
</tr>
<tr>
<td>Majuba 1</td>
<td>1188.31</td>
<td>700</td>
<td>1.70</td>
</tr>
<tr>
<td>Majuba 3</td>
<td>1054.59</td>
<td>1000</td>
<td>1.05</td>
</tr>
<tr>
<td>Makalu</td>
<td>846.38</td>
<td>700</td>
<td>1.21</td>
</tr>
<tr>
<td>Palmer</td>
<td>622.95</td>
<td>700</td>
<td>0.89</td>
</tr>
<tr>
<td>Camden</td>
<td>1281.33</td>
<td>700</td>
<td>1.83</td>
</tr>
<tr>
<td>Amerfoort</td>
<td>1115.8</td>
<td>700</td>
<td>1.59</td>
</tr>
<tr>
<td>Komati headwater</td>
<td>1143.15</td>
<td>700</td>
<td>1.63</td>
</tr>
<tr>
<td>Olifants headwater</td>
<td>1063.67</td>
<td>700</td>
<td>1.52</td>
</tr>
<tr>
<td>Sandspruit headwater</td>
<td>1162.62</td>
<td>600</td>
<td>1.94</td>
</tr>
</tbody>
</table>
3.5.2 M2 compared to ambient concentrations and deposition measurements

M2 ambient values are compared to measured values in Table 3.8 below. M1 values are included for comparative value. Ambient NO was predicted to be less at Kendal and Palmer, while NO$_2$ at Verkykkop, Kendal and Palmer were predicted to be lower than what was measured. SO$_2$ predictions compared very well at all sites, but were lower than measurements.

Deposition values predicted by M2 are compared to measure deposition values from literature in Tables 3.9, 3.10 and 3.11 below. Wet N deposition predictions were lower than measurements, while predicted total N deposition was much lower.

Wet S deposition was predicted to be much higher than the measured values at Amersfoort and Ermelo. This is most likely due to the much higher rainfall predicted by the MM5 data at those locations. In contrast, at sites like Vryheid, Warden and Suikerbosrand wet S values were predicted to be within the acceptable range. Dry S deposition was predicted to be very close to the measured values. Due to the high predicted wet S deposition values at Amersfoort and Ermelo the total S deposition values are much higher than the measured total S values.
### Table 3.8: Measured to modelled (Current [M2] and original) pollutant concentrations

<table>
<thead>
<tr>
<th>Station</th>
<th>Period average</th>
<th></th>
<th></th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>NO</td>
<td>NO₂</td>
<td>SO₂</td>
<td>NO₃</td>
<td>SO₄</td>
</tr>
<tr>
<td><strong>Modelled (M2) µg/m³</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Verkykkop</td>
<td>1.3</td>
<td>0.6</td>
<td>6.7</td>
<td>1.3</td>
<td>1.4</td>
</tr>
<tr>
<td>Elandsfontein</td>
<td>1.5</td>
<td>4.6</td>
<td>21.7</td>
<td>1.6</td>
<td>1.9</td>
</tr>
<tr>
<td>Kendal2</td>
<td>0.4</td>
<td>2.7</td>
<td>22.6</td>
<td>1.7</td>
<td>1.8</td>
</tr>
<tr>
<td>Leandra</td>
<td>0.9</td>
<td>2.8</td>
<td>21.3</td>
<td>1.9</td>
<td>0.8</td>
</tr>
<tr>
<td>Majuba1</td>
<td>1.6</td>
<td>1.1</td>
<td>9.2</td>
<td>1.4</td>
<td>1.83</td>
</tr>
<tr>
<td>Palmer</td>
<td>0</td>
<td>0.8</td>
<td>4.8</td>
<td>1.2</td>
<td>0.8</td>
</tr>
<tr>
<td>Amersfoort</td>
<td>1.6</td>
<td>1.1</td>
<td>9.2</td>
<td>1.4</td>
<td>1.83</td>
</tr>
<tr>
<td><strong>Measured µg/m³</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Verkykkop</td>
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<td>5.4</td>
<td>8.8</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Elandsfontein</td>
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<td>9.2</td>
<td>28</td>
<td>2.4</td>
<td>4.3</td>
</tr>
<tr>
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<td>5.9</td>
<td>13.9</td>
<td>35.4</td>
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<td></td>
</tr>
<tr>
<td>Leandra</td>
<td></td>
<td></td>
<td>30.1</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Majuba1</td>
<td></td>
<td></td>
<td>12.7</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Palmer</td>
<td>2.4</td>
<td>5.6</td>
<td>7.8</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Amersfoort</td>
<td>3</td>
<td>4.9</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td><strong>Ratio (M2 vs Measured)</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Verkykkop</td>
<td>0.93</td>
<td>0.11</td>
<td>0.76</td>
<td></td>
<td></td>
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<tr>
<td>Elandsfontein</td>
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<td>0.50</td>
<td>0.78</td>
<td>0.67</td>
<td>0.44</td>
</tr>
<tr>
<td>Kendal2</td>
<td>0.07</td>
<td>0.19</td>
<td>0.64</td>
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</tr>
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<td>Leandra</td>
<td></td>
<td></td>
<td>0.71</td>
<td></td>
<td></td>
</tr>
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<td></td>
<td></td>
</tr>
<tr>
<td>Palmer</td>
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<td>0.14</td>
<td>0.62</td>
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<td></td>
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<tr>
<td>Amersfoort</td>
<td>0.37</td>
<td>1.88</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td><strong>Ratio (M1 vs Measured) (modelled/measured)</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Verkykkop</td>
<td>0.2</td>
<td>0.5</td>
<td>0.9</td>
<td></td>
<td></td>
</tr>
<tr>
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<td>0.9</td>
<td>1</td>
<td>0.7</td>
<td>0.6</td>
</tr>
<tr>
<td>Kendal2</td>
<td>0.4</td>
<td>0.5</td>
<td>0.8</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Leandra</td>
<td></td>
<td></td>
<td>0.7</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Majuba1</td>
<td></td>
<td></td>
<td>0.8</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Palmer</td>
<td>0.1</td>
<td>0.5</td>
<td>1.1</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Amersfoort</td>
<td>1.1</td>
<td>2</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Station</td>
<td>Measured Wet N Deposition</td>
<td>Predicted Wet N Deposition for M2</td>
<td>Ratio of Predicted to Measured</td>
<td>Details of Measurement</td>
<td></td>
</tr>
<tr>
<td>---------</td>
<td>--------------------------</td>
<td>----------------------------------</td>
<td>-------------------------------</td>
<td>-----------------------</td>
<td></td>
</tr>
<tr>
<td></td>
<td>kg N ha(^{-1})yr(^{-1})</td>
<td>kg N ha(^{-1})yr(^{-1})</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Ermelo</td>
<td>8.3</td>
<td>5.8</td>
<td>0.70</td>
<td>1985-1992 Turner 1993</td>
<td></td>
</tr>
<tr>
<td>Amersfoort</td>
<td>8.4</td>
<td>3.7</td>
<td>0.5</td>
<td>1985-1992 Turner 1993</td>
<td></td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Station</th>
<th>Measured Total N Deposition</th>
<th>Predicted Total N Deposition for M2</th>
<th>Ratio of Predicted to Measured</th>
<th>Details of Measurement</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>kg N ha(^{-1})yr(^{-1})</td>
<td>kg N ha(^{-1})yr(^{-1})</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Amersfoort</td>
<td>10.92</td>
<td>4.4</td>
<td>0.40</td>
<td>Galy-Lacaux et al. 2003</td>
</tr>
</tbody>
</table>
Table 3.10 Comparison between measured and modelled (Current [M2] deposition velocities (Part 2))

<table>
<thead>
<tr>
<th>Station</th>
<th>Measured Wet S Deposition kg N ha⁻¹ yr⁻¹</th>
<th>Predicted Wet S Deposition for M2 kg N ha⁻¹ yr⁻¹</th>
<th>Ratio of Predicted to Measured</th>
<th>Details of Measurement</th>
<th>Measurement Averaging Period</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ermelo</td>
<td>5.9</td>
<td>15.3</td>
<td>2.59</td>
<td></td>
<td>1985-1992</td>
<td>Turner 1993</td>
</tr>
<tr>
<td>Amersfoort</td>
<td>5.7</td>
<td>15.5</td>
<td>2.72</td>
<td></td>
<td>1985-1992</td>
<td>Turner 1993</td>
</tr>
<tr>
<td>Amersfoort</td>
<td>5.3</td>
<td>15.5</td>
<td>2.92</td>
<td></td>
<td>1986-1999</td>
<td>Mphepya et al. 2005</td>
</tr>
<tr>
<td>Amersfoort</td>
<td>5</td>
<td>15.5</td>
<td>3.10</td>
<td></td>
<td>1996-1998</td>
<td>Mphepya and Held, 1999</td>
</tr>
<tr>
<td>Warden</td>
<td>4.4</td>
<td>5.8</td>
<td>1.3</td>
<td></td>
<td>1985-1992</td>
<td>Turner 1993</td>
</tr>
<tr>
<td>Suikerbosrand</td>
<td>8.5</td>
<td>5.76</td>
<td>0.68</td>
<td></td>
<td>Nov 1992 - March 1993</td>
<td>Skoroszewski, 1993</td>
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</tbody>
</table>
Table 3.11 Comparison between measured and modelled (Current [M2] deposition velocities (Part 3))

<table>
<thead>
<tr>
<th>Station</th>
<th>Measured Dry S Deposition</th>
<th>Predicted Dry S Deposition for M2</th>
<th>Ratio of Predicted to Measured</th>
<th>Details of Measurement</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>kg N ha(^{-1}) yr(^{-1})</td>
<td>kg N ha(^{-1}) yr(^{-1})</td>
<td></td>
<td>Measurement Averaging Period</td>
</tr>
<tr>
<td>Elandsfontein</td>
<td>9.4</td>
<td>8</td>
<td>0.85</td>
<td>Mphepya and Held, 1999</td>
</tr>
<tr>
<td>Palmer</td>
<td>3.1</td>
<td>2.8</td>
<td>0.90</td>
<td>Mphepya and Held, 1999</td>
</tr>
<tr>
<td>Elandsfontein</td>
<td>13.1</td>
<td>8</td>
<td>0.61</td>
<td>Zunckel, 1999; Zunckel et al. 1999 - Inferential modelling</td>
</tr>
<tr>
<td>Palmer</td>
<td>3.1</td>
<td>2.8</td>
<td>0.90</td>
<td>Zunckel, 1999; Zunckel et al. 1999 - Inferential modelling</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Station</th>
<th>Measured Total S Deposition</th>
<th>Predicted Total S Deposition for M2</th>
<th>Ratio of Predicted to Measured</th>
<th>Details of Measurement</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>kg N ha(^{-1}) yr(^{-1})</td>
<td>kg N ha(^{-1}) yr(^{-1})</td>
<td></td>
<td>Measurement Averaging Period</td>
</tr>
<tr>
<td>Elandsfontein</td>
<td>13.1</td>
<td>27.7</td>
<td>2.11</td>
<td>Zunckel, 1999</td>
</tr>
</tbody>
</table>
3.5.3 Average Rainfall year (2001) M1 vs M2

General observations for average rainfall year: M2 (2001) are as follows:

- Ambient air concentrations at Elandsfontein correlated well with measured and M1 (2000/1) predicted values.
- SO$_2$ and NO$_2$ gaseous concentrations correlated well with measured and M1 predicted values at most receptor locations, while NO, NO$_2$ and SO$_4^-$ were much lower than M1 predicted values.
- Deposition rates at Verkykerkop, Elandsfontein, Palmer and Amersfoort compared well with M1 predicted rates.
- Total Dry sulphur depositions compare well with M1 predicted rates.

Spatially SO$_2$ predicted ambient concentrations M1 and M2 looks very similar with concentration isopleths located at similar locations. The SO$_4^-$ concentration plots differ considerably. The 1 µg m$^{-3}$ isopleth for M2 covers a smaller area. The M2 NO$_2$ 1 µg m$^{-3}$ isopleth covers a slightly smaller area and the 5 µg m$^{-3}$ isopleths are very closely grouped around the main industrial areas of the central Highveld. The NO$_3^-$ outer isopleths for M1 and M2 are similarly spread out, but M2 has much higher ambient levels of NO$_3^-$ closer to the industrial areas. M2 has a 3 µg m$^{-3}$ isopleth stretching from Emalahleni to the Sasolburg/Vereeniging area, while M1 only goes up to 1.8 to 2 µg m$^{-3}$ for the same region.

The total sulphur deposition plots for both M1 and M2 are similar in shape for the central Highveld, but M2’s 5 kg ha$^{-1}$yr$^{-1}$ isopleth is much larger. The M2 total nitrogen deposition 6 and 8 kg ha$^{-1}$yr$^{-1}$ isopleths cover a much larger area compared to those plotted from M1. These areas stretch from Emalahleni to Sasolburg/Vereeniging area. The wet and dry nitrogen plots are similar to their M1 counterparts, with the higher deposition rates centred close to the industrial areas located on the central Highveld.
In Table 3.12 and Table 3.13 comparison between modelled M1 and M2 ambient concentration and deposition values at specific receptor locations is shown. Spatial plot can be seen in Appendix C: Average Rainfall year (2001).
Table 3.12 Comparison between modelled current (M2 {2001}) and original (M1 {2000/1}) pollutant concentrations for the average rainfall year scenario

<table>
<thead>
<tr>
<th>Station</th>
<th>NO</th>
<th>NO₂</th>
<th>SO₂</th>
<th>NO₃</th>
<th>SO₄</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Modelled (M1)</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>µg m⁻³</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Verkykop</td>
<td>0.3</td>
<td>2.5</td>
<td>7.5</td>
<td>1.3</td>
<td>1.7</td>
</tr>
<tr>
<td>Elandsfontein</td>
<td>2.3</td>
<td>8.5</td>
<td>29.2</td>
<td>1.6</td>
<td>2.5</td>
</tr>
<tr>
<td>Kendal2</td>
<td>2.4</td>
<td>7.4</td>
<td>29.3</td>
<td>1.7</td>
<td>2.6</td>
</tr>
<tr>
<td>Leandra</td>
<td>1.5</td>
<td>6.9</td>
<td>21.9</td>
<td>1.9</td>
<td>2.6</td>
</tr>
<tr>
<td>Majuba1</td>
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<td>3.3</td>
<td>9.9</td>
<td>1.5</td>
<td>1.9</td>
</tr>
<tr>
<td>Palmer</td>
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Table 3.13 Comparison between modelled current (M2 {2001}) and original (M1 {2000/1}) deposition rates for the average rainfall year scenario

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3.5.4 Above Average Rainfall Year (2010) M1 vs M2

Above average rainfall year:

- Ambient air concentrations did not compare well with the M1 (1995/6) modelled concentrations. M1 predicted much higher ambient concentrations at receptor locations.
- Sulphur deposition rates compared reasonably well with M1 predicted rates, but where higher than M1 predicted.
- Dry N deposition rates were predicted to be much higher than what was predicted by M1.
- Lower wet N deposition was also predicted by M2.

Spatially SO$_2$ concentrations plot for both M1 and M2 are very similar. The SO$_4^{2-}$ plot for M2 look very different from the M1 plot. The M1 isopleths range from 1 to 3.5 kg ha$^{-1}$yr$^{-1}$, while for M2 the isopleth range from 0.5 to 1.5 kg ha$^{-1}$yr$^{-1}$. Both NO$_2$ plots shows higher concentrations around the industrial areas, but the M2 plot only goes up to 5 µg m$^{-3}$ while M1 goes up to 10 µg m$^{-3}$. M2’s NO$_3^-$ ambient concentrations mapped similar to M1, with high concentration of NO$_3^-$ (> 4.5 µg m$^{-3}$) over the central Highveld. Nitrogen and sulphur deposition rates for M2 mapped similar to the M1 model predictions.

In Table 3.14 and Table 3.15 comparison between modelled M1 and M2 ambient concentration and deposition values at specific receptor locations is shown. Spatial plot can be seen in Appendix C: Above Average Rainfall Year (2010).
Table 3.14 Comparison between modelled current (M2 {2010}) and original (M1 {1995/6}) pollutant concentration for the above average rainfall year scenario.

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<th>SO₂</th>
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<th>SO₄</th>
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Table 3.15 Comparison between modelled current (M2 {2010}) and original (M1 {1995/6}) deposition rates for the above average rainfall year scenario
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<td></td>
<td>0.92</td>
<td>0.73</td>
<td>0.85</td>
<td>2.31</td>
<td>0.36</td>
<td>0.46</td>
</tr>
<tr>
<td>Kendal2</td>
<td></td>
<td>1.58</td>
<td>0.57</td>
<td>1.51</td>
<td>20.74</td>
<td>2.37</td>
<td>3.27</td>
</tr>
<tr>
<td>Leandra</td>
<td></td>
<td>0.59</td>
<td>0.76</td>
<td>0.63</td>
<td>4.54</td>
<td>0.96</td>
<td>0.99</td>
</tr>
<tr>
<td>Majuba1</td>
<td></td>
<td>1.07</td>
<td>0.80</td>
<td>1.03</td>
<td>2</td>
<td>0.35</td>
<td>0.39</td>
</tr>
<tr>
<td>Palmer</td>
<td></td>
<td>2</td>
<td>2</td>
<td>2</td>
<td>3.56</td>
<td>0.62</td>
<td>1.56</td>
</tr>
<tr>
<td>Amersfoort</td>
<td></td>
<td>1.32</td>
<td>0.84</td>
<td>1.16</td>
<td>3.24</td>
<td>0.39</td>
<td>0.59</td>
</tr>
</tbody>
</table>
3.5.5 Below Average Rainfall Year (2003) M1 vs M2

Below average rainfall year: M2 (2003):

- M2 predicted much higher values of NO while M2 predicted much lower NO\textsubscript{2} values. SO\textsubscript{2}, NO\textsubscript{3}\textsuperscript{-} and SO\textsubscript{4}\textsuperscript{2-} compared well to the M1 predicted values, falling with the 0.5 – 2 ratios.
- Deposition rates for most receptor locations were very similar except at Kendal M2 predicted much higher deposition values.
- Wet S deposition was predicted to be higher than M1 predicted.

It should be noted that the M1 (2006/7) model wind field data contained inaccuracies which affected the spatial patterns of projected ambient concentrations and deposition rates (Scorgie and Kornelius, 2009). This made it difficult to compare to the latest M2 plots. Ambient concentrations and deposition rates spatial variations for this year share similar traits compared to the M2 plots described above.

In Table 3.16 and Table 3.17 comparison between modelled M1 and M2 ambient concentration and deposition values at specific receptor locations is shown. Spatial plot can be seen in Appendix C: Below Average Rainfall Year (2003).
Table 3.16 Comparison between modelled current (M2 {2003}) and original (M1 {2006/7}) pollutant concentrations for the above average rainfall year scenario

<table>
<thead>
<tr>
<th>Station</th>
<th>Period average</th>
<th></th>
<th></th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Modelled (M1)</td>
<td>NO</td>
<td>NO₂</td>
<td>SO₂</td>
<td>NO₃</td>
</tr>
<tr>
<td>Verkykkop</td>
<td>0.1</td>
<td>1.9</td>
<td>6.7</td>
<td>1.5</td>
<td>1.8</td>
</tr>
<tr>
<td>Elandsfontein</td>
<td>0.9</td>
<td>6.9</td>
<td>23.6</td>
<td>2.4</td>
<td>2.9</td>
</tr>
<tr>
<td>Kendal2</td>
<td>1.2</td>
<td>6.1</td>
<td>25.5</td>
<td>2.4</td>
<td>2.8</td>
</tr>
<tr>
<td>Leandra</td>
<td>1.9</td>
<td>9.9</td>
<td>36.5</td>
<td>2.8</td>
<td>2.9</td>
</tr>
<tr>
<td>Majuba1</td>
<td>0.2</td>
<td>2.7</td>
<td>9.4</td>
<td>1.9</td>
<td>2.1</td>
</tr>
<tr>
<td>Palmer</td>
<td>0.1</td>
<td>2.0</td>
<td>7.5</td>
<td>1.3</td>
<td>1.6</td>
</tr>
<tr>
<td>Amersfoort</td>
<td>0.2</td>
<td>2.4</td>
<td>8.7</td>
<td>1.8</td>
<td>2.0</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Station</th>
<th>Period average</th>
<th></th>
<th></th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Modelled (M2)</td>
<td>NO</td>
<td>NO₂</td>
<td>SO₂</td>
<td>NO₃</td>
</tr>
<tr>
<td>Verkykkop</td>
<td>1.6</td>
<td>0.5</td>
<td>8.2</td>
<td>1.1</td>
<td>0.8</td>
</tr>
<tr>
<td>Elandsfontein</td>
<td>2.3</td>
<td>4.5</td>
<td>23.9</td>
<td>2.5</td>
<td>1.7</td>
</tr>
<tr>
<td>Kendal2</td>
<td>0.1</td>
<td>2.7</td>
<td>23.9</td>
<td>5.2</td>
<td>2.0</td>
</tr>
<tr>
<td>Leandra</td>
<td>0.6</td>
<td>2.9</td>
<td>22.8</td>
<td>5.7</td>
<td>2.0</td>
</tr>
<tr>
<td>Majuba1</td>
<td>2.1</td>
<td>0.9</td>
<td>11.3</td>
<td>1.4</td>
<td>1.0</td>
</tr>
<tr>
<td>Palmer</td>
<td>1.1</td>
<td>6.9</td>
<td>15.7</td>
<td>3.6</td>
<td>1.4</td>
</tr>
<tr>
<td>Amersfoort</td>
<td>1.0</td>
<td>2.3</td>
<td>10.8</td>
<td>1.4</td>
<td>1.2</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Station</th>
<th>Period average</th>
<th></th>
<th></th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Ratio (M1/M2)</td>
<td>NO</td>
<td>NO₂</td>
<td>SO₂</td>
<td>NO₃</td>
</tr>
<tr>
<td>Verkykkop</td>
<td>0.06</td>
<td>3.80</td>
<td>0.82</td>
<td>1.36</td>
<td>2.25</td>
</tr>
<tr>
<td>Elandsfontein</td>
<td>0.39</td>
<td>1.53</td>
<td>0.99</td>
<td>0.96</td>
<td>1.71</td>
</tr>
<tr>
<td>Kendal2</td>
<td>12.00</td>
<td>2.26</td>
<td>1.07</td>
<td>0.46</td>
<td>1.40</td>
</tr>
<tr>
<td>Leandra</td>
<td>3.17</td>
<td>3.41</td>
<td>1.60</td>
<td>0.49</td>
<td>1.45</td>
</tr>
<tr>
<td>Majuba1</td>
<td>0.10</td>
<td>3.00</td>
<td>0.83</td>
<td>1.36</td>
<td>2.10</td>
</tr>
<tr>
<td>Palmer</td>
<td>0.09</td>
<td>0.29</td>
<td>0.48</td>
<td>0.36</td>
<td>1.14</td>
</tr>
<tr>
<td>Amersfoort</td>
<td>0.20</td>
<td>1.04</td>
<td>0.81</td>
<td>1.29</td>
<td>1.67</td>
</tr>
</tbody>
</table>
Table 3.17 Comparison between modelled current (M2 {2003}) and original (M1 {2006/7}) deposition rates for the above average rainfall year scenario

<table>
<thead>
<tr>
<th>Station</th>
<th>Modelled (M1) (kg ha(^{-1}) yr(^{-1}))</th>
<th>Modelled (M2) (kg ha(^{-1}) yr(^{-1}))</th>
<th>Ratio (M2/M1)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Wet S</td>
<td>Dry S</td>
<td>Total S</td>
</tr>
<tr>
<td>Verkykkop</td>
<td>3.3</td>
<td>2.3</td>
<td>5.6</td>
</tr>
<tr>
<td>Elandsfontein</td>
<td>12.3</td>
<td>8.2</td>
<td>20.4</td>
</tr>
<tr>
<td>Kendal2</td>
<td>50.8</td>
<td>8.8</td>
<td>59.6</td>
</tr>
<tr>
<td>Leandra</td>
<td>7.4</td>
<td>11</td>
<td>18.4</td>
</tr>
<tr>
<td>Majuba1</td>
<td>8.7</td>
<td>3.2</td>
<td>11.9</td>
</tr>
<tr>
<td>Palmer</td>
<td>3.9</td>
<td>2.2</td>
<td>6.2</td>
</tr>
<tr>
<td>Amersfoort</td>
<td>5.3</td>
<td>3.1</td>
<td>8.3</td>
</tr>
</tbody>
</table>

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3.5.6 Conclusion

Spatial plots predicted by M1 and by M2 show very similar patterns. The general pattern important for this study is that higher concentrations of pollutants are found close to two major industrial areas within the study area. These areas are located within the central Highveld which contains the large coal fired stations (including Kendal, Duvha, Kriel and Matla) and the Vaal Triangle with major plants such as Sasol 1, ArcelorMittal Vanderbijlpark, Lethabo PS and Natref.

Similar predicted deposition values for M1 and M2 at receptor locations are found. Generally lower ambient gas concentrations and higher deposition rates were predicted by M2 due to the higher rainfall predicted by the MM5 data.

Given the model limitations and after comparing deposition results from previous studies M2 will be suitable to study the effect that biogenic NO\textsubscript{x} and H\textsubscript{2}S will have on deposition on the South African Highveld.

3.6 Hydrogen sulphide on the South African Highveld

Various sources located within the South African Highveld contribute to atmospheric H\textsubscript{2}S. These include:

- Fugitive emissions from petrochemical plants (mainly from Sasol located in Secunda).
- Open cast mines.
- Smelting plants.
- Coal fired plants.

An in-situ study done by Igbafe (2007) at Elandsfontein shows that on the Highveld in summer (1.5 - 3 µg m\textsuperscript{-3}), autumn (3 – 4 µg m\textsuperscript{-3}) and winter (3 – 5 µg m\textsuperscript{-3}) there was no or very little variation in the diurnal pattern of atmospheric H\textsubscript{2}S, while during spring (8 – 11 µg m\textsuperscript{-3}) a definite diurnal pattern could be observed with accumulation of H\textsubscript{2}S during night time and reduction during the day time. These patterns can be
most likely explained through rapid thermal oxidation of \( \text{H}_2\text{S} \) during day time and is shown below in Figure 3.3.

When released into the atmosphere \( \text{H}_2\text{S} \) undergoes rapid thermal photo-oxidation (reacting with hydroxyl radical in the presence of sunlight) conversion to \( \text{SO}_2 \) thereby contributing to the sulphur deposition on the Highveld. (Eggleton & Cox, 1978; Wang & Howard, 1990).

Another observation made by Igbafe (2007) was that there was a linear relationship between the mean seasonal concentrations of atmospheric \( \text{H}_2\text{S} \) and sulphate, which meant that atmospheric \( \text{H}_2\text{S} \) contributes to atmospheric \( \text{SO}_2 \).

![Graph showing diurnal variations of mean \( \text{H}_2\text{S} \) concentrations at Elandsfontein for various seasons (Igbafe, 2007)](image)

**Figure 3.3** Diurnal variations of mean \( \text{H}_2\text{S} \) concentrations at Elandsfontein for various seasons (Igbafe, 2007)
3.6.1 Emission scenario

Two major sources both from the Sasol petrochemical plant have been identified (Burger, von Gruenewaldt & Bird, 2014):

- Stack 1 1401.22 g s\(^{-1}\)
- Stack 2 1246.34 g s\(^{-1}\)

The CALPUFF reaction scheme used (RIVAD) does not allow H\(_2\)S gas as an input and thus cannot simulate the thermal oxidation of H\(_2\)S to SO\(_2\). To accommodate the extra SO\(_2\) load that should be attributed by the released H\(_2\)S, the two emission sources were input into CALPUFF as if they were SO\(_2\) sources. This would allow the extra sulphur added through these two stacks to take part in the atmospheric chemical reaction processes that CALPUFF simulates.

To emulate the different seasonal atmospheric patterns mentioned by Igbafe (2007) (seen in Figure 3.3) only a certain percentage of the total emission amount was taken to be converted to SO\(_2\) for each seasonal period as defined above. Because H\(_2\)S is still observed within the atmosphere and the conversion of H\(_2\)S can never be assumed to be 100%. Using Figure 3.3 as reference for the amount H\(_2\)S converted each season the following conversion rates are proposed:

- Summer 70% conversion.
- Autumn 60 % conversion.
- Winter 50 % conversion.
- Spring 40 % conversion.

Using these proposed conversion rates and the reported H\(_2\)S emission rates (see above) final CALPUFF SO\(_2\) emission input values are calculated for each stack and each season and are shown below in Table 3.18.
Table 3.18 H$_2$S conversion rates and input rates as SO$_2$ in CALPUFF

<table>
<thead>
<tr>
<th>Stack</th>
<th>H$_2$S (g s$^{-1}$)</th>
<th>H$_2$S (mol s$^{-1}$)</th>
<th>% Converted</th>
<th>SO$_2$ (mol s$^{-1}$)</th>
<th>SO$_2$ (g s$^{-1}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>1401.22</td>
<td>41.21</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>Summer</td>
<td>70</td>
<td>28.85</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>Autumn</td>
<td>60</td>
<td>24.73</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>Winter</td>
<td>50</td>
<td>20.61</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>Spring</td>
<td>40</td>
<td>16.48</td>
</tr>
<tr>
<td>2</td>
<td>1246.34</td>
<td>36.66</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>Summer</td>
<td>70</td>
<td>25.66</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>Autumn</td>
<td>60</td>
<td>21.99</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>Winter</td>
<td>50</td>
<td>18.33</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>Spring</td>
<td>40</td>
<td>14.66</td>
</tr>
</tbody>
</table>

These emission sources will be run separately from the biogenic emission models. This scenario will be referenced as Model 3 (M3).
3.7 Waterberg Dispersion Model and Emission Inventory

The Waterberg area is located within the Limpopo province within South Africa and is bordered to the west by Botswana. It is a much smaller area than the South African Highveld but also contains a large coal field, which is being mined primarily to supply adjacent coal fired power stations. The emission inventory for this area is based on previous work done by Liebenberg-Enslin et al. (2009).

3.7.1 Emission Sources on the Waterberg Area

The Waterberg area’s largest sources for airborne emissions are from coal fired power stations, domestic fuel burning and vehicle tail pipe emissions.

3.7.1.1 Power Generation

Two coal fired power stations, Matimba and Medupi are located within the Waterberg area. Both are located near Lephalale and obtain their coal from the Waterberg coal field. Matimba power station consists of six 665 MW units to produce a total of 3990 MW. The main source of emissions are two flue stacks located at the station.

Medupi power station was commissioned in 2012 with currently (Aug 2016) only one unit operational. It will have a total power generation capacity of 4800 MW which will consist of six 800 MW units. Initially after Medupi becomes fully operational it will operate for six years without the required flue gas desulphurisation (FGD) to regulate SO$_2$ emissions as prescribed by the Minimum Emission Standards. (Zunckel & Raghunandan, 2014) The emission rates used as input are from this interim period and emissions from both stacks will be included in the deposition simulation. Emission rates are shown below in Table 3.19
Table 3.19 Emissions for Matimba and Medupi (Zunckel & Raghunandan, 2014).

<table>
<thead>
<tr>
<th>Facility</th>
<th>Stack Height (m)</th>
<th>Stack Diameter (m)</th>
<th>Elevation (m)</th>
<th>Stack Exit Temperature (K)</th>
<th>Stack Exit Velocity (m/s)</th>
<th>Emissions (tpa)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Matimba</td>
<td>250</td>
<td>12.82</td>
<td>880</td>
<td>405</td>
<td>24.84</td>
<td>154631 33796</td>
</tr>
<tr>
<td>Matimba</td>
<td>250</td>
<td>12.82</td>
<td>880</td>
<td>405</td>
<td>24.84</td>
<td>154631 33796</td>
</tr>
<tr>
<td>Medupi</td>
<td>220</td>
<td>15.4</td>
<td>912</td>
<td>410.14</td>
<td>15.4</td>
<td>245436 46019</td>
</tr>
<tr>
<td>Medupi</td>
<td>220</td>
<td>15.4</td>
<td>912</td>
<td>410.14</td>
<td>15.4</td>
<td>245436 46019</td>
</tr>
</tbody>
</table>

3.7.1.2 Domestic Household Fuel Burning

From the Community Survey conducted by Stats SA in 2007 energy usage statistics and household numbers were used to determine the number and spatial distribution of households using the various types of fuel, either for cooking or heating purposes. The types of fuel used for domestic sources are wood, paraffin and gas. The Community Survey indicated that no coal was used for cooking or heating within the area (Liebenberg-Enslin et al. 2009).

Diurnal and seasonal trends were also modelled. Diurnal patterns were taken from literature (Annegarn and Grant, 1999), while seasonal trends were mostly affected by residential space heating trends. These trends are mainly affected by how the minimum daily temperature varies throughout the year and were estimated by calculating the amount of “heating-degree-days” (HDD). Figure 3.4 and Figure 3.5 show how the fuel burning activities varies throughout the year and throughout a 24h day cycle.
The total amount of each type of fuel used by each household was calculated by combining typical monthly fuel usage figures (Afrane-Okese, 1999) with the household data stated above. Together with emission factors emission rates could then be calculated as shown in Table 3.20 below.
Table 3.20 Emission factors and calculated NO emissions rates for household fuels (Liebenberg-Enslin et. al., 2010)

<table>
<thead>
<tr>
<th>Fuel</th>
<th>Emission Factors</th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>SO$_2$ (g kg$^{-1}$)</td>
<td>NO (g kg$^{-1}$)</td>
</tr>
<tr>
<td>Paraffin</td>
<td>0.1(a)</td>
<td>1.5(b)</td>
</tr>
<tr>
<td>Wood</td>
<td>0.2(c)</td>
<td>1.3(c)</td>
</tr>
<tr>
<td>Gas</td>
<td>0.01(d)</td>
<td>1.4(d)</td>
</tr>
</tbody>
</table>

Quantified emissions

<table>
<thead>
<tr>
<th>Emission Rates</th>
<th>SO$_2$ (tpa)</th>
<th>NO (tpa)</th>
<th>PM10 (tpa)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Paraffin, wood, gas</td>
<td>0.06</td>
<td>0.63</td>
<td>3.36</td>
</tr>
</tbody>
</table>

Notes:
(a) Based on sulphur content of paraffin (<0.01% Sulphur).
(b) US-EPA emission factors for kerosene usage (EPA, 1996).
(c) Based on US-EPA emission factor for residential wood burning (EPA, 1996).
(d) Based on US-EPA emission factor for residential gas burning (EPA, 1996).

3.7.1.3 Vehicle Tail Pipe Emissions

Here emission rates for the main roads within the area were calculated using the European emission factors, which have been shown to compare very well with emission factors developed for the South African Highveld by Wong (1999) and Stone (2000). Fourteen roads were included in the inventory and are listed in Table 3.21.

The total estimated emissions were estimated to be:
- SO$_2$ at 2.6 tpa.
- NO$_x$115 tpa.
- PM10 1.3 tpa.
Table 3.21 Main roads inventory for the Waterberg area (Liebenberg-Enslin et al., 2010)

<table>
<thead>
<tr>
<th>Road</th>
<th>Average Daily Traffic</th>
<th>% Heavy Vehicles</th>
</tr>
</thead>
<tbody>
<tr>
<td>Road D1675 West of Steenbokpan</td>
<td>75</td>
<td>9.3</td>
</tr>
<tr>
<td>Road D1675 Steenbokpan to Medupi</td>
<td>150</td>
<td>15.9</td>
</tr>
<tr>
<td>Road D1675 East of Medupi</td>
<td>800</td>
<td>12.5</td>
</tr>
<tr>
<td>Road D175 South of Steenbokpan</td>
<td>95</td>
<td>8.6</td>
</tr>
<tr>
<td>Road D175 North of Steenbokpan</td>
<td>120</td>
<td>6.8</td>
</tr>
<tr>
<td>Road D2286 Along Limpopo River</td>
<td>100</td>
<td>5.0</td>
</tr>
<tr>
<td>Road D1925 South of Medupi</td>
<td>25</td>
<td>12.5</td>
</tr>
<tr>
<td>Road D2001 North of Matimba</td>
<td>35</td>
<td>3.0</td>
</tr>
<tr>
<td>Nelson Mandela East of D2001</td>
<td>4460</td>
<td>7.0</td>
</tr>
<tr>
<td>Road D2001 North of D1675 (A)</td>
<td>4310</td>
<td>7.0</td>
</tr>
<tr>
<td>Road D2001 North of D1675 (B)</td>
<td>2290</td>
<td>7.0</td>
</tr>
<tr>
<td>Matimba Turn-off Road</td>
<td>2020</td>
<td>6.3</td>
</tr>
<tr>
<td>Grootgeluk Road</td>
<td>1500</td>
<td>5.4</td>
</tr>
<tr>
<td>Marapong Road (D2816)</td>
<td>790</td>
<td>10</td>
</tr>
</tbody>
</table>

3.7.1.4 Summary of Waterberg sulphur and nitrogen emissions

The Waterberg area largest emissions of NO\textsubscript{X} and SO\textsubscript{2} are due to power generation. Emissions from household burning and vehicle tailpipe will be excluded due to the very small amounts of SO\textsubscript{2} and NO\textsubscript{X} these sources contribute to the total emission rates (Table 3.22).
Table 3.22 Total SO2 an NOX emissions calculated for the Waterberg area

<table>
<thead>
<tr>
<th>Source</th>
<th>SO2 (tpa)</th>
<th>% SO2</th>
<th>NOx (tpa)</th>
<th>% NOx</th>
</tr>
</thead>
<tbody>
<tr>
<td>Power Generation</td>
<td>309262 (a)</td>
<td>38.65</td>
<td>67592 (a)</td>
<td>42.31</td>
</tr>
<tr>
<td></td>
<td>490872 (b)</td>
<td>61.35</td>
<td>92038 (b)</td>
<td>57.62</td>
</tr>
<tr>
<td>Household Burning</td>
<td>0.06</td>
<td>0.00001</td>
<td>0.63</td>
<td>0.0004</td>
</tr>
<tr>
<td>Tailpipe Emissions</td>
<td>2.6</td>
<td>0.0003</td>
<td>115</td>
<td>0.07</td>
</tr>
<tr>
<td>Total</td>
<td>800136.66</td>
<td></td>
<td>159745.63</td>
<td></td>
</tr>
</tbody>
</table>

Notes:
(a) Matimba
(b) Medupi

3.8 Biogenic emissions

Most of the work on NOx emission within Southern Africa has so far been focused on arid environments such as the Kalahari and savannah areas due to their importance to global NOx emissions. Areas such as the grasslands of the Highveld have not been studied extensively and are less well understood (Feig, 2008; Scholes, 1997).

NOx soil emissions for the Highveld and the Waterberg are based on work done by Yienger and Levy (1995), which was used to estimate global NOx emissions. In their study Yienger and Levy (1995) determined NOx flux as a function of soil temperature (T_{Soil}), an emission coefficient that depends on the type of biome (A_{W/D}), a scalar precipitation pulse value (P) and a scalar factor that accounts for the canopy reduction (CR) effect within biomes that tends to reduce the NOx flux to the atmosphere (see equation 19 below).

\[
Flux = f_{W/D}(T_{Soil},A_{W/D}(Biome)) \times P \times CR(LAI,SAI)
\]  

The general calculation process is summed up as follows:
• Breaking up of modelling domain into smaller area sources.
• Identifying the type of land use within each area source and calculating the fraction of land area each type of land use takes up.
• Calculating an emission coefficient ($A_{W/D}$) for each type of land use type for both dry and wet soil conditions.
• Sourcing of ground level atmospheric temperature and rainfall rates for the duration of each of the modelling periods for each cell.
• Converting atmospheric ground level temperatures to soil temperatures.
• Identifying wet and dry conditions for each cell and calculating the resulting NO$_x$ emissions, using the changing soil temperature and rainfall rates throughout the modelling period of a year without taking pulsing events into account.
• Identifying NO$_x$ pulsing events and the size and duration of those events and applying them to the emission rates above.
• Calculating total NO emission rates for each cell in g/s.
• Converting the NO$_x$ emissions into a CALPUFF input file.

Because of the size of the modelling domain and lack of LAI and SAI values for the canopy reduction effect, the canopy reduction factor was assumed to 1. These values varies by season and land use type and would have added to the complexity of the model. Without the added LAI/SAI values the emission calculated will be the emission directly emitted by the soil, without being impeded by plant growth. There is also a lot of uncertainty imbedded in the LAI/SAI model, because of uncertainty in accurately describing the process where NO$_X$ interacts with intercanopy effects (Yienger and Levy, 1995).

Each step is discussed in more detail below.
3.8.1 Area Sources and Land use

Land use was sourced from the CALMET file set up for the CALPUFF simulations. The modelling domain is divided into smaller cells, each one representing the overall land use for that area. The different types of land use assigned by CALMET to these cells are as follows:

- Urban
- Agricultural
- Rangeland
- Forest
- Water
- Barren

To accommodate the limited number of area sources that CALPUFF can process within one input file and to facilitate biogenic calculations within a reasonable amount of time, the modelling domain was divided into larger area sources each consisting of a number of these smaller cells. For each of these area sources the fraction of the different types of land use areas was calculated by counting the number of different types of cells.

3.8.2 Rainfall and Soil Temperature

Several factors such as cloud cover, solar insolation, vegetative cover and soil properties affect the relationship between soil and air temperature. Yienger and Levy made use of empirical relationships that were derived by Williams et al. (1992b) by doing linear least squares fitting to air and soil temperature data to convert the extracted ground level atmospheric temperatures to soil temperatures. These relationships are shown below in Table 3.23.

Large scale atmospheric temperature and rainfall measurements are not available for the Highveld or the Waterberg areas on an hourly basis, but lower atmospheric
temperature and average rainfall values can be extracted for each defined area source from a CALMET file by using the PRTMET program that is part of the CALPUFF modelling suite.

### Table 3.23 Soil and Air Temperature Relationships for different biomes (Williams et al., 1992b)

<table>
<thead>
<tr>
<th>Land Use Type</th>
<th>Estimated Soil Temperature</th>
</tr>
</thead>
<tbody>
<tr>
<td>Natural</td>
<td></td>
</tr>
<tr>
<td>Grasslands</td>
<td>$T_{\text{soil}} = 0.66T_{\text{air}} + 8.8$ (20)</td>
</tr>
<tr>
<td>Forests</td>
<td>$T_{\text{soil}} = 0.84T_{\text{air}} + 3.6$ (21)</td>
</tr>
<tr>
<td>Wetlands</td>
<td>$T_{\text{soil}} = 0.92T_{\text{air}} + 4.4$ (22)</td>
</tr>
<tr>
<td>Agricultural</td>
<td></td>
</tr>
<tr>
<td>Corn</td>
<td>$T_{\text{soil}} = 0.72T_{\text{air}} + 5.8$ (23)</td>
</tr>
<tr>
<td>Wheat</td>
<td>$T_{\text{soil}} = 1.03T_{\text{air}} + 2.9$ (24)</td>
</tr>
<tr>
<td>Soybeans</td>
<td>$T_{\text{soil}} = 1.03T_{\text{air}} + 2.9$ (25)</td>
</tr>
</tbody>
</table>

### 3.8.3 NO$_x$ flux temperature relationships

Soil NO emissions vary due to soil moisture content and temperature. Instead of including a term explicitly for soil moisture in a temperature and NO flux relationship, Yienger and Levy fitted an exponential curve to wet soils and linear curve to dry soils using emission data from available literature for different biomes. To distinguish when each curve can be applied to the soil conditions, the soil is classified as dry when the rain rate drops below 0.1 cm/day and wet if the rain rate is above that.

### 3.8.4 Wet soil NO$_x$ emissions

For wet soils three temperature intervals were chosen to model NO$_x$ emissions as temperature increases: 0 to 10°C, 10 to 30°C and 30°C and up. 0 to 10°C the rise in NO$_x$ to temperature is close to linear, while from 10 to 30°C NO$_x$ flux increases exponentially. Above 30°C the emissions remain constant. Below 0°C it was assumed that no emissions take place. In Table 3.24 below these relationships are shown for each interval.
Table 3.24 Wet soil NOx flux temperature dependencies (Yienger and Levy, 1995)

<table>
<thead>
<tr>
<th>Temperature Range</th>
<th>Function</th>
</tr>
</thead>
<tbody>
<tr>
<td>0 to 10°C</td>
<td>[ f_w = 0.28 \times A_w(biome) \times T ] (26)</td>
</tr>
<tr>
<td>10 to 30°C</td>
<td>[ f_w = A_w(biome) \times e^{0.103\pm0.04 \times T} ] (27)</td>
</tr>
<tr>
<td>&gt; 30°C</td>
<td>[ f_w = 21.97 \times A_w(biome) ] (28)</td>
</tr>
</tbody>
</table>

The k value (0.103±0.04) was assumed by Yienger and Levy to be globally constant and was calculated from a range of exponential temperature dependencies for wide range of biomes for which k values was available. \( A_w \) for each biome was calculated by applying available NO\(_x\) flux data and temperature measurements to equations 6 to 7.

3.8.5 Dry soil NO\(_x\) emissions

Dry soils do not have the same well defined dependencies on temperature that wet soils have. In this case, only two temperature intervals were used to calculate NO\(_x\) flux rate: between 0 and 30°C the flux rate increases linearly and above 30°C a constant flux rate is reached. These relationships are shown in Table 3.25.

Table 3.25 Dry soil temperature relationships (Yienger and Levy, 1995)

<table>
<thead>
<tr>
<th>Temperature Range</th>
<th>Function</th>
</tr>
</thead>
<tbody>
<tr>
<td>0 to 30°C</td>
<td>[ f_d = (A_d(biome) \times T) / 30 ] (29)</td>
</tr>
<tr>
<td>&gt; 30°C</td>
<td>[ f_d = A_d(biome) ] (30)</td>
</tr>
</tbody>
</table>

3.8.6 Pulsing

To distinguish between heavy, long lasting pulses and short term pulses a scheme was developed and is shown below in Table 3.26. As stated above, for rainfall below 0.1 cm/day the soil is dry and no pulse is emitted. For higher rainfall, pulses vary in size and get larger as the rainfall amount increases. A pulse also decays exponentially as time goes on after the initial release. The length of each pulse depends on the amount of rainfall in the area and varies from 3 days to 2 weeks.
Table 3.26: Pulse scheme (Yienger and Levy, 1995)

<table>
<thead>
<tr>
<th>Rain rate (cm/day)</th>
<th>Pulse description (F_N = Normal calculated flux rate)</th>
<th>Event description</th>
</tr>
</thead>
<tbody>
<tr>
<td>Rain &lt; 0.1</td>
<td>No pulse</td>
<td>No Rain</td>
</tr>
<tr>
<td>0.1 &lt; Rain &lt; 0.5</td>
<td>3-day pulse, starting at 5 × F_N with exponential decay</td>
<td>Sprinkle</td>
</tr>
<tr>
<td>0.5 &lt; Rain &lt; 1.5</td>
<td>1 week pulse, starting at 10 × F_N with exponential decay</td>
<td>Shower</td>
</tr>
<tr>
<td>1.5 &lt; Rain</td>
<td>2-week pulse, starting at 15 × F_N with exponential decay</td>
<td>Heavy Rain</td>
</tr>
</tbody>
</table>

The temporal patterns for each type of pulse are shown below in Table 3.27. A pulse will decrease continuously and will not stop if a new pulse starts or if one is already occurring within a specific area.

Table 3.27: NOx pulse temporal patterns, t = days (Yienger and Levy, 1995)

<table>
<thead>
<tr>
<th>Type</th>
<th>Equation</th>
<th>Parameters</th>
</tr>
</thead>
<tbody>
<tr>
<td>No Pulse</td>
<td>P = 1.0</td>
<td></td>
</tr>
<tr>
<td>Sprinkle</td>
<td>P = 11.19 × e^{-0.805[day^{-1}]t}</td>
<td>(1 &lt; t &lt; 3)</td>
</tr>
<tr>
<td>Shower</td>
<td>P = 14.68 × e^{-0.384[day^{-1}]t}</td>
<td>(1 &lt; t &lt; 7)</td>
</tr>
<tr>
<td>Heavy Rain</td>
<td>P = 18.46 × e^{-0.208[day^{-1}]t}</td>
<td>(1 &lt; t &lt; 14)</td>
</tr>
</tbody>
</table>

3.8.7 Estimated Highveld NO_x emissions

3.8.7.1 Area sources and land use

The 350 × 350 km modelling domain was divided into 196 area sources. Each area source consists of 16 to 25 cells, 5 × 5 km large and representing the overall type of land use for that area (see Figure 3.6 below). The fraction of land use type for each area source was calculated by adding the number of cells of the same land use type and then dividing them by the total number of cell within the area source (See Appendix B: Table 8.1 Highveld land use values defined for each cell). It should be noted that the coordinate system used for this area (see Figure 3.6 below) are Lambert Conic Conformal (LCC) due to the fact the standard UTM coordinates became distorted over such a large area.
Figure 3.6 196 defined area sources and land use for the Highveld (LCC coordinates)
3.8.7.2 Emission coefficients

No distinction was made between range land and agricultural land. They were assumed to be the same as calculated for grasslands by Yienger and Levy (1995). Uncertainty on the type of crop grown and the fertilizer loads each year meant that it was impossible to superimpose this over the current land use map. The only other land use types that affected the biogenic calculation were the forest/plantations to the east of the modelling domain, main urban areas within the Gauteng province and some water bodies (mainly the Vaal dam in central Gauteng). Urban and barren land types were assumed to emit no biogenic NOx. The values used for both grassland and forest/plantations are shown below in Table 3.28 below.

Table 3.28 Emission coefficients for the Highveld (Yienger and Levy, 1995)

<table>
<thead>
<tr>
<th>Biome</th>
<th>A_W</th>
<th>A_D</th>
</tr>
</thead>
<tbody>
<tr>
<td>Grasslands</td>
<td>0.36</td>
<td>2.65</td>
</tr>
<tr>
<td>Forest</td>
<td>0.03</td>
<td>0.22</td>
</tr>
</tbody>
</table>

3.8.7.3 Rainfall and temperature

Sourcing of ground level atmospheric temperature and rainfall rates for the duration of each of the three modelling (2001, 2003 and 2010) periods for each cell was done from CALMET as mentioned above. It should be noted that to create a CALMET file, MM5 data is used as input and the rainfall data extracted from the CALMET file would be that predicted by the MM5 file. An average temperature and rainfall value for the entire area covered by each area source was extracted from CALMET for every hour for an entire year. The atmospheric ground level temperatures were then converted to soil temperatures by using the formulas in Table 3.25.
3.8.7.4 Modelling biogenic NO\textsubscript{X} in CALPUFF

For each defined area source an hourly NO\textsubscript{X} emission was calculated using equations 26 to 28 when the soil was wet and equations 29 to 30 when dry. At the same time using the rainfall rates pulsing events were identified and applied when they occur using equations 31 to 34. Finally, for each of the 196 area sources the hourly NO\textsubscript{X} value for every hour for the entire year was then input into CALPUFF using a Buoyant Area Source Emissions File with Arbitrarily Varying Emissions (BAEMARB.DAT) file.

These emissions could then be included into the M2 model to study the impact form biogenic emission on nitrogen deposition on the Highveld. This was done for each of the three modelling periods of 2001, 2010 and 2003. This will be referenced as Model 4 (M4).

Model limitations:

- Rainfall rates were extracted from the CALMET files generated for each of the three M2 modelling periods. Because as has been mentioned above in section 3.5.1 the MM5 data used to generate the CALMET files over predicted the rainfall rates this means the predicted biogenic NO\textsubscript{X} emissions for each modelling period may also be over predicted.
- Canopy reduction was not considered.

3.8.8 Estimated Waterberg NO\textsubscript{X} emissions

3.8.8.1 Area sources and land use

25 area sources were defined for the Waterberg area, each consisting from 475 to 520 cells of 1 km ×1 km. Each cell has land use type attached to it (See Figure 3.7 below). The fraction land area for each type of land use type within each defined area source is calculated in the same manner as for the Highveld. This is
summarised in Appendix B: **Table 8.2** Waterberg land use values defined for each cell. Standard UTM coordinates were used for this area and is also used in Figure 3.7.

### 3.8.8.2 Emission coefficients

Here a distinction was made between rangeland and agricultural land. Grassland emission coefficients were again assigned to the agricultural land use type for the same reasons mentioned above. The rangeland land use type was treated as savanna biome. For this biome emission coefficients were calculated using the same method as Yienger and Levy. Temperature and flux were applied to equations 26, 27, 28 and 29 to calculate an $A_{W/D}$ value. These values were then linearized and from this an average $A_{W/D}$ coefficient was calculated. This process is summarised below in Table 3.28 and Table 3.29.

**Table 3.29 Wet Savanna Biome emission coefficient calculation**

<table>
<thead>
<tr>
<th>Temperature Interval</th>
<th>Flux $\text{NO/m}^2\text{s}$ (ng)</th>
<th>Flux $\text{N/m}^2\text{s}$ (ng)</th>
<th>$T$ ($°C$)</th>
<th>$A_w$</th>
<th>$\ln A_w$</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>10-30 °C</td>
<td>20</td>
<td>28</td>
<td>1,18262</td>
<td>0,167732</td>
<td>-2,40455</td>
<td>Scholes et al., 1997</td>
</tr>
<tr>
<td></td>
<td>1,128</td>
<td>25</td>
<td>0,090306</td>
<td>-3,01766</td>
<td>-2,74939</td>
<td>Feig et al., 2008</td>
</tr>
<tr>
<td></td>
<td>0,611</td>
<td>25</td>
<td>0,048916</td>
<td>-2,87456</td>
<td></td>
<td>Otter et al., 1999</td>
</tr>
<tr>
<td></td>
<td>0,799</td>
<td>25</td>
<td>0,063967</td>
<td>-0,14434</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>0,705</td>
<td>25</td>
<td>0,056441</td>
<td>-2,74609</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>13,026</td>
<td>25</td>
<td>1,04284</td>
<td>0,041947</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>10,812</td>
<td>25</td>
<td>0,86559</td>
<td>-1,65398</td>
<td></td>
<td></td>
</tr>
<tr>
<td>&gt;30 °C</td>
<td>1,645</td>
<td>35</td>
<td>0,074875</td>
<td>-2,59194</td>
<td></td>
<td>Feig et al., 2008</td>
</tr>
<tr>
<td></td>
<td>1,41</td>
<td>35</td>
<td>0,064178</td>
<td>-2,74609</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>1,645</td>
<td>35</td>
<td>0,074875</td>
<td>-2,59194</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>45</td>
<td>41</td>
<td>2,048248</td>
<td>0,716985</td>
<td></td>
<td></td>
</tr>
<tr>
<td><strong>Average</strong></td>
<td></td>
<td></td>
<td>-1,65398</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td><strong>$A_w$ Savanna</strong></td>
<td></td>
<td></td>
<td>0,191287</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>
Figure 3.7 25 defined area sources and land use for the Waterberg area (UTM coordinates)

(Isopleth bars are an artefact generated by the mesh factor used in CALMET file generation)
Table 3.30 Dry Savanna Biome emission coefficient calculation

<table>
<thead>
<tr>
<th>Temperature Interval</th>
<th>Flux (ng N/m²s)</th>
<th>T (°C)</th>
<th>A₀</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>0-30 °C</td>
<td>2</td>
<td>28</td>
<td>2.142857</td>
<td>Scholes et al, 1997</td>
</tr>
</tbody>
</table>

3.8.8.3 Rainfall and temperature

Rainfall could not be sourced from the CALMET file as was the case for the Highveld. In this case, yearly rainfall for Lephala was supplied by the South African Weather Service. Temperature for each area source was sourced from CALMET.

3.8.8.4 Modelling biogenic NOₓ in CALPUFF

Like for the Highveld, an hourly NOₓ rate was calculated for each of the area sources define above. For each of the 25 area sources the hourly NOₓ value for each day for the entire modelling period of 2006 was then input into CALPUFF using a BAEMARBDAT file, which could then be added to the model constructed above for the Waterberg area to study the effect biogenic NOₓ will have on nitrogen deposition for this area.

Model limitations:

- Rainfall used to predict NOₓ represents only a single point and does not consider the spatial rainfall pattern for this the area.
- Canopy reduction was not considered.
4. RESULTS AND DISCUSSION

4.1 Estimated NO\textsubscript{x} emissions for the South African Highveld.

The calculated total annual biogenic NO\textsubscript{x} for the entire modelling domain for the three modelling periods are shown below in Table 4.1, 4.2 and 4.3 and are compared to the other NO\textsubscript{x} sources from the 2001 emission inventory. Biomass burning values from the previous Scorgie and Kornelius (2009) study are included for comparison.

The biogenic NO\textsubscript{x} emitted made up 3.96 %, 4.14 % and 3.34 % of total released NO\textsubscript{x} for 2001, 2003 and 2010 respectively. This is significantly more than is released by household fuel burning, small industrial sources and biomass burning.

The reason for the large amount of nitrogen contributed to the atmosphere is the large area covered by the modelling domain. The entire area is 350 km × 350 km large consisting of 196 smaller sections, each 25 km × 25 km.

Table 4.1 Total Highveld NO\textsubscript{x} emissions for 2001 compared to NO\textsubscript{x} emissions from the 2001 emission inventory.

<table>
<thead>
<tr>
<th>Source group</th>
<th>NO\textsubscript{x} (tpa)</th>
<th>%NO\textsubscript{x}</th>
</tr>
</thead>
<tbody>
<tr>
<td>Major sources(a)</td>
<td>526345</td>
<td>73.83</td>
</tr>
<tr>
<td>Other industrial sources</td>
<td>4099</td>
<td>0.57</td>
</tr>
<tr>
<td>Household fuel burning</td>
<td>3449</td>
<td>0.48</td>
</tr>
<tr>
<td>Vehicle exhaust emissions</td>
<td>147577</td>
<td>20.70</td>
</tr>
<tr>
<td>Biomass burning (wildfires)</td>
<td>3204</td>
<td>0.45</td>
</tr>
<tr>
<td>Biogenic NO\textsubscript{x}</td>
<td>28249</td>
<td>3.96</td>
</tr>
<tr>
<td>Total</td>
<td>712923</td>
<td></td>
</tr>
</tbody>
</table>
Table 4.2 Total Highveld NOx emissions for 2003 compared to NOX emissions from the 2001 emission inventory.

<table>
<thead>
<tr>
<th>Source group</th>
<th>NOx (tpa)</th>
<th>%NOx</th>
</tr>
</thead>
<tbody>
<tr>
<td>Major sources(a)</td>
<td>526345</td>
<td>73.70</td>
</tr>
<tr>
<td>Other industrial sources</td>
<td>4099</td>
<td>0.57</td>
</tr>
<tr>
<td>Household fuel burning</td>
<td>3449</td>
<td>0.48</td>
</tr>
<tr>
<td>Vehicle exhaust emissions</td>
<td>147577</td>
<td>20.66</td>
</tr>
<tr>
<td>Biomass burning (wild fires)</td>
<td>3204</td>
<td>0.45</td>
</tr>
<tr>
<td>Biogenic NO</td>
<td>29540</td>
<td>4.14</td>
</tr>
<tr>
<td><strong>Total</strong></td>
<td><strong>714214</strong></td>
<td></td>
</tr>
</tbody>
</table>

Table 4.3 Total Highveld NOx emissions for 2010 compared to NOX emissions from the 2001 emission inventory.

<table>
<thead>
<tr>
<th>Source group</th>
<th>NOx (tpa)</th>
<th>%NOx</th>
</tr>
</thead>
<tbody>
<tr>
<td>Major sources(a)</td>
<td>526345</td>
<td>74.22</td>
</tr>
<tr>
<td>Other industrial sources</td>
<td>4099</td>
<td>0.58</td>
</tr>
<tr>
<td>Household fuel burning</td>
<td>3449</td>
<td>0.49</td>
</tr>
<tr>
<td>Vehicle exhaust emissions</td>
<td>147577</td>
<td>20.81</td>
</tr>
<tr>
<td>Biomass burning (wild fires)</td>
<td>3204</td>
<td>0.45</td>
</tr>
<tr>
<td>Biogenic NO</td>
<td>24475</td>
<td>3.45</td>
</tr>
<tr>
<td><strong>Total</strong></td>
<td><strong>709149</strong></td>
<td></td>
</tr>
</tbody>
</table>
4.1.1 Temporal Biogenic NO\textsubscript{x} Emission Patterns

The monthly average NO\textsubscript{x} emission rate calculated for the 25 km × 25 km blocks ranges from:

- 2.11 – 7.73 g s\textsuperscript{-1}, with an annual average rate of 4.53 g s\textsuperscript{-1} for 2001.
- 1.94 – 9 g s\textsuperscript{-1} with an annual average rate of 3.96 g s\textsuperscript{-1} for 2010.
- 2.11 – 7.62 g s\textsuperscript{-1} with an annual average rate of 4.78 g s\textsuperscript{-1} for 2003.

The monthly NO\textsubscript{x} emitted is shown for each modelled year in Figures 4.1 to 4.3 below. The highest amounts of NO\textsubscript{x} occur in the spring and autumn months. These are the months when the largest NO\textsubscript{x} pulses are releases on the Highveld. These time periods produce conditions when the soil remains dry for long enough periods for soil nutrients to build up and for large enough rainstorms to wet the soil quickly. When the soil moisture content remains constant (wet or dry) no pulses occur. Total NO\textsubscript{x} emitted for the modelling domain varies between:

- 1388.25 t month\textsuperscript{-1} – 4217.24 t month\textsuperscript{-1} for 2001.
- 988.06 t month\textsuperscript{-1} – 4571.44 t month\textsuperscript{-1} for 2010.
- 1105.95 t month\textsuperscript{-1} – 3924.95 t month\textsuperscript{-1} for 2003.

![Figure 4.1 Seasonal biogenic emissions for 2001](image-url)
4.1.2 Contribution of biogenic NO\textsubscript{x} emission on the nitrogen concentration and deposition on the South African Highveld.

The effect of the biogenic NO\textsubscript{x} emission on ambient nitrogen based gases and on nitrogen deposition at selected receptor locations is shown in Tables 4.7 to 4.11. Main trends are:
• Ambient annual NO concentration contribution varies between 2.59 % and 35.37 % (Table 4.4).

• Ambient annual NO₂ concentration contribution varies between 5.21 % and 9.25 % (Table 4.5).

• Dry nitrogen deposition rates have been affected the most. Biogenic NOₓ added between 1.69 - 6.19 % (Table 4.6).

• Wet deposition rates were affected very little due to the secondary nature of the ammonium salts formed in the atmosphere (0.13 % to 0.75 %) (Table 4.7).

• Total contribution to nitrogen deposition rates ranged from 0.32 % to 1.77 % (Table 4.8).

Dry N deposition rates were the most affected by biogenic NOₓ emissions. This is because the wet N rates are much higher than dry N rates. Comparing the predicted dry N and wet N deposition rates due to biogenic NOₓ at each receptor point the dry N rates are from 1.14 to 2.08 times larger. Another reason could be that during dry periods conditions are favourable for dry deposition after a short rain spell. Wetted surface due to any rainfall would also enhance the removal of rates due to dry deposition.

Table 4.4 Biogenic dry N and wet N comparison for 2001

<table>
<thead>
<tr>
<th>Modelled (2001)</th>
<th>Biogenic Dry N (kg N ha⁻¹ yr⁻¹)</th>
<th>Biogenic Wet N (kg N ha⁻¹ yr⁻¹)</th>
<th>Dry N/Wet N</th>
</tr>
</thead>
<tbody>
<tr>
<td>Verkykkop</td>
<td>0.0346</td>
<td>0.0166</td>
<td>2.08</td>
</tr>
<tr>
<td>Elandsfontein</td>
<td>0.0593</td>
<td>0.0356</td>
<td>1.67</td>
</tr>
<tr>
<td>Kendal2</td>
<td>0.0693</td>
<td>0.0371</td>
<td>1.87</td>
</tr>
<tr>
<td>Leandra</td>
<td>0.0701</td>
<td>0.0463</td>
<td>1.51</td>
</tr>
<tr>
<td>Majuba1</td>
<td>0.0361</td>
<td>0.0215</td>
<td>1.68</td>
</tr>
<tr>
<td>Palmer</td>
<td>0.0159</td>
<td>0.0139</td>
<td>1.14</td>
</tr>
<tr>
<td>Amersfoort</td>
<td>0.0346</td>
<td>0.0223</td>
<td>1.55</td>
</tr>
</tbody>
</table>
### Table 4.5 Percentage ambient NO gas concentrations contributed by biogenic sources

<table>
<thead>
<tr>
<th>Modelled (2001)</th>
<th>Total NO</th>
<th>Biogenic NO</th>
<th>% Biogenic NO</th>
</tr>
</thead>
<tbody>
<tr>
<td>Verkykkop</td>
<td>1.5581</td>
<td>0.2654</td>
<td>17.03</td>
</tr>
<tr>
<td>Elandsfontein</td>
<td>1.511</td>
<td>0.0391</td>
<td>2.59</td>
</tr>
<tr>
<td>Kendal2</td>
<td>0.37082</td>
<td>0.0195</td>
<td>5.25</td>
</tr>
<tr>
<td>Leandra</td>
<td>0.93216</td>
<td>0.0223</td>
<td>2.39</td>
</tr>
<tr>
<td>Majuba1</td>
<td>1.7931</td>
<td>0.2195</td>
<td>12.24</td>
</tr>
<tr>
<td>Palmer</td>
<td>0.014635</td>
<td>0.0052</td>
<td>35.37</td>
</tr>
<tr>
<td>Amersfoort</td>
<td>1.7546</td>
<td>0.1869</td>
<td>10.65</td>
</tr>
</tbody>
</table>

### Table 4.6 Percentage ambient NO$_2$ gas concentrations contributed by biogenic sources

<table>
<thead>
<tr>
<th>Modelled (2001)</th>
<th>Total NO2</th>
<th>Biogenic NO2</th>
<th>% Biogenic NO2</th>
</tr>
</thead>
<tbody>
<tr>
<td>Verkykkop</td>
<td>0.63929</td>
<td>0.0337</td>
<td>5.28</td>
</tr>
<tr>
<td>Elandsfontein</td>
<td>4.8618</td>
<td>0.2545</td>
<td>5.23</td>
</tr>
<tr>
<td>Kendal2</td>
<td>2.9686</td>
<td>0.2747</td>
<td>9.25</td>
</tr>
<tr>
<td>Leandra</td>
<td>3.1033</td>
<td>0.2726</td>
<td>8.78</td>
</tr>
<tr>
<td>Majuba1</td>
<td>1.0235</td>
<td>0.0525</td>
<td>5.13</td>
</tr>
<tr>
<td>Palmer</td>
<td>0.82742</td>
<td>0.0597</td>
<td>7.21</td>
</tr>
<tr>
<td>Amersfoort</td>
<td>1.166</td>
<td>0.0607</td>
<td>5.21</td>
</tr>
</tbody>
</table>

### Table 4.7 Percentage dry N contributed by biogenic sources

<table>
<thead>
<tr>
<th>Modelled (2001) (kg N ha$^{-1}$ yr$^{-1}$)</th>
<th>Dry N</th>
<th>Biogenic Dry N</th>
<th>% Biogenic Dry N</th>
</tr>
</thead>
<tbody>
<tr>
<td>Verkykkop</td>
<td>0.55836</td>
<td>0.0346</td>
<td>6.19</td>
</tr>
<tr>
<td>Elandsfontein</td>
<td>1.8566</td>
<td>0.0593</td>
<td>3.19</td>
</tr>
<tr>
<td>Kendal2</td>
<td>4.0918</td>
<td>0.0693</td>
<td>1.69</td>
</tr>
<tr>
<td>Leandra</td>
<td>4.0998</td>
<td>0.0701</td>
<td>1.71</td>
</tr>
<tr>
<td>Majuba1</td>
<td>0.68906</td>
<td>0.0361</td>
<td>5.24</td>
</tr>
<tr>
<td>Palmer</td>
<td>0.78101</td>
<td>0.0159</td>
<td>2.04</td>
</tr>
<tr>
<td>Amersfoort</td>
<td>0.70625</td>
<td>0.0346</td>
<td>4.91</td>
</tr>
</tbody>
</table>
Table 4.8 Percentage wet N contributed by biogenic sources

<table>
<thead>
<tr>
<th></th>
<th>Wet N</th>
<th>Biogenic Wet N</th>
<th>% Biogenic Wet N</th>
</tr>
</thead>
<tbody>
<tr>
<td>Verkykkop</td>
<td>2.3194</td>
<td>0.0166</td>
<td>0.71</td>
</tr>
<tr>
<td>Elandsfontein</td>
<td>4.7408</td>
<td>0.0356</td>
<td>0.75</td>
</tr>
<tr>
<td>Kendal2</td>
<td>28.955</td>
<td>0.0371</td>
<td>0.13</td>
</tr>
<tr>
<td>Leandra</td>
<td>11.844</td>
<td>0.0463</td>
<td>0.39</td>
</tr>
<tr>
<td>Majuba1</td>
<td>3.0616</td>
<td>0.0215</td>
<td>0.7</td>
</tr>
<tr>
<td>Palmer</td>
<td>1.8876</td>
<td>0.0139</td>
<td>0.73</td>
</tr>
<tr>
<td>Amersfoort</td>
<td>3.2954</td>
<td>0.0223</td>
<td>0.68</td>
</tr>
</tbody>
</table>

Table 4.9 Percentage total N contributed by biogenic sources

<table>
<thead>
<tr>
<th></th>
<th>Total N</th>
<th>Biogenic Total N</th>
<th>% Biogenic Total N</th>
</tr>
</thead>
<tbody>
<tr>
<td>Verkykkop</td>
<td>2.8778</td>
<td>0.051</td>
<td>1.77</td>
</tr>
<tr>
<td>Elandsfontein</td>
<td>6.5975</td>
<td>0.0946</td>
<td>1.43</td>
</tr>
<tr>
<td>Kendal2</td>
<td>33.047</td>
<td>0.1064</td>
<td>0.32</td>
</tr>
<tr>
<td>Leandra</td>
<td>15.944</td>
<td>0.1164</td>
<td>0.73</td>
</tr>
<tr>
<td>Majuba1</td>
<td>3.7507</td>
<td>0.0576</td>
<td>1.54</td>
</tr>
<tr>
<td>Palmer</td>
<td>2.6686</td>
<td>0.0298</td>
<td>1.12</td>
</tr>
<tr>
<td>Amersfoort</td>
<td>4.0017</td>
<td>0.057</td>
<td>1.42</td>
</tr>
</tbody>
</table>
4.2 Contribution of Hydrogen Sulphide emission on simulated deposition

The converted H$_2$S emission released from the Sasol plant near Secunda has a noticeable effect on wet, dry and total sulphur deposition rates. This is not surprising due to the amount of sulphur released from these stacks. The percentage of sulphur contributed to wet, dry and total sulphur deposition at important receptor locations is shown below in Tables 4.9, 4.10 and 4.11. Because CALPUFF cannot simulate the conversion of H$_2$S, it should be noted that assumptions were made on the amounts of H$_2$S converted to SO$_2$ for each season using the atmospheric patterns measured by Igbafe (2007) at Elandsfontein. These were:

- Summer 70% conversion.
- Autumn 60% conversion.
- Winter 50% conversion.
- Spring 40% conversion.

Key findings are:

- From 0.23 – 9.17% (average of 4.85%) was contributed to wet sulphur deposition rates.
- From 2.50 – 8.81% (average of 5.95%) was contributed to dry sulphur deposition rates.
- From 0.32 – 9.68% (average of 5.15%) was contributed to the total sulphur deposition rates.

It should be noted that this is a conservative estimate, because of the assumptions made in the previous section and the limitations of the CALPUFF modelling suite. Not only are there more sources of H$_2$S on the Highveld that were not taken into account, but approximations were made about the H$_2$S conversion to SO$_2$ to allow the added S to take part in the simulated atmospheric chemistry and deposition mechanisms of the CALPUFF modelling suite. But even with the very basic assumptions made to account for the H$_2$S conversion, this shows that H$_2$S will contribute a notable amount of sulphur to the deposition rates which warrant further study.
Table 4.10 Contribution of $\text{H}_2\text{S}$ to wet sulphur deposition rates on the Highveld

<table>
<thead>
<tr>
<th>Modelled (2001) (kg S ha$^{-1}$yr$^{-1}$)</th>
<th>Wet S (M3)</th>
<th>$M_3 + \text{H}_2\text{S}$ emissions</th>
<th>Deposition due to $\text{H}_2\text{S}$</th>
<th>% of Wet S</th>
</tr>
</thead>
<tbody>
<tr>
<td>Verkykkop</td>
<td>8.6</td>
<td>9.31</td>
<td>0.73</td>
<td>7.81</td>
</tr>
<tr>
<td>Elandsfontein</td>
<td>17.7</td>
<td>18.18</td>
<td>0.50</td>
<td>2.77</td>
</tr>
<tr>
<td>Kendal2</td>
<td>190.8</td>
<td>191.25</td>
<td>0.44</td>
<td>0.23</td>
</tr>
<tr>
<td>Leandra</td>
<td>14.1</td>
<td>14.66</td>
<td>0.60</td>
<td>4.12</td>
</tr>
<tr>
<td>Majuba1</td>
<td>19.3</td>
<td>20.50</td>
<td>1.24</td>
<td>6.05</td>
</tr>
<tr>
<td>Palmer</td>
<td>2.2</td>
<td>2.30</td>
<td>0.09</td>
<td>3.83</td>
</tr>
<tr>
<td>Amersfoort</td>
<td>13.9</td>
<td>15.28</td>
<td>1.40</td>
<td>9.17</td>
</tr>
</tbody>
</table>

Table 4.11 Contribution of $\text{H}_2\text{S}$ to dry sulphur deposition rates on the Highveld

<table>
<thead>
<tr>
<th>Modelled (2001) (kg S ha$^{-1}$yr$^{-1}$)</th>
<th>Dry S (M3)</th>
<th>$M_3 + \text{H}_2\text{S}$ emissions</th>
<th>Deposition due to $\text{H}_2\text{S}$</th>
<th>% of Dry S</th>
</tr>
</thead>
<tbody>
<tr>
<td>Verkykkop</td>
<td>2.4</td>
<td>2.60</td>
<td>0.21</td>
<td>7.89</td>
</tr>
<tr>
<td>Elandsfontein</td>
<td>7.2</td>
<td>7.45</td>
<td>0.29</td>
<td>3.87</td>
</tr>
<tr>
<td>Kendal2</td>
<td>7.9</td>
<td>8.10</td>
<td>0.20</td>
<td>2.50</td>
</tr>
<tr>
<td>Leandra</td>
<td>6.9</td>
<td>7.31</td>
<td>0.44</td>
<td>5.98</td>
</tr>
<tr>
<td>Majuba1</td>
<td>3.2</td>
<td>3.50</td>
<td>0.29</td>
<td>8.23</td>
</tr>
<tr>
<td>Palmer</td>
<td>1.6</td>
<td>1.65</td>
<td>0.07</td>
<td>4.34</td>
</tr>
<tr>
<td>Amersfoort</td>
<td>3.4</td>
<td>3.68</td>
<td>0.32</td>
<td>8.81</td>
</tr>
</tbody>
</table>
Table 4.12 Contribution of \( \text{H}_2\text{S} \) to total Sulphur deposition rates on the Highveld

<table>
<thead>
<tr>
<th>Modelled (2001) (kg S ha(^{-1}) yr(^{-1}))</th>
<th>Total S (M3)</th>
<th>M3 + ( \text{H}_2\text{S} ) emissions</th>
<th>Deposition due to ( \text{H}_2\text{S} )</th>
<th>% of Total S</th>
</tr>
</thead>
<tbody>
<tr>
<td>Verkykkop</td>
<td>11.0</td>
<td>11.92</td>
<td>0.93</td>
<td>7.82</td>
</tr>
<tr>
<td>Elandsfontein</td>
<td>24.8</td>
<td>25.63</td>
<td>0.79</td>
<td>3.09</td>
</tr>
<tr>
<td>Kendal2</td>
<td>198.7</td>
<td>199.35</td>
<td>0.64</td>
<td>0.32</td>
</tr>
<tr>
<td>Leandra</td>
<td>20.9</td>
<td>21.97</td>
<td>1.04</td>
<td>4.74</td>
</tr>
<tr>
<td>Majuba1</td>
<td>22.5</td>
<td>24.00</td>
<td>1.53</td>
<td>6.37</td>
</tr>
<tr>
<td>Palmer</td>
<td>3.8</td>
<td>3.95</td>
<td>0.16</td>
<td>4.04</td>
</tr>
<tr>
<td>Amersfoort</td>
<td>17.2</td>
<td>17.84</td>
<td>1.73</td>
<td>9.68</td>
</tr>
</tbody>
</table>

4.3 Waterberg Biogenic NO\(_x\) emissions

Biogenic emissions are only 2.3 % of total NO\(_x\) emissions for the Waterberg area. The modelling domain is much smaller than the domain used for the Highveld. Rainfall for this area is also much less compared to the Highveld.

The main NO\(_x\) emissions, in total comprising 98 % of emissions, are from Matimba and Medupi from the power stations. Because of these large NO\(_x\) emissions from these power station it means the biogenic NO\(_x\) will have very little effect on Nitrogen deposition within the area. The NO\(_x\) emissions for this area are summarised in Table 4.12.

Table 4.13 Total NO\(_x\) emissions for the Waterberg

<table>
<thead>
<tr>
<th>Source</th>
<th>NO(_x) (tpa)</th>
<th>% NO(_x)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Power Generation</td>
<td>159630</td>
<td>97.65</td>
</tr>
<tr>
<td>Household Burning</td>
<td>0.63</td>
<td>0.00039</td>
</tr>
<tr>
<td>Tailpipe Emissions</td>
<td>115</td>
<td>0.07035</td>
</tr>
<tr>
<td>Biogenic Emissions</td>
<td>3721</td>
<td>2.28</td>
</tr>
<tr>
<td>Total</td>
<td>163466.63</td>
<td></td>
</tr>
</tbody>
</table>
The different rainfall patterns suggest that the highest NO\textsubscript{X} emission rates and emissions amounts occur during the summer months when soil moisture conditions are conducive to pulse events. The lowest emissions occur during winter when the soil moisture remains constant. The monthly emission rates and NO\textsubscript{X} amounts are shown Figures 4.4.

![Total Biogenic NO Waterberg Area](image)

**Figure 4.4 Monthly total NO emissions (2006)**

### 4.4 Predicted Highveld Sulphur and Nitrogen Deposition

#### 4.4.1 Spatial Highveld Deposition Model Results

Spatial plots for 2001 (average rainfall), 2010 (above-average rainfall) and 2003 (below average) were made and is presented in Appendix D Spatial Predictions. These results include the biogenic NO\textsubscript{X} emissions from the previous section. The following plots are given:

- Annual average SO\textsubscript{2}, SO\textsubscript{4}, NO\textsubscript{2} and NO\textsubscript{3} concentrations.
- Annual total, wet and dry S deposition.
- Annual total, wet and dry N deposition
4.4.1.1 Average Rainfall Base Case Spatial Results

Annual average ambient SO$_2$ concentration peaks on the central Highveld (up to 15 µg/m$^3$ between Secunda, Bethal and Emalahleni), with smaller isolated areas of up to 30 µg/m$^3$ scattered across within this region. This is because most of the coal-fired power stations (Matla, Kriel, Duvha and Kendal) are located within this area with in addition major factories such as Sasol 2, Sasol 3 and Highveld Steel. A secondary peak is observed over the Vaal Triangle (up to 15 µg m$^{-3}$) due to emissions from Mittal Vanderbijl Works, Sasol 1, Lethabo PS and Natref.

Annual average ambient SO$_4$ concentrations (1.5 µg m$^{-3}$) are spatially offset from the SO$_2$ dispersion described above. This is due to the secondary nature of SO$_4$. A highly localised peak was predicted to the west of Emalahleni of up to and exceeding 2 µg m$^{-3}$.

Annual average ambient NO$_2$ concentrations over the central Highveld were predicted up to 3 µg m$^{-3}$ with scattered locations of up 5 µg m$^{-3}$ and higher. Concentrations up to 6 µg m$^{-3}$ were predicted to be centred over the Vaal Triangle, with lower NO$_2$ concentration of 3 µg m$^{-3}$ stretching from the Vaal Triangle to the south Gauteng metropolitan areas and a small area located close to the Tshwane metropolitan area, which is due to emissions from motor vehicles and household fires.

Annual average ambient NO$_3$ peak values were predicted to be spatially offset due the secondary nature of the pollutant. A 5 µg m$^{-3}$ isopleth stretches between Emalahleni and the Vaal Triangle.

Total annual sulphur deposition peaks (20 kg ha$^{-1}$yr$^{-1}$) occur over the central Highveld between Emalahleni and Standerton and Ermelo, with scattered areas of up to 35 kg ha$^{-1}$yr$^{-1}$ close to Bethal and Secunda. A second elevated area (between 10 kg ha$^{-1}$yr$^{-1}$ and 20 kg ha$^{-1}$yr$^{-1}$) east of the Vaal triangle was also predicted. Most of
the Highveld was predicted to receive between 5 kg ha\(^{-1}\) yr\(^{-1}\) and 10 kg ha\(^{-1}\) yr\(^{-1}\) sulphur.

Total annual wet S deposition had a very similar pattern to the total S deposition described above. Wet S deposition was predicted to range between 20 kg ha\(^{-1}\) yr\(^{-1}\) and 30 kg ha\(^{-1}\) yr\(^{-1}\) close to Bethal and Secunda. A very localised area of wet S (>10 kg ha\(^{-1}\) yr\(^{-1}\)) was predicted over the Vaal triangle. Total annual dry S deposition peaked just west of Emalahleni (> 10 kg ha\(^{-1}\) yr\(^{-1}\)) with lower peak over the Vaal triangle (> 5 kg ha\(^{-1}\) yr\(^{-1}\)).

Total annual nitrogen deposition peaks (> 10 kg ha\(^{-1}\) yr\(^{-1}\)) just west of the central Highveld, but stretches from Emalahleni to west of Standerton. Highly localised total N deposition is observed just south of the Vaal Triangle (> 10 kg ha\(^{-1}\) yr\(^{-1}\)). As is the case for S deposition, total wet N deposition follows a similar spatial deposition pattern to the total N deposition due to large part wet N deposition makes up of the total N deposited on the Highveld. Wet N peaks from 6 kg ha\(^{-1}\) yr\(^{-1}\) to 9 kg/ha/annum just west of the central Highveld. In this case, no second peak is predicted over the Vaal triangle. Total dry N deposition (up to 5 kg ha\(^{-1}\) yr\(^{-1}\)) is centred on the Vaal Triangle and an area stretching between Emalahleni, Secunda and Johannesburg with isopleths ranging from 3 kg ha\(^{-1}\) yr\(^{-1}\) to 5 kg ha\(^{-1}\) yr\(^{-1}\).

4.4.1.2 Above Average Rainfall Year (2010) Spatial Results

The general spatial patterns predicted for 2010 appear very similar to those for 2001, with nitrogen and sulphur deposition peaking over the Highveld and the Vaal Triangle, but with higher and a different spatial rainfall patterns there are some noticeable differences.

For total S deposition, larger areas were predicted within the 10 and 20 kg ha\(^{-1}\) yr\(^{-1}\) isopleths. The 10 kg ha\(^{-1}\) yr\(^{-1}\) isopleth was predicted to be larger for dry S deposition,
and stretches from Emalahleni to just outside Bethal. For wet S deposition, the 20 kg ha\(^{-1}\)yr\(^{-1}\) isopleth was predicted to be larger and covers an area from Emalahleni to close to Standerton.

All N spatial deposition plots (total, wet and dry) showed a third area to the east where N deposition peaked. This area was just outside Carolina and were most likely due to emissions from the Hendrina and Arnot coal-fired power stations. Total N deposition was predicted to have an 8 kg ha\(^{-1}\)yr\(^{-1}\) isopleth similar in size to that predicted in 2001, but this one encompasses both Bethal and Secunda. In this case the 15 kg ha\(^{-1}\)yr\(^{-1}\) isopleth could also be clearly identified, which was not the case for 2001. For the dry N deposition, it was predicted that the 6 kg ha\(^{-1}\)yr\(^{-1}\) isopleth covers a larger area than for 2001 and stretched from the central Highveld to Carolina.

### 4.4.1.3 Below Average Rainfall (2003) Year Spatial Results

For total S deposition, the 35 kg ha\(^{-1}\)yr\(^{-1}\) isopleth over the central Highveld was predicted to be much smaller than for 2001. Similarly, the wet S deposition 20 kg ha\(^{-1}\)yr\(^{-1}\) isopleth was predicted to be concentrated around isolated areas rather than covering one big area as was predicted for wet S in 2001. Rainfall for this period was lowest for this scenario, which would explain the smaller area these isopleth cover.

The 10 kg ha\(^{-1}\)yr\(^{-1}\) isopleth predicted for dry S deposition covers a larger area than for both 2001 and 2010 and stretches from Emalahleni to an area close to Bethal and Secunda. The reason for this could be that during a drier period less atmospheric SO\(_2\) is removed through wet removal processes, which leaves dry removal as the only pathway through which atmospheric SO\(_2\) is removed from the atmosphere. The annual ambient average SO\(_2\) concentration were predicted to have a very similar spatial pattern to the total dry S deposition plot.

The total nitrogen deposition isopleth predicted also covered smaller areas than the plots predicted for 2001 and 2010.
4.4.2 Deposition Rates at Discrete Receptor Locations

Rainfall predicted by MM5 data at each receptor location is shown in Table 3.7. The general trend is that the rainfall predicted for the above average year (wet) is slightly higher than that for the average rainfall year with the below average year (dry) the lowest. It should be noted that the years chosen were based on measured data supplied by the South African Weather Services. It has already been shown in Section 3 that the MM5 data over-predicts the rainfall for the Highveld. It is therefore expected that the deposition rates predicted by this model will be very different from what was predicted by Scorgie and Kornelius (2009).

Predicted deposition rates at receptor locations for wet, average and dry years are shown in Table 4.13, Table 4.14 and Table 4.15. Figure 4.6 to Figure 4.11 show the relation between each type of deposition rate for wet, average and dry years at each receptor location.

The receptor locations for the highest deposition rates are:

- Kendal 2 (198.7 kg S ha\(^{-1}\) yr\(^{-1}\); 33 kg N ha\(^{-1}\) yr\(^{-1}\)).
• Elandsfontein (24.8 kg S ha\(^{-1}\) yr\(^{-1}\); 6.6 kg N ha\(^{-1}\) yr\(^{-1}\)).
• Leandra (20.9 kg S ha\(^{-1}\) yr\(^{-1}\); 15.9 kg N ha\(^{-1}\) yr\(^{-1}\)).
• Makalu (12.8 kg S ha\(^{-1}\) yr\(^{-1}\); 8 kg N ha\(^{-1}\) yr\(^{-1}\)).
• Olifants headwater (29.9 kg S ha\(^{-1}\) yr\(^{-1}\); 7.8 kg N ha\(^{-1}\) yr\(^{-1}\)).

Deposition rates at upwind receptor locations (e.g. Palmer [3.8 kg S ha\(^{-1}\) yr\(^{-1}\); 2.7 kg N ha\(^{-1}\) yr\(^{-1}\)]) located at the north-eastern boundary of the modelling domain were the lowest due to their position.

Downwind receptor locations were predicted to have relatively high sulphur and nitrogen deposition values, but lower than those mentioned above because of the distance from the source region. The receptor locations are:

• Amersfoort (17.2 kg S ha\(^{-1}\) yr\(^{-1}\); 4 kg N ha\(^{-1}\) yr\(^{-1}\)).
• Verkykerskop (11 kg S ha\(^{-1}\) yr\(^{-1}\); 2.9 kg N ha\(^{-1}\) yr\(^{-1}\)).
• Sandspruit catchment (11.1 kg S ha\(^{-1}\) yr\(^{-1}\); 2.8 kg N ha\(^{-1}\) yr\(^{-1}\)).

Generally, higher total sulphur deposition rates were predicted for the wet year and lowest rates for the dry year at most receptor sites. Only at Camden the average rainfall year total S predicted is slightly higher. At Verkykerskop, Majuba, Kendal 2, Amersfoort and the Sandspruit headwater the average rainfall year predicted rates were lower than those for the dry year.

Wet sulphur showed a similar trend to the total sulphur with highest rates during the wet year and the lowest rates during the dry year. This is because the wet deposition accounted for most of the sulphur deposition and is strongly influenced by rainfall. Percentage wet S varies between 53 and 86 % of total S deposition for the average rainfall year. At Majuba 1 and Amersfoort the average year wet S deposition was below that of the dry year predicted deposition rate.

Dry S deposition for wet and dry years were predicted to have very similar rates at receptor locations, while dry S rates for the average rainfall year were generally
lower than both the wet and dry years. At the Olifants river headwater the dry S rate was predicted to be higher than both wet and average years.

Predicted total N deposition rates were predicted to be highest during the wet year. The average year has the second highest values followed by the dry year rates, although at Makalu the average year was predicted to be higher than for both the wet and dry years. Wet N deposition made up the bulk of the total N deposition, and has the same general pattern with the highest predicted rate during the wet year, followed by the average year and then the dry year. Dry N rates for wet, average and dry years were predicted to be very similar.

Figure 4.6 Total S deposition at receptor locations for the South African Highveld
Figure 4.7 Wet S deposition at receptor locations for the South African Highveld

Figure 4.8 Dry S deposition at receptor locations for the South African Highveld
Figure 4.9 Total N deposition at receptor locations for the South African Highveld

Figure 4.10 Wet N deposition at receptor locations for the South African Highveld
Figure 4.11 Dry N deposition at receptor locations for the South African Highveld
Table 4.14 Annual average concentrations and deposition rates at defined Receptor locations for the M4 model (2001)

<table>
<thead>
<tr>
<th>Receptor Site</th>
<th>Annual Average Ambient Air Concentrations (µg m(^{-3}))</th>
<th>Total Annual Deposition (kg ha(^{-1})yr(^{-1}))</th>
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Table 4.15 Annual average concentrations and average deposition at defined Receptor locations for the M4 model (2010)

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<th>Annual Average Ambient Air Concentrations (µg m(^{-3}))</th>
<th>Total Annual Deposition (kg ha(^{-1})yr(^{-1}))</th>
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### Table 4.16 Annual average concentrations and average deposition at defined Receptor locations for the M4 model (2003)

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<th>Total Annual Deposition (kg ha⁻¹yr⁻¹)</th>
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<td>Olifants headwater</td>
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5. CONCLUSION AND RECOMMENDATIONS

5.1 The South African Highveld

5.1.1 Construction of a comprehensive emission inventory and dispersion model of both the South African Highveld.

Sources quantified for this study include:

- Large coal-fired power stations.
- Major and minor industrial sources.
- Vehicle exhaust emissions.
- Household fires.

The model results of the new model (M2) were then compared to measured values and the original model build by Scorgie and Kornelius (2009) (M1) to assess whether the model could be used.

Spatial plots of M1 and M2 were compared. Both models show that higher concentration of pollutants are found close to major industries. These hotspots were located over the central Highveld and in the Vaal Triangle. Similar predicted deposition values for M1 and M2 at receptor locations. Generally lower ambient gas concentrations and higher deposition rates were predicted by M2.

Given the model limitations and after comparing deposition results from previous studies M2 will be suitable to study the effect biogenic NO\textsubscript{x} and H\textsubscript{2}S will have on deposition on the South African Highveld.

Model limitations should be considered.

- The MM5 data used to construct the CALMET files for all three years over predicted rainfall.
- Ammonia emission were not quantified as part as the emission inventory nor were they simulated. Because of this the model (M2) will under predicted dry N deposition. Dry deposition of ammonia has been estimated by Galy-Lacaux et al (2003) to account for roughly 30% of total N deposition.
• Emissions due to biomass burning were not included in the emission inventory.

5.1.2 Comprehensive biogenic NO\textsubscript{X} model for the South African Highveld area to study the effect biogenic NO\textsubscript{X} will any effect on the nitrogen emitted and deposition within these regions.

A comprehensive model was built using work previously done by Yienger and Levy (1995). The modelling domain was divided into 196 smaller 25 km × 25 km areas. Land use values, rainfall and temperature values for those areas were then used to calculate the NO\textsubscript{X} emission for each area for every hour for the entire modelling period. This was done all three modelling periods of 2001, 2010 and 2013.

The biogenic NO\textsubscript{X} emitted made up 3.96 %, 4.14 % and 3.34 % of total released NO\textsubscript{X} for 2001, 2010 and 2013 respectively. This is significantly more than is released by household fuel burning, small industrial sources and biomass burning.

Dry N deposition rates were the most affected by biogenic NO\textsubscript{X} emissions, with higher deposition rates predicted at all receptor locations. The highest NO\textsubscript{X} emissions were predicted during spring and autumn months. Dry nitrogen deposition rates have been affected the most. Biogenic NO\textsubscript{X} added between 1.69 - 6.19 %. Wet deposition rates were affected very little (0.13 % to 0.75 %). Total nitrogen deposition rates ranged from 0.32 % to 1.77 %.

Recommendations for further study:

• Fertilizer used in crops could have a significant effect biogenic NO\textsubscript{X} and NH\textsubscript{4}\textsuperscript{+} volatilization. Land use should therefore be more accurately mapped to distinguish more clearly between agricultural produce fields and grasslands.
• Fertilizer loads (timing and amount of fertilizer added by farmers) should be considered. Large areas of the Highveld are covered with crops. This would affect biogenic NO\textsubscript{X} emissions for the area.
• Accurate biogenic NO flux rates should be determined specifically for the grasslands on the South African Highveld, which would result in a more accurate model.
5.1.3 Quantifying \( \text{H}_2\text{S} \) sources on the Highveld and to find a way of including the \( \text{H}_2\text{S} \) emissions to the deposition model.

Due to the limitation that \( \text{H}_2\text{S} \) is not part of the RIVAD chemical suite of CALPUFF the following assumption was made:

- \( \text{H}_2\text{S} \) emission sources will be inputted into CALPUFF as \( \text{SO}_2 \) sources.
- To emulate the different seasonal atmospheric patterns measured by Igbafe (2007) at Elandsfontein the following conversion rates are proposed and were used to adapt the \( \text{SO}_2 \) emission rates inputted into CALPUFF:
  - Summer 70% conversion.
  - Autumn 60% conversion.
  - Winter 50% conversion.
  - Spring 40% conversion.

The model was run only for 2001 and the following contributions on sulphur deposition was predicted at the receptor locations shown in tables 4.10, 4.11 and 4.12:

- An average of 4.85% was contributed to wet Sulphur deposition rates.
- An average of 5.95% was contributed to dry Sulphur deposition rates.
- An average of 5.15% was contributed to the total Sulphur deposition rates.

This shows that \( \text{H}_2\text{S} \) emission on the South African Highveld do need to be considered in future deposition studies. Recommendations for further study are:

- A more detailed emission inventory should be drawn up that include other industrial point sources and area sources which include mainly petrochemical factories, coking operations at iron steel works and open cast coal mines (Scorgie and Kornelius, 2009).
- Accurate study of atmospheric \( \text{H}_2\text{S} \) conversion rates to \( \text{SO}_2 \) on the South African Highveld.
- The CALPUFF RIVAD chemical reaction scheme should be modified to include the conversion of \( \text{H}_2\text{S} \) to \( \text{SO}_2 \), which will lead a more accurately understanding the effect of \( \text{H}_2\text{S} \) emissions on the Highveld.
5.1.4 Highveld sulphur and nitrogen dispersion and deposition

A comprehensive analysis of the results generated by the M2 model which included the biogenic emissions calculated for the South African Highveld in section 3.8 was done. This was done for the three modelling periods (2001, 2010 and 2003) each representing the different rainfall years specified above. Predicted dispersion of SO₂, SO₄, NO₂ and NO₃ gases was plotted for the area. Predicted dry, wet and total N and S depositions rates were also plotted for the area. Predicted ambient values of the abovementioned gases was reported at specified receptor locations. This was also done for N and S deposition rates at these receptor locations.

5.2 The Waterberg Area

5.2.1 Construction of a comprehensive emission inventory and dispersion model of the South African Waterberg area.

Sources quantified for this study include:

- Large coal-fired power stations.
- Vehicle exhaust emissions.
- Household fires.

Vehicle exhaust emissions and household fires contribute very little to both sulphur and nitrogen emissions and was excluded in the model. Power generation contribute 98% of nitrogen emissions and 99% of sulphur emissions for the Waterberg priority area. Because Medupi is still in process of being constructed, the emission rates used for the model are from this interim period when it will run without the required flue gas desulphurisation (FGD) to regulate SO₂ emissions prescribed by the Minimum Emission Standards.
5.2.2 Comprehensive biogenic NO\textsubscript{x} model for the South African Waterberg area to study the effect of biogenic Nitrogen emitted within this region.

A comprehensive model was build using work previously done by Yienger and Levy (1995). The modelling domain was divided into 25 areas, which consisted of 475 to 520 1 km \times 1 km cells. Land use values, rainfall and temperature values for those areas were then used to calculate the NO\textsubscript{x} emission for each area for every hour for the entire modelling period. This was done for the modelling period of 2006.

Biogenic emissions are only 2.3 % of total NO\textsubscript{x} emissions for the Waterberg area. Because biogenic emissions were so low compared to NOx emission from both the power stations, it was decided that not to run a dispersion model for the Waterberg area. The effect on nitrogen dispersion and deposition will be minimal.

Model limitations:

- Rainfall used to predict NO\textsubscript{x} represents only a single point and did not consider the spatial rainfall pattern for this the area.
- Canopy reduction was not considered.

Recommendations for further study:

- Fertilizer used in crops could have a significant effect biogenic NO\textsubscript{x} and NH\textsubscript{4}\textsuperscript{+} volatilization. Land use should therefore be more accurately mapped to distinguish more clearly between agricultural produce fields and grasslands.
- Fertilizer loads (timing and amount of fertilizer added by farmers) should be considered.
6. REFERENCES


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Scire, JS, Strimaitis, DG and Yamartino, RJ (2000) *A user’s guide for the CALPUFF


## 7. APPENDIX A INDUSTRIAL EMISSION SOURCES

### Table 7.1 Major industrial and Large Coal-fired Power Station point sources input parameters for 2001

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<th>Stack Diameter (m)</th>
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© University of Pretoria
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8. APPENDIX B LANDUSE FOR THE HIGHVELD AND WATERBERG

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© University of Pretoria
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Table 8.2 Waterberg land use values defined for each cell

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9. APPENDIX C MODEL VERIFICATION: SPATIAL AND RECEPTOR COMPARISON

9.1 Average Rainfall year (2001)

The original model (M1) plot by Scorgie and Kornelius is on the left and the current model’s (M2) plot is on the right.
Predicted Annual Average SO4 Concentration (µg/m³) - Average Rainfall Year (2000/1)

Predicted Annual Average SO4 Concentration (µg/m³)
Average Rainfall year 2001

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Table 9.1 Annual average concentrations at defined receptor locations for the original model (M1 {2000/1}) and the current model (M2 {2001})

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<tr>
<th>Receptor Site</th>
<th>SO₂ (M1)</th>
<th>SO₂ (M2)</th>
<th>SO₄ (M1)</th>
<th>SO₄ (M2)</th>
<th>NO (M1)</th>
<th>NO (M2)</th>
<th>NO₂ (M1)</th>
<th>NO₂ (M2)</th>
<th>NO₃ (M1)</th>
<th>NO₃ (M2)</th>
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Table 9.2 Annual average deposition rates at defined receptor locations for the original model (M1 {2000/1}) and the current model (M2 {2001})

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<th>Receptor Site</th>
<th>Total Annual Deposition (kg/ha/year)</th>
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<tr>
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<td>Leandra</td>
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<td>Majuba 3</td>
<td>6.7</td>
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<td>Palmer</td>
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<tr>
<td>Komati headwater</td>
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<td>Olfants headwater</td>
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<tr>
<td>Sandspruit headwater</td>
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9.2 Above Average Rainfall Year (2010)

Annual Average SO2 Concentrations (µg/m³)
- Above-Average Rainfall Year (1995/6)

Predicted Annual Average SO2 (µg/m³)
Below Average Rainfall Year 2010

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Predicted Annual Average NO\textsubscript{2} Concentration (µg/m\textsuperscript{3})
Above Average Rainfall year 2010

© University of Pretoria
Predicted Annual Average NO$_3$ Concentrations ($\mu$g/m$^3$)
Above Average Rainfall Year 2010

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Table 9.3 Annual average concentrations at defined receptor locations for the original model (M1 {2010}) and the current model (M2 {1995/6})

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<th>Receptor Site</th>
<th>Annual Average Ambient Air Concentrations (µg/m³)</th>
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<tr>
<td>Majuba 1</td>
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<td>Sandspruit headwater</td>
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</table>
Table 9.4 Annual average deposition rates at defined receptor locations for the original model (M1 {2010}) and the current model (M2 {1995/6})

<table>
<thead>
<tr>
<th>Receptor Site</th>
<th>Wet S (M1)</th>
<th>Wet S (M2)</th>
<th>Dry S (M1)</th>
<th>Dry S (M2)</th>
<th>Total S (M1)</th>
<th>Total S (M2)</th>
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</tbody>
</table>
9.3 Below Average Rainfall Year (2003)

Predicted Annual Average SO$_2$ (µg/m$^3$) - Below-Average Rainfall Year (2006/7)

Below Average Rainfall year 2003

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Predicted Annual Average SO₄ Concentrations (µg/m²)
- Below-Average Rainfall Year (2006/7)

Below Average Rainfall year 2003
Predicted Annual Average NO₂ Concentrations (µg/m³)
Below Average Rainfall Year 2003

© University of Pretoria
Predicted Annual Average NO\textsubscript{3} Concentrations (µg/m\textsuperscript{3})
- Below-Average Rainfall Year (2006/7)

Below Average Rainfall Year 2003

© University of Pretoria
Predicted Annual Wet Sulphur Deposition (kg/ha/annum)
- Below-Average Rainfall Year (2006/7)

Predicted Total Annual Wet Sulphur Deposition (kg/ha/annum)
Below Average Rainfall Year 2003

© University of Pretoria
Table 9.5 Annual average concentrations at defined receptor locations for the original model (M1 {2006/7}) and the current model (M2 {2003})

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<tr>
<th>Receptor Site</th>
<th>Annual Average Ambient Air Concentrations (µg/m³)</th>
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<td>Palmer</td>
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<td>Camden</td>
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<td>Amersfoort</td>
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Table 9.6 Annual average deposition rates at defined Receptor locations for the original model (M1 {2006/7}) and the current model (M2 {2003})

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<th>Dry S (M2)</th>
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<th>Total S (M2)</th>
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10. APPENDIX D SPATIAL PREDICTIONS FOR THE SOUTH AFRICAN HIGHVELD

10.1 Spatial Predictions Average Rainfall Year (2001)

Predicted Annual Average SO₂ (µg/m³) Average Rainfall year 2001

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10.2 Spatial Predictions Above Average Rainfall Year (2010)
10.3 Spatial Predictions Below Average Rainfall Year (2003)

Predicted Annual Average SO$_2$ (μg/m$^3$)
Average Rainfall year 2003

Predicted Annual Average SO$_4$ (μg/m$^3$)
Below Average Rainfall year 2003