DSPACE

https://dspace.org/

Comparison of air pollution hotspots in the Highveld using airborne data

Ncipha, Xolile Gerald

2011-06-22
http://hdl.handle.net/10539/10166

Downloaded from a DSpace repository. https://dspace.org/

Chapter 1

Overview

Land cover and land use over the Highveld air pollution hotspots which influence air quality is described. Air pollution problems and challenges in air quality management over the Highveld region are mentioned. A discussion of air pollution sources and their emissions constituents over the Highveld region is provided. The characteristics, sources, sinks, diurnal and seasonal variation of aerosols and trace gases under investigation are presented in detail. The influence of meteorology on air pollution levels is discussed. The research goals are also outlined.

Introduction

The Highveld region is an eco-region that lies at the centre of the high interior plateau of South Africa. Its name is derived from it being part of this high lying flat terrain and its wide coverage of grassland species. The surface altitude of the Highveld is varying from approximately 1200 masl to 1800 masl. The Highveld extends from the Western Cape Province to the northeast, covering the whole Free State Province. In the east it is bordered by the Drakensburg, in the west by the arid Karoo and Kalahari, and the low lying bushveld to the north (Coutsoukis, 1996; World Wildlife Fund (WWF), 2001).

The Highveld developed to be an important agricultural and industrial region because of its richness in natural resources (Terblanche *et al.,* 2000; Gauteng Department of Agriculture, Conservation and Environment (GDACE), 2004). The region is a major coal producer and the world's third largest exporter of coal (Coakley, 2000). Its abundance of coal and other mineral resources led it to develop into a major industrial and urban population centre of South Africa (Terblanche *et al*., 2000; North West Department of Agriculture, Conservation and Environment (NWDACE), 2002; GDACE, 2004, Mpumalanga Department of Agriculture and Land Administration (MDALA), 2005b). About 75% of industrial infrastructure in South Africa is based on the Highveld (Freiman and Piketh, 2003). Industries found in this region include power generation plants, refineries, chemical, mining, metallurgical, mineral

processing, agriculture and utility industries (Mpumalanga Department of Agriculture, Conservation and Environment (MDACE), 2003; Coakley, 2000; NWDACE, 2002; MDALA), 2005a; Vaal Triangle Info (VTI), 2005; Save the Vaal Environment (SAVE), 2006).

The social and economic activities in this region exert pressure on the environment, especially the atmosphere. Continued growth in the industrial/manufacturing sector led to increased emissions of air pollutants (GDACE, 2004). Industries such as petrochemical, metallurgical, manufacturing and power generation burn most of South African coal, releasing SO_2 and NO_2 (van Tienhoven,1999), the precursors of fine sulphates and nitrates particles (Held *et al*., 1996). These oxides of nitrogen and sulphur are deposited through dry and wet deposition as acid rain (GDACE, 2004). The industrialised Highveld region contributes approximately 90% of South Africa's scheduled emissions of industrial dust, SO_2 and NO_2 (Freiman and Piketh, 2003). The eastern Highveld is also known as a sulphate particles reservoir (Scheifinger, 1992). This is because electricity generation in South Africa, a process emitting SO_2, is predominantly occurring in the Mpumalanga Province (MDACE, 2003). The emission density of SO_2 from Highveld industries are said to be comparable to that encountered in other large industrial areas of the world (Annnegarn *et al*., 1996a; van Tiehoven, 1999). Wind-blown dust from unrehabilitated mine tailing dumps are of more concern if they are located close to residential places (NWDACE, 2002). In Sasolburg waste coal ash and mine tailings dumps are located near Zamdela a residential township. The coal ash and dust is frequently blown over the Sasolburg town (GroundWork, 2003). Domestic fossil fuel burning is also an important source of SO_2 and NO. The increase in dependence on private vehicles and small public transport unit (minibus taxis) in this region adds to the air pollution problems associated with fossil fuel burning (GDACE, 2004).

The most common natural air pollution source in the Highveld region is veld fires. They occur often in the dry season, autumn, winter and early spring (Eck *et al*., 2003; Crutzen *et al*., 1999; GDACE, 2004). The meteorological conditions associated with veld fires are warm, dry day with strong winds (de Villiers, 2000).

Despite the poor air quality over the Highveld, until recently, the geographical and spatial coverage of air quality data is poor. Air quality monitoring sites are predominantly located in urban areas where the health impacts of air pollution are of main concern (GDACE, 2004). Some local municipalities and industries continuously monitor air quality. However, industries locate their monitoring stations to monitor the effects of their specific activities, rather than to be representative of the air quality of the area. Their monitoring stations distributions normally don't allow determination of the spatial extent of the air pollutants. Air quality monitoring is also performed by some organisations through uncoordinated ad hoc projects. In general it is difficult to obtain a complete spatial picture of ambient air quality in this region because of the isolated and uncoordinated monitoring activities and the lack of sharing of air quality data amongst involved organisations (MDACE, 2003).

An airborne platform has an advantage of providing a complete spatial picture of ambient air quality. Aircraft measurements offer accurate 3-D data over a large area in fine spatial resolution (National Aeronautics and Space Administration (NASA), 1994). They can fill the spatial data gaps created by biased uneven distribution of ground-based air quality monitors (MDACE, 2003). The little restriction in aircraft mobility enables the determination of the horizontal extent of air pollutants. It also has an advantage of measuring air pollution aloft which can influence ground level concentrations of air pollution by being mixed down to the surface (Department of California Environmental Protection Agency (DCEPA), 2006). The purpose of this study is to compare air pollution hotspots over the Highveld region using airborne data of SO_2, NO_X, O_3 and $PM_{2.5}$ aerosols. The air pollution hotspots that are investigated in this study are the Witbank, Secunda, Rustenburg and the Vaal Triangle area, including their surrounding towns. The seasonal variation of air pollution in these hotspots will be compared. This study will also investigate the relationship between airborne and ground based air quality data.

Literature Review

Study areas

Witbank

Witbank is one of the major towns in the Emalahleni area, located in the western part of Mpumalanga on the Highveld. It falls under the Nkangala District Municipality. Emalahleni occupies 2670 km^2 and is mainly grassland, with patches of savanna in the north. The elevation of this area varies from 1301 m to 1700 m. The land cover varies between pristine grassland (with the highest land cover percentage), cultivated commercial dry land and irrigated land, forest plantations, mines and quarries, urban commercial, residential, and industrial land, and a small percentage of water bodies and wet lands. The area is called Emalahleni (coal place) because of its rich coal reserves around Witbank. As a result, coal related industries can be found in the vicinity of Witbank. The most noticeable are four coal-fired power stations, Kendal, Matla, Kriel and Duvha (MDALA, 2005a). These power stations together with the other Mpumalanga coal-fired power stations generate almost 70% of the total electricity generated in South Africa. The other economic activities include mining, agriculture, forestry and manufacturing (MDACE, 2003). Figure 1.1 shows the land use over Witbank area, a detailed description of land use classes are given by Piketh *et al*., 2006.

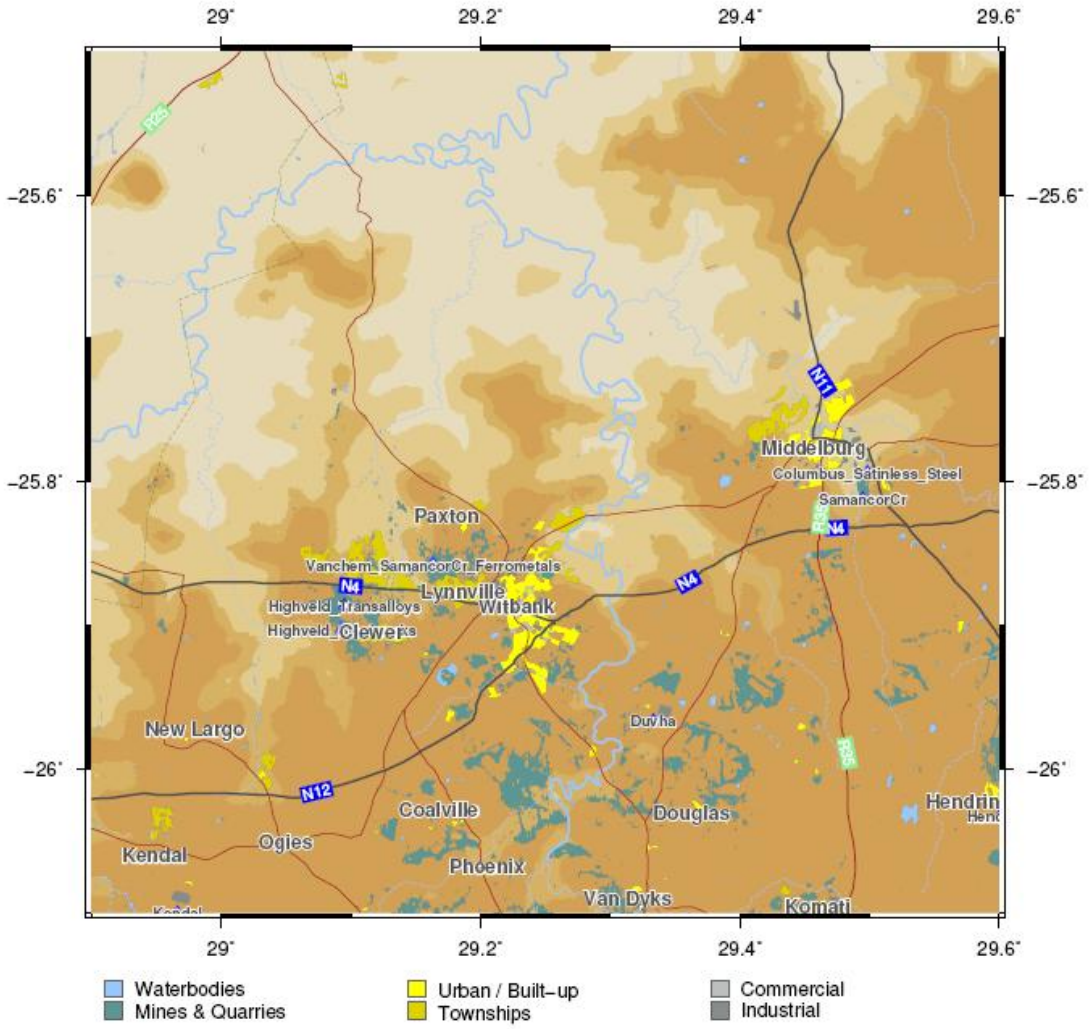


Figure 1.1: Land use over Witbank area (Piketh *et al*., 2006).

Secunda

Secunda is one of the major towns in the Goven Mbeki (Highveld East) area, located in the western part of Mpumalanga, south of Witbank (MDALA, 2005b; MDACE, 2003). It falls under the Gert Sibande District Municipality. The area occupies 2960 km^2 and is mainly grassland. The elevation of this area varies from 1501 m to 1700 m. The land cover varies from pristine grassland (with the highest land cover percentage), cultivated commercial dry land and irrigated land, forest plantations, mines and quarries, urban commercial, residential and industrial land, and a small land cover percentage of water bodies and wet lands. The area has coal and gold deposits. This resulted the development of coal related industries in this area (MDALA, 2005b). The most notable coal dependent industry is the SASOL coal liquefaction plant in Secunda. The plant converts coal into oil and other related

products. It is one of the highly developed synthetic fuel industry in South Africa (MDALA, 2005b; Coakley, 2000). Figure 1.2 shows the land use over Secunda area, a detailed description of land use classes are given by Piketh *et al*., 2006.

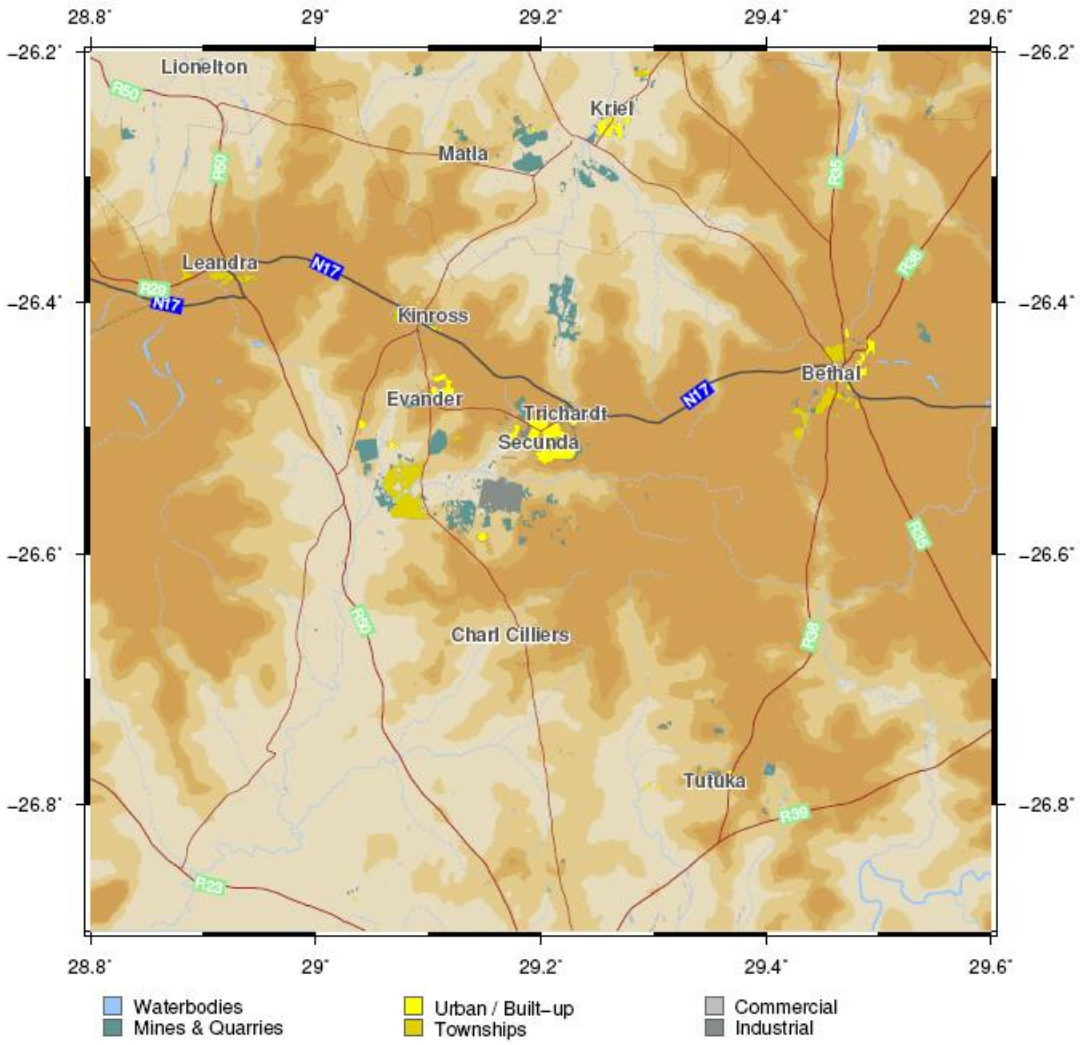


Figure 1.2: Land use over Secunda area (Piketh *et al*., 2006).

Rustenburg

Rustenburg is a big town located in the north-eastern region of the North West province (NWDACE, 2002). It falls under the Bojanala Platinum District Municipality (Bojanala Platinum District Municipality (BPDM), 2005). The natural land cover of the Rustenburg area is savanna and some different types of bushvelds (NWDACE, 2002). Mining in Rustenburg is the most important economic activity. The area houses two of the biggest platinum mines in the world (Pro-Poor Tourism (PPT), 2006; North West Parks and Tourism Board (NWPTB), 2006) and produces

70% of the world's platinum (NWDACE, 2002). Mining of other minerals such as chrome, palladium, nickel and copper also occurs in this area (PPT, 2006; NWPTB, 2006). The mining of these minerals brought about the development of mineral processing industries. Agriculture takes the second position as an important economic activity in the area (NWDACE, 2002). Figure 1.3 shows the land use over Rustenburg area, a detailed description of land use classes are given by Piketh *et al*., 2006.

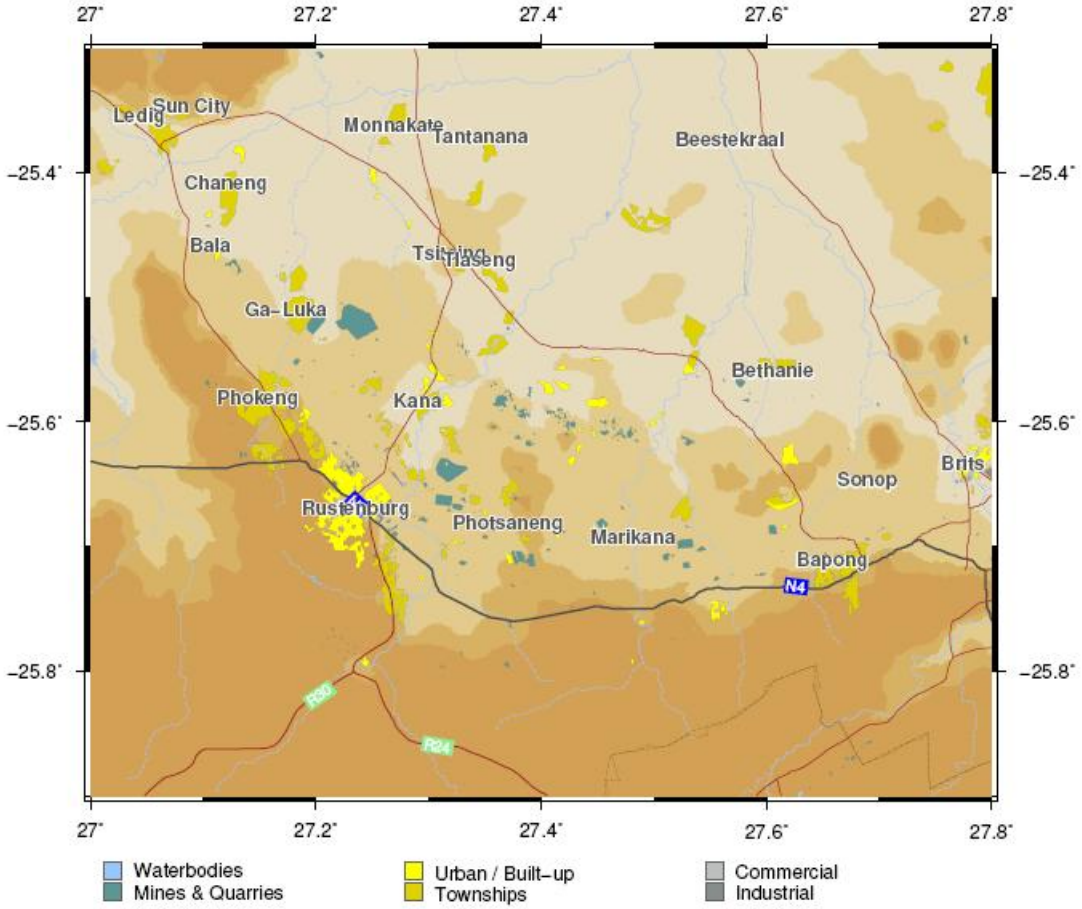


Figure 1.3: Land use over Rustenburg area (Piketh *et al*., 2006).

The Vaal Triangle

The Vaal Triangle is made up of three towns, Vereeniging, Vanderbijlpark and Sasolburg. These towns are located approximately 15 km from each other, and positioned in triangular shape relative to each other (van Tienhoven, 1999). The area was established as an industrial area because of the proximity of water reserves from the Vaal River and coal reserves and it is mainly grassland (WWF, 2001; PPT, 2004; VTI, 2005). The Vaal Triangle is known as an industrial heartland of South Africa

(WWF, 2001; VTI, 2005). The industries that can be found in this area include a power plant, a refinery, chemical, metallurgic, agriculture, mining and utility industries (VTI, 2005; SAVE, 2006). The most notable industries in this area are the Lethabo power plant, the giant steel producer Mittal Steel, and the SASOL's synthetic petrochemical plant (VTI, 2005). Figure 1.4 shows the land use over Vaal Triangle area, a detailed description of land use classes are given by Piketh *et al*., 2006.

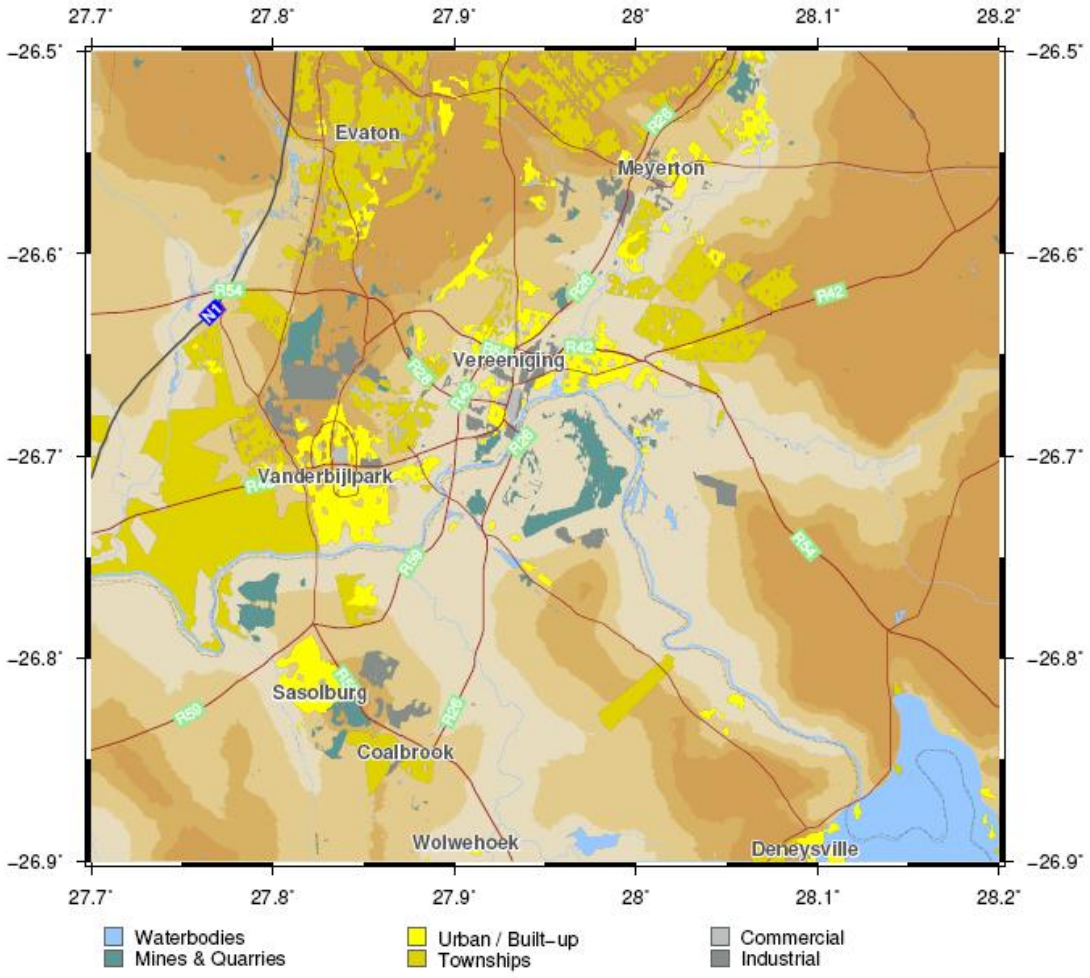


Figure 1.4: Land use over the Vaal Triangle area (Piketh *et al*., 2006).

Air pollution sources and their emission constituents over the Highveld region

Emissions from manufacturing industries include SO_2, NO_X, carbon dioxide (CO_2), carbon monoxide (CO), volatile organic compounds (VOC's) and aerosols (NWDACE, 2002, GDACE, 2004). Mining and mineral processing industries release aerosols, SO_2, NO_X, CO_2, and CO into the atmosphere (NWDACE, 2002). Unrehabilitated mine tailing dumps are a source of wind-blown dust, which may contain radioactive elements (NWDACE, 2002; GDACE, 2004). Agricultural activities release methane (CH_4) and other agrochemicals into the atmosphere (NWDACE, 2002). Veld fires for agricultural field clearance and management also contributes to anthropogenic air pollution in this region (GDACE, 2004). The transport sector is also an important source of air pollutants (NWDACE, 2002; GDACE, 2004). Vehicle exhaust emissions include SO_2, NO_X, CO_2, CO, VOC's and aerosols. They also included lead (Pb) from the leaded fuel, before the leaded fuel production and consumption was prohibited (NWDACE, 2002; GDACE, 2004). Heavy vehicles also cause dust from road surfaces that are not tarred (NWDACE, 2002). Another important source of anthropogenic air pollutants is the domestic consumption of fossil fuel as a source of energy in townships and rural areas, especially in winter. Emissions from fossil fuel burning include aerosols, SO_2, NO, CO_2, CO, VOC's (van Horen *et al*., 1996; Wells *et al*., 1996; Spalding-Fetcher *et al*., 2000; Collins, 2001; NWDACE, 2002; MDACE, 2003; GDACE, 2004).

The notable natural sources of air pollution in this region are the wild veld fires and wind-blown dust (GDACE, 2004). Vegetation fires are globally important source of gases and aerosols (Bertschi *et al*., 2003, Eck *et al*., 2003; Crutzen *et al*., 1999). The emissions from these biomass fires include NO_X, VOC's, CO, CO_2 and aerosols (Bertschi *et al*., 2003; Hobbs *et al*., 2003). Wind-blown dust from uncovered ground is also a source of primary aerosols in this region (Held *et al*., 1996).

Characteristics of the monitored air pollutants

Ozone

Ozone is a colourless, highly reactive gas with a distinctive odour. It is consists of three oxygen atoms and is a principal component of smog at the surface. About 90% of it occurs in the stratosphere and the remaining 10% in the troposphere. Stratospheric O_3 protects life on earth against harmful ultraviolet (UV) radiation (Crutzen *et al*., 1999; Wisconsin Department of Natural Resources (WDNR), 2006; World Meteorological Organisation (WMO), 2006b). In the troposphere O_3 is a greenhouse gas, it absorbs terrestrial infrared light. (Doddridge *et al*., 1992; Frost *et al*., 1998, Intergovernmental Panel of Climate Change (IPCC), 2001; Betts *et al*., 2002; Fiore *et al*., 2002; WMO, 2006b). O_3 has an atmospheric lifetime of approximately 28 days (Seinfeld and Pandis, 2006).

Sources of tropospheric ozone

The sources of O_3 in the troposphere are stratospheric O_3 intrusion and photochemical production from a series of reactions involving photochemical oxidation of hydrocarbons and CO in the presence of NO_X and sunlight (Jacobs *et al*., 1995; IPCC, 2001; Lefohn *et al*., 2001; Fiore *et al*., 2002; Kleinman *et al*., 2002; Hobbs *et al*., 2003; Taubman *et al*., 2004; Huixiang *et al*., 2005; WMO, 2006b; WDNR, 2006). The stratospheric O_3 intrusion into the troposphere is caused by well developed deep low pressure troughs or cut off low (COL) pressure system. Both these synoptic features can extend vertically into the stratosphere, disturbing the tropopause and creating a passage for energy and air mass transfer between the troposphere and stratosphere and vice versa (Fishman and Crutzen, 1978; Garstang *et al*., 1996; Combrink *et al*., 1998; Freiman and Tyson, 2000; Diab *et al*., 2004). Modeling results from the Model of Atmospheric Transport and Chemistry (MATCH) showed that photochemical generation of tropospheric O_3 generally dominates the stratospheric O_3 contribution to the O_3 levels in the troposphere (Crutzen *et al*., 1999). The production of O_3 in the troposphere is induced by solar ultraviolet (UV) radiation of wavelength shorter than about 340 nm. The photons of this UV light break the O_3 molecules into oxygen (O_2) molecules and electronically excited oxygen (O) atoms (Crutzen *et al.,* 1999; Taubman *et al.,* 2004).

$O_3 + h\nu \rightarrow O(^1D) + O_2$ ($\lambda < 340$ nm) .....(1.1)

The majority of the high energy O (‘D) atoms collide with O_2 molecules and get deactivated to ground energy state and form O_3 molecules:

$O + O_2 + M \rightarrow O_3 + M$ .....(1.2)

The remaining excited O (‘D) atoms have sufficient energy to react with water vapour to produce hydroxyl (OH) radicals:

$O(^1D) + H_2O \rightarrow 2OH$ .....(1.3)

The OH radicals can initiate reactions leading to O_3 production through the following reaction pathway (Crutzen *et al*., 1999; Taubman *et al*., 2004):

$CO + OH (+O_2) \rightarrow CO_2 + HO_2$ .....(1.4)

The peroxy radical (HO_2) can then react with nitric oxide (NO) to form nitrogen dioxide (NO_2).

$HO_2 + NO \rightarrow OH + NO_2$ .....(1.5)

NO_2 rapidly photolysis, followed by O_3 formation.

$NO_2 + h\nu \rightarrow NO + O$ .....(1.6)

$O + O_2 + M \rightarrow O_3 + M$ .....(1.7)

Sinks for tropospheric ozone

The OH radical and NO can both act as a sink for surface O_3. The OH radical can initiate reactions leading to surface O_3 destruction if there is insufficient NO to be oxidised to form NO_2 (Poulida *et al*., 1994; Sillman and Samson, 1995; Crutzen *et al*., 1999; Taubman *et al*., 2004).

$CO + OH (+O_2) \rightarrow CO_2 + HO_2$ .....(1.8)

$HO_2 + O_3 \rightarrow OH + O_2$ (O_3 destruction reaction) .....(1.9)

Surface O_3 may react with NO in the absence of, or insufficient OH radicals to regenerate NO_2 (Kley *et al*., 1994; Poulida *et al*., 1994; Hobbs *et al*., 2003, Taubman *et al*., 2004).

$O_3 + NO \rightarrow O_2 + NO_2$ (O_3 destruction reaction) .....(1.10)

Tropospheric O_3 can also be lost through liquid-phase reactions and dry deposition (Ryerson *et al*., 1998).

Tropospheric ozone diurnal variation

Surface O_3 concentration varies diurnally. In the morning O_3 increases after the break-up of the nocturnal low-level inversion layer as the air aloft is mixed downward. During the day the sunlight intensity at the surface increases resulting in the increase of photochemical production of O_3. In the afternoon when the solar radiation intensity is highest, O_3 concentration is at its peak. In the absence of sunlight at night NO_2 cannot be photolysed to generate surface O_3 (Trainer *et al*., 1987; Poulida *et al*., 1994; Annegarn *et al*., 1996a; Betts *et al*., 2002; Taubman *et al*., 2004). At night surface O_3 is depleted by reacting with NO and through surface deposition as it is trapped in a shallow nocturnal inversion layer.

Tropospheric ozone seasonal variation

Surface O_3 has a seasonal variation. It has a broad peak in the dry season that is due to a large photochemical generation occurring in this season (Jacobs *et al*., 1995; Annegarn *et al*., 1996b; Betts *et al*., 2002). In southern Africa the peak concentrations occur in the spring months from August to November (Zunckel *et al*., 2004). The large photochemical production of surface O_3 in the dry season is supported by wide spread fossil fuel burning and biomass burning which both emit significant amounts of NO_X (O_3 precursor) into the atmosphere (Crutzen *et al*., 1999; Betts *et al*., 2002). The seasonal minimum concentration occurs in the wet season in the summer months of December and January (Zunckel *et al*., 2004).

Tropospheric ozone impact on health

O_3 has a negative health impact on humans. Its main target is the respiratory system. The severity of the impacts is determined by the length and frequency of exposure as well as the concentration levels (WDNR, 2006; Fierro, 2001; United States Environmental Protection Agency (USEPA), 2006b). It affects the respiratory system by irritating the mucous membranes of the nose and throat. The inhaled O_3 react with sensitive lung tissues causing several negative health effects (WDNR, 2006). The respiratory illnesses that can be caused by O_3 include the following:

- Asthma aggravation.
- Inflammation of lung tissue.
- Impaired lung function.

- Significant temporary decrease in lung capacity of 15% to over 20% in some healthy adults.
- Impair the body immune system, making people more susceptible to bronchitis and pneumonia.

Tropospheric ozone impact on plants

Tropospheric O_3 is the most important pollutant that has damaging effect on plants (van Tienhoven *et al*., 2004). It negatively influences the ability of plants to produce and store food. As a result plant growth, reproduction and overall plant health are compromised (USEPA, 2006b). O_3 react with plant molecules to form reactive molecules that destroy rubiscon, an enzyme crucial for photosynthesis, but the complete mechanisms by which O_3 damages plants are not yet fully understood (van Tienhoven *et al*., 2004; Giles, 2005). It has been found that many types of vegetation are affected by high surface O_3 concentration exposure. Studies have also found that with regard to agricultural crops, ozone damage has negative economical consequences (Huixiang *et al*., 2005; USEPA, 2006b).

Tropospheric ozone impact on climate

O_3 in the troposphere plays an important role in the atmospheric environment processes (WMO, 2006b). In the free troposphere O_3 is a direct greenhouse gas (Doddridge *et al*., 1992; Frost *et al*., 1998, IPCC, 2001; Betts *et al*., 2002; Fiore *et al*., 2002; WMO, 2006b), and is the third most important greenhouse gas after CO_2 and CH_4 (WMO, 2006b). It alters the surface energy balance, by strongly absorbing infrared radiation in the atmospheric window (wavelength of ~ 9.6 μm) (Betts *et al*., 2002). The global average concentration of tropospheric O_3 is believed to be increasing (Doddridge *et al*., 1992) and that increase is estimated to contribute the third biggest rise in direct radiative forcing since the pre-industrial era (IPCC, 2001). The growth in tropospheric O_3 global budget add to global warming, hence resulting global climate change (Parrish *et al*., 1990; Frost *et al*., 1998; IPCC, 2001; Gauss *et al*., 2003; Liao *et al*., 2006). On the other hand tropospheric O_3 also influences the oxidation capacity of the troposphere by producing OH radicals. The OH radicals are known as the detergents of the troposphere (Crutzen *et al*., 1999). They remove most gases in the atmosphere through chemical reactions preventing them from

accumulating (Crutzen *et al*., 1999; Hobbs *et al*., 2003). This includes greenhouse gases such as CO and CH_4. The removal of these greenhouse gases reduces global warming (Parrish *et al*., 1990; Doddridge *et al*., 1992, IPCC, 2001, WMO, 2006b).

Sulphur dioxide

SO_2 is a colourless gas phase pollutant with a sharp irritating smell, similar to that of a struck match (Chen *et al*., 2001; Queensland Environmental Protection Agency (QEPA), 2006; WDNR, 2006). It is non flammable, and it is soluble in water vapour (Victorian Association for Environmental Education (VAEE), 2005). Its concentration has a large spatial and temporal variation, because of its short atmospheric lifetime of a week and uneven anthropogenic source distribution (Seinfeld and Pandis, 2006; WMO, 2006c).

Sources of sulphur dioxide

The main source of SO_2 in the troposphere is the burning of sulphur containing fuel, like coal and oil. 90-95% of the combustion emissions containing sulphur are in the form of SO_2 (Finlayson-Pitts and Pitts, 1986; QEPA, 2006). On global scale it is estimated that 60% of SO_2 emissions are from coal burning, 30% from petroleum refining and burning, and 10% from other processes like smelting mineral ores (Finlayson-Pitts and Pitts, 1986). The single biggest source of SO_2 in South Africa is the energy sector. It includes electrical power generation, coal mining and gas extraction, as well as petroleum products production from coal and oil (van Tienhoven, 1999; Piketh *et al*., 2004). The source of high SO_2 concentration in residential areas in South Africa is mainly due to coal burning for space heating (Horne, 1978; Annegarn *et al*., 1996a). Natural sources of SO_2 in the atmosphere include volcanic emissions (Horne, 1978, WMO, 2006c). SO_2 can also be a secondary pollutant produced by the oxidation of dimethylsulphide (DMS) from oceans by OH radicals (Huebert *et al*., 1993; WMO, 2006c).

Sinks of sulphur dioxide

SO_2 is removed from the atmosphere by dry and wet deposition. In aqueous phase SO_2 is oxidised by hydrogen peroxide (H_2O_2) in cloud droplets then followed by wet acidic deposition. In gaseous phase OH radicals oxidises SO_2 resulting in gaseous sulphuric acid (H_2SO_4), then followed by H_2SO_4 vapour condensation onto aerosols particles and dry or wet acidic deposition to the environment. The oxidation of SO_2 in the atmosphere leads to the formation of sulphate (SO_4^{2-}) aerosols particles (Brock *et al*., 2002; Taubman *et al*., 2004; WMO, 2006c). The gas phase reactions that lead to SO_4^{2-} aerosols particles in the atmosphere are the following (Taubman *et al*., 2004):

$$OH + SO_2 + M \rightarrow HOSO_2 + M \quad \text{.....(1.11)}$$

$$HOSO_2 + O_2 + M \rightarrow HO_2 + SO_3 + M \quad \text{.....(1.12)}$$

$$SO_3 + H_2O \rightarrow H_2SO_4 \quad \text{.....(1.13)}$$

In cloud droplets dissolved SO_2 is oxidised to form SO_4^{2-} aerosols particles by the following reaction:

$$SO_2 + H_2O_2 \rightarrow H_2SO_4 \quad \text{.....(1.14)}$$

The sulphuric acid can rapidly react with atmospheric ammonia (NH_3) to form SO_4^{2-} particles (Horne, 1978; Brock *et al*., 2002) by the following reaction:

$$H_2SO_4 + 2\ NH_3 \rightarrow (NH_4)_2SO_4 \quad \text{.....(1.15)}$$

Sulphur dioxide diurnal variation

SO_2 has two diurnal variations that are influenced by the diurnal evolution of the mixing layer and the type of SO_2 source (Annegarn *et al*., 1996b; Stehr *et al*., 2000; Chen *et al*., 2001). Domestic coal combustion results in SO_2 diurnal variation with two peaks, one in the morning and the other one in the evening. The peaks are due to stronger source strength in the morning and evening from domestic fossil fuel burning and rush hour traffic emissions. The high concentrations of SO_2 in the morning and evening are aggravated by nocturnal ground level inversions (Annegarn *et al*., 1996b). The other diurnal variation has only one peak that occurs during mid-day, this is a rural or semi-rural feature. The peak is from tall industrial emissions. During mid-day the mixing layer is deep, and SO_2 aloft from tall industrial emissions is mixed down to the surface (Annegarn *et al*., 1996a; Stehr *et al*., 2000; Chen *et al*., 2001).

Sulphur dioxide seasonal variation

SO_2 surface concentration also varies seasonally. In winter SO_2 has peak concentrations and in summer it has the lowest concentrations. The highest concentrations in winter are found in residential areas, where coal burning is used for space heating (Annegarn *et al*., 1996b). The high SO_2 concentrations in winter are also aggravated by the influence of semi-permanent high pressure systems, which cause subsidence over southern Africa (Scheifinger, 1992, Annegarn *et al*., 1996b; Tyson *et al*., 1996; GDACE, 2004).

Sulphur dioxide impact on health

SO_2 can affect the respiratory system and irritate eyes. A brief exposure to high SO_2 concentrations can constrict the bronchi and increase mucous secretion, and make breathing difficult. When SO_2 irritates the respiratory tract, it causes coughing, worsening conditions such as asthma and chronic bronchitis and makes people more susceptible to respiratory tract infections. Children, elderly people, people with chronic lung disease, and asthmatics are especially prone to bad health effects caused by exposure to SO_2 (QEPA, 2006; WDNR, 2006).

Sulphur dioxide impact on plants

SO_2 can affect vegetation directly. It causes injury to many plant species and varieties. SO_2 affect metabolism of vegetation, and plants that are affected can be seen by the changes in their physical appearance. The first impact on plants being exposed to high SO_2 levels is noticeable on their leaves. Depending on the sensitivity of plants, the impact occurs within hours or days of being exposed to high concentrations of SO_2. Noticeable change can occur in sensitive plants at exposure as low as 0.12 ppm for eight hours, for intermediate sensitive plants it can occur at exposure of 0.30 ppm for eight hours (Atmosphere, Climate and Environment (ACE), 2004; WDNR, 2006).

Sulphur dioxide impact on climate

SO_2 in the atmosphere is precursor of fine SO_4^{2-} particles (WMO, 2006c). It is converted to SO_4^{2-} particles through aqueous and gas phase oxidation reactions (Brock *et al*., 2002). These SO_4^{2-} particles constitute a highly solar radiation scattering sulphate-dominated haze, which cools both the atmosphere and the surface (Taubman *et al*., 2004; Taubman *et al*., 2004). The cooling of the surface is caused by the

sulphate-dominated haze by blocking direct solar radiation from reaching the surface (VAEE, 2005).

The fine sulphate particles also behave as cloud condensation nuclei (CCN). They can change the microphysical structure of clouds, to the one with higher droplet concentration and narrower droplet size distribution. This causes an increase in shortwave albedo of clouds and reduces precipitation efficiency, which can lead to longer cloud lifetime and also enhance the reflection of solar radiation to space (Huebert *et al*., 1993; Piketh *et al*., 2004). The sulphate particles are important in determining the Global climate (Huebert *et al*., 1993), it was demonstrated by Brock *et al*., (2002) that their mass increases in the atmosphere are associated with high SO_2 concentrations.

Nitrogen oxides

NO_X is a mixture of nitric oxide (NO) and nitrogen dioxide (NO_2). NO is a colourless, odourless gas and is essentially insoluble in aqueous liquids. It is oxidised in the atmosphere to form NO_2. NO_2 is an odorous, brown gas and is moderately soluble in aqueous liquids. Both these gases are essential components of photochemical smog and NO_2 gives brown haze the redish-brown colour. They both have high spatial and temporal variability due to their short atmospheric lifetime of a day (Seinfeld and Pandis, 2006) and uneven source distribution (IPCC, 2001; QEPA, 2006; USEPA, 2006c; WMO, 2006a).

Sources of tropospheric nitrogen oxides

The compounds of NO_X are simultaneously emitted in combustion processes and other high temperature industrial operations (USEPA, 2006c). A large percentage of tropospheric NO_X are released as NO, which photochemically equilibrates with NO_2 within few minutes. The principal source of NO_X in the atmosphere is fossil fuel burning, with 40% contributed by the transport sector. Emissions of tropospheric NO_X in the free troposphere include aircraft emissions (Crutzen *et al*., 1999; IPCC, 2001). Other important sources of NO_X in the troposphere, but with less certainty in source contribution, are natural sources. They include lightning, microbial activity in soils,

oxidation of tropospheric NH_3, outgassing from oceans and stratospheric input (IPPC, 2001; Doddridge *et al*., 1992).

Sinks of tropospheric nitrogen oxides

The major sink of NO_X in the atmosphere is the oxidation of NO_2 by OH radicals to form nitric acid (HNO_3) and peroxyacetylnitrate (PAN), which are both eventually removed from the atmosphere by dry or wet deposition. Alternative pathways for direct NO_X removal from the atmosphere is through canopy scavenging of NO_X and direct dry deposition of NO_X, HNO_3 and nitrate particles (NO_3^-) to land and ocean surfaces. In addition dry deposition can influence the surface exchanges of (NO_X) and hence influence the release of NO_X into the atmosphere (Parrish *et al*., 1990; IPCC, 2001; WMO, 2006a).

Nitrogen oxides diurnal variation

Surface NO_X have a diurnal variation which has two peaks, occurring in the morning and at night (Annegarn *et al*., 1996b). The diurnal variation is influenced by the evolution of the mixing layer, the type of NO_X source and photochemistry. The peaks of NO_X concentrations are the result of stronger source strength in the morning and evening from domestic fossil fuel burning and rush hour traffic emissions. The high concentrations of NO_X in the morning and evening are aggravated by nocturnal ground level inversions and the absence or weak solar radiation intensity to support chemical consumption of NO_X to produce O_3 (Parrish *et al*., 1990; Doddridge *et al*., 1992; Poulida *et al*., 1994). The minimum concentrations of NO_X occur in the afternoon when solar radiation intensity is maximum, an optimal condition for the photochemical consumption of NO_X in O_3 production (Poulida *et al*., 1994).

Nitrogen oxides seasonal variation

Surface NO_X concentration varies seasonally. In winter NO_X have peak concentrations and in summer it they have the lowest concentrations (Parrish *et al*., 1990; Doddridge *et al*., 1992). The high NO_X concentrations in winter are due to increased emissions from wide spread biomass burning (Crutzen *et al*., 1999; GDACE, 2004), domestic fossil fuel burning (Annegarn *et al*., 1996b) and slower removal by less efficient photochemical processes (Parrish *et al*., 1990; Doddridge *et al*., 1992). The high NO_X concentrations in winter are also aggravated by the

influence of high pressure systems, which cause subsidence. These systems have a high frequency of occurrence in winter (Scheifinger, 1992; Tyson *et al*., 1996; GDACE, 2004). The summer minimum concentrations of NO_X are due to moist unstable conditions that are suitable to rapid pollution dispersion, mixing, deposition by rainfall (Tyson *et al*., 1996; GDACE, 2004), and faster removal by more efficient photochemical processes (Parrish *et al*., 1990; Doddridge *et al*., 1992).

Nitrogen oxides impact on health

High levels of NO_X exposure can cause death, and exposure to low levels can affect the delicate structure of lung tissue. It can disturb the blood's ability to transport oxygen through the body, resulting in headache, fatigue, dizziness, and blue colour to skin and lips. NO_2 is known to be highly toxic to various animals as well to humans. High levels of NO_2 exposure cause damage to the mechanisms that protect the human respiratory tract and it can also increase a person's vulnerability to respiratory infections and asthma. It may cause genetic mutations, damage a developing foetus, and decrease fertility in women. It may also lead to permanent lung damage if the exposure is repeated. On the other hand, exposure to industrial high levels of NO can cause unconsciousness, vomiting, mental confusion, and damage to the teeth. Low level exposure to NO_X can irritate the eyes, nose, throat, and lungs. It can cause coughing, shortness of breath, fatigue, and nausea (QEPA, 2006, WDNR, 2006)

Nitrogen oxides impact on plants

Surface NO_X have a direct and indirect damaging effect on vegetation (Morgan *et al*., 1992; Regional Air Pollution Control Agency (RAPCA), 2006; WDNR, 2006). NO_X directly affect plants by their toxic and irritating effects (RAPCA, 2006). They cause serious harm to plants at certain concentrations. Their effects include bleaching or killing of plant tissue, causing leaves to fall, and reducing plant growth rate (QEPA, 2006; WDNR, 2006). NO_X indirectly damages vegetation when they are converted in the atmosphere into nitric acid, nitrate particles and peroxyacetylnitrate (PAN). These three chemicals end up being dry or wet deposited on the plants surface (Phamornsuwana, 1991; Morgan *et al*., 1992; RAPCA, 2006). The surface deposited nitrate particles from atmospheric NO_X oxidation boost the soil mineral nutrients for plants, but at high levels they are destructive to plants (Morgan *et al*., 1992).

Nitrogen oxides impact on climate

Surface NO_X affect the Earth radiative balance indirectly by catalysing tropospheric O_3 production (IPCC, 2001; WMO, 2006a). Tropospheric O_3 is a greenhouse gas and a strong oxidant (Doddridge *et al*., 1992), its global budget influences global climate by contributing to global warming (Parrish *et al*., 1990; Frost *et al*., 1998). It can also moderate the effects of the other greenhouse gases by initiating the production of OH radicals, the detergents of the atmosphere. The OH radicals remove other greenhouse gases like CO, CH_4 and hydrofluorocarbons (HCFCs) from the atmosphere (Parrish *et al*., 1990; Crutzen *et al*., 1999; IPCC, 2001; WMO, 2006a). NO_2 can be further oxidised in the atmosphere, with nitrate particles being produced (Parrish *et al*., 1990). The nitrate particles directly and indirectly cause a negative surface forcing, resulting a net cooling on the surface (IPCC, 2001; Brink and Schaap, 2005).

Aerosols

Aerosols are a heterogeneous mixture of solid particles and liquid droplets suspended in the atmosphere (Held *et al*., 1996; NASA, 1996). Their size spectrum is characterised by a trimodal size distribution structure (Figure 1.5), the nucleation mode, accumulation mode, and coarse mode particles. Piketh *et al.*, 2004 has defined the three modes size ranges for continental, marine and remote continental environments for summer and winter season. The dominance of any of these particle size modes depend on the particles source. The urban continental environment's air is dominated by aerosol in the fine fraction. Marine environment's air is dominated by particles in coarse mode if it is uncontaminated by air from urban sources. Remote continental air can be dominated by fine particles and or coarse mode particles depending on its location with respect to urban continental and marine source regions (Piketh *et al*., 2004).

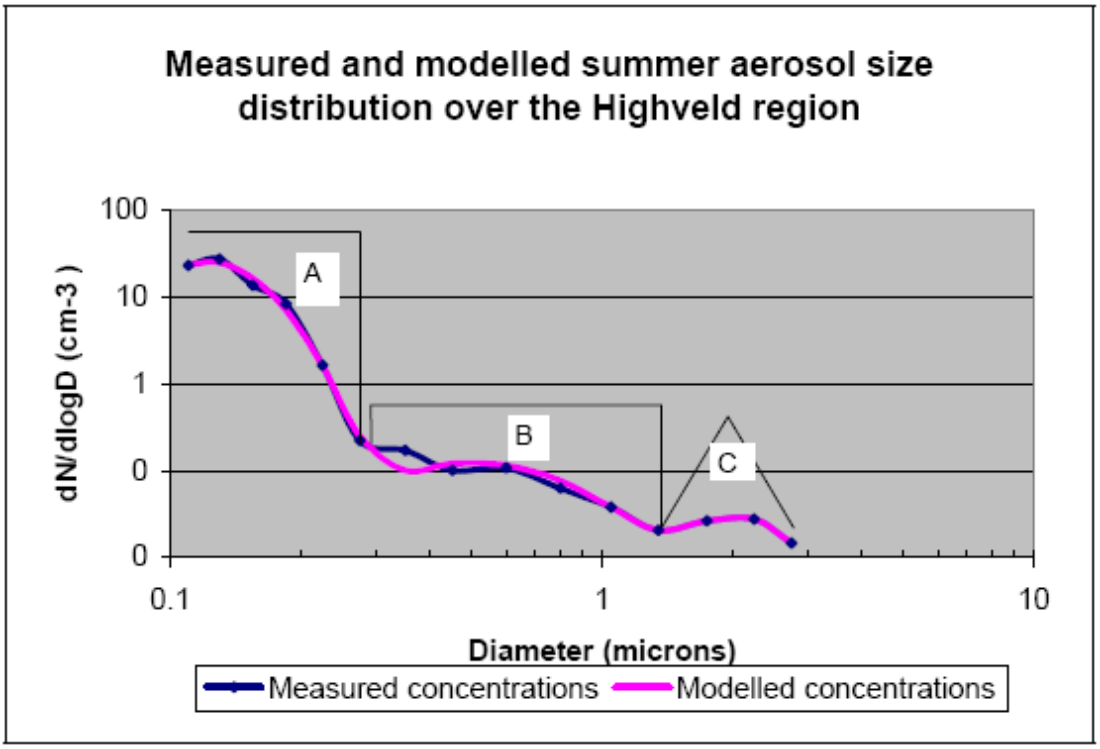


Figure 1.5: The measured and modelled aerosol number size distribution over the Highveld region. The three modes have been defined as nucleation mode aerosols (mode A), accumulation mode aerosols (mode B) and Coarse mode aerosols (mode C) (Piketh *et al*., 2004).

Aerosols in the atmosphere can be classified into two types, primary and secondary aerosols. Primary aerosols are released directly into the atmosphere by the source as particles, and secondary aerosols are formed by physical or chemical changes of gas phase constituents of the atmosphere (Huebert *et al*., 1993; Held *et al*., 1996; Brock *et al*., 2002; Carrico *et al*., 2003). The primary aerosol particles size distribution is tilted to larger aerosols, dominating the coarse mode, which has a diameter greater than 1μm. The secondary aerosol particle dominates the fine fraction, which is ranging between 0.1 and 1 μm in diameter, they mainly originate from chemical processes in the atmosphere (Buseck and Pósfai, 1998; Ross *et al*., 2003). The atmospheric lifetime of aerosols vary from few days to few weeks (Seinfeld and Pandis, 2006).

Sources of aerosols

Primary aerosols and precursors of secondary aerosols in the atmosphere emanate from natural and anthropogenic sources (Held *et al*., 1996). The natural sources include, Aeolian dust, volcanic eruptions, biomass burning and sea sprays (Huebert *et al*., 1993; O' Dowd *et al*., 1997; DeMott *et al*., 2003; Hobbs *et al*., 2003). The anthropogenic sources include, fossil fuel burning, industrial emissions, traffic emissions, and dust from land cover and land use change (Held *et al*., 1996; IPCC, 2001; Carrico *et al*., 2003, WDNR, 2006).

The relative importance of aerosols sources in atmospheric loading is difficult to establish. However some progress has been achieved in estimating the present day annual source strength of aerosol precursors. The NO_X derived aerosols global sources are dominated by fossil fuel burning, biomass burning and lightning. The important global sources for ammonia derived aerosols are domestic animals, agriculture and biomass burning. The SO_2 derived aerosols important global sources are fossil fuel burning, industrial emissions, and volcanic eruptions. Dimethylsulphide (DMS) or hydrogen sulphide (H_2S) derived aerosol's dominant global source is the oceans. Soil dust particles global sources are mainly deserts, dry lakebeds, and arid areas where vegetation have been reduced or soil surface have been disturbed by human activities (IPCC, 2001).

Sinks of aerosols

Aerosols are effectively removed from the atmosphere by rainfall and other forms of precipitation. They can also be dry deposited to the surface by the influence of gravity. The efficiency of both these atmospheric cleansing processes and the aerosols atmospheric lifetime is dependent on the aerosols physical and chemical properties (IPCC, 2001; Brock *et al*., 2002; Carrico *et al*., 2003; Ross *et al*., 2003; ACE, 2004).

Aerosols diurnal variation

At the surface aerosols have a diurnal variation that is influenced by the diurnal evolution of the mixing layer and the type of aerosols source. Low level aerosols sources in close proximity give a diurnal variation with two peaks, one in the morning and the other in the evening. The peaks are due to stronger source strength in the morning and evening associated with fossil fuel combustion and rush hour traffic

emissions. The peak aerosols concentrations in the morning and evening are aggravated by nocturnal ground level temperature inversions, which act as a barrier to vertical dispersion of aerosols allowing build-up of aerosols concentrations. The low concentrations in the afternoon is the result of a deeper mixing layer, diluting aerosols released from the surface (Annegarn *et al*., 1996b; Chen *et al*., 2001, Butler *et al*., 2003; Carrico *et al*., 2003).

Aerosols seasonal variation

Atmospheric aerosols concentrations vary seasonally. In winter they have peak concentrations and in summer they have the minimum concentrations (Annegarn *et al*., 1996b; Butler *et al*., 2003). The high winter concentrations are associated with increased fossil fuel combustion for space heating. The low summer concentrations are associated with atmospheric cleansing effect by rainfall (Annegarn *et al*., 1996b). The high winter concentrations are aggravated by shallow mixing layers caused by the semi-permanent high pressure systems, which cause subsidence (Scheifinger, 1992; Tyson *et al*., 1996; GDACE, 2004; Butler *et al*., 2003).

Aerosols impact on health

Surface aerosols are known to have a negative health effects on humans (Held *et al*., 1996; WDNR, 2006; Peng *et al*., 2008; Hu and Rao, 2009). Almost all of the aerosols inhaled are either exhaled or trapped in the upper areas of the respiratory system. The inhaled aerosols enter the windpipe and lungs, cling to protective mucous and are removed. The other method of removing the inhaled aerosol particles is through coughing. Both these pulmonary clearance methods collectively protect the lungs from most of the inhaled particles (WDNR, 2006). Bigger size aerosols are generally filtered in the nose and throat and they normally don't pose health problems (Peng *et al*., 2008). But smaller particles that can be inhaled having a diameter less than 10 μm, known as PM_{10}, can make their way into the bronchi, lungs and capillaries and settle there (WDNR, 2006; Peng *et al*., 2008). They are known to cause the slowing down of the exchange of oxygen and carbon dioxide in the blood, resulting in shortness of breath. They also strain the heart, because it must work harder to make up for the oxygen loss (WDNR, 2006). In a similar manner particles with diameter less than 2.5 μm ($PM_{2.5}$) also penetrate through gas exchange regions of

the lung and pass through lungs to impact other organs. The $PM_{2.5}$ may cause high plaque deposits in the arteries, resulting in vascular inflammation and atherosclerosis – a hardening of arteries that reduces elasticity, which may lead to heart attacks and other cardiovascular problems. A short exposure to high concentrations of $PM_{2.5}$ particles could significantly contribute to heart disease (Hu and Rao, 2009).

Aerosols impact on plants

Aerosols particles in the atmosphere have both damaging and beneficial effects on vegetation. The type of effect depends on the chemical composition of the deposited matter, the duration and intensity of deposition episodes, the species and genotypes on which the substances are deposited, the physiological condition, structure and stage of maturity of the organism. The damaging effects are caused by phytotoxic aerosols, which chemically destroy plants, and inert aerosols which physically damages plants (Cowling and Linthurst, 1981; Niyogi *et al*., 2004; Health Canada, 2005; USEPA, 2006a). The phytotoxic aerosols contribute to acid rain and acidic clouds and fog. Acid rain destroys vegetation by damaging its leaves, limiting nutrients available to it, and poisoning vegetation with toxic substances like aluminium which is slowly released from the soil. Acid rain limits the mineral nutrients by dissolving them in the soil, and then washing them away before they can be taken up by vegetation. Vegetation lying on mountainous terrain can be exposed to acidic clouds and fog which are often more acidic than rainfall. The acidic clouds and fog wears away the protective waxy coating of the leaves, damaging them and creating brown spots on them. This damage on leaves makes vegetation weak and vulnerable to be attacked by disease or insects that ultimately kill them (USEPA, 2006a).

Inert aerosols physically damages vegetation by smothering their leaf surface, and blocking their stomata by settling or penetrating stomatal apertures. The physical smothering of the leaf surface reduces transmission of solar radiation, which is needed to induce photosynthetic processes. Physical blocking of the stomata reduces stomatal resistance, resulting in higher potential for the uptake of pollutant gases, and it may also affect water vapour exchange (Health Canada, 2005).

The particles in the atmosphere that are beneficial to vegetation consist of chemical elements that are essential for plant growth and development. These elements include Calcium (Ca), Potasium (K), Phosphorus (P), Iron (Fe) and Zinc (Zn). However these micro-nutrient elements can be injurious to plants in excess amounts (Cowling and Linthurst, 1981). Atmospheric aerosols also benefit vegetation by scattering solar radiation, allowing the scattered light to reach more leaves at different levels of each plant. The sunlight induces the uptake of carbon dioxide by the vegetation leaves for photosynthesis. The effect of aerosols in the atmosphere is that it exposes more leaves in the entire plant to sunlight so they can also undergo photosynthesis (Niyogi *et al*., 2004).

Aerosols impact on climate

Aerosols in the atmosphere have an influence on global climate and climate change. Their influence may vary from local to global scale depending on the type of source and its strength. They influence the climate by directly and indirectly interacting with the Earth's radiation budget (NASA, 1996; Buseck and Pósfai, 1998; ACE, 2004). They can either intensify or moderate the effects of greenhouse gases (Buseck and Pósfai, 1998; Ross *et al*., 2003).

Atmospheric aerosols directly influence the transfer of energy in the atmosphere through scattering and absorbing both incoming solar radiation and surface thermal radiation (Buseck and Pósfai, 1998; ACE, 2004). Black carbon or soot strongly absorb in the visible, near ultraviolet and near infrared light spectrum, resulting in a net warming effect at the surface and intensifying the greenhouse effect (Buseck and Pósfai, 1998; Chen *et al*., 2001). The aerosols can also counter act the effect of greenhouse gases by scattering solar radiation, diffusing its radiation intensity reaching the surface, and producing a net cooling effect at the surface. The sulphate particles are considered to be the most important scatterers of solar radiation on global scale (Buseck and Pósfai, 1998), but locally or regionally nitrates can be more important scatterers than sulphates (IPCC, 2001; Brink and Schaap, 2005). Atmospheric aerosols also moderate the effect of greenhouse gases by influencing their deposition. Diffuse light scattered by aerosols enhances the uptake of CO_2 a major greenhouse gas by vegetation (Niyogi *et al*., 2004).

Aerosols indirectly influence the atmospheric energy transfer by acting as cloud condensation nuclei (CCN). At high CCN concentrations, cloud droplet size distribution is dominated by small droplets, which makes clouds more reflective. Clouds with high small droplet concentrations are inefficient in forming precipitation and they have a longer atmospheric lifetime (Ross *et al*., 2003; ACE, 2004; Broekhuizen *et al*., 2006).

Impact of meteorology on air quality levels

Meteorology plays a central role in ambient air quality levels. The overall variation in air pollution levels is determined by the meteorological conditions prevailing (McNider *et al*., 1998). Meteorology influences the emissions of precursors, their transport and mixing, and the photochemical production rates of secondary air pollutants (Tyson *et al*., 1996; McNider *et al*., 1998). The important meteorological parameters affecting air quality levels in the atmosphere are wind, temperature, humidity, cloud cover and precipitation (Vukovich, 1994; McNider *et al*., 1998; Crutzen *et al*., 1999).

Surface winds

Strong surface winds dilute air pollutants in the atmosphere. Wind strength is determined by synoptic pressure gradients, but mesoscale features like topography and other high obstacles can influence surface wind speed (McNider *et al*., 1998). Light surface winds with variable direction cause stagnation and accumulation of air pollutants (Scheifinger, 1992). Light surface winds are normally associated with high pressure systems (Vukovich, 1994), which occur most frequently in winter over subtropical southern Africa (Tyson *et al*., 1996).

Surface temperature

Surface temperature influences air pollution levels. Warm temperatures are associated with high photochemical air pollution levels (Sillman and Samson, 1995). For ozone production, the evaporative emissions of anthropogenic and biogenic volatile organic compounds (BVOC) require warm temperatures (Sillman and Samson, 1995; McNider *et al*., 1998). Surface temperature is often correlated to insolation (McNider *et al*., 1998), which in turn is related to photochemical production of air

pollutants in the atmosphere (Poulida *et al*., 1994; Daum *et al*., 2000). The chemical kinetics rates of photochemical processes in the atmosphere are dependent on temperature (McNider *et al*., 1998).

Humidity, cloud cover and precipitation

Humidity, cloud cover and precipitation also influence the levels of air pollution in the atmosphere. Moisture in the form of vapour is a precursor of OH radicals. The OH radicals, the detergent of the atmosphere, oxidises most gases in the atmosphere. They can initiate reactions leading to formation of photochemical air pollutants and they can also act as a sink of other air pollutants (Crutzen *et al*., 1999; Taubman *et al*., 2004). Clouds affect air pollution levels by reflecting and scattering incoming solar radiation (Earth observatory, 2004), that is needed for photolysis of gases in the atmosphere to produce photochemical air pollutants (Poulida *et al*., 1994; McNider *et al*., 1998; Taubman *et al*., 2004). Cloudy conditions result in less photochemical pollutants, because of suppressed photochemical activity caused by limited solar radiation to drive ozone photochemistry (Betts *et al*., 2002). Water soluble gaseous air pollutants can be oxidised in cloud droplets to produce aerosols (Taubman *et al*., 2004). Precipitation result in low air pollutants concentrations as they are wet deposited to the surface (Betts *et al*., 2002, GDACE, 2004).

Transportation of air pollutants

The total global concentrations of atmospheric constituents are dependent not only on their regional production but also on their transport and mixing (Tyson *et al*., 1996). Transportation of air pollutants from biomass burning products, urban and industrial emissions in the troposphere over southern Africa and adjacent ocean areas, is driven by interacting planetary and synoptic scale features of the general circulation of the southern hemisphere (Garstang *et al*., 1996). The general circulation over southern Africa is dominated by four major circulation types, occurring with different frequencies throughout the year (Figure 1.6). They are semi-permanent subtropical anticyclones, transient mid-latitude ridging anticyclones, westerly baroclinic disturbances and barotropic quasi-stationary tropical easterly waves. The anticyclonic type circulation has a tendency of recirculating air masses at various scales, ranging from subcontinental, regional, and local scale. Transport of air mass at a particular

location is based on the circulation type prevailing at the location of origin of the transport. These circulation types show an annual cycle. The anticyclonic circulations combined together show maximum frequency of occurrence of 80% in midwinter. The easterly tropical disturbance has a maximum frequency of occurrence of 55% in midsummer. Westerly disturbances show little seasonal variation with a peak occurrence of 40% in spring (October) (Garstang *et al*., 1996; Tyson *et al*., 1996).

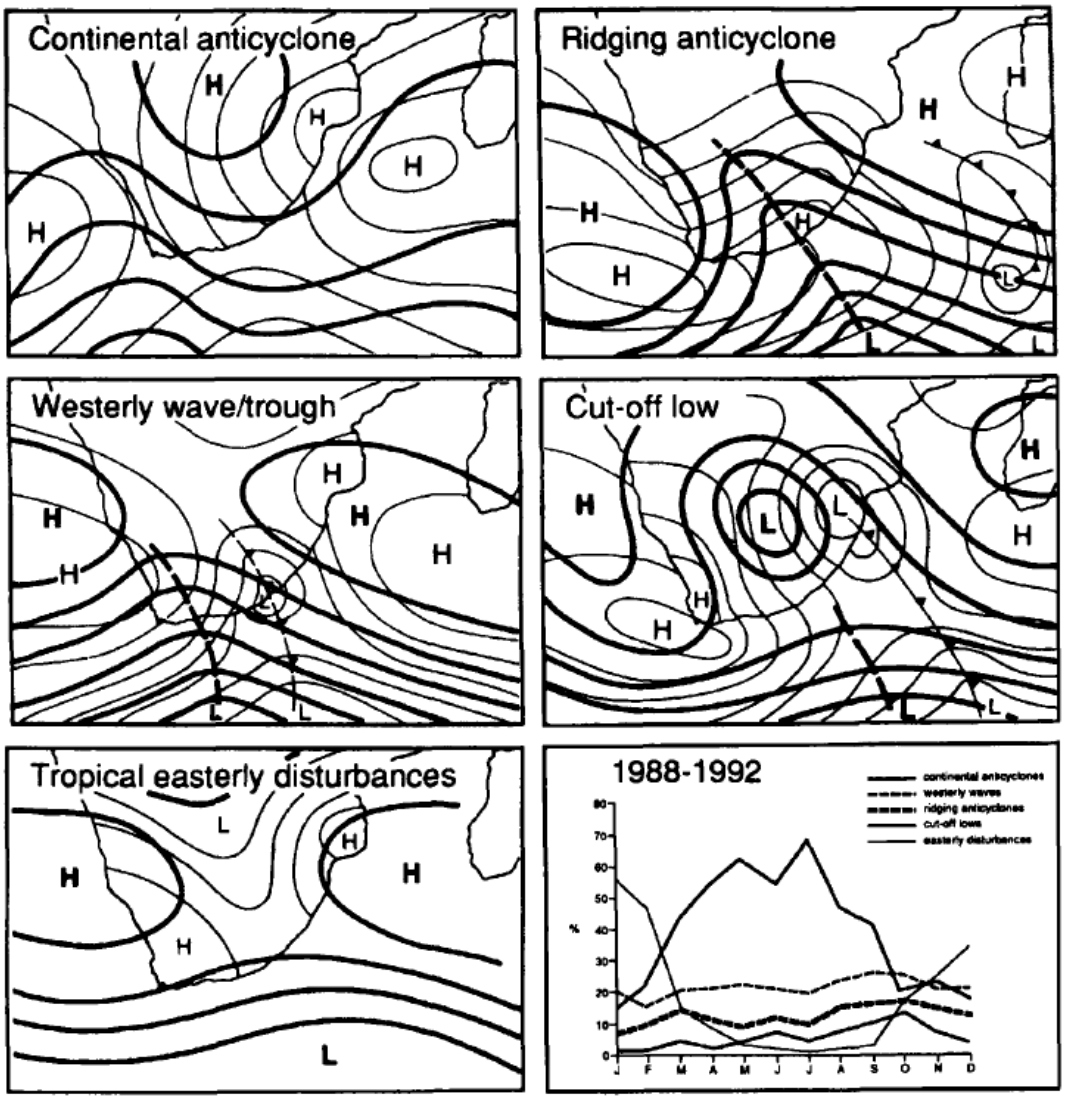


Figure 1.6: Major circulation types affecting southern Africa and their monthly frequency of occurrence over the 5-year period 1988-1992. Heavy lines represent conditions at 500 hPa; light lines represent surface conditions (as sea-level isobars over the oceans and contours of the 850 hPa surface over the subcontinent) (Garstang *et al*., 1996; Tyson *et al*., 1996).

Vertical transport

Southern Africa and the adjacent Atlantic and Indian Oceans are positioned below a region between the Hadley and Ferrel cells of the southern hemisphere general circulation, where large scale subsidence is occurring. (Garstang *et al*., 1996; Tyson *et al*., 1996; Piketh and Walton, 2004). Therefore, the occurrence of this wide spread subsidence is characteristic of southern Africa. During the Southern Africa Fire-Atmosphere Research Initiative (SAFARI) investigation, three wide-spread and temporal persistent stable discontinuities were found to be frequently occurring in the vertical structure of the atmosphere during the dry season. The first discontinuity occurred at the top of the mixing layer and centred at 750-700 hPa (3000 masl; 1500 magl of interior plateau). The second was associated with the main subsidence inversion located on average at about 550-500 hPa level (5000 masl; 3500 magl of interior plateau). The third discontinuity was at about 350 hPa level (8000 masl; 6500 magl of interior plateau), sandwiched by the main subsidence and tropopause. On some days the mixed layer can be deeper and extend up to 500 hPa, after the 700 hPa discontinuity has been broken by the passage of the convective westerly wave disturbance. The 550-500 hPa discontinuity is temporally persistent, and can withstand the passage of shallow westerly disturbances (Garstang *et al*., 1996; Tyson *et al*., 1996). The result of this temporal persistence of multiple stable layers is trapping of surface originated pollutants into altitude bands of air pollution, and limits their vertical transport. But well developed deep low pressure troughs and cut off low (COL) pressure systems, can destabilise the whole column of the troposphere up to the stratosphere. This enables air mass transfer between the troposphere and stratosphere (Garstang *et al*., 1996; Freiman and Tyson, 2000).

Horizontal transport

Five air transport modes can be produced frequently from the three prevalent circulation types mentioned before. They are direct easterly or westerly transport, easterly or westerly transport out of an initially anticyclonic circulation, and anticyclonic recirculation (Garstang *et al*., 1996). The continental circulation type results in two air mass transport modes. The first transport exit the subcontinent into Atlantic Ocean through the northern periphery of the continental anticyclone in the Angolan plume, the second transport exit the subcontinent into the Indian Ocean via the Natal plume. The first transport mode predominates with this circulation type.

Ridging highs from mid-latitudes dominant transport is towards the southern Atlantic Ocean, and the recirculated transport is towards the south into the Indian Ocean. Westerly waves produce a fast transport into the mid-latitudes of the Indian Ocean. Easterly waves produce low level transport exiting the subcontinent into Atlantic Ocean. Continental tropical convection elevates transport to 550 hPa before it subsides quickly to 850 hPa as it reaches the Atlantic Ocean (Garstang *et al*., 1996; Tyson *et al*., 1996).

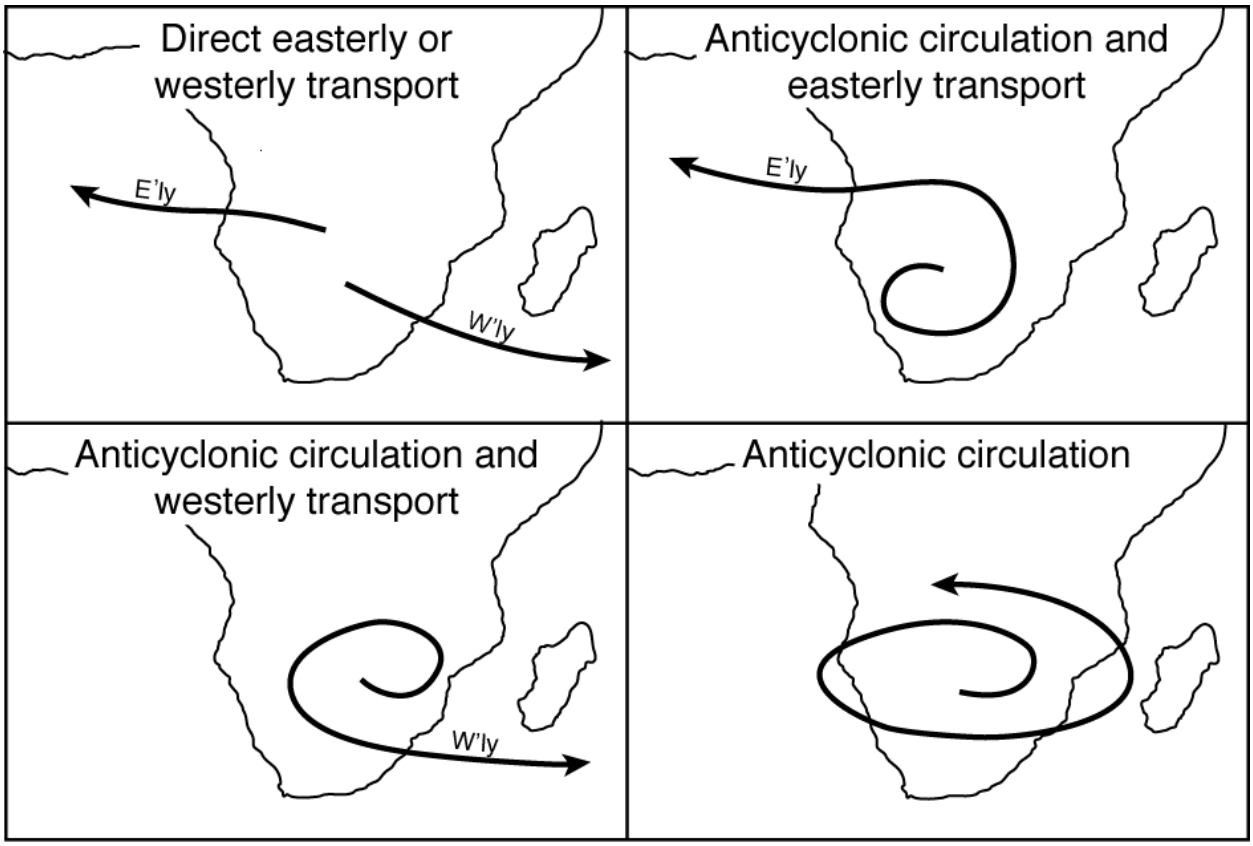


Figure 1.7: Schematic representation of major low-level transport trajectory modes likely to result in easterly or westerly exiting of material from southern Africa or in recirculation over the subcontinent (Garstang *et al*., 1996).

Air transport influencing air pollution levels over the Highveld

The Highveld region is the largest source of anthropogenic air pollutants in southern Africa. The concentration levels of air pollutants in this region are also influenced by air transport into and out of this region. There are four major transport pathways to the Highveld between 850 and 700 hPa (Figure 1.7). They result from the flow from the Atlantic Ocean, Indian Ocean, subtropical Africa, and over southern Africa. Transport from the Atlantic Ocean, which is most likely to be free of industrial emissions,

occurs most frequently (43%). Transport from Indian Ocean (26%) and the African continent (25%) together contribute approximately 50% of all transport to the Highveld. African transport may bring air pollutants from central-southern Africa to the Highveld. Southern Africa transport reaching the Highveld is frequently loaded with previously emitted industrial air pollutants. Air is transported out of the Highveld in two modes. One is direct transport, in which atmosphere constituents are directly transported to the Indian Ocean, Atlantic Ocean, and south Indian Ocean or toward equatorial Africa. The other is recirculated transport, in which material recirculates toward a point of origin over the subcontinent on a regional or subcontinental scale (Freiman and Piketh, 2003; Piketh *et al*., 2004).

Aims of this study

The aim of this study is to compare air pollution hotspots over the Highveld, utilizing the airborne data of SO_2, NO_X, O_3 and $PM_{2.5}$ aerosols. The air pollution hotspots investigated are Witbank, Secunda, Rustenburg and the Vaal Triangle area, including their surrounding towns. They consist of industrialised locations surrounded by urban and rural sites. The specific objectives of this study are to:

1. Compare the air pollution hotspots over the Highveld by identifying the differences and or similarities in O_3, NO_X, SO_2, and $PM_{2.5}$ concentrations.
2. Compare the seasonal variations of O_3, NO_X, SO_2, and $PM_{2.5}$ concentrations over the hotspots.
3. Investigate the relationship between airborne and ground based air pollution data.

****

Land cover and land use of the Witbank, Secunda, Rustenburg and the Vaal Triangle area, which impacts on the air quality over these study areas, has been described. Air pollution problems, sources and the emissions constituents over the Highveld region have been presented. A discussion on the influence of meteorology on air pollution levels has been given in this chapter. Aims and objectives of the study have been outlined in detail.