CHAPTER 1

BACKGROUND

This Chapter gives a general overview of the climate of southern Africa and the aerosol forcing effect. The Zambian Copperbelt and biomass burning products are considered as major contributors of aerosols that traverse the southern Africa subregion. These pollutants join those from the Mpumalanga Highveld in South Africa and are exported out of the continent within major circulation gyres in the region.

1.1 INTRODUCTION

The atmosphere over South Africa is each year laden with large quantities of aerosols and trace gases, for example sulphur dioxide, dust and smoke particles. Trace gases and particulate matter in the atmosphere have large impacts on local and regional visibility and planetary albedo (Houghton, 1995). Airborne particles exert radiative forcing, through the direct cooling effect from scattering of short wave solar radiation and the indirect effect on cloud condensation nuclei and cloud properties (Twomey, 1977; Charlson *et al,* 1987). Their impact on local and regional climate can therefore not be ignored**.**

Particulate and trace gas movement over southern Africa has fallen under considerable scrutiny over the last decade (Annegarn *et al*, 1993; Piketh, 1994; Salma *et al*, 1994; Maenhaut *et al.,* 1996; Tyson *et al.,* 1996; Garstang *et al.*, 1996). Studies have revealed that trace gases and particulate matter from the industrialized Highveld of South Africa (Piketh *et al,* 1999) and from the pyrometallurgical processing of copper in the Democratic Republic of Congo and the Zambian Copperbelt undergo marked recirculation before being either deposited or transported out of the sub-continent off the Atlantic and Indian Ocean and beyond (Meter, 2000). These aerosols are mainly concentrated in the southern African haze layer, which is a ubiquitous sub-continental feature of the lower atmosphere (Tyson *et al*., 1996, Piketh *et al*, 1999). Aerosols from biomass burning are thought to contribute substantially to the total background loading within the layer, almost exclusively during the late winter and austral spring season (Garstang *et al.*, 1996), while industrially derived sulphur and crustally derived aeolian dust are the major summer and winter constituents of the haze layer over Southern Africa (Piketh, 1999).

The extent to which air pollution, particularly industrial sulphur from the Zambian Copperbelt, and biomass burning products from the sub-region in general are transported over South Africa is the focus of this research. Recent findings indicate that sulphate concentrations over Southern Africa, previously attributed solely to South African industrial emissions, have a contribution from the Zambian Copperbelt (Meter, 2000). Climatological conditions in Southern Africa are such that the dominance of anticylonic activity in the region is favourable to the west, and then to the southward transportation

of air as a recirculation plume from the Copperbelt. These pollutants join those originating from other countries in the region and the Mpumalanga Highveld in South Africa to recirculate over the country, and some to exit the continent at ~30º S towards Australasia (Zunkel *et al.*, 2000).

It has been hypothesized that the generation of aerosols from biomass burning has been over-emphasized for South Africa and that in the sub-tropics of Southern Africa south of 20°S other sources are equally as important as contributors to the total aerosol loading of the lower troposphere (Piketh, 1999). The Southern African region is dominated by industrial infrastructure, most of which is coal based, and releases large amounts of sulphur pollutants into the atmosphere. Modeled SO_2 concentrations on the central Highveld of South Africa range between 10 and 50ppb, exceeding 50ppb in source areas. Such high concentrations in sulphur emissions play a large role as climate forcing agents (IPCC, 2001).

1.2. THE RESEARCH HYPOTHESIS

The industrialised Mpumalanga Highveld in South Africa is not the sole contributor to high aerosol loading over South Africa. Pollutants from the Zambian Copperbelt and products of biomass burning in southern Africa have a marked contribution to total aerosol loading over the country more than was originally expected.

The hypotheses to be tested are:

(i) sulphate pollutants from the industrialised South African Highveld are not the sole contributors to high aerosol loading over South Africa and the subcontinent.

(ii) Products of biomass burning make a substantial contribution to aerosol loading over the southern African subcontinent, especially during the dry winter and spring seasons.

(iii) Recirculation is a major feature of the lower troposphere and results in a transboundary transportation of aerosols in the Southern African subregion.

1.3 AREA OF STUDY

The area of study encompasses the southern African subcontinent from the Zambian Copperbelt, more specifically from Kitwe (12.9°S, 28.2°E, 1262m amsl) to the tip of the South African coast at approximately 35°S (Fig. 1.1). The Copperbelt is positioned on an inland plateau approximately 1200 to 1500m amsl. Botswana and Namibia have vast tracts of desert land, stretching to the southwestern parts of South Africa. The Highveld of South Africa is dominated by sulphur producing industrial activities. The vegetation of the southern African region is dominantly scrub savanna, comprising of inland plants that are xerophytic due to the lengthy dry period.

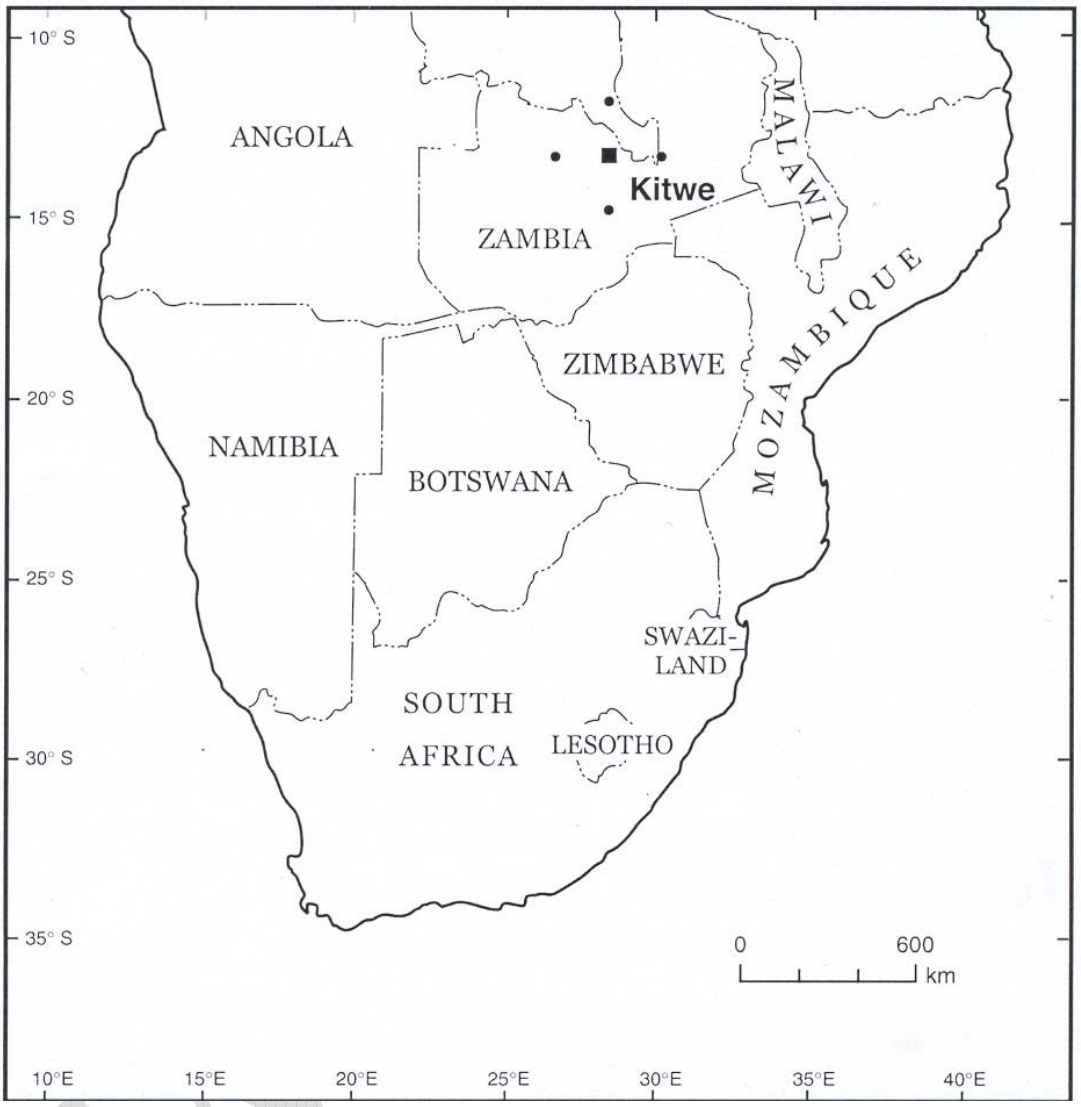

Figure 1.1 The positioning of Kitwe showing the 5-point cluster from which trajectories have been run. The other countries in central and southern Africa are affected by pollutants from the Copperbelt and are producers of biomass burning products.

1.4 LITERATURE REVIEW

The climate of southern Africa, and implications for aerosol transportation.

Southern Africa is characterized by the presence of semi-permanent sub-tropical continental anticyclones that mark the boundary between the Hadley and Ferrell cells (Houghton, 1995). Because of the descending nature of air masses in these regions, the atmosphere is quite stable. Frequently occurring stable discontinuities are found to control vertical transport of aerosols in the sub-tropical atmosphere, the main being the subsidence-induced, ubiquitous and temporarily persistent absolutely stable layers at the ~850, ~700 and ~500 hPa levels in the troposphere (Cosjn and Tyson, 1996; Freiman and Tyson, 2000). The layers at the ~700 and ~500hPa levels have been found to be the most spatially consistent over the entire subcontinent (Cosijn and Tyson, 1996). The absolutely stable layers effectively cap horizontal transport and inhibit any vertically upward transfer of air over the subcontinent (Tyson *et al.,* 1996). The absolutely stable layer has been found to coincide with the African haze layer capped by the equivalent of the mean climatological `500hPa absolutely stable layer (Cosijn and Tyson, 1996), and during a flight undertaken during TRACE-A (Browell, 1993) was continuous from south of Johannesburg to north of Lusaka in Zambia (Figure 1.2).

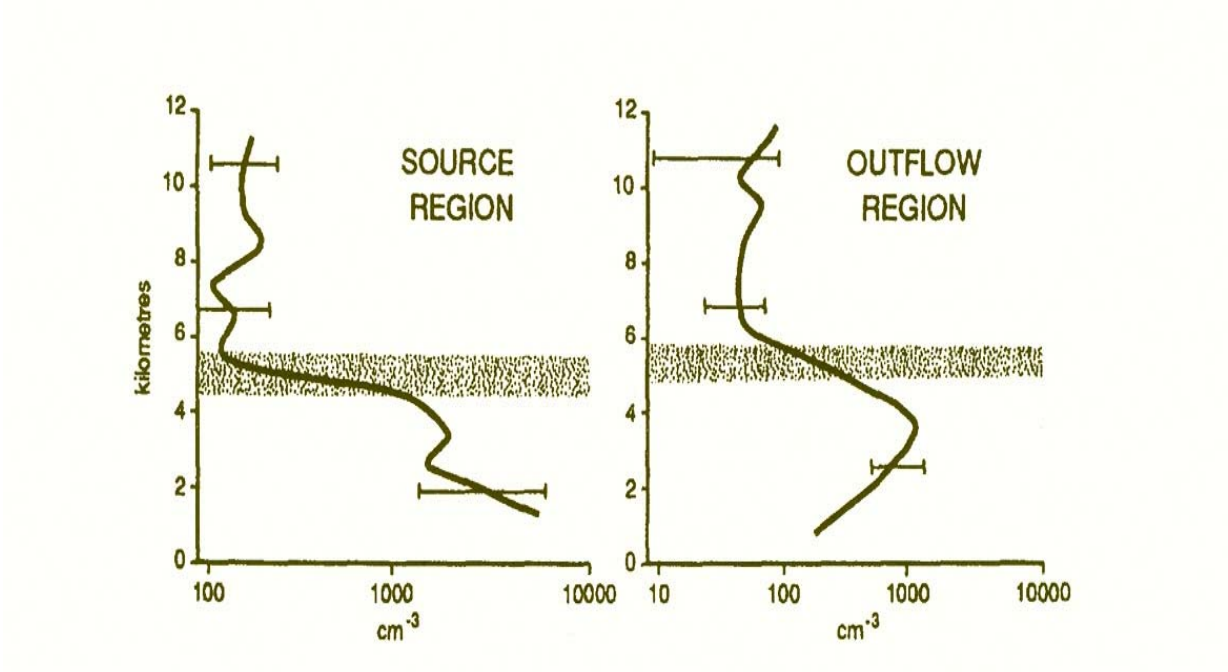

Figure 1.2 The mean 1986-1992 ~500hPa absolutely stable layer as determined by Cosijn and Tyson (1996). Average vertical profiles show aerosol number density observed over aerosol source regions during SAFARI/TRACE-A over the Atlantic Ocean. (After Anderson *et al*., 1996).

Subsidence over the sub-tropical region of Southern Africa also favours radiative inversions (Tyson, 1997; Piketh, 1999). Such situations create obvious implications for local as well as regional transport and recirculation of aerosols and trace gases

The four most common synoptic circulation types over South Africa are semi-permanent continental anticyclones, transient mid-latitude ridging anticyclones quasi-stationary easterly waves, and westerly baroclinic disturbances (Figure 1.3) (Tyson *et al*., 1996, Tyson, 1987; Preston-Whyte and Tyson, 2000). Synoptic structures control vertical

divergence and convergence, which in turn control the structure of the absolutely stable layers.

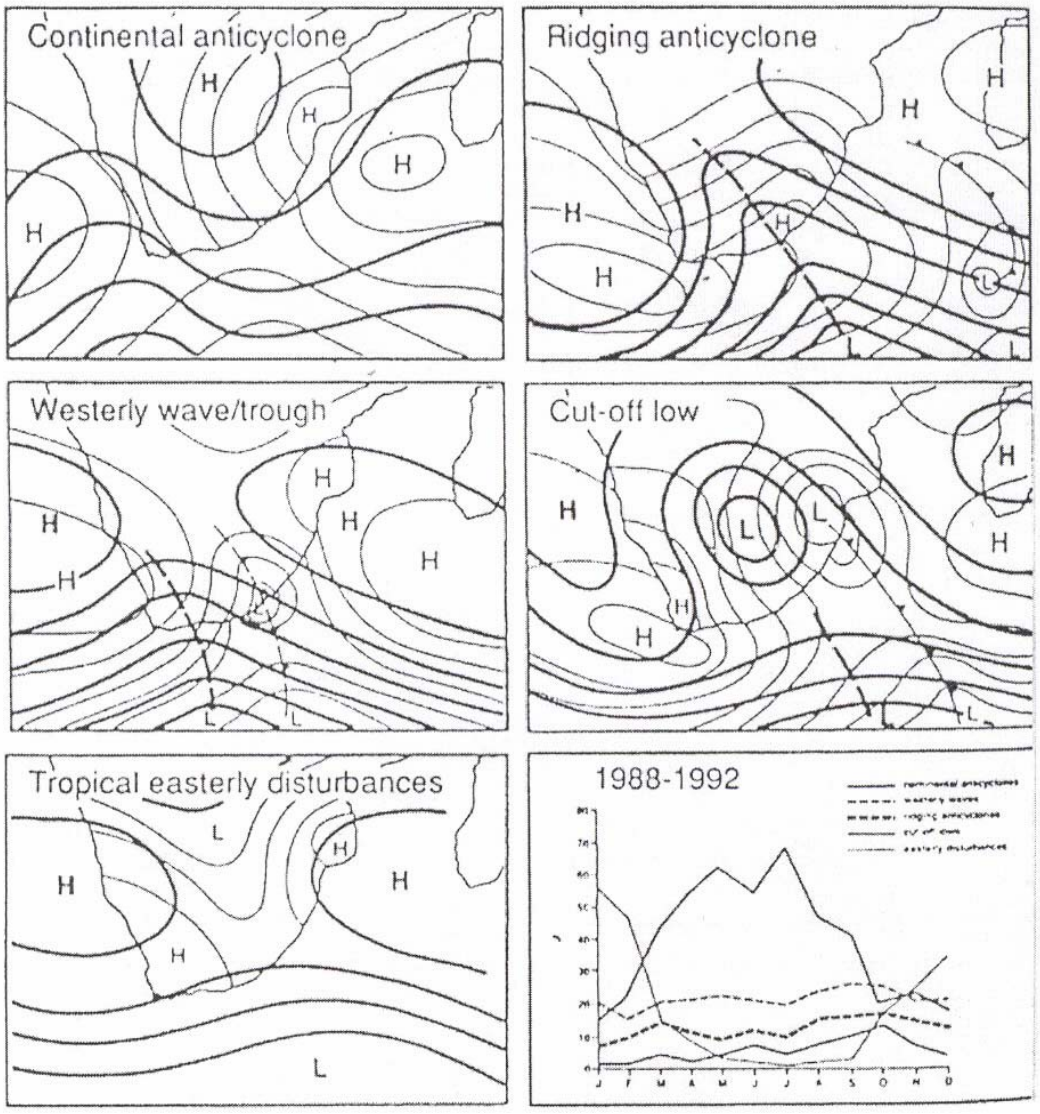

Figure 1.3 Major circulation types over subtropical southern Africa, 1988-1992 (Tyson *et. al*, 1996b).

The direction of air mass transport depends on the spatial domain of the circulation type at any one time (Tyson *et. al.,* 1996, Tyson and Preston-Whyte, 2000). Therefore the long-range transportation of aerosols requires a critical review of the main atmospheric circulation patterns over identified source regions. Such transportation has to be effected by the location of its origin and thus depends on the area in which the circulation type occur preferentially (Tyson *et al*., 1996, Tyson and Preston-Whyte., 2000). An example of this is illustrated in Figure 1.4. To the northwest an easterly wave occurred: elsewhere over the subcontinent a large anticyclone was evident. In the region of the easterly wave, transport into the Atlantic Ocean along the pathway suggested by the circulation type (pathway A) correspondent to actual 875-700 hPa trajectories. (Fig 1.4, upper right). Air originating near the center of the anticyclone could have been expected to recirculate, and trajectories to the lower left of the diagram show that they did. As expected those originating to the south-western side of the antyicyclone exited directly into the south-western Indian Ocean (Figure 1.4, lower right.) (Tyson and Preston-Whyte, 2000).

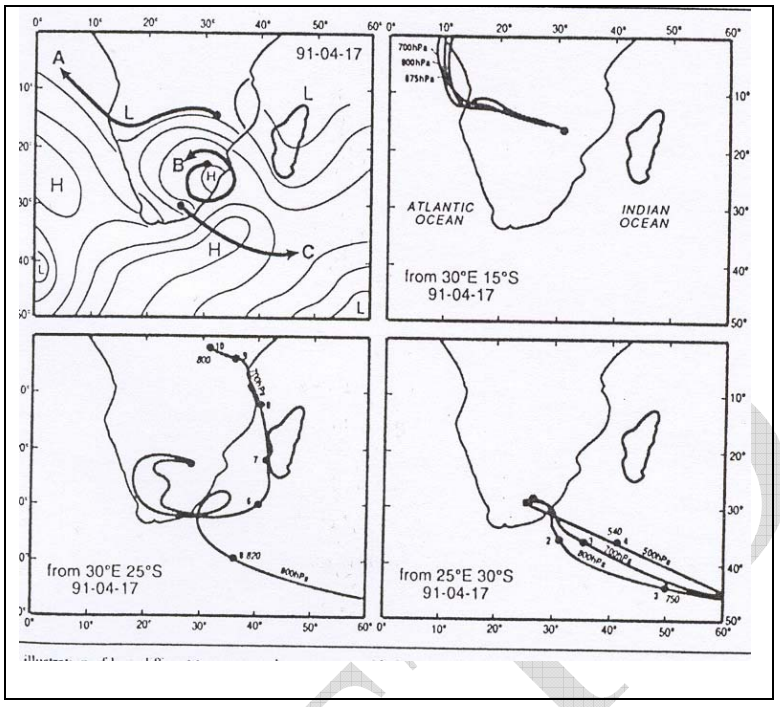

Figure 1.4 An illustration of how different transport may occur with the same synoptic situation (Tyson *et al.,* 1996)

Aerosol and trace gas transportation

The climatology of the air transport plumes which advect aerosols and trace gases out of southern Africa is now well understood (Garstang *et al*., 1996, Tyson *et al*., 1996, Tyson and D'Abreton). Trajectory modeling reveals transport across the Indian Ocean to Australia and New Zealand (Fig 1.5), and on occasions, these plumes may transport dust from southern Africa right across the ocean to Australia. Such transport of aerosols have been modeled in general circulation model transport models (Rayner and Law, 1995) (Fig 1.6) and the existence of clear plumes of carbon-dioxide extending from Africa to Australia have been confirmed (Tyson and D'Abreton, 1998). Aerosol transportation is

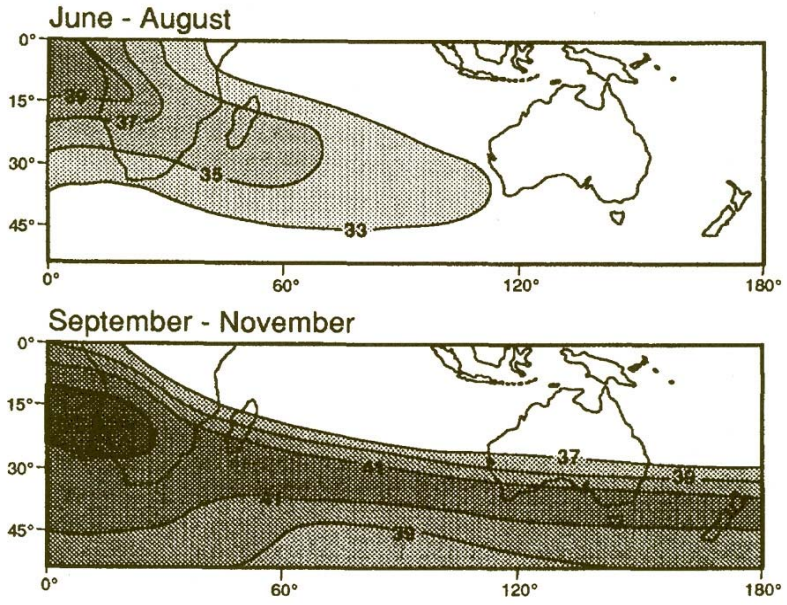

Figure 1.5 Trans-oceanic transportation of residual tropospheric ozone, off the east coast of southern Africa in June-August and September-November (adapted after Fishman *et al.*, 1990)

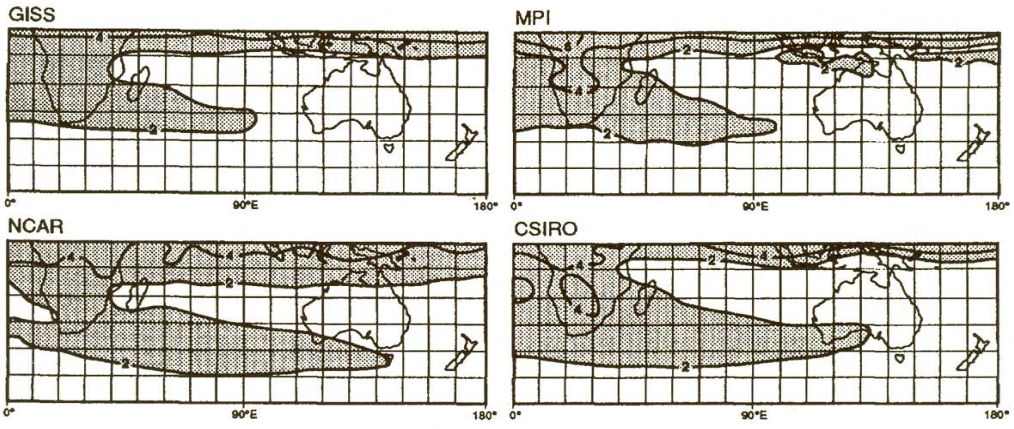

Figure 1.6 GCM transport modeling of carbon dioxide transport in the South Africa Australasian sector of the southern hemisphere for the Goddard Institute of Space Science (GISS), National Centre for Atmospheric Research (NCAR), Marx Planck Institute (MPI) and Commonwealth Scientific and Industrial Research Organisation (CSIRO) models (Rayner and Law, 1995).

therefore of high magnitude and transportation is not only across international boundaries but can also be intercontinental. Fishman *et al.,* (1990) demonstrated that large-scale transport occurs from southern Africa across the West Indian Ocean and that a macro-scale plume of ozone extends over the ocean in winter and spring (Fig 1.5).

The nature and concentration of aerosols and trace gases in the atmosphere plays a large role in determining short and long-term changes in climate. Global and regional atmospheric circulation patterns play a large role in determining the concentrations and long-range transport of pollutants. Atmospheric aerosols have a relatively short residence time as compared to greenhouse gases, but depending on their particle sizes and prevailing atmospheric conditions may be carried over long distances in the atmosphere. For example, in Africa, the presence of mineral dust from the Sahara desert has been reported thousands of kilometers from its source. (Swap *et al,* 1999). Likewise, the transport of continentally-derived material from Africa over the Indian and Atlantic oceans has been established (Swap and Tyson, 1999).

Andreae *et al.*, (1990) and Fishman *et al.* (1991) have shown that aerosols and trace gases may be transported over Africa south of the equator into the tropical South Atlantic Ocean and into the Indian Ocean, and that large-scale recirculation may also occur regionally within parts of Southern Africa. Such long-range transport of pollutants points to the spread of the climatic effects of aerosols even to areas far removed from the source regions. Garstang *et. al.* (1996) have noted that the patterns of aerosol and trace gas

transportation over and out of southern Africa are not as straight forward as was previously envisaged. Recirculation occurs on a massive scale, and the thermodynamic structure of the atmosphere inhibits vertical transport more than was originally expected (Garstang *et al.*1996). Of interest to this paper is that the studies on air mass trajectories during and after SAFARI 92 have shown that approximately 40% of all air parcels that originate over the subcontinent recirculate in an anticyclonic pattern before being transported to the Atlantic (18%) and the Indian Ocean (54%) for all synoptic circulation types (Tyson *et al.*, 1996; Tyson, 1997; Garstang *et al.*, 1996).

Case studies for other regions on the long-range transportation of pollutants in Central and Southern Africa have been carried out (D'Abreton, 1996; Annegarn *et al.*, 1993; Gatebe, 1998, Piketh, 1999, Piketh *et al.,* 1999, Piketh *et al*, 2000, Meter, 2000, Eck *et al.,* 2001, Freiman and Piketh, 2003). These studies have largely revealed that both anticyclonic circulations and westerly disturbances in Southern Africa produce conditions conducive for the transport of aerosols and trace gases away from Africa to the east over the Indian Ocean. Only the easterly wave conditions are likely to favour transport to the west into tropical Atlantic Ocean, and these occur only 4% of the time (Garstang *et al.*, 1996). A case study of air trajectories over the Etosha National Park in Namibia (Garstang *et al.*, 1992), showed that air masses are transported in an anticyclonic nature, and during their movement southeastwards, they pick up further aerosols from all sources, adding to the plume that exits over the Indian Ocean. The Aerosol, Recirculation and Rainfall Experiment (ARREX) has shown that 75% of the recirculated air parcels

exit the continent at approximately 30º S into the Indian Ocean and beyond (Piketh *et. al*., 2002) (Figure 1.7)

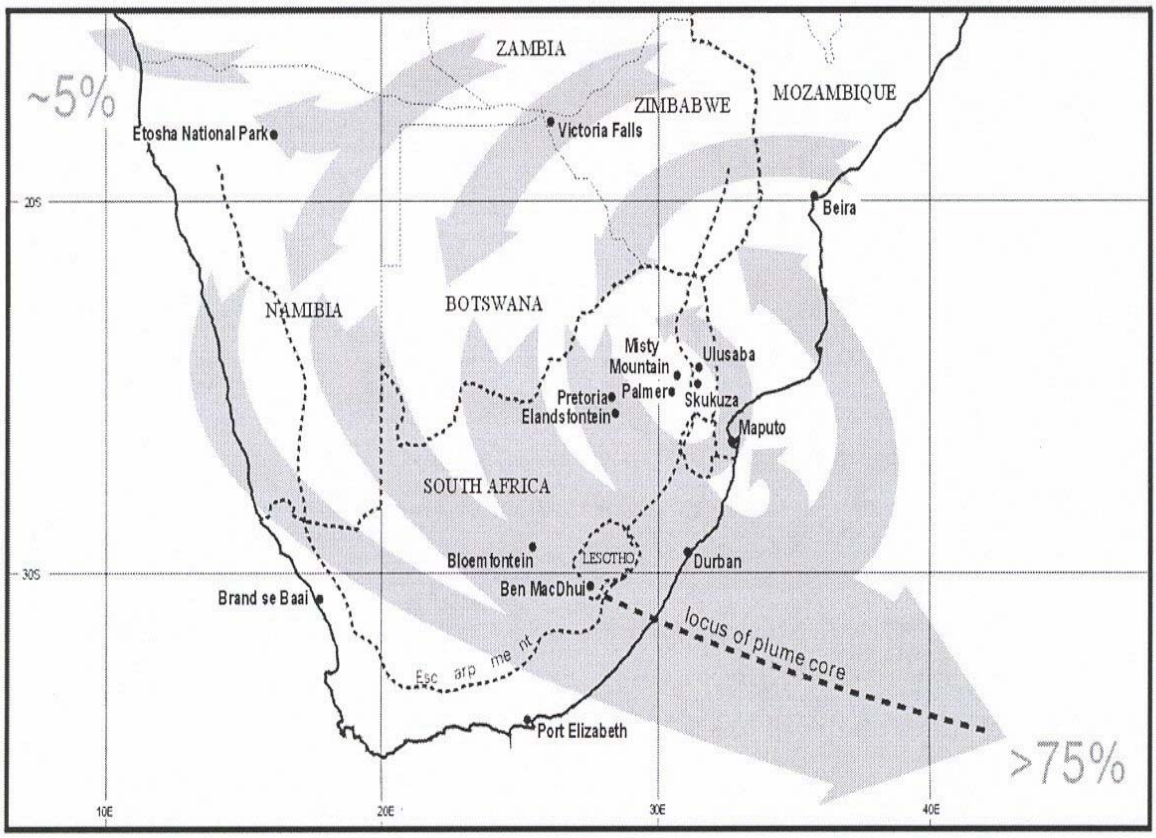

Figure 1.7 Average atmospheric transport pathways over the southern African subcontinent as derived from kinematic trajectories (Piketh *et. al.*, 2002).

Transport through recirculation

Horizontal and vertical dispersion of trajectories from a single point or a cluster of points have been considered using Lagrangian kinematic trajectory modeling (Garstang *et al*., 1996). The method consists of following individual trajectories through vertical meridional and latitudinal planes at specified intervals (Tyson and D'Abreton, 1998). The heights, latitude and longitude of trajectory passage through successive planes are recorded in both eastward and westward directions (Tyson and D'Abreton, 1998). Direct transport is recorded when planes are penetrated directly in the same direction as the initial movement from the source. Recirculation transport occurs when penetrations occur indirectly in a direction opposite to the initial motion from the source, or in the same direction, but in planes behind the source (Tyson and D'Abreton, 1998). Such direct and indirect penetration of planes gives an indication of direct and recirculated transport over a region. The climate of southern Africa is favourable to the recirculartion of aerosols, especially during the winter and austral spring season (Figure 1.8). The transboundary transportation of aerosols from the Copperbelt should be consistent with such observed patterns for the southern African subregion.

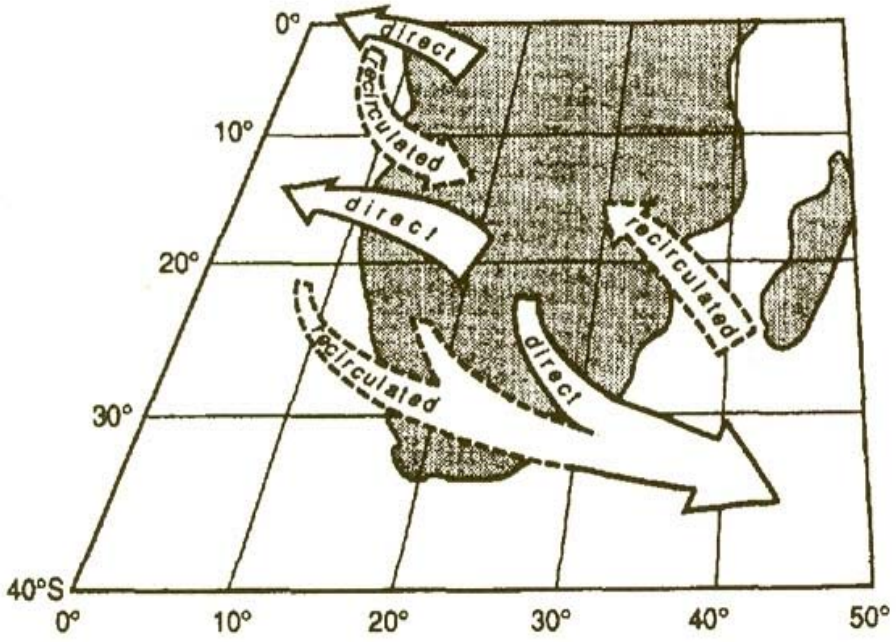

Figure 1.8 Conceptualised direct outflow and recirculating inflow aerosol plumes over the east and west coasts of southern Africa (after Tyson and D' Abreton, 1998).

Sulphate aerosols and radiative forcing

Trace gases and particulate matter in the atmosphere make substantial contributions to atmospheric chemistry, and exert some radiative forcing, from the direct cooling effect from scattering of short wave radiation to the indirect effect of cloud condensation nuclei on cloud properties (Twomey, 1977; Andreae, 1995). The present global average estimate of direct forcing due to anthropogenic aerosols is –0.4 W/m^2 sulphate, -0.2W/m^2 for biomass burning aerosols and –0.1W/m^2 for fossil fuel organic carbon. (IPCC, 2001). The

tropospheric effect of the aerosol forcing factor is comparable in magnitude, but opposite in sign, to the greenhouse effect (Fig.1.8).

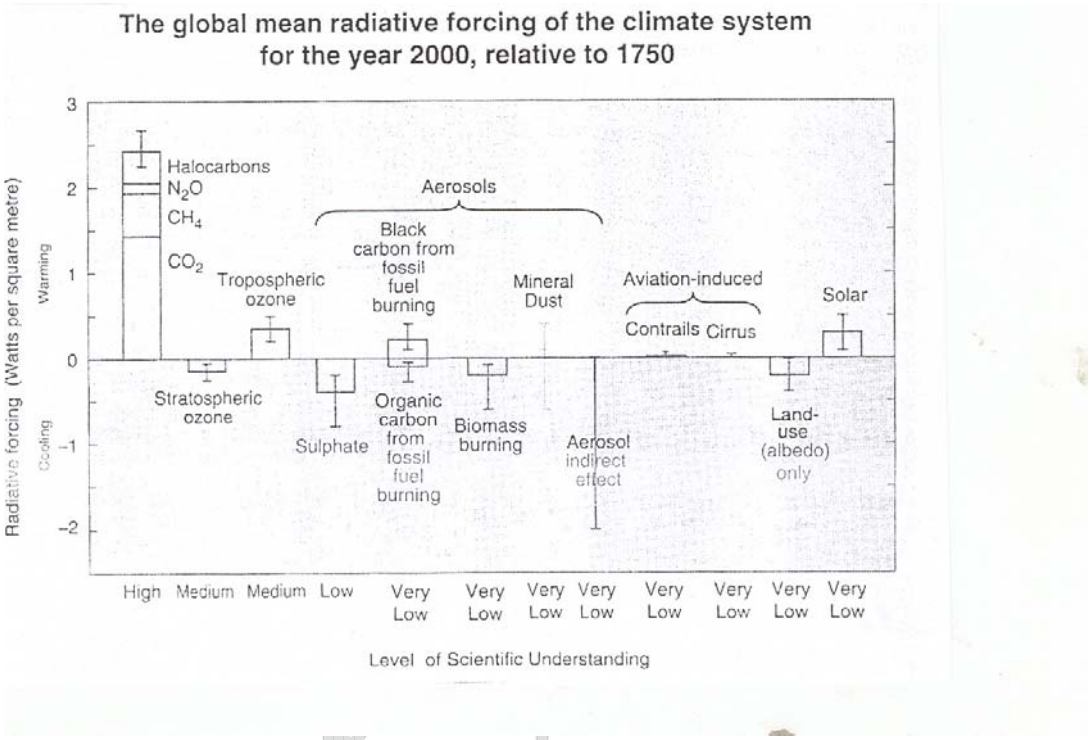

Figure 1.9 Global average estimates of anthropogenic climatic forcing factors due to changes of greenhouse gas aerosol concentrations from pre-industrial times (IPCC, 2001).

The indirect effect of aerosols on cloud condensation nuclei and cloud properties has a shorter period of influence in the Southern African region (Twomey, 1977). The actual indirect effects of aerosols on cloud properties are complex. An increase in the total number concentration of particles in the accumulation mode at a given liquid water content results in a decrease in the mean droplet radius of the cloud, and a corresponding increase in the cloud albedo (Piketh, *et al.* 1999). Charlson *et al.* (1992) calculated that a

30% increase in the cloud droplet number would result in the planetary albedo and a subsequent decrease in the mean surface temperature of about 1.3º C.

The Inter-governmental Panel on Climate Change (IPCC) has noted that regions of high negative forcing coincide with large sulphur emitting sources such as those over Central Africa. (IPCC, 2001). In Central Africa, the main sources of these emissions include the pyrometallurgical processing of copper in the Zambian Copperbelt and the Democratic Republic of Congo (DRC). Sulphate, in its various forms, is a secondary pollutant in the atmosphere. The oxidation of sulphate in the atmosphere can take place either homogeneously or heterogeneously. Homogeneous reactions occur within the same phase, whereas in heterogeneous reactions the processes occur in cloud and fog water, or aerosol droplets involving the transfer of sulphur dioxide and oxidant species to the droplet phase with subsequent liquid phase oxidation, especially during the wet season (Warneck, 1988). The homogenous reaction is thought to be dominant in Southern Africa due to the dry conditions that prevail over the subcontinent for most of the year, with an oxidation rate of sulphur dioxide estimated to be between 3-5% per hour (Piennar *et al.,* 1996). Sulphates have the greatest potential to contributing to climate global aerosol forcing due to their smaller sizes and the fact that they have a longer lifetime in the atmosphere (Charlson *et al.,* 1992). Sulphates are also hygroscopic and therefore have twice the optical scattering per unit mass concentration as dry particles (Charlson *et al.*, 1992).

Besides sulphur pollutants, other important sources of pollutants are those of a carbonaceous nature from biomass burning, and aeolian dust, which is crustally derived. This research will also concentrate on organic aerosols emitted from biomass burning-the second largest source of atmospheric aerosols after sulphur emissions (Andreae, 1977). Satellite imagery shows that biomass burning produces smoke which leads to the formation of smoke clouds (Robbock, 1991). This means that smoke particles absorb incoming short-wave radiation, and are transparent to outgoing terrestrial radiation. This leads to a net cooling of the earth's surface.

This research seeks to determine the seasonality of sulphur emissions from the Copperbrelt and biomass burning products and their potential pathways as determined by regional and synoptic circulation patterns over the sub-region. Kinematic trajectory and the atmospheric vertical structure are used to describe the four dimensional nature of the atmosphere and the extent of direct and recirculated transport of elevated aerosols over Kitwe and the southern African subcontinent. Meridional and zonal walls are created at 2° interval and trajectory hits per grid cell are totaled for each pressure level.

The Zambian Copperbelt as a source of sulphate emissions

Copper production in Central Africa is concentrated on a belt between 10-15° S to 30° E, covering the southern part of the DRC and the northern part of Zambia. This region is the major source of anthropogenic sulphate emissions. Estimates of sulphate emissions for the Copperbelt indicate a decline in emissions from the 1980s (Benkovitz *et al.,* 1996),

whereas there was an increase in emissions from the South African Highveld. Coal combustion for power production was the main cause of this increase in the Highveld. Emission estimates for the Copperbelt follow copper production and its performance in the world market closely. They are also linked to the method used in the refining process. Copper extraction to a sellable product requires ore-beneficiation, either hydro- or pyrometallurgical processing and finally chemical refining (Anon, 1996). The use of the two methods depends on the elements bonded to copper. Because of the addition of sulphur during pyrometallurgical processing, relatively higher amounts of sulphur are released. Sulphur emissions due to this processing, and from bad piping and venting systems are considered to be the major contributors to tropospheric sulphate concentrations (Meter, 2000).

Biomass burning aerosols as climate forcing agents

Many compounds emitted during biomass burning may alter, directly or indirectly, the radiation budget of the atmosphere. While CO_2, NO_x N_2O and NH_3, among others, are directly released by burning processes, ozone is a typical secondary product in these air masses (Helas *et al.,* 1995). Present estimates of emissions from vegetation fires imply that approximately 40% of the global amount of biomass burning takes place in Africa and roughly 40% of that in savanna fires (Andréae, 1991). The contribution of African savanna fires is, therefore, an essential factor in the global atmospheric trace gas inventory.

Human activities over the past century have led to an increase in the extent of biomass burning more than was previously believed. Biomass burning is now recognized as a significant global source of emissions, contributing as much as 40% of gross carbon dioxide and 38% of tropospheric ozone (Levine, 1998).

The major gases produced during biomass burning include many environmentally important gases such as carbon dioxide (CO_2), carbon monoxide (CO), methane (CH_4), oxides of nitrogen (NO_x= nitric oxides (NO) + nitrogen dioxide (NO_2)), and ammonia (NH_3) (Levine, 1998). CO_2 and CH_4 are greenhouse gases, while CO and the oxides of nitrogen lead to the photochemical production of ozone (O_3) in the troposphere. Nitric oxide leads to the chemical production of nitric acid (NHO_3) in the troposphere, which leads to acid precipitation. Particulate matter (usually smaller than 10μm in diameter) comprising of small solid particles of smoke and soot, are also produced during burning and released into the atmosphere (Levine, 1998).

The transportation of biomass burning products

Long-range transport of aerosols and trace gases associated with biomass burning from Africa south of the Equator into the tropical South Atlantic Ocean and south western Indian Ocean has been reported (Andreae, 1990, Levine 1991, Fishman, *et al*., 1991, Pickering *et al*., 1992). There is an accumulation of evidence that biomass burning

products have significant perturbations on atmospheric chemistry, with resultant impacts on the earth's physical and chemical climate (Garstang *et al.* 1996b). Scientific campaigns such as SAFARI-92 and SAFARI-2000 have investigated the emissions from savanna fires in Southern Africa, their transportation across the continent, and the relationship between fires and savanna ecology. Emissions such as CO_2, NO_X and CH_4, and other compounds originating from these fires are suspected to contribute substantially to changes in global biogeochemical processes (Garstang *et al.* 1996b).

Savanna fires in most of western and central Africa are intentional in nature, and result from the activities such as the clearing and burning of tropical forests for various forest and agricultural activities. Many traditional African agricultural systems incorporate annual and biennial burning, which generally occurs during the dry season and then accelerates as the rainy season approaches. Prescribed and natural fires are utilized as management tools for the maintenance of wildlife forage in many national parks and game reserves in southern Africa (Scholes *et al.*, 1996).

Biomass burning products do contribute, albeit minimally, to background aerosol loading of air reaching the country during late winter and spring seasons. Air picks up biomass burning aerosols having passed over the forested areas to the north of 20°S where burning of fossil fuels and wood is prevalent especially during the burning season (August-October). Piketh *et al.* (1999) have estimated that the contribution of biomass burning particulates to the stream of aerosols moving to the Indian Ocean is 1% in summer and reaches a maximum of only 1.2% in winter during the burning season (Piketh *et al.* 1999). Industrial sulphur and aeolian dust are the major constituents of the plume that recirculates and exits the subcontinent at ~25ºS.

South Africa is itself not the major contributor to atmospheric pollution through biomass burning, but other countries in the sub region are (Cahoon *et al.*, 1992; Lindesay *et al.,* 1998). Air masses from the north and northeastern parts of the subcontinent transport a substantial amount of products of biomass burning into the country. Andreae *et al.* (1996) have indicated that the amount of gases emitted from biomass burning may even be comparable to that released from global industrial activities. During the burning months of August-October, air reaching South Africa is most likely to be dominated by biomass burning products. Andreae *et al.* (1997) have also shown that biomass burning accounts for roughly one half of the atmospheric sources of hydrocarbons in southern Africa. Piketh *et al*. (1999) have shown that biomass burning products, together with aeolian dust, marine aerosols and industrial sulphur are the major contributors to total aerosol loading of the lower tropospheric haze layer throughout the year. During the spring season, however, products of biomass burning contribute significantly to the African haze layer between 10° and 20°S (Cahoon *et al.* 1992, Levine *et al.,* 1996, Scholes *et al.,* 1996). However, products of biomass burning have the least contribution to total inorganic-component aerosol loading over the country (Piketh *et al.,* 1999), the highest contributor to total aerosol loading being industrial sulphur, especially during the summer season.

Studies made by the Aerosol Robotic Network (AERONET) as part of the SAFARI 2000 campaign on several sites in Zambia during August-September have yielded insight into the nature and transportation of aerosols in the sub-region. Inter-annual variability in fire

seasonality resulting from meteorological conditions results in variable concentrations of biomass burning aerosols during the dry season and between dry seasons (Eck *et al.*, 2001). Large amounts of burning in the northern parts of the sub-region were observed at all sites in Zambia as a result of human activities. Burning has increased in the last two decades mainly due, in part, to increased human populations. Plumes moving into South Africa from this part of the region are most likely to transport these products of biomass burning into the country.

However, it has been noted that over South Africa, biomass burning appears to be a far less significant source of aerosol production than aeolian dust and industrial processes, except in the immediate vicinity of fires (Tyson and D'Abreton, 1998). High industrial emissions and considerable recirculation beneath and between absolutely stable layers of the atmosphere ensure that industrial aerosols are major contributors to overall background aerosol loadings observed far from source areas (Tyson and D'Abreton, 1998).

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The transportation of sulphate aerosols from copper processing in the Copperbelt and biomass burning products from the burning of savanna fires in the subregion is largely determined by the climatic characteristics of the region. Atmospheric aerosols exert radiative forcing, and are therefore important agents of climatic change. Air masses reaching South Africa from other countries in the subregion are most likely to comprise sulphur pollutants from the processing of copper in the Copperbelt and products of biomass burning from savanna fires. The methodology to be used in determining the transportation pathways of aerosols is described in the next Chapter.