THE TRANSPORT OF POLLUTANTS OVER SOUTH AFRICA AND ATMOSPHERIC SULPHUR IN CAPE TOWN

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Abstract

Cape Town experiences unpleasant pollution (called the Brown Haze) in winter. Sulphur is a major constituent of this haze. To reduce sulphur pollution, and its negative impact on health in Cape Town, air quality management has focussed on identifying local sources and reducing the local emissions of sulphur. Meanwhile, the transport of sulphur pollutants from areas outside Cape Town can contribute to ambient sulphur concentrations. This work studies the transport of sulphur pollutants over South Africa and examines whether Cape Town is a net source of or sink for the pollutant. It shows the link between sulphur emissions on the Mpumalanga Highveld (the most polluted area in South Africa) and sulphur pollution in Cape Town. Two atmosphere chemistry-transport models (RegCM-Chem and WRF-Chem) were used for this study. The models were applied to simulate the atmospheric flow and chemistry transport over South Africa for two years (2001 and 2002), and the results were validated with observations within Cape Town.

The models reproduced observed seasonal variability in atmospheric wind flow and sulphur concentrations over Cape Town. Results from both models show a transport of sulphur pollutants from the Mpumalanga Highveld to Cape Town. While the sulphur pollutants from the Mpumalanga Highveld are transported eastward (toward the Indian Ocean) at middle atmospheric levels, the pollutants are transported south-westward (towards Cape Town) at lower atmospheric levels. In addition, the pollutants are transported from the Mpumalanga Highveld to Cape Town, following the south coast of South Africa, in April. During an extreme sulphur pollution event in Cape Town, there is formation of either a col or a converging flow over the city. These features encourage the accumulation of sulphur over Cape Town. The sulphur flux budget analysis shows that Cape Town can be a net source of or sink for sulphur during an extreme pollution event. This study has application potential in developing policies to reduce sulphur pollution in Cape Town and in other areas of South Africa.

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Chapter 1: Introduction

Poor air quality is a major problem that affects many parts of the world. It damages the environment and human health in industrial areas (like the Mpumalanga Highveld) and in urban centres (like Cape Town). Sulphur dioxide (SO_2), in particular, is considered the most widespread pollutant that threatens environmental and human health (Atkinson & Faith, 1972; Boubel *et al.*, 1996). SO_2 damages human health because it is highly soluble and dissolves on the mucous membranes. Prolonged human exposure to high concentrations of SO_2 may cause respiratory and cardiopulmonary morbidity (Pope *et al.* 2002). This highly soluble pollutant is the main precursor of sulphate (SO_4), which has a longer residence time in the atmosphere. SO_2 also forms hydrogen sulphate (H_2SO_4) that precipitates as acid rain. Acid rain leads to the acidification of soil and water. In addition, it corrodes metals, paintwork and building stones, and inhibits vegetation growth and reproduction (de Kok *et al.* 2007).

To adequately manage air quality through emission controls, it is necessary to identify the sources of the anthropogenic air pollutants. This is no simple task because, in the atmosphere, some pollutants are transformed to form secondary pollutants, and pollutants are dispersed, transported and deposited at a range of scales. This work studies the transport of sulphur pollutants over South Africa and examines whether Cape Town is a net source of or sink for the pollutants.

1.1 The City of Cape Town

The city of Cape Town is a hub of production and transport connecting international, local and inland transport by sea, air, rail and road. It is located on the south-western tip of South Africa at approximately 34°S and 18°30'E, and the surrounding metropolitan area extends over roughly 500 km^2. Like many major cities in developing countries, Cape Town has a growing population (2.89 million in the 2001 to 3.4 million in 2007 (StatsSA 2003; StatsSA 2009)), and local authorities face challenges of providing amenities and encouraging economic growth while protecting the integrity of environmental health, especially from the emission of air pollutants.

Cape Town has a Mediterranean climate with dry, warm summers and cold, wet winters. Summer winds are predominately a strong, cooling, south-easterly flow from the Indian

Ocean that brings some low-level orographic cloud across the Table Mountain complex and is associated with fine weather. Winter weather is characterised by strong north-westerly winds from the Atlantic Ocean and a procession of cold precipitation fronts from the west. The mean winds are driven by the synoptic scale circulation, most commonly the location of the quasi-stationary South Atlantic anticyclone and, in winter, the passage of westerly wave disturbances. These features are discussed further in Chapter two.

1.2 The problem of atmospheric sulphur in Cape Town

The problem of poor air quality has long been a concern in Cape Town. The most well-known air quality problem for Cape Town is the Brown Haze, a layer of pollutants that extends from the surface for a distance into the atmosphere, and that obscures visibility during winter. The Brown Haze, which occurs frequently, is visible evidence of poor air quality in the city. Atmospheric sulphur (sulphur dioxide and its secondary pollutants) contributes to the Brown Haze (Wicking-Baird, De Villiers, and Dutkiewicz 1997). Studies have linked enhanced concentrations of SO_2 in the city with the increased incidence of asthma in local school children (White *et al.*, 2009) and with an increase in morbidity and mortality in Cape Town, where high rates of infection of tuberculosis increase the vulnerability of respiratory health (Batterman, Cairncross, and Huang 1999; Wichmann and Voyi 2012). Hence, how to reduce the impact of atmospheric sulphur on environment and health is a major concern in Cape Town, and local authorities are engaging in various activities to reduce local emissions. The City initiated two Brown Haze studies. The first study, led by the Energy Research Institute, aimed to identify the sources of the visible haze (Wicking-Baird, De Villiers, and Dutkiewicz 1997), while the second study, led by the Climatology Research Group (Piketh *et al.* 2004), aimed to investigate the properties of the haze, so as to assess the threat to health and to inform abatement efforts. In addition, the City monitors ambient SO_2 concentrations on an ongoing basis and promotes the reduction of SO_2 emissions by the regulation of high emission activities, engagement with local industry to employ cleaner technology (of fuel and process), and interventions like roadside checking of diesel vehicle emissions (Muchapondwa 2010). The City also promotes the use of cleaner energy and has created a market for renewable source electricity (City of Cape Town 2012).

In Cape Town, SO_2 emissions have largely been attributed to diesel vehicles and industry (Wicking-Baird, De Villiers, and Dutkiewicz 1997). The oil refinery located near the coast to the north-west of the city centre is a source of concern, as children in nearby schools suffer an

increased incidence of asthma associated with elevated levels of ambient SO_2 (White *et al.* 2009). Other local sources of concern include a glass production factory in the suburb of Bellville and fuel combustion in townships (Walton 2005). However, since sources of atmospheric sulphur can be both local and remote, it is possible that the accumulation of sulphur pollutants transported from areas remote to Cape Town might be the main cause of pollution over the city. Hence, the study investigates whether Cape Town is a net producer or recipient of atmospheric sulphur.

1.3 Aim and objectives

The main aim of this dissertation is to study the transport of sulphur pollutants over South Africa and to investigate whether Cape Town is a net source of or sink for the pollutants, especially during the extreme pollution event. The objectives of study are to:

- study the observed annual and seasonal variations of SO_2 in Cape Town and investigate the role of meteorological conditions on the variations;
- use two atmospheric-chemistry models in simulating and studying the transport of sulphur pollutants over South Africa;
- evaluate the ability of the models in simulating the characteristics of sulphur over Cape Town; and
- calculate sulphur flux budget over Cape Town in examining whether the city is a net sink or source for the sulphur transport.

The dissertation is divided into seven chapters. Chapter 2 presents a literature review on the characteristics of sulphur and air quality studies in South Africa, and Cape Town in particular. Chapter 3 details the methodology of the study, including the description of the observed data, the atmosphere chemistry-transport models and their set-up for the model simulations. Chapters 4 to 6 present and discuss the results of the study; Chapter 4 - the characteristics of the observed sulphur, Chapter 5 - the capacity of the models to simulate the characteristics, and Chapter 6 - the simulation results to discuss transport of sulphur pollutants over South Africa and to make evident the link between sulphur pollution in Cape Town and the transport of sulphur pollutants from the Highveld. Chapter 7 gives the concluding remarks and suggestions for future studies.

Chapter 2: Literature review

This chapter reviews findings from previous pollution studies over South Africa. The review focuses on the characteristics of sulphur pollutants, the transport of sulphur pollutants from Highveld to other regions in South Africa, the meteorological conditions that produce poor air quality in Cape Town, and the modelling of the transport of pollutants over Cape Town.

2.1 Characteristics of sulphur in the atmosphere

Sulphur pollutants occur in the atmosphere as part of a natural sulphur cycle, where sulphur constituents are transformed between different phases and compounds. The most commonly released compounds are sulphur dioxide (SO_2) and dimethysulfide (DMS). SO_2 is released by biomass burning and from geothermal sources such as hot springs and volcanic activity, while DMS is released from the ocean (de Kok *et al.* 2007). However, various studies (WHO 2005; Norman *et al.* 2007; Pope 2000) have shown that natural emissions of sulphur are rarely associated with damage to health or the environment, but that anthropogenic emissions of sulphur are vast and are associated with damage to the built and natural environment, and to human health. The major source of anthropogenic SO_2 emission is the combustion of sulphur-bearing fuel (especially coal and oil) in energy production, oil and petrol refineries, metallurgical works, and motorised vehicles. The highly reactive and soluble nature of SO_2 makes it spread easily in the atmosphere, and SO_2 is known as the most widespread pollution threat to environmental and human health (Boubel *et al.* 1994; Atkinson & Faith 1972).

In the atmosphere, SO_2 is an unstable gas and it readily oxidises to any number of compounds (McElroy 2002; Atkinson and Faith 1972). It is transformed by oxidation and can follow a complex chain of reactions in the aqueous or gas phase. SO_2 is most commonly transformed to sulphate (SO_4), which has a longer residence time than SO_2 in the atmosphere. SO_4 is also released in combustion processes, but in small amounts in comparison to SO_2. The transformation of SO_2 to SO_4 is by oxidation reactions (McElroy 2002; Husain and Dutkiewtcz 1992) which can generally be simplified as:

Gas-phase oxidation $\quad SO_2 + 2OH \Rightarrow H_2SO_4 \Rightarrow SO_4 + H$

Aqueous-phase oxidation $\quad SO_2 + H_2O_2 \Rightarrow SO_4 + 2H$

SO_2 and SO_4 may subsequently form hydrogen sulphate (H_2SO_4), or sulphuric acid as it is known in its aqueous phase, to precipitate as acid rain. In the built environment, the high acidity of sulphuric compounds corrodes metals, paint and building stone (Boubel *et al.* 1994). In the natural environment, its acidifying effect poses a threat to ecology (Pullin 2002). Elevated levels of SO_2 in soil and water may lead to crop failure, as the increase in acidity has been shown to inhibit development of vegetation and the root production of maize (Emberson *et al.* 2001), the staple food in South Africa.

In contrast to SO_2, SO_4 is chemically stable and less reactive than SO_2. SO_4 is not observed by air quality monitoring as it is often coated by fine dust, which prevents it from growing by coalescence to a discernible size (Korhonen *et al.* 2003); thus the dust-coating of SO_4 renders the method of monitoring SO_2 ineffective for monitoring SO_4, because the method is based on the scattering of ultraviolet light by sulphur. However, the dust-coated SO_4 can be observed within observations of particulate matter (PM) as a whole (Chow *et al.* 1993).

The concentrations of SO_2 and SO_4 in the atmosphere exhibit diurnal and seasonal patterns because atmospheric conditions and solar radiation influence the removal of SO_2 and SO_4 from the atmosphere by the production of secondary pollutants. For example, the transformation of SO_2 to SO_4 is increased by an increase of sunlight and water vapour. Various studies (Josipovic *et al.* 2010; Pienaar *et al.* 2011; Meagher and Bailey 1983) have shown that the near-surface concentration of SO_2 is higher in winter than in summer; in contrast, the concentration of SO_4 is higher is summer than in winter. Similarly Scheifinger and Held (1997) found a higher concentration of SO_4 during the day (noontime) than at night. SO_2 and SO_4 are also removed from the atmosphere by direct deposition.

Seasonal deposition rates are explained in part by changes in mean wind regimes, atmospheric stability (Zunckel *et al.* 2000), vegetative cover and the availability of water (owing to the soluble nature of the pollutant). Deposition is described as dry deposition when pollutants settle on surfaces of land, water, buildings and vegetation, or as wet deposition when pollutants are removed from the atmosphere by precipitation (below clouds) or by scavenging (within clouds). The relative rates of wet and dry deposition under varying conditions are complex and have been investigated by Held et al, (1996) and Zunckel *et al.* (2000) over the Mpumalanga Highveld.

2.2 The transport of sulphur pollutants from the Highveld

Previous studies (Scheifinger and Held 1997; Tyson, Kruger, and Louw 1988; Zunckel *et al.* 2000) have shown that the Mpumalanga Highveld is the main source of anthropogenic pollutants in South Africa. The Mpumalanga Highveld, which is located on the plateau in the north-eastern part of South Africa, is amongst the most polluted areas in the world (Munnik 2010). Various studies attribute the source of the pollution in Mpumalanga Highveld to emissions from the mining, processing, and production activities attracted by the rich deposits of ore and coal in the area. In particular, SO_2 emissions are generated by power plants, each of which are fuelled by a dedicated mine and which burn low-grade, high-sulphur-content coal to jointly produce more than 90% of the South Africa's electricity (NER(SA) 2006; Winkler 2009). The area has long suffered severe air pollution. Although, since the 1970s industrial chimney-stack heights have been elevated to encourage dispersion away from the area, Annegarn *et al.* (1996) showed that this strategy only increased the area affected by this pollution, as increased stack heights facilitate the transport of these emissions across the southern Africa region.

Using radiosonde observation, Cosijn and Tyson (1996) showed that vertical mixing over the escarpment (and thus the Mpumalanga Highveld) is influenced by three layers of atmospheric stability. These layers are formed under subsidence imposed by the descending limbs of the Ferrell and Hadley circulation systems. The layers around 300 hPa and 500 hPa prevail throughout the year, while the layer at 700 hPa is intermittently disrupted, releasing pollutants to be trapped between 500 and 700 hPa, or (as more rarely occurs) if the 500 hPa layer is disrupted between 500 and 300 hPa. This explains the haze layer observed over the escarpment around 500 hPa in the aerial Southern African Fire-Atmosphere Research Initiative in 1992 (SARARI-92) (Stein *et al.* 2003). Swap and Tyson (1999) showed that the prevalence of these layers, under the mean anticyclonic circulation, is a highly effective control on vertical mixing.

A fourth layer of atmospheric stability was found by Cosijn and Tyson (1996) around 850 hPa. This lower layer occurs over the thin belt of land between the escarpment and the coast, and to intercept the edge of the escarpment to the south and east (Cosijn and Tyson 1996; Freiman and Piketh 2003; Garstang *et al.* 1996). These stable layers influence the transport of pollutants, as observed by the TRACE-A (Transport and Atmospheric Research Chemistry

near the Equator - Atlantic) campaign in the decoupling of air above (generally westerly) and below the haze (Tyson 1997).

In their trajectory study, Freiman and Piketh (2003) also found a decoupling in the atmosphere between 600 and 700 hPa. Tyson (1997) found that these stable layers have seasonal height, because they are elevated by surface heating in summer. Garstang *et al.* (1996) showed that the stable layers tilt upward towards the north and the east of the country, and downwards to the west and south, with important implications for the transport and recirculation of pollutants.

Previous studies (Freiman & Piketh, 2003; Tyson *et al.*, 1996) showed that the main transport of pollutants from the Mpumalanga Highveld is eastward to the Indian Ocean and to a lesser extent westward in summer to the Atlantic Ocean, and that air is recirculated at various scales over the subcontinent (Garstang *et al.*, 1996; Tyson, *et al.*, 1996). Tyson *et al.*, (1996) used the classification of synoptic charts for the study, while Garstang *et al.* (1996) used trajectory analysis. Tyson *et al.* (1996) describes four major synoptic circulation types driving the transport of aerosols over the region, the two dominant types being semi-permanent, subtropical, continental anticyclone, which is present 70% of the days in the year; and the transient, mid-latitude, ridging anticyclone, which is observed 10 to 15% of days in the year, mainly in July. The studies showed that westerly wave disturbances produce transport of pollutants eastward to the Indian Ocean in winter, while the barotropic, quasi-stationary, tropical easterly disturbances transport pollutants from the northern parts of the country westward to the Atlantic Ocean in summer. The period with most mass flux to the Indian Ocean is May to August. Garstang *et al.* (1996) suggested the dominance of continental anticyclone (especially in winter) and also highlighted pollutant recirculation as a major transport pattern.

Prior to the discovery of the absolute stable layers, it was believed that emissions from the Highveld were either deposited locally over a small area or transported eastward to the Indian Ocean (Piketh *et al.* 2002). However, the discovery of the stable layers, and subsequent studies, showed that emissions of SO_2 from the Highveld area were actually deposited over a larger area around the Highveld and even further away. For instance, in studies reviewed by Tyson *et al.* (1988), the area thought to be affected by emissions from the Mpumalanga Highveld (then known as the Eastern Transvaal) was within 600 km^2 around the area;

however, subsequent work by Scheifinger and Held (1997) showed that a larger area around the Highveld was affected. Industrial emissions of sulphur have been shown by trajectory modelling and by chemical tracer analysis to be transported as far as Kenya (Tyson and Gatebe 2001) and Ascension Island respectively (Tyson, 1997). Hence, it is now understood that atmospheric sulphur may be transported for minutes to days, and across a short distance to thousands of kilometres (Freiman & Piketh, 2003; Piketh & Watson, 2004; Zunckel *et al.*, 2000), and that the effects of SO_2 emissions from the Highveld are experienced not only locally. However, no studies have considered how sulphur pollution in Cape Town may be linked with emission of SO_2 from the Highveld, and that is the focus of this study.

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2.3 The meteorology of air quality in Cape Town

Previous studies have identified and discussed various meteorological processes that favour poor air quality in Cape Town.

2.3.1 Atmospheric inversion

An atmospheric inversion occurs when warm air is trapped below cooler air, thus inhibiting vertical mixing. This favours poor air quality, as emissions released near the Earth's surface are prevented from dispersing upwards. In Cape Town, the atmospheric inversion is documented as a key process for the occurrence of the Brown Haze pollution over the city. Keen (1979) observed the Brown Haze is more commonly associated with surface radiation inversions than with the elevated subsidence inversion. Later, Jury *et al.* (1990) also associated pre-frontal inversions, where cold, wet air in a westerly disturbance moves over warmer air, with pollution episodes. However Wicking-Baird *et al.* (1997), in agreement with Keen (1979), found that the surface inversion, which traps pollutants after long, clear nights in winter, is most associated with the Brown Haze.

2.3.2 Berg winds

Berg winds are slow, dry, off-shore air flows which are conducive to poor air quality. They may flow over cool air and prevent vertical mixing (in a similar fashion to an inversion). The slow movement of Berg winds is also conducive to the accumulation of pollutants. Jury *et al.* (1990) found Brown Haze days in Cape Town to be associated with north-easterlies or berg winds from the arid interior. These winds are brought about over Cape Town under anticyclonic flow ahead of a westerly disturbance (cold front), or by the development of a coastal low (a shallow low pressure system that forms along the west coast of South Africa and which then moves along the coast either to the north or to the south). Berg winds blow for hours to days in late winter and early spring (Preston-Whyte and Tyson 1989), and their low ventilation potential is believed to accumulate pollution in Brown Haze episodes.

2.3.3 Mesoscale and synoptic scale features

Keen (1979) showed that different topographically induced mesoscale features (like the urban heat island, land breeze, and sea breeze) can cause pollutant accumulation in Cape Town. Jury *et al.* (1990) investigated synoptic and mesoscale features that favour air pollution over Cape Town and found that the mesoscale features that induce pollution in Cape Town only form under certain synoptic conditions. For instance, these authors showed that

the ridging of the South Atlantic anticyclone over Cape Town (or over the south-western tip of South Africa) commonly preceded pollution episodes. In addition, they also showed that, under stagnant synoptic conditions, thermal and topographically induced mesoscale features are apparent. Expanding on the study of Jury *et al.* (1990), Stein *et al.* (2003) reviewed studies of pollution meteorology over South Africa and found that in the presence of weak high pressure circulation or col (a pressure saddle or ridge between two high pressure cells), and in the absence of westerly disturbances, local mesoscale features dominate. Hence, these synoptic conditions favour air pollution in Cape Town. In addition, Stein *et al.* (2003) concluded that layers of atmospheric stability created by prevailing anticyclonic conditions commonly trap pollutants below 700 or 850 hPa (within the 1 to 1.5 km above sea level) over coastal areas. These synoptic conditions also create conditions for the recirculation of air and pollution as described by Keen (1979).

On the other hand, low pollutant concentrations in Cape Town are associated with strong winds and cold fronts. The south-easterly wind, commonly called "the Cape Doctor", ventilates the area in summer. In winter although pre-frontal inversions of westerly disturbances concentrate pollutants, Wicking-Baird *et al.* (1997) showed that a front itself is associated with non-haze days. Fronts have a cleansing effect on the atmosphere, as precipitation dissolves and coalesces with pollutants in the atmosphere as 'wash out'. Behind the front, mixing height is increased and flow is southerly, affording good dispersion (Preston-Whyte & Tyson 1989). Summer is a period of lower air pollution potential in Cape Town, as winds are stronger and increased heating of the atmosphere lifts the boundary layer, thus promoting convective mixing (Preston-Whyte and Tyson 1989).

Both synoptic and mesoscale features have been used to explain the meteorology of air quality. Under positive synoptic conditions and atmospheric stability, mesoscale features enhance the accumulation of pollutants. However, no study has investigated the synoptic conditions that favour the transport of pollutants from remote sources (like the Mpumalanga Highveld) to Cape Town.

2.4 Modelling the transport of atmospheric sulphur over South Africa and Cape Town

The problem of insufficient monitoring stations has limited the observational approach in the investigation of how far SO_2 emissions from the Mpumalanga Highveld are transported. This

is because observations have been limited to the area within hundreds of kilometres from the Mpumalanga Highveld (Held and Mphepya 2000; Pienaar *et al.* 2011; Zunckel *et al.* 2000; Josipovic *et al.* 2010). The application of pollution models offers an alternative approach to study the transport of pollutants from the area to other areas in South Africa, or even the world.

Many studies have applied trajectory models, based on the approximate calculation of the transport pathway of a small air parcel, which were consistently used for studies until the late 1990s. Tyson and D'Abreton (1998) used a Langrangian kinematic trajectory model with Light Detection and Ranging (LIDAR) remote sensing observation in comparison with a global climate model (GCM) simulation. They found the GCM simulation lacked a sufficiently high resolution to represent synoptically driven pollutant recirculation over South Africa. Piketh *et al.* (2002) used validated trajectory model simulations and observations from the Ben MacDhui High Altitude Aerosol and Trace Gas Transport Experiment (BHATTEX) in Lesotho (Tyson and D'Abreton 1998) to investigate the extent to which pollutants are transported from industrial emission sources. They found that the level of accuracy of trajectory models in simulating the extent of transport was adequate. In addition, they found that the stable layer at 700 hPa was the most important layer in facilitating the transport.

Freiman and Piketh (2003) employed an ensemble trajectory method to study the transport of pollutants from the Mpumalanga Highveld, and their results broadly agreed with those of Garstang *et al.* (1996). The results showed that 45% of pollutants from the Highveld are transported over the South Indian Ocean and Indian Ocean, and that 33% is recirculated in the atmosphere for two to nine days. In addition, they found that the transportation of pollutants to the South Indian Ocean occurs at all heights, but mostly between 500 and 700 hPa. Using a trajectory model, Freiman and Piketh (2003) found that the wind over the Highveld is mainly northerly and decoupled between 700 and 600 hPa. This decoupling is in agreement with observation in the South African Fire-Atmosphere Research Initiative (SAFARI-92) (Tyson 1997).

Van der Merwe (1998) used a trajectory model to investigate the dispersion of SO_2 over the Highveld and to assess the distribution of SO_2 and SO_4 under the major circulation. The author found that the area affected by the emissions was greater than previously thought, up to 600 km from the source. Freiman and Piketh (2003), however, argue that trajectory

models are of limited use under turbulent conditions, and that these models do not incorporate equations to account for the chemical reactions between the pollutants.

Eularian pollution models overcome this shortcoming of trajectory models. Zunckel *et al.* (2000) used a Eularian model, the Multi-scale Atmospheric Transport and Chemistry model (MATCH) to investigate sulphur deposition, and showed that the results agreed with observations. Although, the MATCH model failed to accurately predict quantities of the pollutant, the modelling method was found to be satisfactory for showing the pattern of deposition (and also of transport). Zunckel *et al.* (2000) concluded that concentrations of atmospheric sulphur in any location in southern Africa may be linked to emissions from other parts of the region. However, over the past decade, the Eularian chemistry-transport or dispersion models have become more sophisticated. The MATCH model was limited to only two land-use categories, land and sea. The latest atmosphere-chemistry models have more vertical levels, more land-use categories, and include parameters for the topography and equations for chemistry to take into account the changing conditions of the atmosphere.

Some studies have applied advanced Eulerian models to study pollution over Cape Town. As part of the Khayelitsha Air Pollution Strategy initiated in 1997 (Muchapondwa 2010), the Cape Town Air Quality Monitoring Group employed an atmosphere chemistry-transport model, in addition to ambient monitoring, to study the problem of continually high levels of particulate matter with a diameter of less than 10 microns (PM10). Zunckel *et al.* (2004) developed a photochemical dispersion model, the Dynamic Air Pollution Prediction System (DAPPS) intended for the prediction of ambient pollution based on local emissions sources and meteorological forecasting. The project was however abandoned owing to a lack of funding (personal communication, Eugene Cairncross).

Walton (2005) used a Eularian model to study the contribution of major local sources to the Brown Haze and to investigate meteorological conditions that contributed to Brown Haze formation. However, the focus of Walton (2005) was on the influence of local emissions on Cape Town pollution. Eularian models can be run at a regional scale to incorporate synoptic conditions and provide an ideal tool to investigate the source of pollutants that may be remote as well as local in origin. This study adopts two advanced Eulerian models to study the transport of pollutants from the Mpumalanga Highveld and atmospheric sulphur in Cape Town.

Chapter 3: Methods

3.1 Observed data

This study used meteorological and pollution observations from five City of Cape Town Air Quality monitoring network stations (Figure 1b). The stations measure ambient concentrations of SO_2 and other pollutants hazardous to human health and ecology (City of Cape Town 2005), as well as temperature and wind speed. The stations are located in the areas of Bellville South, Bothasig, City Hall, Goodwood and Tableview. Although SO_2 is also measured at Killarney and Athlone, data from these stations are not appropriate for a study of ambient air quality, as Killarney is very near the oil refinery and considered to indicate source emissions, and the station at Athlone is used for calibration purposes only.

Depending on the seasonal wind direction, all sites are at times on the downwind side of various local emission sources. Bellville South is an area of mixed residential, commercial and industrial land use. The major source of SO_2 emissions is a glass manufacturing factory (Walton 2005). Goodwood is a mixed residential and commercial area with nearby industry to the south-east and south-west. The nearby national road, the N2, carries commuter traffic from Cape Town's northern suburbs to the City, and another busy national road, the N7, passes along the south side of this area. Aircraft traffic at Cape Town International is another source of SO_2 emissions and the monitoring stations at Bellville South and Goodwood are the nearest in proximity. The City Hall station is adjacent to the city's busy taxi rank, bus station and rail terminus, hence vehicular and rail transport are the main source of SO_2 emission, especially during the morning and evening commute.

Vehicular emissions are a lesser source near the Bothasig and Tableview monitoring stations, as they are located along arterial roads. These stations are in proximity to each other and are in residential areas that have grown alongside industrial areas. The major nearby source of SO_2 is the local oil refinery, Caltex, which was commissioned in 1966, when settlement nearby was sparse. Seasonal wind for Cape Town is known to be south-east in summer and north-west in winter, and Bothasig is thus mainly downwind of Caltex in winter and Tableview is downwind in summer. Another notable source of SO_2 in the area is the waste-water treatment plant in Potsdam, an area neighbouring Tableview.

Dispersion of pollutants between the stations may vary owing to the topography and proximity to the coast. Bothasig and Tableview are located near the coast and are surrounded by flat topography; hence these stations are likely to experience enhanced dispersion. The topography at the City Hall station is least favourable for dispersion. Although the site is near the coast, the area is on the leeward side of the Table Mountain complex and at times pollutants accumulate in the area. The topography around Bellville South and Goodwood is flat to undulating and conducive to dispersion, although further from the ventilating effect of winds off the ocean.

The data is comprised of hourly average concentrations of SO_2, the wind speed in metres per second (m s^{-1}), and temperature in degrees Celsius (°C) from January 2001 to December 2009. The data is analysed to identify patterns of temporal variation in concentrations and corresponding causes in patterns of meteorological data. Diurnal variation is analysed starting at 0000 SLT (standard local time), then average monthly observations are used to identify seasonal variation. Finally, the annual averages are analysed to identify any inter-annual trends.

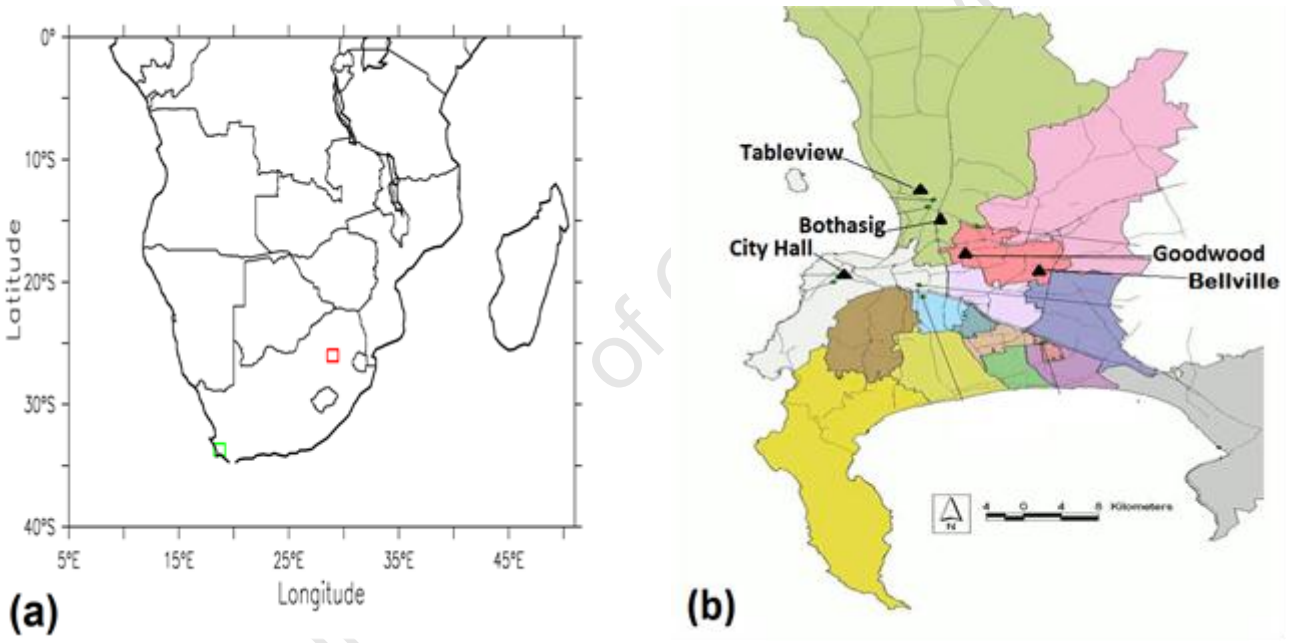

Figure 1: Study domain: (a) Map of southern Africa showing Cape Town area (green box) at the south western tip of South Africa and Mpumalanga Highveld area (red box) in the interior; (b) The City of Cape Town air quality network showing the location of observation stations. The location of the five stations (Bellville South, City Hall, Goodwood, Bothasig and Tableview) used in the study are indicated with triangles (Δ). source: www.capetown.co.za with some modifications

3.2 Models description and set-ups

For this work, two atmospheric-chemistry models, RegCM-Chem and WRF-Chem, are used to simulate the climate and pollution transport over southern Africa. Both models allow online coupling of atmospheric and chemistry parameters, and their climate components have been tested over Southern Africa (Sylla *et al.* 2009; Crétat *et al.* 2011). The major difference between the dynamics of the two models is that RegCM-Chem is a hydrostatic model, while WRF-Chem is a non-hydrostatic model. This means RegCM-Chem uses approximations for some transfer in the vertical and thus uses less computational power. Using two models provides the opportunity to compare results and assess the degree of inter-model variability.

3.2.1 RegCM-Chem

The International Centre for Theoretical Physics (ICTP) Regional Climate model with chemistry (RegCM-Chem), as described in Pal *et al.* (2007) and Giorgi and Anyah (2012), is a hydrostatic, sigma-coordinate following model. This study uses the most recent version, RegCM-Chem version 4, and from the various options for physics and chemistry parameterisations, the following parameterisation schemes are selected: the community climate model version 3 (CCM3) scheme (Kiehl *et al.* 1996) for radiation, the mass-flux cumulus scheme of Grell *et al.* (2005) with Fritsch and Chappell (1980) closure, and the planetary boundary layer parameterisation of Holtslag and Boville (1993). Land surface–atmosphere interactions are treated using the Biosphere-Atmosphere Transfer Scheme 1E (BATS 1E) (Dickinson *et al.*, 1993) which is based on Monin Obukhov similarity relations (Monin and Obukhov 1954).

For the chemistry, the photochemical Carbon Bond Mechanism-Z (CBM-Z), (Zaveri and Peters 1999) is selected. Photolysis is based on the Tropospheric Ultraviolet-Visible Model (TUV) scheme developed by Madronich and Flocke (1999) (the Madronich scheme (1987) for WRF-Chem). For dry deposition, RegCM-Chem uses the Community Land Model 4 (CLM4) developed after Wesley (1989), and wet deposition follows the Model for Ozone and Related chemical Tracers [MOZART global model (Emmons *et al.* 2010)]. Wet removal in the atmosphere (scavenging in clouds) is not included in RegCM-Chem. For a detailed discussion of the gas-phase chemistry in RegCM-Chem, please refer to Shalaby *et al.* (2012).

The RegCM-Chem simulation is set up with a 35 km horizontal resolution. The domain is centred at 33°S and 24°E. It extends with the Lambert conformal projection from 16.62°W to

54.41°E across 189 grid points, and from 10.5°S to 40.45°S across 97 grid points. In the vertical, the domain spans 18 levels, sigma following near the surface and more flat towards the top of the model. Initial and lateral boundary meteorological conditions were provided by ERA-Interim 1.5° x 1.5° gridded reanalysis data from ECMWF (European Centre for Medium-Range Weather Forecasts) and emissions data with a 1° x 1° resolution was provided by the RCP (Representative Concentration Pathways) emissions dataset (IIASA 2012).

3.2.2 WRF-Chem

The Weather Research and Forecasting model, WRF-Chem version 3.2, as described by Skamarock *et al.* (2005), and by Grell *et al.* (2005), is non-hydrostatic. Similar options to those used in setting up RegCM-Chem are selected where possible, namely the RRTM (Rapid Radiation Transfer Model) longwave radiation scheme (Mlawer *et al.*, 1997), the Dudhia (1989) shortwave radiation scheme, the Grell 3d ensemble cumulus scheme (which expands on Grell and Devenyi (2002), and the Yonsei University scheme (YSU), (Hong *et al.*, 2006) for the planetary boundary layer. The photochemical Carbon Bond Mechanism-Z (CBM-Z) (Zaveri and Peters 1999) is selected and photolysis is based on the Madronich scheme (Madronich, 1987). Dry deposition is resolved using the ''flux-resistance'' method (Wesley, 1989), except for the deposition of sulphates, which follows (Erisman, Van Pul, and Wyers, 1994). The wet scavenging option is turned off and same time-step of 2.5 minutes is used for both the chemistry and meteorology components.

The WRF-Chem simulation is run at a 35 km horizontal resolution. The domain was also centred at 33°S and 24°, again with a Lambert conformal projection. The grid extends from 16.69°W to 56.1°E across 188 grid points, and from 12.82°S to 37.54°S across 96 grid points. The vertical is divided into 33 levels, also sigma following near the surface, and each level is flatter than the one below. Initial and boundary conditions are also provided by 1.5° x 1.5° gridded reanalysis data from ECMWF, while emissions data at a 1.5° x 1.5° resolution are provided by Edgar version 4.2 (European Commission Joint Research Centre 2011).

It is imperative that suitable meteorological and emissions data is selected for modelling studies. To this end the ECWMF meteorological data has been assessed to be suitable in pollution transport (D'Abreton 1996; Piketh *et al.* 2002) and climate studies over southern Africa (Sylla *et al.* 2009).

The emissions data-sets are used globally for pollution and climate change research. Different data-sets were used in each of the models, as no single data-set was in a format appropriate for use in both models. The selection of the emissions data-set for each model was based on appropriateness of content (i.e. biogenic emissions were excluded) and were recent releases actively being used for collaborative work by respected institutions. Although the emissions between the data-sets are quantitatively different, the temporal uniformity means that it would nonetheless be possible to distinguish patterns of transport of the pollutants (even though a comparison of actual pollutant concentrations would not be meaningful). Thus, the differing data-sets and the temporal uniformity in each data-set are not limitations on this study, as the focus of the study is not the exact quantity of the pollutant, but rather the pattern of the transport of the SO_2 and whether the transport is associated with observations of ambient SO_2 in Cape Town.

The models are run from January 2001 to December 2002, and the analysis of the observation data for this period is used to validate the simulation results in Chapter 5. Following this, Chapter 6 investigates the characteristics of the simulated sulphur and atmospheric conditions over South Africa.

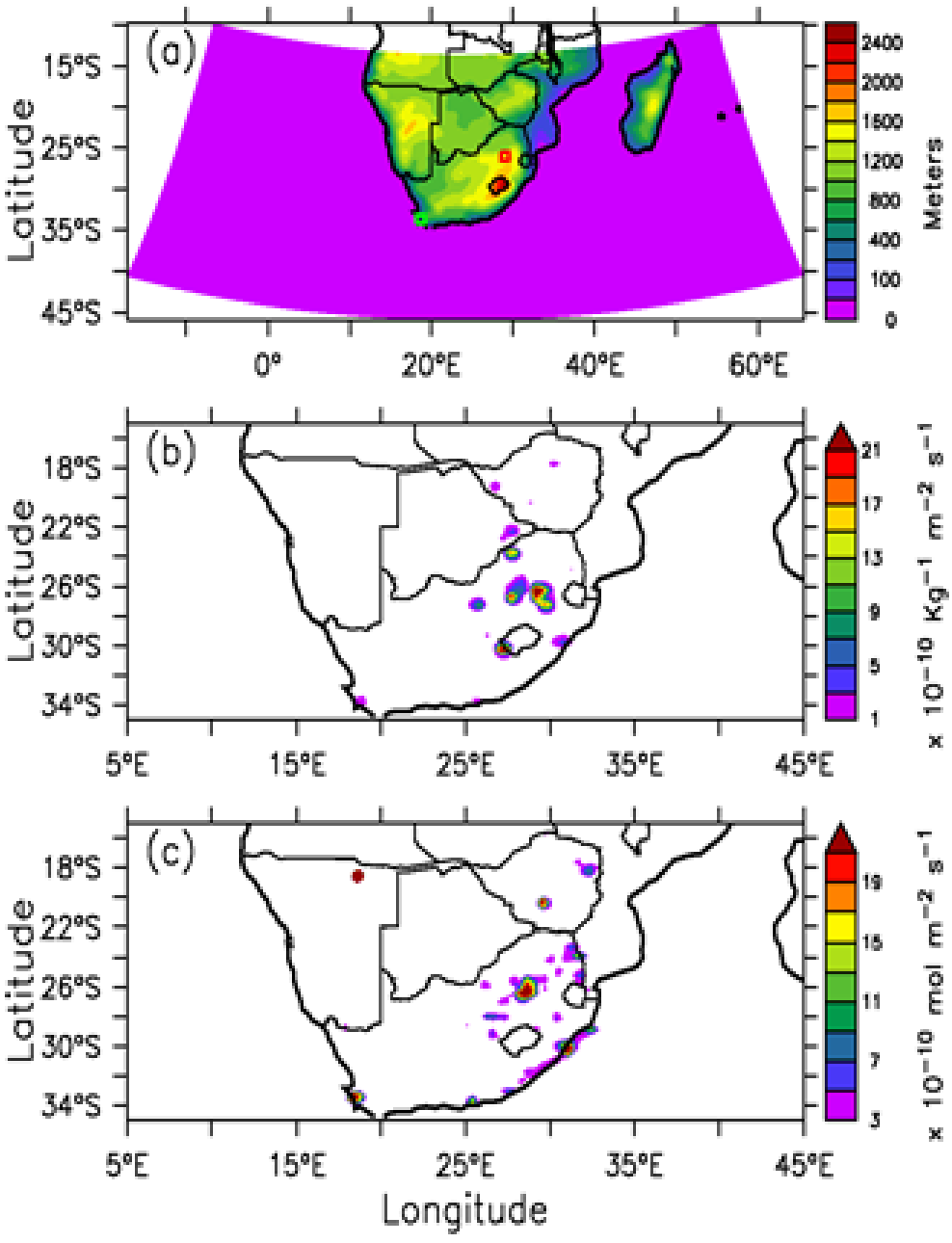

Figure 2: The RegCM-Chem and WRF-Chem simulation indicating domain (panel a) showing the southern Africa topography (in meters) as seen by the models; and the SO_2 emission distribution use the RegCM-Chem (panel b x 10^{-10} Kg^{-1} m^{-2} s^{-1}) and in the WRF-Chem (panel b; x 10^{-10} Mol m^{-2} s^{-1}) simulations. The location of Cape Town and Mpumalanga Highveld are indicated with green and red boxes, respectively, in panel (a)

Chapter 4: Results and discussion Part 1: Characteristics of the observed sulphur and atmospheric conditions in Cape Town

4.1 Diurnal variations

Analysis of the hourly mean SO_2 observations for the period from January 2000 to December 2009 shows two distinct peaks in the diurnal variation (Figure 3a), a morning peak around 0800 SLT and an evening peak around 1600 to 2000 SLT. The distinction between the two morning and evening peaks is clear at all the stations, but weak at the Bothasig station. The morning peak occurs at about the same time at all the stations but varies in magnitude from station to station. The concentration of the morning peak is higher than the evening peak and varies among the stations. This peak occurs earlier at City Hall than at other stations, but the value is highest at the Bellville South Station. The rate of industrial SO_2 emissions can be assumed to be constant and thus is not the cause of the peaks. Therefore transport may be the main producer of SO_2, apart from industry. The occurrence of peaks in the morning and evening can be attributed to the high volumes of commuter traffic, both road and rail, as people rush to work and school in the morning and return home in the evening. The morning commute is generally around the same time and over a shorter period (as businesses and schools open around the same time) compared to the evening commute (as businesses and schools close at different times). Thus this may partly explain why the morning SO_2 peak is higher than the evening one.

Nevertheless, atmospheric conditions may also contribute to concentration in the morning peak. Stable atmospheric conditions, which produce a shallow temperature inversion, can inhibit pollution dispersion. This is most common in the morning, and disappears in the afternoon, as the warmed land surface warms the air above (Figure 3b). Although the removal of the inversion layer cannot be identified from ground level observations alone, the rapid decrease in SO_2 concentrations in the afternoon, when the temperatures are high, can be partly attributed to the removal of the inversion layer. In addition, the weak wind speed in the morning over the stations (Figure 3c) would enhance pollution concentration, as there is reduced dispersion of pollutants. All the stations experience lowest concentration in the afternoon when temperature and wind

speed are highest; hence, buoyant atmospheric conditions enhance pollution dispersion and clear the atmosphere. High levels of SO_2 at individual monitoring stations can be associated with nearby emitters.

4.2 Seasonal variations

The monthly mean SO_2 observations, for the period 2000 to 2009, show that pollutant concentration is influenced by atmospheric conditions. The peaking of SO_2 is between April and July for all the stations (Figure 4a). This can be attributed to poor atmospheric dispersion during this period as a result of relative low temperatures (Figure4b) because of temperature inversion which inhibits pollution dispersion vertically. Also, low wind speeds (Figure 4c) during this period are favourable for high SO_4 concentration, as they inhibit horizontal dispersion of pollutants. The atmospheric conditions associated with low SO_2 concentrations are relatively high temperatures, and high wind speeds.

4.3 Inter-annual variability

Analysis of the annual mean SO_2 observations for the period 2000 to 2009 shows that the concentration of SO_2 in Cape Town varies from year to year. There are notable peaks between 2004 and 2007 (Figure 5a), and these vary between the stations. While City Hall shows two concentration peaks (in 2004 and 2007), concentrations at other stations (Bothasig, Bellville South, and Goodwood) peak in 2006. The inter-annual variability of atmospheric conditions may enhance or inhibit dispersion in some years. An increase in temperature inversions in 2006 is believed to account for high air pollution in that year (personal communication, Ian Gildenhuys, Specialised Environmental Health Services, City of Cape Town). A lack of continuous atmospheric data in some years prevents direct comparison of the inter-annual SO_2 concentrations and atmospheric conditions. Hence, in light of limited data, the study uses atmosphere chemistry-transport models to better study the SO_2 budget for Cape Town.

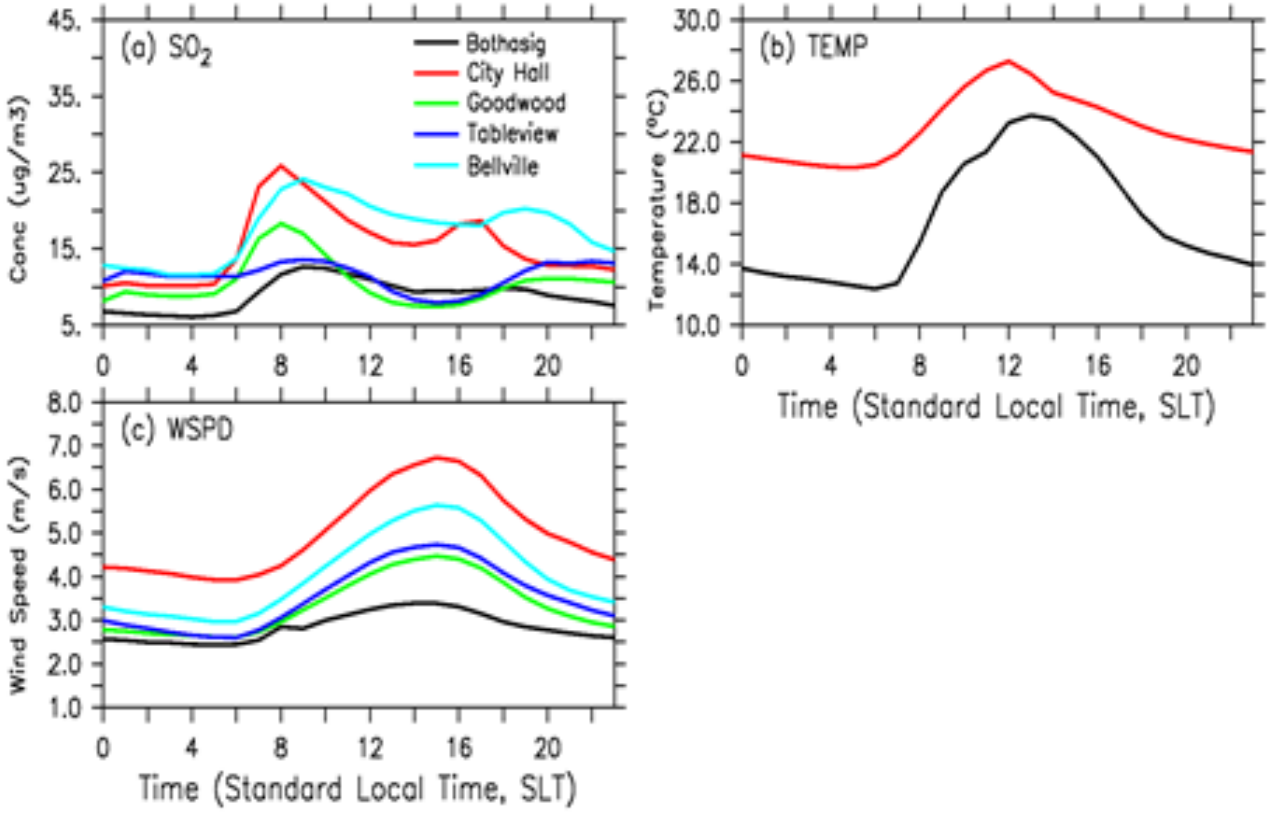

Figure 3: Diurnal variation of the observed (a) SO_2 concentration, (b) temperature, and (c) wind speed at five monitoring stations in Cape Town

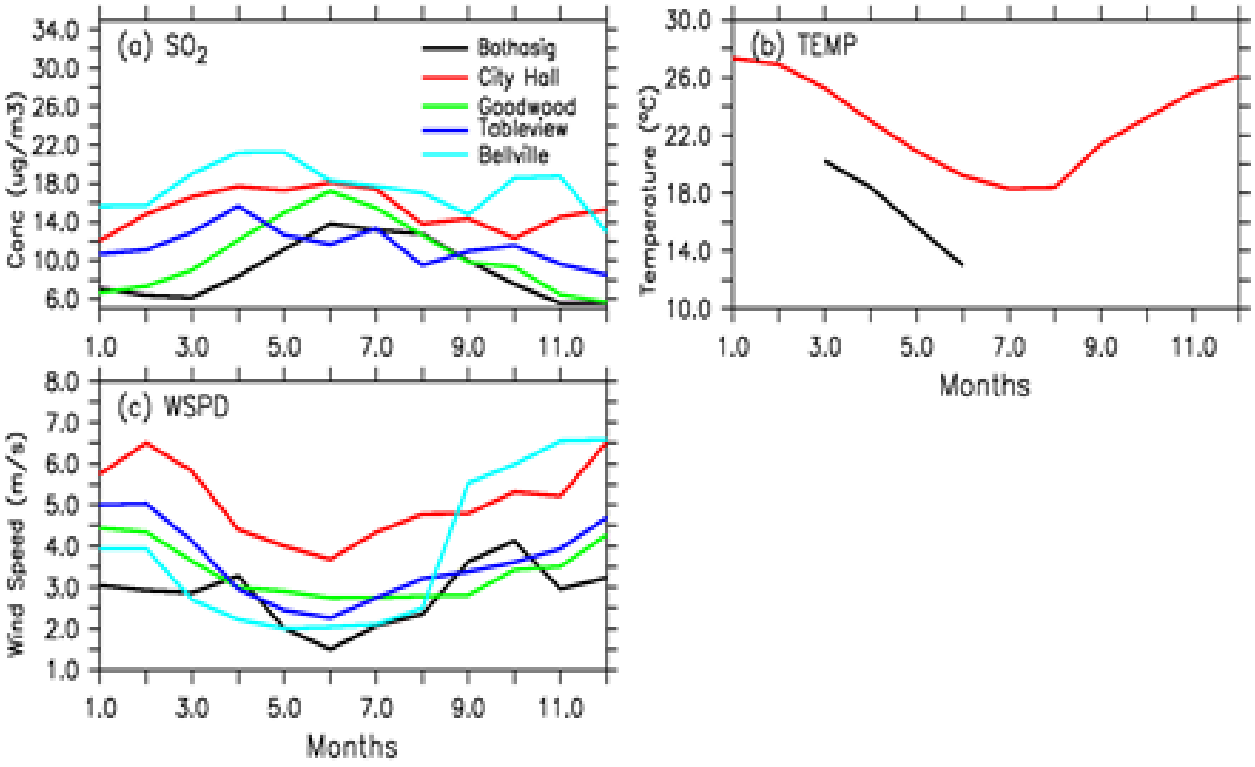

Figure 4 : The seasonal variation of observed (a) SO_2 concentration, (b) temperature, and (c) wind speed at five monitoring stations in Cape Town

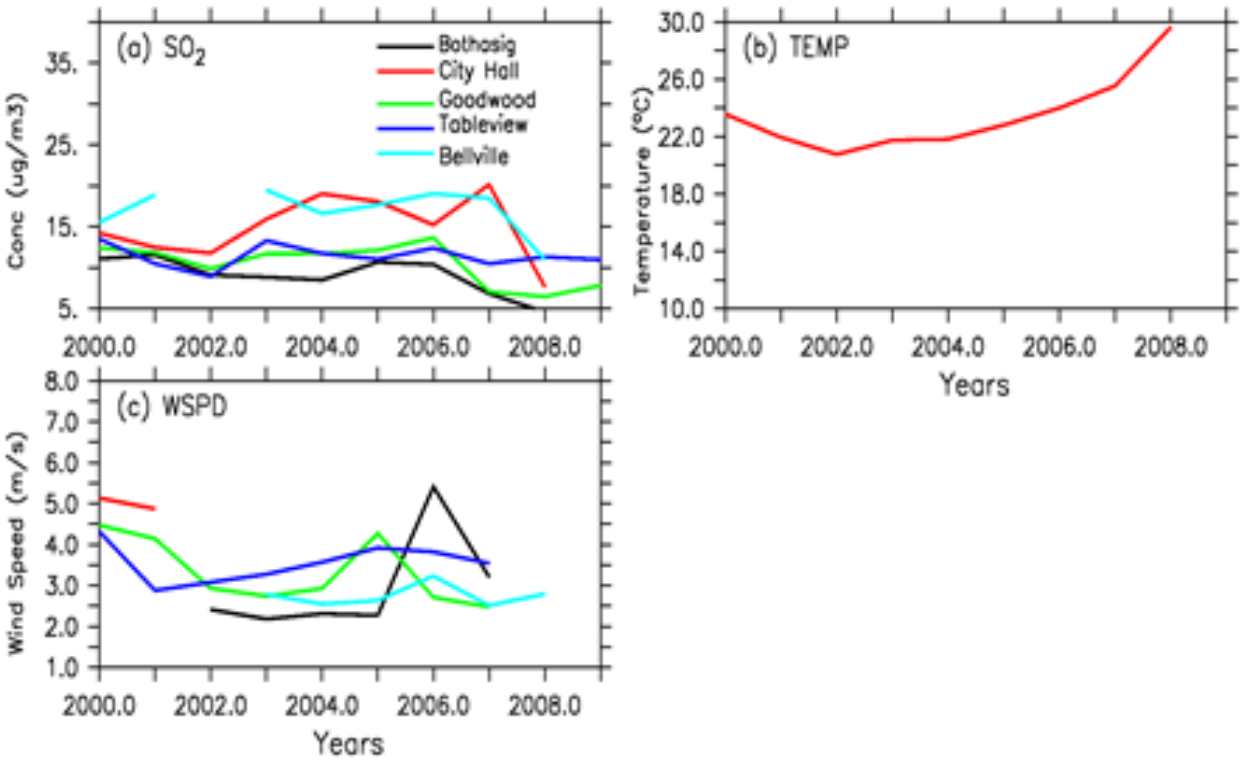

Figure 5: The inter-annual variation of observed (a) SO_2 concentration, (b) temperature, and (c) wind speed at five monitoring stations in Cape Town

Chapter 5: Results and discussion Part 2: Model validation

In order to validate the models, the simulated SO_2 concentration and atmospheric parameters are compared with those observed for January 2001 to December 2002. The evaluation focuses on how well the models reproduce area averages of seasonal and daily mean variations of the simulated variables over the area 18°21'E to 18°55'16''E and 33°28'24''S to 34°55'S. This area incorporates the locations of the five monitoring stations, i.e. Bellville South (33°54'56''S 18°38'36''E), Bothasig (33°51'28''S 18°32'27''E), City Hall (33°55'30''S 18°25'26''E), Goodwood (33°54'08''S 18°33'54''E) and Tableview (33°49'10''S 18°30'51''E). The observation data used for the validation is the mean of the observations at the five monitoring stations. While the area of the simulation domain used for the validation includes the location of the five stations, as an area it is of much lower resolution. This means that the simulation area may include locations with especially high emissions (for example the petrochemical refinery) and very low emissions. This difference in resolution may give bias to the simulation data, hence it is important to note that the validation focuses not only on absolute values but also on the temporal pattern in the observation and simulation output.

There is a generally good agreement between the simulated and observed seasonal variation of the SO_2 (Figure 6a), although both models overestimate SO_2 and there are some discrepancies between the two models. The models are able to simulate the maximum and minimum SO_2 concentrations in autumn (MAM) and winter (JJA) respectively, although RegCM-Chem underestimates the values of these extremes. Both models underestimate SO_2 concentration in summer (OND). The main discrepancy between the simulated and observed SO_2 occurs in October to December (OND). The observed SO_2 shows a decreasing trend in OND, but that of simulated SO_2 increases. In addition, the observed SO_2 shows a secondary peak in SO_2 in August, but neither of the models capture this, and both models underestimate the concentration of SO_2 in August. Although there is no SO_4 observation data to evaluate how well the models simulate SO_4, it is noteworthy that both models agree on the concentration of SO_4 in January to March

(JFM) and in April to December, but disagree for the period April to August (from mid-autumn through winter).

It is difficult to explain the disagreement in SO_2 observation and simulation data (or between the two sets of model results) using the available meteorological observation (surface temperature and wind speed). Figure 6(c) shows that both models replicate the observed temperature pattern but with a cold bias in all the months; the highest biases in JFM and OND. While it might be tempting to attribute the positive bias of simulated SO_2 in OND to the cold bias in the season, either because a colder surface temperature might imply stronger stability and higher concentration or it might imply less intense sunlight and thus a lower removal of SO_2 by oxidation to SO_4, this does not explain why in other periods (i.e. JFM) simulated SO_2 shows very good agreement with observation, despite the cold bias. Figure 6(d) shows a big discrepancy between the simulated and observed seasonal variation of wind speed, except in JFM. The models overestimate the wind speed in all months; the greatest difference is in May to August, when the models overestimate the wind speed by about 4.0 ms^{-1}. However, this also does not explain the pattern of discrepancy and agreement between the observed and simulated SO_2. Nevertheless, the disagreement between the simulated SO_4 in RegCM-Chem and WRF-Chem in April to August may be attributed to differences in the treatment of deposition or transformation between the models.

Correlation of values of the simulated daily mean SO_2 with daily mean observation (Figure 7) shows weak correlation, while the simulated standard deviation of the daily mean SO_2 for both models is very close to that of the observed values. Figure 7 shows a comparison of the simulated daily mean of SO_2 (circles), temperature (square), and wind speed (triangle) for each of the models in comparison to observations (the observations are in black, RegCM-Chem in blue and WRF-Chem in red.). With RegCM-Chem, the correlation coefficient is 0.4 and the normalised standard deviation is 0.9; with WRF-Chem, the correlation coefficient is 0.35 and the normalised standard deviation is 1.2. Both models perform best in simulating temperature, and least well in simulating wind speed. For temperature, in both models, the correlation coefficient is about 0.8 and the normalised standard deviation is about 0.8, while for wind speed the correlation coefficient is about 0.6 for RegCM-Chem and 0.4 for WRF-Chem. The simulated standard

deviations are higher than that of the observed by a factor of 2. In general, the RegCM-Chem, simulation gives better results than WRF-Chem in terms of correlation coefficient and standard deviation of daily mean SO_2, temperature and wind speed.

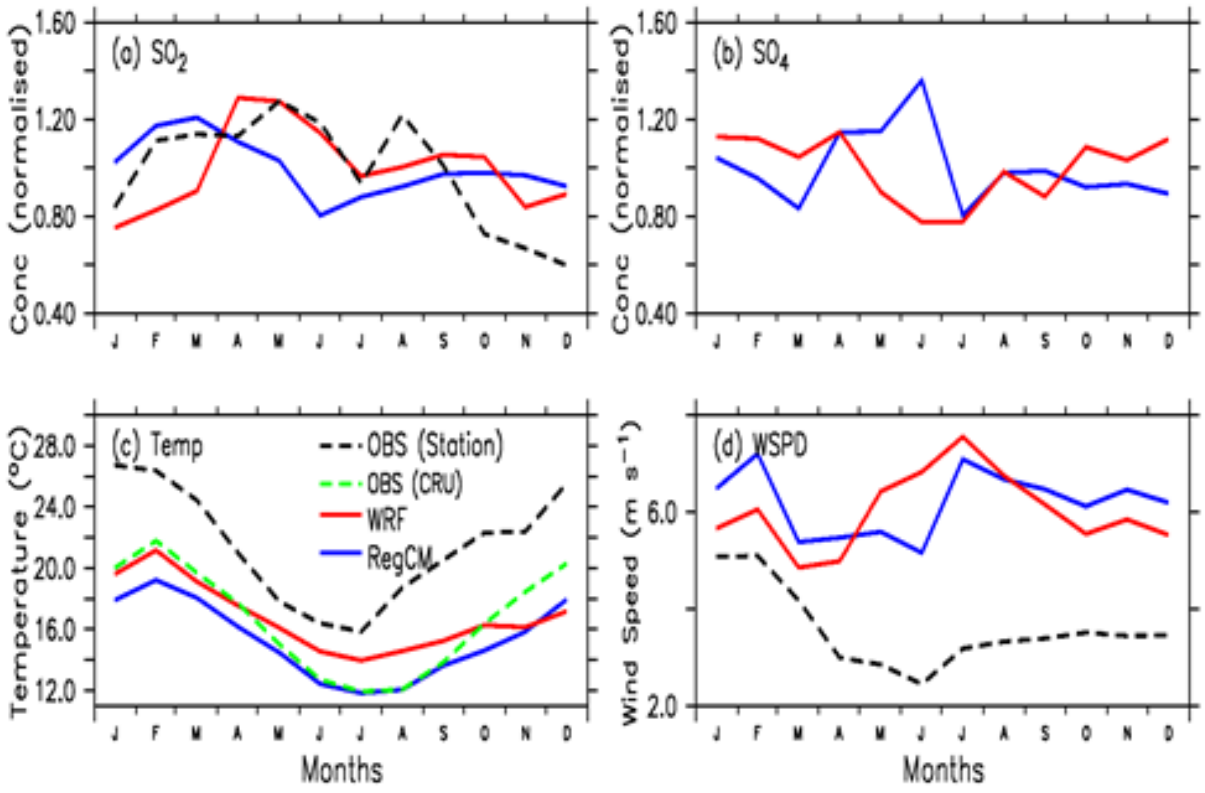

Figure 6: Seasonal variation of observed and simulated (a) SO_2, (b) SO_4, (c) temperature (°C) and (d) wind speed (m s^{-1}) for 2001-2002. The SO_2 and SO_4 values are normalised with their annual mean values for easy comparison

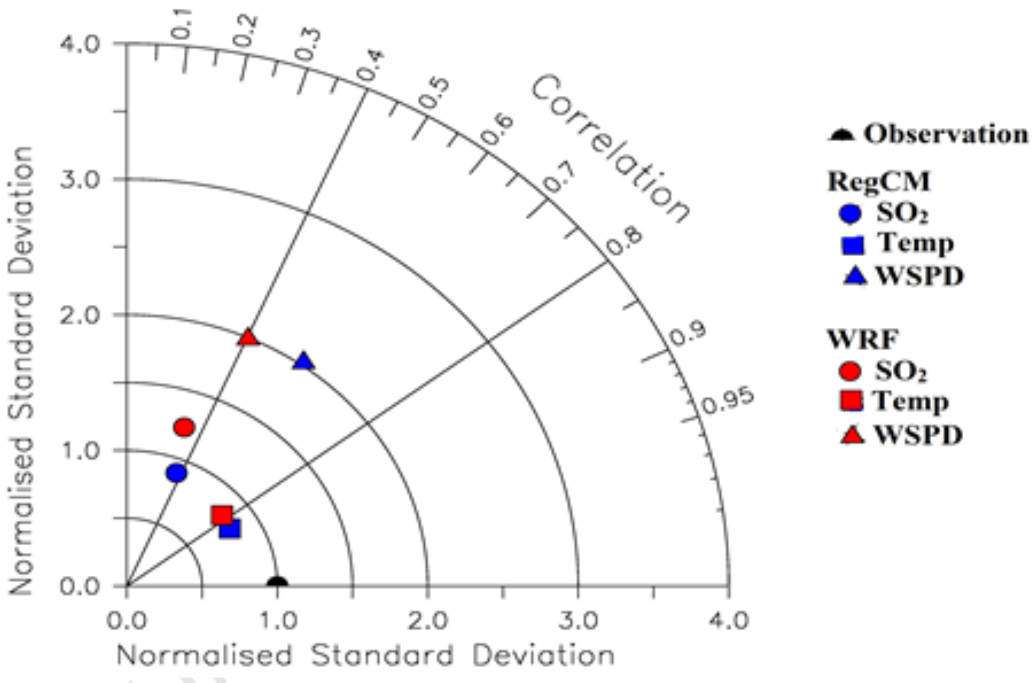

Figure 7: Comparison of the simulated and observed daily mean of SO_2 (circles), temperature (square), and wind speed (triangle). The observations are in black, RegCM-Chem in blue and WRF-Chem in red

Chapter 6: Results and discussion Part 3: Characteristics of the simulated sulphur and atmospheric conditions over South Africa

In this chapter, the mean annual simulated concentrations near the surface are analysed to assess whether the spatial distribution reveals links between major sources of SO_2 emissions in South Africa (the Mpumalanga Highveld) and the city of Cape Town. To identify patterns of pollutant transport, the mean annual simulated concentrations around the lowest layer of atmospheric stability, around 700 hPa, are analysed. Thereafter the seasonal pattern of pollutants and the meteorological drivers of mixing and transport across the country at the near surface level are investigated. The analysis focuses on identifying dominant drivers of high concentrations of atmospheric sulphur over Cape Town.

6.1 Annual mean around 700 hPa

The simulated SO_2 and SO_4 mean concentrations at upper levels (around 700 hpa) (Figure 8 and Figure 9) show an eastward transport of sulphur over South Africa by anti-cyclonic flow. RegCM-Chem simulates the maximum concentrations of SO_2 and SO_4 over the Mpumalanga Highveld, an area of intense industrial activity in South Africa (Zunckel *et al.* 2000). Upper panels of Figure 8 Figure 9 clearly show that, at the upper level, some of the simulated sulphur in RegCM-Chem and WRF-Chem is transported directly to the Indian Ocean by the westerly flow, while some is recycled over southern Africa by anti-cyclonic flow. This is consistent with previous studies (Freiman and Piketh 2003; Garstang *et al.* 1996; Piketh *et al.* 2002; Tyson and D'Abreton 1998) which show that 39% of pollutants from the Mpumalanga Highveld is transported to the Indian Ocean and 33% is recycled over the sub-continent.

6.2 Annual mean near the surface

However, the transport of simulated SO_2 and SO_4 near the earth's surface (the models' lowest grid point) over South Africa in both RegCM-Chem and WRF-Chem is different from that of upper levels, because wind patterns differ (Figure 8c and d; Figure 9c and d). Although, the peak concentration is still over the Mpumalanga Highveld (as at the upper level), the annual wind

pattern at surface level shows a strong north-easterly flow over the Mpumalanga Highveld and weak northerly flow over the southern half of South Africa (in contrast to that of the upper levels). This suggests a near surface southward transport of sulphur from the Mpumalanga Highveld. This is consistent with Freiman and Piketh (2003) that 6% of Highveld pollutant is transported by northerly flow south to the South Indian Ocean, except that in the present study the percentage may be higher because concentration of the pollutant is highest near the surface. In addition, RegCM-Chem and, to a lesser extent, WRF-Chem suggest that most of the sulphur transported by the northerly flow to the South Indian Ocean may be trapped along the south coast owing to the convergent flow between the northerly and southerly winds. As a result, there is a band of high SO_2 concentration along the coastline. However, it not clear from these figures if the band of pollutant along the coast is transported westward into, or eastward from, Cape Town. The model results also agree that the peak of SO_4 concentration is over the Mpumalanga Highveld, but with a lower concentration than that of SO_2 and there is no band of SO_4 along the coastline, as is evident for SO_2.

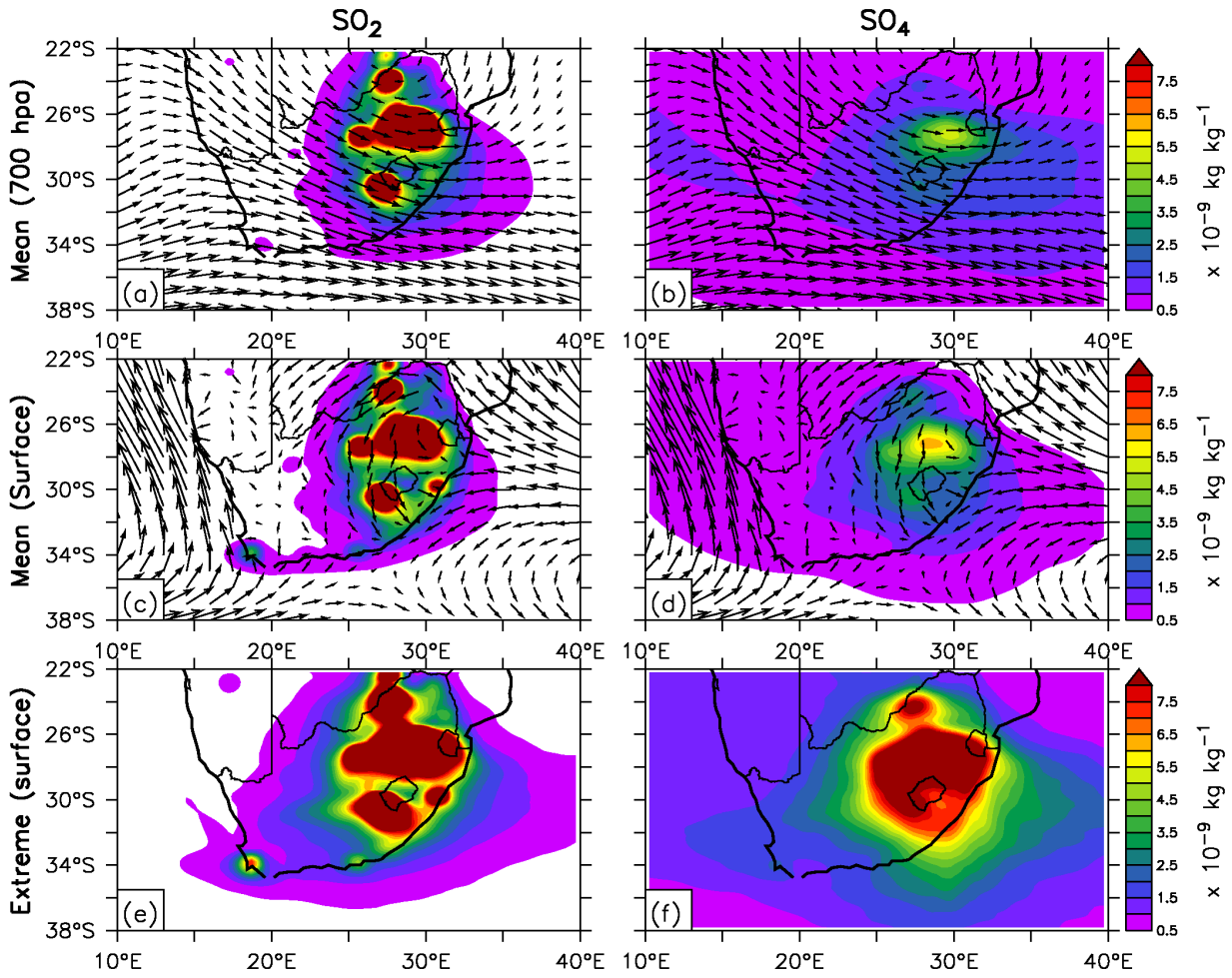

Figure 8: The simulated (RegCM-Chem) annual mean (a, b, c, and d) and extreme (e and f) concentrations for SO_2 (left panels) and SO_4 (right panels) over South Africa at 700 hPa level (top panels) and near surface (middle panels) for 2001 and 2002. The extreme concentration is defined as 95 percentile of the daily mean concentration

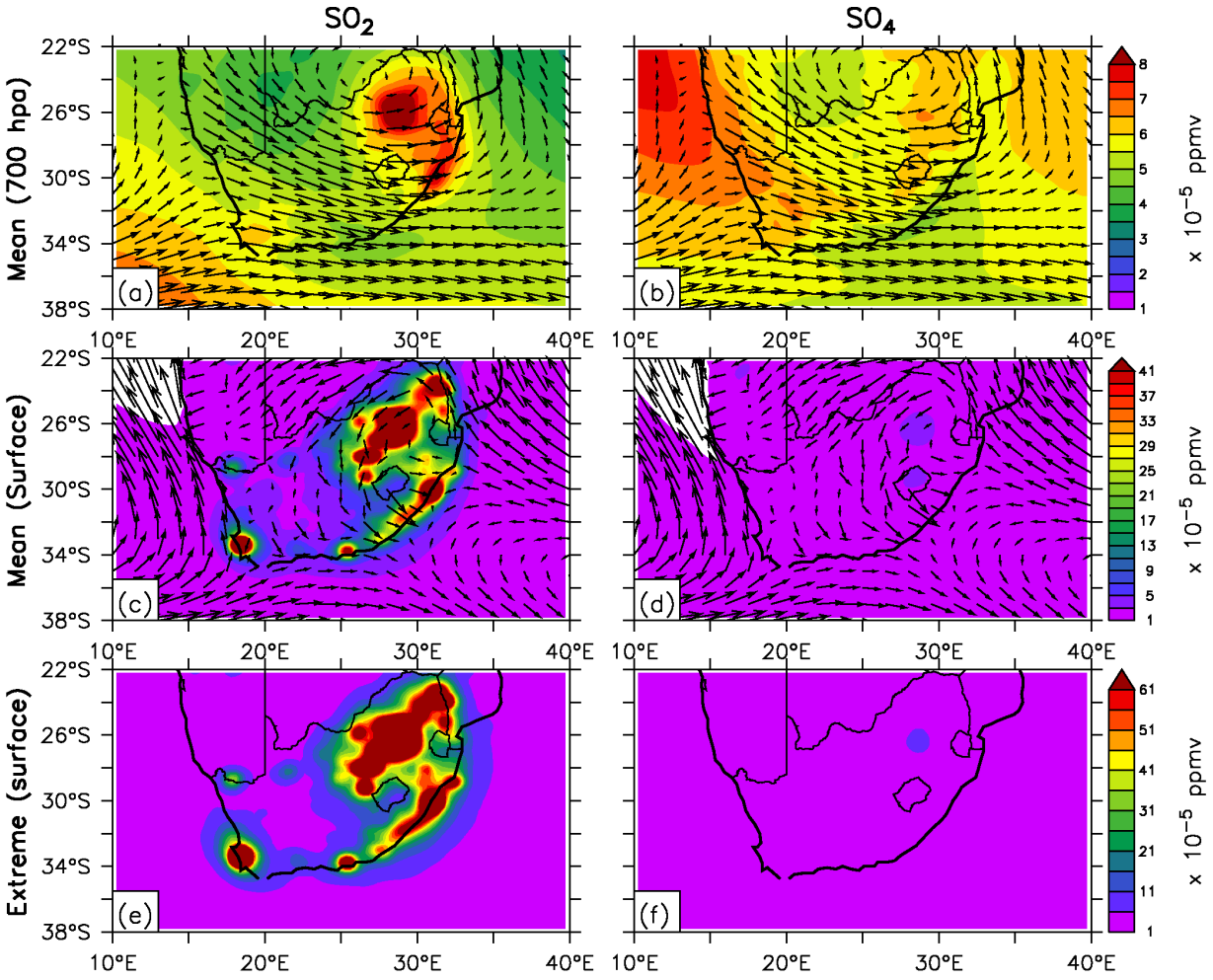

Figure 9: The simulated (WRF-Chem) annual mean (a, b, c, and d) and extreme (e and f) concentrations for SO_2 (left panels) and SO_4 (right panels) over South Africa at 700 hPa level (top panels) and near surface (middle panels) for 2001 and 2002. The extreme concentration is defined as 95 percentile of the daily mean concentration

6.3 Seasonal variation

Error! Reference source not found.and Figure 12 show that the simulated SO_2 concentration (2001 and 2002) over South Africa are generally similar over the whole country for all the months in both models. Both models exhibit a seasonal variability; however, over the western half of the country, there is a weak seasonal variation in SO_2 in WRF-Chem. Both models agree that, over the Mpumalanga Highveld, SO_2 concentrations have negative anomalies (i.e. below the annual mean) in October to January (summer) and positive anomalies (i.e. above the annual mean) in April to August (mid-autumn to winter). This is consistent with the prevailing atmospheric conditions over the region. For instance, in summer (represented by the November climatology plots in Figure 10), the surface air is warmer over the continent than over the surrounding oceans, and the inversion layer is elevated above the eastern mountain range (i.e. the escarpment). This will enhance the vertical mixing of near surface pollutants and, at the same time, allow inland penetration of the near surface easterly flow from the Indian Ocean (Figure 10a and c), bringing in less polluted marine air to dilute the concentration of SO_2 over the north-eastern part of the country (i.e. the Mpumalanga Highveld). On the other hand, from April to August (winter) (represented by the June climatology plots in Figure 10), the inversion layer is below the peak of the escarpment (Figure 10b and d). Hence, there is less vertical mixing of SO_2 and the easterly near surface flow cannot penetrate inland with the fresh air, but moves around the Drakensberg mountain range along the eastern edge of the escarpment, southward along the coastline or northward towards Mozambique as described in Rouault *et al.* (2000). The negative SO_2 anomalies in summer can also be attributed to an increase in the removal of SO_2 owing to the gas-phase reaction; and vice versa in winter. This is consistent with Josipovic *et al.*, (2011) who show that the removal of SO_2 in the production of SO_4 is higher in summer (with longer and more intense sunlight) than in winter.

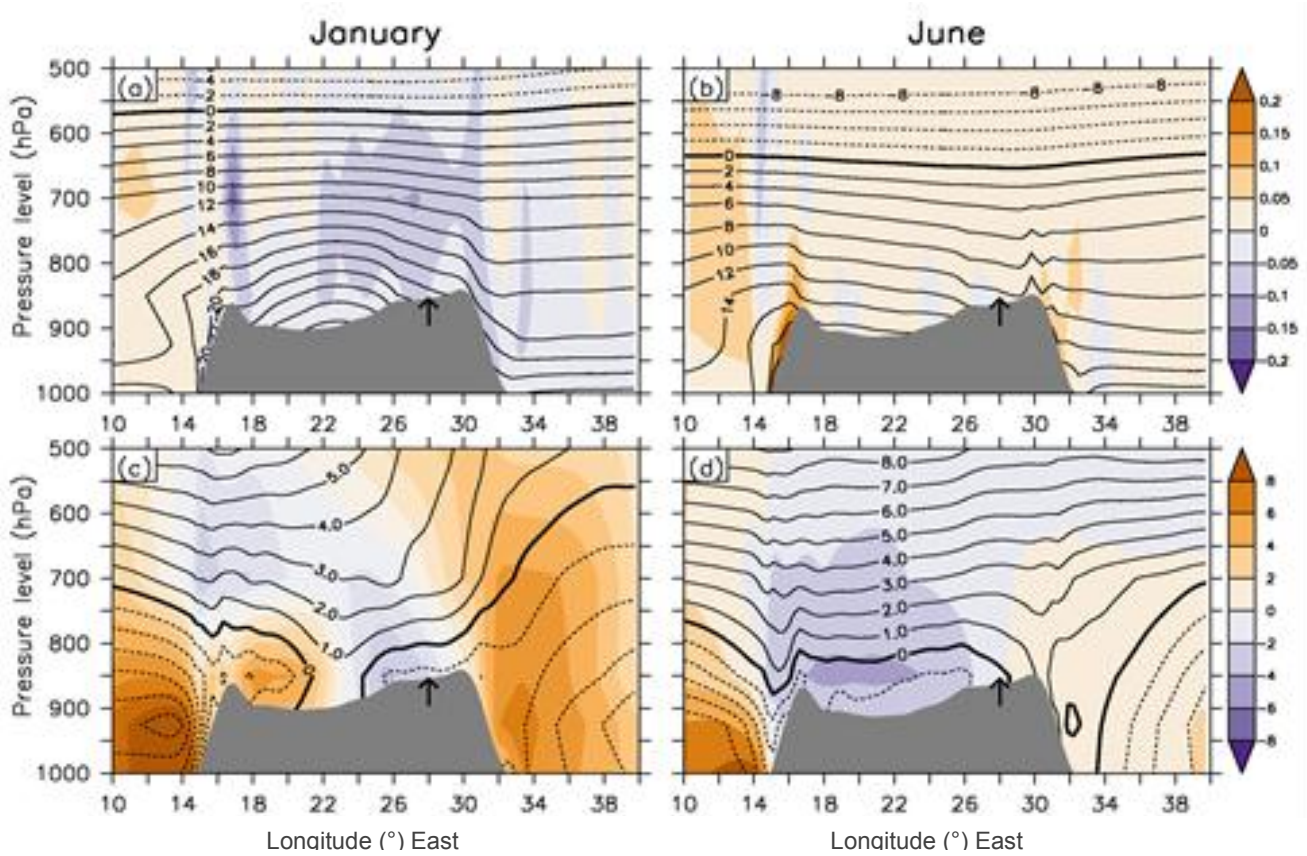

Figure 10: Vertical cross section of temperature (°C; contours in upper panels), vertical wind component (x 100 mb s^{-1}; shaded in upper panels), zonal wind component (m s^{-1}; contours in lower panels), and meridional wind component (m s^{-1}; shaded in lower panels) at latitude 26°S in June and November (2001 and 2002). Topography is shown in grey colour and the location of the Highveld indicated with arrow (↑)

Over the southern half of the country, the models exhibit similar SO_2 patterns (Figure 11 and Figure 12) except that the seasonal variation of SO_2 is weaker in WRF-Chem than in the RegCM-Chem simulation. For instance, in January and February, RegCM-Chem shows strong positive anomalies, while WRF-Chem shows weak negative anomalies; and in May and June, RegCM-Chem shows strong negative anomalies, but WRF-Chem shows weak positive anomalies. The strong positive anomalies over this area in summer (January and February; RegCM-Chem) can be attributed to the transport of SO_2 from the Mpumalanga Highveld by the surface easterly flow, while the strong negative anomalies over the area in winter (May and June; RegCM-Chem) can be attributed to the transport of fresh air from the north-west part of the country.

Over Cape Town, the models agree that SO_2 concentration has positive anomalies from April to June (autumn to early winter) and negative anomalies from November to January; but disagree on the signs of the anomalies in other months. For instance, while RegCM-Chem shows positive anomalies of SO_2 concentrations in March and negative anomalies from January to September, WRF-Chem shows the opposite (especially in September), despite the general agreement between the monthly mean wind patterns of the models in August and September. This implies that the discrepancies in the simulated SO_2 may be due to differences in the chemistry (rather than the dynamics) of the models. Negative anomalies in SO_2 concentration over the area in winter (July to September) may be because of increased removal of SO_2 from the atmosphere by oxidation within clouds and by wet deposition, because SO_2 is highly soluble and winter is the main rainfall period for Cape Town. This process, however, is stronger in RegCM-Chem than in WRF-Chem. Nevertheless, both models show positive anomalies in Cape Town and along the southern coast of South Africa in April, along with convergence and stagnant flow over the area. These atmospheric conditions encourage the accumulation of pollutants. In addition, there is transport of SO_2 from the north-eastern part of the country along the coastline towards Cape Town in April in both models.

There is no agreement between the models as to the seasonal pattern of SO_4 concentrations over South Africa. In fact, the models show opposite signs in monthly anomalies (Figure 13 and Figure 14). While RegCM-Chem simulates SO_4 with positive anomalies over most of the country from May to September and negative anomalies in other months, WRF-Chem simulates the opposite.

In RegCM-Chem, the anomalies of SO_4 have the same pattern as those of SO_2; but in WRF-Chem, the pattern is opposite to that of SO_2 anomalies, except in March, April and September. In both models the SO_4 concentration is mainly driven by the rate of gas-phase transformation from SO_2 to SO_4. However, it appears that in RegCM-Chem, this process is driven by the amount of available SO_2 (i.e. the higher the SO_2, the higher the SO_4); but in WRF-Chem, the process is driven by the availability of OH, which is driven by the intensity and duration of sunlight (more sunlight increases photolysis and subsequently OH). This explains the uniform positive anomaly of SO_4 in summer and the negative anomaly of SO_4 in winter in WRF-Chem simulation, which is consistent with Josipovic *et al.* (2011) findings that SO_4 production is higher in summer than in winter. The lack of this characteristic in RegCM-Chem simulation may mean that OH concentrations in this model are not sensitive to sunlight. However time constraints for this work limit further investigation into the cause of the discrepancy here.

There is strong agreement between the models as to the pattern of vertical velocity across the country and over Cape Town. RegCM-Chem produces output for omega (Figure 15), which is a suitable proxy, as it is converse to vertical velocity, while WRF-Chem shows vertical velocity (Figure 16). Along the coast, vertical velocity is downwards in late autumn and winter (May to August), and upward in late spring and summer (November to February).

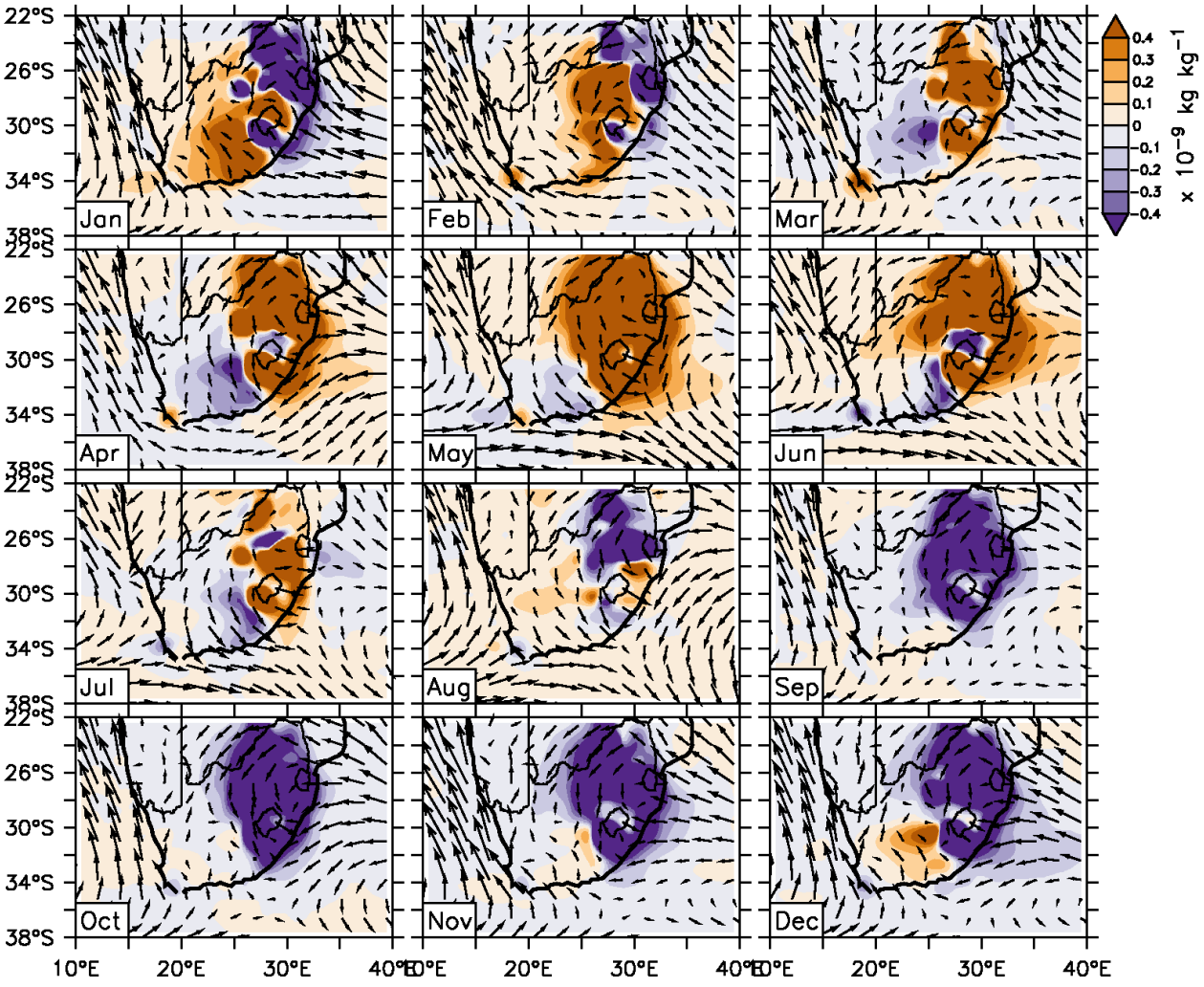

Figure 11: The monthly anomalies SO_2 concentration (10^{-9} kg kg^{-1}) and associated winds (actual) as simulated by RegCM-Chem for 2001 and 2002

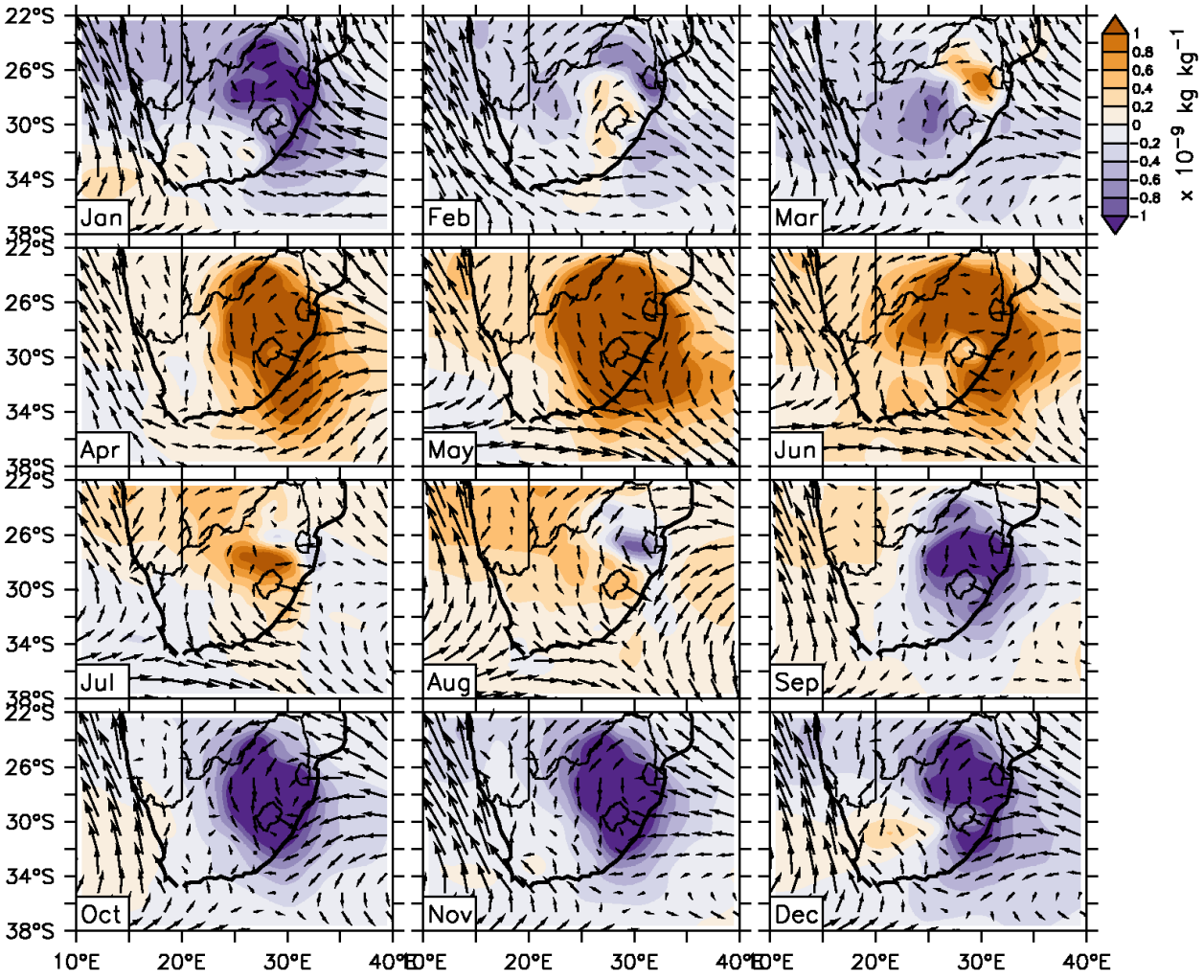

Figure 12: The monthly anomalies SO_2 concentration (x10^{-4} ppmv) and associated winds (actual) as simulated by WRF-Chem for 2001 and 2002

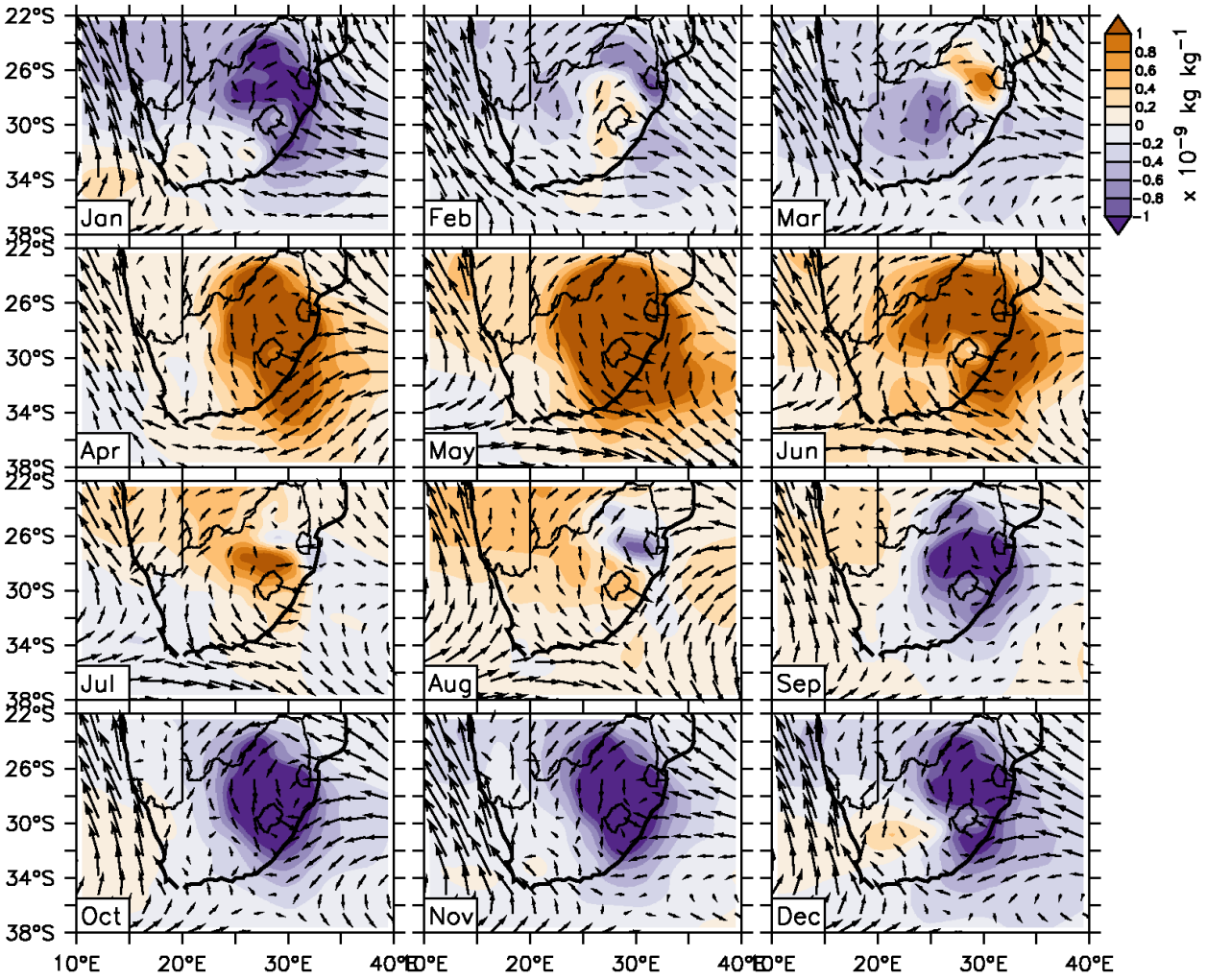

Figure 13: The monthly anomalies of SO_4 concentration (10^{-9} kg kg^{-1}) and the associated winds (actual) as simulated by RegCM-Chem for 2001 and 2002

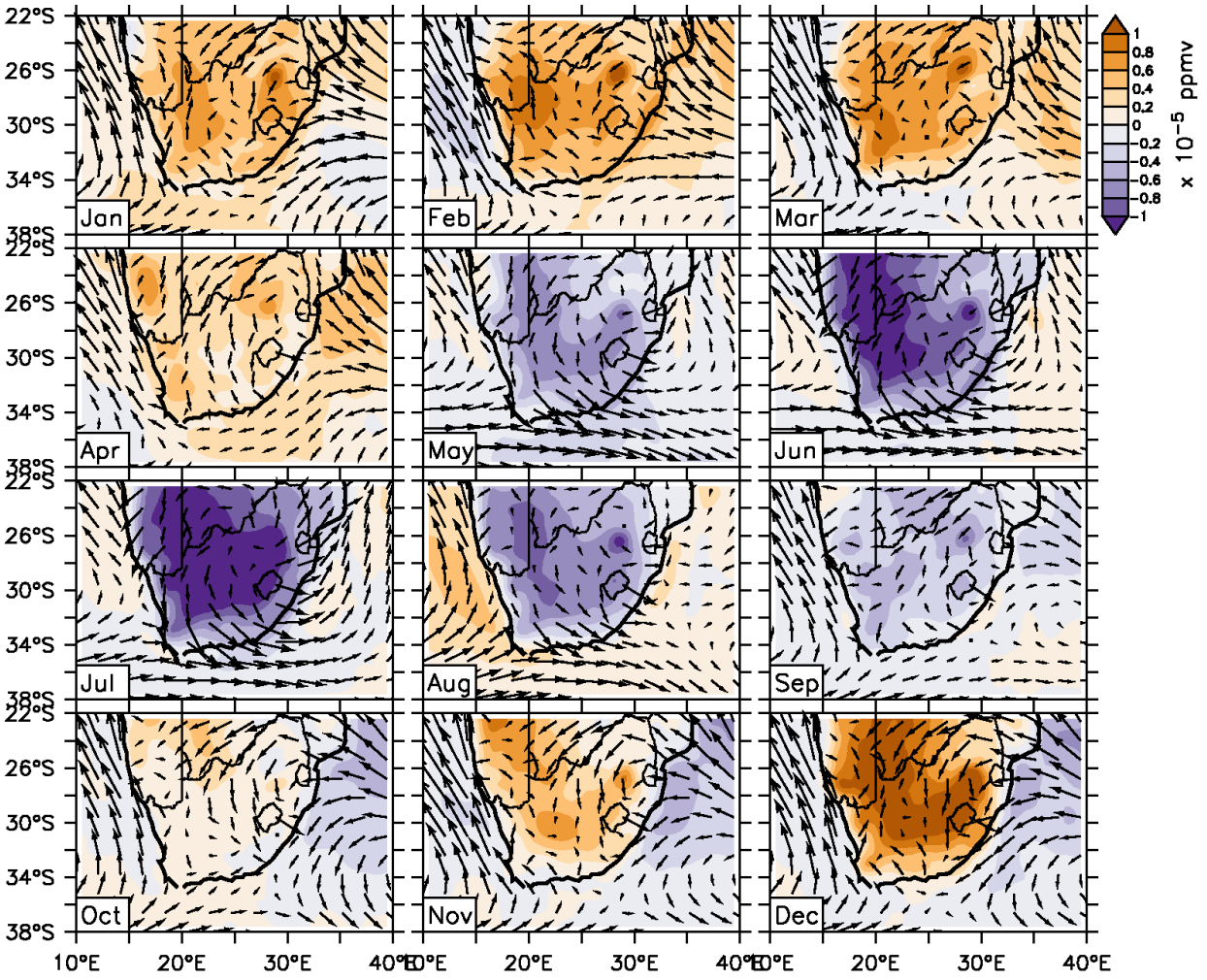

Figure 14: The monthly anomalies of SO_4 concentration (10^{-5} ppmv) and the associated winds (actual) as simulated by WRF-Chem for 2001 and 2002

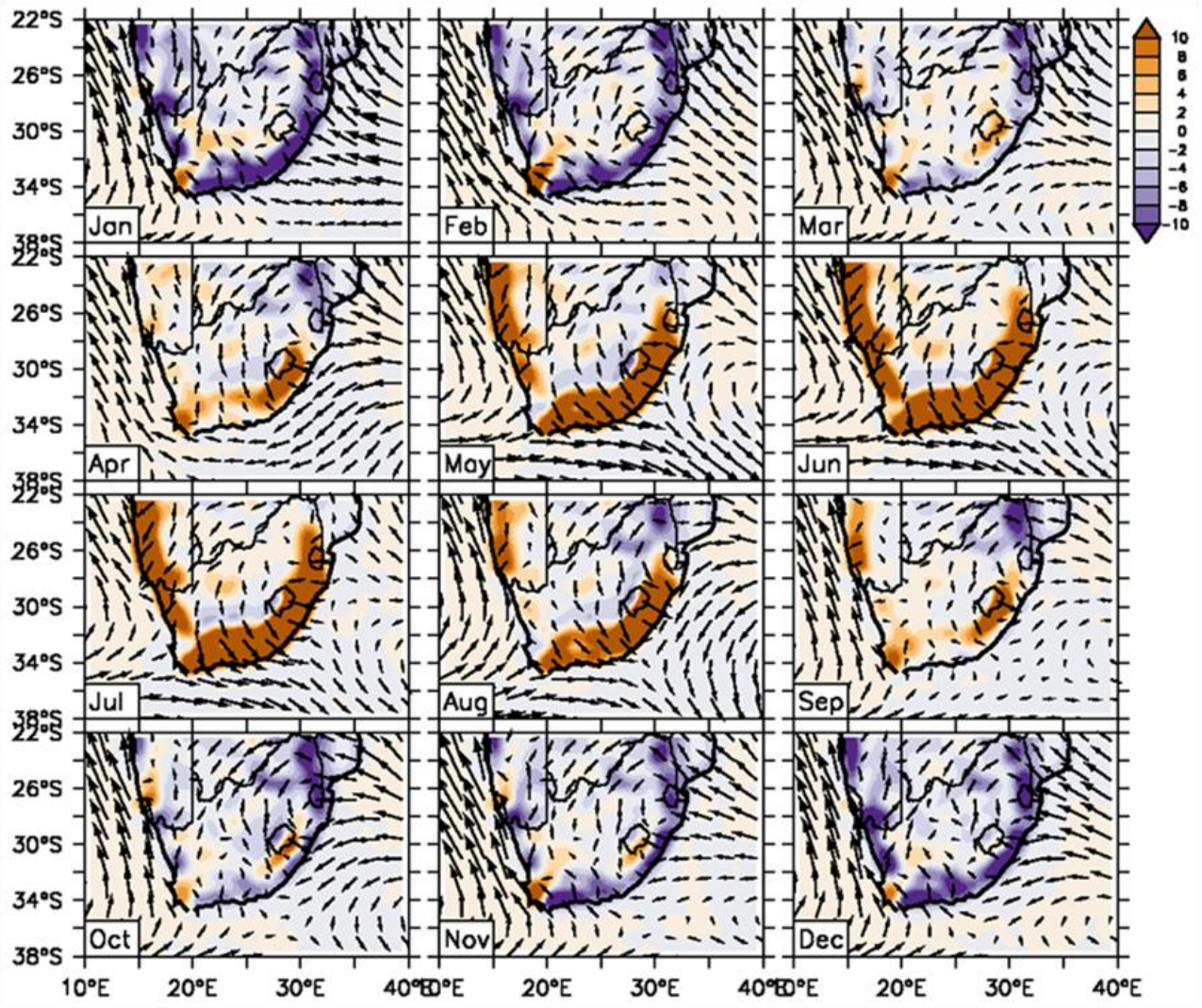

Figure 15: Mean omega (hPa s^{-1}) (the proxy for vertical velocity) around 850 hPa as simulated by RegCM-Chem for 2001 and 2002

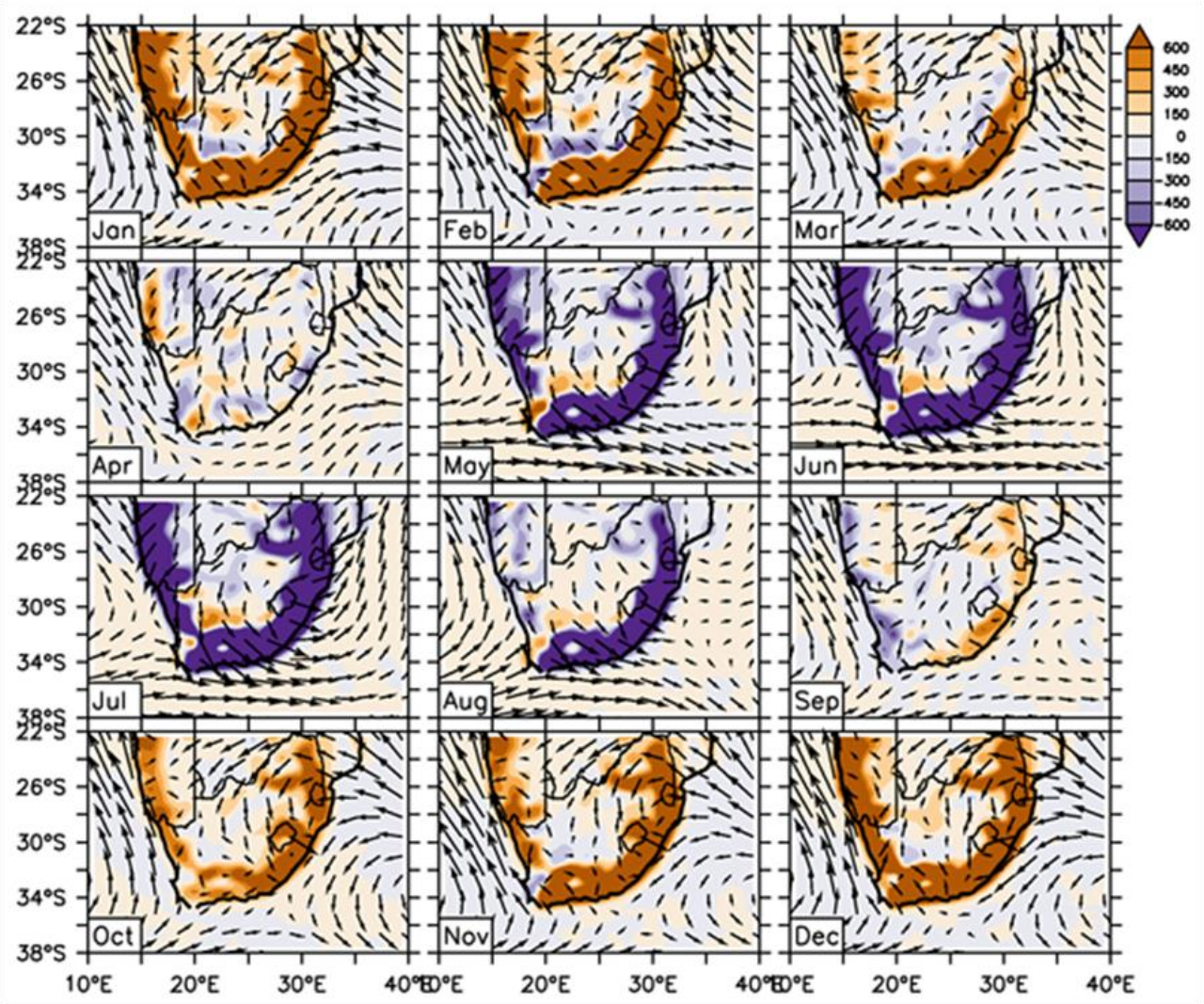

Figure 16: Mean vertical velocity (m s^{-1}) as simulated by WRF-Chem for 2001 and 2002

6.4 Extreme events

The time series of simulated SO_2 and SO_4 over Cape Town (specifically the area of the domain used for model validation in this study and as described in Chapter 5) for 2 years (2001 and 2002)(Figure 17) shows some consistencies between the models as to the months in which highest concentrations or exteme events (in excess of the 95^{th} percentile) occur. In both years, RegCM-Chem identifies April, May and August as months with extreme events for SO_2 while WRF-Chem identifies May, August and September. Both models agree on the occurrence of SO_2 extreme events in June and November 2001, and in February, April, June, September, and December 2002. In the RegCM-Chem simulation, the highest frequency of SO_2 extreme events occur in May (30%), April (22%), and June (11%); but in the WRF-Chem simulation, they occur in May (27%), April (17%), September (13%) and November (13%) (Figure 18). Hence, both models agree that most SO_2 extreme events (more than 42%) occur in April and May.

Meanwhile, there is no agreement between the models as to when the SO_4 extreme events occur. In both years, RegCM-Chem simulates the occurrence of the extreme events as frequent in April, May, June, July, August, and September, but WRF-Chem simulates the occurrence mostly in April, and October. In the RegCM-Chem simulation, the highest percentage of SO_4 extreme events occur in May (20%), July (17%), August (14%) and September (14%); but in the WRF-Chem simulation, they occur in April (31%), October (15%) and December (15%). Hence, for concentrations of SO_4, while RegCM-Chem suggests that most of the extreme events (74%) occur in May to September, WRF-Chem suggests that most of the extreme events (96%) occur in October to April. Unfortunately, there are no SO_4 observations to substantiate either set of results.

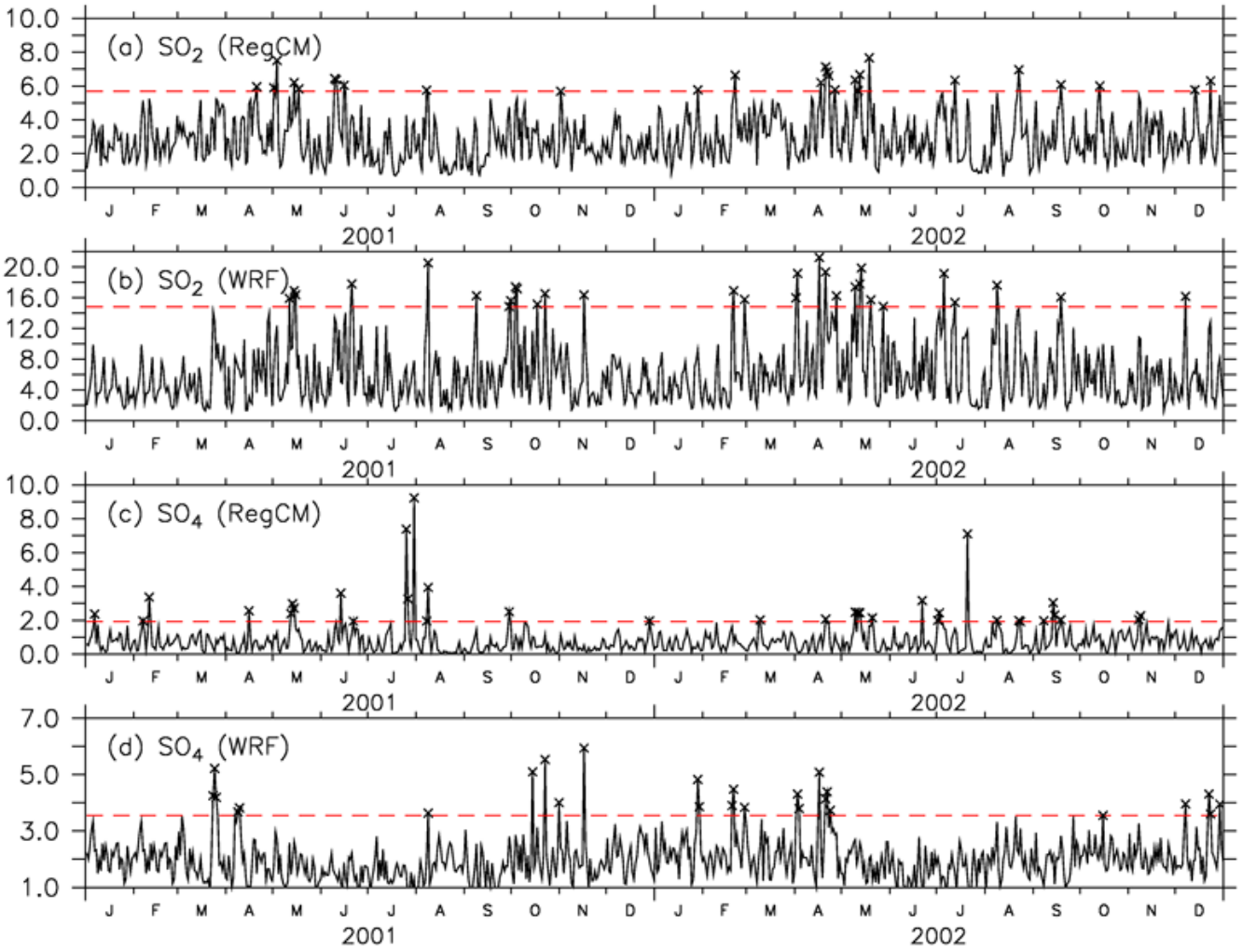

Figure 17: Monthly variations occurrence of the extreme concentration events for SO_2 and SO_4 over Cape Town as simulated by RegCM-Chem and WRF-Chem for 2001 -2002

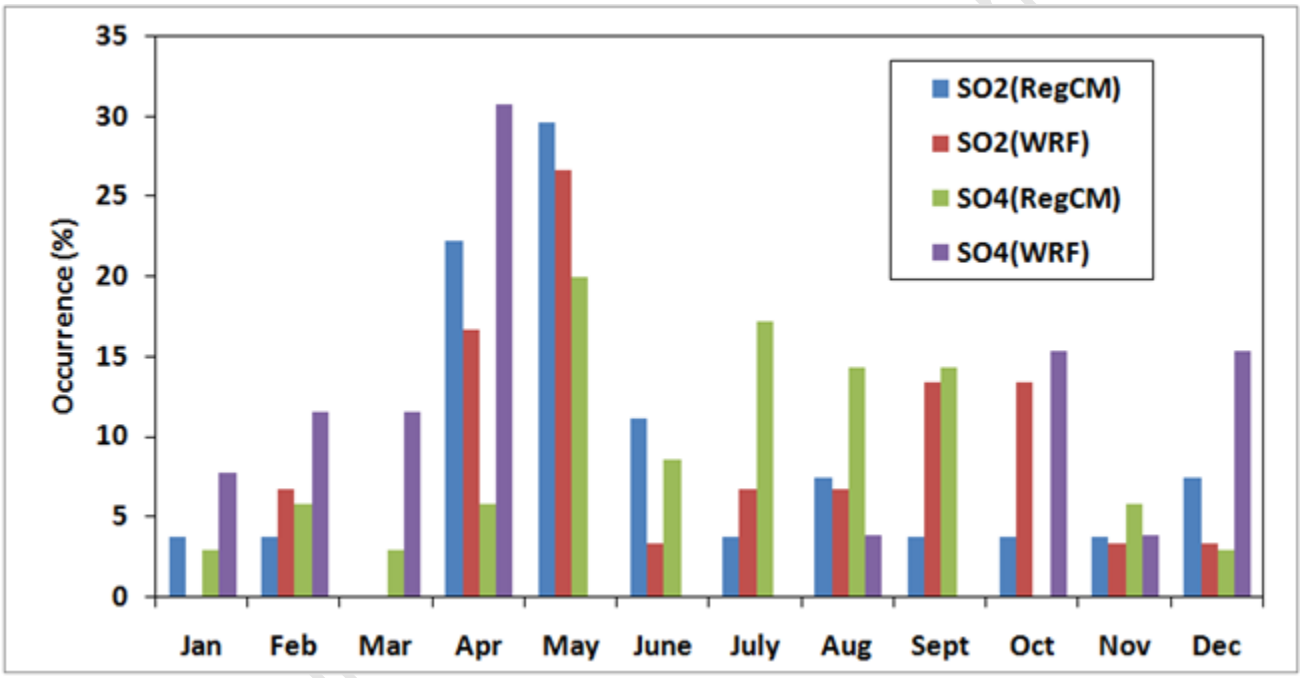

Figure 18: Monthly variations of occurrence of the extreme concentration events for SO_2 and SO_4 over Cape Town as simulated by RegCM-Chem and WRF-Chem for 2001-2002

Figure 19 presents the composite SO_2 and SO_4 concentrations (and associated wind patterns) during the four months with the highest number of extreme events in each model. The figure reveals that the wind pattern associated with extreme concentrations of SO_2 or SO_4 in Cape Town is the formation of a col (with or without subsidence motion) over the area. A col (indicated on synoptic charts as a saddle or ridge in isobars) is usually associated with a calm or light variable wind, which causes stagnation of air flow and the accumulation of atmospheric pollutants. Several studies have shown that col conditions are ideal for producing poor air quality (Stein *et al.* 2003). Figure 20 shows wind patterns that suggest a col, or in some cases, convergence over the area of Cape Town. In some cases, the col is associated with subsidence (negative vertical velocity) which will inhibit the vertical mixing of the pollutant. However, without the subsidence, the col can still produce extreme concentrations (in excess of the 95th percentile) in pollution for Cape Town (Figure 19). In some case the col is associated with easterly flow along the southern coastline and northerly flow from the continent. Both winds may transport pollutants from the north-east, and their convergence along the coast might lead to the development of a band of pollution along the coast; the band of pollution visible in some plates in Figure 19. In the absence of a col, WRF-Chem suggests that a strong north-easterly flow can transfer SO_2 and SO_4 directly from the Mpumalanga Highveld to increase local concentrations and contribute to an extreme event of SO_4.

During periods of high atmospheric sulphur in Cape Town, the area could act as a source or as a sink. RegCM-Chem suggests that Cape Town is a sink during periods of extreme SO_2 in April and May, and during the extreme SO_4 events in May, July, August and September. During these months, there is a strong meridional convergence of air flow and the accumulation of sulphur, because the location of a col over Cape Town, and the convergence of northerly with southerly (or even with zonal) flow. WRF-Chem suggests that Cape Town receives more SO_4 than it exports during extreme events of SO_4 in December. On the other hand, Cape Town acts mainly as a source of SO_2 in June and August (according to the RegCM-Chem simulation), and in April, May, September and November (according to the WRF-Chem simulation).

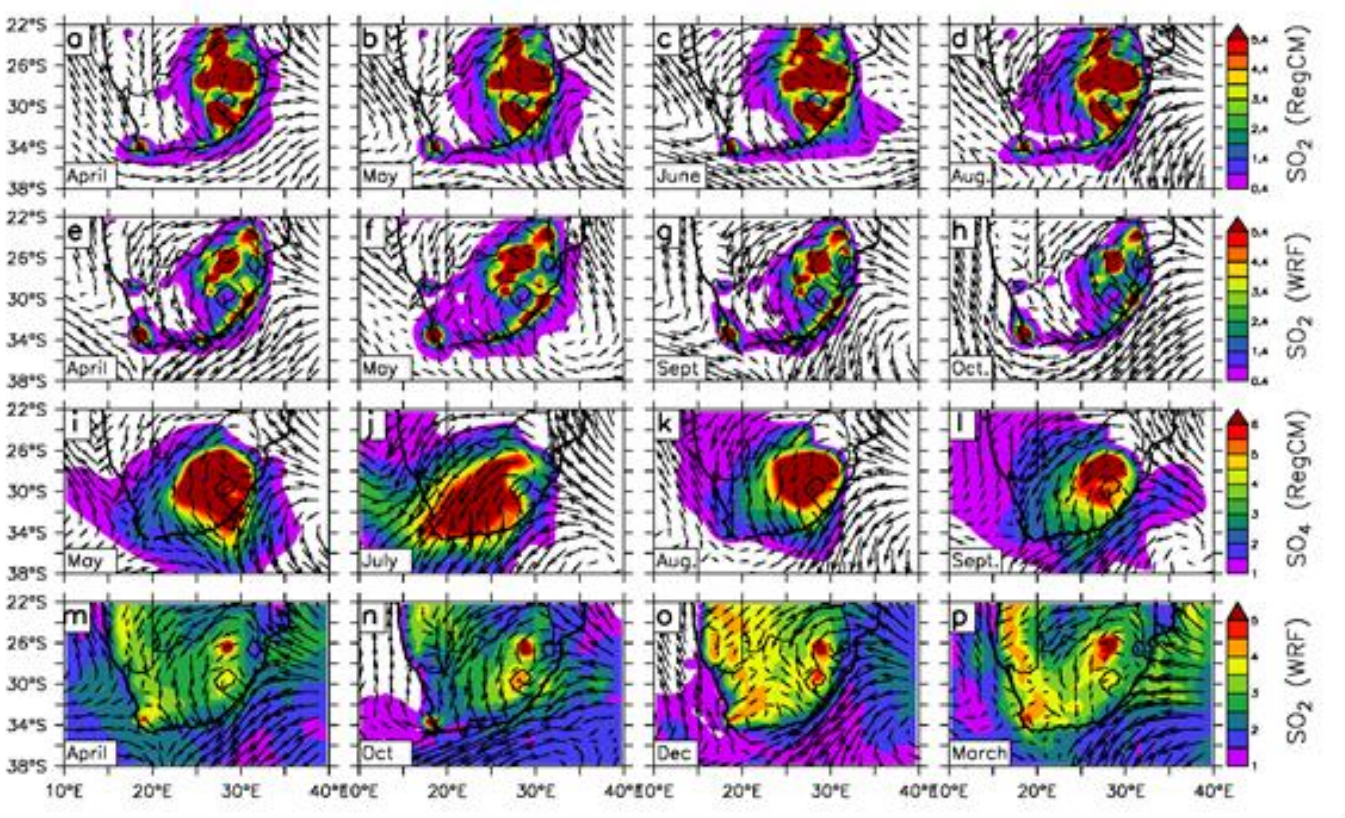

Figure 19: Composite SO_2, SO_4 concentration and wind pattern during months of sulphur extreme events over Cape Town during 2001 and 2001 as simulated by RegCM-Chem (units: x 10^{-9} kg kg^{-1}) and WRF-Chem (unit: units: x 10^{-4} ppmv)

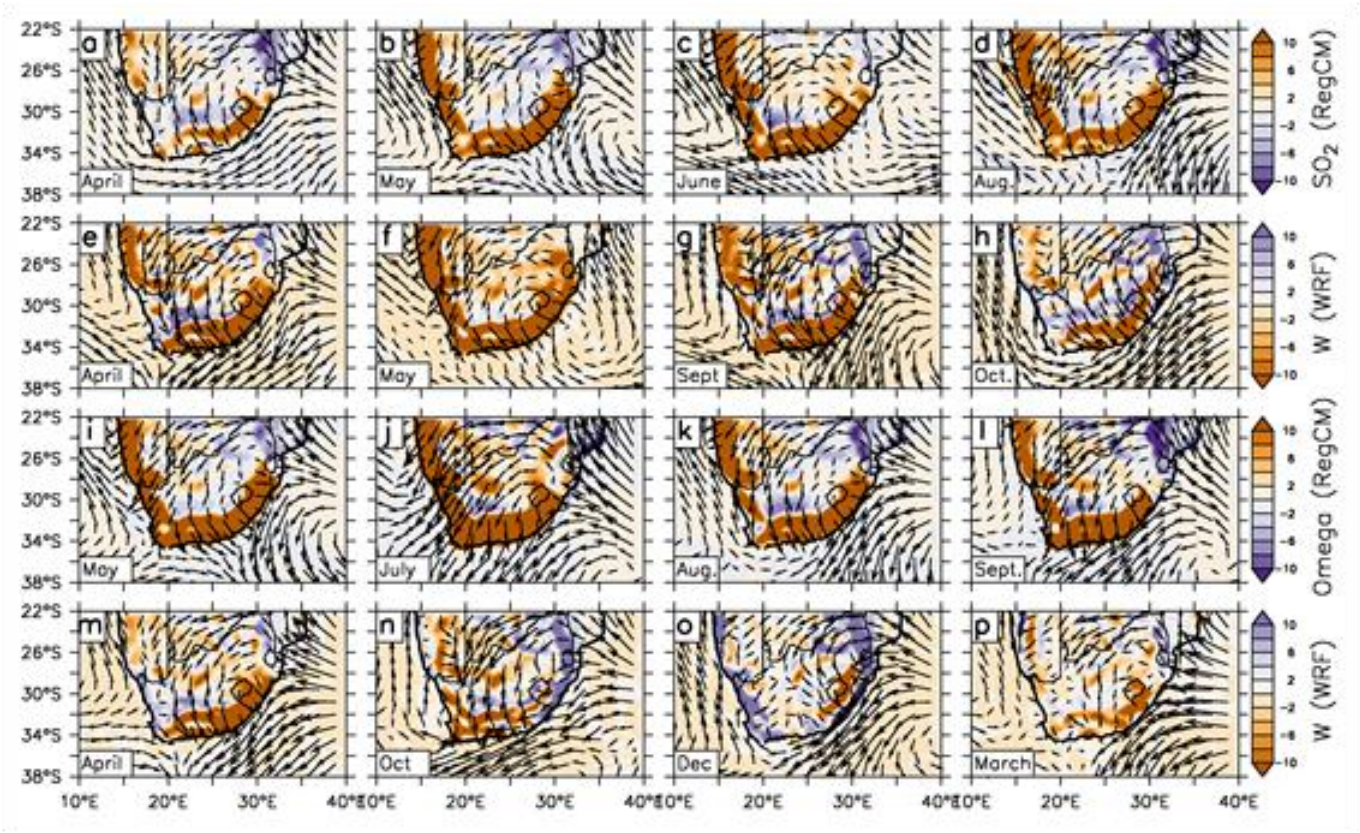

Figure 20: Composite vertical motion and wind pattern during months of sulphur extreme events over Cape Town during 2001 and 2001 as simulated by RegCM-Chem (omega; x 10^{-4} mb s^{-1}) and WRF-Chem (w; x 10^{-3} m s^{-1})

6.5 Flux budget analysis

Flux budget analysis was used to calculate net flux of the pollutants (SO_2 and SO_4) over the Cape Town box area (18.2° to 19.2°E and 33.2° to 34°S; the area described in Chapter 5 for validation) and to examine whether the city acts as a net source of or sink for the pollutants. The net flux (FNet) is defined as:

$$FNet = (FE - FW) + (FN - FS); \qquad (1)$$

where FE, FW, FN, and FS are the pollutant fluxes at the east, west, north and south boundaries of Cape Town, respectively. A positive zonal flux (FE or FW) implies a westerly pollutant flux, while a negative zonal flux means the opposite. A positive meridional flux (FN or FS) denotes a southerly pollutant flux, while a negative zonal flux means the opposite. A positive net flux indicates divergence of a pollutant over the city, meaning that the city is a net source for the pollutant. A negative net flux indicates convergence (or accumulation) of pollutants over the city, meaning that the city is net sink for the pollutant.

Figure 21 shows the climatological mean net flux (for 2001 and 2002) of SO_2 and SO_4 over the area of Cape Town. For SO_2, the RegCM-Chem simulation shows that Cape Town acts as a source in all the months; but WRF-Chem suggests that Cape Town acts as a source from September to April, and as a sink in winter (May to August; with the strongest negative flux in June), when the inflow of SO_2 by northerly flow is more than the outflow in all directions. For SO_4, RegCM-Chem depicts Cape Town as a net source during all months. However, WRF-Chem shows that Cape Town is a sink of SO_4 in July and August owing to strong convergence of SO_4 in the meridional component.

Figure 22 shows a composite of the SO_2 and SO_4 net fluxes over Cape Town area during extreme events as previously indicated (Figure 17) for each of the RegCM-Chem and WRF-Chem simulations. The flux budget indicates that periods of frequent extreme events of simulated SO_2 coincide with periods when convergence is more common; in three of the four months selected for extreme events (April, May, and August for RegCM-Chem, and May, September, and October

for WRF-Chem). The flux budget indicates that SO_4 extreme events over Cape Town coincided with convergent flow over the area in all months in the RegCM-Chem simulation (May, July, August, and September), and in two of the four months (March and December) in the WRF-Chem simulation. Convergent flow, and col, are likely attributing factors to periods of high atmospheric sulphur concentrations in Cape Town, which may be caused by local and remote sources of emissions.

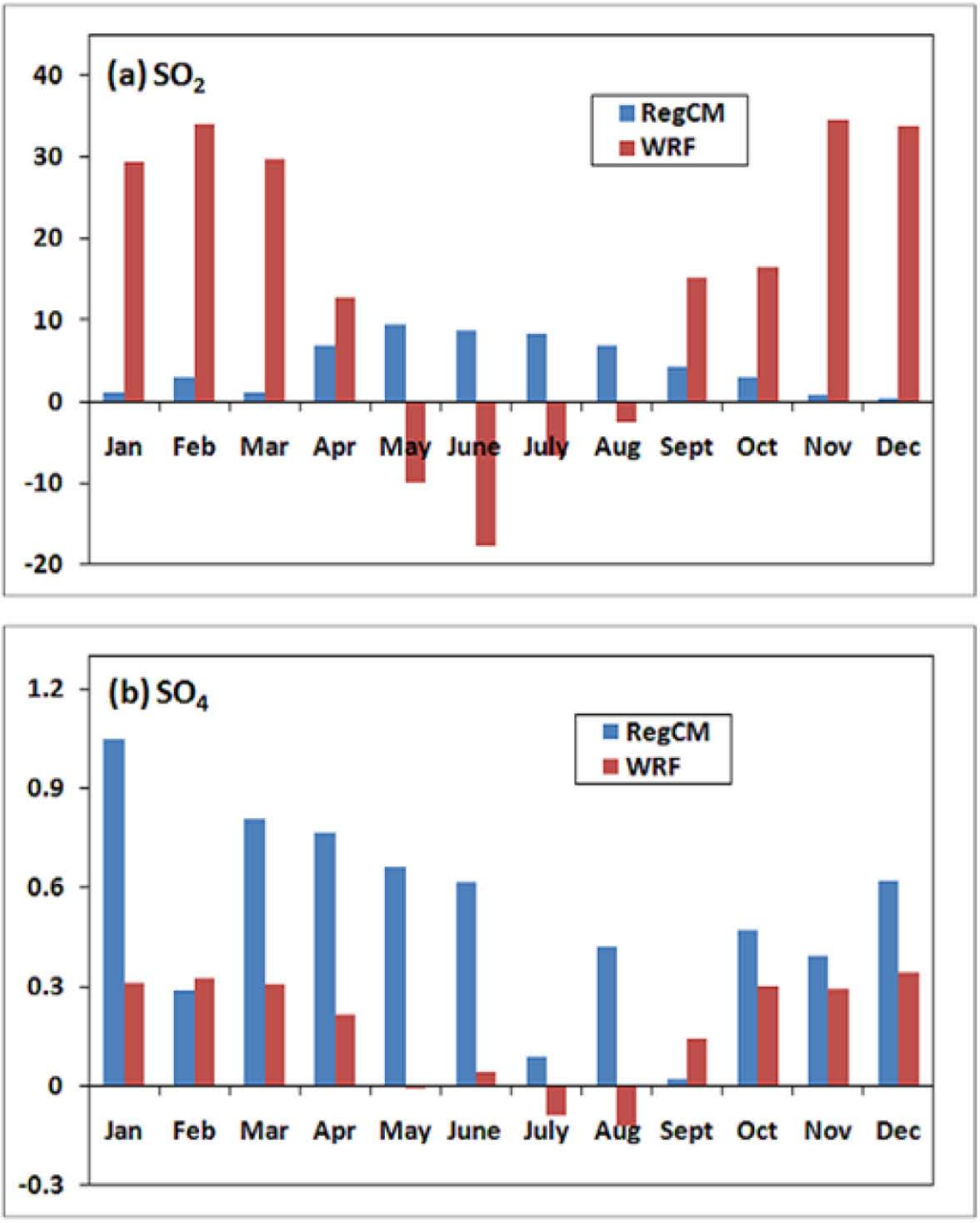

Figure 21 Monthly variation in net fluxes of (a) SO_2 and (b) SO_4 over Cape Town area as simulated by RegCM-Chem and WRF-Chem for 2001 and 2002

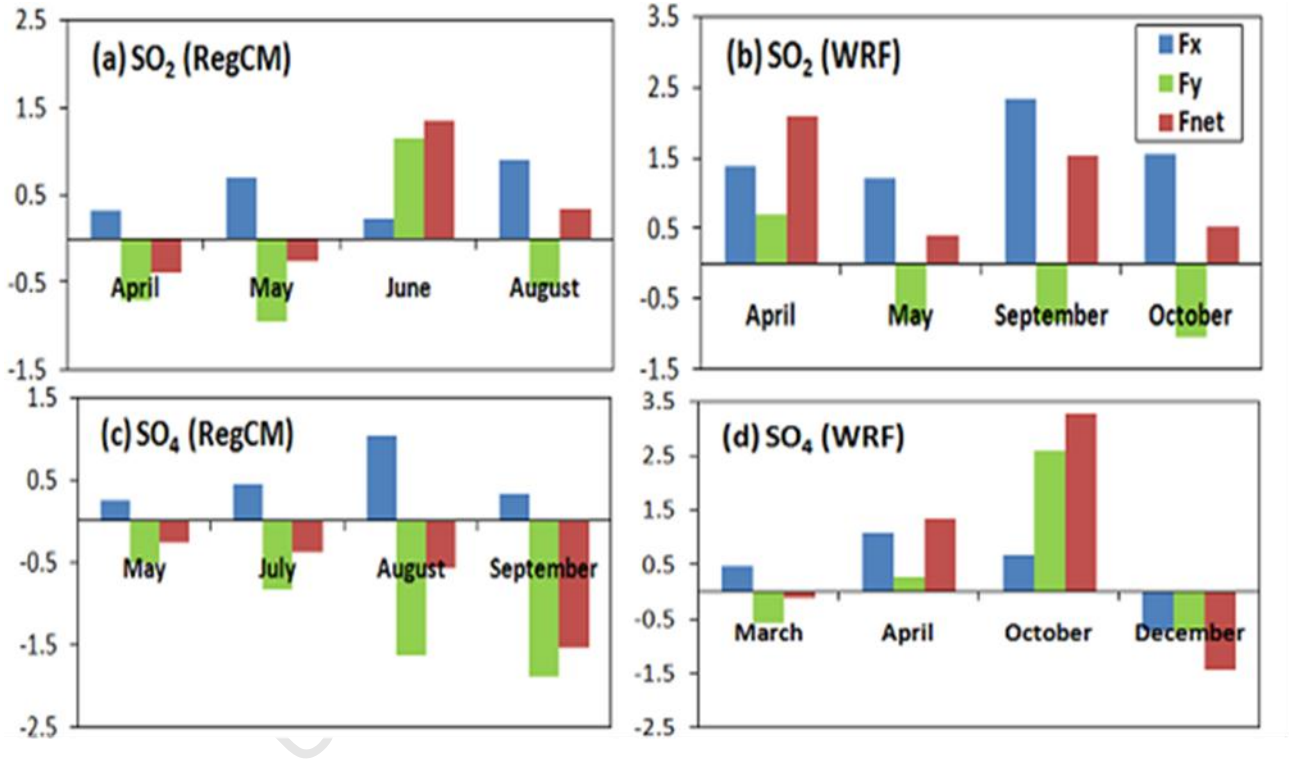

Figure 22: Composite of (a) SO_2 and (b) SO_4 net fluxes over Cape Town area during extreme events as simulated by RegCM-Chem and WRF-Chem for 2001 and 2002. Fx and Fy indicate net fluxes along zonal and meridional directions, while Fnet (= Fx + Fy) indicates convergence (negative values) or divergence (positive values) over the city area

Chapter 7: Conclusion

Anthropogenic emissions of atmospheric sulphur are harmful to the health of the environment and of people. While Cape Town regulates local industrial emissions, air quality may be affected by the transport of remote emissions over long distances to the area. To protect air quality by regulating emissions, the sources and causes of high ambient concentrations of sulphur must be identified. The purpose of this research was to establish whether local emissions are the sole source of poor air quality in Cape Town, or if remote emissions from the major source area in the country, the Mpumalanga Highveld, contribute to periods of unusually high concentrations.

The objectives of this dissertation have been achieved. It is evident that the main source of emissions in South Africa is generated over the Highveld during the time that high sulphur concentrations (extreme events) are observed in Cape Town; however, this is not conclusive that Cape Town acts as a sink for emissions from the Highveld. Following the objectives of this work:

- from analysis of observation, the seasonal variation of SO_2 in Cape Town is associated with the wind speed and most likely with the occurrence of temperature inversion. It was not possible to assess the inter-annual variation of SO_2;

- two atmospheric-chemistry models were used to simulate the meteorology and chemistry transport of sulphur pollutants over South Africa;

- the models were found to simulate quite well the seasonal characteristics of sulphur and wind speed over Cape Town (albeit better for some months than others). The models both show weakness in simulating the pattern of SO_2 for October to December, although the cause of this has not been identified;

- there are considerable unexplained differences between the two models, specifically for SO_2 in October to December and in the seasonal pattern of SO_4 concentration. The cause of the differences, might be identified if observations of SO_4 were available; and

- calculation of the sulphur flux budget over Cape Town established that the city is a net source for most of the year but, that during periods of high atmospheric sulphur in Cape Town, the area does act as a sink.

Both models used in this study agree with previous findings that concentrations of atmospheric sulphur are highest near the source of the emissions. The models consistently reproduced observed seasonal variability in atmospheric wind flow and sulphur concentrations over Cape Town.

In conclusion, the models suggest that flow may well transport atmospheric sulphur along a band of elevated concentration along the southern coastline of the country or directly from the north-east of the country to Cape Town. Both models agree that during periods of high sulphur pollution in Cape Town the corresponding wind regimes indicate either convergence or col conditions over the area. The simulation results indicate that during most months Cape Town is a net source of atmospheric sulphur, but that in some months more atmospheric sulphur is transported to than away from Cape Town, and Cape Town acts as a sink for atmospheric sulphur emitted elsewhere.

7.1 Suggestions for future research

The findings of this dissertation can be used in application of chemistry modelling to inform pollution abatement policy. Important points to consider include the following:

- The source of the inconsistencies between the models RegCM-Chem and WRF-Chem for SO_4 concentrations should be identified to contribute to model development. Work would benefit from validation of simulation results for SO_4.

- Future work would benefit from the use of a temporally differentiated emissions inventory based on observation; this would be an important development towards providing quantitative results for transported pollutants, thus improving the quality of information for air quality management strategy. This suggestion highlights the importance of work towards improving emissions inventories.

- For the analysis of transport driven by synoptic conditions and meteorological features, finer resolution simulation results can be achieved by using nesting capabilities (a finer resolution domain within, and receiving input from, a larger parent domain) within the atmosphere-chemistry transport models.

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