An integrated systems approach towards air quality management in the Vaal Triangle Priority Area

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PREFACE

Format of the Thesis

This thesis is submitted for examination in article format in accordance with the academic rules of the North-West University, in which provision is made for the article model. Work done for this thesis was carried out by the author between 2016 and 2020 and includes original data that has never been published or previously submitted for degree purposes to any university.

The conceptualisation, research, organisation and writing of the thesis and journal articles were conducted by the author. In cases where work by other researchers has been used, such work is acknowledged appropriately in the text.

Three unpublished articles are included in the thesis. Of these three, one has been submitted for review. As per requirements laid out by the North-West University regulations, a PhD candidate must submit at least one article to a reputable journal before the PhD thesis is submitted for examination.

Outline of the Thesis

This thesis is presented in seven chapters, and a description of each chapter is provided below:

Chapter 1: Introduction

This chapter includes the specific problem at hand and motivation regarding the study. The aim and objectives are presented.

Chapter 2: Literature review

This chapter gives a background to particulate air pollution. Various air quality management techniques for particulate matter are outlined.

Chapter 3: Data and methods

This chapter provides a description of the methods and data analysis used in each article.

Chapter 4: Evaluating the potential of remote sensing imagery in mapping ground-level fine particulate matter ($PM_{2.5}$) for the Vaal Triangle Priority Area.

The chapter comprises of an article that focuses on exploring the potential of satellite remote sensing as an air quality management tool for monitoring ground-level particulate matter concentrations.

Chapter 5: Updated $PM_{10-2.5}$ and $PM_{2.5}$ source apportionment for the Vaal Triangle air pollution priority area, South Africa

The chapter consists of an article which is focused on identifying the main sources of particulate air pollution, with a particular focus on low-income settlements.

Chapter 6: Integrated assessment of strategies to reduce air pollution in the Vaal Triangle Priority Area, South Africa

This chapter contains an article that is focused on employing an integrated assessment model to identify cost-effective control measures for particulate air pollution.

Chapter 7: Conclusions

This chapter discusses the conclusions of the study and highlights important findings from the study. Recommendations for future research are provided.

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Tanya and Tinashe, daddy is coming home!

“Nothing in life is to be feared, it is only to be understood. Now is the time to understand more, so that we fear less.” **- Marie Curie -**

In loving memory of my father,

Elisha Muyemeki (1961-2005),

I hope I've made you proud and became the person you wanted me to be.

ABSTRACT

The quality of air breathed in South Africa is of great concern, especially in the industrialised region of the Vaal Triangle where particulate matter (PM) concentrations are high. Long term exposure to PM is associated with serious adverse health impacts, including respiratory illnesses, cardiovascular morbidity and premature mortality. Rapid urbanisation combined with industrialisation, increase in vehicle ownership and continual use of coal and wood as primary domestic energy carriers have contributed to the deterioration of air quality in the Vaal Triangle Airshed Priority Area (VTAPA). Despite the establishment of an air quality management plan and implementation of mitigation measures, the current approach to managing air quality in the VTAPA has not produced the desired outcomes. PM levels still remain well above national ambient air standards. This is in part due to a lack of implementation oversight and allocation of resources. Financial restrictions have also made a contribution; with the current economic performance of South Africa, more cost-efficient strategies are required to manage air pollution in the VTAPA.

Air quality management is multidisciplinary in nature. An approach that examines different pollutant emissions, their contributions to atmospheric concentrations, potential control measures and their associated costs in an integrated system is therefore required. For this reason, an integrated approach to air quality management was performed for this study in order to identify alternative policy interventions that will reduce air pollution with minimal costs to the South African economy.

Monitoring is a key component of air quality management. However, the number of air quality monitoring stations distributed in the VTAPA is inadequate to capture the full-scale variability of pollutants. Adding more stations to the current monitoring network will require substantial financial resources. The paper ***"Evaluating the potential of remote sensing imagery in mapping ground-level fine particulate matter ($PM_{2.5}$) for the Vaal Triangle Priority Area"*** (Chapter 4) in this study, seeks to examine the potential of satellite remote sensing, a cheaper and more spatially descriptive alternative, in monitoring $PM_{2.5}$ across the VTAPA. The satellite remote sensing method was able to identify $PM_{2.5}$ concentration clusters consistent with known source emission regions in the VTAPA. Temporal analysis of $PM_{2.5}$ using the satellite remote sensing technique shows that concentrations are still above national ambient air standards. For satellite remote sensing to be effectively exploited as a tool for air quality monitoring in unmonitored regions in the VTAPA, further research on improving the precision and accuracy of satellite-retrieved $PM_{2.5}$ is necessary.

Source apportionment is an important process during the initial phase and review stage of air quality management as it helps in identifying the sources that need to be prioritised for control. The paper ***"Updated $PM_{10-2.5}$ and $PM_{2.5}$ source apportionment for the Vaal Triangle air pollution priority area, South Africa"*** (Chapter 5) in this study, identified the main sources contributing to PM loading in the VTAPA using the receptor modelling technique. It was found that although the VTAPA is a highly industrialised region, localised sources may have a greater impact on PM loading in low-income settlements. Source contributions varied across low-income settlements, and it is recommended that individual plans will need to be designed to manage air quality in these areas. Decision-makers will need to prioritise dust, industry, domestic coal burning, wood and biomass combustion sources in the new air quality management plan for the VTAPA.

The paper *"**Integrated assessment of strategies to reduce air pollution in the Vaal Triangle Priority Area, South Africa"*** (Chapter 6) in this study, uses the GAINS model to integrate data on pollutant emissions, their contributions to atmospheric concentrations, potential control measures and their associated costs so as to devise a management strategy that effectively reduces PM levels in the VTAPA to within national acceptable standards. Comparisons between the current and alternate management strategy showed that significant air quality improvements would not take place in the medium term if the policy interventions that are presently there for the VTAPA were to continue. Implementation of more stringent controls will lower emissions by more than half, and decrease concentrations to within recommended air quality limits. The negative health impacts from PM exposure are likely to be drastically reduced in the near future if a stricter approach is adopted. However, operational costs for this alternative management strategy will almost be doubled in comparison to the current approach.

This study makes an important contribution to the current body of knowledge by identifying a more cost-effective approach to air quality management in the VTAPA. It is recommended that future studies should include climate change policies in order to identify air pollution reduction measures that have the co-benefit of reducing greenhouse gas emissions.

Keywords: Integrated assessment modelling, source apportionment, GAINS model, satellite retrievals, VTAPA

LIST OF ABBREVIATIONS

ADDAS	Airborne Dust Dispersion Model from Area Sources
AOD	Aerosol Optical Depth
APPA	Atmospheric Pollution Prevention Act
AQMP	Air quality management plan
AS	Alternative Scenario
$C_2O_4^{2-}$	Oxalate ion
Ca^{2+}	Calcium ion
CH_3COO^-	Acetate ion
CH_4	Methane
Cl^-	Chloride ion
CLE	Current Legislation Scenario
CMB	Chemical Mass Balance
CO	Carbon monoxide
CSIR	Council for Scientific and Industrial Research
DEA	Department of Environmental Affairs
DMS	Dimethyl sulphide
EC	Elemental carbon
EMEP	European Monitoring and Evaluation Programme
ERI	Emission Reduction Impacts
ESP	Electrostatic precipitators
GAINS	Greenhouse Gas and Air Pollution Interactions and Synergies model
GCR	Gauteng city-region
GDP	Gross domestic product
GWR	Geographically Weighted Regression
HCl	Hydrochloric acid
$HCOO^-$	Formate ion
HNO_3	Nitric acid
IAM	Integrated Assessment Modelling
IC	Ion Chromatography
IIASA	International Institute for Applied Systems Analysis
INC	Incremental
IOA	Index of agreement
K^+	Potassium ion
kt	kiloton
LPG	Liquefied petroleum gas
MB	Mean Bias
Mg^{2+}	Magnesium ion
MISR	Multi-angle imaging spectroradiometer
MODIS	Moderate resolution imaging spectroradiometer
MSW	Municipal solid waste
MT	Mass-Transfer
Na^+	Sodium ion

NAAQS	National Ambient Air Quality Standards
NAEIS	National Atmospheric Emission Inventory System
NEMAQA	National Environmental Management: Air Quality Act
NH_3	Ammonia
NH_4^+	Ammonium ion
NO_2	Nitrogen dioxide
NO_3^-	Nitrate ion
NO_X	Oxides of nitrogen
OC	Organic carbon
PAF	Population attributable fraction
PM	Particulate matter
$PM_{10-2.5}$	Coarse fraction of particulate matter
$PM_{2.5}$	Fine fraction of particulate matter
PMF	Positive Matrix Factorization
REF	Reference year
RR	Relative risk
SAAQIS	South African Air Quality Information System
SANRAL	South African National Roads Agency SOC Ltd
SeaWIFS	Sea-viewing Wide Field-of-view Sensor
SEM	Scanning electron microscopy
SO_2	Sulphur dioxide
SO_4^{2-}	Sulphate ion
SPARTAN	Surface particulate matter network
SSA	Sub-Saharan Africa
µg	Microgram
US-EPA	U.S. Environmental Protection Agency
UTM	Universe Transverse Mercator
VKT	Vehicle kilometre travelled
VOC	Volatile organic compounds
VTAPA	Vaal Triangle Airshed Priority Area
WFGD	Wet flue gas desulphurisation
XRF	X-Ray Fluorescence
yr	Year

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CHAPTER 1 INTRODUCTION

1.1 Background to air quality management in South Africa

Air pollution in South Africa has long been recognised as a serious threat to humans and the environment (Naiker *et al.*, 2012). The country's heavy reliance on coal has not only contributed to economic and social development, but it has also led to high emissions of greenhouse gases per capita, and criteria air pollutants including sulphur dioxide (SO_2), oxides of nitrogen (NO_x) and particulate matter (PM) (Klausbruckner *et al.*, 2018). Across many parts of South Africa, especially in urban areas, PM air pollution is of great concern (Altieri & Keen, 2019). Sources of PM include industries, coal and wood burning in residential areas, mining, transport, biomass burning and fugitive dust. Air quality management in South Africa is hinged on the fundamental rights that guide legislation and policy. These rights entitle South Africans "*to an environment that is not harmful to their health or well-being*" and *"to have the environment protected, for the benefit of present and future generations*" (South Africa, 2013). In order to guarantee this, air pollution levels within the country must be kept within acceptable limits that have minimal health effects.

From 1965 to 2004, air pollution in South Africa was managed through the Atmospheric Pollution Prevention Act, 1965 (APPA). This act adopted a "best practicable means" approach towards controlling air pollution in which maximum emission limits were not enforced (Naiker *et al.*, 2012). Under APPA, provisions were made for the control of certain industrial activities using a registration process. Mechanisms for the control of vehicles, dust emissions from mining and smoke control from fuel-burning appliances were also provided. The registration certificates provided to industries outlined the stipulated conditions under which the scheduled activity could take place. These certificates neither imposed emissions limits to emitters nor did they provide a clear outline for controlling air pollution (Winstanley, 2010). APPA was regarded as an ineffective legislative measure in managing air pollution in South Africa due to the following reasons:

- APPA focused on individual source emissions control instead of having and achieving ambient air quality standards and thus disregarded the cumulative effects of air pollution.
- APPA was mainly targeting point source emissions from specific industries instead of addressing pollution from all sources.
- APPA did not set targets or standards for air quality.
- APPA had inefficient mechanisms to ensure compliance and enforcement of control measures.

In response to these challenges encountered with APPA, the South African government made legislative changes which resulted in APPA being repealed and substituted with the National Environmental Management: Air Quality Act, 2004 (NEMAQA). This new legislation saw a shift in policy from source-based air pollution control to an outcomes-based approach aligned with international best practises (Centre for Environmental Rights, 2017). The provisions of NEMAQA are outlined below:

- NEMAQA provides the assessment of the state of air quality through monitoring of ambient concentrations
- NEMAQA provides the government with an important regulatory tool that allows for the declaration of regions exceeding ambient air quality guidelines as priority areas.
- NEMAQA provides for the establishment of ambient and emissions standards.
- NEMAQA provides for the development of air quality management plans at each government level.

Efforts to manage air quality in South Africa through the regulatory mechanisms specified in NEMAQA have been made. The Highveld, Vaal Triangle and Waterberg-Bojanala areas were identified as regions exceeding ambient air quality guidelines and declared priority zones (South Africa, 2013). Air quality management plans were developed for these priority areas. Minimum emission standards for PM, SO_2 and NO_x were established in 2010 and revised in 2013 (Klausbruckner *et al.*, 2016). National ambient air quality standards (NAAQS) for pollutants were set up and enforced in all spheres of government. However, despite these and other undertakings air quality management in South Africa still remains a challenge, as witnessed by the observed high levels of PM in priority areas (Altieri & Keen, 2019). Inadequate political buy-in at all spheres of government has led to decision-makers giving less attention to air quality issues and allocating fewer resources (Tshehla & Wright, 2019). The inconsistency in compliance report checking and the lack of follow-ups to non-compliance by authorities have also contributed to air quality objectives not being met (Department of Environmental Affairs, 2019).

Improvements in the management of air quality in South Africa will require the weighing of environmental, economic, and social interests in decision making through assessing the multiple dimensions of air pollution (van Bree *et al.*, 2007). This will enable the development of effective air quality policies required in order to meet national regulatory standards and legislation. However, this has to be accompanied by proper monitoring, adequate implementation and consistent reviewing of air quality management plans.

1.2 Motivation

After being declared a priority area in terms of the NEMAQA, an air quality management plan (AQMP) detailing possible intervention strategies for the Vaal Triangle Airshed Priority Area (VTAPA) was published on May 29, 2009 (Department of Environmental Affairs and Tourism, 2009). A mid-term review of the VTAPA AQMP in 2013, revealed that in spite of the regulatory measures put in place, air pollution continues to be a problem in this region (Department of Environmental Affairs, 2013). Although air quality in this priority area has improved, PM concentration levels still remain above the national ambient air quality standards and global health guidelines (Altieri & Keen, 2019). Substantial investments are therefore needed to further reduce emissions and decrease the remaining health risks associated with air pollution. However, financial constraints are one of the reasons that the VTAPA AQMP failed to meet the desired outcomes in the first place (Scorgie, 2012). With the current economic climate in South Africa, it is necessary to come up with health-effective and cost-efficient control strategies to further improve air quality in a cost-effective manner.

An integrated approach that assesses the technological, economic, ecological and public health aspects of air pollution is required. Such an approach should be able to evaluate future air quality based on the assessment and comparison of different air quality policies and control strategies taking into account economic costs (Amann *et al.*, 2011; Rafaj *et al.*, 2018). Even in times of economic austerity, this approach is capable of designing cost-effective mitigation measures (Reis *et al.*, 2012). In the case of air quality management in the VTAPA, data on emissions from different pollutants, their contributions to concentrations and exposure, potential mitigation measures and their associated costs will be analysed together for this study, with a special focus on PM. This will help in identifying which PM source types need to be prioritised, which control strategies to put in place and their cost implications to the economy of South Africa.

1.3 Aim and objectives

This study aims to perform an integrated assessment of air quality in the Vaal Triangle Airshed Priority Area.

The objectives of the study are:

1. To explore the spatial variations and temporal trends of particulate matter concentrations in the Vaal Triangle Airshed Priority Area
2. To identify and quantify the relative contributions of sources contributing to particulate matter loading in low-income settlements of the Vaal Triangle Airshed Priority Area

3. To assess the potential impacts of emission reduction strategies on air quality in the Vaal Triangle Airshed Priority Area.

CHAPTER 2 LITERATURE REVIEW

2.1 Particulate air pollution

Particulate air pollution can be described as the suspension in the air of a mixture of small solid particles and liquid droplets, in quantities that can be harmful to human or animal life (Tan, 2014). These particles constitute of a wide range of matter including combustion end products such as fumes, soot and smoke, and natural particulates such as aerosolised sea salt, wind-blown dust, debris from decomposing plant and animal life, pollen grains, fungal spores and particles from volcanic and geothermal eruption (Godish, 2004). These particles, when inhaled, can lead to adverse health outcomes such as respiratory illnesses and premature mortality (Liu *et al.*, 2016).

2.2 Characteristics of particulate matter

2.2.1 Nature of particulate matter

Based on the way that they are formed, particle pollutants can be classified into primary and secondary particles. Primary particles occur as a result of direct emission from the source such as unpaved roads, construction sites, stone crushing, smokestacks, waste burning and forest fires (Vallero, 2014). The concentration of primary particles in the atmosphere is controlled by the rates of emission, transportation, dispersion and removal of these particles in the atmosphere (Popoola *et al.*, 2018). Secondary particles emanate from the reaction of precursor gases (SO_2, NO_x, NH_3) and non-methane volatile organic compounds (VOC) in the atmosphere (Tan, 2014). This will result in either the development of new particles or the growth of existing particles. The formation of secondary particles in the atmospheric environment is influenced by the concentrations of precursor gases and other gaseous reactive species (e.g., ozone, hydroxyl radical), conditions in the atmosphere, and interactions with cloud or fog droplets (Steinfeld & Pandis, 1998).

In comparison to primary particles, secondary particles have a longer residence time in the atmosphere as they are smaller (Popoola *et al.*, 2018). In the atmosphere, primary and secondary particles experience significant physical and chemical transformations as a result of interactions between particles, gaseous reactions, clouds and rain, and photochemical reactions (Tomasi & Lupi, 2017). This results in incessant changes to their particle size distribution and chemical composition. Primary particle effects are mostly localised, whilst the effects of secondary particles are mainly regional and experienced over a wide area (Vallero, 2014).

2.2.2 Physical properties

These include the size distribution and mass concentration of particulates. Size distribution is an important attribute as it is the main factor that determines the behaviour of PM. Size distribution

influences the transportation and removal of particles in the atmosphere, and the toxicity of particles towards human beings (Wiseman & Zereini, 2011). Particle size is usually expressed in diameters. However, as particles are not necessarily spherical and have varying densities they are defined in terms of an equivalent diameter based on the particle behaviour in a divergent airflow, a property which is known as the *aerodynamic diameter* (Godish, 2004).

The size for particles spans over a wide scale, from as small as 0.005 µm to as large as 100 µm diameter (Finlayson-Pitts & Pitts, 2000). However, the suspended fraction is essentially less than 40 µm (Steinfeld & Pandis, 1998). PM is classified into two main categories based on aerodynamic diameter. This includes fine particles (small particles) with an aerodynamic diameter ranging between 0.1 µm and 2.5 µm and coarse particles (large particles) which have an aerodynamic diameter ranging from 2.5 µm to 10 µm (Anderson *et al.*, 2012). The fine and coarse particles together have a bimodal size distribution in which the minimum concentration lies between 1 and 3 µm. These particles have varying characteristics (Table 2-1).

Table 2-1: Comparison between coarse and fine particulate matter properties (modified from Panyacosit, 2005).

	Coarse particles	**Fine particles**
Formation and sources	Break-up of solids and droplets Erosion of land Suspension of dust Re-suspension of road debris (tyre/brake wear) Ocean spray Ash (black smoke) from uncontrolled combustion Construction and demolition Disturbance of surfaces (agriculture, mining, quarrying, unpaved roads) Biogenic emissions (pollen, fungal spores)	Condensation of atmospheric gases Coagulation of ultrafine particles Reactions of component gases of particles Evaporation of water droplets containing dissolved gases Combustion of fossil and biomass fuels Industrial processes (smelters, refineries, steel mills, mining)
Composition	Organic and elemental carbon Sulphates Nitrates Chlorides Oxides of crustal elements Sea salt Plant and animal debris Bacteria	Organic and elemental carbon Sulphates Nitrates Ammonium Metals Organic compounds Water Bacteria Viruses
Physical characteristics	Large mass	Large surface area High particle number
Spatial/temporal variability	High	Low
Atmospheric lifetime	Minutes to days	Days to weeks

Distance travelled	Usually <10 km	100s to 1 000s km
Removal process	Gravitational deposition Scavenging by rain	Cloud droplet formation

Coarse particles normally occur as a result of different mechanical forces such as abrasion and fragmentation (Gieré & Querol, 2010). These particles have a higher mass concentration as compared to fine particles and generally comprise of earth crust materials and fugitive dust from roads and industries (Vallero, 2014). Coarse particulates include materials such as mineral dust, sea salt, biogenic particles such as pollen and fungal spores. Due to their large grain size, these particles have high settling velocities, leading to shorter residential time in the atmosphere. The duration of large particles in the atmosphere is short (minutes to days) with a small distance range of 1 to 10 km (Hinds, 1982). During days of high wind activity, the fallout of coarse particles is offset by re-entrainment.

Fine particles have a greater particle number distribution than larger particles and consist of converted precursor gases, combustion particles and, organic and metal vapours that have condensed (Akbar & Kojima, 2003). These smaller particles have a longer lifespan in the atmosphere, from days to weeks and can travel long distances (100 to 1 000 km). Fine particles can be divided into three size fractions or modes: Nucleation, Aitken and accumulation modes (Figure 2-1).

Nucleation mode particles have diameters ranging from 1 nm (0.001 µm) to 10 nm (0.01 µm). Particles in this mode are formed through the condensation of hot vapours (e.g. flue gas) or through the nucleation of atmospheric gases (Steinfeld & Pandis, 1998). Aitken mode particles have diameters ranging from 10 nm (0.01 µm) to 100 nm (0.1 µm) and are formed through gas-to-particle transformations and hot vapour condensation on nucleation mode particles (Gieré & Querol, 2010). Due to their minuscule size, particles in the nucleation and Aitken mode have the highest particle numbers but make up for only a marginal fraction of the total PM volume or mass (Steinfeld & Pandis, 1998). Particles in the accumulation mode are the largest fine-size portion having diameters that range from 0.1 µm to 2.5 µm. These particles form as a result of the coagulation of particles in the smaller mode or through condensation of vapours on existent particles (Godish, 2004).

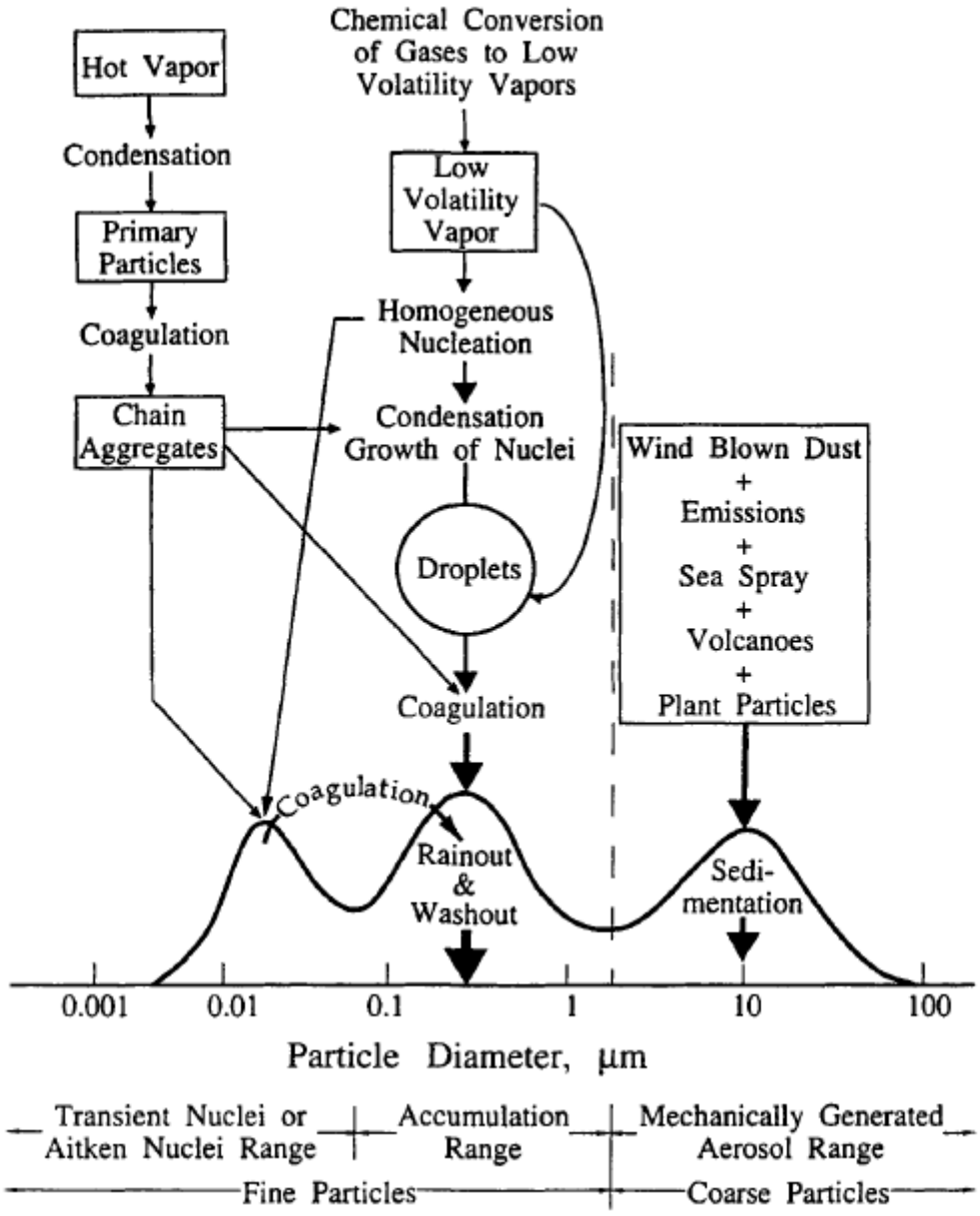

Figure 2-1: Typical size distribution of particle surface area of an atmospheric aerosol (Source: Seinfeld & Pandis, 2016)

2.2.3 Chemical properties

Understanding the chemical species that make up PM is important in identifying and quantifying sources of atmospheric particulate pollution, as well as the health risk towards human beings (Stanek *et al.*, 2011). The chemical composition of atmospheric particles is dependent on the source emission and the processes which drive the formation of these particles (Tomasi & Lupi, 2017). Atmospheric PM can be divided into the following categories:

(i) Sea-salt particles: This is the main component of marine PM. They occur when sea-spray droplets evaporate into the atmosphere. Sea-spray droplets form when air bubbles inside

whitecaps burst due to wind entrainment, and due to the tearing of droplets from wave surfaces. Sea salt is predominantly composed of ions of chloride (Cl^-), sodium (Na^+), and sulphate (SO_4^{2-}), with some magnesium (Mg^{2+}) (Tang *et al.*, 1997).

(ii) Industrial dust, mineral dust, volcanic ash and dust: These entities have varying compositions (e.g. silicates, oxides, sulphates, carbonates, alloys, glass) as their generation processes and sources differ (Seinfeld & Pandis, 2016).

(iii) Primary biogenic aerosol particles: This consists of biological materials (e.g. bacteria, fungal spores, pollens, plant fragments and debris) which are mainly coarse in size (Després *et al.*, 2012). These materials are composed mainly of organic carbon and other elements (e.g. potassium, phosphorus, silicon) in minor quantities.

(iv) Combustion-derived carbonaceous particles: These particles are mainly composed of elemental carbon (EC) and organic carbon (OC). OC is a complex mixture constituting of a wide range of organic compounds that form as a result of biomass burning (of wood, leaves, crops and forests) or through the condensation of less volatile gases. Elemental carbon (which can also be referred to as black carbon, graphitic carbon, or soot) consists of carbonaceous materials which have a high solar radiation absorption capacity (Seinfeld & Pandis, 2016). EC forms through different combustion processes.

(v) Secondary inorganic aerosol particles: These comprise of salts (e.g. NO_3^-, SO_4^{2-}, and NH_4^+) that form from precursor gases originating from natural (e.g. marine dimethyl sulphide (DMS)) or anthropogenic (e.g. SO_2, NO_x, NH_3, HCl) sources (Allen *et al.*, 2019). The reaction between gaseous precursors and mineral dust or sea-salt particles may result in the presence of different cations in small quantities.

(vi) Secondary organic aerosol particles: These particles form in the atmosphere through the chemical reaction of organic gases (VOCs) that originate from biogenic (mostly forests and phytoplankton) and anthropogenic sources (Hallquist *et al.*, 2009).

2.3 Sources of particulate matter in the VTAPA

The current air quality management plan for the VTAPA has identified the following sources as important contributors to particulate matter loading:

2.3.1 Industries

Air pollution from industries is a major environmental concern in the VTAPA. Industries account for over 80% of SO_2 and NO_x, and 60% of PM_{10} emissions in the VTAPA (Department of Environmental Affairs, 2013). Most of these industries are found in the Vanderbijlpark,

Vereeniging, Sasolburg and Meyerton areas. These industries include the iron and steel and ferroalloy sector, and petrochemical sector. Coal, coking coal and heavy fuel oil are the main energy sources used to drive these industries and have contributed significantly to particulate emissions in the VTAPA (Department of Environmental Affairs, 2013).

The main iron and steel manufacturers in the VTAPA are ArcelorMittal steel (Vanderbijlpark and Vereeniging Works) and Davsteel (Cape Gate). Samancor Metalloys (Meyerton) is the only ferroalloy facility found in the region. Ca, Al, Mg, Si, K, Fe, Mn and Zn are typical marker elements found in metal smelters (Taiwo *et al.*, 2014). Sasol Chemical Industries, Natref and Omnia fertilisers are the main petrochemical facilities that are located in the Sasolburg area (Department of Environmental Affairs and Tourism, 2009). Trace elements emitted from petrochemical industries based on crude oil refining include metals such as Ni and V (Luo *et al.*, 2018).

2.3.2 Power generation

Eskom's Lethabo power station is the sole large power generation source in the VTAPA. This power plant operates on large quantities of low-grade coal to generate electricity (Department of Environmental Affairs and Tourism, 2009). The burning of low-quality coal at Lethabo has resulted in over 8 000 tonnes of PM_{10} being emitted on an annual basis (Pretorius *et al.*, 2017). Despite being retrofitted with electrostatic precipitators (ESP) to reduce particulate emissions, Lethabo routinely experiences upset conditions as it is operating past its designed lifespan. A health risk assessment study for the Vaal Triangle-Highveld Priority Areas show that, in 2016, exposure to $PM_{2.5}$ pollution from Lethabo power station contributed an estimate of 57 to 122 premature deaths (Gray, 2019). Products released from coal-fired power stations include SO_2, NO and Cl^- (Qiu *et al.*, 2016; Zou *et al.*, 2017). When released into the atmosphere, SO_2 and NO can undergo secondary transformation to form NO_3^- and SO_4^{2-}.

2.3.3 Domestic fuel burning

Although the majority of the houses in the VTAPA are electrified, households continue to burn fuel to meet part of their energy demands as electricity is costly. The majority of the population resides in low-income settlements, which include Evaton, Sebokeng, Sharpville, Boipatong, Bophelong and Zamdela who cannot afford to consistently pay for the high monthly electricity tariffs (Department of Environmental Affairs and Tourism, 2009). Low-grade coal, wood and paraffin are deemed to be cheaper alternatives for cooking, lighting and space heating. Apart from being low-priced, access to coal is easy as low-income settlements in the VTAPA are located within the vicinity of coal mines and local coal traders. Domestic coal and wood burning are an important source of PM, especially during stable winter months, when there is a strong need for space heating (Barnes *et al.*, 2009). Diurnal and seasonal characteristics of residential burning

show that combustion activities peak during the early morning and evening, and during stable winter months as a result of increased demand for space heating (Nkosi *et al.*, 2017).

Although PM emissions from the residential sector are significantly lower (8% of PM_{10} emissions) than those from industries and the power generation sector, the PM exposure levels from domestic fuel burning are considerably higher as particulates are emitted at significantly lower heights and in enclosed dwellings where people live (Department of Environmental Affairs, 2013; Department of Environmental Affairs, 2019). Domestic fuel burning has significant negative effects on the health of humans. An estimated 540 000 people die prematurely each year in sub-Saharan Africa due to household air pollution exposure (International Energy Agency, 2017). Chemical profiles from coal combustion in households, other than the carbonaceous species that dominate, usually consist of S, Cl^-, NO_3^- and SO_4^{2-}, while those from wood-burning contain an abundance of K^+ ions (Matawle *et al.*, 2014).

2.3.4 Fugitive Dust

Fugitive dust is defined as dust emitted into the atmosphere through an unrestricted flow stream (Pianalto & Yool, 2013). The prevailing dry climatic conditions in the urban environs of South Africa makes fugitive dust a potential nuisance. Dust generated through abrasion and entrainment can be a traffic safety concern as these particles obscure visibility for vehicles (Van Pelt & Zobeck, 2007). This dust can also be toxic to humans as it may contain pollutants originating from anthropogenic sources. A study into the composition of road dust in the Tshwane Metropolitan region of South Africa reported elevated levels of trace metals which include Cd, Pb, Cu, Zn, Mn and Cr (Okonkwo et al., 2006). These potentially lethal substances were introduced into the soil via deposition of pollutants emitted by industries and vehicle exhausts. For the Vaal Triangle, a source inventory performed in the 1990s also identified fugitive dust as one of the main sources of PM (Annegarn & Scorgie, 1997).

2.3.5 Mines and ash dumps

Three mines are located in the VTAPA, New Vaal Colliery, Sigma Colliery and Glen Douglas Dolomite Mine. These mines extract coal through the open cast method. Emissions of fugitive dust are the main source of PM from these mines, from activities such as drilling and blasting, bulldozing, materials handling and vehicle entrainment on unpaved roads (Department of Environmental Affairs and Tourism, 2009). The fugitive dust comprises mineral matter with elemental composition Al, Si, K, Ca and Fe, and coal dust opencast mining operations may also emit SO_2, CO and NO_x pollutants from spontaneous combustion in tailings storage facilities (Walton & Ngcukana, 2009). Ash dumps and coal stockpiles at Eskom's Lethabo power station are a source of fugitive dust.

2.3.6 Vehicle emissions

The increased transport demand as a result of the rising population in urban areas has led to a rise in vehicle numbers in South Africa. Based on previous baseline assessments, emissions from vehicles in the VTAPA is an important source of air pollution (Thomas, 2008). Vehicles are the second highest contributors to NO_x air pollution in the VTAPA, accounting for 11% of emissions (Department of Environmental Affairs, 2013). PM, SO_2 and NO_x emissions from vehicles are as a result of fuel combustion, particularly from diesel engines. Diesel exhausts are classified as carcinogenic to humans (Silverman, 2017). Particulates can also be released as a result of the wearing and tearing of brakes and tyres, and these particles usually contain Zn, Mn and Fe (Jan Kole *et al.*, 2017). Apart from combustion PM can also be released into the atmosphere through the re-entrainment of road dust by vehicles.

2.3.7 Waste burning

Emissions from waste combustion are of concern in the high population density areas of the VTAPA. Low-income settlements are associated with high volumes of waste being generated. Refuse collection in these areas is infrequent, which forces residents to find alternative means of disposal apart from through formal landfills (Department of Environmental Affairs and Tourism, 2009). Options include burying refuse, discarding of litter in open spaces and the unsanctioned burning of waste (Department of Environmental Affairs, 2013). The latter activity is more detrimental to air quality as particulates can be released through waste burning. The type of waste burned is important as carcinogenic heavy metals such as Cd and Cr can be released into the atmosphere (Verma *et al.*, 2016). These heavy metals are emitted as a result of the burning of discarded electronic devices such as cellphones, remote controls and computer monitors (Gangwar *et al.*, 2019).In low-income residential areas, waste is usually not sorted before disposal (Oelofse *et al.*, 2018). Domestic waste can end up being mixed with garden waste. In addition, domestic waste is usually dumped in open spaces where there is vegetation. This can lead to waste burning mixing being accompanied by biomass burning. Waste in the form of plastics, papers as well as tyres is usually openly burned in open spaces and near minibus stations by street vendors, especially during winter for warmth (Rodseth *et al.*, 2020).

2.3.8 Biomass burning

Largescale agricultural burning in the VTAPA plays a significant role in ambient air quality, especially during drier winter months. Combustion of organic plant material results in large amounts of air pollutants being emitted in a short time span. These pollutants include CO, CH_4, NO_x, SO_2, PM and VOCs (Department of Environmental Affairs, 2019). Biomass burning also takes place in the domestic and municipal setting so as to control grass encroachment. Chemical

profiles for biomass burning are usually characterised by a high abundance of K and K^+ and minor contributions of SO_4^{2-} and NO_3^- (Chuang *et al.*, 2016).

2.4 Health impacts of particulate matter

Epidemiological research into the effects of air pollution has identified PM as a major health threat (Liu *et al.*, 2016). The presence of inhalable particulate matter in the atmosphere in high concentrations could expose humans to respiratory allergies and cardiovascular diseases. There is strong evidence to support that PM is a source of several health problems including increased severity in the symptoms of asthma and pulmonary diseases, reduced lung performance, heart-related complications and premature mortality (Feng *et al.*, 2016). The health effects of PM depend on the chemical composition and physical size of particles as well as length of exposure (Kim *et al.*, 2015).

Generally, coarse particles do not pass the upper respiratory tract when inhaled, as they tend to settle quickly on the trachea or the bronchi (Atkinson *et al.*, 2010). Fine particles, however, can enter considerably deep into the lower respiratory tract and lodge in the alveoli where toxins attached to the fine particles can be released and absorbed into the bloodstream (Fu *et al.*, 2011). An overview of some of the adverse health effects associated with PM inhalation is provided in Figure 2-2.

Chemical composition plays an important role in determining the toxicity of particulates (Davidson *et al.*, 2005). Particulates containing transition metals, including iron, can cause damage to the pulmonary tissue by increasing the production of reactive oxygen species in human cells (Valavanidis *et al.*, 2008). Other heavy metals, such as lead, are associated with cardiovascular diseases, specifically hypertension (Mordukhovich *et al.*, 2012).

The health effects of PM vary can according to the duration of exposure, which can either be short-term (hours, days) or long-term (months, years). Short-term exposure is associated with eye irritations, respiratory illnesses and increased hospital admissions for ischemic heart disease and heart failure (Morris, 2001). Health outcomes as a result of long-term exposure to PM include a greater risk of lung cancer and cardiovascular morbidity and mortality (Anderson *et al.*, 2012).

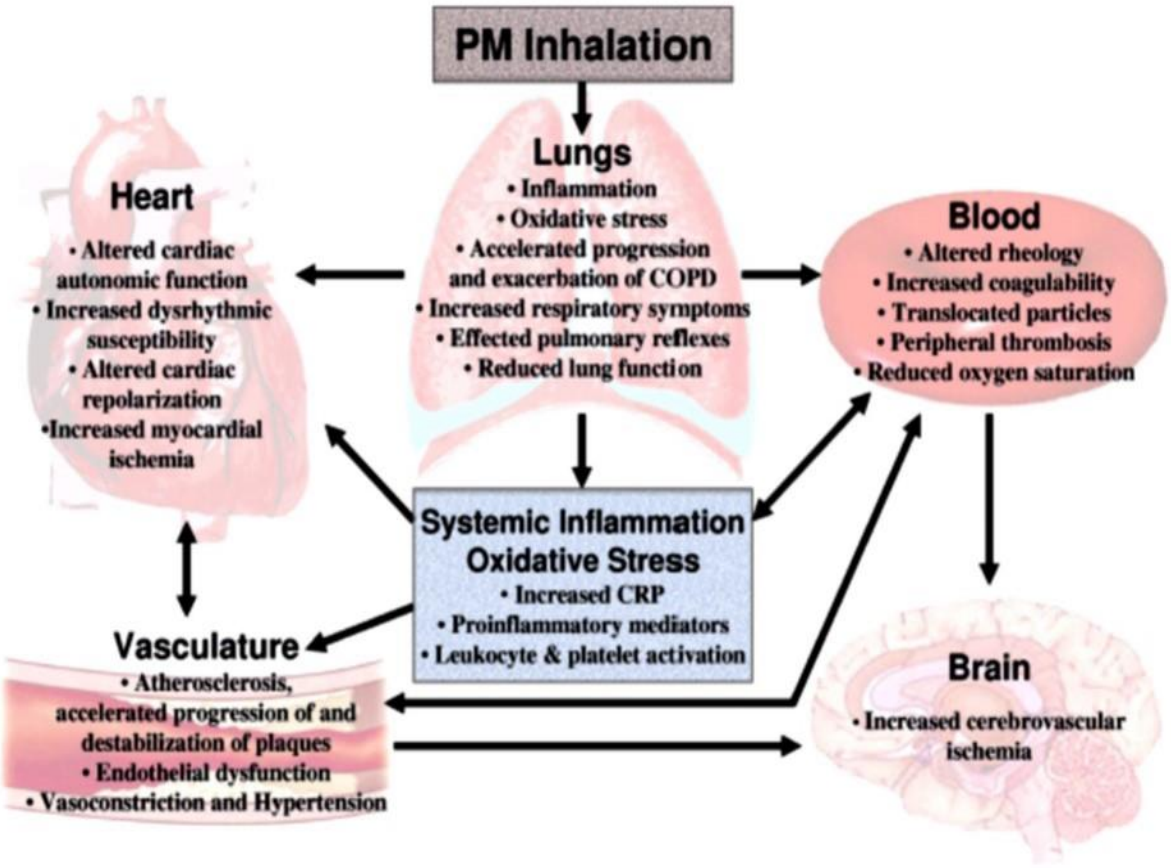

Figure 2-2: Health effects of PM inhalation on the human body (Source: Pope & Dockery, 2006).

2.5 Management of particulate matter

2.5.1 Emissions estimates

There is a major concern over the level of particulate emissions in South Africa as PM is regarded as a priority pollutant that can have significant negative impacts on air quality and human health. Obtaining accurate PM emission estimates is important for establishing air quality trends and identifying priority emission sources (Simon *et al.*, 2008). These estimates can be used for policy purposes to assess the effectiveness of mitigation strategies in attaining national emission targets (Jyethi, 2016). Emission estimates can also be used as input for modelling atmospheric dispersion (Simon *et al.*, 2008). Currently, a complete bottom-up emission inventory for South Africa's air pollutants is non-existent. This is due to the absence of up-to-date emissions data resulting from inconsistent reporting by source emitters. In an attempt to tackle this problem, the Department of Environmental Affairs for South Africa launched the internet-based National Atmospheric Emission Inventory System (NAEIS) in 2014. All major emitters with atmospheric emission licenses are now required to submit emission inventory reports annually (Mdluli, 2014).

Global emission sources are currently being used to assess trends in air quality for South Africa (Henneman *et al.*, 2016). These include the Emission Database for Global Atmospheric Research (EDGAR) version 4.3 and the Greenhouse Gas and Air Pollution Interactions and Synergies (GAINS) model. EDGAR is a bottom-up global emissions inventory developed by the Joint Research Centre of the European Commission in partnership with the Netherlands Environmental Assessment Agency (Janssens-Maenhout *et al.*, 2013). EDGAR contains historical emissions for air pollutants and greenhouse gases, from combustion and non-combustion sources, for all countries. The approach used to calculate these emissions is clear and consistent, allowing for comparisons between countries (Crippa *et al.*, 2018). The GAINS model is an integrated assessment framework that provides emission estimates and abatement options for 172 country regions (Amann *et al.*, 2011). Emission estimates are calculated using activity data for energy use, industrial combustion and processes, and agricultural activities obtained from international and national statistics (Amann, 2012). A comparison between EDGAR and GAINS is shown in Table 2-2:

Table 2-2 Description and comparisons between the GAINS model and EDGAR global emission inventory

Name	**Source sectors**	**Pollutant species**	**Spatial resolution**	**Temporal resolution**	**Reference**
EDGAR	energy, industrial processes, on-road and off-road transport sources, agriculture, large-scale biomass burning, and other anthropogenic sources	CO_2, SO_2, CO, PM_{10}, NO_x, NH_3, NMVOC, CH_4, N_2O, HFCs	0.1° x 0.1°	1970 – 2012	Janssens-Maenhout *et al.* (2013)
GAINS	energy, industrial combustion and processes, on-road and off-road transport sources, residential, and agriculture	CO_2, SO_2, CO, TSP, PM_{10}, $PM_{2.5}$, PM_1, BC, OC, NO_x, NH_3, VOC, CH_4, N_2O, F gases	0.5° x 0.5°	1990 – 2050 (5-year increment, projection starting in 2015)	Amann *et al.* (2011)

2.5.2 Satellite Retrievals

Routine PM concentration measurements are traditionally done at surface monitoring stations that can provide valuable information for air quality forecasting (Sinha *et al.*, 2015). Due to high capital and operational costs, ground-based monitoring stations are irregularly distributed and have limited spatial coverage (Munir *et al.*, 2016). Satellites can be used to supplement ground-based PM measurements through their ability to repetitively monitor geographic phenomena over large spaces, including remote areas (Engel-Cox *et al.*, 2004). Improvements in the interpretation of satellite aerosol retrievals have enabled for the estimation of surface PM at high spatial and

temporal resolutions. This has the potential for satellite retrievals to be used in monitoring long term trends of ambient PM (Duncan et al., 2014).

Aerosol optical depth (AOD) is a highly robust parameter that is widely used to estimate surface PM concentrations from satellites (Chu *et al.*, 2016). It is a dimensionless parameter that describes the degree of light extinction by aerosols along the integrated atmospheric vertical column from Earth's surface to the top of the atmosphere (Streets *et al.*, 2013). An AOD of 0.01 represents a very bright, clear blue sky. AOD values ranging between 2 and 3 are indicative of heavy aerosol loading from extreme events like dust storms or fires (National Oceanic and Atmospheric Administration, 2019). Moderate Resolution Imaging Spectroradiometer (MODIS) and Multi-angle Imaging Spectroradiometer (MISR) are the most widely used satellite instruments for converting AOD to ground-level PM mass concentration as they have global coverage with moderate spatial resolutions (van Donkelaar *et al.*, 2006; Liu *et al.*, 2007).

Research on applying satellite remote sensing for estimating and monitoring surface PM began in the mid-2000s. This involved investigating the relationship between total column AOD and surface PM concentrations. Early studies through linear regressions developed simple empirical relationships between these two variables (Liu *et al.*, 2005; Liu *et al.*, 2007). Other studies combined meteorological variables such as temperature, relative humidity, wind speed, wind direction and mixing layer height with satellite AOD retrievals in an attempt to improve the quality of PM estimations (Koelemeijer *et al.*, 2006). In recent works by van Donkelaar *et al.* (2015), AOD retrievals from MODIS, MISR and SeaWIFS (Sea-viewing Wide Field-of-view Sensor) instruments were merged with AOD simulated from the GEOS-Chem chemical transport model to generate surface PM mass concentration estimates. These estimates were then globally adjusted using a Geographically Weighted Regression (GWR) model. Although numerous studies (Chu *et al.*, 2016) have shown that satellite remote sensing can be used to produce PM estimations with a reasonable degree of agreement with ground measurements, converting satellite retrieved AOD to global PM concentrations is still a challenge as meteorological variables and aerosol composition varies from one region to another (Stirnberg *et al.*, 2018).

2.5.3 Source apportionment

Source apportionment is a process that involves identifying emission sources and determining the proportional contribution of each source to observed concentrations at selected receptor sites (Pant & Harrison, 2012). This is important in air quality planning, as priority can be given towards managing important sources through the identification of sources and the apportionment of their contributions (Thunis *et al.*, 2019). Source apportionment is typically conducted through receptor modelling to identify PM sources. Receptor models are mathematical techniques that use

multivariate statistics about physical and chemical characteristics of PM to infer contributions from different emission sources at specific receptor locations (Watson *et al.*, 2002). Receptor models can be classified into two groups: microscopic and chemical.

Microscopic methods focus on analysing features associated with individual particle formation in the ambient air. These techniques include scanning electron microscopy (SEM) and automated SEM (Pant & Harrison, 2012). Application of these techniques on a large-scale is difficult, as results from microscopic methods are not quantitative (Hopke, 2016). Chemical methods identify different source types based on the chemical composition of PM particles from ambient air. The compositional profiles of trace elements, ions (cations and anions) and elemental/organic carbon for specific sources are used as markers to distinguish and to ascertain the contributions of sources at receptor sites. Chemical methods have been widely used in PM source apportionment studies worldwide (Taiwo *et al.*, 2014). The most commonly used chemical methods are the Chemical Mass Balance (CMB) and Positive Matrix Factorization (PMF).

2.5.3.1 Chemical Mass Balance

The CMB receptor model is based on the principle of conservation in which the particulate mass recorded at the receptor site is equal to the summation of contributions from each of the sources (Hopke, 2016). Before applying this method, the chemical signatures of all relevant sources for the area under study must first be determined. The fundamental assumptions of the CMB model are:

- the source profiles are constant over the ambient and source sampling period
- chemical species do not interact with each other (i.e. they add linearly)
- the number of chemical species is greater than or equal to the number of sources
- source compositions are linearly independent of each other
- measurement errors are random, uncorrelated and normally distributed.

The model equation for CMB is given below:

$$\boldsymbol{C_I = \sum_{j=1}^{p} a_{ij} S_j, 1, n}$$

Equation 2-1: Chemical mass balance

where: C_i is the ambient concentration of the species I; a_{ij} is the fractional concentration of species i in the emissions from source j; S_j is the total mass concentration contributed by source j; p is the number of sources; n is the number of species, with $n \geq p$.

The CMB has been employed on several occasions to evaluate the contribution of different sources to PM pollution (Gupta *et al.*, 2007; Gummeneni *et al.*, 2011; Srimuruganandam & Shiva Nagendra, 2012; Huang & Wang, 2014; Martínez-Cinco *et al.*, 2016). The strengths of the CMB model are:

- key source contributions are quantified from elemental, ionic, and carbonic measurements
- source subtypes can be quantified using single-particle and organic compound measurements
- errors on source contribution estimates can be quantified from input data, uncertainties and collinearity of source profiles.

Despite being a commonly used model for source apportionment, the CMB has some limitations, which include:

- a generalisation of sources is difficult as source profiles are usually site-specific, i.e. the composition of soil dust can vary according to location
- source profiles are time-dependent so measurements over shorter periods may not fully capture data that is representative of particular sources, i.e. vehicle emissions can change in the long term due to changes in fuel composition, vehicle design and emission control technology
- knowledge on the emission profiles for every relevant source is required – this data is not commonly available
- analysis in the CMB can be impaired by collinearity among similar sources
- unknown or new sources are not directly identified; therefore, a substantial fraction of the measured pollutant load may not be resolved.

2.5.3.2 Positive Matrix Factorization

The PMF model is a multivariate factor analysis tool that deconstructs the matrix of speciated sample data into two matrices: factor contributions and factor profiles (Zou *et al.*, 2017). PM species observations are expressed as the summation of contributions from a number of non-time varying source profiles (Reff *et al.*, 2007). Unlike the CMB, the PMF does not require any a

priori knowledge on source composition. The selection of the number of factors in a PMF is subjective and will depend on the knowledge of the sources commonly found in the region of study (Gupta *et al.*, 2012). The model for PMF is expressed in Section 5.2.4.

The fundamental assumptions of the PMF model are:

- compositions of emission sources are constant throughout the ambient sampling periods at the receptors
- chemical species include in PMF do not react with each other and are linearly additive
- source compositions are linearly independent of each other
- the number of analysed chemical species is greater than or equal to the number of sources
- a large data set is normally required in which the number of samples exceeds significantly the number of species analysed

The PMF is a powerful and widely used multivariate receptor model with advantages that include:

- data on emission sources profiles are not required
- missing data or observations below detection limits are handled by assigning them low weights to reduce their influence during the analysis
- missing sources can be identified using this model.
- Data with a heavily positive skewed distribution can be included in the analysis by down weighting extreme points to reduce their influence

Like any other receptor model, the PMF has its limitations which are:

- a large dataset on measured concentrations is required
- there is a subjective determination on the number of factors to be retained
- inference of emission sources is made by interpreting these factors
- knowledge of source profiles is required to validate the representativeness of the computed source profiles and errors in the estimated source contributions
- an analysis is limited by the number of species measured at the receptor sites.

2.5.4 Dispersion modelling

Atmospheric dispersion models are mathematical simulations used to mimic the dispersion of air pollutants by tracing the emission pathways of a pollutant from its source to the receptor based on the physical and chemical processes taking place in the atmosphere (Bluett *et al.*, 2004). These models can be applied to investigate the impacts of emissions from specific sources on pollutant concentrations nearby (Awasthi *et al.*, 2006). Atmospheric dispersion models are also important regulatory tools in assessing compliance with air quality standards and regulations (South Africa, 2014). Unlike traditional ground-based monitoring which is limited to specific locations, these models are used to assess air quality in unmonitored sites.

There are different forms of models used to simulate the dispersion of pollutants which include Gaussian, Eulerian and Lagrangian models. Gaussian models are the simplest and most commonly used. Gaussian models are divided into plume or puff. Plume models describe dispersion under the assumption of steady-state conditions, whilst puff models simulate the evolution of puffs over time as a function of spatial and temporal changes in meteorological conditions (Forehead & Huynh, 2018). Examples of Gaussian based dispersion models include AERMOD and CALPUFF. A limitation of the Gaussian model is that it does not take into account low wind conditions and sites within proximity to the source i.e. distances less than 100 m (Holmes & Morawska, 2006). Eulerian and Lagrangian models have advanced features. Eulerian models calculate the dispersal of pollutants on a three-dimensional grid of cells fixed in space, whilst Lagrangian models perform similar simulations but on grid cells moving with the wind flow (Capelli *et al.*, 2013).

Most modern-day dispersion models operate on computer-based programs. As input, these models require data on the geometric configurations of emission sources and receptors, emission rates and meteorological parameters (wind speed and direction, ambient temperature, and stability conditions) of the study area. The step-by-step procedure for operating a dispersion model is shown in Figure 2-3:

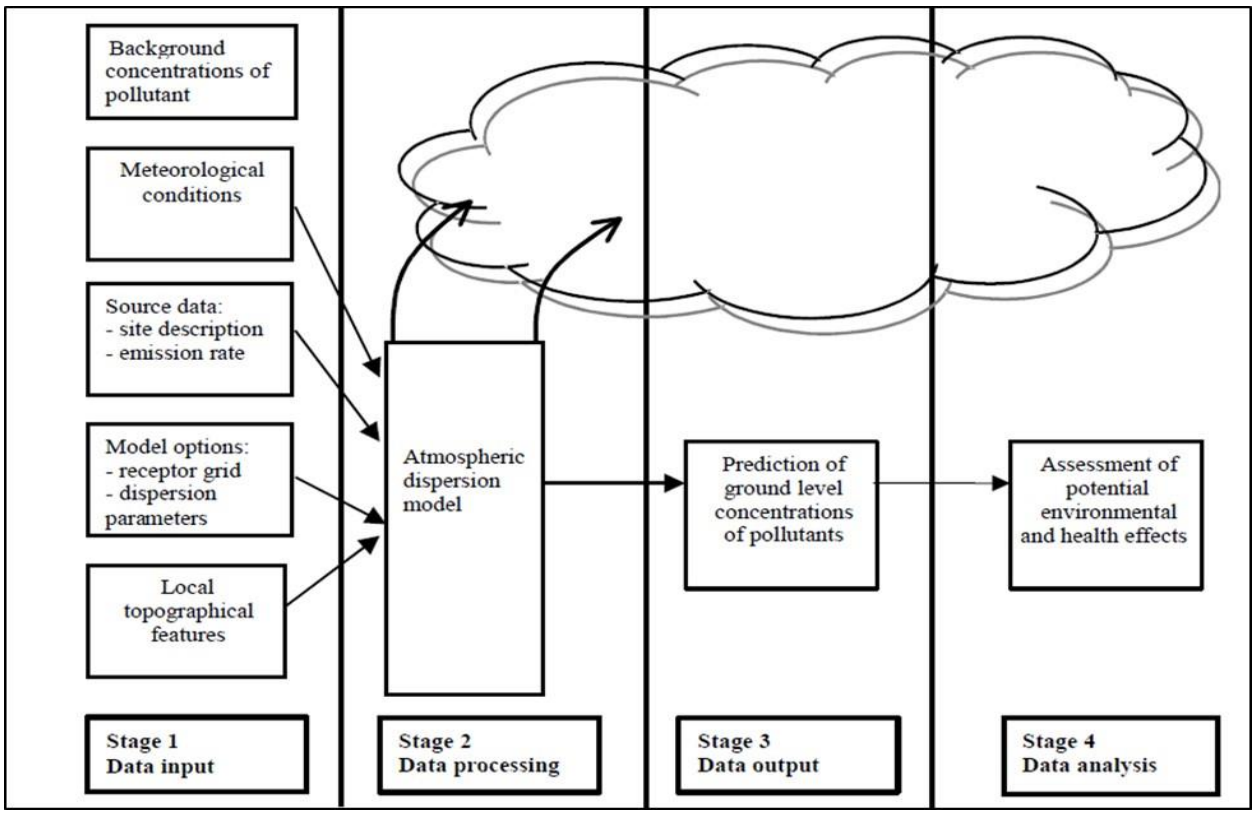

Figure 2-3: Overview of the procedure used when operating a dispersion model (Source: Bluett *et al.,* 2004).

For the purpose of this study, the CALPUFF dispersion model will be discussed in detail in Section 3.4.1.

2.5.4.1 Advantages of atmospheric dispersion modelling

Atmospheric dispersion models have been extensively applied to simulate spatial and temporal distributions of air pollutant concentrations at varying scales. There are a number of reasons that make these models suitable for air quality assessments:

- air quality information is generated at high spatial and user-specific temporal resolutions
- operating a network of ambient monitoring stations will cost more than estimating pollutant concentrations using atmospheric dispersion models
- the effectiveness of specific emission control strategies can be assessed through multiple scenario analyses
- can be used to evaluate source-receptor relationships.

2.5.4.2 Limitations of atmospheric dispersion modelling

Like any other mathematical model, atmospheric dispersion models have their limitations which impair on accuracy. These limitations include:

- Predictions are limited by the incorrectly calculated emission data, inaccurate meteorological parameters and inadequate knowledge on proper model configuration by the user
- Model performance is limited by the model's treatment of physical and chemical processes, i.e. Gaussian models do not take into consideration low wind conditions.

2.5.5 Integrated assessment modelling

Integrated Assessment Modelling (IAM) is the process of combining knowledge from multiple disciplines in order to elicit feasible solutions that can be used to inform policy (Peduzzi *et al.*, 2018). In the case of air quality planning, IAM models combine data on source emissions, atmospheric dispersion, abatement measures and their implementation costs (Carnevale *et al.*, 2012). IAM models like GAINS have been used in the EU for intergovernmental negotiations concerning long-range transboundary air pollution (Miranda *et al.*, 2016). Application of IAM models for air quality management in South Africa is still in its infancy. These tools have the potential to be used to validate compliance with air quality targets in South Africa. For this study, the GAINS model was used to justify the need for an integrated assessment for managing air quality in South Africa.

2.5.5.1 GAINS model

The GAINS model is an integrated assessment model developed by the International Institute for Applied Systems Analysis (IIASA). The purpose of this model is to translate scientific insights on air quality and climate change into applicable policy (Klaassen *et al.*, 2005). This is done by combining information on emission projections and control costs, atmospheric dispersion characteristics and environmental sensitivities to air pollution and climate change (Carnevale *et al.*, 2012). The model operates in two modes which are the scenario analysis and optimisation mode. The scenario analysis mode gives insights into the possible environmental outcomes based on an assumed pattern of emission controls and, economic and energy developments. In the optimisation mode, the minimal-cost allocation of emission controls to meet environmental targets are systematically identified (Kelly, 2006). The GAINS model has different components. The first component is centred on anthropogenic activities and emissions, the second on emission mitigation options and their costs, the third on atmospheric dispersion, and the fourth on the air quality impacts. These components are discussed in the following sections except for the air

quality impacts (on health), which are described in Section 6.2.8. Figure 2-4 shows the framework for the GAINS model:

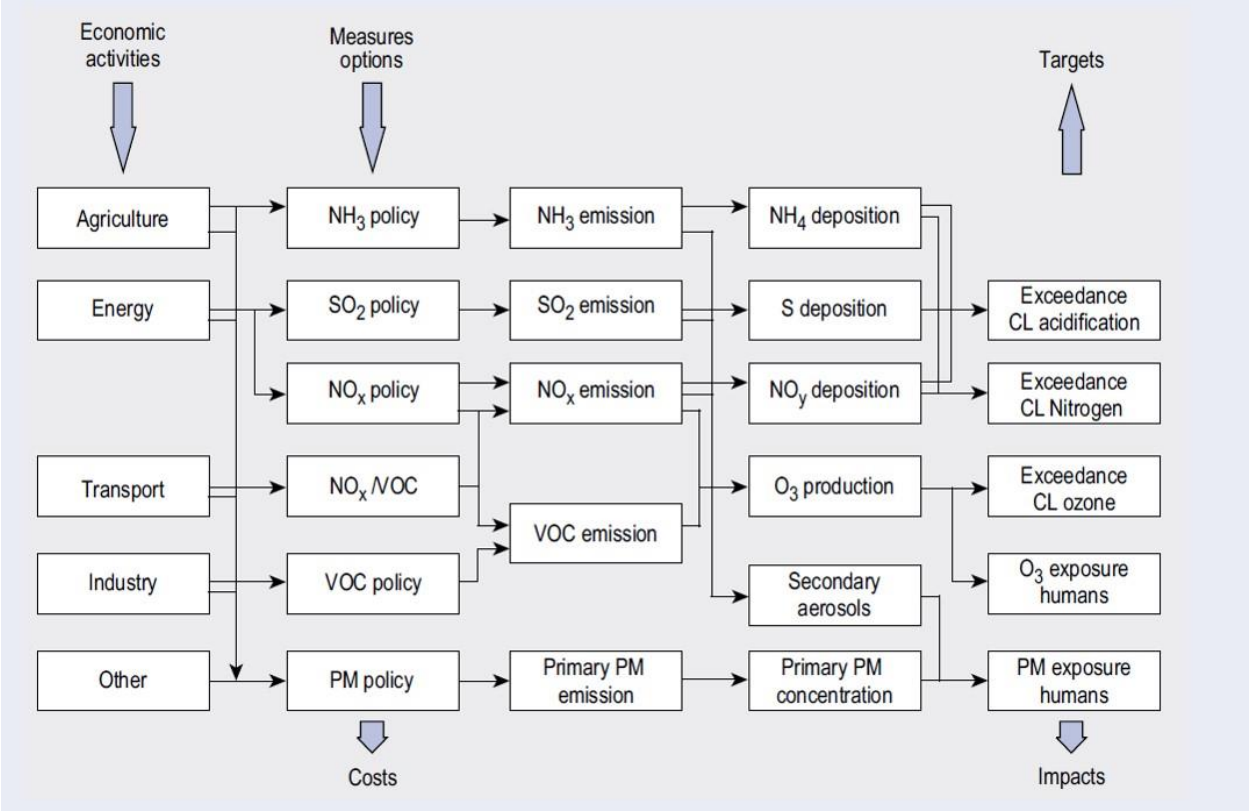

Figure 2-4: Overview of the GAINS modelling procedure (Source: Winiwarter, 2005).

2.5.5.2 Emissions projections in GAINS

Emission forecasts are based on socio-economic drivers and energy projections. Socio-economic drivers include population growth, economic growth (i.e. GDP) and expected growth of key sectors or activities (Kelly, 2006). These parameters are important in determining the likely pathways of polluting activities. Energy use plays an important role in the generation of air pollutants (International Energy Agency, 2017). Information on the type of fuel used and the degree of consumption for a particular sector/activity is critical for energy forecasting. Socio-economic parameters and energy forecasts are exogenous and have to be fed into the GAINS model. A detailed description of how emissions are calculated in the GAINS model is given in Section 6.2.2.

2.5.5.3 Emission control options and cost calculation in GAINS

The GAINS model uses a set of technical and structural measures to reduce emissions. Technical measures involve the use of end-of-pipe options that capture emissions at their sources before they are released into the atmosphere (Amann, 2012). This includes control strategies such as

flue gas desulphurization (FGD) and the application of high-efficiency dedusters. However, these measures can neither alter the anthropogenic driving forces of these emissions nor can they induce structural changes to the composition of energy systems and agricultural activities (Purohit *et al.*, 2013). For structural measures, the level at which energy services are supplied to consumers is similar but with lower resultant pollution. These measures alter polluting levels but do not have an impact on anthropogenic activities. Substituting coal for liquefied petroleum gas (LPG) in the domestic sector is a typical example of such measures (Amann *et al.*, 2011).

Associated costs for each emission control option are calculated under the notion of a free market in which abatement technologies are assumed to be available to all countries at the same costs (Amann, 2012). The parameters used in calculating the implementation costs of emission controls are abatement efficiencies, unit investments costs and, fixed operating and maintenance costs. Cost curves that relate the emission reduction levels with associated costs for a given pollutant are produced (Klaassen *et al.*, 2005).

2.5.5.4 Atmospheric dispersion in GAINS

In order to determine the fate and impacts of air pollutants in a particular region, it is necessary to trace the path of emissions to their end-points. The GAINS model relies on the Unified European Monitoring and Evaluation Programme (EMEP) Eulerian model for atmospheric dispersion and deposition simulations (Simpson *et al.*, 2012). National emissions data from GAINS is used for a 'country-to-grid' EMEP model analysis which illustrates the air quality impacts of changing emissions on individual grid cells (Kelly, 2006). GAINS does not assign individual emissions to each grid cell and then simulate pollutant dispersion grid by grid. Instead, a reduced form of the EMEP model comprising source-receptor relationships based on several hundred full EMEP runs from emissions by individual sources are used to model dispersion (Amann, 2012). Pollutant dispersion is modelled on a 50 km x 50 km grid resolution.

CHAPTER 3 DATA AND METHODS

3.1 Introduction

The data and methods employed are explained in detail within each article (chapter 4 - 6). This chapter intends to briefly provide a description of the methods used and, in some cases, to provide supplementary information not included in the articles themselves. For this study, the following conceptual framework (Figure 3-1) was followed, highlighting where each objective fits in:

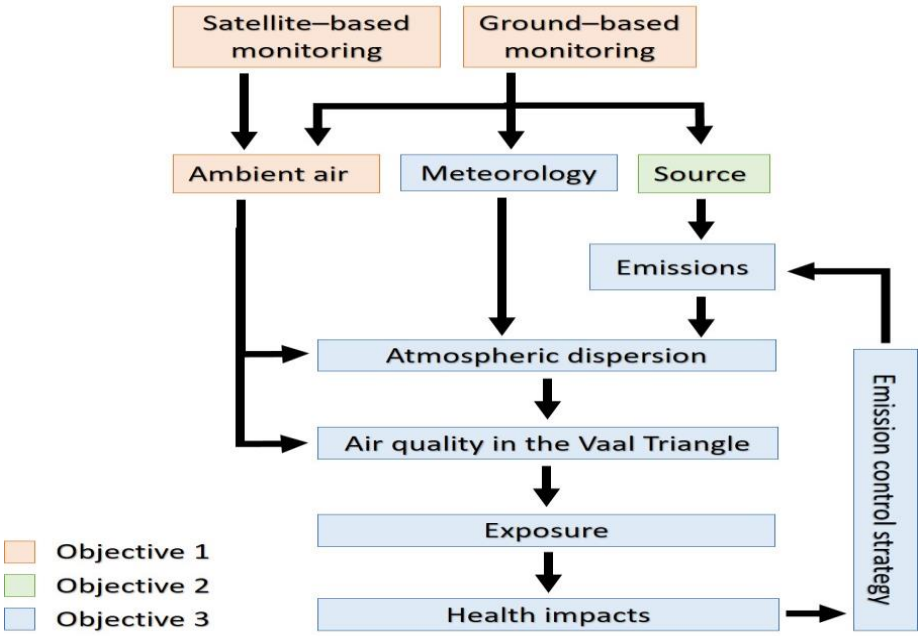

Figure 3-1: Conceptual framework outlining the process to be followed in each research objective (modified from the Department of Environmental Affairs and Tourism, 2007).

3.2 Objective 1: To explore the spatial variations and temporal trends of particulate matter concentrations in the Vaal Triangle Priority Area

The purpose of this article was to justify the need for the inclusion of satellite remote sensing in monitoring spatial and temporal changes in $PM_{2.5}$ concentrations in the VTAPA. High resolution (1 km x 1 km) satellite-derived $PM_{2.5}$ concentration data for the period 2007-2016 were acquired from the Atmospheric Composition Analysis Group of Dalhousie University, Canada (http://fizz.phys.dal.ca/~atmos/martin/) and ground-truthing was employed using local monitoring station data for the six sites within the VTAPA obtained from the South African Air Quality Information System (SAAQIS). The method used by Atmospheric Composition Analysis Group at Dalhousie University to estimate $PM_{2.5}$ concentrations is explained in the following sections and follows the methodology of van Donkelaar *et al.* (2015).

3.2.1 Calibration of retrieved and simulated Aerosol Optical Depth

Remotely sensed data from NASA's MODIS (MODerate resolution Imaging Spectroradiometer), MISR (Multi-angle Imaging SpectroRadiometer) and SeaWIFS (Sea-viewing Wide Field-of-view Sensor) instruments were used in the retrieval of the Aerosol Optical Depth (AOD). For the MODIS data, the Dark Target (DT) and Deep Blue (DB) retrieval algorithms were applied to retrieve AOD at 10 km resolutions. SeaWIFS AOD at 13.5 km resolutions was obtained using the DB algorithm. The MISR algorithm was applied to the MISR data to retrieve AOD at 17.6 km resolutions (van Donkelaar *et al.*, 2016). The AOD retrievals from the different satellite platforms were then calibrated using observational data from the AERONET (Aerosol Robotic Network) sun photometer global network so as to create a globally continuous AOD field on 10 km x 10 km grids.

Additional AOD was simulated using the GEOS-Chem chemical transport model. A vertical bias correction was applied to this simulated AOD using CALIOP aerosol extinction vertical profiles. Similar to the AOD retrievals, the simulated AOD was also calibrated using AERONET observations. The GEOS-Chem chemical transport model was then used to simulate the spatially varying geophysical relationship between AOD and $PM_{2.5}$ (van Donkelaar *et al.*, 2015).

3.2.2 Estimation of $PM_{2.5}$ using retrieved and simulated AOD

Based on the simulated AOD-$PM_{2.5}$ relationships, simulated AOD and AOD retrievals from the satellite instruments were used to estimate global $PM_{2.5}$ concentrations. These estimated $PM_{2.5}$ concentrations (from simulated and retrieved AOD) were then combined through the use of a weighted mean (van Donkelaar *et al.*, 2015).

3.2.3 Globally corrected $PM_{2.5}$ estimates

Global adjustments to these $PM_{2.5}$ estimations were made by applying a Geographically Weighted Regression (GWR) model based on $PM_{2.5}$ data from ground observations across the globe. This model takes into account spatial variations of predictor coefficients by assigning weights to estimate-observation pairs at various geographic locations based on their inverse-squared distance from individual points of interest (van Donkelaar *et al.*, 2016). The predictors used for the GWR include urban land cover, sub-grid elevation difference and aerosol composition. These adjusted $PM_{2.5}$ estimations are then were interpolated onto 1 km x 1 km grids.

3.3 Objective 2: To identify the sources contributing to particulate matter loading in low-income settlements of the Vaal Triangle Priority Area

The article sought to identify the sources that contribute to PM loading in the VTAPA through receptor modelling. The procedure used in this study is described in the sections to follow.

3.3.1 Instrumentation

The MCZ (MicroPNS Type Dichoto LVS16, Umwelttechnik MCZ GmbH, Bad Nauheim, Germany) dichotomous low volume sampler (Figure 3-2) is an automated dual-filter air sampler for the simultaneous and sequential collection of PM in the coarse and fine fraction. Found on the sampler are a PM_{10} sampling head, a virtual impactor and two separate mass flow controlled gas streams for coarse particles (15 L/min) and fine particles (1.7 L/min). This split flow structure of the virtual impactor allows a PM_{10} aerosol to be separated into the coarse and fine fraction for subsequent collection onto two separate membrane filters with a diameter of 47 mm. The sampler allows for the holding of 32 filter cartridges, 16 in the coarse fraction compartment and 16 in the fine fraction compartment.

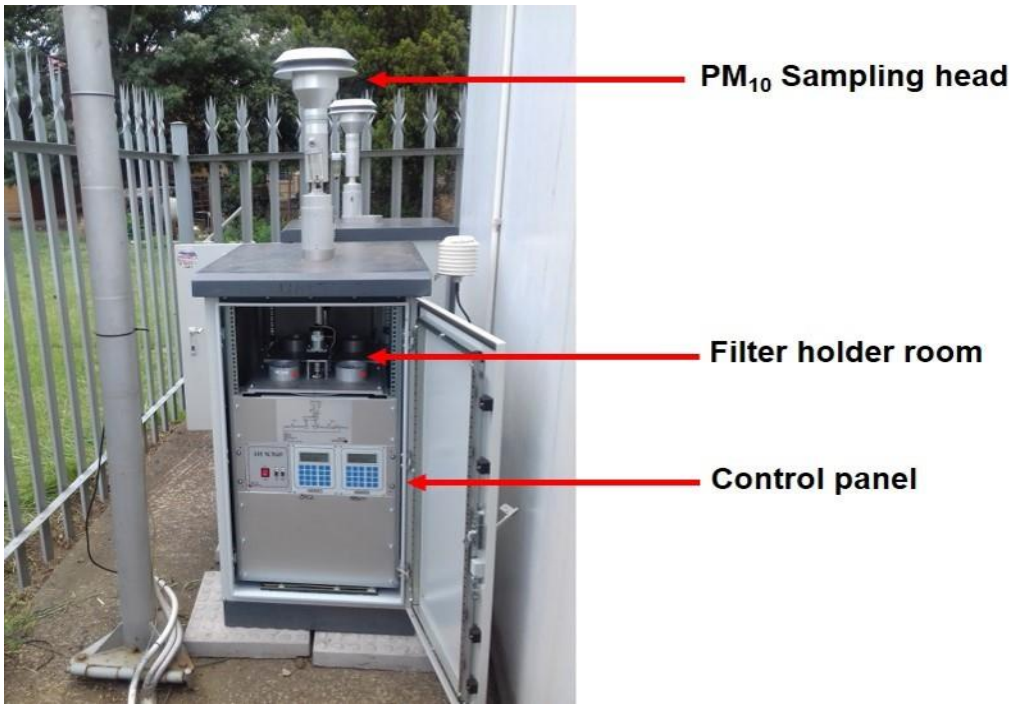

Figure 3-2 Internal and external structure of an MCZ dichotomous low volume sampler.

3.3.2 Sampling strategy

The MCZ dichotomous low volume sampler was used to sample ambient PM concentrations. Samples were collected daily at 12-hour intervals (between 10:00 AM and 10:00 PM and between 10:00 PM and 10:00 AM) using pre-weighed 47 mm Teflon filters with a pore size of 2 µm. A lag time of one minute between sampling periods was set to allow the sampler to sequentially change samples from the exposed filter to the next filter in line. Sampling was carried out for 16 days in summer, winter and spring 2018 (Table 3-1). Upon completion of sampling, exposed Teflon filters were reweighed and elemental, and ionic analyses were carried out on them. The results obtained

from these analyses were then fed into the PMF receptor model so as to determine the sources contributing to PM loading in the VTAPA.

Table 3-1 Dates on which campaigns were conducted at the sampling sites.

Season	Sampling dates
Summer	28 February 2018 to 21 March 2018
Winter	20 June 2018 to 6 July 2018
Spring	13 September 2018 to 3 October 2018

3.3.3 Local source profile development

Fugitive dust is a significant source of ambient PM whose composition can vary across space. As a result, it is necessary to characterise site-specific profiles. Soil samples were collected at 25 sites in the VTAPA (Figure 3-3), in triplicate, from 9 May 2017 to 25 May 2017. Table 3-2 gives a summary of the sampling locations and source categories.

Table 3-2 Location and description of the soil grab sampling sites.

Point	Sampling area	Description	Eastings	Southings
1	Nancefield train station, Klipspruit	Footpath (gravel road) close to driving school	27,9069	-26,2523
2	Between Orlando and Diepkloof East	Football pitch devoid of grass	27,9344	-26,2467
3	Diepkloof Zone Luvhuvhu Street	Open space	27,9399	-26,2457
4	Diepkloof Zone Point 4	Open space close to shops	27,9512	-26,2411
5	Kliprivier Business park	Truck stopping spot (parking lot)	28,0688	-26,4165
6	Kliprivier walking path	Footpath (gravel road) close to industries	28,0820	-26,4183
7	Meyerton Park	Open space close to shops	27,9955	-26,5584
8	Three Rivers East	Unpaved road shoulder	28,0092	-26,6567
9	Three Rivers, Almond Street	Open space	27,9956	-26,6558
10	Three Rivers, Poplar Street	Open space	27,9885	-26,6581
11	Three Rivers, Palm Street	Footpath (gravel road)	27,9816	-26,6511
12	Zamdela Phase 3, Kgotsong	Unpaved road	27,8569	-26,8672
13	Zamdela Phase 3, Iraq	Unpaved road	27,8591	-26,8696
14	Zamdela Phase4, Iraq	Unpaved road	27,8707	-26,8701
15	Zamdela Phase1, Iraq	Unpaved road	27,8745	-26,8622
16	Zamdela paved road	Paved road	27,8482	-26,8465
17	Somerspost Street, Zamdela, 1949	Paved road	27,8548	-26,8449
18	Sharpeville, Dubula Drive	Open space	27,8713	-26,6912
19	Sharpeville, Lebowa Primary School	Unpaved road	27,8798	-26,6886

Point	Sampling area	Description	Eastings	Southings
20	Sharpeville, Rooistein Street	Unpaved road	27,8763	-26,6851
21	Sharpeville, Seliba Primary School	Unpaved road shoulder close to a school entrance	27,8799	-26,6828
22	Evaton, Kanana informal settlement	Unpaved road	27,8334	-26,5514
23	Sebokeng, 111732 Nguna Street	Unpaved sideways along houses	27,8685	-26,5536
24	Sebokeng, Mantanzima Street	Unpaved road	27,8482	-26,5641
25	Sebokeng, Nwetla Street	Unpaved road	27,8440	-26,5775

Surface soils were collected at depths of 10–20 mm using a garden trowel. For paved and unpaved surfaces loose surface material was swept onto a dustpan using a clean hard stifled brush. Each time a sample was collected, the sampling equipment was cleaned so as to avoid sample contamination. Samples collected were put in labelled polyethylene zipper locking bags.

In the laboratory, samples were sieved using 707, 105, 74, and 44 µm mesh sieves in preparation to be sampled in a resuspension chamber. In the resuspension chamber, each sieved sample (100 mg) was scattered and suspended by blowing clean filtered air (at a flow rate of 33.33 L/min) into the chamber for 3 minutes. The suspended material was then sampled for 20 minutes through size-selective inlets ($PM_{10-2.5}$ and $PM_{2.5}$) and collected onto pre-weighed Teflon (pore size of 2 µm) membrane filters of 47 mm in diameter The operating flow rates for the sampling inlets were determined based on the cyclone specifications, with both $PM_{10-2.5}$ and $PM_{2.5}$ inlets having flow rates of 16.67 L/min.

Exposed filters were weighed again so as to determine their mass concentrations. These filters were then chemically analysed to determine their elemental and ionic compositions. The chemical profiles obtained from these filters were then used to assist in identifying PM sources based on output results from the PMF receptor model.

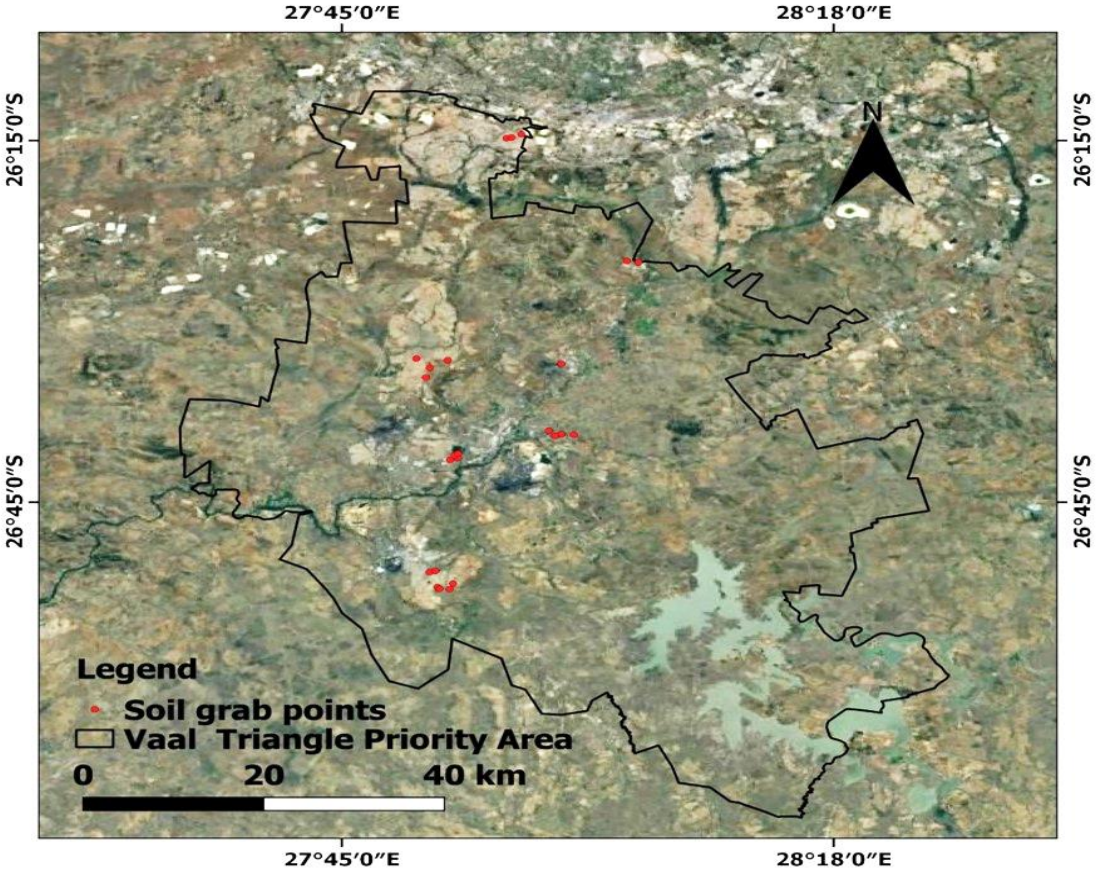

Figure 3-3 Map showing the location of the soil grab sampling sites in the VTAPA.

3.3.4 Chemical analyses

3.3.4.1 X-Ray Fluorescence

The X-Ray Fluorescence (XRF) analytical technique was utilized to determine the composition and concentration of trace elements in the exposed Teflon filter samples. A PANalytical AxiosmaX wavelength dispersive x-ray fluorescence spectrometer (WD-XRF) was used to carry out this analysis. This spectrometer consists of a rhodium x-ray tube with a 4 kW generator. Seven trays each with the capability to hold eight 37mm stainless steel sample cups are also attached to the instrument.

Aerosol samples were placed in filter holders and put into the 37mm stainless steel sample cups. These samples are then analysed by being exposed to an excitation condition in which x-rays produced from the spectrometer interact with atoms in the specimen leading to the ejection of electrons in the inner shells (Research Triangle Institute, 2009). Outer shell electrons will then move to occupy the vacant inner shells and in the process x-rays characteristic of the element found in the sample are emitted and detected by the spectrometer (Figure 3-4). The following elements were analysed using the spectrometer: Li, Na, Mg, Al, Si, P, S, Cl, K, Ca, Ti, V, Cr, Mn,

Fe, Co, Ni, Cu, Zn, Ga, Ge, As, Se, Br, Rb, Sr, Y, Zr, Nb, Mo, Pd, Ag, Cd, In, Sn, Sb, I, Cs, Ba, Ce, W, Pt, Au, Hg, Tl, Pb, and Bi.

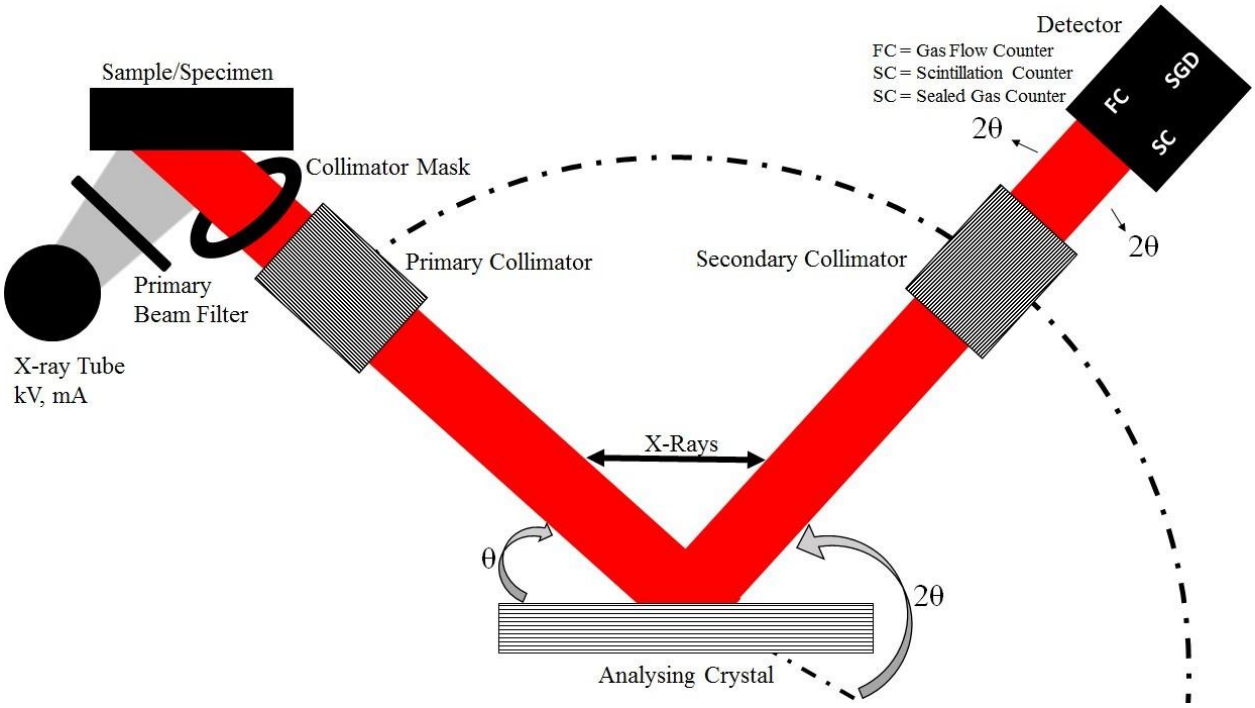

Figure 3-4 Analytical design of the wavelength dispersive x-ray fluorescence spectrometer (Adapted from Willis *et al.*, 2014)

3.3.4.2 Ion Chromatography

Ion Chromatography (IC) was used to determine the composition and concentration of ionic chemical species on the exposed Teflon filters. A Dionex ICS-3000 IC system consisting of two flow lines was used for IC analysis. One flow line was used for the detection of anion species and the other flow line to detect cation species. Before IC analysis commenced, the filter samples were leached in 10 ml de-ionised water in an ultrasonic bath for 30 minutes. Certified stock solutions obtained from Industrial Analytical was used in the preparation of five standards, ranging from 20 ppb – 500 ppb. Filter samples were then analysed for the following water-soluble ionic species: F^-, Cl^-, SO_4^{2-}, NO_3^-, CH_3COO^-, $HCOO^-$, $C_3H_5O_2^-$, $C_2O_4^{2-}$, Na^+, NH_4^+, K^+, Mg^{2+} and Ca^{2+}. Detection limits were calculated by using several laboratory blanks. In order to avoid contamination, this procedure was conducted in a stabilised room. The components for the Dionex ICS-3000 IC system and the concentration profiles for the anionic and cationic species are respectively illustrated in Table 3-3 and Table 3-4 below:

Table 3-3 A summary of the Dionex ICS-3000 IC system components utilised in the chemical analysis of the exposed filter samples

	Anions	**Cations**
Analytical Column	IonPak™ AS-15 4x250 mm	IonPak™ CS-15 3x250 mm
Guard Column	IonPak™ AG-15 4x50 mm	IonPak™ CG-15 3x50 mm
Loop Size (µL)	500	100
Suppressor	AERS 500 4 mm	CERS 500 2 mm
Carbonate Removal Device	CRD 200 4 mm	None
Detector	Conductivity	Conductivity
Eluent	KOH (Potassium Hydroxide)	MSA (Methane Sulphonic Acid)
Auto sampler	Dionex AS-DV	Dionex AS-DV

Table 3-4 The concentration profiles and suppressor currents used for the analysis of water-soluble ionic aerosol species

Anions (99mA) **Multistep concentration profile**		**Cations (30mA)** **Isocratic concentration profile**	
Time = 0.00 min	5 mm	Time = 0.00 min	22.5 mm
Time = 15.00 min	5 mm	Time = 50.00 min	22.5 mm
Time = 17.00 min	40 mm		
Time = 40.00 min	40 mm		
Time = 42.50 min	5 mm		
Time = 50.00 min	5 mm		

3.4 Objective 3: To assess the potential impacts of emission reduction strategies on air quality in the Vaal Triangle Priority Area

This article explored emission pathways under different management scenarios for the VTAPA. The GAINS model was used to identify a set of emission reduction strategies that when applied over time, could effectively reduce $PM_{2.5}$ concentrations to within national ambient air quality standards. The implementation costs for these mitigation strategies were calculated in GAINS. In order to determine the air quality and human health effects of reductions under different emission management scenarios, $PM_{2.5}$ concentration data was required. The GAINS emissions modelling procedure is briefly explained in this chapter. A more detailed description of the GAINS model methodology outlining the scenario design, implementation cost and health effects calculations is given in Section 6.2.2. Dispersion modelling for the GAINS model analysis was performed exogenously and the procedure used for the model simulations is outlined in detail in the subsequent sections.

3.4.1 Emissions modelling

The GAINS model was used to model emissions for the VTAPA for the period 2015 – 2035. This model requires energy and non-related energy activity data as input to generate emissions. However, the data available for the VTAPA is incomplete. Therefore, input data for the GAINS model was collected from three sources which include, (i) the Gauteng energy use data from the University of Stuttgart's Institute for Energy Economics and the Rational Use of Energy (Tomaschek *et al.*, 2012), (ii) South Africa provincial energy use data from the University of Cape Town's Energy Research Centre (Energy Research Centre, 2018) and, (iii) waste generation data from the Gauteng waste information system (Gauteng Department of Agriculture and Rural Development, 2018). Data from the University of Stuttgart's Institute for Energy Economics and the Rational Use of Energy was gathered from national statistics, e.g. fuel sales, electricity sales, economic output and employment figures. Data from the University of Cape Town's Energy Research Centre was compiled from various energy statistics. This energy data from the three sources were arranged into the GAINS model data template, uploaded to the GAINS model from which emissions of $PM_{2.5}$, SO_2 and NO_X for the VTAPA were generated.

3.4.2 Dispersion simulations

Regulations pertaining to dispersion modelling in South Africa recommend a number of models that can be used to monitor compliance (South Africa, 2014). Selection of the appropriate model to be used is determined by the scope, purpose and the level of assessment required by the study. In regards to the level of assessment, the tiered approach based on South African Draft Regulations Regarding Air Dispersion Modelling (Notice 1035 of 2012 in terms of NEMAQA (Act No. 39 of 2004) is used (South Africa, 2014). Level 1 assessments give estimations of air quality impacts based on a worst-case scenario. Level 2 assessments are required for situations where spatial and temporal distributions of pollutant concentrations and depositions need to be assessed. Level 3 assessments are more complex as they take into account irregular plume trajectories, variations in turbulent mixing, various source types and chemical transformation.

For this study, simulations were done using the Department of Environmental Affairs (DEA) recommended US-EPA approved CALMET/CALPUFF dispersion model as it supports Level 3 assessments. CALPUFF is a multi-layer, multi-species, non-steady-state Lagrangian Gaussian puff model that can simulate the spatial and temporal varying effects of meteorological conditions on pollutant transportation, transformation, and removal (Scire *et al.,* 2000). CALPUFF has the capability to simulate dispersal of pollutants over a large region taking into account chemical transformations that primary pollutants may undergo (Rood, 2014). The CALPUFF model consists of three main modules: CALMET, which is a meteorological pre-processing model, the CALPUFF dispersion model and CALPOST, which is a post-processing model.

CALMET is a diagnostic meteorological model that provides parameterised treatments of terrain effects (which include slope flows, terrain channelling and kinematic effects) that account for variations wind patterns (Whitford, 2009). Three-dimensional meteorological fields are generated in CALMET and are then used in CALPUFF for the advection of puffs containing material emitted from modelled sources (Rood, 2014). CALPOST is used to compute time-averaged concentrations and deposition fluxes based on CALPUFF outputs. Figure 3-5 gives an overview of the CALPUFF modelling procedure.

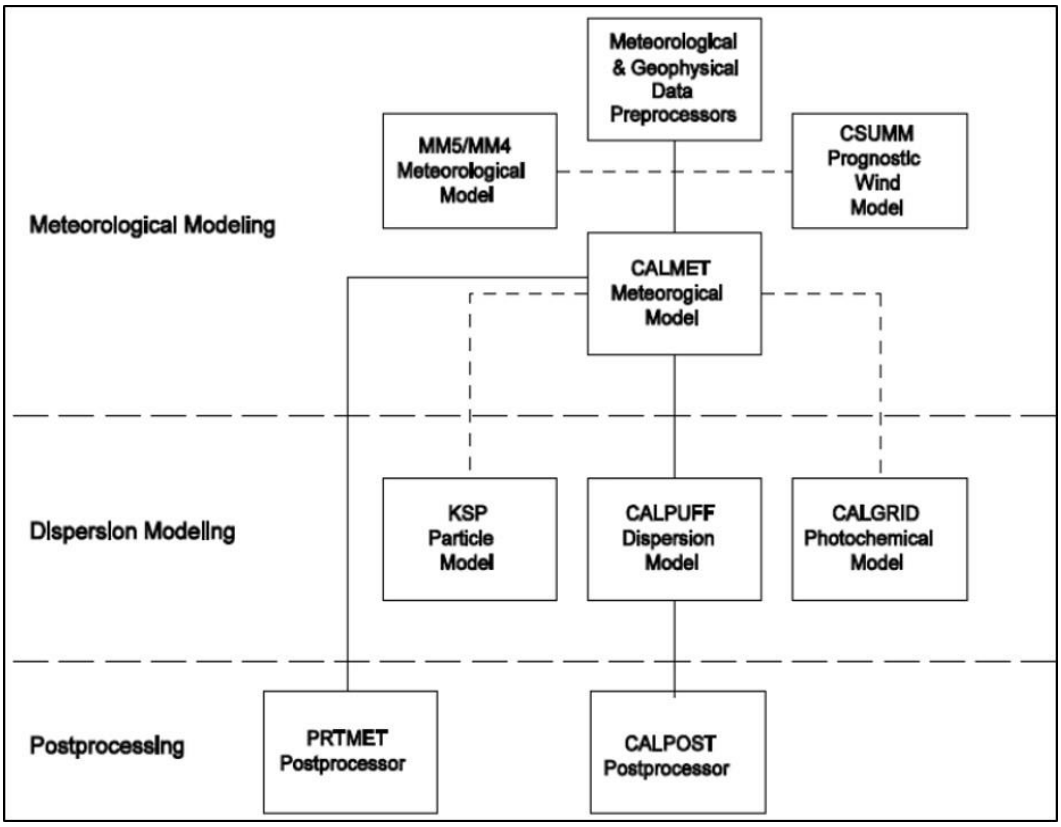

Figure 3-5: Overview of the CALPUFF modelling process (Source: Scire *et al.,* 2000).

3.4.3 CALMET modelling

The CALMET modelling domain for the VTAPA was 200 km by 200 km with a horizontal grid resolution of 1 km x 1 km (Figure 3-6). The modelling domain covered the entire VTAPA and most of the Gauteng region. The Universe Transverse Mercator (UTM) coordinate system was used for the VTAPA modelling domain. CALMET was run for three consecutive years in one model run (January 2014 to December 2016).

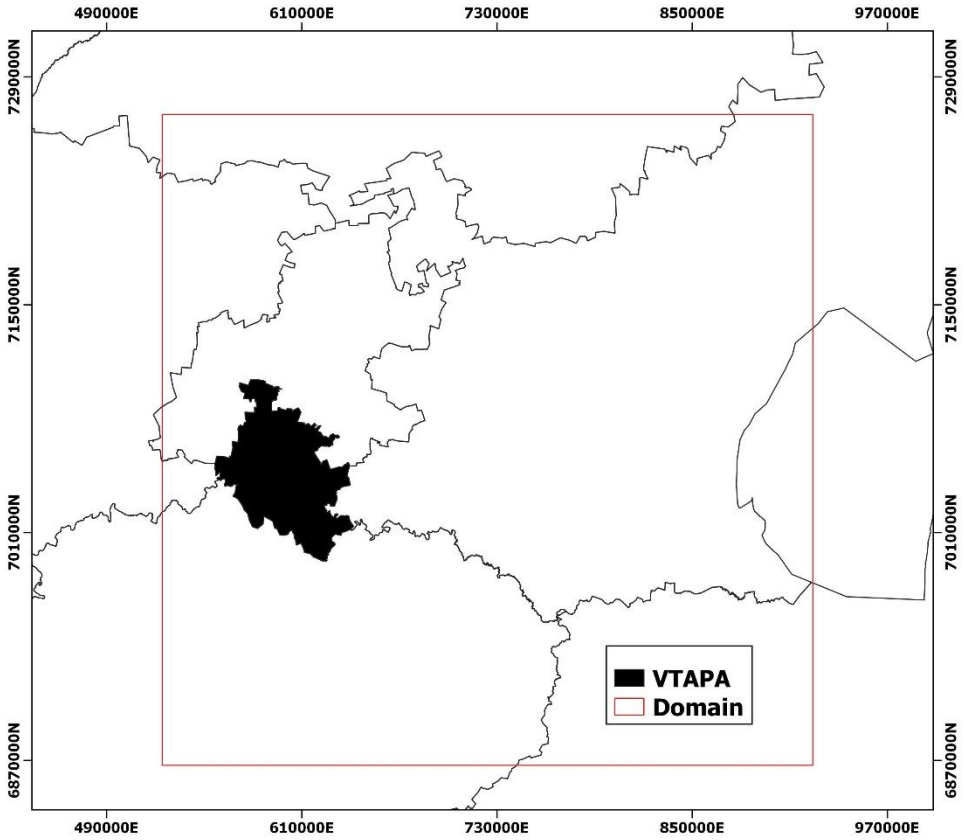

Figure 3-6 CALPUFF modelling grid domain for the VTAPA.

3.4.4 CALPUFF modelling

For this study, the CALPUFF version 7 was used to simulate the dispersion of the following pollutants: SO_2, SO_4, NO_x, ammonium nitrate (NH_4NO_3), NO_3 and $PM_{2.5}$. The choice of pollutants is based on the Mesopuff-II chemical transformation scheme. The model settings used in CALPUFF are detailed in Table 3-5. Central points of the 2011 South African census data small area level geographic units were used as discrete receptors in the model domain. Point locations for the monitoring stations found within the VTAPA were also used as receptors for ground-truthing purposes. In order to determine the individual contribution of each emission source type, sources were modelled separately in CALPUFF.

Table 3-5 Model settings used for running the CALPUFF dispersion model.

Variable	Model option utilised
Map projection	WGS-84 UTM Global coverage
Chemical transformation scheme	MESOPUFF 2 Scheme
Night-time conversion rates (percent/hour)	SO_2 = 0.2, NO_x loss rate = 2, HNO_3 formation rate = 2.0
Wind speed profile	ISC Rural

Variable	Model option utilised
Plume rise	Stack-tip downwash, Transitional plume rise, and partial plume penetration modelled
Plume element	Puff
Dispersion option	Dispersion coefficients use turbulence computed from micro-meteorology
Terrain effects	Terrain adjustment method applied to gridded and discrete receptors - partial plume path adjustment

3.4.5 Post-processing

CALPOST version 7.1.0 was used to estimate the time-averaged concentrations for the six pollutants modelled in CALPUFF. CALSUM version 7.0.0 was used to sum the individual contributions of each emission source type.

3.4.6 Model evaluation

CALPUFF simulations for the reference year (2015) were evaluated by comparing modelled values with annual averages from six national monitoring stations in the VTAPA. These stations are Diepkloof, Kliprivier, Sebokeng, Sharpeville, Three Rivers and Zamdela. According to U.S. EPA, CALPUFF predictions tend to have a better correlation with observations when modelled values are within a factor of 2 of the observations (Table 3-6). As shown in Table 3-6 modelled values of five stations have a reasonable agreement with actual monitoring values. The large deviation from the observed value exhibited at the Diepkloof site could be as a result of inherent uncertainties related to the random nature of the atmosphere and its representation by the WRF model (US EPA, 2005). These inherent uncertainties can lead to a ±50% deviation from measured concentrations.

Table 3-6 Comparison of simulated concentrations by CALPUFF and monitoring concentrations in the VTAPA

Station	Modelled $PM_{2.5}$ ($\mu g/m^3$)	Observed $PM_{2.5}$ ($\mu g/m^3$)	Ratio
Diepkloof	53	21	2.5
Kliprivier	32	35	0.9
Sebokeng	31	29	1.1
Sharpeville	31	36	0.9
Three Rivers	32	26	1.2
Zamdela	34	30	1.1

CHAPTER 4 EVALUATING THE POTENTIAL OF REMOTE SENSING IMAGERY IN MAPPING GROUND-LEVEL FINE PARTICULATE MATTER ($PM_{2.5}$) FOR THE VAAL TRIANGLE AIRSHED PRIORITY AREA

Journal article - Preface

Author list, contributions and consent

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The majority of the work was conducted by the first author, **L. Muyemeki,** who was responsible for data processing, analysis, interpretation, and writing the manuscript. Co-author contributions were as follows: S. J. Piketh and R. Burger supervised the study and assisted in interpretation of data and writing the manuscript.

The author has been given written permission to submit the manuscript for degree purposes (see Annexure II).

Formatting and status of the article

The article was configured according to the journal requirements of the *Clean Air Journal*; to which it was submitted. The article is presented in the style, format and length stipulated by the journal. The guide for authors used in the preparation of the article is available at https://journals.assaf.org.za/index.php/caj/about/submissions. The article is currently (March 2020) in the review process.

Thesis objective addressed: To explore the spatial variations and temporal trends of particulate matter concentrations in the Vaal Triangle Airshed Priority Area

Air quality decisions are made based on reliable monitoring data. These data are used in assessing compliance with regulatory ambient air quality standards and to evaluate the effectiveness of emissions mitigation strategies. In South Africa, air quality monitoring is conducted using ground-based instrumentation which due to limited financial resources are placed in areas of poor air quality. Although ground-based measurements have high accuracy, air quality monitoring stations are sparsely distributed within South Africa. This results in monitoring data being spatially under-represented. Satellite remote sensing offers the opportunity to monitor air quality over large scales. Due to their broad coverage and fine spatial resolutions, satellite observations can be used to provide a better understanding of the regional distribution of air pollutants. This technology can be used to complement ground-based measurements. In this

regard, this article seeks to evaluate the potential of using satellite remote sensing in monitoring the spatial and temporal variations of $PM_{2.5}$ concentrations in the VTAPA.

Abstract

The quality of air breathed in South Africa is of great concern, especially in industrialised regions where $PM_{2.5}$ concentrations are high. Long term exposure to $PM_{2.5}$ is associated with serious adverse health impacts. Traditionally, $PM_{2.5}$ is monitored by a network of ground-based instruments. However, the coverage of monitoring networks in South Africa is not dense enough to fully capture the spatial variability of $PM_{2.5}$ concentrations. This study explored whether satellite remote sensing could offer a viable alternative to ground-based monitoring. Using an eight-year record (2009 to 2016) of satellite retrievals (MODIS, MISR and SeaWIFS) for $PM_{2.5}$ concentrations, spatial variations and temporal trends for $PM_{2.5}$ are evaluated for the Vaal Triangle Airshed Priority Area (VTAPA). Results are compared to corresponding measurements from the VTAPA surface monitoring stations. High $PM_{2.5}$ concentrations were clustered around the centre and towards the south-west of the VTAPA over the highly industrialised cities of Vanderbijlpark and Sasolburg. Satellite retrievals tended to overestimate $PM_{2.5}$ concentrations. Overall, there was a poor spatial agreement between satellite-retrieved $PM_{2.5}$ estimates and ground-level $PM_{2.5}$ measurements. Root mean square error values ranged from 6 to 11 $\mu g/m^3$ and from -0.89 to 0.32 for the correlation coefficient. For satellite remote sensing to be effectively exploited for air quality assessments in the VTAPA and elsewhere, further research to improve the precision and accuracy of satellite-retrieved $PM_{2.5}$ is required.

Evaluating the potential of remote sensing imagery in mapping ground-level fine particulate matter ($PM_{2.5}$) for the Vaal Triangle Airshed Priority Area

4.1 Introduction

At a global scale, air pollution is ranked fourth amongst the leading risk factors to human health, with recent estimates linking it to nearly 5 million premature deaths (Stanaway *et al.*, 2018; Health Effects Institute, 2019). In sub-Saharan Africa, particularly in the urban areas, the deterioration in air quality as a result of rapid urbanisation, population growth and industrial expansion is evident (Amegah & Agyei-Mensah, 2017; Fayiga, Ipinmoroti, & Chirenje, 2018). Of great concern to public health, are the levels of fine particulate matter ($PM_{2.5}$) in the cities of sub-Saharan Africa, which are amongst the highest in the world (Fayiga et al., 2018; Katoto et al., 2019). Long-term exposure to high levels of $PM_{2.5}$ is harmful to humans as it can lead to increased severity in the symptoms of asthma and chronic obstructive pulmonary disease (Guaita *et al.*, 2011; Feng *et al.*, 2016; Hamanaka & Mutlu, 2018; Loxham & Nieuwenhuijsen, 2019).

In South Africa, air pollution is an important issue, especially in industrialised regions like the Vaal Triangle Airshed Priority Area (VTAPA) where strong economic growth has taken place (Naiker *et al.*, 2012; Zhu et al., 2012). The VTAPA routinely experiences poor air quality as a result of strong emissions from industries, residential burning, vehicles and fugitive dust sources coupled with unfavourable meteorological conditions that have led to the accumulation of $PM_{2.5}$ in high concentrations (Annegarn & Scorgie, 1997; Scorgie *et al.*, 2003). To improve air quality and public health in the VTAPA, the Department of Environmental Affairs (DEA) developed an air quality management plan that outlined strategies to reduce emissions from key sources (South Africa, 2009; Tshehla & Wright, 2019). Furthermore, air quality monitoring stations were placed in areas in the VTAPA where National Ambient Air Quality Standards (NAAQS) are being exceeded. This is so as to assess pollution trends and to ascertain whether concentrations of $PM_{2.5}$ and other pollutants are being kept within the regulatory limits (Ngcukana, 2016; Altieri & Keen, 2019). However, the main drawback with these monitoring stations is that they only provide concentration data at specific points, leaving large areas uncovered (Engel-Cox *et al.*, 2004; Tian & Chen, 2010; Hu *et al.*, 2014). Intra-urban variability of $PM_{2.5}$ concentrations is therefore not accounted for. In order to capture the full-scale variability of $PM_{2.5}$ concentrations in the VTAPA, there is need for a vast network of monitoring stations. However, this requires large financial resources (Munir *et al.*, 2016).

Satellite remote sensing can provide repeated observations of atmospheric pollution at large spatial scales. The monitoring of air pollutants using satellite observations is gradually gaining more attention in atmospheric pollution studies (Duncan et al., 2014; Hersey *et al.*, 2015; Di *et al.*,

2019; Yi *et al.*, 2019; She *et al.*, 2020). Advancements in regional algorithms have allowed for the large scale retrieval of $PM_{2.5}$ concentrations at fine spatial resolutions that have a reasonable agreement with ground measurements (van Donkelaar *et al.*, 2015; van Donkelaar *et al.*, 2016). These retrievals have been successfully used to assess long term spatial-temporal patterns of $PM_{2.5}$ in regions experiencing poor air quality such as China and Saudi Arabia (Lu *et al.*, 2017; Munir *et al.*, 2016; Shi *et al.*, 2012). In the case of South Africa, knowledge on the applicability of remote sensing to monitor air pollution levels is insufficient. A regional case study by Kneen *et al.* (2016) revealed that satellite technology has the potential to offer a practical and credible option to ground-based monitoring in South Africa. However, further investigation is still required in order to have more concrete evidence to advocate the use of remote sensing for air quality monitoring in South Africa. This study evaluated the potential use of remote sensing imagery for air quality assessment in the VTAPA using a publicly available high resolution remotely sensed $PM_{2.5}$ concentration global dataset developed by van Donkelaar *et al.* (2015). This dataset begins in 1998 and ends in 2016. Spatial variations in $PM_{2.5}$ concentrations were examined, and temporal trends were explored.

4.2 Methods

4.2.1 Study region

The VTAPA is an industrialised region lying on the South African Highveld, a high central plateau of South Africa that forms part of the grassland biome. It stretches from southern Gauteng down to the northern section of the Free State province with an area of over 4 900 km^2. The region is host to industries including iron and steel manufacturers (ArcelorMittal steel and Davesteel), FerroAlloy (Samancor Metalloys) and petrochemical companies (Sasol Chemical Industries, Natref and Omnia Fertilisers). Eskom's Lethabo coal-fired power station is located in this region.

The VTAPA has a population of 3.1 million people with most of its inhabitants residing in south-western Johannesburg, Soweto, Sebokeng, Sharpeville, Vereeniging, Vanderbijlpark, Sasolburg and Zamdela.

4.2.2 Satellite-derived data

Global satellite-derived $PM_{2.5}$ concentration data with a high spatial resolution (1 km x 1 km) covering a 10-year period from 2007 to 2016 was obtained from the Atmospheric Composition Analysis Group of Dalhousie University (http://fizz.phys.dal.ca/~atmos/martin/). This global dataset was generated by merging satellite retrievals of AOD (Aerosol Optical Depth) from the NASA MODIS (MODerate resolution Imaging Spectroradiometer), MISR (Multi-angle Imaging SpectroRadiometer) and SeaWIFS (Sea-viewing Wide Field-of-view Sensor) instruments with

AOD simulated using the GEOS-Chem chemical transport model in order to produce $PM_{2.5}$ estimates (van Donkelaar *et al.*, 2016). The $PM_{2.5}$ estimates were then calibrated by means of a Geographically Weighted Regression (GWR) based on ground observations (van Donkelaar *et al.*, 2015). These estimations have a good agreement with 210 ground-based $PM_{2.5}$ observations (R^2 = 0.81, slope of 0.82) from the United States of America, Canada and Europe.

Based on previous research, uncertainty may exist in the global $PM_{2.5}$ data as a result of the satellite retrieval method (van Donkelaar et al., 2015). Existing studies have resolved this by applying a three-year average as an annual average (Han *et al.*, 2015; Peng *et al.*, 2016; Shisong *et al.*, 2018). For this research, three-year moving averages were applied to the satellite retrievals from the period 2009 to 2016. A subset of the global $PM_{2.5}$ dataset for each year was extracted to cover the VTAPA study area using the Integrated Land and Water Information System (ILWIS) program (ITC, 2011). At each point location of the ground monitoring sites, the mean values of a square of 3 × 3 pixels (3 km × 3 km area) were extracted from the satellite retrievals.

4.2.3 Ground measured data

The VTAPA air quality monitoring network consists of six stations (Figure 4-1), which are located in Diepkloof (26.2507S, 27.9564E), Kliprivier (26.4203S, 28.0849E), Sebokeng (26.5878S, 27.8402E), Sharpeville (26.6898S, 27.8678E), Three Rivers (26.6583S, 27.9982E) and Zamdela (26.8449S, 27.8551E) and have been operational since 2007. Based on the simulated spatial distribution of air pollutants from dispersion modelling these monitoring stations, except for Kliprivier, are considered to be located in high PM_{10} concentration zones (Thomas, 2008; Department of Environmental Affairs and Tourism, 2009; Ngcukana, 2016). Hourly $PM_{2.5}$ concentration data (2007–2016) for all the VTAPA monitoring sites except for Diepkloof (data is incomplete) was acquired from the South African Air Quality Information System (SAAQIS). Although the data were provided in a quality-controlled form, further quality checks were conducted to remove negative values. For consistency with the satellite retrievals, three-year moving averages were also applied to the ground $PM_{2.5}$ concentration data from the period 2009 to 2016.

4.2.4 Satellite retrieval accuracy assessment

Satellite retrievals for $PM_{2.5}$ were compared with the ground measurements from all monitoring stations for the period 2009 to 2016 using the following performance evaluation metrics: root mean square error (RMSE) and correlation coefficient (R). These statistics were computed in R statistical software using the modstat function in the Open Air package (Carslaw & Ropkins, 2012).

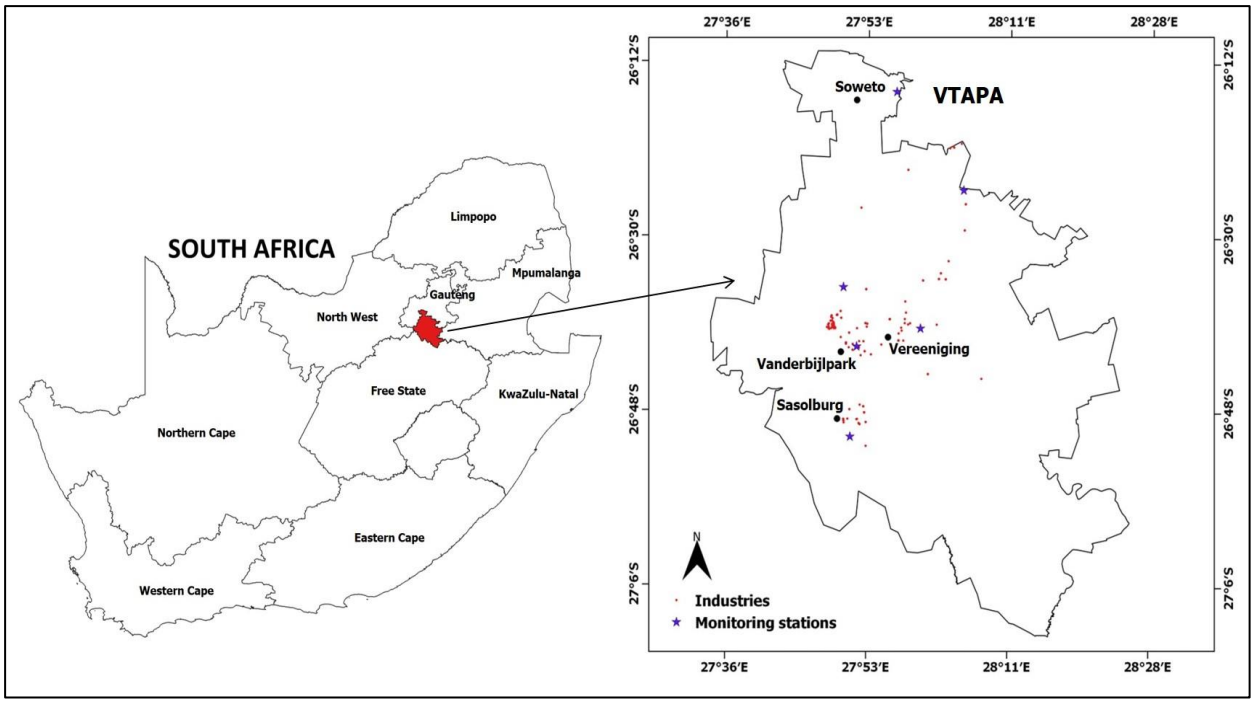

Figure 4-1: Study area map showing the location of the VTAPA in South Africa.

4.3 Results and discussion

4.3.1 Comparison of temporal variations between satellite-retrieved and ground measured $PM_{2.5}$ concentrations

Comparisons between satellite-retrieved and ground measured $PM_{2.5}$ concentrations (three-year moving averages) for the six monitoring stations in the VTAPA for the period 2009–2016 are shown in Figure 4-2. It can be observed that in three cases, there was an overestimation of observed $PM_{2.5}$ concentrations by the satellite retrievals. The exceptions were for the Kliprivier and Sebokeng stations, where ground measurements were much higher than the satellite-retrieved estimates. Both satellite retrievals and ground measurements showed that $PM_{2.5}$ concentrations for all sites were above the annual National Ambient Air Quality Standards (NAAQS).

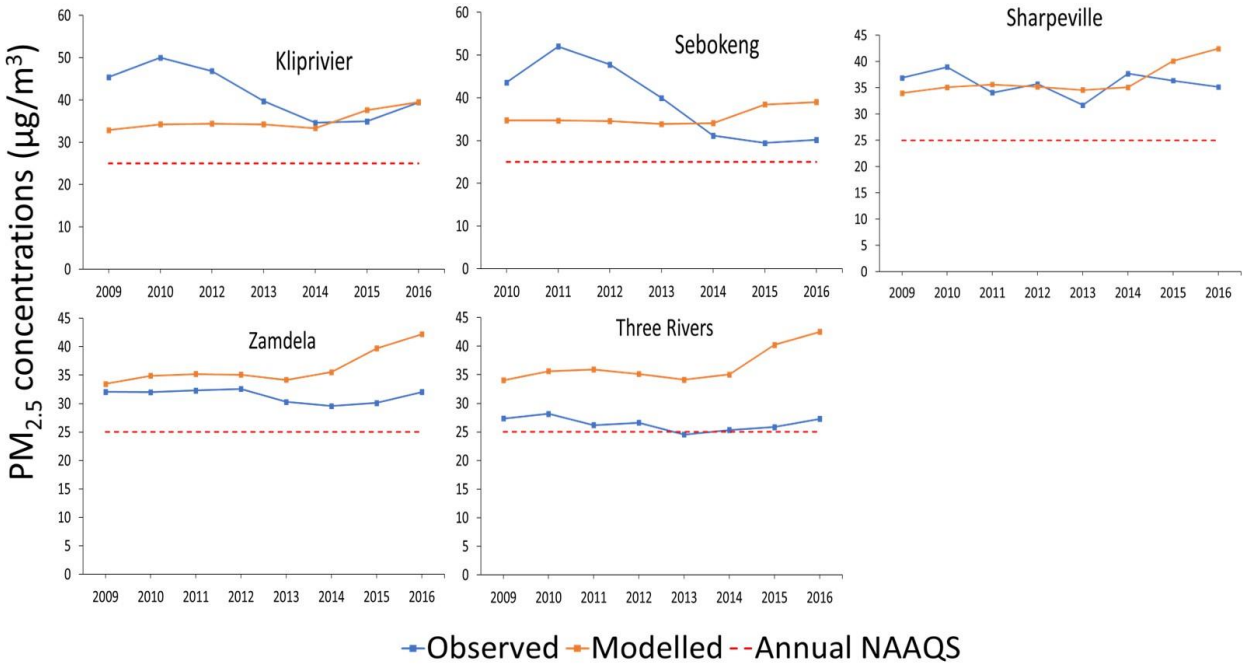

Figure 4-2: Comparisons between satellite-retrieved and ground measured $PM_{2.5}$ concentrations at Kliprivier, Sebokeng, Sharpeville, Zamdela and Three Rivers stations from 2009 to 2016.

Figure 4-3 shows the eight-year averages for satellite-retrieved and ground observed $PM_{2.5}$ concentrations at all stations. The Sharpeville station had the smallest offsets with a difference of 0.7 µg/m^3 between observed measurements and estimates retrieved from satellite imagery. Fairly high offsets are observed for the Sebokeng (-4 µg/m^3), Zamdela (5 µg/m^3) and Klipriver (-6 µg/m^3) stations. The performance of the satellite retrieved model is less encouraging for the Three Rivers station that has a considerably large offset of 10 µg/m^3.

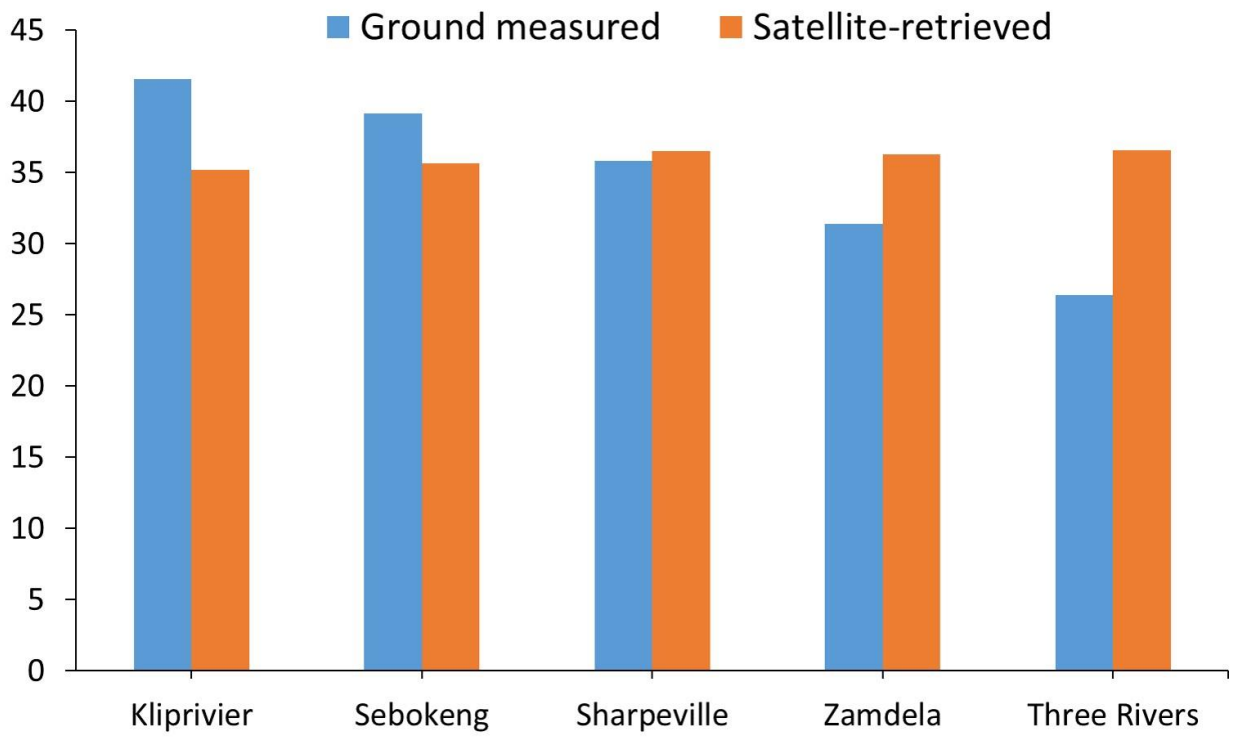

Figure 4-3: Comparison of the ground measured and satellite-retrieved 8-year averaged $PM_{2.5}$ concentrations at low-income settlements (Kliprivier, Sebokeng, Sharpeville, and Zamdela) and suburban area (Three Rivers).

The clustering of air quality monitoring stations in areas deemed to be experiencing pollution concentrations above national air quality guidelines could have contributed to the constant overestimation of $PM_{2.5}$ concentrations throughout the VTAPA. The location of monitoring stations in the VTAPA includes domestic coal-burning suburbs and a busy highway intersection. This lack of spatial representation will lead to mainly high values being used in the calibration of $PM_{2.5}$ estimates in the GWR model. Therefore, in order to improve the accuracy of the GWR model for satellite retrievals, the positioning of ground-based stations in South Africa needs to be optimised so as to have monitoring data that is more spatially representative.

The relationship between AOD and $PM_{2.5}$ is an important source of uncertainty in satellite retrieval accuracy as the AOD-$PM_{2.5}$ relationship can vary across space and countries. This spatial and temporal variation in the relation between AOD and $PM_{2.5}$ is mainly as a result of changes in the vertical distribution of aerosols in the atmosphere (Stirnberg *et al.*, 2018). This could have contributed to some of the inconsistencies between ground measured and satellite-derived $PM_{2.5}$ concentrations for the VTAPA. There is a need for an independent assessment of the AOD-$PM_{2.5}$ relationship through an integrated monitoring strategy like SPARTAN (Surface PARTiculate mAtter Network) in which $PM_{2.5}$ monitoring instruments are collocated with ground-based sun

photometers for AOD measurements (Snider *et al.*, 2015). This will help in evaluating AOD-$PM_{2.5}$ model accuracy and enhance $PM_{2.5}$ estimates from satellite AOD retrievals.

4.3.2 Satellite retrieval performance evaluation

Statistical comparisons between satellite-retrievals and ground measurements show that RMSE values ranged from 5 to 10 µg/m^3, with an average of 8 µg/m^3, indicating a significant difference between ground measured and satellite-retrieved $PM_{2.5}$ values (Table 4-1). R values for the years 2009–2012 ranged from -0.76 to -0.90, demonstrating strong negative correlations between satellite retrievals and ground-based measurements. These inverse relationships, however, do not imply a good agreement between satellite-retrieved $PM_{2.5}$ estimates and ground-level $PM_{2.5}$ measurements. R values from the period 2013 to 2016, displayed weak correlations between ground measured and satellite-retrieved $PM_{2.5}$ concentrations.

Table 4-1: $PM_{2.5}$ satellite retrieval performance evaluation for the period 2009 ─ 2016.

Year	RMSE (µg/m^3)	R
2009	7	-0.76
2010	9	-0.89
2011	10	-0.90
2012	9	-0.89
2013	6	-0.23
2014	5	-0.31
2015	9	-0.31
2016	10	-0.32

4.3.3 Spatial variations of satellite-derived $PM_{2.5}$ concentrations

Variations in $PM_{2.5}$ concentrations (three-year moving averages) over the VTAPA from 2009 to 2016 are presented in Figure 4-4. The average $PM_{2.5}$ concentrations in this region increased significantly by 25% from 33 µg/m^3 in 2009 to 41 µg/m^3 in 2016. This large increase took place mainly from 2015 to 2016, during which the highest concentrations (41 µg/m^3–44 µg/m^3) were observed in the VTAPA. The high $PM_{2.5}$ concentrations from 2015 to 2016 could possibly be due to increased AOD resulting from changes in aerosol mass transport during the *El Niño* episodes experienced in South Africa. *El Niño* events can increase regional aerosol concentrations by altering atmospheric circulation systems which leads to changes in the transport and removal of aerosols (Yu *et al.*, 2019). Wang *et al.*, (2019) found a positive link between the *El Niño* Southern Oscillation (ENSO) index and $PM_{2.5}$ concentrations in North China during the 2015/2016 *El Niño*

event. Mean $PM_{2.5}$ concentrations in North China were significantly higher in 2015 (51 µg/m^3–95 µg/m^3) as compared to 2017 (41 µg/m^3–74 µg/m^3).

Similar spatial patterns for $PM_{2.5}$ concentrations in the VTAPA are observed throughout the period (2009–2016) in which the area of highest PM pollution is concentrated around the centre and slightly down towards the south-western region of the VTAPA. These spatial patterns are comparable to those observed by Thomas (2008), who modelled PM_{10} concentrations in the VTAPA using a dispersion model. Due to the low dispersion potential of pollutants in the VTAPA, high $PM_{2.5}$ concentrations are clustered in the central to south-west region over the cities of Vanderbijlpark and Sasolburg, where heavy industrial (iron and steel, ferroalloy and petrochemical) activities, domestic burning and mining operations are taking place. Spatial distributions for the VTAPA show that high $PM_{2.5}$ concentrations are observed in the northern part of the area. The main sources for $PM_{2.5}$ in this northern area are residential combustion from the Soweto township, vehicle emissions and windblown dust from gold mine dumps.

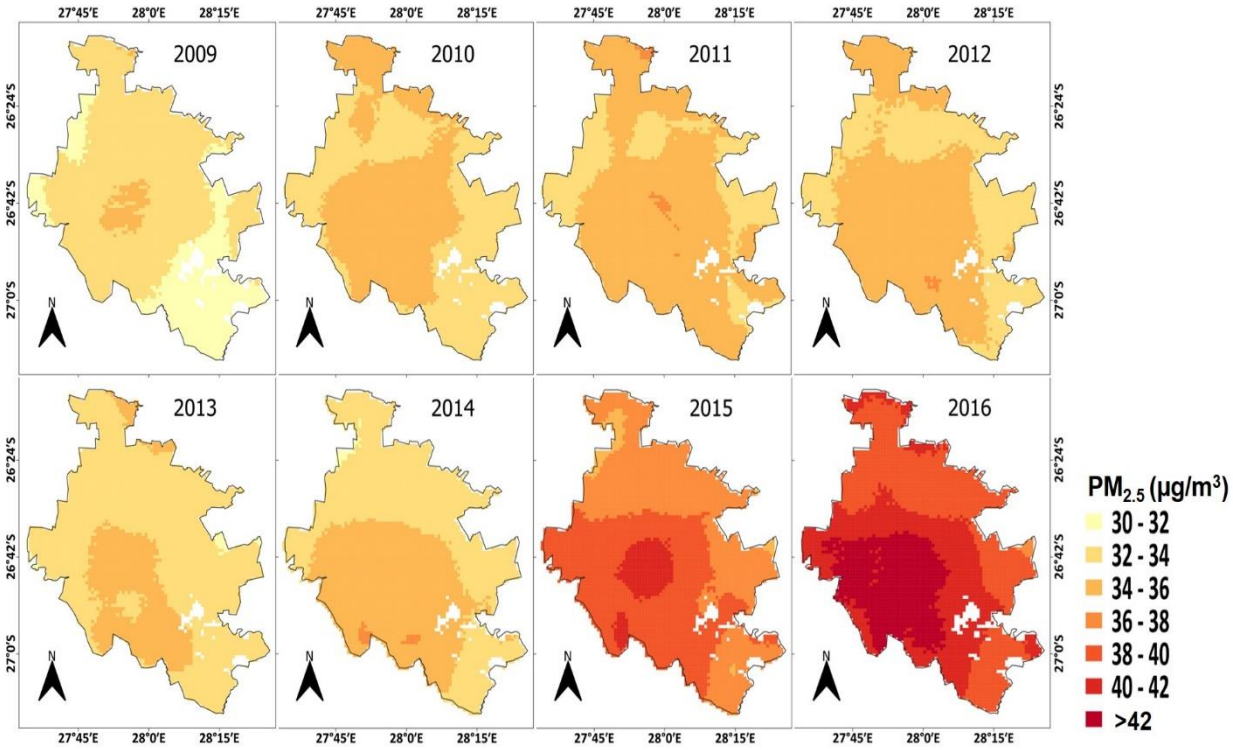

Figure 4-4: Spatial distributions of annual $PM_{2.5}$ concentrations in the VTAPA from 2009 to 2016.

4.4 Conclusion

This study evaluated the potential value of satellite remote sensing as a viable alternative to $PM_{2.5}$ ground-based monitoring in the VTAPA. There was a poor agreement between satellite-retrieved

$PM_{2.5}$ estimates and ground-level $PM_{2.5}$ measurements. Satellite retrievals tended to overestimate $PM_{2.5}$ concentrations resulting in inflated values throughout most of the VTAPA. In addition, the spatial resolution of the satellite retrievals is too coarse to provide useful spatial distribution information to fill the gaps in the sparse ground-based monitoring networks. For satellite remote sensing to be effectively exploited for air quality assessments in the VTAPA and elsewhere, further research to improve the precision and accuracy of satellite-retrieved $PM_{2.5}$ is required. This includes an independent assessment of the relationship between AOD and $PM_{2.5}$ in South Africa and the use of fine-scale satellite imagery such as Landsat (30m spatial resolution) for retrieving $PM_{2.5}$ estimates instead of medium resolution images such as MODIS.

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Chapter conclusion

The objective of this paper is to explore the spatial variations and temporal trends of particulate matter concentrations in the Vaal Triangle Priority Area

The main conclusions extracted from this study are:

- For satellite remote sensing to be a more viable option in future air quality monitoring programs, efforts have to be made to improve on its precision and accuracy
- Ground-based measurements reveal that air quality in the VTAPA is still a cause for concern as $PM_{2.5}$ concentrations still remain above national ambient air quality limits.

The next step in this study is to identify the sources contributing to the high $PM_{2.5}$ concentrations present in VTAPA so as to determine which sources need to be given more attention during the design of an air quality plan. This objective is addressed in the following chapter.

CHAPTER 5 UPDATED $PM_{10-2.5}$ AND $PM_{2.5}$ SOURCE APPORTIONMENT FOR LOW-INCOME SETTLEMENTS IN THE VAAL TRIANGLE AIRSHED PRIORITY AREA, SOUTH AFRICA

Journal article - Preface

Author list, contributions and consent

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The majority of the work, which included data gathering, analysis, interpretation and writing the manuscript was done by the first author, **L. Muyemeki.** S. J. Piketh and R. Burger, who co-authored this manuscript assisted in data explanation and reviewing the manuscript.

The co-authors of the article have been notified by the author of the intent to submit the PhD in article format and have given their consent which is attached in Annexure II.

Formatting and status of the article

The article was structured according to the journal stipulations of *Atmospheric Pollution Research*; to which it will be submitted. The article is presented in the style, format and length required by the journal. The guide for authors that was used to prepare this article is available at https://www.elsevier.com/journals/atmospheric-pollution-research/1309-1042/guide-for-authors. The article has not yet been submitted for publication.

Thesis objective addressed: To identify the sources contributing to particulate matter loading in low-income settlements of the Vaal Triangle Airshed Priority Area

Airborne PM is one of the major pollutants found in the VTAPA. Exposure to high levels of PM can increase the risk to residents in the VTAPA experiencing cardiovascular and respiratory problems. In order to design control strategies that will effectively reduce ambient concentrations to within regulatory limits, quantitative information on air pollution sources which includes their compositions and contributions to PM loading is required. This information can be acquired through the use of source apportionment models. Source apportionment models are important tools that can improve the understanding of the link between PM source emissions, ambient concentrations and their associated health effects. This study, therefore, seeks to identify the main sources of PM loading in the VTAPA, with a special focus on low-income settlements which have the highest receptors vulnerable to the effects of air pollution. The results from this study

will assist in determining which ambient PM sources need to be given more priority during the drafting of an air quality plan.

Abstract

The Vaal triangle, like other priority areas in South Africa, has a worrying air pollution problem. A comprehensive understanding of particulate matter sources in the different size fractions will assist in the selection and implementation of appropriate air pollution control strategies. Aerosol samples in the coarse ($PM_{10-2.5}$) and fine ($PM_{2.5}$) fraction were collected for three seasons at four sites. Three of the study sites were located in densely populated low-income suburbs (Sebokeng, Sharpeville and Zamdela) and the fourth site was located in a low population density area (Kliprivier). The elemental and ionic compositions of these samples were used to identify and characterise the sources contributing to particulate loading. The results demonstrated that the highest seasonal median concentrations of $PM_{10-2.5}$ (116 µg/m^3) and $PM_{2.5}$ (88 µg/m^3) were observed in Sharpeville during the winter, and the lowest concentrations of $PM_{10-2.5}$ and $PM_{2.5}$ were detected in the summer. At all sites, there was a high abundance of crustal elements in $PM_{10-2.5}$ and a dominance of coal and biomass combustion-related elements in $PM_{2.5}$. Eight sources of PM were identified using the Positive Matrix Factorization (PMF) receptor model. Dust related (33–55%) and secondary aerosols (17–20%) were the major contributing sources in $PM_{10-2.5}$. For $PM_{2.5}$, contributions were predominantly from coal-burning (60%>) for Sebokeng and Sharpeville and from secondary aerosols (51–90%) for Kliprivier and Zamdela.

Updated $PM_{10-2.5}$ and $PM_{2.5}$ source apportionment for low-income settlements in the Vaal Triangle Airshed Priority Area, South Africa

5.1 Introduction

Over the past decades, South Africa has experienced strong economic growth, industrial expansion and rapid urbanisation. This has led to the emergence of cities characterised by high population densities and, high industrial and traffic activities. Air pollution is a serious environmental problem in these urban areas and has attracted widespread attention from the public as a result of its negative effects on humans (Amegah & Agyei-Mensah, 2017). Pollution from particulate matter (PM) is of primary concern in South Africa. Exposure to PM is the fourth leading human health risk factor and is linked to over 5 million premature deaths all over the world (Stanaway *et al.*, 2018). Exposure to PM, especially $PM_{2.5}$, over long periods is dangerous to humans as inhaled particles will penetrate deep into the lungs and increase the risk of morbidity and premature mortality due to cardiopulmonary diseases and lung cancer (Norman *et al.*, 2007; Feng *et al.*, 2016). Effective strategies are urgently needed to improve air quality and address the health risks associated with PM. Acquiring reliable and comprehensive information on the main sources of PM is the first key step required to achieve this (Thunis *et al.*, 2019).

Source apportionment is an air quality management tool that can provide statistical information about source contributions which is important in the formulation of mitigation strategies for PM (Zhu *et al.*, 2018). Source apportionment air quality management includes three different approaches which include the Emission Reduction Impacts (ERI), Mass-Transfer (MT) and Incremental (INC) methods (Thunis *et al.*, 2019). The ERI method is a form of sensitivity analysis that compares the difference in modelled concentration results based on the baseline emissions of a source to that based on reduced emissions of the same source (Thunis *et al.*, 2018). This method is used for air quality simulations in the GAINS (Greenhouse Gas and Air Pollution Interactions and Synergies) model (Amann *et al.*, 2011). MT involves the use of receptor models to identify and quantify emission sources based on multivariate statistical inference (Hopke, 2016). The MT method is often used in source apportionment studies (Taiwo *et al.*, 2014). The approach used in the INC involves calculating the difference in concentrations from a specific site that is influenced by a source with that from another site that is not influenced by the same source (Lenschow *et al.*, 2001).

Attempts have been made in South Africa to apportion sources and their contributions, however, a few studies have reported on PM sources (Mathuthu *et al.*, 2019). Engelbrecht *et al.* (2002) used the Chemical Mass Balance (CMB) model to compare PM source contributions from residential coal and low-smoke fuels used in the township of Qalabotjha. The CMB model was

also applied to identify the PM sources contributing to air pollution in Kwadela township(van den Berg, 2015). Recently, Tshehla & Djolov (2018) used the Positive Matrix Factorization (PMF) receptor model to apportion PM sources in an industrialised rural area in the Limpopo province. A thorough understanding of the different compositions and contributions of PM will assist air quality planners in assigning precedence to key pollutant sources.

In 2006 the Vaal Triangle was classified as an *air pollution priority area* due to public health concerns over the elevated levels of air pollution faced in this region (Department of Environmental Affairs, 2019). The major local sources found in the Vaal Triangle Airshed Priority Area (VTAPA) include industries, residential areas, vehicles, waste and windblown dust (Department of Environmental Affairs, 2018). These sources occur within proximity to one another. In 2009, an Air Quality Management Plan (AQMP), detailing possible intervention strategies for the VTAPA was published (Department of Environmental Affairs and Tourism, 2009). The first 5-year cycle review of this AQMP in 2013 revealed that, despite efforts made, air pollutant concentrations were still above national ambient air quality standards (Department of Environmental Affairs, 2013). This was due to inadequate implementation of air quality controls (Department of Environmental Affairs, 2013). The second 5-year cycle of the VTAPA AQMP is currently in review. Target air quality limits still have not been met as daily, and annual average PM concentrations remain above the national standards (Department of Environmental Affairs, 2019). A revised source apportionment study is required to establish an updated understanding of the current sources contributing to PM and identify opportunities for further emission reductions.

This study, therefore, seeks to achieve the following objectives: (a) to explore the temporal and spatial variations of PM in the townships of the VTAPA; (b) to determine the elemental and ionic compositions of PM in the townships of the VTAPA, and (c) to identify and apportion the main sources contributing to PM pollution in the townships of the VTAPA.

5.2 Material and methods

5.2.1 Sampling sites

The VTAPA is situated on the high central inland plateau of South Africa with terrain elevations ranging between 1300 and 1900masl. The VTAPA stretches from the southern Gauteng to the northern section of the Free State province. The land-use in this region includes commercial, industrial, residential and low-intensity agricultural activities, all situated within close vicinity to one another. Four sites in the VTAPA were selected for this study. These sites (Figure 5-1) were selected based on a baseline assessment that identified these sites as experiencing ambient PM concentrations above national air quality guidelines (Thomas, 2008). Sebokeng (26.5879S, 27.8410E), Sharpeville (26.6810S, 27.8677E) and Zamdela (26.8449S, 27.8551E) sites are

situated inside densely populated low-income settlements, while Kliprivier (26.4203S, 28.0849E) site is in a low population density area.

Sebokeng is a low-income settlement in the VTAPA. The main sources of emissions in the area include iron and steel processes at ArcelorMittal and Davsteel, commercial boilers, small industrial activities and domestic solid fuel burning. The low-income settlement of Sharpeville located between Vereeniging Central Business District and Vanderbijlpark. The dominant emissions sources in this area are domestic solid fuel burning and metallurgical industries which include ArcelorMittal and Davsteel. Zamdela is a low-income settlement situated close to the town of Sasolburg. The main sources of pollution of air pollution this area include Sasol Chemical Industries Complex, Natref, Omnia Fertiliser, Karbochem, Safripol and Sigma Colliery. Domestic fuel burning is also an important emission source in Zamdela. Kliprivier is a low-density region located on the boundary of the Midvaal Local Municipality and Ekurhuleni Metropolitan Municipality. Grasslands and cultivated commercial fields make up most of land use in the area.

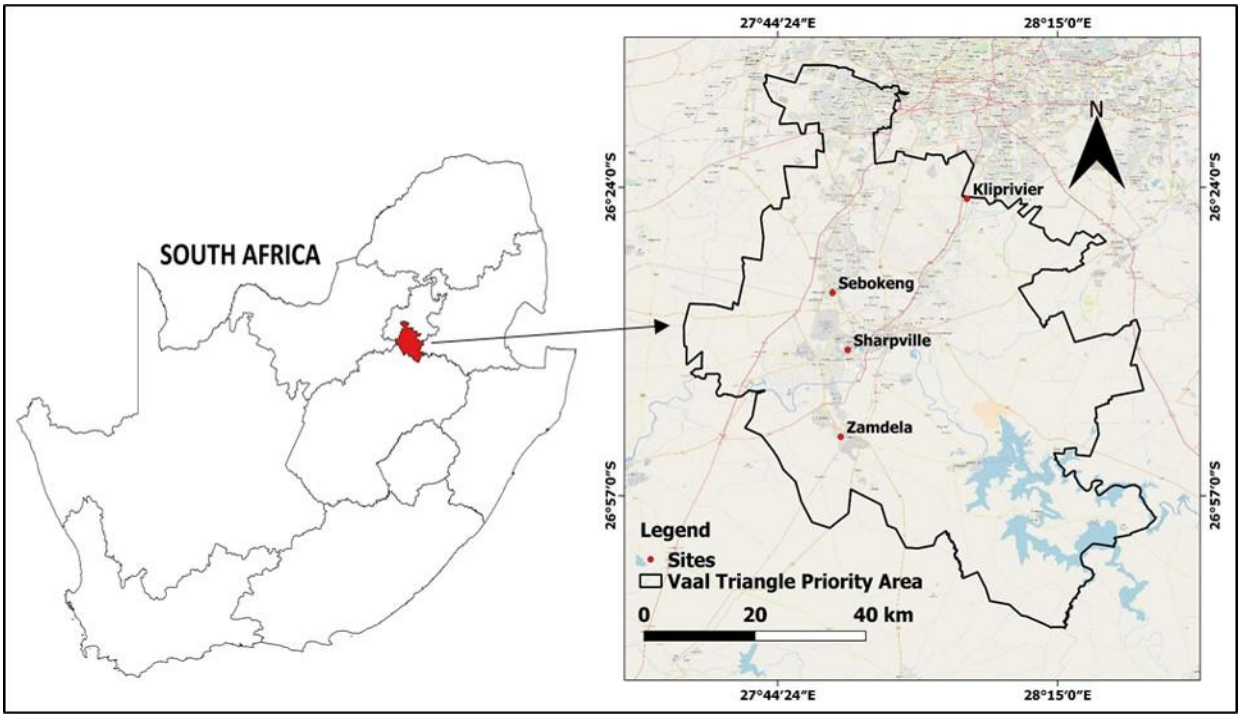

Figure 5-1: Study area map showing the location of the VTAPA in South Africa and the four sampling sites within the VTAPA.

5.2.2 Sampling strategy

Sampling was performed simultaneously at all of the sites for 16 days, in summer (February-March 2018), winter (June-July, 2018) and spring (September-October, 2018). Dichotomous low volume samplers (MicroPNS Type Dichoto LVS16, Umwelttechnik MCZ GmbH, Bad Nauheim, Germany) with split-flow rates of 1.7 L/min (for fine particles) and 15 L/min (for coarse particles)

were employed for the simultaneous and sequential collection of particulate matter in the fine ($PM_{2.5}$) and coarse ($PM_{10-2.5}$) fraction on Teflon filter membranes. Two consecutive continuous 12-hour samples for each size fraction were collected daily to enable comparisons between day (10:00 AM—10:00 PM) and night concentrations (10:00 PM—10:00 AM). In total, 768 Teflon filters were sampled for the entire campaign. Soil samples for different source categories were also acquired from different locations across the VTAPA. These samples were sieved, resuspended and analysed in the laboratory so as to determine their source profiles. These profiles were used to assist in identifying PM sources based on output results from the receptor modelling.

5.2.3 Chemical analyses

5.2.3.1 Elements

Trace elements on the Teflon filters were chemically analysed using X-Ray Fluorescence (XRF). XRF is a non-destructive procedure involving the interplay between x-ray photons and the elements found in the PM species leading to the discharging of electrons (Research Triangle Institute, 2009). This will result in the release of x-rays that are unique for the individual element.

5.2.3.2 Ions

Water-soluble ionic species on the Teflon filters were analysed using Ion Chromatography (IC). A Dionex ICS-3000 IC system consisting of two flow lines was used for IC analysis (Conradie *et al.*, 2016). One flow line was used for the detection of anion species and the other flow line to detect cation species. Before chemical analysis was commenced, the filters were leached in 10 ml de-ionised water in an ultrasonic bath for 30 minutes. Five standards, ranging from 20 ppb to 500 ppb, were prepared using certified stock solutions obtained from Industrial Analytical.

5.2.4 Positive Matrix Factorization (PMF) model analysis

$PM_{10-2.5}$ and $PM_{2.5}$ source contributions to ambient air particulate concentrations in the VTAPA were quantified using PMF. The PMF model is a multivariate factor analysis tool that deconstructs the matrix of speciated sample data into two matrices: factor contributions and factor profiles (Paatero *et al.*, 2014). This is a well-tested receptor model that has been applied globally (Bove et al., 2016; Chuang et al., 2016; Crilley et al., 2017). For this study, the PMF (Version 5.0) was performed to obtain quantitative source profiles and mass contributions (Norris *et al.*, 2014). The PMF model equation can be expressed as follows:

$$X_{ij} = \sum_{k=1}^{p} g_{ij} f_{kj} + e_{ij}$$

Equation 5-1 Positive Matrix Factorization

where X_{ij} is the concentration of species j measured on sample i; p is the number of factors contributing to the samples; f_{kj} is the concentration of species j in factor profile k; g_{ik} is the relative contribution of factor k to sample i, and e_{ij} is the error of the PMF model for the species j measured on sample i,

In order to run PMF, the model requires sample chemical species concentration values and uncertainty estimates for each species. Uncertainty estimates were conducted by dividing the limit of quantification from the mass concentration for each species in order to obtain a fractional value. Then an uncertainty is assigned to each species based on the specific range the fractional value of a particular species falls into. The PMF model was run multiple times for all sites using elemental and ionic composition data for $PM_{10-2.5}$ and $PM_{2.5}$.

The species used in the model were chosen according to the signal-to-noise (S/N) criterion. Species with S/N values greater than 2 were classified “strong”, while those within the 0.2 – 1.9 range were categorised as “weak”. Species with S/N values less than 0.2 are defined as “bad” variables and were removed from the analysis. The optimal number of factors for each site were selected based on (i) the number of runs/iterations, (ii) knowledge of sources affecting the study area, (iii) distributions of the scaled residuals and, (iv) the Qtrue/Qrobust ratio (Table 5-1) (Vossler *et al.*, 2016). Species with symmetrically distributed scaled residuals within a range of -3 to +3 indicates a good model fit. The Qtrue/Qrobust ratio is useful in determining the influence of outliers on the model. A ratio above 1.5 indicates that outliers may have a disproportionate effect on the model and will need to be down weighed (Gupta *et al.*, 2012; Weber *et al.*, 2019). A source type was assigned to each factor based on known representative indicator chemical species and source profiles obtained from the US EPA SPECIATE database (https://www.epa.gov/air-emissions-modeling/speciate-2). (Engelbrecht *et al.*, 2002; Simon *et al.*, 2010)

Table 5-1 Overview of the Qtrue/Qrobust values for Klipriver, Sebokeng, Sharpeville and Zamdela

	Summer		**Winter**		**Spring**	
	$PM_{10-2.5}$	**$PM_{2.5}$**	**$PM_{10-2.5}$**	**$PM_{2.5}$**	**$PM_{10-2.5}$**	**$PM_{2.5}$**
Kliprivier	1.24	1.00	1.43	1.00	1.08	1.00
Sebokeng	1.03	1.14	1.21	1.00	1.06	1.00
Sharpeville	1.00	1.00	1.37	1.24	1.12	1.00
Zamdela	1.46	1.11	1.48	1.00	1.17	1.11

5.3 Results

5.3.1 Spatial and temporal variations of $PM_{10-2.5}$ and $PM_{2.5}$ mass concentration

It can be observed that the highest seasonal median values for $PM_{10-2.5}$ were experienced in Sharpeville during the winter season for both the day (95 µg/m^3) and night (116 µg/m^3) time (Figure 5-2). Similarly, for $PM_{2.5}$ the highest seasonal median values are also observed in winter, with Sebokeng (68 µg/m^3) having the maximum concentrations during the day time and Sharpeville (88 µg/m^3) peaking during the night. Peak concentrations experienced in winter could be as a result of extensive domestic solid fuel burning for cooking and heating. In comparison to winter, PM pollution was also considerably higher in spring. For $PM_{10-2.5}$, concentration median values for spring were highest in Sebokeng during the day (92 µg/m^3), and in Sharpeville (during the night (97 µg/m^3). $PM_{2.5}$ levels were also high in spring with median values for the day peaking in Sharpeville (59 µg/m^3) and maximum night time median concentrations being experienced in Sebokeng (75 µg/m^3). For all seasons, both during the day and night, $PM_{2.5}$ median values for Kliprivier were generally lower than Sebokeng, Sharpeville, and Zamdela. The lowest seasonal median values for $PM_{10-2.5}$ and $PM_{2.5}$ were experienced in summer

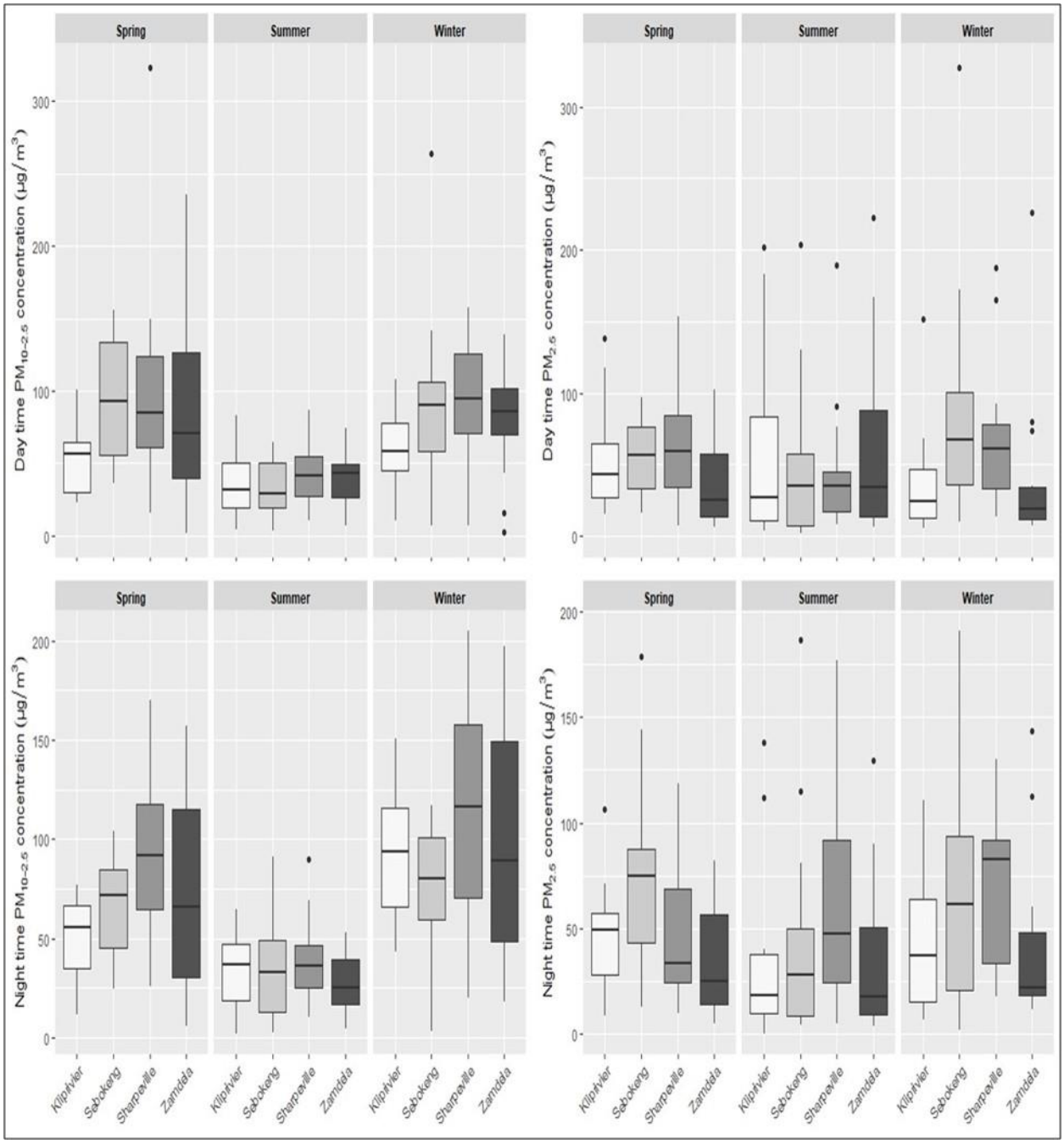

Figure 5-2: Day and night time seasonal range of $PM_{2.5}$ and $PM_{10-2.5}$ mass concentration at all the sampling sites.

5.3.2 $PM_{10-2.5}$ and $PM_{2.5}$ chemical composition

The elemental and ionic contents of $PM_{10-2.5}$ and $PM_{2.5}$ for summer, winter and spring at the four sampling sites are shown Figure 5-3, Figure 5-4 and Figure 5-5 respectively. Statistical summaries of the elemental and ionic species for each site are given in Annexure I.

5.3.2.1 Elements

Analysis of the data revealed that at all sites, crustal elements which include Si, Mg, Al, Ca, Na, S and Fe contributed the most towards $PM_{10-2.5}$ both during the day and night for all seasons. These crustal elements were highest in spring which is a season associated with strong winds in South Africa. The abundance of Si, Mg, Al, Ca, Na, and Fe in $PM_{10-2.5}$ indicate that dust is dominant at these sites. There is also a strong presence of S in $PM_{2.5}$ for both day and night at all sites during all three seasons, implying that coal combustion is an important contributor to atmospheric PM. There is an abundance of K and Zn in the $PM_{2.5}$ during the days and nights of winter and spring. These elements could have been emitted as a result of wood and biomass burning. Fe, Cr and Ni were dominant in $PM_{2.5}$ during the day and night for Kliprivier (summer) and Zamdela (winter and spring). Fe, Cr and Ni were also significant contributors of $PM_{2.5}$ during the summer nights at Kliprivier and Sebokeng. This suggests that industries and vehicles make a significant contribution to ambient PM.

5.3.2.2 Ions

Ionic compositions for $PM_{10-2.5}$ and $PM_{2.5}$ revealed that SO_4^{2-}, NH_4^+ and F^- are the dominant species for summer, winter and spring at all sites. The occurrence of these ionic species could be a result of coal combustion from industries and to a lesser extent, residential solid fuel burning. In addition to this, the strong presence of NO_3^- in $PM_{10-2.5}$ and $PM_{2.5}$ during the day and night in winter could suggest an industrial origin. SO_4^{2-}, NH_4^+, F^- and NO_3^- concentrations are highest in winter mainly as a result of increased coal combustion. High Na^+, Mg^{2+} and Ca^{2+} abundances were observed for $PM_{10-2.5}$ suggesting the possibility of marine and crustal origin sources. Wood and biomass burning is also an important source as indicated by the abundance of in K^+ in $PM_{2.5}$, especially in winter where it is highest due to the need for space heating

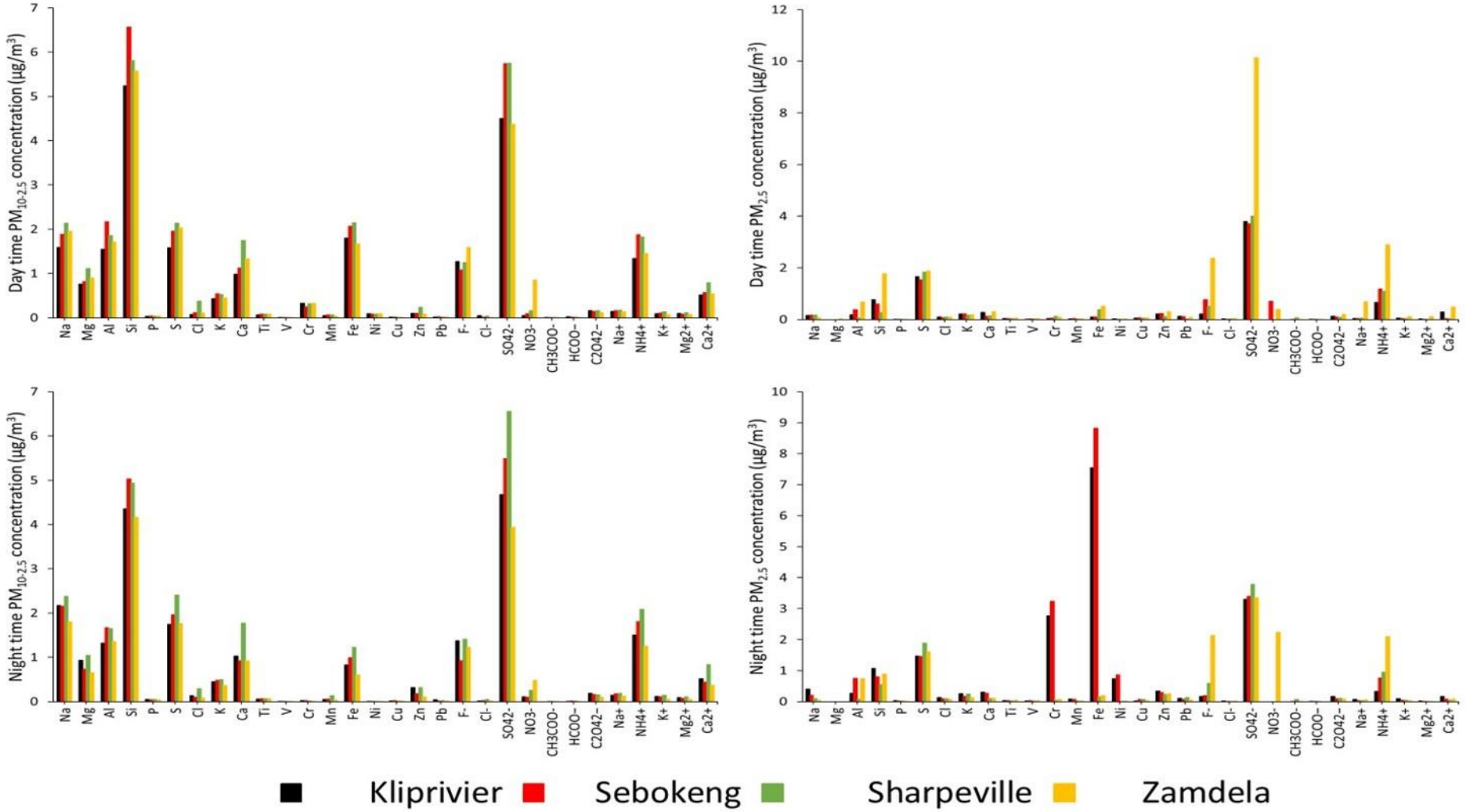

Figure 5-3: Summer day (top left and right) and night time (bottom left and right) average elemental and ionic composition of $PM_{10-2.5}$ and $PM_{2.5}$ at all the sampling sites.

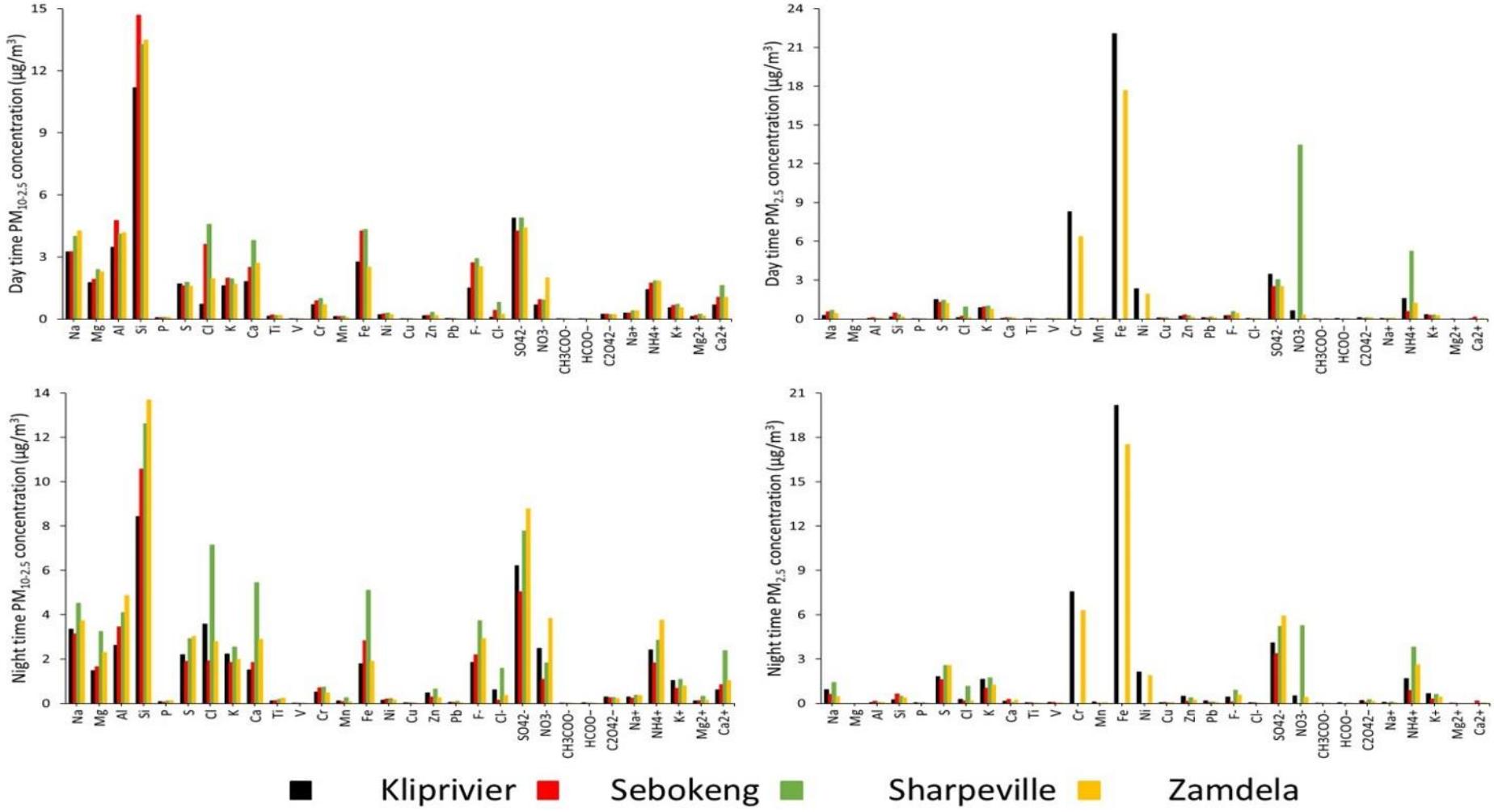

Figure 5-4: Winter day (top left and right) and night time (bottom left and right) average elemental and ionic composition of $PM_{10-2.5}$ and $PM_{2.5}$ at all the sampling sites.

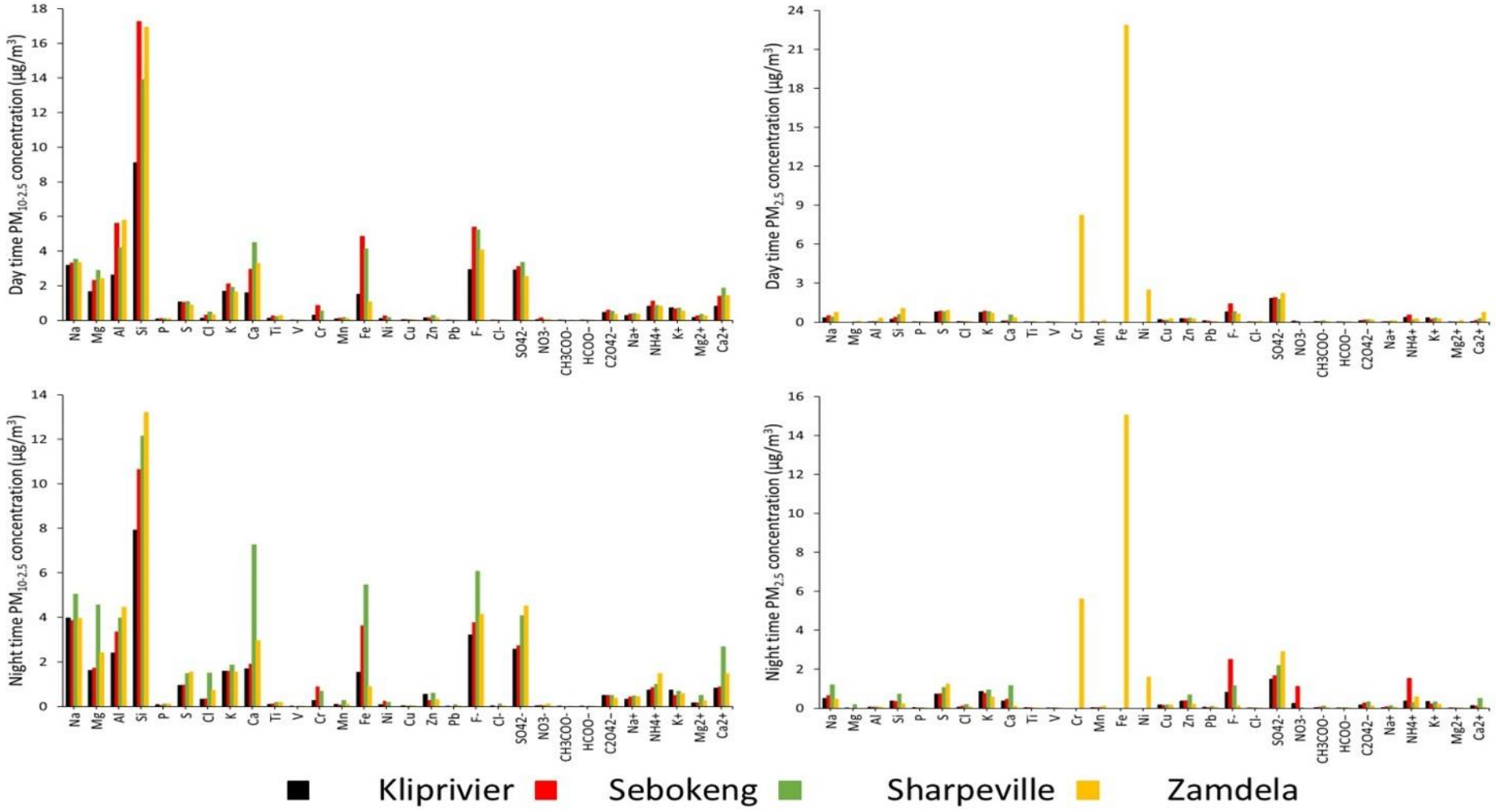

Figure 5-5: Spring day (top left and right) and night time (bottom left and right) average elemental and ionic composition of $PM_{10-2.5}$ and $PM_{2.5}$ at all the sampling sites.

5.3.3 Apportionment of sources identified by PMF

Eight factors were determined based on representative indicator chemical species. These identified potential sources included industry, coal burning, wood and biomass burning, waste burning, dust-related, vehicles, secondary aerosols and aged sea salt (Figure 5-6). Figure 5-7 presents the source apportionment results for the four sampled sites and shows the variations in contributions based on the three seasons.

5.3.3.1 $PM_{10-2.5}$ and $PM_{2.5}$ sources

The industry source is typically characterised by strong contributions from Zn, Fe, Pb, Ni, Cr, Mn and V. These elements are typically associated with smelters and metallurgical industries (Dall'Osto *et al.*, 2013). The metal element V is mainly associated with heavy fuel oil combustion (Yu *et al.*, 2013). Coal, coking coal and heavy fuel oil are the main energy sources that drive industries in the VTAPA (Department of Environmental Affairs and Tourism, 2009). Coal burning is an important source identified through PMF. For the VTAPA, this source is generally associated with burning in low-income households and industries. The coal-burning source is highly loaded with Cl^-. This ion is mainly from ammonium chloride (NH_4Cl), which occurs as a result of the rapid reaction between HCl and NH_4^+ in the atmosphere (Chen *et al.*, 2014). With coal being the primary energy source in South Africa, coal-burning can be regarded as the largest potential source of HCl.

Wood and biomass burning are characterised by a high content of K and K^+ and minor contributions of SO_4^{2-} and NO_3^-. K^+ is widely recognized as an indicator of biomass burning as it is released during the plant combustion process (Chuang *et al.*, 2016). For the low-income settlements in the VTAPA, K^+ is more representative of wood combustion as wood is an important energy source for cooking and space heating (Department of Environmental Affairs, 2019). Biomass burning occurs in the VTAPA through burning of open spaces used for agricultural activities (Department of Environmental Affairs and Tourism, 2009).

Refuse collection in the low-income settlements of South Africa is infrequent and has resulted in the pile-up of solid waste into heaps. As a measure to reduce these heaps, residents have resorted to burning waste (Department of Environmental Affairs, 2019). The waste burning source identified through PMF contained high values of NH_4^+ and small contributions from Cl^- and K^+. The occurrence of Cl^- could be as a result of the presence of salt-containing foodstuff and chlorine-based materials in domestic waste disposals. Residents in low-income settlements often don't sort their waste before disposal. This can result in domestic waste being mixed with garden waste, thus likely explaining the presence of K^+ in the waste burning source.

The dust-related source was identified in the coarse fraction and was characterised by crustal elements which included Ca, Mg, Si, Al, Fe, Ti and Mn. The metals elements found in the dust-related source could also be associated with resuspended dust resulting from motor vehicle entrainment on unpaved roads in the low-income settlements. Other metals such as Cr and V were also present in the dust-related source and could be as a result of soil contamination from industrial emissions (Okonkwo *et al.*, 2006). NO_3^-, Pb, Zn, Mn and Fe were characteristic of the vehicle source. The Zn, Mn and Fe metal elements found in this source are associated with both petrol and diesel-fueled vehicles (Squizzato *et al.*, 2018). These elements are also associated with brake, tear and engine wear (Park *et al.*, 2019). Zn is a common additive found in lubricating oils and can be emitted through combustion by diesel engines (Yu *et al.*, 2013).

The secondary aerosol source mainly consisted of SO_4^{2-}, NO_3^- and NH_4^+, which was formed through the chemical transformation of SO_2, NO_x and NH_3 pollutants originating from other direct sources. The presence of secondary aerosols in the VTAPA could also be as a result of long-range transportation by air masses from the sea. The aged sea salt source was characterised by high loadings of Na and low Cl levels. The lack of Cl in the aged sea salt source could result from Cl displacement in sea salt particles by acidic pollutants (H_2SO_4 and HNO_3) leading to the formation of sulfate and nitrate salts (Laskin *et al.*, 2012).

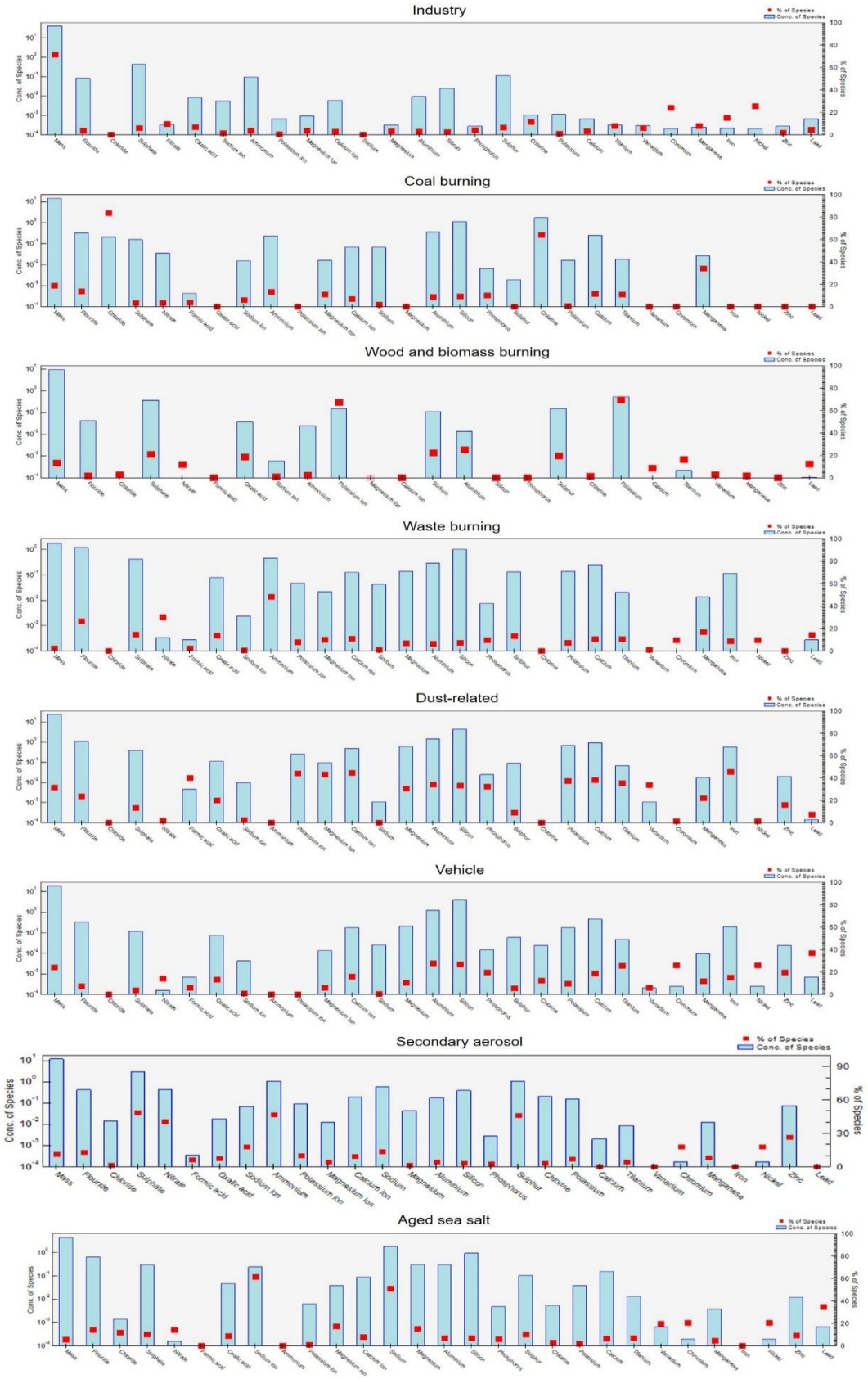

Figure 5-6: Source profiles identified through PMF analysis

5.3.4 Seasonal contributions

Source apportionment results (Figure 5-7) reveal that for $PM_{10-2.5}$, dust-related is a major source at Kliprivier (32% (16 µg/m^3) — 52%(18 µg/m^3)), Sebokeng (31% (24 µg/m^3) — 68%(23 µg/m^3)), Sharpeville (34%(14 µg/m^3) — 49%(50 µg/m^3)), and Zamdela (19%(15 µg/m^3) — 65%(17 µg/m^3)). Dust-related contributions show relatively higher concentrations in summer and spring, and lower concentrations in winter. This is consistent with the seasonal patterns of winds experienced in the central and northern parts of South Africa (Kruger *et al.*, 2016). For the coarse fraction, secondary aerosols have an important contribution at Kliprivier (12%(9 µg/m^3) — 32%(16 µg/m^3)), Sebokeng (11%(9 µg/m^3) — 14%(11 µg/m^3)), Sharpeville (12%(12 µg/m^3) — 35%(15 µg/m^3)), and Zamdela (29%(24 µg/m^3) — 32%(25 µg/m^3)). For summer this could mainly be as a result of the regional transportation of secondary sulphate and nitrate aerosols from coal-fired power stations from the industrial region of Mpumalanga whilst for spring, secondary aerosols could be from the intensive agricultural region of the Free State province. (Tesfaye *et al.*, 2011).

Coal combustion and vehicles are sources prominent in the coarse fraction. Coal combustion accounts for 4%(3 µg/m^3) to 19%(7 µg/m^3), 6%(2 µg/m^3) to 19%(14 µg/m^3), 8%(8 µg/m^3) to 14%(13 µg/m^3), and 7%(6 µg/m^3) to 16%(13 µg/m^3) of $PM_{10-2.5}$ mass concentrations in Kliprivier, Sebokeng, Sharpeville, and Zamdela, respectively. Vehicles account for 11%(8 µg/m^3) to 20%(7 µg/m^3), and 16%(5 µg/m^3) to 25%(19 µg/m^3) of $PM_{10-2.5}$ mass concentrations in Kliprivier and Sebokeng, respectively. Vehicles contributed to 10% of $PM_{10-2.5}$ mass concentrations for both summer and winter at Sharpeville (4 µg/m^3 and 10 µg/m^3). In winter vehicles contributed 14%(11 µg/m^3) of $PM_{10-2.5}$ mass concentrations at Zamdela.

Coal-burning, secondary aerosols, wood and biomass burning, and industries are the key PM sources in the fine fraction. Coal-burning is the main source of $PM_{2.5}$ air pollution in Sebokeng and Sharpeville, contributing over 60% for all three seasons with the highest concentrations being experienced in winter (57 µg/m^3 and 73 µg/m^3). Secondary aerosols are a key $PM_{2.5}$ source in Zamdela with contributions ranging from 24%(4 µg/m^3) to 67%(6 µg/m^3). Secondary aerosols are also an important $PM_{2.5}$ source at Kliprivier. The contribution from secondary aerosols for all three seasons in Kliprivier varied from 17%(5 µg/m^3) to 22%(5 µg/m^3). Industries account for 5%(1 µg/m^3) to 11%(3 µg/m^3), 7%(5 µg/m^3) to 14%(10 µg/m^3), 10%(9 µg/m^3) to 12%(7 µg/m^3), and 18%(3 µg/m^3) to 35%(6 µg/m^3) of $PM_{2.5}$ mass concentrations in Kliprivier, Sebokeng, Sharpeville, and Zamdela, respectively.

Wood and biomass burning is an important source identified in both fractions, accounting for 15%(15 µg/m^3 in winter) to 25%(23 µg/m^3 in spring) and 6%(5 µg/m^3 in winter) to 14%(11 µg/m^3

in spring) of $PM_{10-2.5}$ mass concentrations in Sharpeville and Zamdela, respectively. The higher contributions in spring for both Sharpeville and Zamdela are consistent with the biomass burning patterns in South Africa which occur during late winter and early spring (Hersey *et al.*, 2015). Wood and biomass burning accounts for 26%(20 μg/m^3 in winter) and 17%(13 μg/m^3 in spring) of $PM_{10-2.5}$ mass concentrations in Kliprivier and Sebokeng, respectively. In the fine fraction, wood and biomass burning accounts for 72%(22 μg/m^3) to 84%(8 μg/m^3), 2%(1 μg/m^3) to 13%(9 μg/m^3), 4%(3 μg/m^3) to 6%(4 μg/m^3), and 32%(6 μg/m^3) to 49%(8 μg/m^3) of PM mass concentrations in Kliprivier, Sebokeng, Sharpeville, and Zamdela respectively. For Kliprivier, Sebokeng, and Sharpeville, the concentrations of the wood and biomass burning source were highest in spring as extensive biomass burning activities take place during August and September (Tesfaye *et al.*, 2011). Regional transportation also plays a significant role during the same period as biomass burning emissions originating from Zambia, Angola, Mozambique and Zimbabwe are transported to South Africa (Hersey *et al.*, 2015).

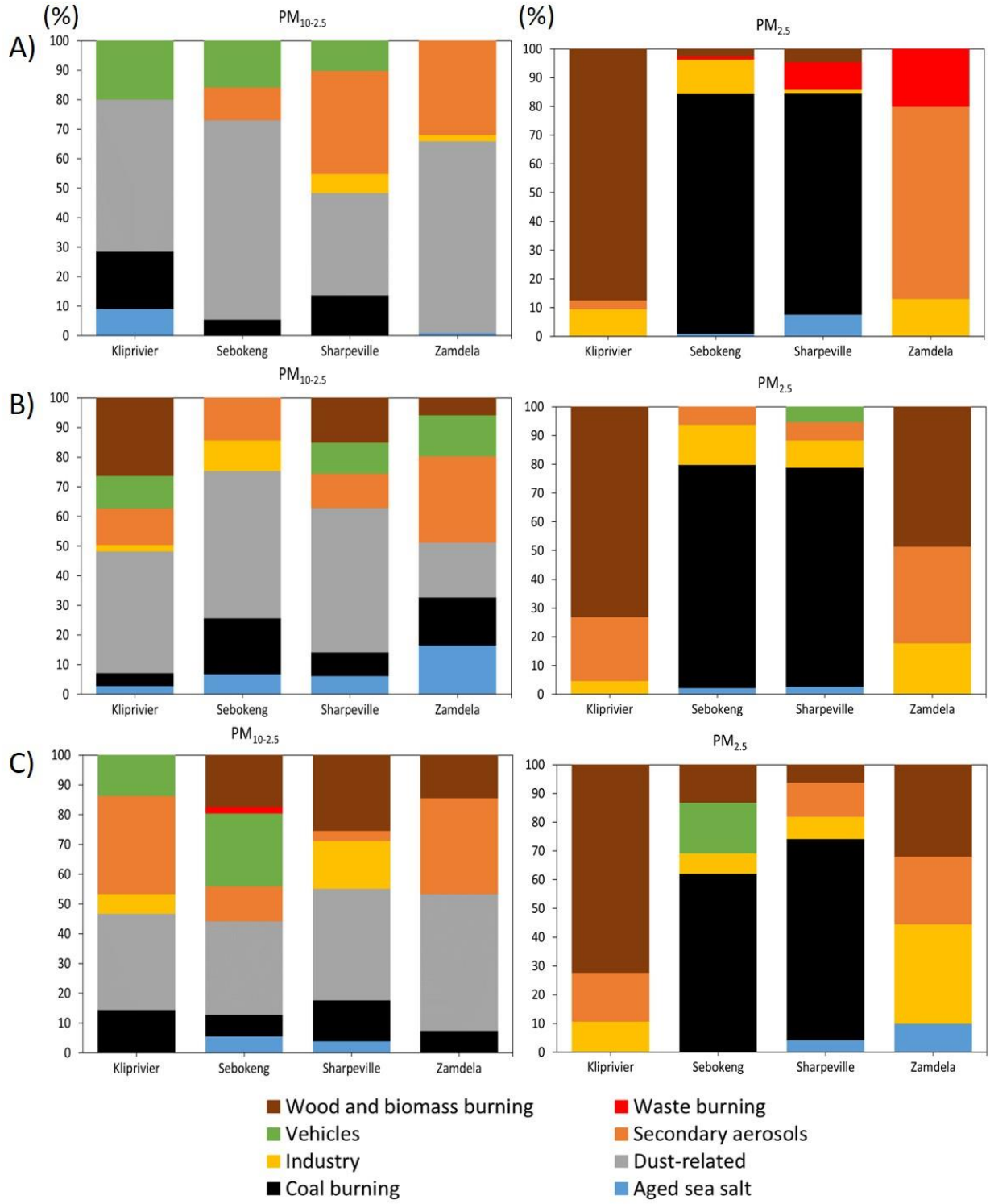

Figure 5-7: Relative (%) source contributions for $PM_{10-2.5}$ and $PM_{2.5}$ at all sites for A) summer B) winter and C) spring.

5.4 Discussion

This study has demonstrated that particulate air pollution remains a significant issue in the VTAPA. The air quality conditions in the low-income settlements of Sebokeng, Sharpeville and

Zamdela are harmful to the health to the residents. There were significant temporal variabilities in PM levels at these low-income settlements with average $PM_{2.5}$ on most occasions exceeding the 24-hour NAAQS (40 µg/m^3). Peak concentrations were experienced in winter when demand for space heating is high.

Contributions to PM in the low-income settlements of the VTAPA were primarily associated with anthropogenic activities. In the coarse fraction, the dust-related source (mainly composed of aluminium silicates) was the main contributor to ambient PM pollution, accounting for over 30% of concentrations at all sites except Zamdela. Elemental compositions for $PM_{10-2.5}$ also show an abundance of dust-related elements in the form of Si, Mg, Al, Ca, Na, and Fe. This source could have been generated locally through resuspension of soil, ash dumps, cement manufacturing and construction works as well as through regional transportation of dust aerosols (Tesfaye *et al.*, 2011).

In the fine fraction, secondary aerosols are important contributors to ambient PM pollution in Kliprivier and Zamdela. For Zamdela, the secondary aerosols are likely to have an industrial origin as Sasol Chemical Industries Complex is located within the vicinity of this area. The presence of secondary aerosols at Kliprivier is likely to be from coal-fired power stations. Kliprivier is also representative of transboundary pollution as it located at the boundary of the VTAPA. Apart from secondary aerosols, wood and biomass burning is also an important PM source in Kliprivier and Zamdela. In Zamdela, the concentrations of wood and biomass burning were highest in winter and this could be due to the extensive use of wood by households for space heating. Wood is the main solid fuel source for cooking and space heating in Zamdela (Statistics South Africa, 2016). High wood and biomass burning contributions in Kliprivier could be likely from burning activities at grasslands and cultivated commercial fields. Coal-burning had the highest contributions in Sebokeng and Sharpeville with peak concentrations being experienced in winter. These results are expected as domestic fuel combustion in low-income settlements is higher during winter due to the high demand for space heating (Adesina *et al.*, 2020).

5.5 Conclusion

$PM_{10-2.5}$ and $PM_{2.5}$ aerosol samples were collected for three seasons at four sites in the VTAPA industrial/urban region and were chemically analysed. Elemental and ionic compositions for these samples show an abundance of crustal elements in $PM_{10-2.5}$ and a predominance of coal and biomass combustion-related elements in $PM_{2.5}$ at all sites. Eight sources for $PM_{10-2.5}$ and $PM_{2.5}$ were resolved and identified using the PMF model and include industry, coal burning, wood and biomass burning, waste burning, dust-related, vehicles, secondary aerosols and aged sea salt. For the coarse fraction, dust-related and secondary aerosols were the major contributing sources.

In the fine fraction, secondary aerosols and coal burning had the highest contributions. Wood and biomass burning is an important source in both the coarse and fine fraction.

The present study has demonstrated the importance of source apportionment tool in the management of air quality management in the townships of the VTAPA. For Kliprivier, appropriate abatement strategies should focus on reducing emissions from dust and vehicles. The main emission sources to target in Sebokeng are dust, vehicles and domestic coal burning. In Sharpeville, focus should be on reducing emissions from domestic coal burning, dust, industry and vehicles. Abatement strategies in Zamdela should focus on industry and dust emission sources. Reducing the strength of these sources will benefit residents in the VTAPA by lowering PM exposure and improving air quality.

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Chapter conclusion

The objective of this article is to identify the sources contributing to particulate matter loading in low-income settlements of the Vaal Triangle Airshed Priority Area

From the above article, the following main conclusions were drawn:

- Although the VTAPA is a heavily industrialised region, industrial sources do not predominantly contribute to ambient PM in low-income settlements. Unregulated sources such as windblown dust, domestic coal burning and, wood and biomass combustion have considerable contributions
- Results from this study can assist in the design of local municipality air quality management plans for the VTAPA.

With the identification of the sources contributing to PM loading in the VTAPA, the next step in the thesis is to then identify suitable, cost-effective strategies to reduce emissions from these sources. The potential impact of these emissions reduction strategies on air quality in the VTAPA will also be assessed. This objective is addressed in the next chapter.

CHAPTER 6 INTEGRATED ASSESSMENT OF STRATEGIES TO REDUCE AIR POLLUTION IN THE VAAL TRIANGLE AIRSHED PRIORITY AREA, SOUTH AFRICA

Journal article - Preface

Author list, contributions and consent

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The majority of the work was done by the first author, **L. Muyemeki,** who collected, analysed and interpreted the data, and wrote the manuscript. Co-author contributions were as follows: S. J. Piketh and R. Burger were the promoters of the study, who helped in interpreting the data and reviewing the manuscript. P. Rafaj and G. Kiesewetter assisted with data processing and analysis.

All the co-authors gave written permission for the manuscript to be submitted as part of the PhD (see Annexure II).

Formatting and status of the article

The article was organised according to the journal specifications of *Air Quality, Atmosphere & Health*; to which it will be submitted. The article is presented in the style, format and length prescribed by the journal. The guide for authors that was followed in the preparation of the article is available at https://www.springer.com/journal/11869/submission-guidelines. At the time when this PhD was submitted for examination, this article had not yet been submitted for review, but the intent is to submit it afterwards.

Thesis objective addressed: To assess the potential impacts of emission reduction strategies on air quality in the Vaal Triangle Airshed Priority Area

The use of Integrated Assessment Models to support the design of effective air quality management plans has increased in the last two decades. By bringing together data on emissions from different pollutants, atmospheric dispersion, mitigation controls and their costs to implement, different air quality policies can be compared. This will aid in selecting the most appropriate management option, which has the least impacts on the economy of a particular area. The air quality management plan for the VTAPA is currently in its second stage of review, and the targets set from this plan still have not been met. Incorporating Integrated Assessment Models into the

review process will assist in decision making. In this regard, the purpose of this study is to explore different air quality policy scenarios for the VTAPA using an Integrated Assessment Model methodology so as to identify a suite of control strategies that will optimise benefits to humans and the environment at the least financial costs.

Abstract

South Africa currently faces a serious problem of air pollution in its cities. The approach used to manage air quality has not yet met the desired outcomes. In this study, potential mitigation options for ambient $PM_{2.5}$ in the Vaal Triangle Airshed Priority Area (VTAPA) – one of the most industrialised regions in South Africa - were explored using the GAINS (Greenhouse Gas and Air Pollution Interactions and Synergies) model Based on projections of economic activities and population, changes in $PM_{2.5}$ concentrations were quantified, applying different assumptions on application rates of end-of-pipe control technologies for key emission sources of $PM_{2.5}$ and its precursor gases NO_x and SO_2. Ambient $PM_{2.5}$ concentrations were used to estimate potential human health benefits of emission reductions. Our findings reveal that $PM_{2.5}$ concentrations for the VTAPA will not reach recommended health limits by 2035 under the current legislation. By introducing stringent controls, emissions could be reduced by more than half and national air quality standards could be attained. Trajectories show that implementation costs in the alternative scenario would be nearly twice as much as that for the current policies. Analysis using the GAINS interface has demonstrated the value of assessing the multiple dimensions of air pollution using an integrated approach to provide evidence-based support for policy decision making.

Integrated assessment of strategies to reduce air pollution in the Vaal Triangle Airshed Priority Area, South Africa

6.1 Introduction

Air pollution is recognised as a serious environmental problem for South Africa. Of particular concern are the high levels of particulate matter (PM) that have been recorded in some regions of the country (Feig *et al.*, 2019). In 2016, the death rate attributed to ambient air pollution in South Africa was was 41 per 100 000 population (World Health Organization, 2018). Additionally, the rising levels of nitrogen oxides (NO_x), partly attributed to an upsurge of vehicle use, has become an emergent air quality issue. Between December 2008 and January 2019, South Africa experienced a considerable increase in vehicle numbers from approximately 9 to 12 million (eNaTIS, 2019). NO_x emissions are not only an important component in secondary PM generation but also contribute to ozone formation, which in large amounts is harmful to humans (Karagulian *et al.*, 2015).

Current environmental legislation in South Africa seeks to reduce air pollution to acceptable human health levels by providing regulatory tools that will assist in achieving compliance with national ambient air quality standards (South Africa, 2013). One such instrument is proclaiming problem areas as a priority with stricter legislative requirements in which explicit management strategies are to be adopted. South Africa currently has three priority areas with the Vaal Triangle Airshed Priority Area (VTAPA) being the first to be proclaimed on 21 April 2006 (South Africa, 2006). PM levels in this priority area frequently exceed national ambient air quality standards with low-income settlements such as Sharpeville experiencing annual $PM_{2.5}$ concentration levels above 70 µg/m^3 (Govender & Sivakumar, 2019). In order to tackle air quality issues in the VTAPA, an air quality management plan (AQMP) outlining the proposed control strategies for the region was developed in 2009. Interventions proposed in the AQMP focused on reducing emissions from industries, households, motor vehicles, mines and waste burning activities through enforcing regulations and adopting mitigation technologies (Department of Environmental Affairs, 2009). Efforts to lower emissions have been mainly focused on the industrial sector and have led to some industries experiencing substantial reductions. However, these undertakings have not significantly cut back exposure to pollutants in residential areas (Department of Environmental Affairs, 2013).

A mid-term review of the VTAPA AQMP revealed that the desired outcomes for this priority area had not been met, as concentrations are still above national ambient air quality limits (Department of Environmental Affairs, 2013). This is in part due to a priority issue; compared to other risks like poverty, malnutrition and non-pollution related diseases such as HIV/AIDS, air pollution is still not considered an important priority and thus given less attention by the government (Naiker *et al.*,

2012). Additionally, there is a lack of follow up by authorities, and there are no penalties for failure to implement the recommended interventions in the AQMP (Department of Environmental Affairs, 2019). Financial constraints have always been a big concern for air quality management (Scorgie, 2012). Mitigation technologies are costly for industries and with South Africa currently experiencing economic challenges, fewer financial resources are allocated to air pollution control than other key strategic areas. Therefore, to tackle air quality issues under the current economic climate and competing priorities, there is a need to identify alternative policy interventions that improve air quality with minimal costs to the South African economy.

Integrated assessment tools like the GAINS (Greenhouse Gas and Air Pollution Interactions and Synergies) model can offer practical solutions to tackle the air quality management challenges currently faced in the VTAPA. The GAINS model has the capability to evaluate and compare different air quality policies and control strategies for a country according to their economic and social costs (Amann, 2012; Rafaj *et al.*, 2018). Through the assessment of emission levels under different control and activity scenarios, the risk of error when selecting mitigation options can be significantly reduced. The GAINS model has been successfully applied in different regions around the world, including developing countries such as Pakistan (Purohit *et al.*, 2013) and India (Amann *et al.*, 2017). In South Africa, the GAINS model has been used to assess emission trajectories under different management options at the country level (Henneman *et al.*, 2016). However, given the high variability in the concentrations of pollutants observed across South Africa, it is important to look at emissions at a more local scale (Scorgie, 2012). In this study, a local version of the GAINS model is employed in the VTAPA to quantify the impacts of mitigation options under different policy scenarios and determine the induced air quality and health impacts resulting from emission reductions.

6.2 Methods

6.2.1 Study area

The VTAPA is an industrial region experiencing strong emissions from industries, residential combustion, motor vehicles and mining (Department of Environmental Affairs, 2019). This area spans from the southern part of the Gauteng province or Gauteng city-region (GCR) to the northern part of the Free State province. The VTAPA (Figure 6-1) has a population of over 3 million people with more than 60% of its inhabitants concentrated in low-income areas where the burning of solid fuels is a common practice (Department of Environmental Affairs and Tourism, 2009). Waste combustion, biomass burning and fugitive dust are also important air pollution sources in the VTAPA.

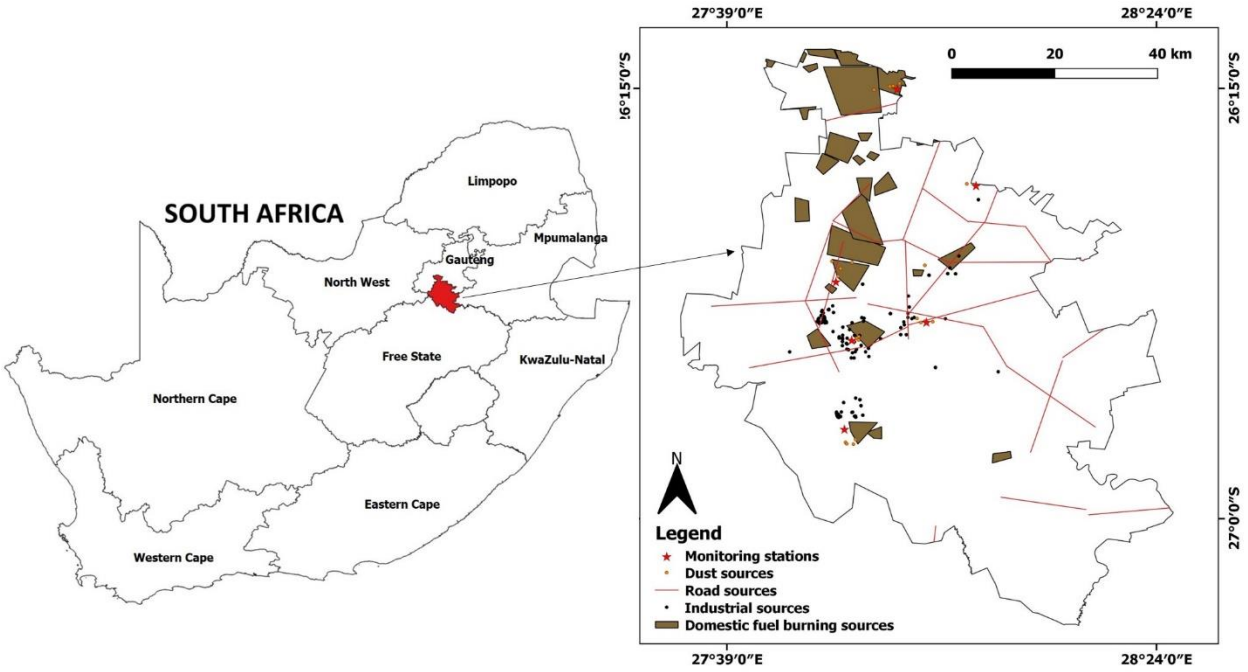

Figure 6-1: Map of the study region. The shaded red region represents the VTAPA and the zoomed shows the emission sources found within the VTAPA.

6.2.2 GAINS model

The GAINS model is a techno-economic tool developed by the International Institute for Applied Systems Analysis (IIASA). It is used to simulate emission trajectories for major air pollutants based on future economic growths (Amann, 2012). Emissions for major pollutants (SO_2, NOx, PM_{10}, $PM_{2.5}$, NH_3 and VOCs) and greenhouse gases (CO_2, CH_4, N_2O) from various activities (including power generation, industrial combustion and processes, transport, residential combustion and agriculture) are quantified in the model.

Four parameters are used to calculate emissions of air pollutants in the GAINS model, including sectoral activity data, uncontrolled emission factors, reduction efficiency of the abatement technologies, and the implementation rates of these measures. Sectoral activity data includes activities from different sectors and are expressed as primary energy units. Uncontrolled emission factors are the emissions per unit activity if no emission abatement measure is applied. The reduction efficiency and implementation of rates of abatement measures are expressed as a percentage of uncontrolled emission factors, and as a percentage of the activity data considered, respectively. Emission calculation in the GAINS model is expressed in Equation 6-1 (Amann *et al.*, 2011):

$$\boldsymbol{Em_{i,j}(t) = \sum_{k,m} act_{j,k}(t) * ef_{i,j,k,m} * X_{i,j,k,m}}$$

Equation 6-1: GAINS model

where; $Em_{i,j}(t)$ = emission of pollutant *i* in region *j* in year *t*; $act_{j,k}(t)$ = activity level of type *j* in region *k* in year *t*; $Ef_{i,i,k,m}$ = emission factor of pollutant *i* for activity *k* in region *j* after application of control measure *m*; $X_{i,j,k,m}$ = implementation rate of the abatement measure *m* for pollutant *i* for activity *k* in region *j*,

6.2.3 Data input for GAINS

The GAINS model requires energy activity information as input data into generating emissions. However, the data available for the VTAPA is incomplete. Therefore, publicly available energy activity data for the Gauteng province was used in GAINS as most of the VTAPA falls within this region. The energy activity data used was obtained from the greenhouse gas inventory and energy balance study for the Gauteng province which was conducted by the University of Stuttgart's Institute for Energy Economics and the Rational Use of Energy (Tomaschek *et al.*, 2012). These data provide the most comprehensive and complete energy activity statistics for the Gauteng region. The data was gathered from national statistics, e.g. fuel sales, electricity sales, economic output and employment figures. The source categories covered in this dataset include industries (which included power generation), domestic fuel burning, and vehicles. Since the lower southern segment of the VTAPA extends into the Free State province, large emitting sources (Eskom Lethabo power station, Sasol and Natref) found in this portion were also included in the modelling exercise. Activity data for these sources were obtained from the University of Cape Town's Energy Research Centre (Energy Research Centre, 2018). Data for non-energy related processes, specifically waste generation, were obtained from the Gauteng waste information system (Gauteng Department of Agriculture and Rural Development, 2018). All the activity data were arranged into the GAINS model data template, uploaded to the GAINS model from which emissions of $PM_{2.5}$, SO_2 and NO_X for the VTAPA were generated. Wind-blown dust and biomass burning emissions were included for this study. However, the GAINS model does not cover these sources, therefore, emission projections for wind-blown dust and biomass burning were carried out exegenously using emissions data obtained for Council for Scientific and Industrial Research (CSIR) (Department of Environmental Affairs, 2019).

6.2.4 Scenario design

A set of scenarios were developed for the VTAPA to explore the potential of reducing the health impacts of air pollutants by altering the energy activity mix and applying abatement measures to different sectors. Emissions reduction pathways for these scenarios were assessed in the medium-term (20-year period), in which 2015 was taken as the reference year (REF). Emissions for the VTAPA were compared with emissions for the whole of South Africa (Rafaj *et al.*, 2018) so as to show the contribution of the VTAPA in relation to the rest of South Africa.

6.2.5 Current Legislation Scenario (CLE)

The CLE scenario is based on energy projections taken from World Energy Outlook for 2017 (International Energy Agency, 2017). This scenario involves the careful enforcement of current pledges and plans, taking into consideration the existing institutional, political and economic barriers that South Africa is facing. It assumes that there will be an adoption of regulations and compliance with emission standards as of 2015. It also takes into account regulation issues such as the postponed compliance to emission standards by some industries, and the delayed implementation of Euro 5 fuel standards in the mobile sector (Henneman *et al.*, 2016). Reductions in this scenario will be significant for SO_2, but comparatively smaller for $PM_{2.5}$ and NO_x; in addition, emission cuts are limited to specific sectors. The emission trajectories under the CLE are also anchored on projected GDP (United States Dollars) and population growth (Figure 6-2).

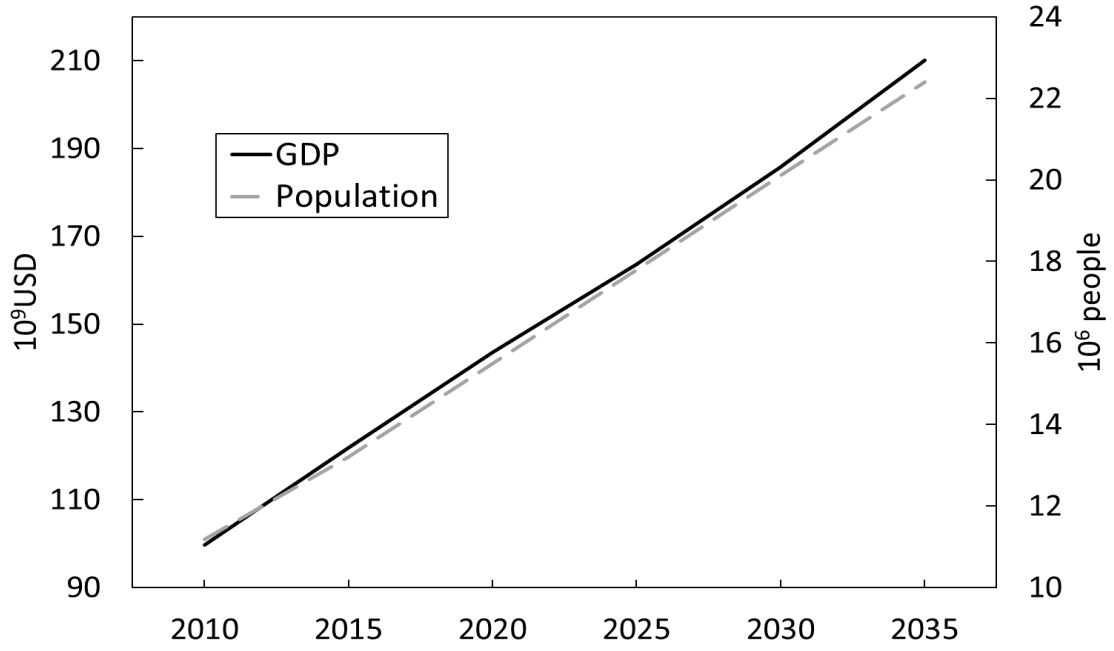

Figure 6-2: GDP and population projections for the Gauteng province.

6.2.6 Alternative Scenario (AS)

The AS shows the extent to which emission levels can be further reduced by altering the fuel mix and applying the best available technologies. This scenario attempts to give a scope of the potential benefits that can be achieved through the full implementation of abatement measures in the VTAPA by 2035. In order to determine the most efficient technologies to use in the AS, the abatement strategies available in the GAINS model for reducing $PM_{2.5}$, SO_2 and NO_x emissions were analysed side by side for each sector. The resultant controls applied in the AS scenario are listed in Table 6-1.

Table 6-1: List of controls applied to different emission sources in the AS scenario.

Emission sources	Pollutant targeted	Control description
Domestic combustion	$PM_{2.5}$	Substituting coal with LPG in households
Waste combustion	$PM_{2.5}$, SO_2, NO_x	Ban on open municipal solid waste (MSW) burning
Industry	$PM_{2.5}$	Installation of fabric filters (high-efficiency dedusters)
Industry	SO_2	Installation of wet flue gas desulphurisation (WFGD) technologies
Industry	NO_x	Installation combustion modification and selective catalytic reduction technologies
Vehicles	$PM_{2.5}$, SO_2, NO_x	Application of EURO 6 standards to petrol and diesel vehicles
Windblown dust	$PM_{2.5}$	Use of dust suppressants

Associated costs for each control strategy are calculated under the notion of a free market, in which abatement technologies are assumed to be available to all countries at the same costs. For this study, implementation costs for abatement technologies were calculated for each scenario in GAINS at a 4% interest rate.

6.2.7 Dispersion modelling

Fine particulate matter concentrations for the GAINS model are usually calculated with a reduced form of the European Monitoring and Evaluation Programme (EMEP) chemistry transport model at resolutions of 50 km x 50 km. However, given the fine spatial scale of the VTAPA, fine pertinent details will be lost in the EMEP coarse resolution pixel matrix. CALPUFF is a non-steady-state Lagrangian Gaussian puff model that can simulate the dispersion of pollutants over a large domain (>50 km) at fine spatial resolutions (1 km x 1 km) (Scire *et al.,* 2000; Rood, 2014). A

gridded (1 km x 1 km) emission inventory for the VTAPA was obtained from the CSIR for South Africa (Department of Environmental Affairs, 2019) (Annexure II). These gridded emissions were used to quantify the changes in $PM_{2.5}$ (primary and secondary) concentrations for key sectors based on the different set of control strategies.

6.2.8 Health effects

Health effect assessments for air quality management involve quantifying the fraction of the affected population attributing to fine-PM concentration levels in a region. This can help in determining the potential number of deaths that could be prevented if PM exposure were reduced to a sufficient level. The methodology that was used to estimate the current and future mortality attributed to ambient PM pollution is based on the Global Burden of Disease studies (Lim *et al.*, 2012). This approach involves combining the population exposure distribution to ambient $PM_{2.5}$ with the relative risk at each level of exposure so as to obtain the population attributable fraction (Rao *et al.*, 2012). The disease burden attributed to ambient $PM_{2.5}$ (AB) is then determined by multiplying the population attributable fraction (PAF) with the health endpoint. For this study, the health endpoint of interest is total mortality.

$$\boldsymbol{PAF = [P * (RR - 1)]/[P * (RR - 1) + 1]}$$

where *P* = proportion of the population exposed to a given level of $PM_{2.5}$; and *RR* = Relative risk associated with exposure to ambient air pollution.

$$\boldsymbol{AB = PAF * health\ endpoint}$$

Total population exposures to ambient $PM_{2.5}$ for the VTAPA were calculated as fractions of the projected population data from the GAINS database for South Africa. A theoretical minimum risk exposure value of 10 µg/m^3 was used for this analysis as it is the World Health Organisation's recommended air quality limit for annual $PM_{2.5}$ (WHO, 2006). Census data was used to extract the specific death rate for the VTAPA which was then used to estimate the baseline mortality for current and future scenarios.

6.3 Results

6.3.1 Emission comparisons

Figure 6-3 shows emissions comparisons between the VTAPA and the whole of South Africa. $PM_{2.5}$, SO_2 and NO_x pollutants in the VTAPA contribute 4, 8, and 9% of the total emissions in South Africa respectively.

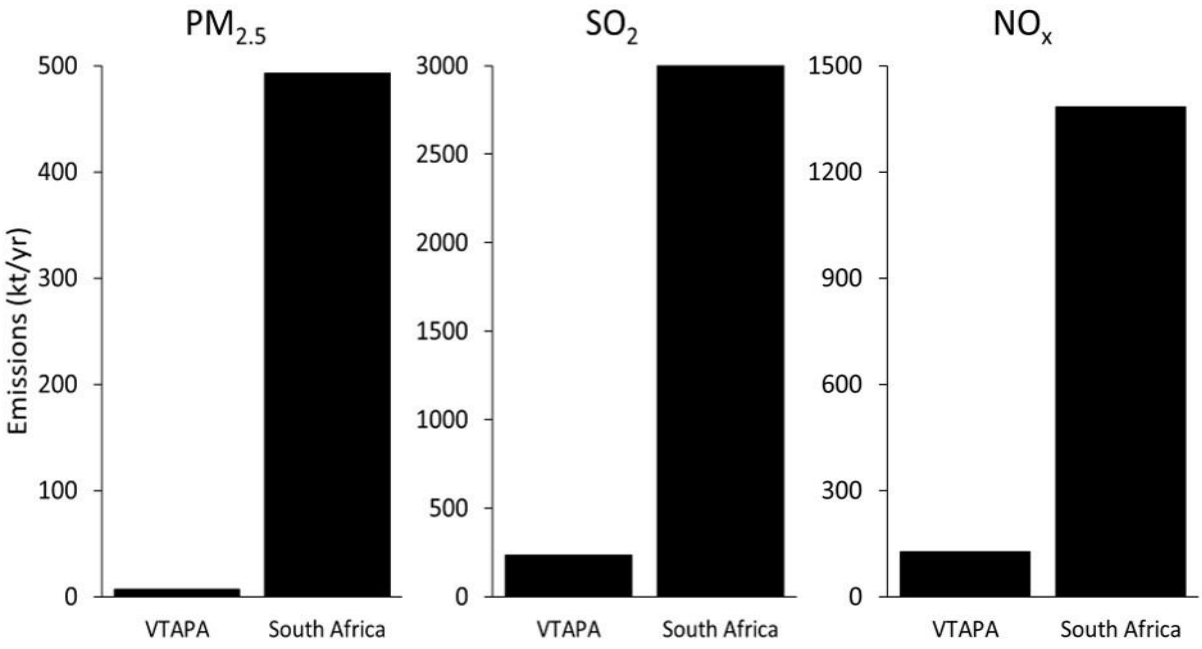

Figure 6-3: Emission comparisons by pollutant between VTAPA and South Africa

Emission contributions for $PM_{2.5}$ vary between the VTAPA and South Africa (Figure 6-4). Industries are the dominant contributors for the VTAPA, while for the whole of South Africa, domestic combustion is the main source of $PM_{2.5}$ emissions. Despite these differences in relative source contributions, industries remain an important source of $PM_{2.5}$ for the whole of South Africa. Windblown dust from mine tailings, stockpiles and ash storage facilities is an important source category for the VTAPA. Industries are the dominant SO_2 sources for both the VTAPA and South Africa. For NO_x emissions, industries and vehicles are the most important sources in the VTAPA and the whole of South Africa as well. Given these variations in emission source contributions between the VTAPA and South Africa, local intervention strategies are likely to be more effective for the city level compared to regional or national policies.

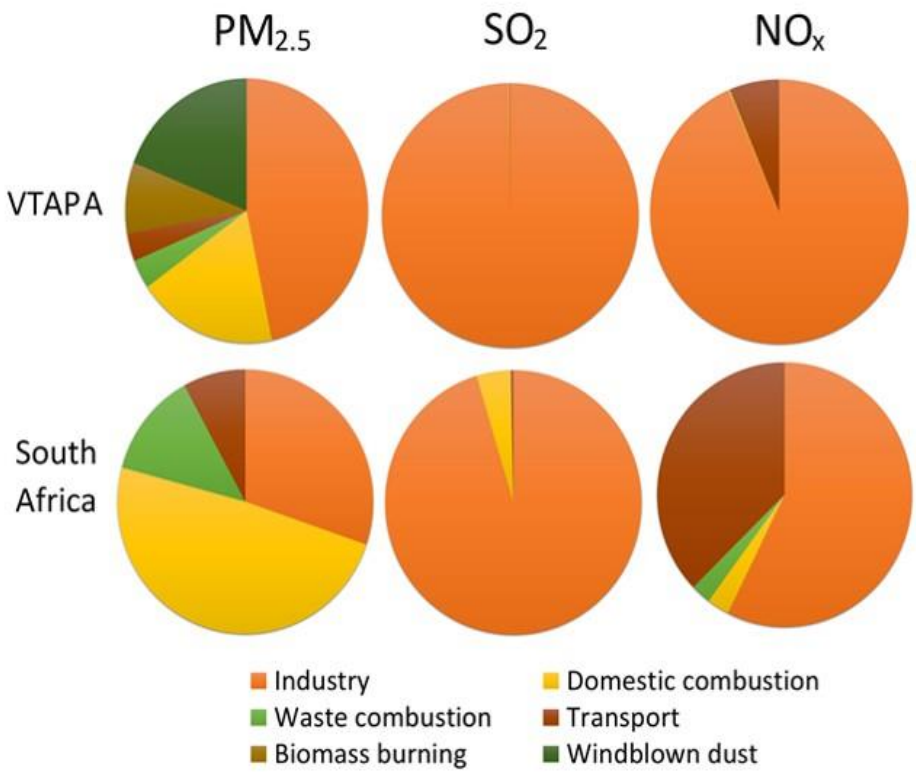

Figure 6-4: Comparisons of emission source contributions by pollutant between VTAPA and South Africa.

6.3.2 Sectoral reductions in the VTAPA

Figure 6-5 shows the changes in $PM_{2.5}$ emissions in the CLE and AS scenarios by sector relative to the year 2015. Under current legislation, emissions stay essentially constant (4% decrease by 2035), while in the AS they are projected to decrease significantly in 2035, with $PM_{2.5}$ dropping by 66%. Under the existing legislation, $PM_{2.5}$ emissions in the domestic sector will experience a steady decline as a result of the increased electrification of households in low-income settlements. However, it should be noted that electrification will not necessarily result in the total disuse of solid fuels in low-income settlements. Due to economic reasons and population growth, an increase in electricity tariffs could discourage a complete switch from solid fuels to electricity.

Replacing coal with Liquefied Petroleum Gas (LPG) in low-income settlements is an option that will complement the electrification programme and could result in the elimination of $PM_{2.5}$ emissions from the domestic sector by 2035. However, for this strategy to be effective, there is a need for a consistent supply of LPG and adherence to government price cap regulations of this fuel. Without such measures in place, residents in low-income settlements will continue to use solid fuels as LPG will be costly.

Industrial emissions in the CLE scenario are predicted to gradually increase due to growth in industries. However, if fabric filters are installed in all industries, a 70% drop in $PM_{2.5}$ emissions could take place by 2035. Waste generation in urban areas is expected to increase in the VTAPA due to the ever-rising population in the region. This can lead to an increase in $PM_{2.5}$ emissions for the waste sector. Enforcing a ban on the burning of municipal solid waste (MSW) could potentially reduce emissions in this sector by 83%. It should be noted that for this strategy to be effective, the ban on MSW burning must be accompanied by regular monitoring, efficient refuse collection and changes in social perceptions.

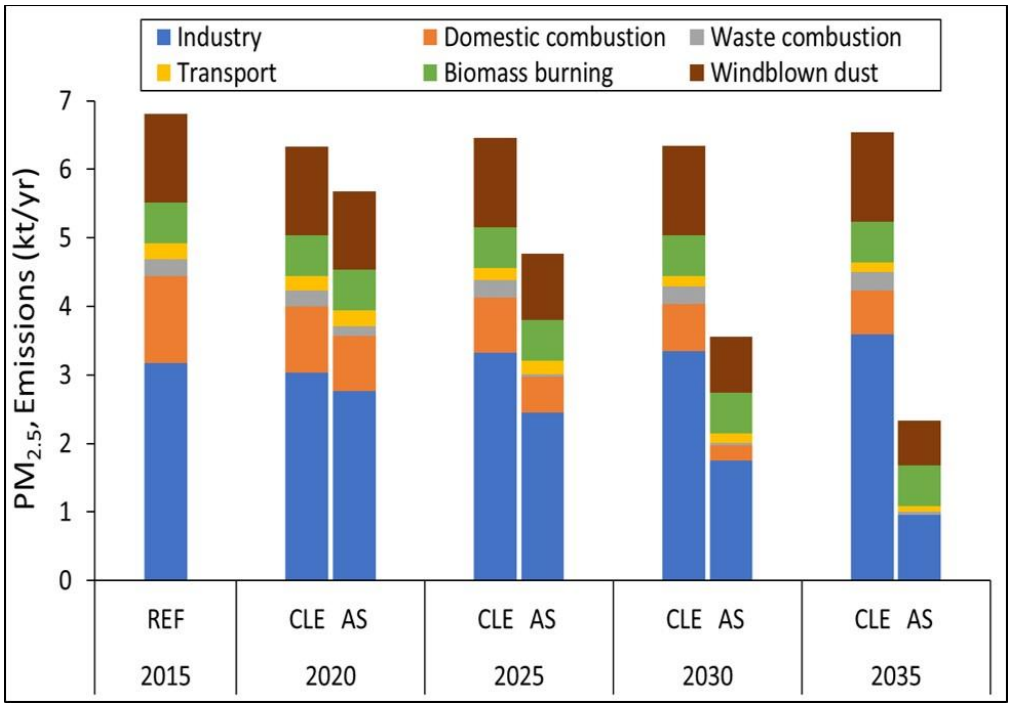

Figure 6-5: $PM_{2.5}$ emissions by sector in the VTAPA, for current legislation (CLE) and alternative (AS) scenarios.

Figure 6-6 shows the impacts of sectoral targeted abatement measures on NO_X emissions in the VTAPA. The industrial sector is the chief NO_X contributor, accounting for over 90% of the total emissions. In the AS scenario, combustion modification and installation of selective catalytic reduction technologies to all industries, as well as the switch to Euro 6 fuel and engine standards in the transport sector will yield a reduction in NO_X emissions by 57%, with the majority of the reduction taking place in the industrial sector. However, in the CLE scenario, emissions for NO_X are projected to gradually rise by 10% due to industrial growth and slow implementation of the Euro 5 emission standard in the transport sector.

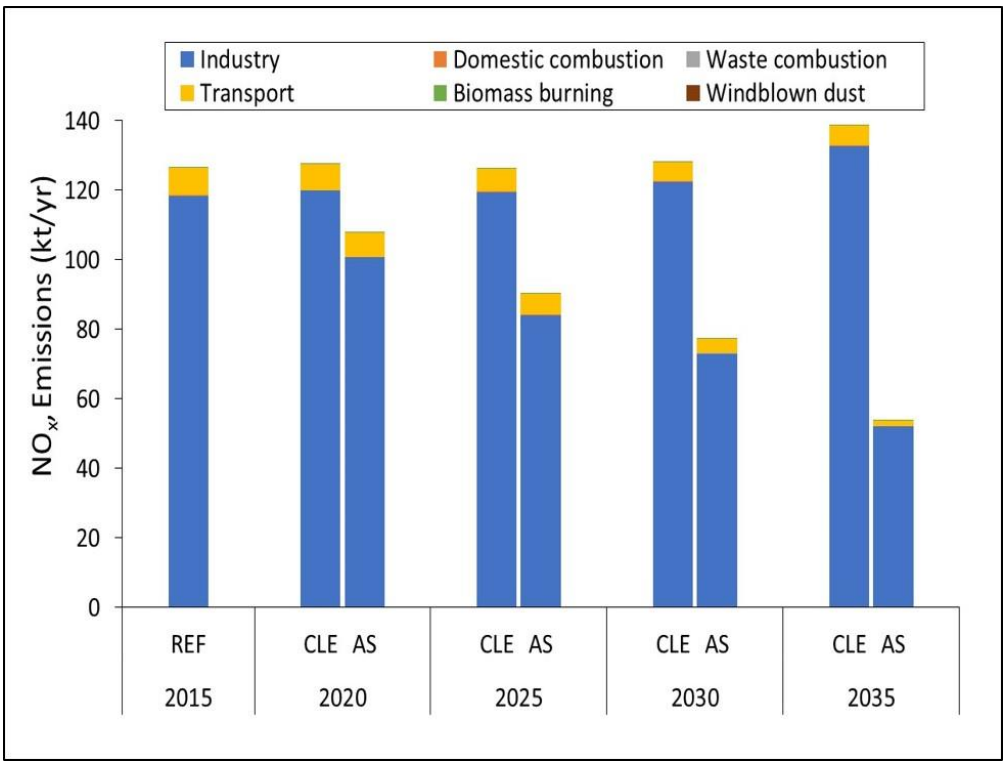

Figure 6-6: NO_x emissions by sector in the VTAPA, for current legislation (CLE) and alternative (AS) scenarios.

SO_2 emissions in the CLE scenario (Figure 6-7) are expected to decrease, considerably dropping by 44% from 2015 to 2035. The high SO_2 emissions reduction results from the application of stringent controls (mainly dry flue gas desulphurisation technologies) in the industrial sector. Emission reduction potential in the AS scenario is greater than in the CLE, as SO_2 emissions are expected to drop sharply by 68% in the VTAPA. This as a result of the adoption of wet flue gas desulphurisation technologies in the industrial sector.

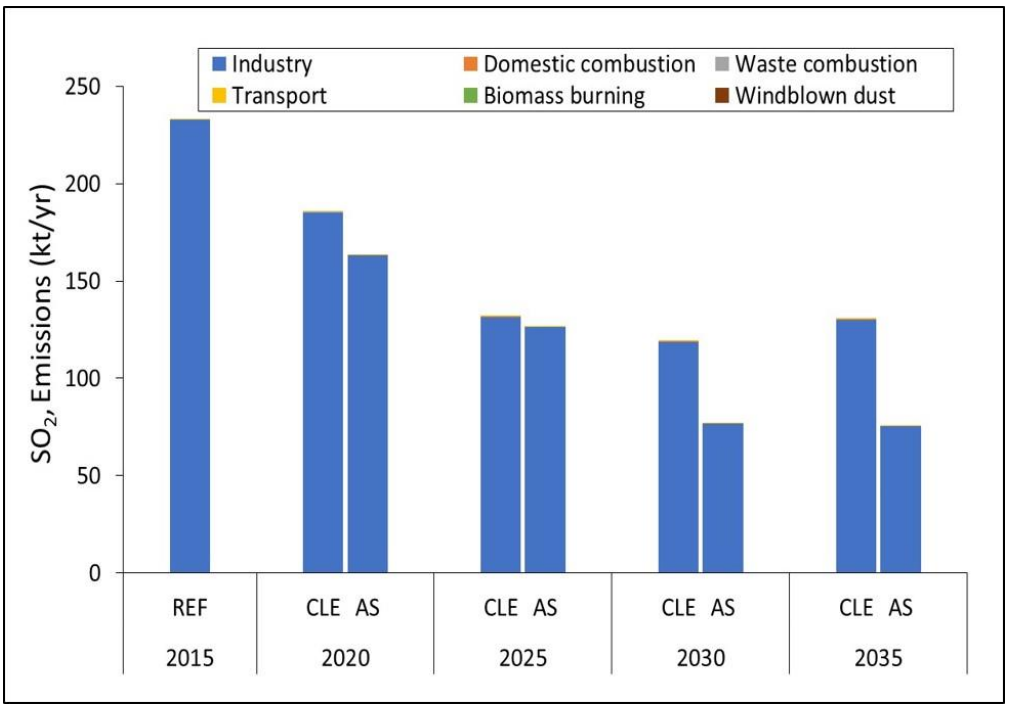

Figure 6-7: SO_2 emissions by sector in the VTAPA, for current legislation (CLE) and alternative (AS) scenarios.

6.3.3 Spatial reductions in the VTAPA

Figure 6-8 presents the spatial distribution of annual $PM_{2.5}$, NO_x and SO_2 emissions for the baseline year and the future year for the CLE and AS scenarios as used in the CALPUFF model simulations. High levels of $PM_{2.5}$ emissions are concentrated in the central part of the VTAPA due to industrial (mineral processing, storage and handling) and coal mining operations. A substantial proportion of $PM_{2.5}$ emissions also occur in the northern regions, with domestic combustion from the low-income settlements of Soweto and windblown dust from gold mining tailings storage facilities, accounting for these high emission intensities. In the CLE scenario, $PM_{2.5}$ emissions in the highest emitting grid cell are expected to drop slightly from 837 to 804 tons/year, and drastically fall to 367 tons/year in the AS. As shown in Figure 6-8, high emission intensities for NO_x and SO_2 are experienced in the central and south-western zones of the VTAPA due to power generating sources (Eskom's Lethabo power plant and Sasol's Sasolburg operations). NO_x emissions in the highest emitting grid cell for the CLE are expected to rise from 33 614 to 36 854 tons/year. Conversely, in the AS scenario, NO_x emissions in the highest emitting grid cell will significantly fall to 14 252 tons/year. SO_2 baseline emissions in the highest emitting grid cell amounting to 78 745 tons/year are expected to drop considerably in the CLE and AS to 44 047 and 25 501 tons/year respectively.

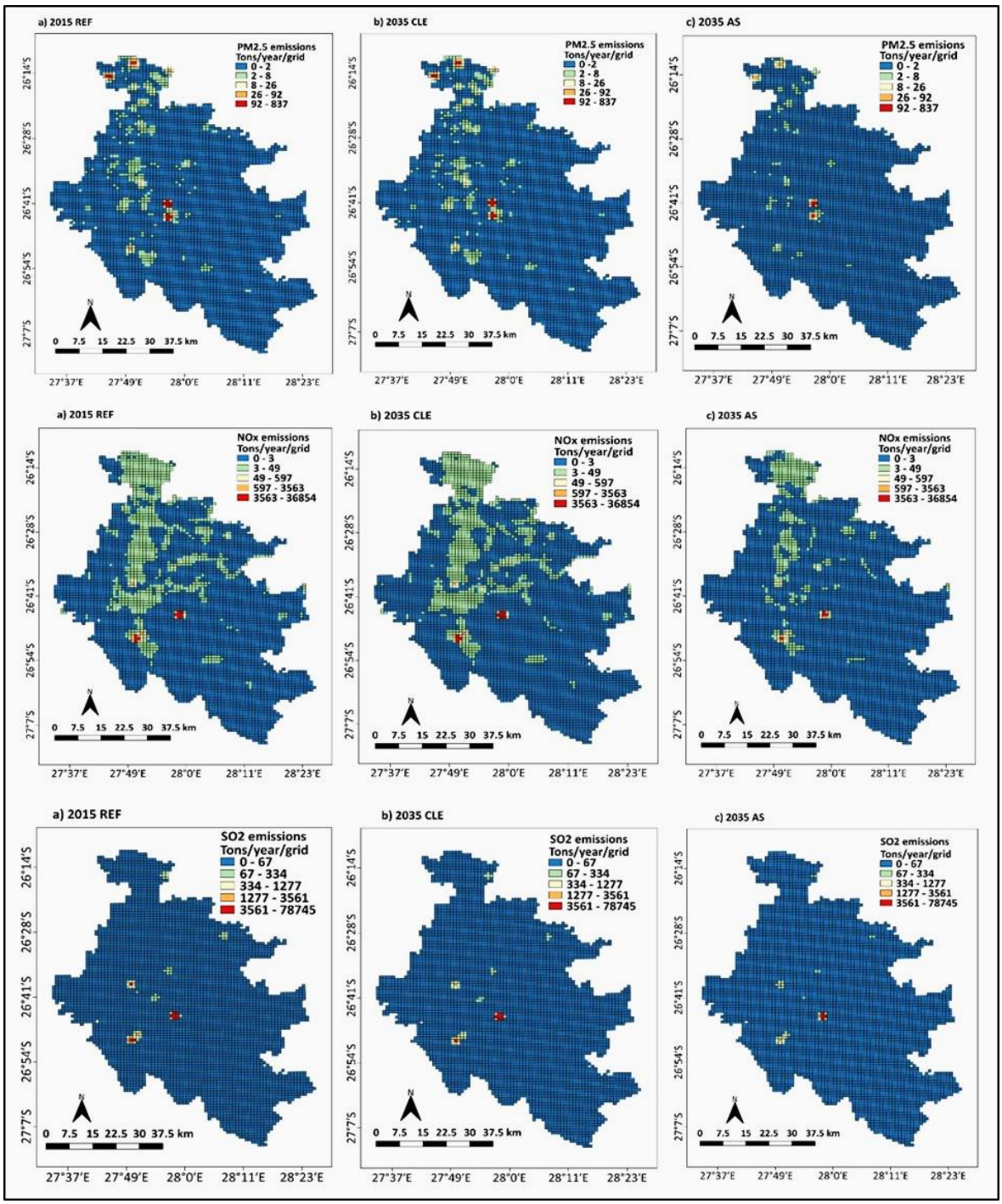

Figure 6-8: Spatial emission distribution for $PM_{2.5}$, NO_x and SO_2 (rows) in the VTAPA, in the different scenarios (columns).

6.3.4 Emission control costs

Implementation costs of controls applied for each scenario were estimated and compared against the projected GDP for the Gauteng province. This was done for technologies in the transport and industrial sector. The GAINS model does not calculate implementation costs for switching from Coal to LPG in the domestic sector and the use of dust suppressants. As expected, control costs

in the AS scenario are higher by almost a factor of 1.6 (Figure 6-9) in comparison to the CLE scenario, and these costs will total 0.6% of GDP by 2030. For the CLE, control costs are estimated at 0.4% of GDP by 2030.

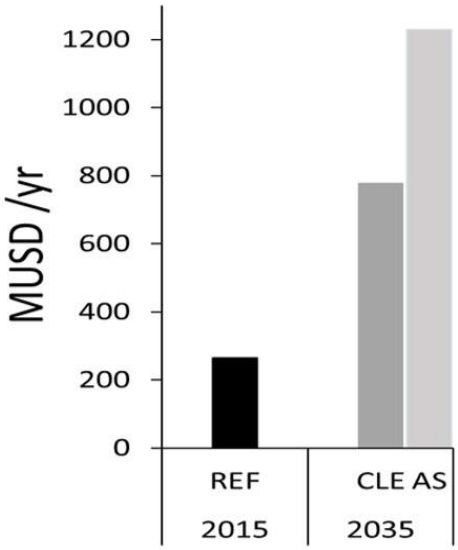

Figure 6-9: Estimated implementation costs for controls in the VTAPA under the CLE and AS scenarios.

6.3.5 $PM_{2.5}$ concentrations in the VTAPA

Table 6-2 illustrates the changes in annual mean and maximum $PM_{2.5}$ concentrations for the different modelled emission sources. Our results suggest that emphasis should not be solely placed on controlling industrial emissions even though it constitutes a major portion of the overall $PM_{2.5}$ concentrations. It can be observed that domestic combustion (32%) has similar contributions to that from industries (34%). Biomass burning and vehicles also have significant contributions. Under the CLE scenario, by the year 2035, annual mean $PM_{2.5}$ concentrations for the industrial and waste sectors are expected to increase, by 1.5 and by 0.3 µg/m^3, respectively. The greatest reductions in the AS scenario are observed in the domestic and industrial sectors.

Table 6-2: Projected changes in annual mean and maximum $PM_{2.5}$ concentrations (µg/m^3) by sector in the VTAPA under the CLE and AS scenario.

Sector	REF		CLE (2035)		AS (2035)	
	Average	Maximum	Average	Maximum	Average	Maximum
Biomass burning	4.6	7.8	4.6	7.8	4.6	7.8
Domestic combustion	10.7	23.4	5.4	11.8	0	0
Industry	11.1	14	12.6	15.8	3.4	4.2

Sector	REF		CLE (2035)		AS (2035)	
	Average	Maximum	Average	Maximum	Average	Maximum
Vehicles	3.5	7.8	2.1	4.6	1.3	2.9
Windblown dust	1	3.6	1	3.6	0.5	1.8
Waste combustion	2.2	5.2	2.5	5.9	0.4	0.9

The overall annual mean $PM_{2.5}$ concentrations from all the emission sectors are shown in Figure 6-10. It can be observed that $PM_{2.5}$ concentrations for the VTAPA (for the year 2015) are not only above national standards (20 µg/m^3), but they are also well above WHO guideline values (10 µg/m^3). Large parts of the VTAPA will experience concentrations ranging from 30 to 50 µg/m^3. The highest $PM_{2.5}$ concentrations are observed in the northern part of the VTAPA where domestic burning is a major emission source. High levels of $PM_{2.5}$ are also in the central to south-west region of the VTAPA where industrial activities, domestic burning and mining operations take place. Under the CLE scenario, $PM_{2.5}$ concentrations are expected to remain above acceptable air quality limits throughout the VTAPA, experiencing only a marginal decrease. More favourable reductions are experienced in the AS scenario. $PM_{2.5}$ concentrations are expected to reduce significantly by 69%. This is mainly due to reductions in domestic burning emissions which reduces 32% of $PM_{2.5}$ concentrations in the VTAPA. Although under the AS scenario, national limits are met throughout the entire region, there are some parts in the northern section of the VTAPA where concentrations that will still remain above WHO guideline values.

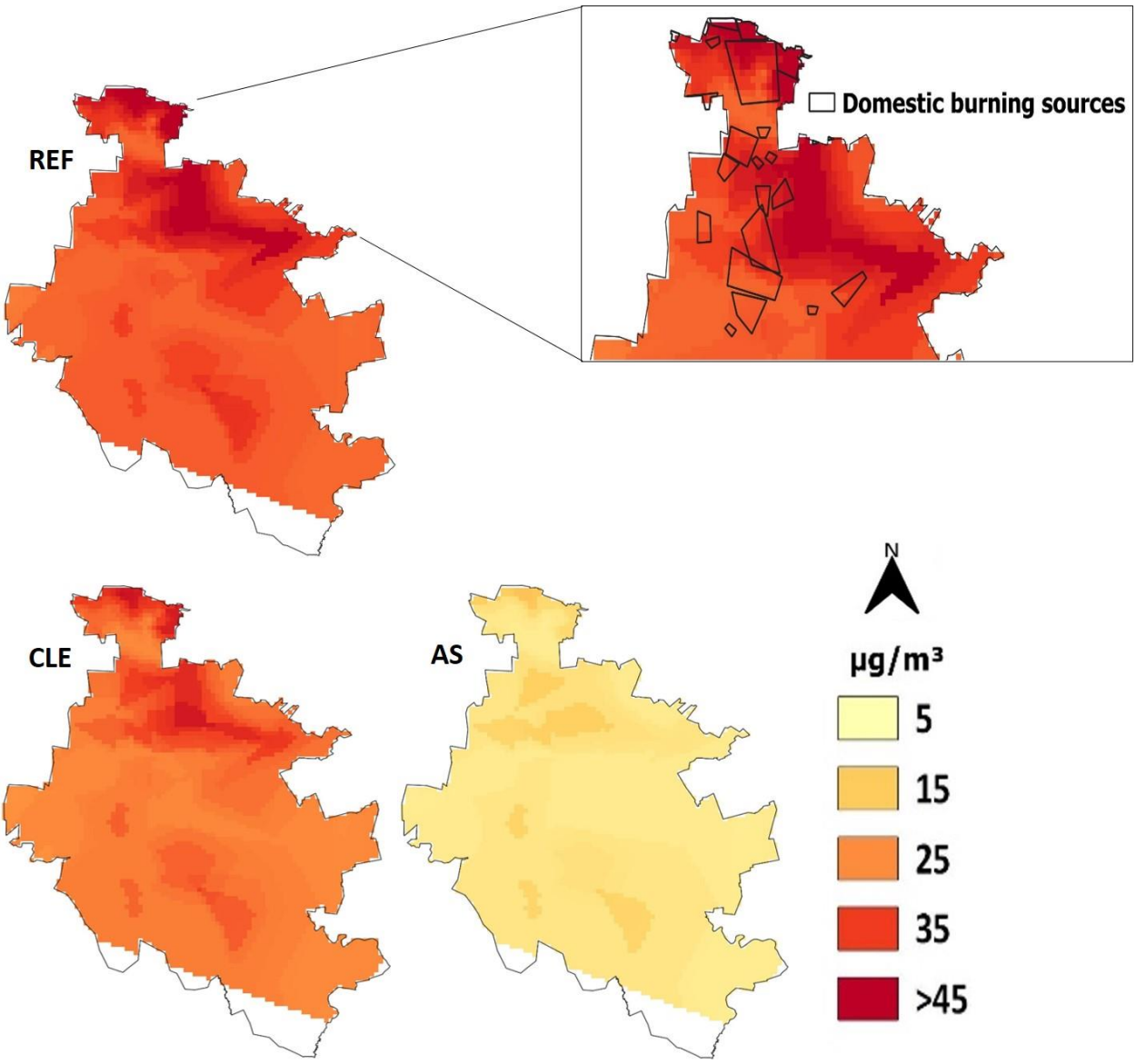

Figure 6-10: Estimated annual $PM_{2.5}$ concentrations across the VTAPA for the year 2015 (left) and for the year 2035 in the CLE (centre) and AS (right) scenario. Zoomed area shows $PM_{2.5}$ concentrations at domestic burning sources.

6.3.6 Effects on human health

Table 6-3 shows health effects findings in terms of the number of deaths attributable to ambient $PM_{2.5}$ pollution in the VTAPA. If the current policies are applied, a minor decrease (8.5%) in the number of deaths will be experienced by 2035. These small health gains are due to the increase in $PM_{2.5}$ concentrations from the industrial and waste sector resulting from the growth of industries and the population. However, in the AS scenario, significant health gains are experienced in which deaths are reduced by over 95.5%, relative to the minimal change in the CLE scenario. This is mainly attributed to the elimination of emissions from domestic fuel burning. By reducing

emissions from a low-level source, exposure to $PM_{2.5}$ is significantly reduced. Stringent measures will, therefore, need to be applied in the VTAPA to avoid future negative health impacts.

Table 6-3: Estimated premature deaths in population >30 years attributed to ambient $PM_{2.5}$ pollution in the VTAPA under the CLE and AS scenario.

Scenario (year)	Central estimate (RR =1.06) with upper and lower 95% confidence intervals	Deaths per 100 000 of population attributable to PM	% reduction relative to REF scenario
REF (2015)	4 700 (1 700 – 7 800)	41	–
CLE (2035)	4 300 (1 500 – 7 300)	–	8.5
AS (2035)	210 (70 – 390)	–	95.5

6.4 Discussion

South Africa's attempt to improve the livelihoods of its inhabitants has resulted in economic advancement taking priority over environmental issues. Poor air quality is a major health concern in the cities of South Africa. Emissions from industries, coal and wood burning in residential areas, open waste and biomass burning have contributed to high concentrations of $PM_{2.5}$, NO_x and SO_2 that are well above national air quality limits (Scorgie, 2012). With the current rate of urbanisation, population growth and economic development taking place in the cities and without stringent control policies, the quality of air is likely to further deteriorate leading to the future worsening of the health burden.

This study is an initial attempt to identify suitable policy interventions that could effectively reduce the emissions of pollutants in the VTAPA. By comparing the effectiveness of different air quality management approaches, it can be observed that air pollution control under the existing legislation is not sufficient to achieve compliance with national air quality standards. Our results suggest that given the increasing energy demand if the proposed controls in the current policies are to be fully implemented there will be a minor decrease in $PM_{2.5}$ and NO_x emissions. However, for SO_2, emissions are expected to be halved by the year 2035, due to stringent controls in the centralised industrial/power sector.

A variety of strategies are available to curb the negative effects associated with the rate of urban and economic development in the VTAPA. In the AS scenario, strict implementation of control measures such as the adoption of advanced control technologies, a switch from 'dirty fuels' to less polluting fuels and the banning of domestic waste burning in open spaces can effectively

lower $PM_{2.5}$ and $PM_{2.5}$ precursors emissions by more than half. The biggest reductions are expected to be experienced in the domestic sector through a switch from coal to LPG. This will have a considerable effect on reducing $PM_{2.5}$ exposures in low-income settlements. Although implementation costs for such fuel switches in domestic combustion are not calculated in GAINS, a study for the VTAPA done by the Department of Environmental Affairs for South Africa shows that substantial financial resources are required to fully implement a switch from coal to LPG use (Department of Environmental Affairs, 2018). Additionally, the supply of LPG is inconsistent and government price cap regulations are not strictly adhered to. Industries will also experience substantial reductions but the technologies needed to combat air pollution in the VTAPA are costly. However, despite these challenges, the environmental and health benefits to society will greatly outweigh these costs, as seen by the potential reduction in the number of deaths due to $PM_{2.5}$ exposures. A total of 4490 premature deaths attributable to ambient $PM_{2.5}$ exposure can be avoided by 2035.

6.5 Conclusion

The current research for the VTAPA demonstrates the value of integrated assessment modelling as an evidence-based scientific tool for supporting decision making through the analysis of climate change, energy, air pollution, health and economics as a complex system. Although the modelling exercise has illustrated how stringent controls can reduce emissions and concentrations, and can improve human health, the adoption of these measures still requires considerable financial resources. In order to fully exploit the health benefits of emission reductions without compromising the economy, careful selection of policy interventions - also including those aiming at clean energy access, waste management, land-use practices and greenhouse gases mitigation - will need to be considered. This study has shown that future emissions and the costs associated with different management strategies can and need to be assessed at a local scale for South Africa.

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Chapter conclusion

The objective of this study is to assess the potential impacts of emission reduction strategies on air quality in the Vaal Triangle Airshed Priority Area

The above study drew the following main conclusions:

- Under the current management strategy for the VTAPA, Air quality targets for $PM_{2.5}$ will not be attained by the year 2035
- By introducing stringent controls into the VTAPA, $PM_{2.5}$ concentrations will drop to levels within the regulatory limits by the year 2035. However, the costs to implement these controls will cost nearly twice as much as compared to the current control strategies
- By eliminating emissions from domestic burning, $PM_{2.5}$ concentrations and premature deaths are significantly reduced in the VTAPA.

CHAPTER 7 CONCLUSIONS

Air quality in the VTAPA has attracted public attention due to its elevated pollutant concentrations, specifically PM levels which are above national health limits. Rapid urbanisation, coupled with industrial expansion, increase in vehicle numbers and continued use of coal and wood as a domestic energy source has contributed to the deterioration of air quality in this region. Air pollution control initiatives in the VTAPA have not produced the desired outcomes. PM concentrations are still alarmingly high and further emission reductions are required to minimise the health impacts of air pollution in this region. Air quality management is multidisciplinary in nature. Collective analysis of air quality, health and cost-benefits aspects is therefore required. For this reason, an integrated approach to air quality management was performed for this study in order to produce the relevant information that will support decision-making for the VTAPA. This was achieved by firstly examining the potential of satellite retrievals in assessing the spatial variations and exploring temporal trends of $PM_{2.5}$ concentrations for the VTAPA. It is necessary to examine past concentration trends so as to predict future outcomes. Next, a source apportionment for the VTAPA was carried out to determine the main sources of PM and their contributions. Lastly, a proposed management strategy for the VTAPA was evaluated by comparing its emission reductions, induced air quality and health impacts relative to projections from the current management strategy. The major findings for each objective and corresponding manuscript will be discussed in the following sections.

7.1 Research objective 1

The objective of the first manuscript, entitled ***"Evaluating the potential of remote sensing imagery in mapping ground-level fine particulate matter ($PM_{2.5}$) for the Vaal Triangle Airshed Priority Area"***, was to explore the potential use of satellite retrievals in assessing the spatiotemporal trends of $PM_{2.5}$ over the VTAPA.

The main findings for this research objective are summarised as follows:

- For remote sensing to become a viable air quality monitoring option, the precision and accuracy of satellite retrievals for $PM_{2.5}$ need to be improved in order to prevent the overestimation of $PM_{2.5}$ concentrations. This can be achieved by:
    - Optimising the positioning of ground-based stations so as to have monitoring data that is spatially more representative
    - Collocating $PM_{2.5}$ monitoring instruments with ground-based sun photometers to assess the AOD-$PM_{2.5}$ relationships independently. This can assist in improving $PM_{2.5}$ estimates from satellite AOD retrievals

- Monthly satellite retrievals for $PM_{2.5}$ concentrations need to be developed so that seasonal variations and trends can be captured.

The main conclusions from this study are that satellite remote sensing is currently not a practical and credible option to ground-based monitoring in the VTAPA. Based on ground measurements, $PM_{2.5}$ concentrations in the VTAPA have not significantly reduced since the inception of the VTAPA AQMP and still remain above national and global ambient air standards. With the current management strategies, $PM_{2.5}$ concentrations are likely to remain above air quality limits.

7.2 Research objective 2

The objective of the second manuscript titled ***"Updated $PM_{10-2.5}$ and $PM_{2.5}$ source apportionment for the Vaal Triangle Airshed Priority Area, South Africa"***, was to identify the main sources contributing to PM loading in the VTAPA through a receptor model approach. This analysis was done for both $PM_{10-2.5}$ and $PM_{2.5}$.

The major findings from this investigation are summarised below:

- $PM_{2.5}$ concentrations both during the day and night for all sites and seasons generally exceeded the NAAQS 24-hour for $PM_{2.5}$
- The highest seasonal concentrations for both $PM_{10-2.5}$ and $PM_{2.5}$ were experienced in winter
- Dust-related elements which include Si, Mg, Al, Ca, Na, S and Fe, contributed the most towards $PM_{10-2.5}$, both during the day and the night for all seasons, whilst for $PM_{2.5}$, S, K, Fe, Cr and Ni have significant contributions
- SO_4^{2-}, NH_4^+ and F^- are the dominant ionic species for both $PM_{10-2.5}$ and $PM_{2.5}$ for all seasons
- Dust-related and secondary aerosols were the main contributing sources for $PM_{10-2.5}$, whilst coal burning and secondary aerosols were significant $PM_{2.5}$ contributors
- Wood and biomass burning is an important source for both $PM_{10-2.5}$ and $PM_{2.5}$, especially during the spring season.

The main conclusions from this study are that although being an industrialised region, low-level emission sources have a significant impact in low-income settlements as observed by the dominance of coal burning in Sebokeng and Sharpeville. Dust, industry, domestic coal burning, wood and biomass combustion are the key sources in the VTAPA that need to be prioritised by

decision-makers. Source contributions differ across low-income settlements. Therefore, individual plans should be developed for each settlement.

7.3 Research objective 3

Careful selection of control strategies for the VTAPA through the assessment and comparison of different mitigation options requires the support of an integrated modelling system. The third objective of this study is addressed in the manuscript titled *"**Integrated assessment of strategies to reduce air pollution in the Vaal Triangle Airshed Priority Area, South Africa**"*, where an integrated assessment model was used to determine cost-efficient options to reduce emissions and achieve compliance to air quality standards. This model was also used to evaluate the effectiveness of current air quality policies for the VTAPA.

The main findings for this research objective are summarised as follows:

- Under the current management strategy for the VTAPA, $PM_{2.5}$ concentrations will not reach recommended air quality limits by 2035
- An alternative management strategy in which more stringent controls are applied will significantly reduce emissions by more than half and annual NAAQS for $PM_{2.5}$ will be attained
- The implementation costs for the alternative management strategy are nearly twice as much as those for the current policies
- Although the operational costs for the alternative management strategy are higher compared to current policies, the negative health impacts are significantly reduced.

The main conclusions drawn from this study are that in order for $PM_{2.5}$ concentrations to be reduced to within national air quality limits by 2035, emission reductions should not be solely from the industrial sector, but also from low emission sources such as domestic fuel burning, vehicles and biomass burning. Integrated assessment modelling through assessing the multiple dimensions of air pollution is an important evidence-based scientific tool for supporting decision making.

7.4 Contribution to the current body of knowledge

The traditional approaches to policy analysis have not offered enough leverage in dealing with the complex problem of air pollution. More needs to be done in order to effectively reduce air pollution levels to within nationally accepted standards. This study makes an important contribution to the current body of knowledge by integrating the technical, environmental, economic and health aspects of air pollution. The findings from this study will assist policy

planners in designing management plans that prioritise key sources in the VTAPA. Through an integrated approach, policymakers will be able to make more informed decisions when it comes to selecting control strategies for improving air quality in the VTAPA without compromising the economy. Future studies should investigate climate change policies in order to prioritise air pollution reduction measures that have the co-benefit of reducing greenhouse gas emissions

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ANNEXURE I: STATISTICAL SUMMARY OF ELEMENTAL AND IONIC CONCENTRATIONS

Supplementary table 1: Day time mean $PM_{10-2.5}$ elemental and ionic concentrations and standard deviation at all sampling sites during the summer period

Site	Kliprivier		Sebokeng		Sharpeville		Zamdela	
	Mean	±SD	Mean	±SD	Mean	±SD	Mean	±SD
Na	1.59	1.22	1.89	1.09	2.14	1.69	1.95	1.51
Mg	0.76	0.54	0.82	0.39	1.11	0.73	0.90	0.62
Al	1.55	1.25	2.17	0.99	1.86	1.04	1.72	1.02
Si	5.24	3.92	6.57	2.80	5.81	3.15	5.57	3.13
P	0.03	0.02	0.04	0.02	0.04	0.02	0.04	0.02
S	1.58	1.23	1.95	1.13	2.14	1.28	2.03	1.47
Cl	0.07	0.05	0.11	0.07	0.37	0.44	0.11	0.11
K	0.43	0.26	0.54	0.21	0.52	0.28	0.45	0.22
Ca	0.99	0.66	1.12	0.39	1.75	1.14	1.33	0.70
Ti	0.06	0.05	0.09	0.04	0.08	0.05	0.08	0.04
V	0.00	0.00	0.00	0.01	0.00	0.01	0.00	0.00
Cr	0.32	1.24	0.25	0.92	0.31	1.20	0.32	1.24
Mn	0.05	0.04	0.07	0.08	0.07	0.04	0.03	0.03
Fe	1.80	3.45	2.07	2.84	2.14	3.29	1.67	3.54
Ni	0.09	0.34	0.08	0.30	0.08	0.32	0.09	0.35
Cu	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01
Zn	0.10	0.08	0.10	0.08	0.24	0.29	0.08	0.08
Pb	0.01	0.02	0.02	0.03	0.01	0.02	0.02	0.02
F^-	1.27	0.87	1.08	0.40	1.24	0.67	1.59	0.93
Cl^-	0.05	0.16	0.01	0.01	0.04	0.05	0.01	0.01
SO_4^{2-}	4.50	3.76	5.74	3.55	5.75	3.51	4.37	3.90
NO_3^-	0.04	0.09	0.09	0.14	0.16	0.22	0.86	1.95
$CH3COO^-$	0.01	0.03	0.00	0.00	0.00	0.00	0.00	0.00
$HCOO^-$	0.02	0.05	0.01	0.01	0.02	0.01	0.01	0.01
$C_2O_4^{2-}$	0.16	0.08	0.14	0.11	0.16	0.08	0.12	0.08
Na^+	0.14	0.15	0.16	0.11	0.17	0.14	0.13	0.13
NH_4^+	1.34	1.18	1.88	1.28	1.83	1.07	1.45	1.30
K^+	0.09	0.05	0.11	0.05	0.13	0.08	0.08	0.06
Mg_2^+	0.10	0.15	0.09	0.04	0.12	0.07	0.07	0.06
Ca_2^+	0.52	0.36	0.57	0.23	0.79	0.53	0.54	0.41
PM	36.15	23.6	32.22	21.3	42.89	23.29	39.85	18.84

Supplementary table 2: Day time mean $PM_{2.5}$ elemental and ionic concentrations and standard deviation at all sampling sites during the summer period

Site	Kliprivier		Sebokeng		Sharpeville		Zamdela	
	Mean	±SD	Mean	±SD	Mean	±SD	Mean	±SD
Na	0.15	0.20	0.17	0.29	0.16	0.31	0.06	0.19
Mg	0.00	0.00	0.00	0.00	0.00	0.00	0.02	0.09
Al	0.18	0.17	0.38	1.19	0.07	0.09	0.67	2.34
Si	0.77	0.54	0.60	0.55	0.27	0.25	1.77	5.12
P	0.01	0.01	0.01	0.02	0.01	0.01	0.01	0.03
S	1.66	1.17	1.54	1.07	1.83	1.08	1.88	1.20
Cl	0.09	0.11	0.07	0.07	0.09	0.12	0.09	0.10
K	0.22	0.12	0.21	0.14	0.18	0.12	0.19	0.44
Ca	0.28	0.32	0.13	0.20	0.15	0.48	0.31	0.55
Ti	0.03	0.05	0.03	0.04	0.04	0.04	0.05	0.10
V	0.01	0.04	0.03	0.05	0.02	0.05	0.04	0.06
Cr	0.03	0.14	0.05	0.15	0.13	0.35	0.08	0.24
Mn	0.02	0.05	0.03	0.04	0.03	0.04	0.03	0.05
Fe	0.09	0.36	0.10	0.38	0.37	1.13	0.52	1.22
Ni	0.02	0.05	0.00	0.02	0.01	0.05	0.03	0.09
Cu	0.05	0.12	0.06	0.10	0.05	0.08	0.07	0.11
Zn	0.22	0.26	0.24	0.27	0.11	0.16	0.31	0.28
Pb	0.13	0.24	0.11	0.18	0.03	0.08	0.08	0.13
F^-	0.22	0.64	0.76	2.10	0.51	0.74	2.36	3.72
Cl^-	0.03	0.05	0.00	0.01	0.02	0.02	0.03	0.04
SO_4^{2-}	3.80	2.95	3.71	2.95	4.01	2.67	10.14	23.13
NO_3^-	0.00	0.00	0.71	2.83	0.00	0.00	0.39	0.81
$CH3COO^-$	0.00	0.00	0.00	0.00	0.07	0.05	0.01	0.03
$HCOO^-$	0.00	0.00	0.00	0.00	0.01	0.01	0.01	0.02
$C_2O_4^{2-}$	0.12	0.08	0.09	0.11	0.09	0.06	0.20	0.32
Na^+	0.04	0.02	0.03	0.08	0.06	0.07	0.69	2.31
NH_4^+	0.66	0.73	1.18	1.90	1.10	1.05	2.90	6.20
K^+	0.06	0.04	0.03	0.03	0.04	0.02	0.12	0.24
Mg_2^+	0.02	0.04	0.01	0.04	0.00	0.00	0.11	0.29
Ca_2^+	0.29	0.38	0.04	0.14	0.03	0.08	0.48	1.47
PM	56.44	65.66	47.10	53.85	72.33	114.64	82.85	96.04

Supplementary table 3: Night time mean $PM_{10-2.5}$ elemental and ionic concentrations, and standard deviation at all sampling sites during the summer period

Site	Kliprivier		Sebokeng		Sharpeville		Zamdela	
	Mean	±SD	Mean	±SD	Mean	±SD	Mean	±SD
Na	2.17	1.37	2.16	1.19	2.37	1.51	1.80	1.47
Mg	0.93	0.82	0.73	0.46	1.04	0.74	0.66	0.50
Al	1.32	0.79	1.66	0.86	1.65	1.00	1.36	0.77
Si	4.35	2.60	5.03	2.46	4.94	3.03	4.17	2.46
P	0.05	0.03	0.04	0.01	0.05	0.02	0.04	0.02
S	1.74	1.27	1.96	1.29	2.40	1.28	1.76	1.18
Cl	0.13	0.16	0.10	0.12	0.29	0.30	0.09	0.10
K	0.44	0.25	0.48	0.20	0.50	0.27	0.36	0.18
Ca	1.03	0.76	0.93	0.55	1.77	1.03	0.93	0.45
Ti	0.05	0.03	0.07	0.03	0.07	0.04	0.06	0.03
V	0.00	0.01	0.00	0.01	0.00	0.01	0.00	0.01
Cr	0.02	0.04	0.02	0.04	0.02	0.03	0.01	0.02
Mn	0.05	0.03	0.06	0.04	0.13	0.23	0.03	0.02
Fe	0.82	0.52	0.99	0.47	1.23	0.64	0.60	0.32
Ni	0.01	0.01	0.01	0.01	0.00	0.01	0.00	0.01
Cu	0.01	0.02	0.02	0.02	0.01	0.01	0.01	0.01
Zn	0.31	0.26	0.18	0.14	0.32	0.20	0.11	0.05
Pb	0.04	0.04	0.02	0.02	0.02	0.03	0.01	0.02
F^-	1.37	0.79	0.92	0.40	1.41	0.72	1.22	0.64
Cl^-	0.01	0.02	0.02	0.05	0.04	0.07	0.01	0.02
SO_4^{2-}	4.67	3.58	5.49	3.79	6.56	3.65	3.94	2.96
NO_3^-	0.11	0.26	0.10	0.15	0.25	0.40	0.49	0.87
$CH3COO^-$	0.00	0.00	0.00	0.01	0.00	0.00	0.00	0.00
$HCOO^-$	0.00	0.00	0.01	0.01	0.01	0.01	0.00	0.00
$C_2O_4^{2-}$	0.18	0.09	0.16	0.10	0.15	0.06	0.10	0.07
Na^+	0.15	0.11	0.17	0.13	0.18	0.13	0.12	0.13
NH_4^+	1.50	1.37	1.80	1.34	2.09	1.20	1.25	0.98
K^+	0.12	0.08	0.11	0.06	0.14	0.09	0.06	0.04
Mg_2^+	0.09	0.08	0.07	0.04	0.11	0.08	0.05	0.04
Ca_2^+	0.51	0.41	0.44	0.31	0.84	0.57	0.37	0.25
PM	34.12	18.6	35.99	26.1	39.45	20.25	27.61	14.45

Supplementary table 4: Night time mean $PM_{2.5}$ elemental and ionic concentrations, and standard deviation at all sampling sites during the summer period

Site	Kliprivier		Sebokeng		Sharpeville		Zamdela	
	Mean	±SD	Mean	±SD	Mean	±SD	Mean	±SD
Na	0.40	0.56	0.20	0.28	0.09	0.20	0.04	0.16
Mg	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Al	0.26	0.52	0.75	1.98	0.08	0.15	0.74	2.58
Si	1.07	1.33	0.80	1.43	0.55	0.49	0.89	0.51
P	0.03	0.02	0.02	0.03	0.02	0.01	0.01	0.01
S	1.46	0.95	1.46	1.01	1.89	1.00	1.60	1.23
Cl	0.12	0.12	0.09	0.11	0.09	0.08	0.07	0.07
K	0.26	0.27	0.17	0.17	0.24	0.14	0.14	0.12
Ca	0.31	0.76	0.27	0.51	0.10	0.21	0.10	0.27
Ti	0.03	0.04	0.03	0.05	0.02	0.03	0.04	0.06
V	0.02	0.04	0.03	0.05	0.02	0.04	0.03	0.04
Cr	2.76	10.92	3.24	12.79	0.04	0.16	0.07	0.27
Mn	0.08	0.20	0.07	0.21	0.02	0.04	0.02	0.04
Fe	7.55	29.83	8.83	34.85	0.15	0.60	0.20	0.79
Ni	0.74	2.88	0.86	3.41	0.01	0.03	0.01	0.04
Cu	0.02	0.05	0.07	0.08	0.07	0.12	0.04	0.07
Zn	0.34	0.52	0.29	0.21	0.23	0.25	0.25	0.29
Pb	0.11	0.27	0.07	0.16	0.12	0.19	0.05	0.09
F^-	0.17	0.66	0.19	0.40	0.59	0.82	2.14	2.56
Cl^-	0.02	0.04	0.00	0.00	0.01	0.01	0.02	0.03
SO_4^{2-}	3.30	2.11	3.39	2.57	3.78	2.71	3.35	2.78
NO_3^-	0.00	0.00	0.00	0.00	0.00	0.00	2.23	5.10
$CH3COO^-$	0.00	0.00	0.00	0.00	0.07	0.04	0.01	0.01
$HCOO^-$	0.00	0.01	0.00	0.00	0.01	0.01	0.00	0.00
$C_2O_4^{2-}$	0.17	0.11	0.10	0.07	0.10	0.09	0.09	0.05
Na^+	0.07	0.10	0.03	0.04	0.03	0.03	0.06	0.06
NH_4^+	0.33	0.45	0.76	0.71	0.95	0.76	2.10	2.15
K^+	0.09	0.06	0.04	0.03	0.05	0.03	0.03	0.03
Mg_2^+	0.02	0.06	0.01	0.02	0.00	0.01	0.00	0.01
Ca_2^+	0.17	0.36	0.08	0.22	0.04	0.08	0.07	0.14
PM	73.56	164.88	42.70	49.64	62.30	53.25	34.52	36.23

Supplementary table 5: Day time mean $PM_{10-2.5}$ elemental and ionic concentrations and standard deviation at all sampling sites during the winter period

Site	Kliprivier		Sebokeng		Sharpeville		Zamdela	
	Mean	±SD	Mean	±SD	Mean	±SD	Mean	±SD
Na	3.23	2.18	3.25	1.97	4.00	1.91	4.26	2.58
Mg	1.76	0.98	1.91	1.00	2.38	1.09	2.27	0.94
Al	3.46	1.73	4.75	2.80	4.11	1.51	4.16	2.19
Si	11.18	6.03	14.69	8.98	13.26	5.01	13.48	6.40
P	0.07	0.03	0.07	0.04	0.09	0.04	0.09	0.04
S	1.70	1.11	1.61	1.00	1.77	1.00	1.58	0.99
Cl	0.71	0.58	3.61	4.34	4.57	4.42	1.95	2.19
K	1.60	0.65	1.98	1.04	1.94	0.92	1.68	0.73
Ca	1.80	0.93	2.49	1.51	3.79	2.23	2.70	1.33
Ti	0.14	0.07	0.19	0.12	0.18	0.07	0.18	0.10
V	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01
Cr	0.69	1.49	0.88	1.61	0.99	1.77	0.70	1.51
Mn	0.12	0.07	0.12	0.07	0.14	0.08	0.06	0.04
Fe	2.75	4.79	4.25	5.53	4.33	6.05	2.52	4.77
Ni	0.21	0.45	0.26	0.47	0.29	0.51	0.21	0.45
Cu	0.02	0.01	0.01	0.02	0.02	0.01	0.01	0.01
Zn	0.17	0.09	0.18	0.10	0.32	0.33	0.17	0.10
Pb	0.03	0.04	0.02	0.04	0.03	0.04	0.01	0.03
F^-	1.49	0.64	2.73	1.90	2.92	1.32	2.54	1.27
Cl^-	0.08	0.11	0.42	0.61	0.80	1.20	0.23	0.37
SO_4^{2-}	4.87	2.84	4.26	3.07	4.89	2.94	4.40	2.81
NO_3^-	0.69	0.39	0.94	1.14	0.92	0.68	2.00	1.87
$CH3COO^-$	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
$HCOO^-$	0.02	0.01	0.01	0.01	0.01	0.01	0.01	0.01
$C_2O_4^{2-}$	0.23	0.11	0.23	0.11	0.22	0.14	0.22	0.10
Na^+	0.29	0.23	0.28	0.23	0.40	0.23	0.40	0.30
NH_4^+	1.42	0.93	1.73	1.32	1.84	1.25	1.82	0.99
K^+	0.55	0.27	0.66	0.41	0.71	0.40	0.55	0.32
Mg_2^+	0.13	0.08	0.17	0.10	0.23	0.12	0.16	0.09
Ca_2^+	0.68	0.36	1.05	0.61	1.63	0.86	1.05	0.61
PM	61.03	28.7	89.73	61.7	94.65	39.37	82.45	38.80

Supplementary table 6: Day time mean $PM_{2.5}$ elemental and ionic concentrations and standard deviation at all sampling sites during the winter period

Site	Kliprivier		Sebokeng		Sharpeville		Zamdela	
	Mean	±SD	Mean	±SD	Mean	±SD	Mean	±SD
Na	0.29	0.23	0.56	0.47	0.68	0.64	0.42	0.35
Mg	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Al	0.04	0.03	0.09	0.17	0.06	0.08	0.03	0.03
Si	0.18	0.15	0.47	0.64	0.33	0.33	0.16	0.12
P	0.01	0.01	0.01	0.01	0.01	0.02	0.01	0.01
S	1.49	0.87	1.30	0.95	1.44	0.90	1.22	0.90
Cl	0.09	0.07	0.21	0.28	0.94	2.02	0.11	0.15
K	0.87	0.42	0.94	0.56	1.01	0.58	0.76	0.50
Ca	0.04	0.12	0.11	0.27	0.11	0.26	0.08	0.18
Ti	0.02	0.03	0.03	0.05	0.04	0.04	0.01	0.02
V	0.02	0.04	0.01	0.03	0.01	0.02	0.04	0.06
Cr	8.30	17.95	0.00	0.00	0.00	0.00	6.36	13.73
Mn	0.04	0.10	0.00	0.01	0.02	0.04	0.06	0.11
Fe	22.07	47.57	0.00	0.00	0.00	0.00	17.68	38.09
Ni	2.33	5.01	0.00	0.00	0.00	0.00	1.92	4.13
Cu	0.07	0.11	0.07	0.11	0.11	0.11	0.01	0.05
Zn	0.21	0.26	0.33	0.32	0.26	0.28	0.14	0.17
Pb	0.10	0.16	0.07	0.12	0.18	0.22	0.10	0.20
F^-	0.25	0.40	0.27	0.62	0.59	1.02	0.44	0.58
Cl^-	0.04	0.06	0.01	0.02	0.00	0.01	0.03	0.06
SO_4^{2-}	3.46	2.35	2.52	2.48	3.03	2.30	2.50	2.25
NO_3^-	0.63	2.46	0.00	0.00	13.46	31.20	0.32	0.74
$CH3COO^-$	0.00	0.01	0.01	0.05	0.00	0.00	0.00	0.01
$HCOO^-$	0.03	0.03	0.00	0.00	0.00	0.01	0.01	0.01
$C_2O_4^{2-}$	0.11	0.08	0.03	0.04	0.09	0.12	0.11	0.07
Na^+	0.05	0.06	0.01	0.03	0.03	0.09	0.06	0.09
NH_4^+	1.60	1.39	0.59	0.81	5.24	9.42	1.22	0.96
K^+	0.34	0.18	0.29	0.23	0.32	0.21	0.25	0.21
Mg_2^+	0.00	0.00	0.01	0.03	0.00	0.00	0.00	0.00
Ca_2^+	0.00	0.01	0.17	0.45	0.00	0.00	0.04	0.15
PM	36.44	36.89	84.62	78.33	67.73	49.16	38.29	54.66

Supplementary table 7: Night time mean $PM_{10-2.5}$ elemental and ionic concentrations, and standard deviation at all sampling sites during the winter period

Site	Kliprivier		Sebokeng		Sharpeville		Zamdela	
	Mean	±SD	Mean	±SD	Mean	±SD	Mean	±SD
Na	3.35	1.73	3.14	1.75	4.51	1.62	3.73	1.73
Mg	1.47	0.51	1.65	0.77	3.24	1.75	2.30	1.40
Al	2.62	0.89	3.45	1.52	4.09	2.04	4.86	4.25
Si	8.42	2.60	10.57	4.67	12.62	6.11	13.69	10.06
P	0.07	0.02	0.06	0.03	0.11	0.05	0.13	0.10
S	2.18	1.03	1.90	1.11	2.92	1.21	3.01	2.39
Cl	3.57	2.55	1.91	1.86	7.13	5.12	2.79	2.82
K	2.22	0.91	1.84	1.08	2.53	1.39	1.99	1.13
Ca	1.52	0.62	1.84	0.83	5.45	3.08	2.89	2.14
Ti	0.11	0.04	0.13	0.06	0.18	0.09	0.23	0.22
V	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01
Cr	0.51	1.39	0.71	1.52	0.73	1.58	0.47	1.30
Mn	0.11	0.05	0.08	0.07	0.26	0.18	0.08	0.06
Fe	1.80	4.09	2.84	5.02	5.09	6.26	1.89	3.80
Ni	0.15	0.40	0.20	0.44	0.21	0.45	0.14	0.38
Cu	0.02	0.01	0.02	0.03	0.02	0.02	0.02	0.02
Zn	0.47	0.29	0.28	0.21	0.65	0.45	0.25	0.22
Pb	0.04	0.03	0.04	0.04	0.09	0.10	0.02	0.03
F^-	1.85	0.71	2.18	0.99	3.74	1.92	2.91	1.10
Cl^-	0.62	0.67	0.14	0.17	1.58	1.63	0.36	0.42
SO_4^{2-}	6.21	2.78	5.03	3.04	7.77	3.51	8.78	6.82
NO_3^-	2.49	1.90	1.08	0.99	1.83	1.35	3.84	1.89
$CH3COO^-$	0.01	0.01	0.00	0.00	0.01	0.00	0.00	0.00
$HCOO^-$	0.02	0.01	0.01	0.01	0.01	0.01	0.01	0.01
$C_2O_4^{2-}$	0.29	0.10	0.25	0.11	0.27	0.14	0.21	0.12
Na^+	0.28	0.15	0.24	0.15	0.38	0.14	0.35	0.17
NH_4^+	2.41	1.06	1.82	1.04	2.84	1.23	3.75	2.24
K^+	1.03	0.48	0.68	0.49	1.08	0.70	0.78	0.51
Mg_2^+	0.11	0.05	0.13	0.06	0.32	0.18	0.16	0.09
Ca_2^+	0.62	0.27	0.85	0.41	2.39	1.27	1.03	0.68
PM	92.18	33.5	73.01	37.6	113.32	59.42	95.65	60.18

Supplementary table 8: Night time mean $PM_{2.5}$ elemental and ionic concentrations, and standard deviation at all sampling sites during the winter period

Site	Kliprivier		Sebokeng		Sharpeville		Zamdela	
	Mean	±SD	Mean	±SD	Mean	±SD	Mean	±SD
Na	0.92	0.63	0.62	0.40	1.42	0.86	0.45	0.44
Mg	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Al	0.04	0.05	0.15	0.34	0.07	0.10	0.07	0.14
Si	0.24	0.14	0.65	1.24	0.48	0.32	0.37	0.49
P	0.03	0.02	0.01	0.01	0.04	0.05	0.02	0.03
S	1.82	0.91	1.59	0.93	2.56	1.03	2.57	1.98
Cl	0.28	0.18	0.16	0.14	1.16	1.48	0.17	0.15
K	1.62	0.77	1.03	0.82	1.73	1.03	1.22	0.89
Ca	0.13	0.39	0.26	0.41	0.07	0.20	0.22	0.36
Ti	0.04	0.06	0.03	0.06	0.03	0.03	0.02	0.03
V	0.06	0.07	0.05	0.07	0.02	0.03	0.05	0.07
Cr	7.55	16.25	0.00	0.00	0.00	0.00	6.28	13.55
Mn	0.10	0.21	0.01	0.03	0.01	0.03	0.06	0.12
Fe	20.16	43.36	0.00	0.00	0.00	0.00	17.52	37.73
Ni	2.11	4.55	0.00	0.00	0.00	0.00	1.88	4.04
Cu	0.05	0.05	0.07	0.11	0.04	0.09	0.07	0.09
Zn	0.49	0.40	0.15	0.20	0.39	0.24	0.22	0.25
Pb	0.18	0.25	0.08	0.21	0.09	0.17	0.06	0.15
F^-	0.43	0.53	0.10	0.22	0.88	1.23	0.57	0.64
Cl^-	0.03	0.04	0.03	0.04	0.00	0.02	0.02	0.04
SO_4^{2-}	4.09	2.36	3.38	2.22	5.20	2.43	5.91	5.07
NO_3^-	0.51	1.77	0.00	0.00	5.25	17.16	0.40	1.10
$CH3COO^-$	0.01	0.02	0.01	0.04	0.00	0.00	0.00	0.01
$HCOO^-$	0.03	0.02	0.00	0.00	0.00	0.01	0.01	0.01
$C_2O_4^{2-}$	0.20	0.12	0.06	0.07	0.24	0.18	0.14	0.10
Na^+	0.07	0.07	0.00	0.01	0.10	0.24	0.03	0.05
NH_4^+	1.67	1.07	0.87	0.70	3.81	4.54	2.60	1.89
K^+	0.66	0.36	0.30	0.32	0.61	0.44	0.42	0.36
Mg_2^+	0.00	0.02	0.02	0.07	0.00	0.00	0.00	0.00
Ca_2^+	0.00	0.00	0.18	0.41	0.01	0.05	0.04	0.09
PM	43.55	31.46	62.78	48.45	128.53	187.10	39.37	37.73

Supplementary table 9: Day time mean $PM_{10-2.5}$ elemental and ionic concentrations and standard deviation at all sampling sites during the spring period

Site	Kliprivier		Sebokeng		Sharpeville		Zamdela	
	Mean	±SD	Mean	±SD	Mean	±SD	Mean	±SD
Na	3.18	1.31	3.31	1.17	3.53	1.50	3.34	1.96
Mg	1.66	0.78	2.30	0.98	2.88	1.79	2.41	1.86
Al	2.63	1.21	5.61	2.57	4.19	2.16	5.78	5.15
Si	9.10	4.16	17.25	7.35	13.91	7.19	16.92	13.93
P	0.07	0.03	0.09	0.04	0.10	0.05	0.11	0.09
S	1.06	0.53	1.04	0.53	1.08	0.57	0.89	0.68
Cl	0.12	0.11	0.31	0.24	0.47	0.46	0.31	0.33
K	1.67	1.04	2.10	1.03	1.91	1.02	1.64	1.28
Ca	1.60	0.86	2.95	1.41	4.49	3.22	3.30	2.69
Ti	0.11	0.05	0.25	0.10	0.21	0.11	0.28	0.26
V	0.01	0.01	0.02	0.01	0.01	0.01	0.01	0.01
Cr	0.30	1.20	0.85	1.83	0.55	1.52	0.00	0.00
Mn	0.08	0.05	0.13	0.07	0.17	0.10	0.09	0.06
Fe	1.51	3.40	4.84	5.05	4.14	4.23	1.06	1.19
Ni	0.09	0.37	0.26	0.57	0.16	0.45	0.00	0.00
Cu	0.03	0.01	0.03	0.02	0.03	0.02	0.03	0.02
Zn	0.15	0.11	0.15	0.07	0.28	0.20	0.15	0.10
Pb	0.01	0.03	0.02	0.02	0.02	0.02	0.01	0.02
F^-	2.93	1.11	5.37	1.32	5.22	1.36	4.06	2.40
Cl^-	0.01	0.01	0.01	0.02	0.01	0.02	0.01	0.01
SO_4^{2-}	2.91	1.49	3.10	1.73	3.35	1.71	2.54	2.03
NO_3^-	0.03	0.08	0.14	0.35	0.03	0.05	0.06	0.13
$CH3COO^-$	0.00	0.00	0.01	0.00	0.00	0.00	0.00	0.00
$HCOO^-$	0.02	0.01	0.01	0.01	0.02	0.01	0.01	0.01
$C_2O_4^{2-}$	0.46	0.22	0.59	0.17	0.53	0.19	0.35	0.21
Na^+	0.28	0.13	0.37	0.15	0.38	0.15	0.36	0.21
NH_4^+	0.82	0.46	1.11	0.66	0.85	0.52	0.82	0.68
K^+	0.72	0.49	0.65	0.38	0.71	0.38	0.55	0.42
Mg_2^+	0.16	0.07	0.26	0.11	0.35	0.19	0.27	0.19
Ca_2^+	0.81	0.39	1.39	0.63	1.86	0.95	1.43	1.43
PM	52.16	23.1	93.10	43.8	102.48	70.77	84.32	63.69

Supplementary table 10: Day time mean $PM_{2.5}$ elemental and ionic concentrations and standard deviation at all sampling sites during the spring period

Site	Kliprivier		Sebokeng		Sharpeville		Zamdela	
	Mean	±SD	Mean	±SD	Mean	±SD	Mean	±SD
Na	0.33	0.35	0.50	0.26	0.40	0.40	0.75	1.28
Mg	0.00	0.00	0.00	0.00	0.00	0.01	0.08	0.29
Al	0.02	0.02	0.06	0.03	0.08	0.09	0.31	0.67
Si	0.23	0.21	0.37	0.29	0.58	0.31	1.05	2.35
P	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01
S	0.79	0.46	0.83	0.49	0.80	0.52	0.90	0.59
Cl	0.04	0.04	0.05	0.07	0.05	0.07	0.06	0.07
K	0.75	0.64	0.83	0.61	0.83	0.59	0.69	0.43
Ca	0.08	0.19	0.09	0.15	0.54	0.77	0.34	0.90
Ti	0.02	0.03	0.01	0.02	0.04	0.05	0.02	0.03
V	0.03	0.04	0.02	0.03	0.02	0.04	0.03	0.04
Cr	0.00	0.00	0.00	0.00	0.00	0.00	8.25	17.08
Mn	0.02	0.05	0.03	0.06	0.03	0.04	0.12	0.21
Fe	0.00	0.00	0.00	0.00	0.00	0.00	22.87	47.36
Ni	0.00	0.00	0.00	0.00	0.00	0.00	2.47	5.11
Cu	0.20	0.14	0.13	0.16	0.17	0.16	0.24	0.17
Zn	0.28	0.30	0.26	0.23	0.32	0.26	0.25	0.33
Pb	0.10	0.30	0.09	0.21	0.05	0.14	0.05	0.08
F^-	0.78	1.92	1.41	2.97	0.81	0.85	0.63	1.29
Cl^-	0.01	0.02	0.03	0.05	0.00	0.01	0.09	0.14
SO_4^{2-}	1.82	1.09	1.90	1.27	1.77	1.20	2.21	1.45
NO_3^-	0.07	0.30	0.05	0.19	0.00	0.00	0.00	0.00
$CH3COO^-$	0.03	0.03	0.05	0.06	0.11	0.08	0.04	0.06
$HCOO^-$	0.00	0.00	0.01	0.04	0.03	0.01	0.05	0.11
$C_2O_4^{2-}$	0.09	0.09	0.18	0.14	0.18	0.10	0.16	0.25
Na^+	0.02	0.02	0.03	0.05	0.07	0.07	0.11	0.15
NH_4^+	0.38	1.03	0.56	1.38	0.19	0.26	0.25	0.40
K^+	0.34	0.28	0.23	0.18	0.31	0.21	0.24	0.19
Mg_2^+	0.00	0.00	0.00	0.01	0.00	0.01	0.15	0.37
Ca_2^+	0.04	0.10	0.14	0.32	0.27	0.37	0.77	1.78
PM	52.72	35.81	74.04	77.21	63.24	39.54	40.26	33.81

Supplementary table 11: Night time mean $PM_{10-2.5}$ elemental and ionic concentrations, and standard deviation at all sampling sites during the spring period

Site	Kliprivier		Sebokeng		Sharpeville		Zamdela	
	Mean	±SD	Mean	±SD	Mean	±SD	Mean	±SD
Na	3.98	1.86	3.87	2.38	5.05	2.31	3.96	2.65
Mg	1.61	0.61	1.74	0.53	4.56	3.32	2.42	1.42
Al	2.39	1.17	3.36	1.27	3.97	1.74	4.45	2.96
Si	7.91	3.75	10.66	3.90	12.15	5.03	13.21	8.19
P	0.08	0.03	0.06	0.03	0.11	0.06	0.11	0.08
S	0.94	0.33	0.97	0.48	1.47	0.44	1.56	1.13
Cl	0.31	0.40	0.36	0.38	1.51	2.11	0.71	0.65
K	1.58	0.82	1.61	0.81	1.86	0.84	1.56	0.89
Ca	1.68	0.89	1.92	0.78	7.27	6.70	2.96	2.06
Ti	0.10	0.05	0.14	0.05	0.18	0.08	0.19	0.12
V	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01
Cr	0.28	1.11	0.89	1.93	0.68	1.86	0.00	0.00
Mn	0.10	0.07	0.08	0.06	0.27	0.19	0.10	0.09
Fe	1.52	3.22	3.64	5.41	5.46	5.63	0.88	0.93
Ni	0.08	0.34	0.26	0.56	0.19	0.51	0.00	0.00
Cu	0.03	0.03	0.02	0.02	0.03	0.02	0.02	0.02
Zn	0.55	0.52	0.29	0.29	0.59	0.37	0.30	0.27
Pb	0.02	0.02	0.01	0.02	0.08	0.16	0.02	0.04
F^-	3.22	1.01	3.78	1.74	6.07	2.48	4.14	2.07
Cl^-	0.02	0.05	0.01	0.01	0.10	0.19	0.02	0.03
SO_4^{2-}	2.57	1.05	2.74	1.52	4.08	1.36	4.51	3.36
NO_3^-	0.02	0.09	0.06	0.17	0.05	0.09	0.11	0.18
$CH3COO^-$	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
$HCOO^-$	0.01	0.00	0.01	0.01	0.02	0.01	0.01	0.01
$C_2O_4^{2-}$	0.49	0.18	0.51	0.18	0.50	0.17	0.37	0.19
Na^+	0.32	0.22	0.44	0.32	0.48	0.28	0.44	0.34
NH_4^+	0.73	0.40	0.86	0.69	1.00	0.50	1.47	1.07
K^+	0.74	0.43	0.52	0.34	0.69	0.37	0.58	0.37
Mg_2^+	0.16	0.06	0.18	0.07	0.50	0.36	0.26	0.14
Ca_2^+	0.81	0.38	0.89	0.35	2.68	2.01	1.48	1.48
PM	51.16	20.0	66.94	23.7	100.59	48.55	74.13	49.15

Supplementary table 12: Night time mean $PM_{2.5}$ elemental and ionic concentrations, and standard deviation at all sampling sites during the spring period

Site	Kliprivier		Sebokeng		Sharpeville		Zamdela	
	Mean	±SD	Mean	±SD	Mean	±SD	Mean	±SD
Na	0.50	0.26	0.64	0.43	1.20	0.83	0.46	0.40
Mg	0.01	0.04	0.00	0.00	0.20	0.41	0.00	0.00
Al	0.04	0.03	0.06	0.05	0.07	0.08	0.06	0.05
Si	0.38	0.17	0.35	0.11	0.73	0.30	0.22	0.18
P	0.03	0.01	0.01	0.01	0.02	0.02	0.02	0.01
S	0.73	0.27	0.74	0.39	1.05	0.38	1.24	0.96
Cl	0.05	0.09	0.10	0.11	0.19	0.30	0.07	0.10
K	0.86	0.47	0.76	0.58	0.93	0.57	0.57	0.43
Ca	0.36	0.69	0.47	1.01	1.17	1.74	0.11	0.40
Ti	0.02	0.05	0.04	0.06	0.02	0.04	0.02	0.03
V	0.02	0.04	0.02	0.03	0.03	0.06	0.01	0.02
Cr	0.00	0.00	0.00	0.00	0.00	0.00	5.60	15.31
Mn	0.03	0.05	0.02	0.04	0.06	0.07	0.10	0.18
Fe	0.00	0.00	0.00	0.00	0.00	0.00	15.05	41.12
Ni	0.00	0.00	0.00	0.00	0.00	0.00	1.60	4.38
Cu	0.16	0.13	0.14	0.12	0.16	0.12	0.17	0.15
Zn	0.36	0.29	0.39	0.34	0.69	0.42	0.21	0.22
Pb	0.05	0.29	0.04	0.06	0.08	0.17	0.06	0.15
F^-	0.82	2.23	2.50	3.61	1.16	1.13	0.12	0.23
Cl^-	0.01	0.03	0.02	0.03	0.00	0.01	0.01	0.03
SO_4^{2-}	1.50	0.73	1.67	1.10	2.20	0.96	2.90	2.54
NO_3^-	0.26	1.02	1.11	2.55	0.03	0.10	0.00	0.00
$CH3COO^-$	0.03	0.03	0.04	0.06	0.11	0.08	0.03	0.04
$HCOO^-$	0.00	0.00	0.01	0.02	0.03	0.02	0.00	0.02
$C_2O_4^{2-}$	0.17	0.14	0.27	0.14	0.32	0.22	0.10	0.09
Na^+	0.03	0.03	0.07	0.08	0.12	0.22	0.04	0.05
NH_4^+	0.36	1.26	1.53	2.38	0.29	0.33	0.59	0.98
K^+	0.35	0.24	0.22	0.18	0.33	0.21	0.20	0.15
Mg_2^+	0.00	0.00	0.01	0.02	0.01	0.02	0.01	0.02
Ca_2^+	0.14	0.31	0.11	0.20	0.51	0.73	0.05	0.09
PM	45.70	25.18	71.14	44.96	61.54	63.98	34.75	25.08

ANNEXURE II: SPATIAL EMISSIONS QUANTIFICATION

Industries

For the VTAPA, industrial sources represent mostly licensed stationary facilities that submit emission reports annually. Significant emitters can be broadly classified into three categories which include, iron and steel manufacturers, chemical and petrochemical sectors, and power generation. Emissions for these industries were quantified based on activity data obtained from information reported by the National Atmospheric Emission Inventory System (NAEIS) for the 2017 calendar year (Department of Environmental Affairs, 2015).

Domestic combustion

This involved a top-down method in which national gas, paraffin and coal consumption data was downscaled into smaller geographical units using Community Survey 2016 and Census 2011 data (Statistics South Africa, 2012; Statistics South Africa, 2016). In the case of wood fuel use, a bottom-up approach was used in which household wood fuel consumption was aggregated into larger geographical units. Domestic combustion emissions were then calculated using emission factors from different literature

Vehicles

Both a top-down and a bottom-up approach was used in the calculation of vehicle emissions. In the bottom-up approach, road count data obtained from SANRAL national counts for 2016 and GAUTRAINS Gauteng Manual counts for 2015 were used in determining the vehicle kilometre travelled (VKT). For the top-down method, provincial fuel sales and fuel efficiency data were used to estimate vehicle kilometre travelled (VKT). The COPERT model was then used to quantify vehicle emissions (Ntziachristos & Samaras, 2009).

Waste combustion

Waste combustion emissions were estimated using data on waste per person and composition for the VTAPA (Gauteng Department of Agriculture and Rural Development, 2018). From the data, the amount of waste burned is quantified and then multiplied with the emission factors for waste. Emissions from landfills and wastewater treatment centres were not quantified as input data for their quantification is not available for the VTAPA.

Windblown dust

Windblown dust emissions were estimated for mine waste facilities, product stockpiles, and ash storages. The Airborne Dust Dispersion Model from Area Sources (ADDAS) was used in the emissions quantification (Burger, 2011). Material particle density, moisture content, particle size distribution and site-specific surface characteristics were used as inputs into this model.

Biomass burning

Biomass burning emissions from large scale agricultural burning and natural fires for the year 2016 were obtained from the Fire INventory from NCAR (FINN) dataset (Wiedinmyer *et al.*, 2011). Corrections were made to the FINN data in which fires that were apportioned incorrectly to surface coal mines and large hot/reflective rooftops were removed through a masking procedure.

Data processing

The spatial emissions data were obtained in a GIS format. Subsets for the VTAPA extracted using QGIS. Attribute tables for each emission category were obtained from the VTAPA subsets and used as input into the CALPUFF model.

ANNEXURE III: PERMISSION FROM CO-AUTHORS

13 March 2020.

To whom it may concern.

Dear Sir/Madam.

We, the undersigned and co-authors of the manuscript,

L. Muyemeki, S.J. Piketh and **R. P. Burger**, 2020. Evaluating the potential of remote sensing imagery in mapping ground-level fine particulate matter ($PM_{2.5}$) for the Vaal Triangle Priority Area

Herewith give permission that this manuscript can be submitted as part of Luckson Muyemeki's PhD thesis. Although we were involved with the conceptualization of this work, Luckson Muyemeki was primarily responsible for the execution and documentation of this research.

Sincerely

Stuart Piketh

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Roelof Burger

roelof.burger@nwu.ac.za

13 March 2020.

To whom it may concern.

Dear Sir/Madam.

We, the undersigned and co-authors of the manuscript,

L. Muyemeki, S.J. Piketh and **R. P. Burger**, 2020. Updated $PM_{10-2.5}$ and $PM_{2.5}$ source apportionment for the Vaal Triangle air pollution priority area, South Africa

Herewith give permission that this manuscript can be submitted as part of Luckson Muyemeki's PhD thesis. Although we were involved with the conceptualization of this work, Luckson Muyemeki was primarily responsible for the execution and documentation of this research.

Sincerely

Stuart Piketh

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13 March 2020.

To whom it may concern.

Dear Sir/Madam.

We, the undersigned and co-authors of the manuscript,

L. Muyemeki, S.J. Piketh, R. P. Burger, P. Rafaj and **G. Kiesewetter**, 2020. Integrated assessment of strategies to reduce air pollution in the Vaal Triangle Priority Area, South Africa

Herewith give permission that this manuscript can be submitted as part of Luckson Muyemeki's PhD thesis. Although we were involved with the conceptualization of this work, Luckson Muyemeki was primarily responsible for the execution and documentation of this research.

Sincerely

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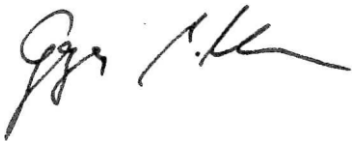

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