Source apportionment of ambient particulate matter in Kwadela, Mpumalanga

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“You cannot affirm the power plant and condemn the smokestack, or affirm the smoke and condemn the cough”

- Wendell E. Berry, Environmental activist, 1981.

PREFACE

Poor air quality conditions in low income communities are of great concern in many developing countries. The resulting impacts of air pollution include respiratory problems amongst the poor and premature deaths. Improving air quality in poor residential areas will also lead to the improvement in live standards. This research attempts to highlight the importance of domestic fuel burning practices in South Africa and the need for mitigation strategies.

This dissertation is written in an article format and is divided into five chapters. Chapter 1 introduces the role of domestic fuel burning in South Africa. A comprehensive project description is provided and the aim of this study is clearly stated. In the literature review all the components relevant to particulate matter occurrences are discussed along with all the polluting sources. Chapter 2 outlines the data acquisition methods, equipment used, analysis procedures along with all the calculations used. Chapter 3 illustrates the role and importance of domestic fuel burning emissions in South Africa. The particulate matter levels were quantified in Chapter 4 and the contribution of every polluting source illustrated. All the results were summarised and discussed in Chapter 5. The methodology (Chapter 2) was briefly summarized in Chapter 3 and Chapter 4 due to the article format of this dissertation.

This research was part of a project with the aim of establishing the baseline of air quality conditions in a low income community. Air quality measurements were undertaken in Kwadela due to the prevalence of coal combustion practices. Interventions were implemented to minimize coal burning occurrences for heating purposes. The effects of these interventions were tested by measuring the air quality in the winter and summer. The overall aim of this project was to control emissions from residential fuel burning and to improve the quality of life of the local residents.

Project deliverables of this dissertation includes a submission to a South African Journal and conference presentations. Chapter 3: “Domestic fuel use in South African low income settlements” has been submitted to the South African Geographical Journal (Manuscript ID: RSAG-2015-0056) for review. This paper was also accepted in the annual conference proceedings of the National Association for Clean Air (NACA) and presented at the conference (Umhlanga, 8-10 October 2014). Chapter 4 of this research

“Source Apportionment of ambient particulate matter in Kwadela, Mpumalanga” was presented in a poster at the NACA conference (Bloemfontein, 1-2 October 2015). A prize for the best scientific poster has been awarded for this work by the NACA panel.

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“To my Lord, Jesus Christ, who made all the impossible, practical.”

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ABSTRACT

The importance of domestic fuel burning emissions was extensively explored in this research. The aim of this research was to identify the main cause of particulate matter in Kwadela, Mpumalanga. Low income communities, such as Kwadela, are notorious for emitting particulate matter into the atmosphere, from domestic coal burning. People in poor settlements tend to use a mixture of energy carriers such as wood, coal, animal dung and paraffin. Indoor and outdoor pollution from household combustion emissions influence a large fraction of South Africa's population. Effects from these emissions are intensified in dense areas with severe health and environmental consequences.

Fuel burning practices was firstly investigated on a national scale. The fuel use patterns and spatial distribution of fuel burning settlements was determined by using the South African 2011 census data. Proximity analyses on the variables that affected fuel choice were undertaken by means of Geographic Information Systems. Statistical R-square calculations were conducted to demonstrate the relationship between the determining factors and the number of fuel users. From these analyses it was found that informal and traditional households can be classified as fuel burning settlements. Determinants that could classify typical fuel burning settlements were identified and the number of people and communities affected by these emissions were calculated. An upper and lower limit calculation method was applied to determine the fraction of the population that was exposed to air pollution. According to the lower limit calculations 14 199 261 people are exposed to indoor air pollution and 19 148 085 to outdoor pollution. The upper limit estimates showed that 28 398 522 people were affected by indoor air pollution 38 296 170 by outdoor pollution. The fraction of people affected by indoor and outdoor pollution was thus 54.8% and 73.9% respectively.

The local domain of this study involved the characterisation of the ambient air quality in Kwadela. The air quality of two sampling periods was investigated; a winter campaign that extended from 21/07 to 29/07 and 05/08 to 12/08 (2014) as well as a summer period from 27/03 to 14/04 (2015). Fine (<2.5µm) and coarse (10µm≤adµm≥2.5µm) particles were collected using the Gent Stacked Filter Units with a PM_{10} cut size inlet. Each filter unit was exposed for 12 hours (06h00-18h00 and 18h00-06h00). These filters were consequently analysed for their chemical characteristics through inductively

coupled plasma-mass spectrometry, X-ray fluorescence and Ion chromatic system analyses. The most abundant species found in the winter samples were sulphate, sodium, nickel and ammonium. Ambient coarse particulate matter sampled during the summer was mainly compiled of copper, sulphur, silicon and cadmium species. Gravimetric results for the winter and summer samples showed that maximum concentrations obtained in the coarse fractions were 86.6 µg/m^3 (average of 25.1 µg/m^3) and 64.58 µg/m^3 (average of 24.68 µg/m^3) respectively. The measured fine fraction had a maximum of 170.2 µg/m^3 for the winter (average 42.3 µg/m3) and 22.25 µg/m^3 (average of 14.81 µg/m^3) during the summer campaigns. Several of these samples exceeded the national air quality standards. Higher gravimetric masses sampled during the winter can be explained by the local community's fuel combustion practices. More coal burning occurred for heating purposes due to colder ambient temperatures.

Lastly, the samples were analysed using Chemical Mass Balance to apportion contributions from different sources. The thirteen sources that influenced Kwadela's air quality were identified. Residential coal combustion was the foremost polluter that contributed a total of 738.49 µg/m^3 to all the samples. The other polluting sources were diesel motor vehicles with a total contribution of 116.12 µg/m^3, refuse/wood combustion with 37.01 µg/m^3, paved road dust with 34.68 µg/m^3 and biomass burning with 31.04 µg/m^3. Coal combustion was thus by far the greatest source of air pollution.

Domestic fuel burning practices should therefore be controlled in order to achieve sustainable, clean air standards.

Key terms: domestic fuel combustion, particulate matter, source apportionment, chemical mass balance model

OPSOMMING

Die belang van huishoudelike brandstof gebruik was breedvoerig ondersoek in hierdie studie. Die doel van hierdie navorsing was om die hoof oorsaak van lugbesoedeling in Kwadela, Mpumalanga te identifiseer. Lae inkomste gemeenskappe, soos Kwadela, is berug vir die vrystelling van vastestof partikels weens hulle verbrandings praktyke. Mense in hierdie nedersettings is geneig om ’n verskeidenheid van brandstowwe te gebruik soos; hout, steenkool, beesmis, gas en paraffien. Binnehuise en buitenste lugbesoedeling van hierdie uitlaatgasse beïnvloed ’n groot proporsie van die Suid-Afrikaanse populasie. Verhoogde effekte van uitlaatgasse word ervaar in digte areas, met erge omgewings en gesondheid nagevolge.

Huishoudelike brandstof gebruik was eerstens op ’n nasionale skaal ondersoek. Die gebruikers patrone en ruimtelike verspreiding van nedersettings wat afhanklik was van alternatiewe energie bronne was bepaal deur gebruik te maak van die 2011 sensus data. Afstands-analises van die faktore wat brandstof gebruik beïnvloed was uitgevoer deur Geografiese Inligtingstelsels. Statistiese R-kwadraat berekeninge was gebruik om die verhouding tussen die bepalende faktore en die aantal brandstof gebruikers te demonstreer. Vanuit hierdie berekeninge was daar gevind dat informele en tradisionele huishoudings geklassifiseer kan word as tipiese brandstof verbruikers. Faktore wat gebruik kan word om tipiese brandstof verbruikende areas te klassifiseer was geïdentifiseer en die aantal mense en gemeenskappe wat deur die lugbesoedeling geraak word was bepaal. ’n Boonste en onderste limiet berekening was gedoen om die proporsie van mense wat aan besoedeling blootgestel was te bepaal. Volgens die onderste limiet berekeninge was 14 199 261 mense blootgestel aan binnehuise lugbesoedeling en 19 148 085 aan buite lug besoedeling. Ongeveer 28 398 522 mense was geraak deur binnehuise lugbesoedeling volgens die boonste limiet berekeninge en 38 296 170 deur buite lugbesoedeling. Die hoeveelheid mense geraak deur binne en buite lugbesoedeling was dus 54,8% en 73,9% onderskeidelik.

Die plaaslike fokus van hierdie studie het die karakterisering van lugpartikels in Kwadela behels. Die lugkwaliteit van twee steekproewe was ondersoek: ’n winter steekproef vanaf 21/07 tot 29/07 en 05/08 tot 12/08 (2014) asook ’n somer steekproef van 27/03 tot 14/04 (2015). Fyn (<2.5µm) en growwe (10µm<adµm>2.5µm) partikels was versamel deur die Gent filter eenheid. Elke filter pak was blootgestel vir ’n 12 uur periode (06h00-

18h00 en 18h00-06h00). Daaropvolgend was die filters deur ICP-MS, XRF en ICS metodes ontleed vir die chemiese samestelling. Die volopste chemiese spesies gevind in die winter monsters was SO_4^{2-}, Na, Ni en NH_4^+. Die lugdeeltjies wat versamel was in die somer was hoofsaaklik saamgestel uit Cu, S, Si en Cd spesies. Die gravimetriese ondersoeke van die winter en somer steekproewe toon dat die maksimum gemete konsentrasies van growwe lugdeeltjies was 86.6 µg/m^3 (gemiddeld 25.1 µg/m^3) en 64.58 µg/m^3 (gemiddeld 24.68 µg/m^3). Die fyn lugdeeltjies het ’n maksimum gewig van 170.2 µg/m^3 vir die winter getoon (gemiddeld 42.3 µg/m^3) en 22.25 µg/m^3 (gemiddeld van 14.81 µg/m^3) vir die somer monsters. Verskeie van die monsters het die nasionale lugkwaliteit standaarde oorskry. Die hoër gravimetriese massas wat in die winter versamel was kan toegeskryf word aan die gemeenskap se verbrandings praktyke. Meer steenkool was verbrand in die winter vir verhittings doeleindes.

Laastens was die lugprofiele verder geanaliseer in die CMB om die bydra van die besoedelings bronne in aanmerking te bring. Dertien lugbesoedelings bronne was geïdentifiseer. Steenkool verbranding was die grootste bron van lugbesoedeling wat ‘n totaal van 738.49 µg/m^3 tot al die monsters bygedra het. Die ander besoedelingsbronne was diesel voertuie met 116.12 µg/m^3, vullis/hout verbranding met 37.01 µg/m^3, teerpad stof met 34.68 µg/m^3 en biomassa verbranding met 31.04 µg/m^3. Steenkool verbranding was dus by ver, die hoof oorsaak van lugbesoedeling. Huishoudelike brandstof verbranding moet dus beheer word om skoon, volhoubare lugkwaliteit standaarde te handhaaf.

Sleutelterme: huishoudelike brandstof verbranding, partikelstof, bron toewysing, chemiese massabalans model

TABLE OF CONTENTS

LIST OF TABLES

LIST OF FIGURES

LIST OF ABBREVIATIONS

Al	Aluminium
API	Air pollution Index
APM	Airborne particulate matter
AsS	Arsenic sulphite
BC	Black carbon
BDL	Below detection limit
C	Carbon
Ca	Calcium
$CaCl^{+}$	Calcium chloride
Cd	Cadmium
CH_4	Methane
Cl	Chlorine
CMB	Chemical mass balance
CO	Carbon monoxide
CO_2	Carbon dioxide
Cu	Copper
DEA	Department of Environmental Affairs
DEAT	Department of Environmental Affairs and Tourism
DF	Degrees of freedom
DOC	Dissolved organic acids
EC	Elemental carbon
Fe	Iron
FeS	Iron(II) sulphide
GHG	Greenhouse gasses
GIS	Geographical Information Systems
HCl	Hydrochloric acid
Hg	Mercury

HNO_3	Nitric acid
HPA	Highveld priority area
ICP-MS	Inductively coupled plasma–mass spectrometry
ICS	Ion chromatographic system
ISE	Ion selective electrodes
K	Kalium
LPG	Liquefied petroleum gas
MEC	Member of executive council
Mg	Magnesium
N	Nitrogen
N_2O	Nitrous oxide
Na	Sodium
NEM:AQA	National Environmental Management: Air Quality Act
NH_3	Ammonia
NH_4^+	Ammonium
NO_{3-}	Nitrate
NO_x	Nitrogen oxides
OC	Organic carbon
PAH	Polycyclic aromatic hydrocarbons
Pb	Lead
PCA	Principal component analysis
PDF	Probability density function
PM	Particulate matter
PMF	Positive matrix factorization
PO_x	Phosphates
S	Sulphur
SAL	Small area layer
SANS	South African National Standard

SCE	Source contribution estimate
SFU	Stacked Filter Unit
Si	Silicon
SO_2	Sulphur dioxide
SO_2	Sulphur dioxide
SO^3	Sulphur trioxide
SO_4^{2-}	Sulphate
SO_x	Sulphur oxides
TB	Tuberculosis
Th	Thorium
Ti	Titanium
TM	Total mass
TSM	Total suspended matter
Tstat	T-statistics
U	Uranium
UNFCCC	United Nations Framework Convention on Climate Change
USEPA	United States Environmental Protection Agency
VOC	Volatile organic compounds
WHO	World Health Organization
XRF	X-ray fluorescence

CHAPTER 1:

OVERVIEW

The purpose of chapter one is to provide insight into domestic fuel burning in South Africa. The project description introduces the problem statement, scope, aim and research questions used in this research. The literature study describes the effects of anthropogenic pollution on air quality, the impacts of air pollution, the importance of domestic fuel burning and historical air quality measurement techniques.

1.1 Introduction

The amount of ambient particulate matter (PM) in South Africa is a cause of concern in terms of air quality (DEA, 2013). Atmospheric PM emerges from anthropogenic activities such as power generation, domestic wood and coal burning, agricultural and waste burning, manufacturing, vehicle emissions and aeolian dust. High ambient PM levels in certain areas can primarily be attributed to emissions from household fuel combustion, power stations and the industrial sector (Scorgie, 2012). In addition to aerosol loadings, other pollutants are also released continuously, which results in the national air quality standards being exceeded. An example of such pollutant is the high concentrations of sulphur dioxide (SO_2) found in certain areas. Greenhouse gasses (GHG) that are emitted from these practices cause impediments in the international attempt to prevent dangerous human interference of the global climate. Poor air quality conditions are detrimental to the environment and to the well-being of humans. The implications of air pollution are aggregated in heavy industrialised areas and low-income settlements (Matooane et al., 2004). Numerous studies have indicated that the respiratory problems in these poor communities may be linked to ambient PM (Jimoda, 2012). These health consequences are intensified in developing countries such as South Africa, due to high population densities, malnutrition and poverty (Wright, 2011).

It is therefore critical to research the air quality within low-income settlements and to determine all the polluting sources within the proximity of these settlements.

Previously disadvantaged and low-income population groups in South Africa are often located adjacent to industrial areas so that residents can easily commute by means of public transport or by foot to jobs (Matooane et al., 2004; DEA, 2013; Masekoameng, 2014). These areas are notorious for poor air quality due to all the manufacturing operations. Apart from industrialisation and mining operations, domestic fuel burning is an important contributor to air pollution. Low-income households rely on alternative fuels such as wood, dung, waste and coal combustion for cooking and heating purposes (Nkomo, 2005). These conducts can be ascribed to the high cost of electricity and inadequate supply of power by the government. Recent research carried out in poor communities show that 47% of the inhabitants complained about the cost of electricity, 13% about the poor quality of the delivery of electricity and 19% stated that supply is insufficient (Alastair & Mhlanga, 2013). These poor communities are thus obligated to use other fuel types as energy alternatives. In South Africa, approximately 950 000 of households use coal as energy source, which constitutes 3% of the total coal combustion that takes place (Balmer, 2007). Assessments of fuel usage in Mpumalanga revealed that 13.8% of the households used dirty fuels for electricity, 30.7% for cooking and 42.4% for heating purposes (SSA, 2011). Dirty fuels are widely used by poor communities as primary or secondary energy source, partially due to limited knowledge of the possible health impacts. The combustion of wood, dung or coal is one of the main contributors to air pollution, and the effects are amplified when used within poorly ventilated houses. Houses are therefore exposed to indoor air pollution (concentrated pollutants released during cooking or heating practices) as well as outdoor air pollution (pollutants released from various other sources such as power stations).

A variety of sources contribute to air pollution, exposing the population to air that may be harmful to their health and well-being (DEA, 2013). Chronic respiratory diseases that are caused or aggravated by air pollution are the primary causes of mortalities in South Africa (WHO, 2007; WHO, 2010;Wright et al., 2011). It is reported that children who are exposed to high levels of indoor pollution are more susceptible to respiratory infections (Barnes et al., 2011). This statement may be supported by the number of respiratory problems detected in children younger than five years of age. Children from the

Highveld area are found to suffer more from respiratory diseases than the children in less polluted areas (Wright et al., 2012). Some of the detrimental effects on health that result from domestic fuel emissions include carbon monoxide (CO) poisoning, suffocation, irritation of the eyes, nose and throat as well as asthma and Tuberculosis (TB) (Balmer, 2007). Residential fuel-burning also emits PM, with associated health problems such as premature mortality, chronic respiratory diseases and cardiovascular conditions (Ni et al., 2012, Naidoo et al., 2013). It is thus important to quantify particulate matter levels throughout South African low income communities.

The figure implies that the amount of suspended PM in South Africa increased since 1994 (Figure 1-1). Even though the data indicated an increase in ambient aerosols, this figure is misleading as the number of gauging stations increased during the same period. The new monitoring stations were constructed within polluted areas and show that the PM levels exceeded the national standards in certain areas. Despite the progress made in reducing PM_{10} levels from 2008, the amount of suspended particulates still requires effective management and mitigation strategies.

The Government implemented the National Environmental Management: Air Quality Act (NEM:AQA) (DEA, 2004) in order to prevent pollution and manage air quality effectively. This act consists of standards and requirements for achieving and promoting sustainable development. The more recent Air Quality Amendment Act states that all processes and operations that affect the air quality must obtain an atmospheric emission license. Both point and non-point sources are important for the adequate control of ambient air quality (Piketh & Burger, 2013). Determining and quantifying ambient air quality and obtaining accurate results is one of the major challenges in South Africa (DEA, 2012).

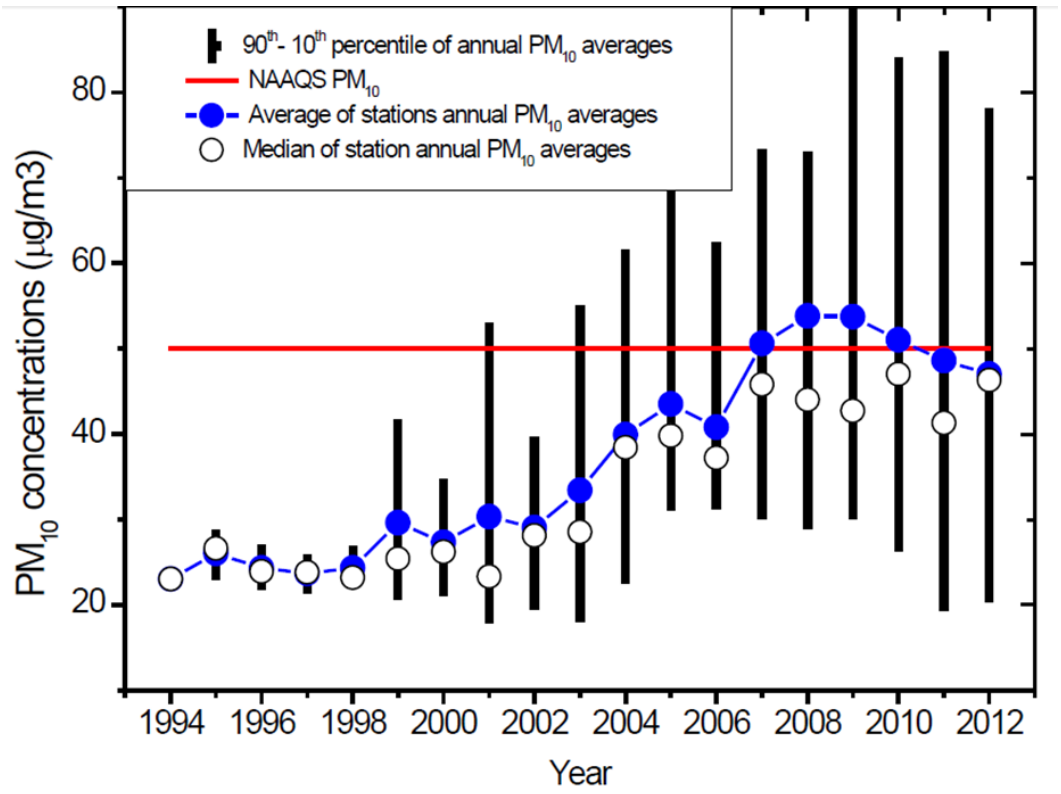

Figure 1-1: Average annual atmospheric PM_{10} concentration in South Africa from 1994 to 2012 (DEA, 2013).

The purpose of this research was to investigate the characteristics of ambient $PM_{2.5}$ (fine particulates) and PM_{10} in a low-income, coal-burning settlement in Mpumalanga. In addition to the coal-burning practices, all the other polluting sources were identified and their proportional contribution calculated. A small, low-income community, Kwadela, was selected as study site due the coal- burning practices of the community as well as the various other emitters located within a close range. Source apportionment methods were applied to identify the main polluter where both ambient PM and particulates from all the possible sources were considered.

The importance of this research is highlighted in Chapter 3 where the fuel use patterns are spatially illustrated. Households that use different fuel types were surveyed in addition to the reasons for using the specific energy alternatives. These results also illustrate the number of households affected by residential fuel-burning. This dissertation clarifies the significance of the impact of domestic fuel-burning in South Africa that needs to be considered by policy makers. Air quality policies should include all point and non-point sources in air quality management strategies to create a cleaner environment

1.2 Project description

1.2.1 Problem Statement

Air quality management and control in South Africa is complicated due to the lack of sufficient air pollution statistics and information (DEAT, 1999). The NEM:AQA was implemented in 2004 to regulate air quality, and to prevent pollution and ecological degradation (DEA, 2004). Other air quality standards include the Standard for Air Quality (SANS 1929) which aims to limit the emission of common air pollutants and the South African National Standard (SANS) 20049 for targeting vehicle emissions (South Africa, 2008; RSA, 2011). Last year, the Air Quality Amendment Act was issued to provide for the consequences of illegal emissions, along with an improved pollution prevention plan for the evaluation, monitoring and reporting of pollution. Even though legislation has been developed, insufficient data and measurements hampered its implementation. These regulations target certain polluters in order to improve air quality, but not all sources are considered. All the possible polluting sources should therefore be identified and included in national management plans.

For the effective monitoring of air quality, it is necessary to distinguish between indoor and outdoor pollution and to identify the contributing sources of each (DEA, 2013). Indoor gasses may be linked to domestic fuel-burning practices which affect the health of the household members. Outdoor air pollution, on the other hand, is associated with a higher variety of polluting sources. These outdoor emissions affect more people with health impacts that vary according emission levels and chemical components released.

Mines and power stations are notorious for contributing to the amount of suspended particles in the atmosphere. However, certain areas are polluted mainly as a result of domestic fuel-burning emissions (Annegarn et al., 1998; Engelbrecht et al., 2001; Engelbrecht et al., 2002; Mdluli et al., 2005; Worobeic et al., 2011 and Piketh & Burger, 2013). Low-income settlements are dependent on energy alternatives for cooking and heating purposes. The fuels that are commonly burnt include wood, paraffin, low-grade coal, gas and animal dung. Other sources that contribute to outdoor pollution include vehicle emissions, construction operations, agricultural activities, waste burning and fugitive dust (Obioh et al., 2013). These emissions have a negative impact on the respiratory health of the population. Health impacts are intensified within low-income

communities. These poor communities commonly lack proper health care and the necessary education to understand the severity of air pollution. It is therefore important to identify the root cause of pollution in order to improve the standard of living of the community.

The sources of pollution differ from one region to the other in South Africa and the number of emitters and pollution levels also vary. In order to fully understand air pollution and to pinpoint a contributor of respiratory health problems in poor communities, a quantification method was applied. This effective measurement technique was used to calculate the exact contribution of each source. The lack of measuring air pollutants and attributing them to their resources are an issue for mitigating air pollution consequences. This method could provide original statistical data and source profile information as a solution in air quality disputes.

1.2.2 Research Scope

This research focused on the practice and importance of domestic fuel- burning in South Africa to local air pollution. Within this scope, two investigations were conducted. Firstly, the energy alternatives used by poor communities were surveyed. Secondly, the ambient air quality of a poor community was analysed to illustrate the impact of PM released during household fuel combustion.

Fuel burning settlements were identified from census (2011) data sets and presented at a small area layer (SAL) scale. The demographic census information was assessed to determine the motives behind fuel choice. The number of possible affected people was consequently calculated for an upper case limit and a lower case limit.

Another method to describe fuel usage patterns involved the quantification of different source emissions. Even though a number of sources release atmospheric pollutants, domestic fuel burning is notorious for being the worst polluter in certain areas. In order to identify the root cause of air quality problems in a low income community, source apportionment methods were applied. Ambient particulates were sampled in the Kwadela community and analysed chemically. The PM samples were divided into two categories according to size: coarse fractions and fine fractions. The $PM_{2.5}$ and PM_{10}

samples were collected twice a day throughout a period of two weeks. These ambient samples were analysed chemically and statistical calculations were applied. The chemical compositions from source emissions were obtained from the United States Environmental Protection Agency (USEPA) database and previous research articles. Variations in the collected samples were explained by differences in source profiles and meteorological conditions. This study dealt with the meteorological conditions such as rain, wind speed and direction as well as variation in temperature that could have affected the samples.

1.2.3 Research Objectives

Primary Objective:

The aim of this research is to evaluate the importance of domestic coal combustion to poor air quality in low income settlements.

Research Questions:

- What are the domestic fuel use patterns throughout South Africa?
- How many people are affected by residential fuel burning emissions?
- What are the characteristics of ambient particulate matter in a low income settlement such as Kwadela?
- What sources contribute to air pollution in Kwadela?

LITERATURE REVIEW

1.3 Particulate matter characteristic

The atmosphere is a compilation of gaseous substances and aerosol particles (Piketh, 2000). Aerosols are commonly defined as atmospheric suspensions of liquid or solid particles that were released through either natural or anthropogenic sources. Particles could be classified into two groups in accordance to their source of origin. Primary particles are a result of direct emissions from natural sources. Dust, salt, soot and pollutants emitted from industrial plants or volcanoes are all examples of primary particles (Tyson & Preston-Whyte, 2000). Chemical reactions and condensation of water vapours are processes that give rise to secondary particles. These aerosols may form through inorganic gas conversions such as sulphur(S), ammonia salts (NH_3), nitrogen (N) or phosphates (PO_x). The nature, origin and transport of particles are difficult to define due to the unpredictability of their occurrences. Aerosol particles are ubiquitous and characterized by their altering properties. These include composition, optical properties, chemical properties and shape. These ambient PM may be used as references for linking the properties to their source of origin. For the purpose of this research the chemical composition and size distribution of suspended particles were used in source apportionment models.

1.3.1 Composition of particulates

Size and chemical composition influence the number of particles at a given time and point of space (Friedlander, 1970). Particulates may be considered as toxins when they consist of various toxic chemicals (Kelly & Fussell, 2012). The chemical elements associated with PM can be found internally or on the particulate surface, which illustrates its complexity. $PM_{2.5}$ is mainly compiled of soluble components such as sulphate (SO_4^{2-}), nitrates (NO_3^-) and ammonium (NH_4^+) along with the organic carbon (OC) and elemental carbon (EC) insolubles (Aneja et al., 2006). Chemicals are commonly found in the atmosphere as a result of anthropogenic activities (Smeets et al., 2000). It is plausible that the chemical constituents in PM may illustrate health and environmental impacts better than gravimetric results (Stanek et al., 2011). The common chemicals found in PM_{10} and $PM_{2.5}$ are illustrated in Figure 1-2:

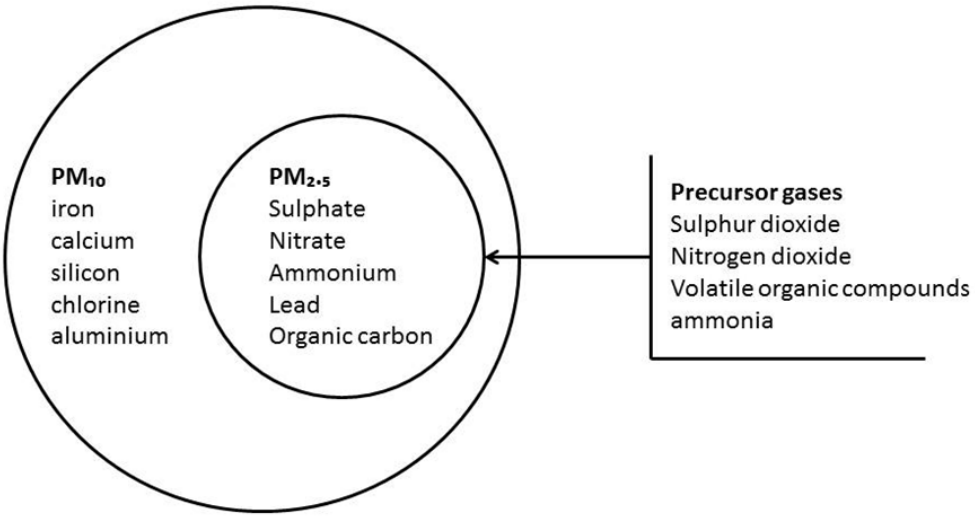

Figure 1-2: The common chemicals found in ambient particulate matter (Government of Canada, 2012).

The chemical composition of particles may be used to link ambient samples to their sources because of the consistency of the major chemical components of the sources (Friedlander, 1970, Aneja et al., 2006).The composition of ambient samples corresponds to the chemical composition of source emissions (Table 1). The chemical composition of ambient $PM_{2.5}$ and PM_{10} can be anticipated with a ($>^{\pm}10\%$) precision, (Chow, 1995).

Assigning ambient PM to their sources is possible, because every source can be distinguished by its unique chemical composition (Table1-1) (Watson et al., 1997). The source profiles were obtained from the EPA database or local sampling (see Chapter 4). All the sources that are included in this study will be discussed later in this chapter. The polluting source profiles were used to point out the main cause ambient PM in the selected study site. The receptor modelling procedure was applied to find the dominant source profile. A more simplified method for determining source contributions is gravimetric investigation.

Table 1-1: Chemical compositions of PM in source emissions (Watson et al., 1997).

Source type	Dominant particle size	Chemical abundances in percent mass			
		<0.1%	0.1 to 1%	1 to 10%	>10%
Paved road dust	Coarse	Cr, Sr, Pb, Zr	$SO_4^=$, Na^+, K^+, P, S, Cl, Mn, Zn, Ba, Ti	Elemental Carbon (EC), Al, K, Ca, Fe	Organic Carbon(OC), Si
Unpaved Road dust	Coarse	NO_3^-, NH_4^+, P, Zn, Sr, Ba	$SO_4^=$, Na^+, K^+, P, S, Cl, Mn, Ba,Ti	OC, Al, K, Ca, Fe	Si
Agricultural soil	Coarse	NO_3, NH_4^+, Cr, Zn, Sr	$SO_4^=$, Na^+, K^+, S, Cl, Mn, Ba, Ti	OC, Al, K, Ca, Fe	Si
Motor vehicle	Fine	Cr, Ni, Y	NH_4^+, Si, Cl, Al, Si, P, Ca, Mn, Fe, Zn, Br, Pb	Cl^-, NO_3^-, $SO_4^=$, NH_4^+, S	OC, EC
Vegetation burning	Fine	Ca, Mn, Fe, Zn, Br, Rb, Pb	NO_3^-, $SO_4^=$, NH_4^+, Na^+, S	Cl^-, K^+, Cl, K	OC, EC
Residential Oil Combustion	Fine	K^+, OC, Cl, Ti, Cr, Co, Ga, Se	NH_4^+, Na^+, Zn, Fe, Si	V, OC, EC, Ni	S, $SO_4^=$
Coal-Fired Boiler	Fine	Cl, Cr, Mn, Ga, As, Se, Br, Rb, Zr	NH_4^+, P, K, Ti, V, Ni, Zn, Sr, Ba, Pb	$SO_4^=$, OC, EC, Al, S, Ca, Fe	Si
Oil fired power plant	Fine	V, Ni, Se, As, Br, Ba	Al, Si, P, K, Zn	NH_4^+, OC, EC, Na, Ca, Pb	S, $SO_4^=$
Smelter Fine	Fine	V, Mn, Sb, Cr, Ti	Cd, Zn, Mg, Na, Ca, K, Se	Fe, Cu, As, Pb	S
Marine	Fine and coarse	Ti, V, Ni, Sr, Zr, Pd, Ag, Sn, Sb, Pb	Al, Si, K, Ca, Fe, Cu, Zn, Ba, La	NO_3^-, $SO_4^=$, OC, EC	Cl^-, Na^+, Na, Cl

Particle size distributions and the amount of total suspended particles (TSP) are the most popular characteristics used in studies because of the high expenses involved in chemical analyses and the challenges involved in measurement (Chow, 1995). Gravimetric analyses are used to determine the amount of suspended particles in an area (Walton et al., 2013), and were used to set national legislative standards and limits. In this research it is important to apply particle settings by using gravimetric measurements. This information will serve as a guideline of the distance over which particles could remain suspended. Large particle sizes indicate nearby source contributions whereas smaller particle sizes could be linked to sources at a further distance. The time that aerosols remain suspended depend on their size.

Large particles may remain in the atmosphere for a few days while the duration for small particles varies from weeks up to several years in the troposphere (Tyson and Preston-Whyte, 2000). Smaller particles tend to diffuse or remain suspended while larger particles have a more prominent gravimetric setting (Montoya, 2013). Smaller particles may travel further than the larger particles and sources closer to the sampling site will contribute more coarse particles than sources located further. Airborne particles change in size according to interactions with other particles and substances. These particulates are typically measured by equivalent sphere diameter, due to their 3-dimensional properties. The size distribution of aerosols range from nucleation (0.01 µm – 0.1 µm) to accumulation (0.1 µm – 1 µm) or coarse particles (≥ 2-3 µm) (Figure 1-3). Particulate matter is regulated according to two categories: coarse particles with a diameter between 2.5 µm – 10 µm and fine particles with a smaller diameter than 2.5 µm (Montoya, 2013).

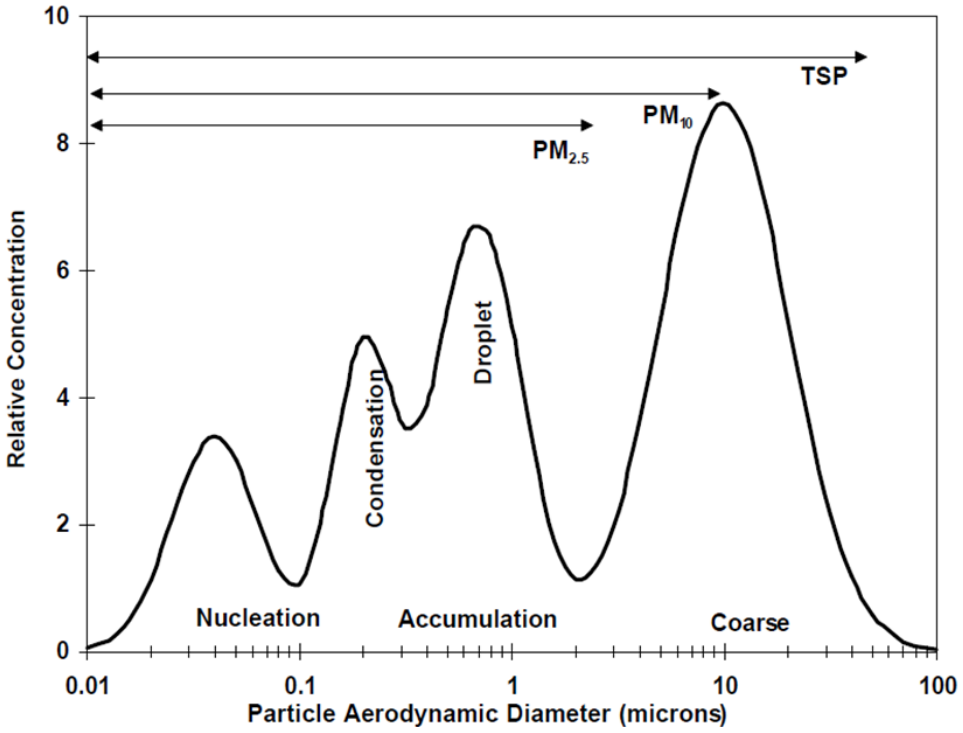

Figure 1-3: Size distributions in ambient air (Chow, 1995).

The nucleation range refers to fine particles that originate from source combustion, gas-to-particle conversions and the condensation of unstable species or ground dust (Chow & Watson, 1994; Watson et al., 1997). The particle size of the accumulation mode is between 0.1 µm ≥ PM ≤ 2.5 µm and is caused by combustion processes and smog. Coarse particles are mainly encrusted to their sizes due to activities such as road dust and drilling (Graham, 2004). These aerosols have a short residence time in the atmosphere compared to gaseous substances, but still affect the global climate. Receptor samples are a representation of transport processes that occurred and of the gravimetric setting of particles. Due to the highly variable nature of particle suspension in the atmosphere, transport processes over South Africa must be taken into consideration, (see Piketh, 2000). In order to understand aerosol occurrences, the deposition rate must be explained in terms of the typical transport processes.

1.3.2 Transportation systems of aerosols

The dominant horizontal air mass movement in South Africa occurs in an anti-cyclonic circulation (Preston-Whyte and Tyson, 1989). The anti-cyclonic movements are more prevalent in the winter when particles may remain suspended for a number of days days. The main winter circulation occurs from the Indian Ocean to south-eastern Africa (Garstang et al., 1996; Annegarn et al., 2002). In order to explain aerosol movement in South Africa, the Highveld regional level could be used as a baseline (Freiman & Piketh, 2002). In addition to the prevalent anticyclonic circulation, the transport patterns in the Highveld may be defined by westerly disturbances that occur approximately 20-40% of the year and easterly disturbances that occur 30-50-% per year. Industrial aerosols and trace gasses are transported to the Highveld from a southerly direction (D' Abreton and Tyson, 1996; Piketh et al., 1998; Freiman and Piketh, 2002). The aerosol and trace gas dispersal from the Highveld to other parts of South Africa influences the different air masses (Freiman & Piketh, 2002).

In order to manage the air quality of a region efficiently, the amount of particulate matter must be regulated. The chemical compositions of ambient particles may be used to determine which sources contribute to air pollution, and to control the emissions. Gravimetric analyses are used to determine the amount of PM in the atmosphere at a given time. In order to enforce control and mitigation measures, the dispersal and transport processes of the specified area must be well-defined. This section argues that particle characteristics may be used effectively for air quality monitoring and analyses.

1.4 The link between particulate matter and the energy sector

South Africa joined the United Nations Framework Convention on Climate Change (UNFCCC) in August 1997 with the aim of preventing anthropogenic sources influencing the global climate system (DEA, 2014). South Africa was recognized as the only African country that, while it produces energy on a regional scale in Mpumalanga, the industrial sector is classified as one of the largest in the world (Josipovic et al., 2011). The energy sector emits carbon dioxide (CO_2) and methane (CH_4) pollutants from oil and natural operations, CO_2, CH_4 and nitrous oxide (N_2O) from spontaneous combustions, CH_4

from abandoned mines and CO_2, CH_4 and N_2O from water-borne navigation. During the evaluation of the different polluting sources it was detected that the fossil fuel category was responsible for the majority of pollutants (Figure 1-4).

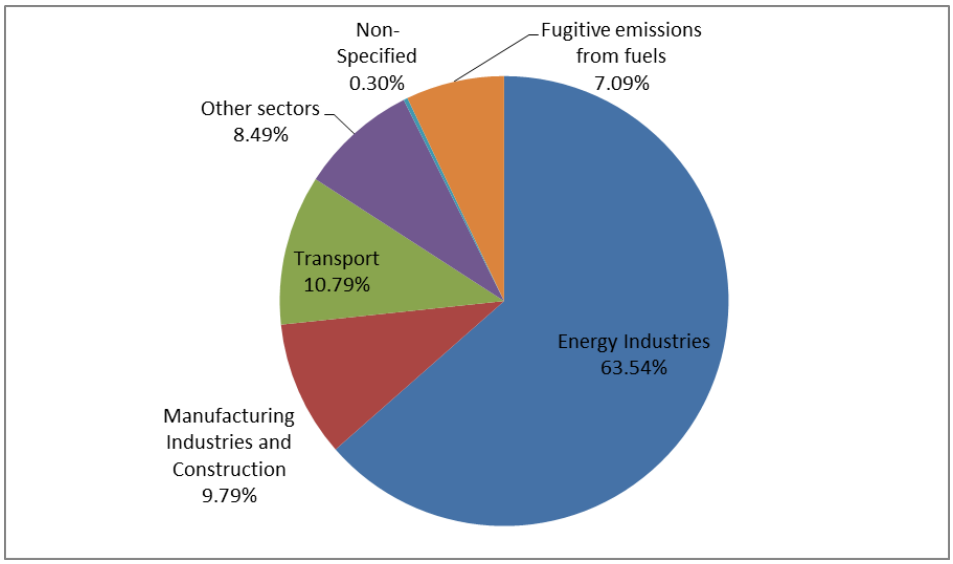

Figure 1-4: The compilation of the average source emissions in the energy sector from 2000 - 2010 (DEA, 2014).

1.4.1 Industrial emissions of South Africa

The concern over controlling emissions from fossil fuel combustion has increased ever since global warming became of serious international concern and air quality management commenced. Controlling emissions requires effective air management which involves establishing limitations and standards as well as implementing mitigation strategies. South Africa must prevent pollution as the country is highly susceptible to droughts and subsequent reduced crops, floods, increased growth of invasive species and disease outbreaks caused by global warming. The biggest source, namely 60% of GHG emissions in South Africa, is the energy sector, (Inglesi-Lotz and Blignaut, 2011). From this, the production of synthetic fuels is accountable for 84% of the energy sector's emissions (Winkler & Marquand, 2009). Apart from the significant impact of synthetic fuel production, the burning of coal is a major factor in the excitation of energy. Throughout the country's history, the energy sector has been highly dependent on coal combustion to provide for industrial ambitions and human needs (Figure 1-5).

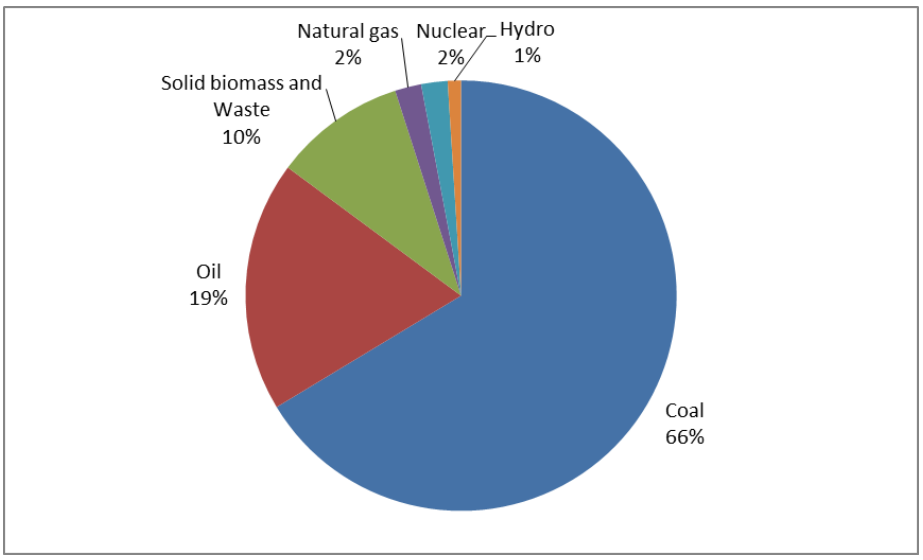

Figure 1-5: Primary energy supply in South Africa 2010 (U.S. EIA, 2013).

The energy sector is responsible for a variety of pollutants that are released into the atmosphere of which coal combustion is the largest contributor. This reliance on coal sources can be seen in the total GHG pollutants. Coal combustion releases sulphur oxides (SO_x), nitrogen oxides (NO_x), CO and volatile organic compounds (VOC) into the atmosphere and conveyer belts from these mining activities are responsible for discharging large amounts of fine coal dust. Carbon dioxide loads have increased by 41% from 1994 – 2008 and this can be ascribed to energy supplied by coal-fired systems (Meyer & Odeku, 2009). Power stations are the main cause of atmospheric SO_x emissions. Sulphur dioxide (SO_2) aerosols is notorious for contributing to global warming by acting as a radiative forcer (Emberson et al., 2012). The mining industry on the other hand is responsible for emitting aerosols consisting out of major pollutants such as SO_x, NO_x, CO, VOCs, CH_4, lead (Pb) and other hazardous metallic substances (Mangena & Brent, 2006). All mining operations contribute to the amount of PM in the atmosphere as they involve earth-moving processes.

Particular matter occurrences in the atmosphere contribute to the amount of GHG (Brasseur et al., 2003). PM may cause the warming of the atmosphere because it has heterogeneous properties which influence the radiative budget (Lydia, 2010). Particulates have a direct impact on global climate by either scattering or absorbing shortwave and long-wave radiation and by forcing radiation back into space by

reflection. Indirect impacts of aerosols refer to the influence particles have on cloud condensation nuclei. These impacts affect cloud radiation, cloud lifetime and the liquid-water balance cycle (Lydia, 2010). The potential climate outcome of aerosols and precursors may be illustrated through radiative forcing (Figure 1-6). These pollutants have varied impacts on the climate due to differences in physical and chemical properties. Some particulates cause negative radiative forcing whereas others, such as black carbon, cause the warming of the atmosphere.

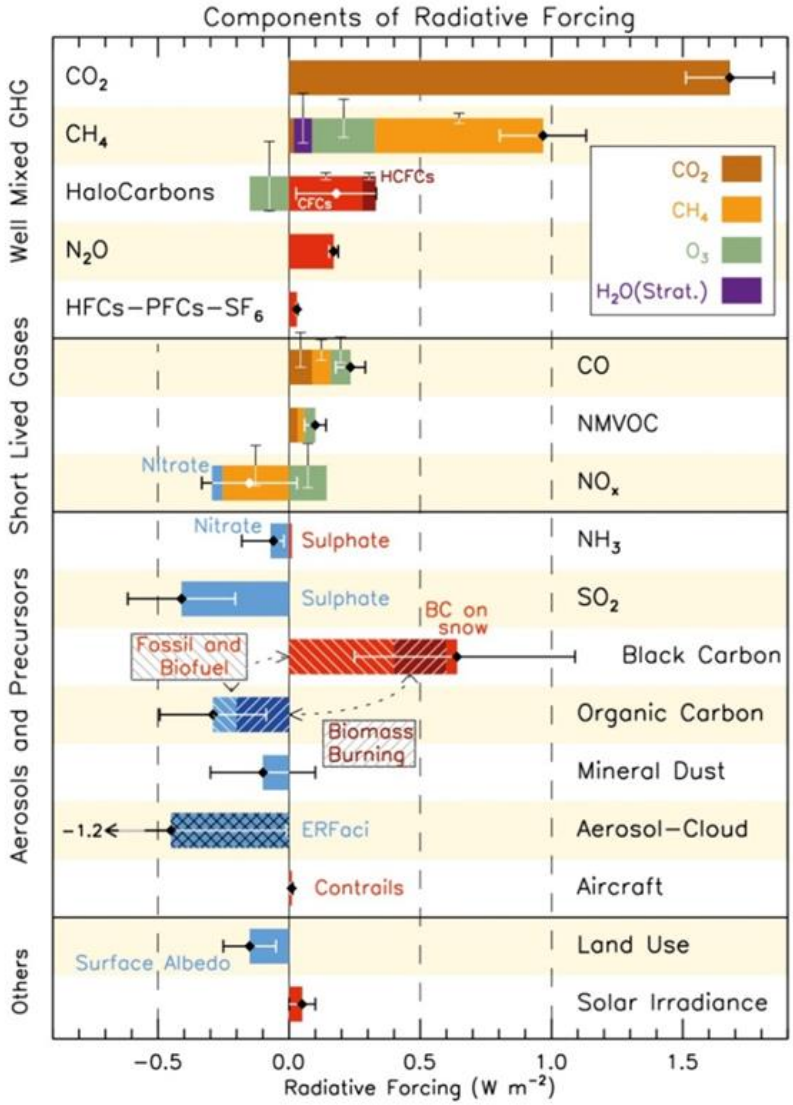

Figure 1-6: Anthropogenic forcing (Myhre et al., 2013).

The emissions of PM raise concerns because South Africa has high levels of suspended aerosols in particular areas (Laakso et al., 2008; Vakkari et al., 2011;

Venter, 2011). The high number of PM in the atmosphere contributes to global warming and may be ascribed partially to the industrial activities of the energy sector. It is important to note that even though the energy sector is one of the main causes of air pollution, there are still a number of other sources that should be taken into consideration.

1.5 Health impacts from particulate matter pollution

The burden of health-related problems caused by ambient aerosols is higher in developing countries. PM, like any other pollutant found in the atmosphere, is linked to a series of health impacts. It is vital to focus the research scope of health-related problems on a local level (Kaonga & Ebenso, 2011). The local scope involves the low-income communities that are dependent on solid fuels, but are affected by the air pollution. Various studies have indicated a linear relationship between PM concentrations and respiratory health problems (Jimoda, 2012). Communities with high suspended aerosol levels have reported more respiratory and cardiovascular problems. Studies conducted in these communities show that more cases of hospitalisation occurred in areas with higher ambient aerosol concentrations (Dockery et al., 1993; Künzli et al., 2000; Brunekreef & Holgate, 2002; Terblanche, 2009, Jimoda, 2012). An increased hospitalization rate of 0.45% to 4.7% was calculated for a 10 µg/m^3 increase in coarse PM (Health Canada, 1998; Terblanche, 2009). This rate was calculated for the general level of coarse ambient particulates found in urban areas which ranged between 25 µg/m^3 to 50 µg/m^3. Low-income communities are more vulnerable to respiratory infections from suspended aerosols, but are, at the same time, branded as main contributors, due to their cooking and heating practices. PM is commonly discharged through a variety of anthropogenic activities with life-threatening consequences. Understanding the effects of PM and illustrating the incidences in South Africa is an important aspect towards improving the living standards of its residents.

1.5.1 Causes of health problems

The health effects associated with particle matter are proportional to particle characteristics such as size and chemical composition. It is also important to take the frequency, period and degree of PM exposure into account (Terblanche, 2009). The danger of aerosols is influenced by the afore-mentioned factors such as particle size, chemical composition and the concentration of the aerosols that are inhaled. PM is made up of various chemical substances, each with its own health impacts. Thus, the chemicals found in certain ambient particles may worsen the effects on human health.

Table 1-2: Constituents of PM's effect on health (Jimoda, 2012).

Heavy metal	**Min. risk level**	**Toxicity effects**
Lead	Blood lead levels below 10 micrograms per decilitre of blood.	Impairment of neurological development, suppression of the haematological system (anaemia), kidney failure, immunosuppression etc.
Mercury	Below 10 microgram per decilitre of blood; oral reference dose (Rfd) 4 mg/kg/day.	Gastrointestinal and respiratory tract irritation, renal failure, neurotoxic.
Cadmium	Below 1 microgram per decilitre of blood.	Local irritation of the lungs and gastrointestinal tract, kidney damage and abnormalities of skeletal system.
Arsenic	Oral exposure of 0.0003 mg/kg/day.	Inflammation of the liver, peripheral nerve damage - neuropathy, cancer of the liver, skin and lungs, irritation of the upper respiratory system pharyngitis, laryngitis, rhinitis, anaemia, cardiovascular diseases.

Table 1-2 summarises the heavy metals found in aerosols with serious toxic effects on the human body. Apart from the chemical constituents in PM, the duration of exposure and the particle sizes should also be taken into account.

The aerodynamic properties of particles - such as shape and density - determine the likelihood of it being deposited in the lungs (Kaonga & Ebenso, 2011). Smaller particles penetrate deeper into the pulmonary system (the gas-exchange regions) whereas larger particles (PM_{10}) could settle in the bronchi and lungs. Small particles are more dangerous due to their ability to penetrate deeper and the subsequent increase in the risk of interference with cellular activity (Benson, 2012). The size of particulates determines the inhalation fraction and the depth of penetration (Figure 1-7).

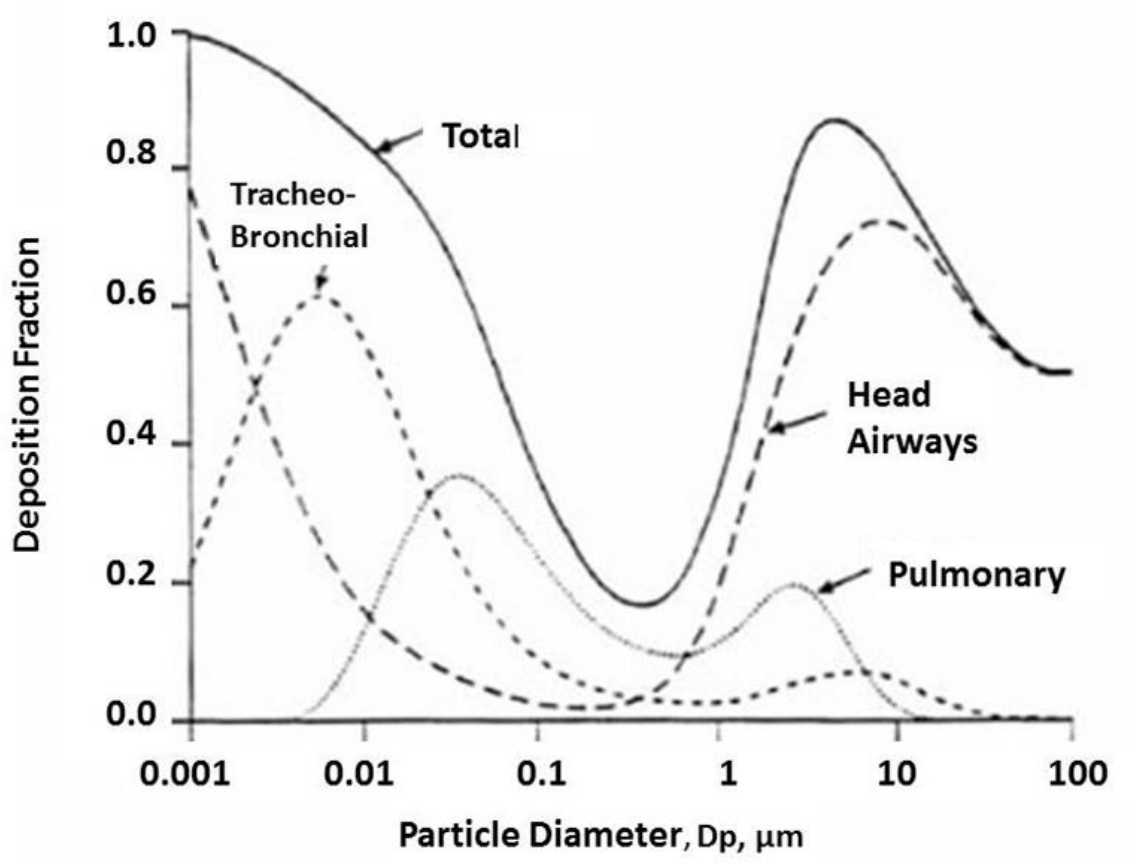

Figure 1-7: The influence of particle size and the deposition fraction in humans (Benson, 2012).

Particles smaller than 0.1 µm undergo random movements (Brownian Motion) in the arteries which results in chronic respiratory problems (Figure 1-8) (Jimoda, 2012). $PM_{2.5}$ cause vascular inflammations and atherosclerosis by infiltrating the arteries, (Pope & Burnett, 2002). Inhalation of $PM_{2.5}$ is extremely dangerous and even short-term exposure can lead to cardiovascular diseases (Brown et al., 1950; Turco, 1971; Boubel et al., 1994; Pope et al., 2002; Bradhawar et al., 2004; *Panda* et al., *2013*). PM_{10} are less invasive, but may still cause critical chronic diseases. Inhalation of PM may cause asthma, immuno-suppression, nausea, cardiovascular infections, lung cancer, fibrotic lung diseases and may lead to premature deaths (Ogola et al., 2001; Pope & Burnett, 2002; Kampa & Castanas, 2008; Kaonga & Ebenso, 2011). Other effects include insulin resistance, oxidative stress, vascular and visceral inflammation, alteration of vasomotor tone, adiposity and atherosclerosis, (Xu et al., 2011). Both fine ($PM_{2.5}$) and coarse particles (PM_{10}) may contribute to lung cancer pathogenesis (Xu et al., 2011). The exposure to PM may induce blood platelets which could lead to the incapacity to restore vessel damage (Kallaf, 2011). The result will then be arterial thrombosis. Thus, the longer the exposure time and the smaller the particle size, the worse the effect on human health. It is important to note that exposure – response characteristics can vary between the different pollutants. Particular adverse health impacts are associated with long-term exposure (months to years) and other with short-term exposure (hours to days (Cairncross et al., 2007).

Health problems are more prevalent in low-income communities as diseases are more likely to spread at a faster rate because health care is limited. Household air pollution that is caused from cooking with alternative energy sources is responsible for over 4 million premature deaths (WHO, 2014). Inhalation of PM (soot) causes more than 50% of the premature deaths in South Africa's population. An estimated 17% of lung cancer deaths are a result of household air pollution and the use of solid fuels. Household coal smoke contains a dangerous carcinogen and wood smoke is mutagenic (Norman et al., 2007). Poorly manufactured stoves used for cooking are responsible for significant indoor emission rates. More women and children are exposed to these emission rates due to poor ventilation within houses. People who are exposed to high PM_{10} concentrations over a 24-hour period show an increased mortality risk of 8% for every 50 µg/m^3, (USEPA, 2004; Terblanche 2009). It is estimated that the inhalation of an average of 50 µg/m^3 PM may mean that 1 – 8 people out of 100 may have an increased

mortality risk: An air pollution index (API) has been applied widely in order to assess the impacts of the ambient concentrations on health more efficiently (Cairncross et al., 2007).

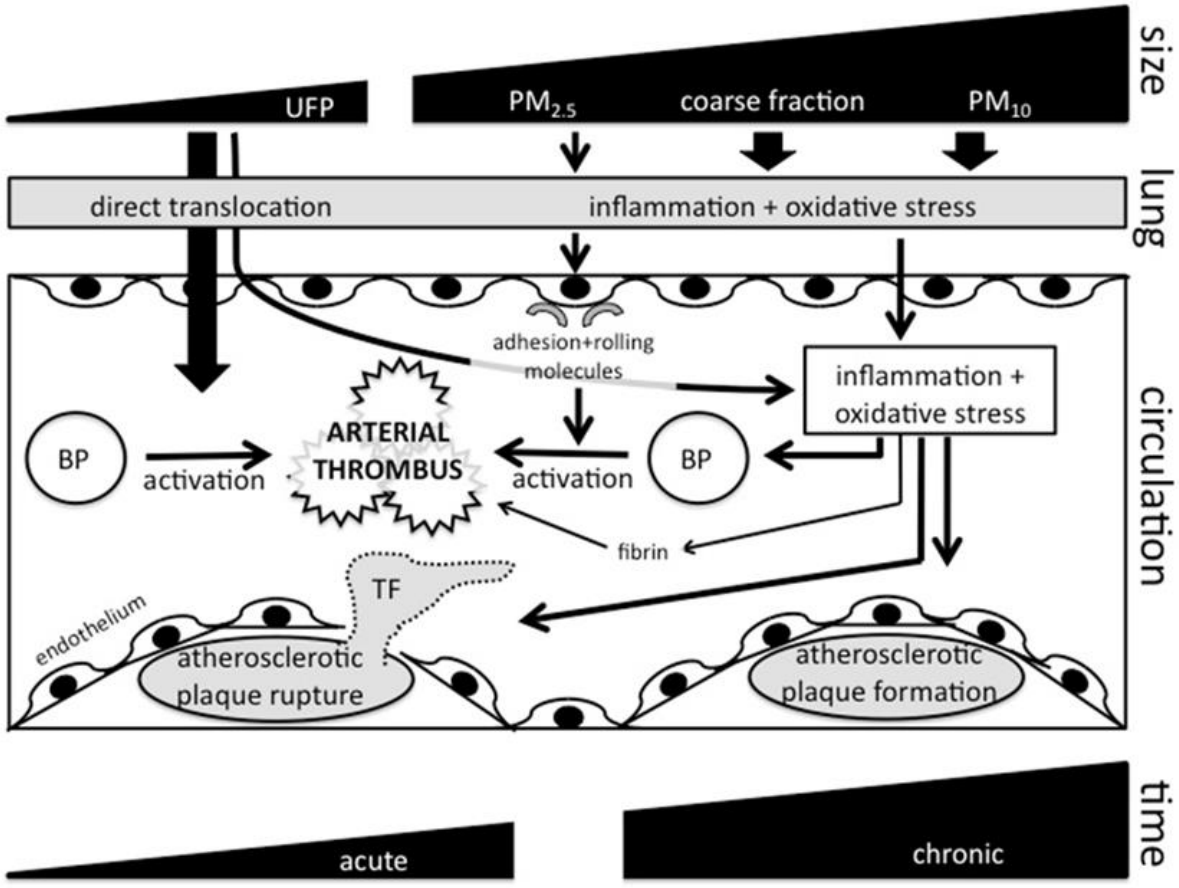

Figure 1-8: Indicating the time factor and particle sizes which results in arterial thrombosis (Kallaf, 2011).

The health impact of exposure to PM is critically important and should be addressed. Low-income communities are more vulnerable to respiratory diseases because they rely more on alternative energy sources. In order to determine whether indoor air pollution is the main cause of respiratory illnesses and premature deaths in low-income communities, the sources that emit aerosols must be identified and quantified.

1.6 Ambient particulate matter of low income settlements

An estimated 2.4 billion people use ‘dirty’ fuels such as wood, coal, gas, paraffin and animal dung as a primary energy source for cooking and heating purposes (IEA, 2002; Heltberg, 2005). In poor countries, indoor pollution leads to critical health problems owing to household fuel combustions (Begum et al., 2009; Zhou et al., 2011). A large proportion of poor populations are continuously exposed to these pollutants over long periods. Fuel combustion typically occurs in poorly ventilated houses, which intensifies the concentration of pollutants. Concentrations of pollutants can reach levels higher than 10-20 times the safety limits (Heltberg, 2005). Other issues to consider are the low efficiency of the fuels burnt, typically a low-grade coal, as well as the traditional stoves that are used. These fuel-burning practices result in serious cases of indoor air pollution as well as outdoor pollution. Understanding residential fuel-burning practices is crucial for setting policies to combat the indoor air quality issue.

1.6.1 The impacts of domestic fuel burning in developing countries

Indoor air pollution is notorious for causing serious health problems in developing countries (Bruce et al., 2000; Ezzati and Kammen, 2001; Mekonnen and Köhlin, 2009). An estimated 1.5 million premature deaths could be ascribed to indoor air pollution (IEA, 2006; Mekonnen and Köhlin, 2009). Indoor air pollution were responsible for more deaths than any other infectious diseases between the years 1997 and 1999 (Ezzati and Kammen, 2001). Indoor air pollution is the main cause for infant fatalities in developing countries (Begum et al., 2009). Other health impacts that were identified were low birth weight, higher rates of perinatal mortalities and the occurrence of cataracts (Bruce et al., 2000).

Solid fuels are the main source of energy for the Chinese population (Zhang et al., 2007). According to analyses conducted in China, approximately 80% of the rural population use biomass as a household energy source. Ninety percent of the rural Sub-Saharan African population use solid fuels as the main energy source (Zhou et al., 2014). In urban areas an estimated 75% of the population use solid fuels as the primary

energy source. Issues caused by residential fuel-burning in India worsened over time (Gupta et al., 2006).

Domestic fuel-burning is the main source of indoor air pollution in China and contributes to numerous health problems. The World Health Organization (WHO) reported in 2007 an estimated 420 000 premature deaths as a result of indoor air pollution (Zhang & Smith, 2007). The main health consequences include illnesses such as lung cancer, lung function reduction, poisonous coal endemics, CO poisoning, weakening of the immune system and chronic pulmonary diseases. More than 3 million deaths resulted from these fuel-burning practices in Sub-Saharan Africa. A study in Guatemala showed effects such as acute respiratory illnesses, pulmonary diseases, cancer and eye problems (Heltberg, 2005). Outdoor pollution that results from smoke vented through chimneys is reported to influence human productivity. A domestic fuel-burning study in Bangladesh reported that women and infants generally experience more serious side-effects from indoor air pollution as they tend to spend more time indoors, cooking in areas where pollutants are inhaled for long periods of time (Begum et al., 2009). Typical pollutants inhaled are products of incomplete combustion, which include CO, NO_2 and PM (Zhang et al., 2007). The poor communities' exposure levels are very high and pollution levels often exceed ambient air quality standards.

Eighty percent of the total global exposure to suspended particles occurs indoors. It is of utmost importance to assess all aspects regarding domestic fuel use (Ezzati and Kammen, 2001). Due to the high exposure levels and the number of people affected, research on fuel-use determinants was undertaken.

1.6.2 The motives behind residential fuel choice

Previous studies linked fuel choices to income levels through the 'energy ladder concept'. The energy ladder model assumes that fuel users will move to more sophisticated fuel types as their monthly household income increases (Heltberg, 2005). This concept implies that one fuel type is replaced by another, thus the transition from one type of fuel to the next. Fuel types were arranged according to household preferences and based on physical characteristics in the energy ladder model (Hiemstra-van der Horst and Horvorka, 2008; Van der Kroon et al., 2013). This concept

consists of three phases where a user moves from one phase to the next in relation to increases in incomes (Figure 1-9). Households will change from biomass fuels such as firewood, agricultural waste and animal waste to transition fuels such as charcoal, kerosene and coal with an increase in income. The last phase includes fuels such as liquefied petroleum gas (LPG), electricity and bio-fuels, which are more costly, but have a higher efficiency, cause less pollution and require less labour for collection (Masera et al., 2000; Van der Kroon et al., 2013). The usage of LPG is described as a clean energy fuel and effective replacement of dirty fuels (Kojima, 2011).

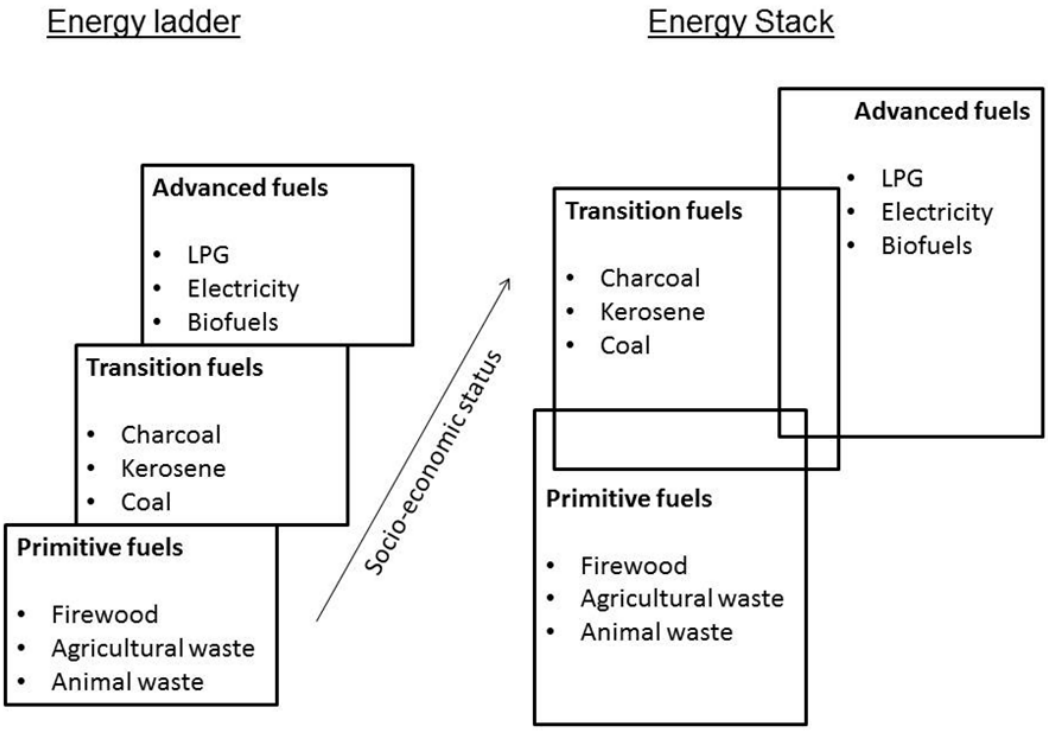

Figure 1-9: The energy ladder and energy stack model in the transition process (Van der Kroon, 2013).

The energy ladder concept suggested that households move from one fuel type to another fuel type, but recent studies showed that households use a mixture of fuels (Zhang & Smit, 2007; Mekonnen, 2009). Multiple studies illustrated that fuel use can't be classified in a linear chain. Fuel use should rather be explained by the concept of fuel-stacking (Leach, 1992; Davis, 1998; Karekezi and Majoro, 2002; Campbell et al., 2003; Brouer and Falcoa, 2004; Heltberg, 2004; Martins, 2005; Arnold et al., 2006; Van der Kroon, 2013). This concept shows that households tend to use multiple fuels at the same time, and tend to change their fuel use portfolios with an increase in income

(Masera et al., 2000). The basic fuel type will still be used, but cleaner, more efficient fuel types will be added to the household's energy consumption file.

Other aspects should also be considered when researching the motives behind the consumption of a certain fuel type (Heltberg, 2005). This includes the fact that choices depend on a variety of factors, such as income levels, preferences and the purpose of the fuel type. These determinants include fuel resource availability, cost of fuel, opportunity costs, type of household, season, electricity supply, gender of the head of the household, number of people living in a household, age of the main household member, the purpose of combustion, time-frame needed for cooking, level of education, and traditional lifestyles (Heltberg, 2005; Mekonnen, 2009; Ogwuche, 2013). A study of the choices dictating the fuel type used in Bangladesh showed that seasonal and economic influences were equally important (Begum et al., 2009). The fuel choices in developing countries are regulated by different factors, with varying scales of influence.

Table 1-3 is a summary of the numerous investigations conducted in developing countries on fuel-use determinants. The first determining factor of fuel type used was the income level of a household. Low-income households in Ouagadougou tended to rely on wood as the primary energy source (Ouedraogo, 2005). Thus, poor users relied on a more accessible and easily collectable alternative. Research on the impacts of higher-income levels in Ethiopia found that the number of fuel types used increased (Mekonnen, 2009). In contrast with the energy ladder model, households changed the quantity of fuel types instead of switching to a more efficient energy source. This finding suggests that people use fuel types based on their habits, the type of appliance in possession or the type of food being cooked. An economical aspect, such as household income, was the dominant factor influencing fuel choice in the Himalayas (Mustaq, 2014). In Nigeria, an increase in income levels did not influence a household's use of charcoal (Ogwuche, 2013). The reason for this is that poor communities mostly rely on charcoal because sources of wood are further away and in richer households charcoal competes with modern fuels for cooking purposes. Households in Malawi changed their fuel type to a more convenient, efficient and cleaner type of fuel with an increase in income (Falcão, 2008). According to these investigations income levels may be used as the primary indicator of fuel choice, but all other determining elements should also be considered.

Table 1-3: Determinants of fuel choice in developing countries.

Determinant	Influence on fuel choice	Research area	Reference
Income levels	• Low incomes are linked to wood usage	Ouagadougou	Ouedraogo (2005)
	• Higher income levels result in higher quantities of fuels used	Ethiopia	Mekonnen (2009)
	• No Influence on charcoal choice	Nigeria	Ogwuche (2013)
	• The most influencing factor	Himalayas	Mustaq (2014)
	• Will use a more convenient clean fuel type with an increase in income	Malawi	Falcão (2008)
Level of education	• Users switch to LPG with higher levels in education	Guatemala	Heltberg (2005)
	• Formal education were linked to LPG and charcoal usage	Ouagadougou	Ouedraogo (2005)
	• Educated members use non-solid fuels	Ethiopia	Mekonnen (2009)
	• Have no influence on fuel type preferred	Himalayas	Mustaq (2013)
	• Have the smallest impact on choice	Nigeria	Ogwuche (2013)
Gender of the household head	• Females are associated time-consuming fuels	Guatemala	Heltberg (2005)
	• Females use either solid fuels, or a mixture of solid and non-solid fuels	Ethiopia	Mekonnen (2009)
	• Females are linked to higher solid fuel consumption	Himalayas	Mustaq (2014)
Age of the household head	• Older people use more solid fuels	Ethiopia	Mekonnen & Kohlin (2008); Mekonnen (2009)
	• Younger generations rely on charcoal	Nigeria	Ogwuche (2013)
	• Opposing effects		Van der Kroon et

			al., (2013)
Family size	• Large family sizes are linked to wood, smaller families use charcoal	Ouagadougou	Ouedraogo (2005)
	• Larger families use more fuels	India	Guptilla & Kohlin (2003)
	• No effect on fuel choice	Ethiopia	Mekonnen (2009)
	• Smaller households have a higher per capita fuel usage	Developing countries	Knight & Rosa (2012)
	• Larger families use more fuels	Himalayas	Mustaq (2013)
	• Larger families use more fuels	Nigeria	Ogwuche (2013)
Ethnic groups	• Igneous groups are linked to wood usage	Guatemala	Heltberg (2005)
Religion	• Religion is the most important determinant of fuel choice	Himalayas	Mustaq (2013)
Employment rate	• More unemployment are associated with more solid fuel combustion	Himalayas	Mustaq (2013)
Location of a household	• Households in rural areas use more wood	Zambia	Nyembe (2011)
	• Households in rural areas uses biomass fuel	Ethiopia	Mekonnen & Kohlin (2008)
	• Use fuels within the closest distance	Ghana	Zhou (2011); Zhou et al., (2014)
Purpose for fuel use	• Coal are used for cooking water	China	Zhang & Smith (2007)
Seasons	• Different fuel types are used in seasons	China	Zhang & Smith (2007)

Availability of resources	• Cow dung are used by the people with agricultural residues	Himalayas	Mustaq (2013)
	• Households situated near coal selling areas are more dependent on coal	Nigeria	Ogwuche (2013)
	• Households with more livestock are linked to a higher solid fuel consumption	Himalayas	Mustaq (2013)

The level of education in Guatemala was a strong determinant of fuel type used (Heltberg, 2005). The higher the level of education in this area the more likely it became that a household would use less polluting energy sources such as LPG. LPG is regarded as a more efficient form of fuel. Educated members of Ethiopian households use non-solid fuel types as the main energy source (Mekonnen, 2009). A clear correlation was found between fuel choice and post-secondary education level, whereas general educational level didn't relate to a certain fuel type in Ethiopia. Members with an education level higher than the secondary level were associated with cleaner fuel types. Thus, higher education is associated with cleaner, better fuel choices. The education level of the household head in the Himalayas didn't influence the fuel choice of the households (Mustaq, 2013). Nigerian households showed a similar pattern. Of all factors included in the research, the literacy level of the household head had the least influence (Ogwuche et al., 2013). The formal educational level of Ouagadougou members was positively associated with LPG and charcoal usage (Ouedraogo, 2005).

The gender of the household head had a weak effect on fuels used in Guatemala (Heltberg, 2005). In Guatemala the females were associated with use of wood as energy source which involves time-consuming methods such as collecting and combustion practices. Female-headed households in Ethiopia tended to rely on solid fuels or a mixture of solid and non-solid fuels for cooking and heating purposes (Mekonnen, 2009). In Himalayan households, on the other hand, an increase in the number of females in a household led to a higher rate of solid fuel consumption (Mustaq, 2013).

The age of the household head had a twofold effect on fuel usage. These opposing effects have been identified in a recent study of fuel- use determinants (Van der Kroon et al., 2013). According to this finding, a household with older members may be

described as a wealthier household that will use cleaner fuel types. On the other hand, older generations tend to continue using a certain fuel type out of habit, or due to the additional costs that will be needed for new appliances. The age of the household head in Nigeria indicated that the younger generations tended to use more charcoal than older people (Ogwuche et al., 2013). In Ethiopia, the older generation are more likely to use solid fuels for cooking and heating (Mekonnen & Kohlin, 2008; Mekonnen, 2009). Thus, the older the head of the household, the more likely it was that they would use solid fuels.

In the urban Ouagadougou, a correlation was found between family size and differences in the use of LPG, charcoal and firewood (Ouedraogo, 2005). According to this study poor households were linked to larger family sizes and wood as the preferred fuel type. Charcoal, on the other hand, was used by smaller family sizes, which represented wealthier households. Ethiopian fuel preferences couldn't be defined by family sizes (Mekkonen, 2009). The expected results were that larger families would use more solid fuels, because of the labour associated with collection. A study conducted in the Himalayas found that the larger the families, the more fuel would be used (Mustaq, 2013). This relates to the fact that larger quantities of food must be cooked in larger families (Ogwuche, 2013). Similar results were obtained in India where larger families had higher fuel use consumption (Guptilla and Kohlin, 2003). A study conducted in all developing countries found, on the contrary, that countries with smaller households have a higher fuel wood-usage per capita (Knight and Rosa, 2012).

Differences in the choice of fuel types were found between ethnic groups in Guatemala (Heltberg, 2005). Indigenous groups used wood as their main fuel type for cooking and heating practices, which contrasted with other ethnic groups (Van der Kroon et al., 2013).

Religion is the most important social aspect that influenced fuel choice in the Himalayas (Mustaq, 2013). The proportion of Muslim households using solid fuels was 88.9%, in contrast to Hindu with 58% and Sikh with 8.5%. This finding may be ascribed to the large family sizes of the Muslim population.

The more unemployed members per household in the Himalayas meant a higher occurrence of fuel combustion (Mustaq, 2013). The creation of job opportunities would therefore decrease fuel use.

The location of a household may be used to identify the type and amount of fuel used (Nyembe, 2011). Previous studies show that rural households tended to use more wood, whereas rural households relied on cleaner energy types. The practices in Ethiopia support this statement by showing that rural areas were more dependent on biomass fuels than the urban households (Mekonnen and Kohlin, 2008). The proximity to energy sources determined the fuel used by communities (Zhou, 2011; Zhou et al., 2014). The more accessible the energy sources, the higher the probability that it was chosen for cooking or heating practices.

The purpose of energy usage, such as cooking, water heating, space heating and heating of beds, may determine fuel-type selection (Zhang and Smith, 2006). Heating practices dominated the fuel combustion applications in China. Long-term cooking tasks, such as heating water for tea, was usually completed on coal stoves.

Seasonal changes may be linked to certain fuel combustion occurrences. Households in China changed fuel types according to seasonal and daily variations (Zhang and Smith, 2007). Higher emission rates were recorded during winter, as a result of residential fuel use. This finding implies that fuels are combusted for heating purposes (Gupta et al., 2007; Geng et al., 2013; Yu et al., 2013).

The availability of resources showed a direct positive correlation with the fuel type used in households. Households in the Himalayas that used cow dung had abundant agricultural residues (Mushtaq et al., 2014). It was found that households located further from forests used fewer wood sources. The communities in Nigeria that were in close proximity to coal-selling yards were more dependent on coal as the main energy source (Ogwuche, 2013). Residents in the Himalayas with livestock are linked to higher solid-fuel combustions (Mustanq, 2013).

The original stove price influenced the fuel-use pattern in Ghana (Zhou, 2011; Zhou et al., 2014).

The motives behind fuel choice vary globally. Some factors determined high usage rates and others factors had no influence. It is thus important to undertake these studies in every country facing air quality problems caused by domestic fuel-burning practices. Once the factors are identified, mitigation strategies and policies may be formulated to create a cleaner environment and improve the lives of, especially, the poor population.

1.7 Source apportionment: The Chemical Mass Balance Model

Ambient PM is deposited through either natural or anthropogenic sources. Numerous sources may subsequently be responsible for the aerosol loadings in a specific region. In order to control air pollution effectively, knowledge of the causes is needed. All sources should be identified and the proportion of contribution then calculated. To assess PM levels effectively, both quantitative and qualitative information on source profiles is needed (Vallius et al., 2008). Source apportionment methods may be used to show the influence of every source on the ambient aerosol levels (Figure 1-10). In South Africa, three categories of source apportionment techniques have been developed, namely data evaluation, dispersion models and receptor models (Viana et al., 2008).

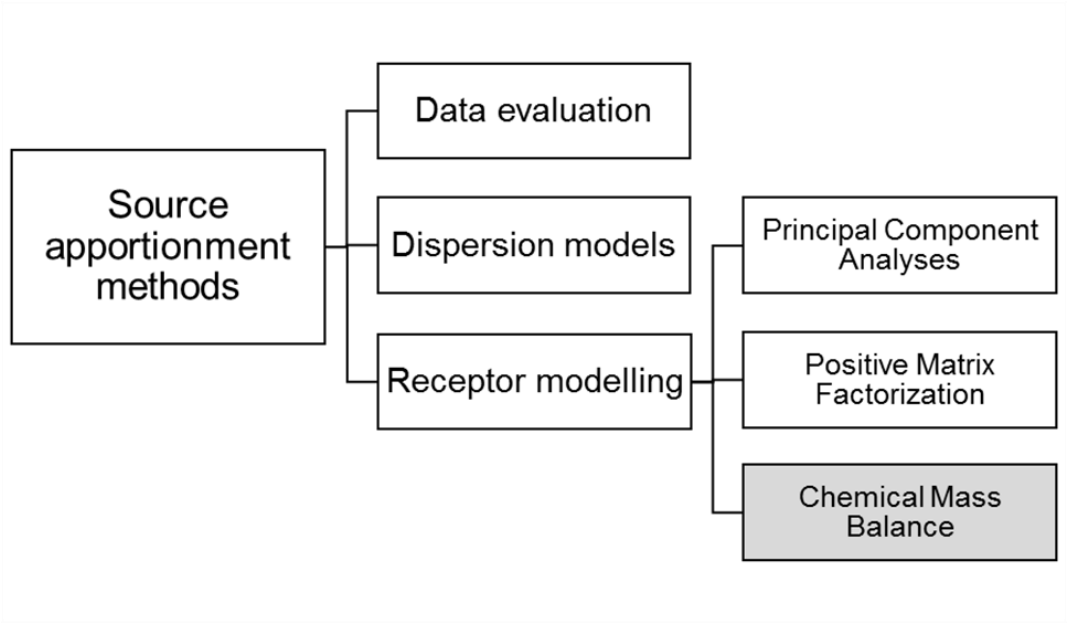

Figure 1-10: Source apportionment methods used in South Africa.

1.7.1 Comparison between the different source apportionment techniques

The data evaluation method is the most simplified source apportionment technique where numerical data is screened and used to identify sources. Conversely, dispersion models are more complex and use the source emission profiles as an input file to determine ambient concentrations (Fragkou et al., 2012). Receptor models, on the other hand, use receptor concentrations as input in order to calculate the source's contributions. A variety of receptor models are commonly used for source apportionment purposes, which includes Principal Component Analysis (PCA), the Chemical Mass Balance model (CMB) and Positive Matrix Factorization (PMF) (Steyn & Rao, 2010). PCA and PMF are briefly described below but emphasis is placed on the CMB model, which was applied in the present research.

PCA makes use of statistical methods to calculate the constituents of PM by the conservation of mass law (Walton et al., 2013). Information on the source profiles is provided by calculating the similarity between the chemical substances found frequently in a variety of emissions from a specific source. This method implies that there is a link between pollutant species loads and the compositions of source emission profiles (Almeida et al., 2006). The emission profile of a certain source may be distinguished from other emissions profiles, when determining the pollutants and their concentration. Another method is used to obtain particle composition data. These data are analysed through various axis rotations (Hopke & Cohen, 2011). The only input file required for this method is the measured concentrations of source emissions. PCA is based on the assumption that source emissions are constant over a time; that no alternative chemical reactions occur, that source profiles dos not interact, errors are random and that there are more chemical constituents than the number of sources (Walton et al., 2013). This method has the advantage of transforming complex datasets into lower dimensions in order to reveal the similarities. PCA is a good method for ascertaining which typical chemicals originate from which source, but cannot be used for source apportionment analyses and for quantifying source contributions. PCA and PMF are also known as factor analysis techniques (Paatero & Tapper, 1993). PCA differs from PMF primarily in terms of the form of output; PCA categorises the data into groups, whereas the PMF output shows no hierarchy.

PMF is a multivariate factor analysis method that utilises two matrices: source profiles and source contributions (Norris et al., 2008). PM data is used as input in this model in order to determine which sources contributed to the ambient samples (Hemann et al., 2009). This analysis tool thus determines which source type is represented by which chemical profile. The source characterisation components, known as factors, are used to identify sources. Experimental uncertainties, measured data and standard deviations are used with the benefit of introducing the limitation of non-negativity (Comero et al., 2009). The advantage of PMF analysis is that it may also be used for non-representative data, such as data below the detection limit. PMF is similar to the CMB model but there are differences in their results (Ke et al., 2008). In previous studies the results from PMF were used as an input file for the CMB model (Begum et al., 2007).

The CMB model combines the chemical and physical features to quantify and determine the presence of source particles and gases in receptor samples (Walton et al., 2013). The model subsequently provides insight into the area's air quality. The contributions are quantified by using chemically distinct source emissions rather than quantifying every individual polluter (Begum et al., 2007). This model operates by analysing every individual sample separately and determining the source contribution of each sample. Samples may differ due to differences in wind direction and speed, emission rates, waste and disposal burning, and coal combustion for cooking or heating (Watson et al., 1990).

The CMB model was employed in the present research to apportion ambient $PM_{2.5}$ and PM_{10} samples to their sources. This model has the ability to show the exact contribution of all identified sources to the air pollution of a specified area. The main polluter influencing the health of residents in Kwadela was subsequently identified.

1.8 Chemical analyses: inductively coupled plasma–mass spectrometry, x-ray fluorescence and ion chromatography

A variety of techniques may be employed to determinate the chemical composition of environmental samples. Examples of such methods are traditional spectroscopic wet-chemistry methods, such as calorimetry, gravimetric analysis, turbidmetry and

electrochemical techniques consisting of ion selective electrodes (ISE) and amperometric titrations. Problems associated with these methods include interferences and limited sensitivity (Jackson, 2000).

The inductively coupled plasma–mass spectrometry (ICP-MS) or the x-ray fluorescence (XRF) technique was applied to determine the presence of elemental compounds within the ambient air samples. Soluble fractions in ambient air samples were obtained with an ion chromatographic system (ICS). The chemical information obtained from the samples is needed as an input file for CMB modelling. This section will describe the history and efficiency of these methods.

1.8.1 Advantages of inductively coupled plasma–mass spectrometry

ICP-MS was first introduced in the 1980's and has been used as an accurate, versatile detection procedure (Pröfrock and Prange, 2012). Compared with other chemical detection methods, ICP-MS is the preferred method owing to its various advantages. This technique is an efficient elemental detector and is well-known for its excellent quantitative analytical capabilities and analytical chemistry (Ammann, 2007; Hassib et al. 2008). Benefits associated with ICP-MS include the ability to operate with both simple and complex matrices with a low matrix intrusion as well as the ability to obtain isotopic information. ICP-MS has the ability to perform quantitative control, where only the relevant chemical species are consequently investigated. Another distinctive property of this method may be ascribed to the plasma component, with the highest collision rate of ions, because of high ion densities (Ammann, 2007). In addition, contamination is prevented because of lower susceptibility to salts and solvent loads. The presence of salts and solvents are known to cause blockages in the instruments. ICP-MS is preferred because of its higher accuracy (through low detection limits) and superior detection capabilities, (Wolf, 2005). Most elements in the periodic table can be detected by ICP-MS (Figure 1-11).

The results obtained from the plasma ion source show the elements contained within the sample (Ammann, 2007). In the present study the chemical elemental footprint acquired for every individual sample was compared with the different source profiles in order to identify the dominant polluting source at Kwadela.

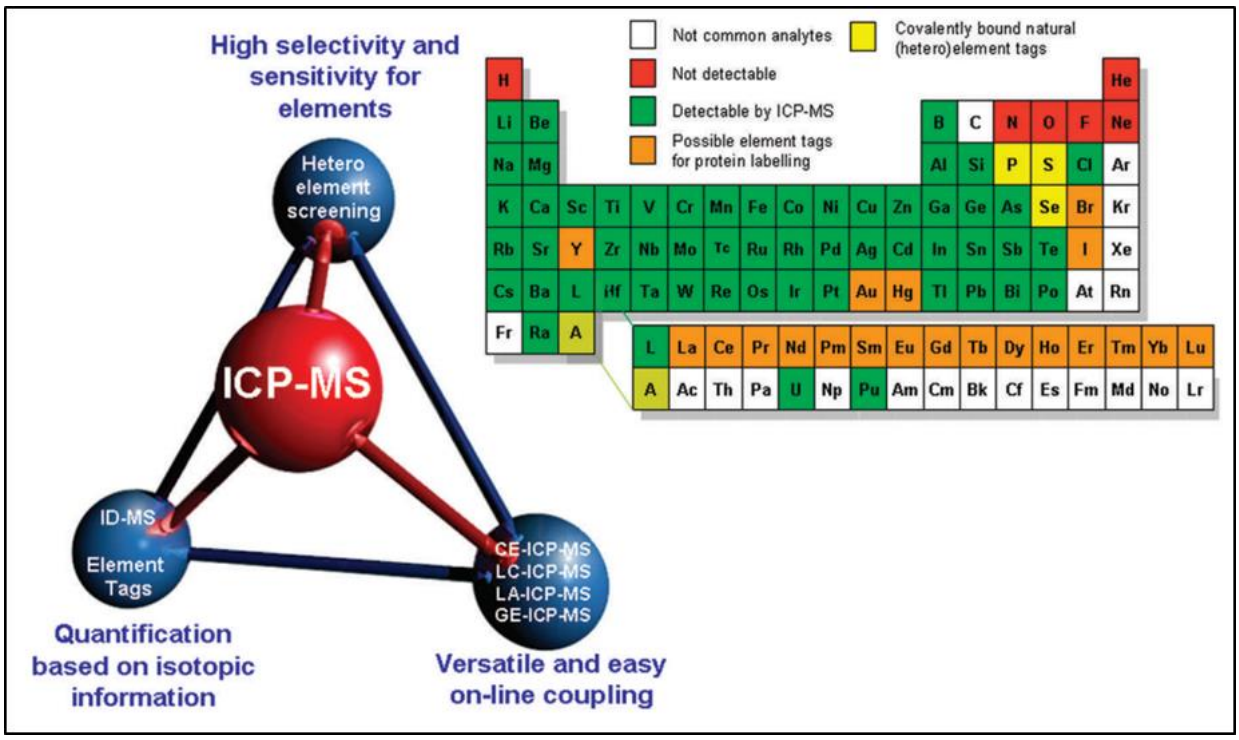

Figure 1-11: Application and detection capabilities of inductively coupled plasma–mass spectrometry (ICP-MS) (Pröfrock and Prange, 2012).

1.8.2 X-ray fluorescence analysis

The XRF method was introduced in the 1913's were Henry Moseley contributed to numbering elements in the periodic table (Shackley 2011). This procedure involved observing k line transitions as it moved across an X-ray spectrum. This method were improved up to a point were different elements are identified from different sample states. XRF are generally used for a wide range of commercial applications and a prevalent tool in the species detection field.

The main advantage of XRF is the non-destructive analysis method. Samples can thus be stored for future reference, of send for different analyses. Secondly, minimal preparation is required before analyses. Samples must be diluted in most of the other techniques, which can be time-consuming. This method is also ranked above other elemental detection procedures for being cost effective and easy to use. The precision of detection are high with accurate, reproducible results (Brouwer, 2010).

1.8.3 The ion chromatography method

The ICS method was developed by Hamish Small in 1975 for the separation and conductometric quantification of ion elements (Small et al., 1975; Jackson, 2000). The introduction of this method served as a foundation for the development of ionic solubility determination. Compared with other spectroscopic methods, ICS offers the advantage of cation analysis for metal determination. The ability to differentiate between the states of metal oxides renders this method superior to alternative methods (Heberling et al., 1990; Schnell et al., 1998; Jackson, 2000). Many different research fields employ this technique, which has been approved by a wide range of regulatory organisations. Most of these government-approved techniques are based on analyses of cations (Table 1-4). Thus, ICS is a reliable, regulatory-approved method for the efficient extraction and identification of ions in air quality samples.

Table 1-4: Regulatory IC methods used in the USA (Jackson, 2000).

Method	**Analytes**
EPA Method 300.0 (A)	F, Cl, NO2, Br, NO3, PO4, SO
EPA Method 300.0 (B)	BrO_3, ClO_3, ClO_2
EPA Method 300.1 (A)	F, Cl, NO_2, Br, NO_3, PO_4, SO_4
EPA Method 300.1 (B)	BrO_3, Br, ClO_3, ClO_2
ASTM D 4327-97	F, Cl, NO_3, Br, NO_2, PO_4, SO_4
Standard Methods[11] 4110	Cl, NO_3, Br, NO_2, PO_4, SO_4
EPA Method 300.6	Cl, NO_3, PO_4, SO_4
ASTM D 5085-90	Cl, NO_3, SO_4
EPA Method B-1011	NO_2, NO_3
EPA SW-846 9058b	ClO_4
EPA Method 218.6	Hexavalent chromium (CrO_4^{-2})
EPA SW-846 7199	Hexavalent chromium (CrO_4^{-2-})
ASTM D 5257-93	Hexavalent chromium (CrO_4^{-2-})
ASTM D19.05.03.23b	Na, NH_4, K, Mg, Ca
EPA Method 300.7	Na, NH_4, K, Mg, Ca
EPA Method 200.10c	Cd, Co, Cu, Pb, Ni, U, V
EPA Method 200.13c	Cd, Co, Cu, Pb, Ni

1.9 Sources of particle matter in the South African Highveld

A variety of sources emit aerosols into the atmosphere. These causes of ambient air pollution include industrial emissions, domestic fuel-burning vehicle emissions, and a variety of other point and non-point sources (Cairncross et al., 2007). In order to evaluate air quality and determine PM quantities in a certain area, a clear description of all contributing sources are needed. Identifying sources releasing airborne particulate matter (APM) and estimating the exact amount of PM are both main objectives in atmospheric research (Begum et al., 2007). The amount and characteristics of every source's emissions will differ over temporal and geographic scales due to a number of influences (Cairncross et al., 2007; Lu et al., 2010).

1.9.1 Sources of particulate matter

Even though chemical profiles are generated for emissions, it may vary from one region to another. For the purpose of the present study, the sources that could cause air pollution in a low-income community, Kwadela, were identified. These sources include agricultural activities and vehicle emissions, waste disposal, dust particles, coal dust, domestic fuel-burning and power stations (Figure 1-12). Kwadela is situated amongst various polluting sources, each within a 50 km range. When evaluating these sources, it is also important to consider their distance from the sampling location. The proximity of sources will give an indication of whether PM_{10} or $PM_{2.5}$ will be found in receptor samples.

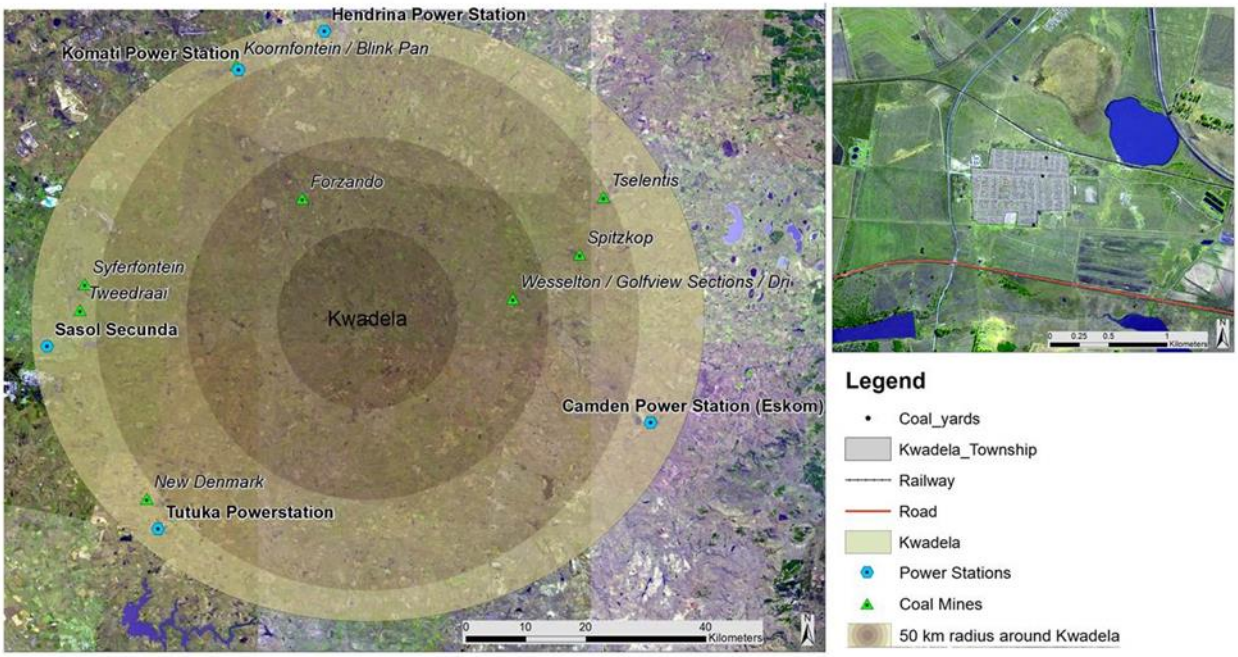

Figure 1-12: Sources contributing to PM in Kwadela, Mpumalanga, are illustrated within a 50 km range of Kwadela. The image on the right shows the sampling site, yards within Kwadela where coal is sold and the proximity of the railway and N17 road.

1.9.1.1 Vehicle emissions

The N17 is a highway that passes by Kwadela and carries high traffic volumes (Figure 1-12). The emissions from vehicles (especially diesel vehicles) contribute significantly to the amount of $PM_{2.5}$ in the atmosphere (Mizohata et al., 1995). The emissions from diesel/gasoline engine- operated vehicles are recognised by high concentrations of black carbon (BC) and S together with magnesium (Mg), aluminium (Al), silicon (Si) and iron (Fe) dust elements (Maricq et al., 2002; Kim et al., 2003; Begum et al., 2004; Begum et al., 2007). Other emissions classifiable in the transport sector are dust on the road, tyre, brake and clutch wear, as well as corrosion of vehicle parts (Wåhlin et al., 2006). Given the close proximity of the N17 to Kwadela, it is important to consider all emissions caused by the transport sector.

1.9.1.2 Refuse burning

Refuse-burning is seen as a significant source of air pollution in South African dense, low- income communities (Nuwarinda, 2007; Pauw, 2008; Walton, 2013). The combustion of domestic waste masses contributes significantly to PM and is responsible for a variety of chemical compounds, air toxics and ash (Li et al., 2012). These aerosols contain heavy metals, VOCs, dioxins and a variety of hazardous substances, such as hydrochloric acid (HCl), polycyclic aromatic hydrocarbons (PAH) and BC (Li et al., 2012; Park et al., 2013). Refuse-burning ought to be undertaken in controlled environments and at certain temperatures to minimise impacts. The problem with these local combustion practices is that garbage is typically burnt at lower temperatures than required. Pollutants released from refuse burning are a health hazard for residents of Kwadela since it comprises of highly toxic substances (Annegarn et al., 1998).

The residents of Kwadela burns excess waste, which is a major source of air pollution (Figure 1-13). Refuse-burning practices are more common in the winter to provide heat to the poor residents Refuse burning is usually accompanied by biomass burning (veld fires). Products of biomass-burning include K, Br, Rb, C and ZN species (Maenhaut, 1995; Duan et al., 2004). Veld fires contribute to the amount of primary and secondary aerosols in ambient PM. Gases emmited form veld fires include secondary pollutants such as CO_2, CH_4, CH_3, N_2O and NO_x (Artaxo, et al., 1998). Trace elements emitted from biomass- burning can impact PM on a global scale.

Figure 1-13: Local community members burning excessive waste in Kwadela (Van den Berg 04/2014).

1.9.1.3 Dust particles

Dust in general is defined as particles with a smaller diameter than 1000 µm that can be found, either in suspended form or deposited on a surface area (Moja & Mnisi, 2013). These dust particulates are commonly referred to as aeolian dust. The influence of dust on climate systems and air quality remains a neglected area of study and is poorly quantified (Maher et al., 2010). aeolian dust is highly complex and varies spatially and temporally (Goudie, 2008; Maher et al., 2010). The mobilisation of these particulates varies according to climate factors, the atmosphere and terrain properties. The entrainment properties of soil detachment are illustrated in Figure 1-14, which shows that the main cause of aeolian dust is turbulent wind flow. Human activities mainly cause such soil erosion and aggregate the presence of dust particulates in the atmosphere.

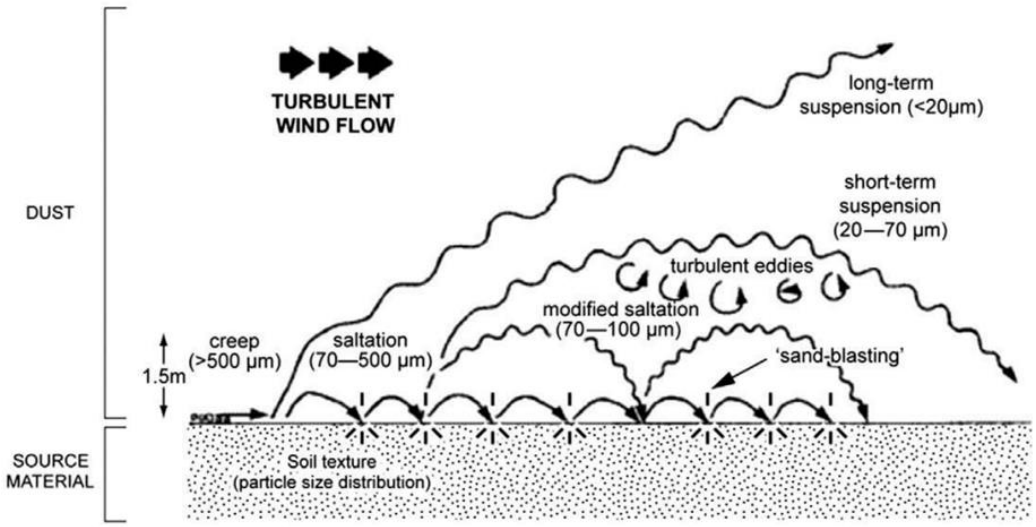

Figure 1-14: Entrainment properties of aeolian dust (Maher et al., 2010).

Aeolian dust at the study site (Kwadela) could be derived from various sources. The typical sources were categorised into four groups: paved road dust, unpaved road dust, agricultural dust and coal dust.

1.9.1.3.1 Road dust

Not all roads in Kwadela are paved and transportation causes the release of dust particles into the atmosphere. The roads in poor residential areas such as Kwadela are usually unpaved which causes that more dust particulates are released. A study conducted in a South African Township (Leandra) identified dust as one of the major sources of PM (Mugabo, 2011). Road dust is linked to the amount of airborne PM (Luhana et al., 2004). Each of the identified sources have a different chemical profile but are mainly composed of carbon (C), Si, calcium (Ca), Al and Fe (USEPA, 2014). The amount of dust particles in the atmosphere peaks during winter and is minimal during the rainy season (Begum et al., 2007). This occurrence is caused by lower rainfall during winter. According to numerous studies, dust is a significant contributor to $PM_{2.5}$ and PM_{10} (Rogge et al., 1993; Chow et al., 1995; Schauer, et al., 1996; Ruellan & Cachier, 2001; Luhana et al., 2004).

1.9.1.3.2 Agriculture and land use

The Kwadela township is located amid farms and therefore may be influenced by emissions from agricultural activities (Figure 1-12). Some of the local community members farm with livestock, such as pigs and goats, as well as maize crops on the outskirts of the township. Agriculture and land-use activities may contribute to the amount of $PM_{2.5}$ and PM_{10} through wind erosion of fertile soil, livestock emissions, chemical fertilisers and the burning of crop residues.

Particulates from dust may result from vehicles and machinery driving on unpaved roads on the farms together with wind, which may cause the emission of excessive fertile particles into the atmosphere. Agricultural soil is more likely to contribute to air pollution during the dry season and generally have higher chemical concentrations during these periods. The reason for this is the addition of compost to the soil. Soil is composed of Al, Si, kalium (K), Ca, titanium (Ti) and Fe (Watson et al., 2001; Begum et al., 2007). These particles could also contain pesticides, chemicals and plant matter (Walton et al., 2013). Pesticides may cause the direct emission of particles that are mostly composed of organic constituents. According to the literature, some of these particles contain volatile components, such as xylene, emulsifiers, diluents and other organic solvents (USEPA, 1995; Walton et al., 2013).

The amount of particles caused by agricultural activities depends on the season and on meteorological conditions. The winter is drier and will cause the release of higher quantities of PM particles. Windier conditions and dry periods are thus responsible for higher amounts of suspended particles.

1.9.1.4 Coal dust

Kwadela is a low-income community that is still dependent on alternative, cheaper energy sources for cooking and heating purposes. Coal is distributed to the households from four plots in this township (Figure 1-12). The loading and transporting of coal to households may result in the discharge of coal particulate matter. Coal dust is mainly composed of Si and Al particles (USEPA, 2014). Coal mines are also responsible for releasing coarse PM in this region.

Eight coal mines are located within a 50 km radius of Kwadela: Koornfontein, New Denmark, Syferfontein, Spitzkop, Tselentis, Tweedraai, Frozando and Wesselton. The handling, storage and transportation activities associated with these mines may cause an increase in total airborne PM comprised mainly of secondary species such as CO_2 and CH_4 (Letete et al., 2009). Coal dust is an important source that contributes to PM in Kwadela and should thus be included in the CMB.

1.9.1.5 Domestic fuel burning

The local members of Kwadela are still reliant on dirty fuels as an energy source for cooking and heating purposes. Electricity expenses, lack of electrification and the abundant availability of coal and wood resources are all reasons why the community prefers alternative energy sources during winter. Coal is the predominant fuel type used by residents owing to its abundant availability. The community uses a monthly average of 216.38 kg coal during the winter months and 117.03 kg during summer (Burger & Piketh, 2015). The percentage of residents that use coal during winter is 74.9% and 54.1% of the residents use coal for cooking during summer. Coal and wood combustion increases during winter because it is used for both thermal heating and cooking (Figure 1-15). Domestic coal combustion is a major cause of pollution in low-income settlements (Zunckel et al., 2006; Nuwarinda, 2007). Products released from coal combustion include S, Cl^- and secondary species NO_3^- and SO_4^{2-}.

Figure 1-15: Typical stoves used for cooking and heating purposes in Kwadela, Mpumalanga (Van den Berg, 08/2014).

The burning of alternative energy sources result in the emission of CO, NO_2, SO_2, suspended aerosols and PAH (Walton et al., 2013). Coal combustion emits high loads of BC and S (Davy et al., 2011). The burning of wooden products releases large amounts of PM into the atmosphere, along with other inorganic and organic chemicals. Even though the combustion of coal primarily discharges fine PM (in the sub-micrometre range), it also affects coarse particles by agglomeration or condensation (Hedberg et al., 2002; Davy et al., 2011). Impure coal is commonly used because it is cheap and abundant, but is notorious for its constituents such as alumina-silicates, co-deposited with hydrocarbon plant soils, iron(II) sulphide (Ferrous Sulfide)(FeS), arsenic sulphite (AsS) and heavy metals such as thorium (Th) and uranium (U) (Nuwarinda, 2007). Domestic fuel-burning evidently has serious environmental and health impacts.

1.9.1.6 Power stations

Power stations are considered one of the largest contributors to poor air quality in the Highveld region. Four coal-fired power stations are located within a 50 km radius of Kwadela: the Camden, Tutuka, Komati and Hendrina power stations (Figure 1-12). These point sources release a mixture of particles of different sizes (Lu et al., 2010). Emission control technologies aim to reduce emissions of components such as SO_2, NO_x, PM, mercury (Hg), SO_3 and other trace metals (Moretti & Jones, 2012). Monitoring and measurement of the concentrations of these pollutants is done on a regular basis. Typical pollutants measured are PM, Hg, CO, CO_2, N_2O, NO, NO_2 and SO_2 (Walton et al., 2013).

The dominant pollutants released during coal combustion are BC and S (Davy et al., 2011). Coal consists of C, N, S, ash, Hg and additional elements that undergo chemical reactions with other elements during combustion processes. Secondary pollutants such as NO_x, SO_2 and SO_3 are also discharged from combustion activities (Table 1-5). An important aspect to take into consideration is the equipment that is used during these combustion activities as well as the process that follows. Equipment is built for a specific process and each type of machinery used will be responsible for releasing different pollutants. The process followed should also be considered when assessing the combustion emissions (Moretti & Jones, 2012). Power stations are responsible for a variety of pollutants, and the exact amounts may be determined through source apportionment analyses.

Table 1-5: Pollutants that result from coal combustion (Moretti & Jones, 2012).

Raw coal substances	**Combustion result**	**Equipment used**
Carbon (C)	Heat, Steam, CO_2	Boilers
Nitrogen (N)	NO_x	Burners and SCR Systems
Sulphur (S)	SO_2	Wet and Dry FGD
	SO_3	Sorbent injection, Wet ESP
Ash	Ash	Ash handling, Soot blowers, precipitators, Fabric filters
Mercury	Hg^{++}, Hg^0	Coal additive, PAC injection, wet FGD enhancement systems

1.9.1.7 Secondary aerosols

Secondary pollutants need to be considered in air quality analysis because it is considered as a main constituent fine PM (Fine et al., 2015). The dominant secondary species are SO_4^{2-}, NO_3, $(NH_4)^2$ SO_4. The abundances of these species varies over time and space as external factors such as meteorology, source emissions, thermodynamics and transport systems changes. Sulphate species are formed from the oxidation of S species and precursor gas SO_2 emitted from combustion sources. The transformation of SO_2 into SO_4^{2-} could also be a result of aqueous-phase chemistry that occurs in clouds or rainwater. $(NH4)^2$ SO^4 species are formed when sulphuric acid (H_2SO_4) reacts with ammonia (a product of vehicle emissions and agricultural activities). The amount of NH_3 in the atmosphere largely influences the production of $(NH_4)(NO_3)$ from HNO_3 species (Fuzzi et al., 2015). NO and NO_x are directly emitted from anthropogenic combustion. These species undergoes gas to phase reactions with hydroxyl radical (OH) to form NO_3. This process occurs during the day when photochemistry is higher. NO_3 could consequently react with marine species to from $NaNO_3$. Atmospheric reactions are classifies as either heterogeneous or homogeneous (Piketh, 2000). Reactions between

species in the same phase are known as homogeneous whereas heterogeneous processes are the opposite. Heterogeneous processes occur in the presence of liquid such as clouds and fog. Dry conditions are associated with homogeneous reactions. Reactions in South Africa are usually homogeneous due to the climate conditions. Including secondary aerosols in analyses is of upmost importance, due to its abundance in ambient PM, throughout the country, during all the seasons.

All the above mentioned sources have a distinct chemical and elemental profile, which can be used in source apportionment analyses. All possible polluters that influence the aerosol loadings in Kwadela were identified and the emission profiles were obtained. The contribution of each source and the presence of secondary species was determined in chemical analysis and the chemical mass balance model.

CHAPTER 2:

DATA AND METHODOLOGY

The purpose of this chapter is to explain the data acquisition methods and equipment used for this research. Analyses procedures are defined, and all associated calculations and formulas used are given and explained. The Chemical Mass balance model used during the investigation is described comprehensively. This model was chosen in order to achieve the main objective of this research. All data components and requirements are also clarified.

The importance of domestic fuel use in South Africa was explained by means of two approaches. Firstly, an investigation was conducted based on fuel use patterns and the number of people affected by air pollution. The scope of this study was on a national scale and all South African fuel users were considered. Secondly, the quality of the air of a low-income community, located close to power stations, mines and other polluting sources was investigated. The selected study domain was based on a local scale. These two investigations were respectively categorized as domestic fuel use in South Africa and source apportionment.

2.1. Study site information

A small, low-income community within the Highveld region was selected for this research. The reason for this is that the Highveld Priority area (HPA) is notorious for its poor air quality as a result of the large number of polluting sources (DEA, 2011; Masekoameng, 2014 and Luhunga, 2013). The air of this region encompasses particulates from commercial, mining, industrial, residential and agricultural sources. Major industrial and urban areas are situated within the Middelburg, Witbank and Secunda cities (Scorgie et

al., 2012) (Figure 2-1). Apart from all these emissions were the impacts of domestic fuel burning also researched.

The Highveld plateau is approximately 1600 m above sea level and the southern region is dominated by the Vaal Basin which is 1400 m above sea level (Lydia, 2010). The land area covers 31 106 km^2 and is recognised by the Department of Environmental Affairs (DEA) as a pollution hotspot or priority area. A notice for approving the HPA air quality management plan was submitted to the DEA in 2011 (DEA, 2011). The air quality problem areas in the HPA are Emalahleni, Kriel, Steve Tshwete, Ermelo, Secunda, Ekurhuleni, Lekwa, Balfour and Delmas. The topography and industrial sources are illustrated in Figure 2-2.

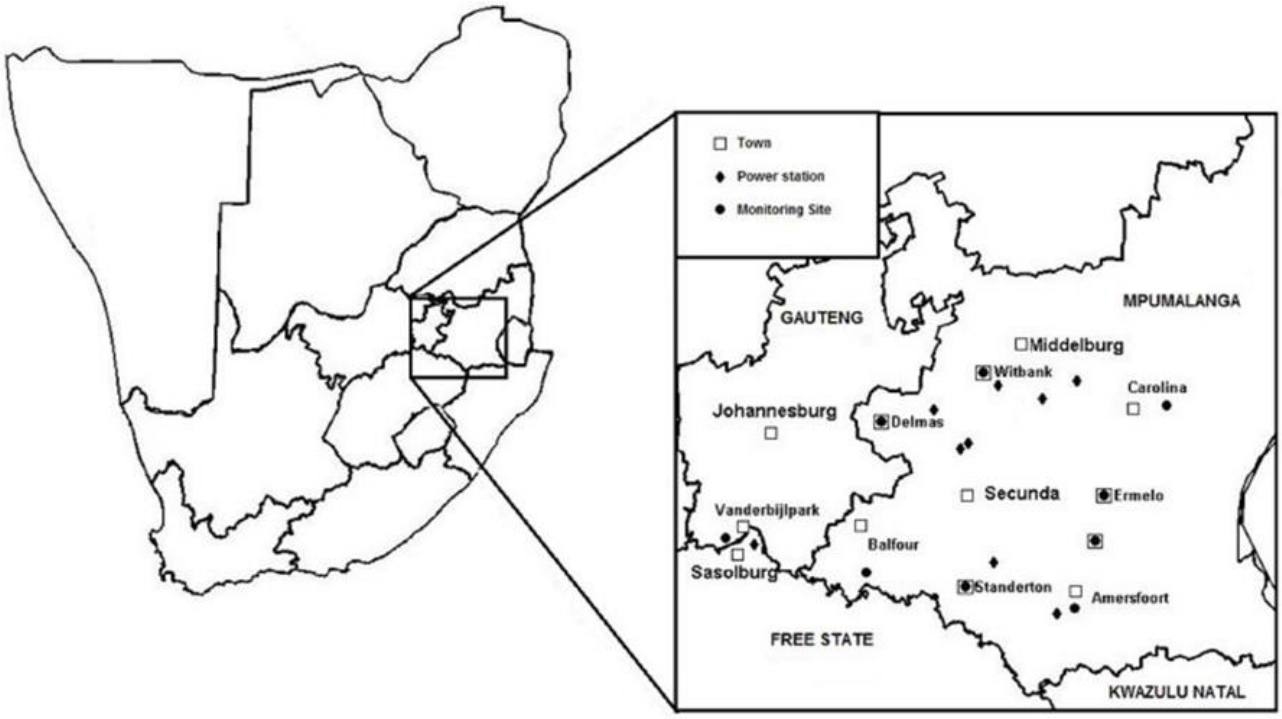

Figure 2-1: Map of the Highveld priority area illustrating major point sources, monitoring stations and towns (Lourens et al., 2011).

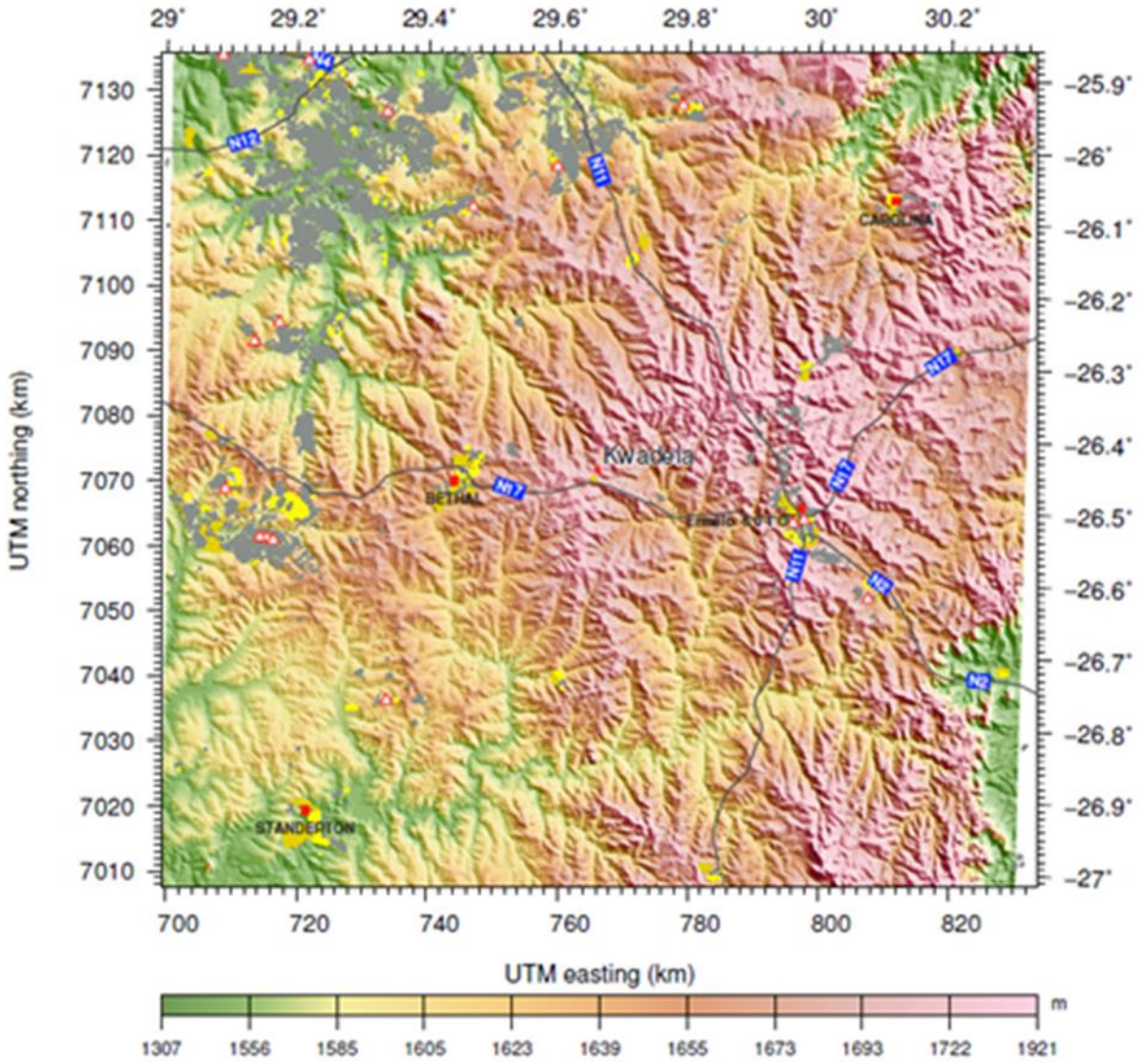

Figure 2-2: Illustrating the topography of the Highveld, the Kwadela township and surrounding industrial sources in red triangles (Burger & Piketh, 2015).

The Kwadela low-income settlement is located at 26°46'3072'' S and 29°66'3138'' E, and is situated between the Bethal and Davel towns in Mpumalanga (Figure 2-3). A total number of 3777 people reside in this community. This area is part of the Govan Mbeki Municipality district. The N17 (a main road) passes south of Kwadela and a railway is situated to the north. Agricultural lands enclose this residential area, with grasslands as the dominant vegetation cover. It can therefore be deduced that a variety of polluting sources are within a 50 km radius of Kwadela and will impact the ambient air quality.

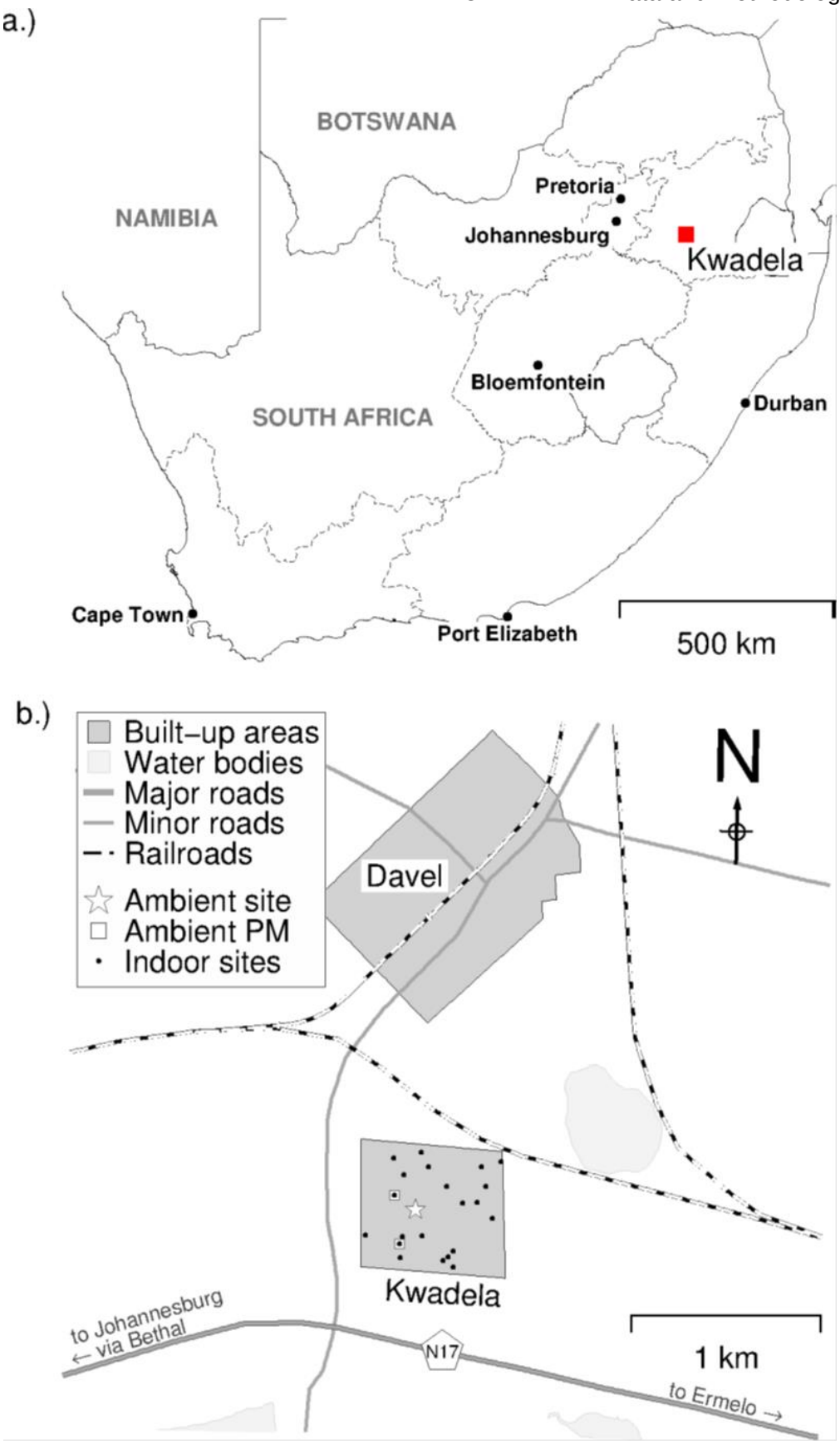

Figure 2-3: The location of Kwadela a) in Mpumalanga and b) on a local scale showing the surrounding area (Piketh and Burger, 2013).

2.2. Domestic fuel use in South Africa

The significance of residential fuel combustion in South Africa is often neglected and the influence of local combustion emissions is not taken into account. Low income communities that depend on biomass fuels were identified and their fuel use habits were investigated. This study was undertaken by using the distinct datasets; census data and spatial information. The factors that influence fuel choice were categorized and the number of affected households and people were determined. Regression modelling and statistical calculations were used to illustrate the importance of residential combustion practices (Figure 2-4).

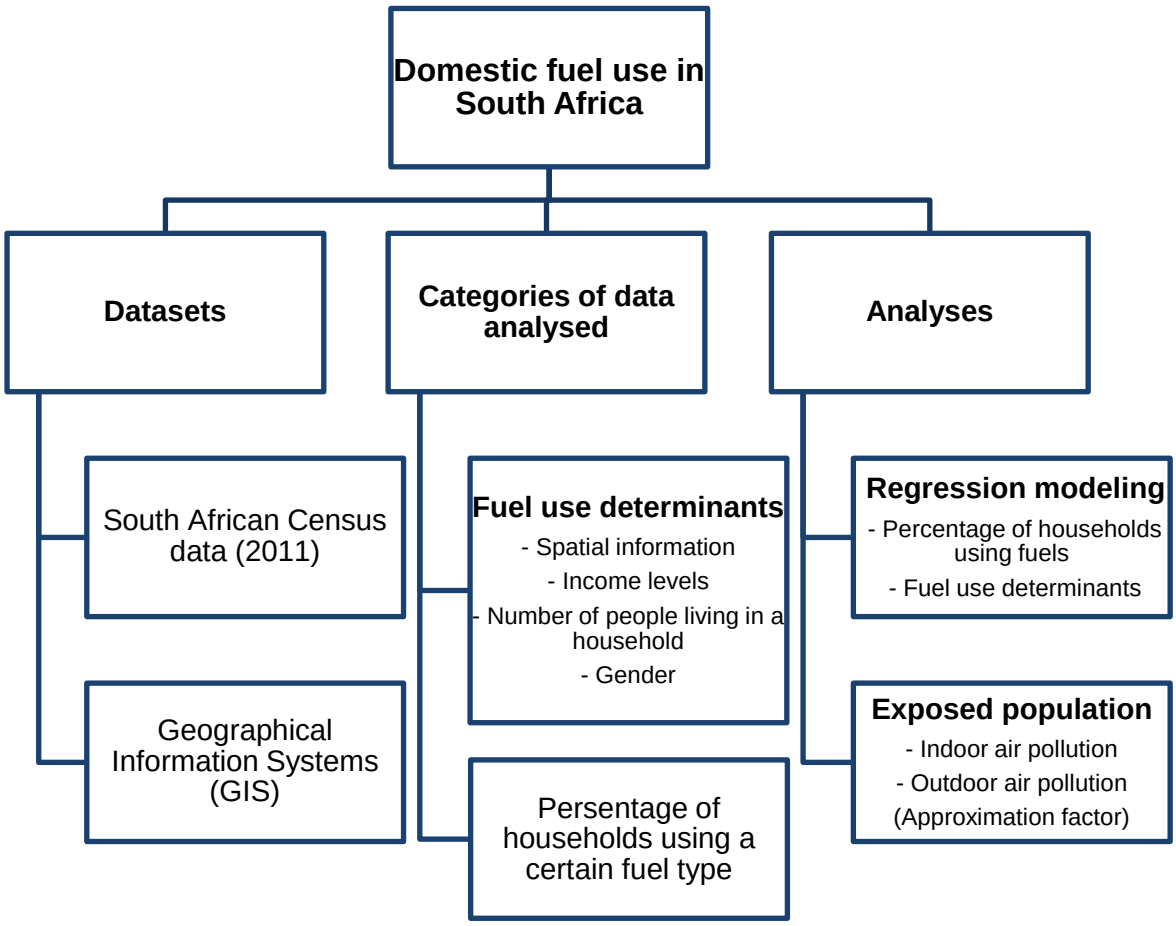

Figure 2-4: Method used to determine domestic fuel use in South Africa.

2.2.1 Information and datasets

The trends in domestic fuel use and the number of people affected by domestic fuel burning emissions were determined by evaluating South African census data on a SAL scale. This information represented the most recent census information (2011) and demonstrated the influence of dirty fuels on the local and national population. Consequently data from Geographic Information Systems (GIS) layers were used for the spatial analyses. The selected GIS layers comprised of vegetation, temperature and location proximity data.

2.2.2 Analyses of South African fuel use data

Regression modelling and exposure determinations were conducted from these datasets. The regression model is a statistical method that was used to calculate the relationship between fuel use determinants and the number of fuel users. Regression modelling finds correlations between two or more variables. The following equation was used:

$$\gamma = \beta_0 + \beta_1 X + \in$$

γ	variable
X	predictor variable
$\beta_0 + \beta_1$	model regression coefficients
$\in$	disturbance/error

The initial possible ‘predictors’ of fuel use included the number of people living in a household, the gender of the head of the household, annual household incomes, the number of people with chronic diseases, appliances used for cooking, enumeration type of the residence, as well as the temperatures, vegetation and coal mines in the vicinity. These predictors (variables) were compared to the number of fuel users on a SAL scale (predictor variable).

The proportion of fuel users in South Africa was accordingly calculated along with the number of people affected by these practices. An upper limit and lower limit were generated. The lower limit involved calculating the number of people exposed to indoor and

outdoor pollution respectively. The number of people exposed to indoor pollution was calculated by multiplying the number of fuel burning households with the median of household members.

$$Exposed\ Population_{In.} = N_{FB} \times \bar{X}_{Res}$$

N_{FB} Number of fuel burning households

$\bar{X}_{Res}$ Average number of people in a household

The outdoor air pollution represented the people who live within the SAL where more than 30% of the households burned solid fuels.

$$Exposed\ Population_{Out.} = SAL_{(>30\%)} \times N_{Res}$$

$SAL_{(>30\%)}$ SAL's where >30% of the households burn fuels

N_{Res} Number of people living in the selected SAL's

For the upper limit, the results were multiplied with a correction factor, to take the uncertainties of the census questionnaire into account. The rationale behind this correction factor was the fact that the census questions only allowed people to identify the main fuel type they used. People in low-income settlements tend to use a mixture of fuels to suit their needs (Pauw et al., 2013).

2.3. Source apportionment analyses

2.3.1 The Chemical Mass Balance Model

The CMB model was employed to apportion ambient $PM_{2.5}$ and PM_{10} samples to their sources. The chemical and physical features were combined to quantify and determine the presence of source particles and gasses in receptor samples (Walton et al., 2013). This model thus provided insight into Kwadela's air quality. The CMB quantifies contributions by using chemically distinct source emissions rather than quantifying every individual polluter (Begum et al., 2007). Every individual sample was analysed separately and the source contribution of every sample was determined. The samples' constituents differed due to variations in wind direction and speed, emission rates, waste and disposal burning, and coal combustion for cooking or heating practices (Watson et al., 1990). A summary of CMB inputs and outputs are given below (Figure 2-5).

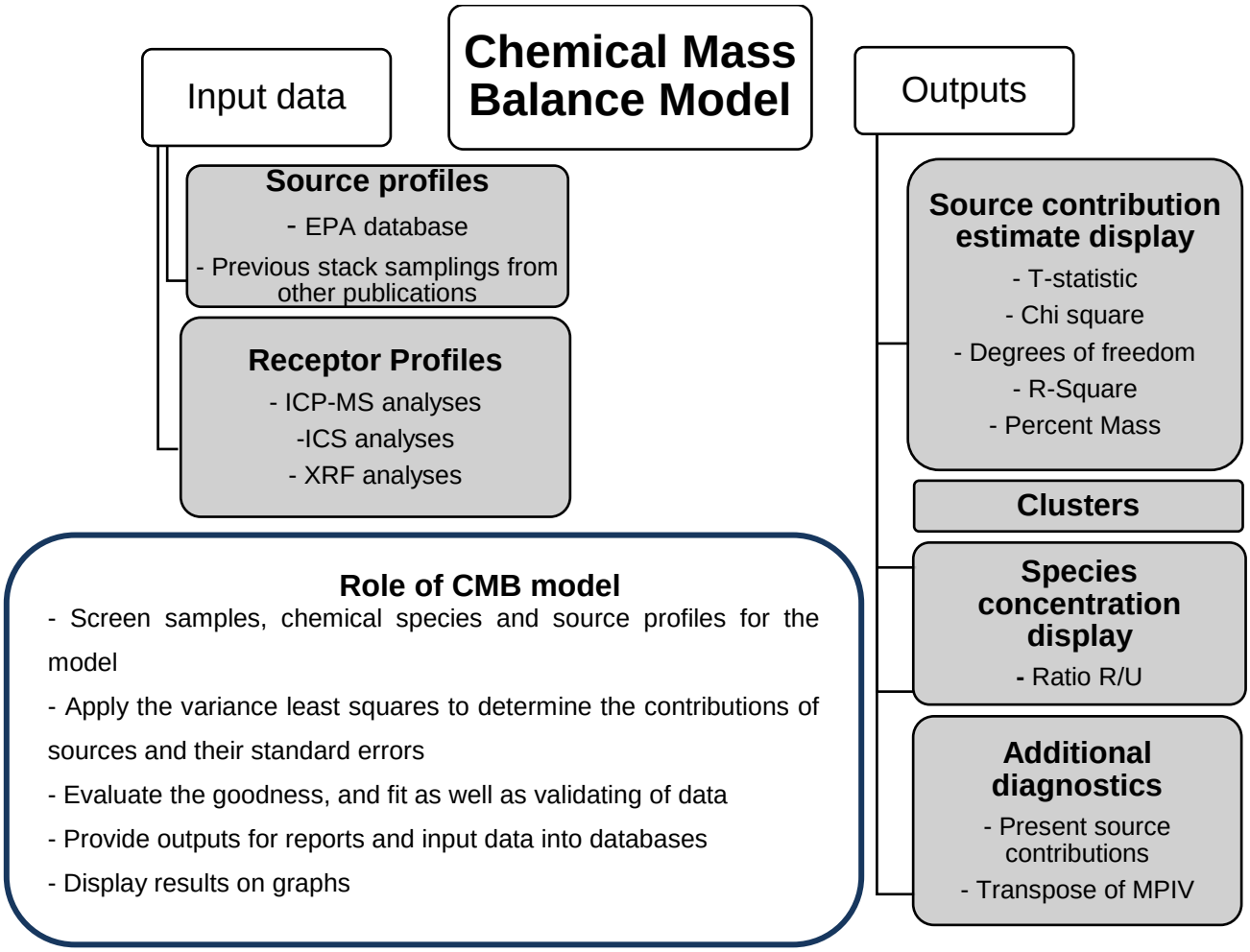

Figure 2-5: A summary of the CMB model (Watson et al., 1990).

The procedure of the CMB can be explained in 5 steps. A least-squares solution of linear equations was used to demonstrate the chemical species of each receptor concentration as a linear sum of the source profiles (Fujita et al., 2007). The equation is expressed as follows (Walton et al., 2013):

$$C_i = \sum_{j=1}^{p} a_{ij\,S_j}, i=1,n$$

Where:

C_i ambient concentration of species (i)

a_{ij} is the fraction concentration of species (i) in emissions from source (j)

S_j the total mass concentration contributed by source (j)

p number of species

n number of species where n ≥ p

The goodness and fit of for the least squares equation was determined and the data validated. The output data were presented along with all the data that were entered into databases. The fit measurement was calculated in the CMB through the following algorithm;

$$Fit = Wt_{chisqr} \times \frac{2}{chisqr} + Wt_{Rsqqr} \times Rsqr + Wt_{pcmass} \times \frac{pcmass}{100} + Wt_{fracest} \times Frac_{Est}$$

$chisqr$ Chi-square

$Rsqr$ R-square

$Pcmass$ percent mass

$Frac_{Est}$ the ratio of the number of fitting sources to the total number of fitting sources

Wt the weight accorded to each of these performance measures

The CMB model used several procedures to evaluate the data and to determine the contribution of sources to receptor concentrations. There were two types of input data files

that were required in the CMB model. The first file was the source profiles which were collected from the USEPA database and previous studies conducted in South Africa. Ambient samples of fine and coarse particles were collected with Ghent Stacked Filter Units (SFU) and 47-mm Nucelopore polycarbonate filters. The elemental constituents within the PM samples were obtained through ICP-MS analyses and ionic elements through the ICS method. The performance measures used to validate source contributions included the source contribution estimates, clusters, species concentration display and additional diagnostics. The source contribution estimate (SCE) is the foremost output where the sum of the concentrations is an estimate of the total mass concentration. If the source emissions remained constant over time with no interaction with other emissions, these species in the source contributions will equal the sum of the receptor concentrations. The total mass of measured species in the receptor sample was calculated as follow:

$$C = \sum_{J=1}^{J} D_j \times E_j = \sum_{j=1}^{J} S_j$$

C total mass measured at the receptor

J number of sources

E_j emission rate

S_j source contribution

D_j dispersion factor

The elemental concentrations were then determined by the following formula:

$$C_i = \sum_{J=1}^{J} F_{ij} \times S_j$$

C_i composition of elemental concentrations

F_{ij} fraction of source contribution

Standard errors of data were displayed through t-statistic (Tstat) values to validate ambient data and source profiles as well as indicating the similarities between profiles. Tstat values provided the ratio of the SCE compared to the standard error. The applicability of the source profiles were assessed through chi-square, degrees of freedom, r-square and percent mass analyses (Gillies et al., 2008). Chi-square involves the weighted sum of squares of the differences between the measured and calculated elemental species. A value <1 shows a good fit of the species whereas larger >1 indicates a poor fit. The following equation was used to determine the chi - square:

$$Chi\ square = x^2 = \frac{1}{I-J}\sum_{i-1}^{I}\left[\frac{\left(C_i - \sum_{j=1}^{J} F_{ij}\ \ S_j\right)^2}{V_{eii}}\right]$$

I = number of chemical species

V_e = Diagonal matrix of effective variances

The degrees of freedom (DF) performance measure were required for the statistical testing of chi-square values. The DF value was obtained by subtracting the number of fitting sources from the number of fitting species. R-Square results indicated the inconsistency in measured concentrations that may be explained by the inconsistency in species concentrations. This value is expressed as a ratio. The R-square value ranges from 0 to 1, where values closer to 1 indicate better SCE's compared to the measured species. These results were obtained through:

$$R\ square = 1 - i = \frac{[(I-J)x^2]}{\left[\frac{\sum_{i=1}^{I} C_i^2}{V_{eii}}\right]}$$

Mass percentage shows the number of calculated source species found within the total measured mass. The target value was 100% which indicated that measured mass can be explained fully by source emissions. The mass percentage required dividing the mass of source contributions by the total measured mass (Ct):

$$Percent\ mass\ (\%) = \frac{\left(\sum_{j=1}^{J} S_j\right)}{C_t} \times 100$$

Clusters were displayed to indicate similarities between source profiles or uncertainties of individual source profiles (Watson et al., 1990). The presence of individual mass concentrations in source contribution estimates were displayed in the species concentration output. This display showed which sources didn't fit into the equation, or are missing. The R/U ratio explained the degree to which the calculated and measured concentrations differed, through uncertainty values.

These output files indicated the source contribution to ambient concentrations and the validity of data. The results obtained must comply with the pre-set ranges in order for the data to be useful and accurate for evaluation.

The CMB model is based on six assumptions (Watson et al., 1990). The postulation is that source emissions remained constant and that no interaction occurred between the chemical species during the sampling period. The model accepts that all contributing sources were identified and characterized and that the emissions were linearly independent of one another. The identified sources must be equal to, or more than, the, chemical species and all uncertainties were random.

2.3.2 Data capture of receptor samples

The ambient particulate matter data were used as an input file in the chemical mass balance model in order to illustrate the causes of pollution in the selected area. This particular sampling area was selected due to its location in the vicinity of several polluting sources such as mines, vehicle emissions and domestic fuel- burning.

The ambient samples were collected according to the Maenhaut method (Maenhaut et al., 1994). A SFU was used which contained two Nuclepore filters (Figure 2-6). This SFU concept allowed for the separate collection of different aerosol sizes. The 0.4 µm filter captured fine particles and the 0.8 µm filter collected coarse particles. The SFU was placed in a black plastic container that was kept upright on a 1.6 m steel tripod. A rain cover was necessary to prevent the samples from obtaining moisture from possible rainstorms. The SFU was connected to the monitoring station by plastic transparent tubing in order to allow suction from the Rotameter that enabled the sampler to calculate the exact amount of sampled air. An hour meter and gas meter indicated the time and the flow-rate of sampled air respectively. A total number of 32 samples were acquired simultaneously in the winter and 28 samples were collected in the summer. The duration for each sample was 12 hours from 06:00 am to 06:00 pm and vice versa. Sampling was undertaken during the winter and summer period to determine the impact of emissions from fuel-burning practices and the other polluting sources.

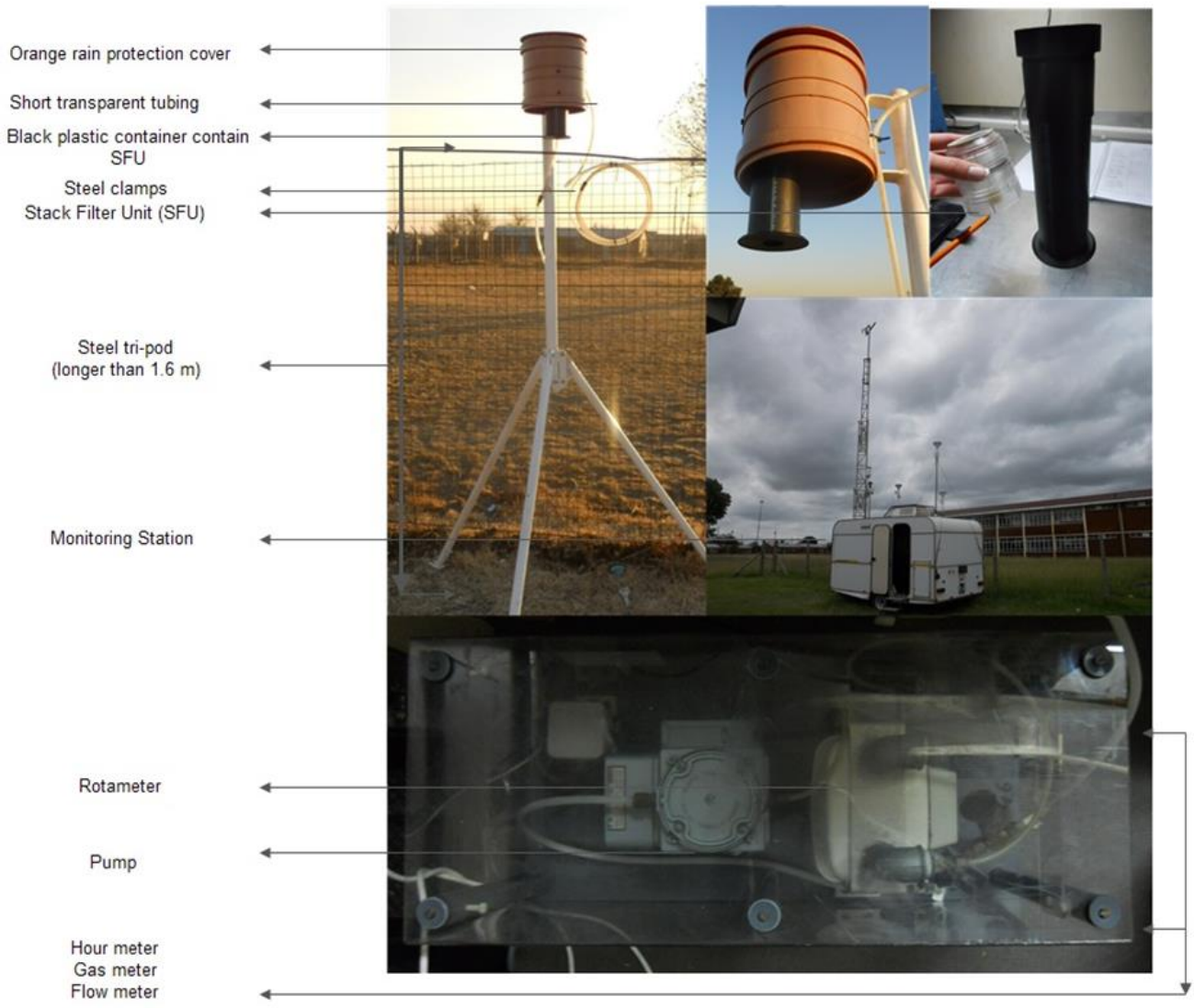

Figure 2-6: Schematic diagram of sampling PM_{10} and $PM_{2.5}$ according to the stack filter unit approach.

All possible unforeseen variables were noted in order to validate or explain results. An example of a variable was the power outages that occurred during this investigation. This influenced the volume of air sampled and the time duration of the affected sample. Nocturnal power interruptions of that ranged from 20 minutes to 1 hour occurred on 21/07, 24/07, 26/07 and 11/08. Load shedding occurred during the summer sampling campaign on 27/03. Other problems encountered include equipment failure on 27/07 during the winter. A new pump was installed 14 minutes later and sampling continued. Other interferences could have involved intrusions to the SFU, for example kids playing next to the sampling unit.

2.3.3 Source profiles

A variety of polluting sources emit particulate matter into the atmosphere. The sources responsible for possible aerosol loading in Kwadela included agricultural soil particulates, emissions from gasoline and diesel motor vehicles' exhausts, heavy-duty diesel vehicles, biomass burning, and particles from paved and unpaved roads, coal dust, wood and coal combustion as well as power plant fly-ash. The profiles of the chemical species contained in every identified polluter were included in the CMB model.

Source profiles were obtained from the USEPA database and from previous source apportionment studies. The reference and representativeness of each source profile were explained comprehensively in Table 2-1.

Table 2-1: Explanations of the representativeness of the selected profiles (Walton, 2013).

Source type	Source profile reference	Description
Soil	Engelbrecht et al., (2002)	Soil acts as sinks for atmospheric pollutants which lead to the accumulation of deposited pollutants over a long period (Quinn et al., 2009). The chemical compositions of soil from Sasolburg were subjected to pollution from Sasol, domestic coal combustion and other emissions (Maponya and Rampedi,

		2013). Similar polluting sources were identified in close proximity to Kwadela. This soil sample was collected 10 km south-east of Sasolburg and could thus be representative of the soil found in Kwadela.
Gasoline motor vehicle	Engelbrecht et al., (2002)	This sample was collected from the exhaust of an idling pick-up truck. The profile is representative of all the vehicles with petrol combustion emissions, passing on the N17.
Diesel motor vehicles	Profile 3462 and 3463 in the EPA database	Emissions form 174 diesel vehicles were sampled while trucks accelerated and decelerated at a constant speed. Six trials were conducted with all the trucks. This profile represents diesel motor vehicles driving past Kwadela.
Heavy duty diesel vehicles	Profile 322032.5 and 3220310 in the EPA database	This profile is representative of heavy duty trucks driving on the N17. Samples were collected from a dilution sampler and a constant volume sampler.
Marine aerosols	Profile 431010 and 431012.5 from the EPA database	These profiles were selected to present the possible occurrence of marine aerosols. A Cl: Na ratio of 1 and the other seawater elements were used as an estimate.
Biomass burning	Engelbrecht et al., (2002); Annegarn (1998)	This profile was chosen carefully to represent the smoke that results from field fires. A sampling campaign was conducted for grass that was collected in the Mpumalanga region. Another refuse profile was obtained from

		refuse-burning profiles sampled in Soweto.
Paved road	Profile 4346 and 4347 from the EPA database	Paved road dust is represented by a source profile compiled of five samples collected from San Antonio.
Unpaved road dust	Profile 4348 and 4349 from the EPA database	A composition of two profiles collected at Guadalupe was selected to be used as an unpaved dust profile.
Coal dust	Profile 212042.5 and 2120410 from the EPA database	This profile is a composite of PM that results from coal storage and handling.
Coal combustion	Engelbrecht et al., (2002); Annegarn (1998)	Four different coal profiles obtained through stack sampling in South Africa were used in the CMB. Low-income communities generally use poor D-grade coal for cooking and heating purposes. The first two profiles represented the pollutants found in ash, sulphur and volatile continents from combustion in stoves and braziers. The first profile was obtained from a coal stove's chimney and the second profile from a hood that was connected to a dilution stack sampler. The next coal profile was generated from three measured coal samplings in Soweto. Lastly, a negative organic carbon profile was obtained from sampling coal combustion practices.
Refuse/Wood combustion	Profile 423032.5 and 4230310 from the EPA database	A total number of sixteen samples were collected from eight sources and the average of the chemical species was calculated. Approximately 45% of the emissions were from fireplaces and 55% from stoves.

Power plant fly-ash	Engelbrecht et al., (2007)	Stack sampling (with a height of 200 m) was conducted at Lethabo Eskom Power Station with a dilution sampler. This sample was used to represent the Eskom power stations surrounding Kwadela within a 50 km radius.
Secondary particulates	Profiles 2540910 and 2540925 for ammonium nitrate as well as profiles 2541010 and 254102.5 for ammonium sulphate from the EPA database.	Secondary particulates are a result of chemical reactions and condensation of water vapours in the atmosphere. Because of the number of industrial sources and the possibility that chemical reactions may occur the ammonium nitrate and ammonium sulphate species were tested in the CMB.

2.3.4 Gravimetric analyses

Both size fractions of the Nuclepore filters were weighted individually prior and subsequent to the sampling period. The purpose of these analyses was to determine the mass of PM samples captured for each trial. All the samples were weighted in a stabilized environment with a micro-balance with a sensitivity of 1 µg (Figure 2-7). Samples were weighed 3 times and the averages of the before sampling weighs and after sampling weighs were respectively calculated. This balance provides stable accurate weighting results due the isolated weighting chamber. This balance has a temperature sensitivity drift of ± 1.0 ppm/°C and stability of ±0.0001%.

Figure 2-7: Mettler Toledo balance used to weight the filters.

2.3.5 Chemical analyses

Chemical analyses were conducted after the gravimetric analyses. These chemical experiments delivered the elemental composition data found in every ambient PM sample. The ICP-MS or XRF methods were used to obtain the elemental compositions and the ICS method was employed for the soluble fractions in ambient PM samples.

2.3.5.1 Sample preparation

Before the employment of the ICP-MS and ICS, pre-analysis had to be undertaken. The IC-preparation procedure involved extraction of particulates from the filter in a nitric acid and hydrochloric acid solution (van de Wiel, 2003). Prior to extraction, the nucleopore filter was cut in half in a stabilized controlled room. One half of the filter was placed in an Erlenmeyer flask for analysis and the other half was saved for other investigations. The filter was then boiled in 20ml nitric acid (HNO_3) and 40ml deionised water in order to extract the particulates. Subsequently, an amount of 5 ml concentrated HCl was added to the solution and the solution was refluxed for 3 hours (Figure 2-8). The solution was stirred continuously throughout the reflux procedure. Lastly, the solution was diluted up to a 100ml calibration mark of a volumetric flask.

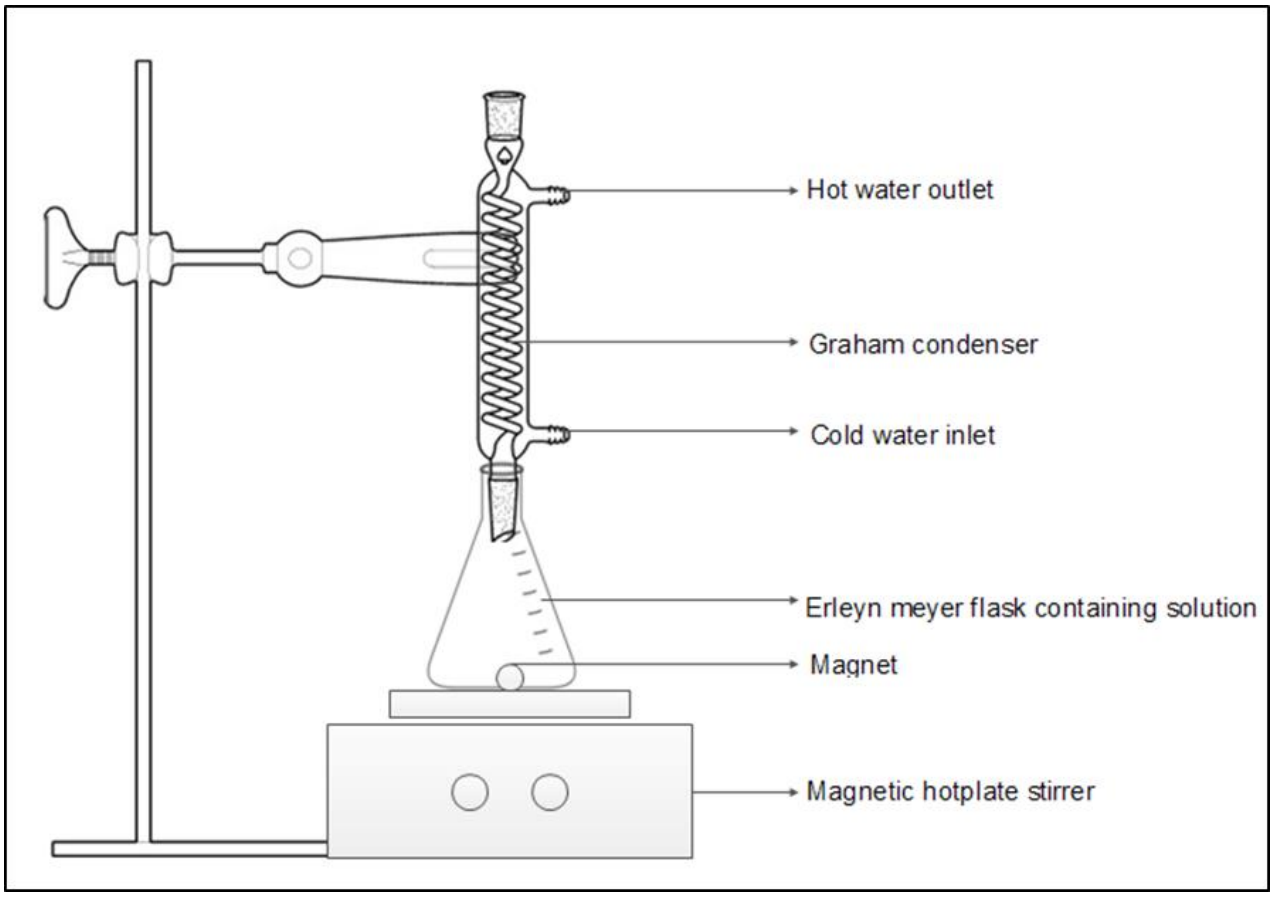

Figure 2-8: The ICP-preparation method while refluxing the solution.

2.3.5.2 The inductively coupled plasma–mass spectrometry method

ICP-MS involves extraction, dissolution or acidification procedures in order to free the PM in a solution. Solutions with a resolution range between 5 to 240 m/z (m = relative mass of an isotope; z = charge number) are typically analysed with this technique (Rüdel et al., 2011). The ICP-MS collects elemental information by joining high temperatures (± 5500 °C-15) with a mass spectrometer (Wolf, 2005; Ammann, 2007). ICP transfigures the atoms in the sample to ions, which are detected by the mass spectrometer after being fragmentised. This process involves the breaking of ion chemical bonding. The course of evaporation, separation of the atoms and the ionization can only take place in the presence of noble gasses such as argon (Pröfrock and Prange, 2012). The ion source with all the components along with the interference is demonstrated in Figure 2-9.

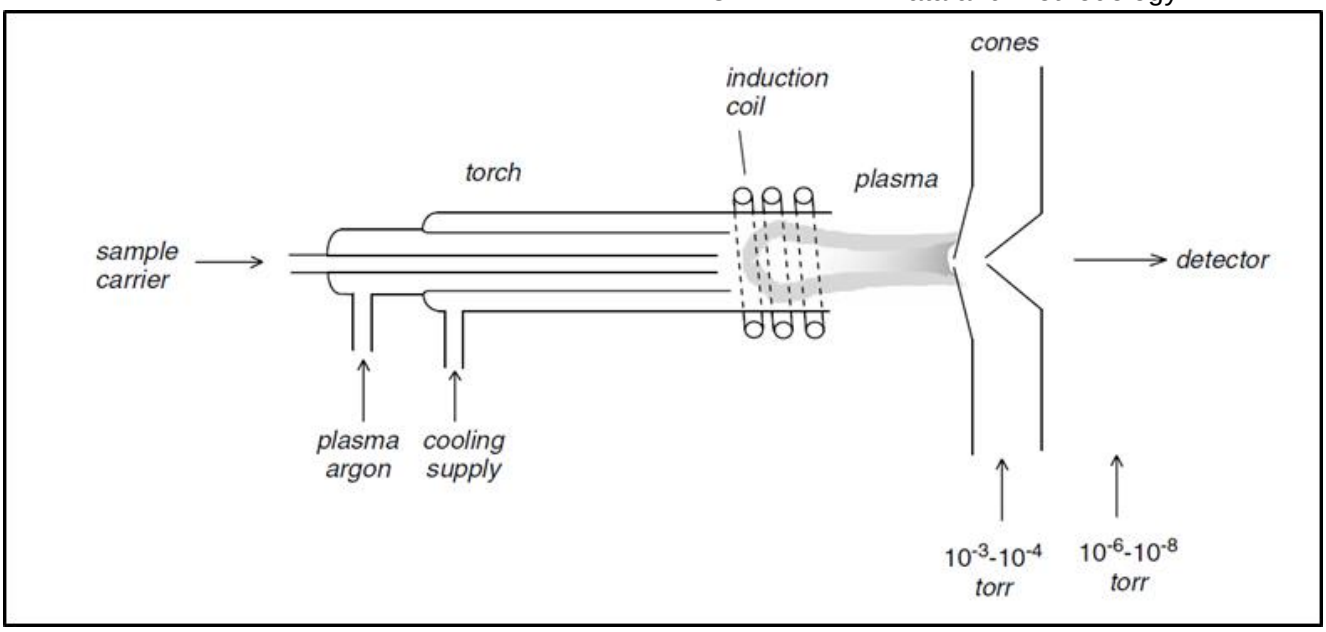

Figure 2-9: A basic illustration of the plasma source and an interference of the ICP- MS method (Ammann, 2007).

Interferences found in the ICP-MS procedure can be divided in two categories: spectral interferences and non-spectral interferences (Van de Wiel, 2003). Spectral interferences are divided further into sub-categories such as interferences from isobaric elements as well as doubly-charged ion and isobaric molecule interferences. Isobaric interfaces exist due to different elements which have similar mass-to-charge ratios. The ICP-MS has trouble separating these species due to insufficient resolution of the MS.

The method's incapacity to split elements is solved by using a correction factor for interfering elements (Table 2-2). The interferences caused by doubly-charged ions and isobaric molecules occur when ions with more than one atom or ionic charge are present in the solution. An example of such occurrence includes 40 calcium chloride ($CaCl^{+}$) ions. A reliable correction factor has not yet been found. Non- spectral interferences are linked to difficulties in nebulisation and transport processes as well as in the ion-transmissions. In order to prevent this interference, solid deposition should be avoided. The total solid levels of the sample analysed should not exceed the volume of 2.000 mg/L. In such a case the sample should be diluted to an adequate concentration.

Detection limits were a pre-requisite for obtaining elemental footprint data. The detection limits were determined by the laboratory and varied for each study because of differences in blank contamination and the laboratory facilities available.

These detection limits were used as an input file in the ICP-MS to identify and determine the mass of elements within the individual samples (Table 2-3). The output data of these analyses were the chemical species contained within each ambient air

2.3.5.2.1 Limitations of the inductively coupled plasma–mass spectrometry

Inaccurate data were obtained from ICP-MS analyses. The elemental species concentrations were approximately hundred times lower than other elemental concentrations used in previous source apportionment analyses such as a study conducted in Lephalale (Walton, 2013) and Soweto (Annegarn, 1998). A more detailed discussion follows in Chapter 4.

Table 2-2: Detection limits used for ICP-MS analysis.

Element	Mass (µg)
Ag	1.88E-05
Al	5.44E-04
As	1.01E-04
Au	7.52E-05
B	4.67E-04
Ba	1.32E-04

Be	1.90E-04
Ca	7.52E-03
Cd	1.28E-04
Co	2.92E-05
Cr	1.72E-04
Cu	4.60E-04
Fe	3.22E-03
Hg	5.08E-04
K	2.54E-02
Mg	2.87E-04
Mn	3.21E-04
Mo	1.70E-05
Na	1.06E-03
Ni	1.46E-04
P	6.88E-04
Pb	1.81E-04
Pd	2.75E-05
Pt	1.73E-04
Sb	2.05E-05
Se	2.38E-03
Sr	2.30E-05
Ti	5.38E-04
Tl	3.48E-05
U	1.94E-05
V	7.08E-06
Zn	6.78E-04

2.3.5.3 The X-Ray Fluorescence technique

XRF methods were applied to determine the trace elements for the summer. An interaction between x-ray photons and the elements in the PM species causes a discharge of electrons (RTI, 2009). This reaction is responsible for releasing x-rays that are unique for individual species. An X-ray is produced by a Coolidge tube by using an RH anode (Formenti et al., 2010). Inserting filters with different widths are used to control the spectrum. Differences found in the electron-binding energies between the two interacting

element are equivalent to the produced x-ray. The x-ray band is used to identify the element and the electron binding energies illustrates the atomic number. The quantity of an element is calculated by the measuring the intensity of the x-rays at a given energy point.

A simplified diagram of an XRF instrument is given in Figure 2-10. An anode that correlates to a certain target is paired for detection purposes. A spectrum is created on the computer screen to show the present species.

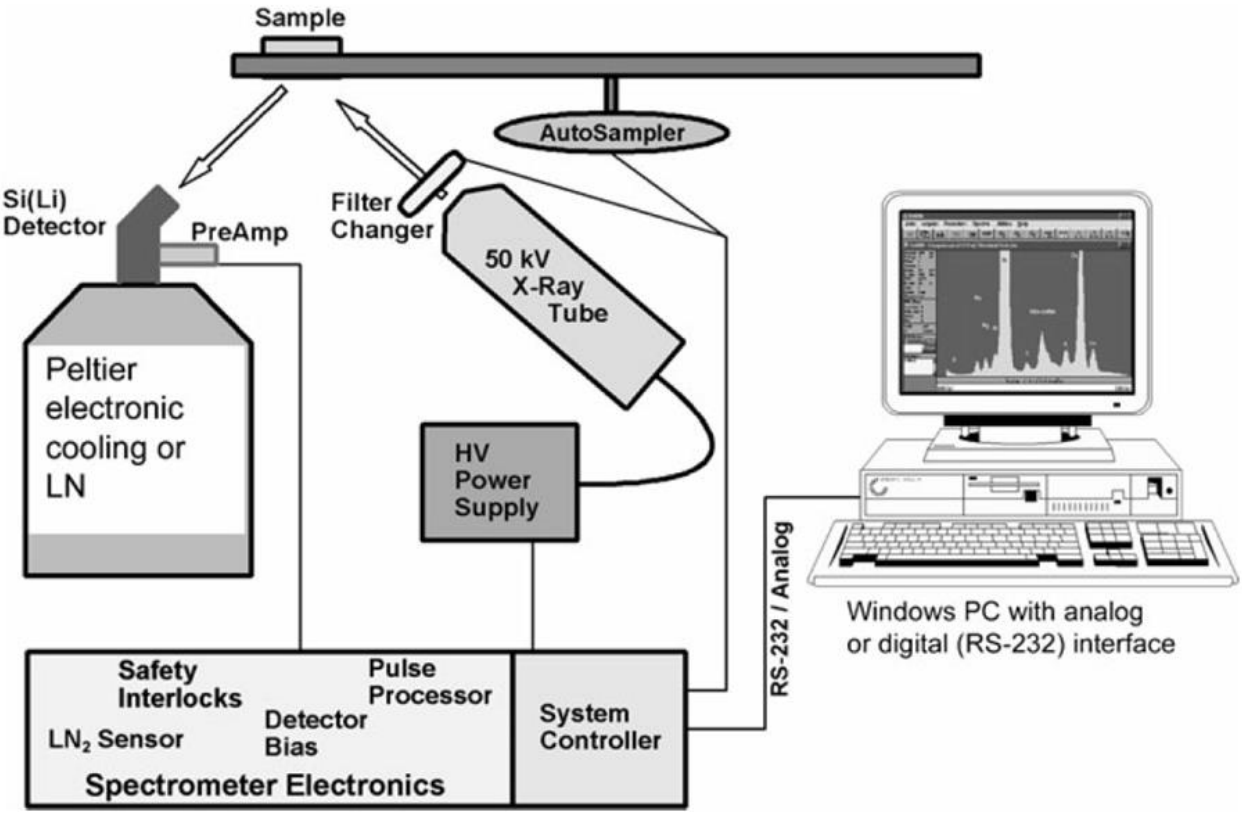

Figure 2-10: Systematic diagram of the XRF equipment (Shackley, 2011).

A pre-requisite of species identifications are the detection limits used. The detection limits for the XRF analyses are given in Table 2-4: Detection limits vary according to equipment used, species specifications and lab preferences.

Table 2-3: Detection limits used in XRF analysis.

Elements	Mass (µg)
Cl	0.2576
S	1.0304
Ca	4.3792

Fe	2.7048
Cd	2.336
Na	0.644
Mg	0.1288
Al	0.9016
Si	1.8032
As	0.9016
P	0.2576
K	0.2576
Ti	0.5152
Mn	0.2576
Zn	0.644
Cr	0.1288
V	0.1288
Pb	0.2576

2.3.5.4 The ion chromatography method

The ICS was used to perform ion analyses by applying a supressed or non-suppressed conductivity recognition (Dionex, 2009). This system contains an eluent to remove the ions from the liquid (Figure 2-11). The sample injection is where the sample is inserted into a sampling loop. The role of the pump is to separate the sample ions by pressuring the sample and eluent into different pathways. Consequently, the sample flows through a suppressor where the ions are detected. The detection process involves a conductivity cell where the electrical conductance of the sample produces a certain signal. This signal is then transmitted to a data collection region where the signal area is analysed and processed.

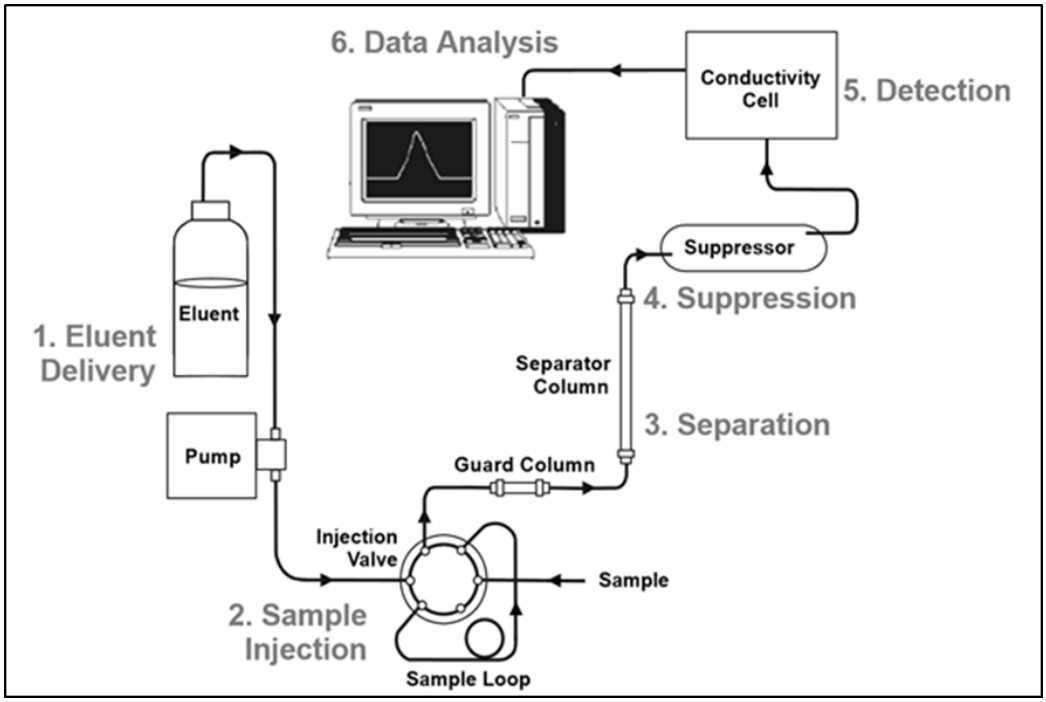

Figure 2-11: Systematic presentation of the ICS method (Dionex, 2009).

The detection limits are important for recognizing the ionic elements. These limits were used in the ICS equipment and for further analyses of the uncertainty of species (Table 2-5).

Table 2-4: Detection limits used for ICS analyses.

Ionic specie	Mass (µg)
Cl^-	21.24

Na^+	3.47
Ca^{2+}	3.32
F^-	1.55
NH_4^+	3.42
Mg^{2+}	11.04
K^+	2.88
NO_3^-	69.32
SO_4^{2-}	140.26

The ICS is an effective method for obtaining the anions and cations from the filter extracts (Makonnen, 2014). The ionic elements obtained from this technique were further converted into the pre-requisite requirements for the CMB. The data thus represent the ions found in the ambient PM samples.

2.3.5.5 Data manipulation

Data preparation and screening measures were undertaken for the ambient elemental concentrations and the detection limits. The first step involved replacing zero and negative values as well as the data species below the detection limit (BDL) (Polossar et al., 1998). These species were replaced by half the detection limit. The following step involved the conversion of the data into a suitable format, as required by the CMB model.

The receptor concentrations were received in an mg/L format and were converted to micrograms per cubic metre (µg/m³). The following formula was used for this calculation:

$$AS_{(\mu g/m^3)} = \left(\frac{As_{(mg/L)} \times Vs \times Df}{Sv}\right) \times 2$$

$AS_{(\mu g/m^3)}$ ambient sample converted (µg/m^3)

$As_{(mg/L)}$ chemical species of ambient sample (mg/L)

$Vs_{(L)}$ sample volume analysed (0.1 L)

Df = dilution factor for the analysed samples (1.54)

$SV_{(\mu g/m^3)}$ volume of air sampled per specie

Further conversion of detection limits were required to calculate the uncertainties of individual samples. The uncertainty values were needed alongside the ambient sample results in the CMB model. The XRF detection limits were received in µg/cm^2. This conversion involved dividing the limits by the filter area (Maenhaut, 1994) and the volume of air sampled. The detection limits for ICP-MS and ICS were converted from ppm (parts per million)/parts per billion (ppb) to micrograms per cubic metre (µg/m^3) by means of the following formula (UNEP and UNHSP, 2005; SKC, 2009):

$$DL_{(\mu g/m^3)} = \frac{\left(\frac{M_r \times DL_{(ppm)}}{24.45}\right)}{x}$$

$DL_{(\mu g/m^3)}$ detection limits converted to correspond to the sample units (µg/m^3)

M_r molecular mass per chemical species (g/mol)

$DL_{(ppm)}$ detection limits as used in analysis (ppm)

x volume of air sampled (µg/m$^{3)}$

Subsequently, a fraction was calculated for each species. These fractions were obtained by dividing the detection limit by the ambient chemical species.

The fraction values were categorized into three groups and a percentage value was applied to each fraction. Percentages were then multiplied by the individual species' data to determine the uncertainties of each sample (Table 2-6). Higher uncertainty values are ascribed to samples closer to the detection limits. The calculated chemical species of ambient samples and their uncertainty values were used as an input file in the CMB.

Table 2-5: Percentages applied to fractions.

Fraction per specie	Percentage (%)
0 – 0.3	5%
0.3 –0.8	10%
>0.8	30%

Ambient particulates, the source emission profiles and all the involved chemical species were filtered for modelling purposes (Watson et al., 1990). The percentage of species within each sample illustrated whether all the important species were detected. The percentage of species detected within each sample were calculated and compared to other samples. This calculation was done using the mass of chemical species and the total measured mass of the constituents in a sample:

$$Chemical\ species_{\ (\%)} = \frac{\sum Mass\ of\ detected\ elements}{TM} \times 100$$

TM total mass of $PM_{2.5}$/ PM_{10}

The last step of the screening process involved omitting species from the dataset with poor detection or with a species with numerous values BDL. This calculation is called the signal-to-noise ratio were the following formula was applied for every chemical species:

$$SN\ Ratio = \frac{\sum Specie\ concentration}{DL_{avg} \times Number\ of\ BDL}$$

DL_{avg} average of the detection limit

$Number\ of\ BDL$ number of the species below the detection limit

2.4. Meteorological overview

The evaluation of ambient particulate matter on a local scale should be accompanied by meteorological explanations. High wind speeds are associated with more suspended dust, (Egami et al., 1989). A greater dispersion of lower pollutants will also be a result of increased wind speeds. Temperatures influences the surface wind speed where hot surface mix with air masses and increase convection (Mugabo, 2011). Cooler temperatures on the other hand caused more pollution near the surface with associated lower wind speeds. Temperatures also influence the rate of chemical reactions. Relative humidity influences the rate of secondary species formation and chemical reactions that occurs in the atmosphere. These weather components included in this study are wind speed, wind direction, temperature and rainfall (Table 2-7). All the necessary equipment was installed in a mobile weather station at the secondary school in Kwadela.

The sampling rate for all the instruments was based on 1 minute intervals. Meteorology data that was measured during the winter sampling period (21/07 to 29/07 and 5/08 to 12/08 in 2014) and summer sampling period (27/03 to 13/04 in 2015) were extracted and used to explain particulate matter loadings.

Table 2-3: Instruments used for meteorological monitoring.

Parameter	**Instrument**	**Range**	**Precision**
Temperature	Vaisala HMP60	40 ◦C to 60 ◦C	±0.6 ◦C
Humidity	Vaisala HMP60	0 to 100%	±5%
Wind	RM Young 05103	0 - 100 m/s	±3 m/s or 1 %
Rainfall	RM Young 52203	0.1 mm per tip	2% to 3%

Day to day weather conditions are determined by subtropical, tropical and temperate features of atmospheric circulation patterns (Preston-Whyte and Tyson, 1989). Synoptic weather conditions are classified in three categories: fine-weather and mildly disturbed conditions, tropical disturbances associated with tropical easterly flow and temperate

mid-latitude disturbances associated with westerly flow. The synoptic patterns experienced during the sampling campaigns were compared to the ten patterns in Figure 2-12, to determine whether a link could be established between the amount of PM and the synoptic weather conditions.

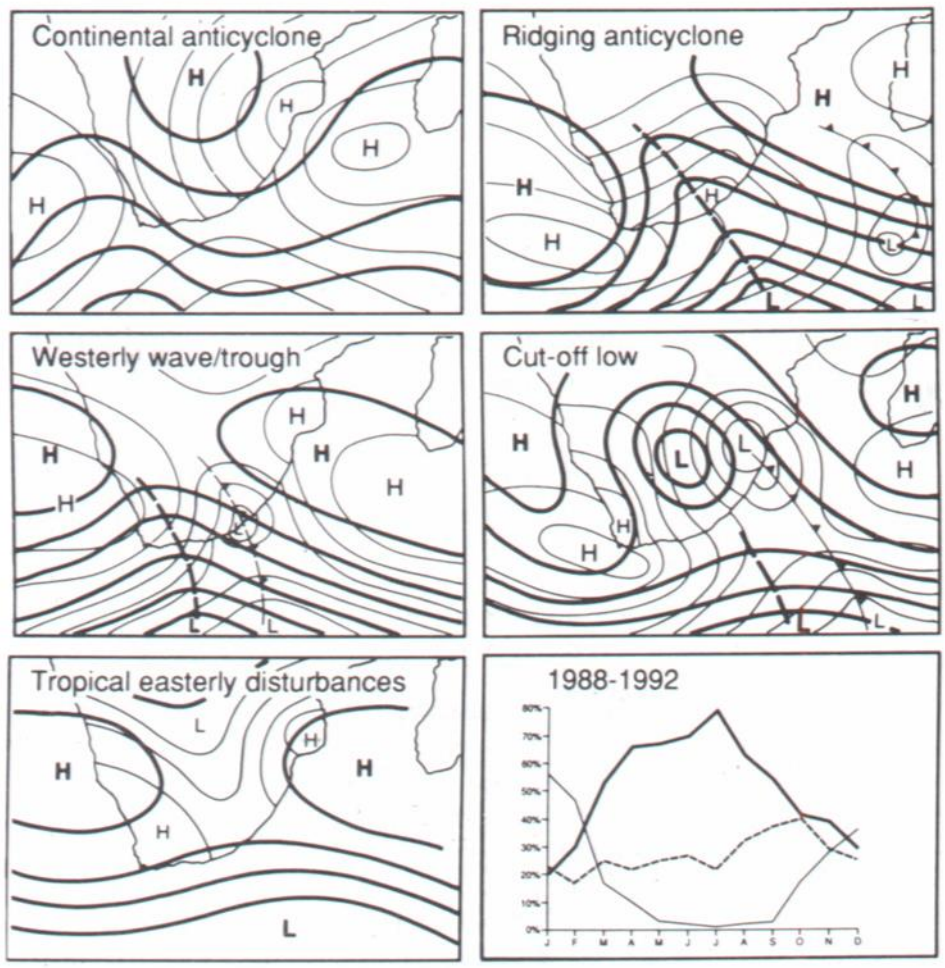

Figure 2-12: The major synoptic circulation types that influence South Africa's climate and their monthly occurrence (Pikteh, 2000).

2.5. Limitations of the methodology

Chemical analysis for ambient particulate matter was undertaken by means of the ICP-MS, XRF and ICS methods. The different methods that were followed caused variations in results. Different chemical elements were also researched for the two sampling campaigns.

In order to obtain comparable results it is important to be consistent in sampling techniques followed and the types of chemical elements tested.

CHAPTER 3:

DOMESTIC FUEL USE IN SOUTH AFRICAN LOW INCOME SETTLEMENTS

This chapter demonstrates the importance of domestic fuel burning practices in South Africa. The fuel use patterns are illustrated in a spatial context. Factors influencing fuel choice are defined and the fraction of the population exposed to these practices was calculated. This study investigated the first two research questions outlined in Chapter 1.

3.1. Background

Residential fuel combustion is one of the primary air quality concerns in South Africa. The mines in South Africa were conventionally blamed for the outdoor air pollution. Recent studies shifted these views towards the residential fuel combustion that occurs on a local scale. Serious environmental effects and chronic diseases are consequences of household fuel burning (Barnes et al., 2006). Emissions from these activities have received much attention due to major health impacts found within poor communities. The 2011 census data illustrated that 45.5 % of the South African population lived in poor communities and could be associated with fuel burning practices (SSA, 2014). These combustion activities are hazardous and highly polluting. Pollutants released from domestic fuel burning causes that the PM levels exceeds the national air quality standards.

Household coal consumption represents 2% of the total coal usage in South Africa, but is responsible for an estimated 25% of the total PM (Scorgie et al., 2004). Paraffin burning emits high levels of SO^2, CO and NO^2 into the atmosphere, which contributes to the amount of GHG (Bailie et al., 1999; Barnes et al., 2009). Indoor air pollution causes serious health problems because the combustion practices occurs within poorly

ventilated houses (DEA, 2008). An estimated 2489 deaths occurred in 2000 due to indoor air pollution (Norman et al., 2007). The indoor air quality of fuel burning households is far more polluted compared to the houses that use electricity. Residential fuel burning causes that people are being exposed to outdoor PM levels that is higher than the national standards for 20%-40% of the days per year. PM levels of an area are a good indicator of air quality. Health problems occurred in areas with high PM levels that range from irritations to serious chronic cardiovascular complications.

A variety of factors could be used describe the type of fuel used in poor communities. Previous studies showed that the former approach of explaining fuel choices by means of only the income levels is inadequate (Mekonnen & Köhlin, 2009). The motives behind fuel preference in developing countries differs geospatially, and can be attributed to a number of root causes. Identifying these factors is of upmost importance because communities in these countries suffer from more severe health problems. This can be explained by their increasing population densities, social influences, poor living conditions and the high rates of urbanization experienced (Matooane et al., 2004). The factors that influenced fuel choice in developing countries include their financial positions, seasonal changes, accessibility of fuel type, demographical characteristics and social-economic factors. The low income communities of a country such as Barkino Fase showed that the low income population relied on wood, while the higher income households preferred charcoal (Ouedraogo, 2006). Population groups of India and the Himalayas used non-commercial fuels such as animal waste, crops and dung when other fuels were too expensive (Sood & Mitchell, 2011; Mushtaq et al., 2014). A study in Bangladesh found that both, economical influences and seasonal trends determined the type of fuel used in communities (Begum et al., 2009). The fuel types that were preferred for cooking purposes in Ethiopia were a result of the type of food being cooked, the timeframe needed for cooking and type of appliance used (Mekonnen & Köhlin, 2009). The income levels of Zambian households and social-economic factors were defined as equal important determinants within the country (Nyembe, 2011). Previous South African studies identified poor electricity supply (Kimemia & Annegarn, 2012), economical factors (Nkomo, 2005; Nuwarinda, 2007; Pauw et al., 2008; Kimemia & Annegarn, 2012; Ogwuche & Asobo, 2013), the availability of cheaper low-grade fuel forms (Engelbrecht et al., 2002; Nuwarinda, 2007), seasonal variations (Barnes et al., 2009), population characteristics (Mushtaq et al., 2014), types of dwelling and

appliances (Scorgie et al., 2003; Pauw et al., 2008) as the factors that influences fuel choice.

This research explored the possible determinants behind fuel choices, towards characterizing typical fuel burning settlements and households in South Africa. The choice of fuel type used by households relates to, inter alia, the economic costs of the energy carriers and equipment, availability of fuel, proximity to sources and demographic characteristics. In order to mitigate air pollution it is important to have estimations on how many people are exposed to indoor and outdoor air quality.

3.2. Fuel use determinants

Driving factors could pressure households to use a specific fuel type. Examples of such determinants are the availability of resources, income levels, geographical variability, temperatures and population characteristics. One important determinant is the type of appliances used in a household (Pauw, et al., 2008). Traditional iron stoves, welded stoves or braziers were typically used for coal and wood combustion. A deleterious aspect of these stoves is the slow transfer of heat and longer burning periods are required for cooking. Iron stoves were used for thermal heat generation. Households preferred traditional stoves during the winter because it provided heat, warm water and the possibility of cooking food the same time. Stoves were passed on to family members, which means that poorly ventilated stoves and chimneys remained in use (Balmer, 2007).

Another influential factor of fuel choice is the type of dwelling (Scorgie et al., 2003). Household types in South Africa range from shacks in informal settlements to brick houses on their own yard. Informal settlements and traditional households are not always equipped with the required infrastructure to be fully electrified for all applications. Traditional households could thus be associated with solid fuel usages whereas brick houses are generally electrified.

Certain demographic characteristics, on the other hand, could also be used to identify fuel burning houses. The number of people living in a house showed a positive correlation against the usage of dirty fuel types (Nyembe, 2011). The gender of the

household head correspondingly played part in the fuel type preferred (Mekonnen & Köhlin, 2009).

Poor people are dependent on heating methods to stay warm during cold periods. The time of day and seasonal period showed which energy type was being used and the time of use. A previous study conducted in low income communities illustrated that fuels were more intensively used during the winter months and were more common in colder areas (Barnes et al., 2009).

The location and availability of resources in South Africa could indicate which fuel type were used where. The accessibility of energy carriers was seen as a key factor for determining the type of fuel used. Wood for example was used in rural areas, where available, and coal users were located around coal mines. Wood sources are limited in the grassland biomes (Figure 3-1). Coal mines are clustered in Highveld region with two mines in the Northern Limpopo. This figure implies that most of the coal users would be located in the Highveld. Animal dung would be a secondary choice in grassland areas where wood is unavailable.

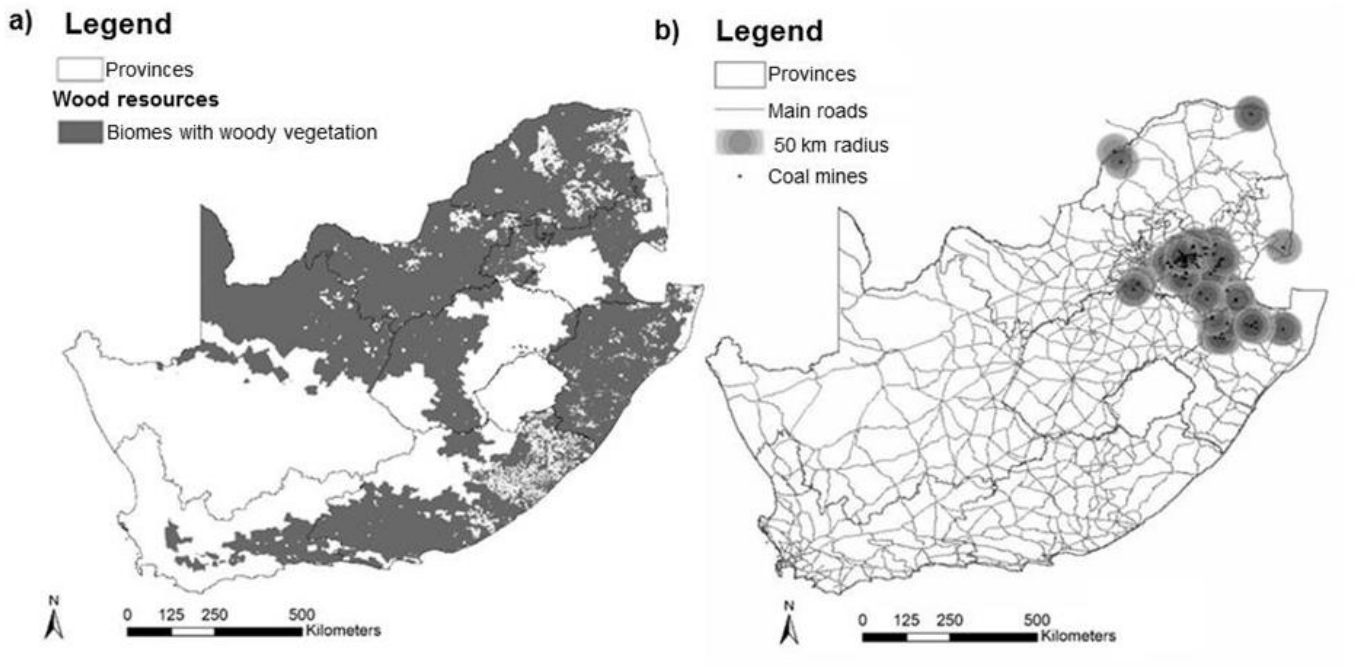

Figure 3-1: a) The spatial illustration of available wood resources and b), coal mines with an enclosing 50 km radius in South Africa.

3.3. South African fuel use trends

The 2011 census data illustrates that electricity supply to South African households has increased substantially since 1996 (Table 3-1).

Table 3-1: Energy usage trends for cooking and heating purposes in South Africa (Census, 2011).

	Electricity	**Gas**	**Paraffin**	**Wood**	**Coal**	**Animal dung**	**Total**
Cooking Fuels							
1996	4265305	286658	1943862	2073221	320830	106068	9059571
2001	5761355	284295	2394919	2292674	310058	110970	11205706
2011	10674090	507390	1227027	1806813	104157	45300	14447757
1996 – 2001 (% change)	35.07	(0.83)	23.20	10.59	(3.47)	4.62	23.69
2001 – 2011 (% change)	85.27	(78.47)	48.77	(21.19)	(66.41)	(59.18)	28.93
Heating Fuels							
1996	4030849	107690	1294963	2417723	735632	84447	9059571
2001	5493022	124982	1641457	2758863	734455	83058	11205706
2011	8503108	357063	1230222	2203384	293949	48252	14450162
1996 – 2001 (% change)	36.27	16.06	26.76	14.11	(0.16)	(1.64)	23.69
2001 – 2011 (% change)	54.80	185.69	(25.05)	(20.13)	(59.98)	(41.91)	28.95361

The census data showed that electricity usage for cooking applications have increased by 85.27% from 2001 to 2011 and by 54.8% for heating. Still, in 2011 52.49% of

informal and traditional households used energy alternatives for cooking purposes and 51.21% for heating. Gas combustion for cooking declined by 78%, but has increased by a high number of 185.69% for heating purposes. The usage patterns of cooking fuels displayed a faster declination rate than heating fuels. Even though a decrease in paraffin, wood, coal and animal dung for cooking were reported, this rate is not sustainable. Current electricity supply in South Africa is inadequate and more power outages occur. Thus, people will be obligated to rely on dirty fuels.

Electricity usage patterns and fuel burning trends for cooking purposes varied over the South African extend (Figure 3-2). The Eastern Cape, Free State, Kwazulu-Natal, Mpumalanga and North-West showed that the number of houses using electricity increased at a faster rate than the population growth. The main solid fuel type preferred in Mpumalanga, the Eastern Cape and Kwazulu-Natal were wood. The high portion of household's that dependent on paraffin decreased over the years in Gauteng and the Free State. Households in the North-West and Western Cape were more likely to use paraffin for cooking purposes. People in the Northern Cape tended to rely more on wood and paraffin compared to the other fuels. Limpopo had wood combustion occurrences which was meaningfully high, and should be addressed.

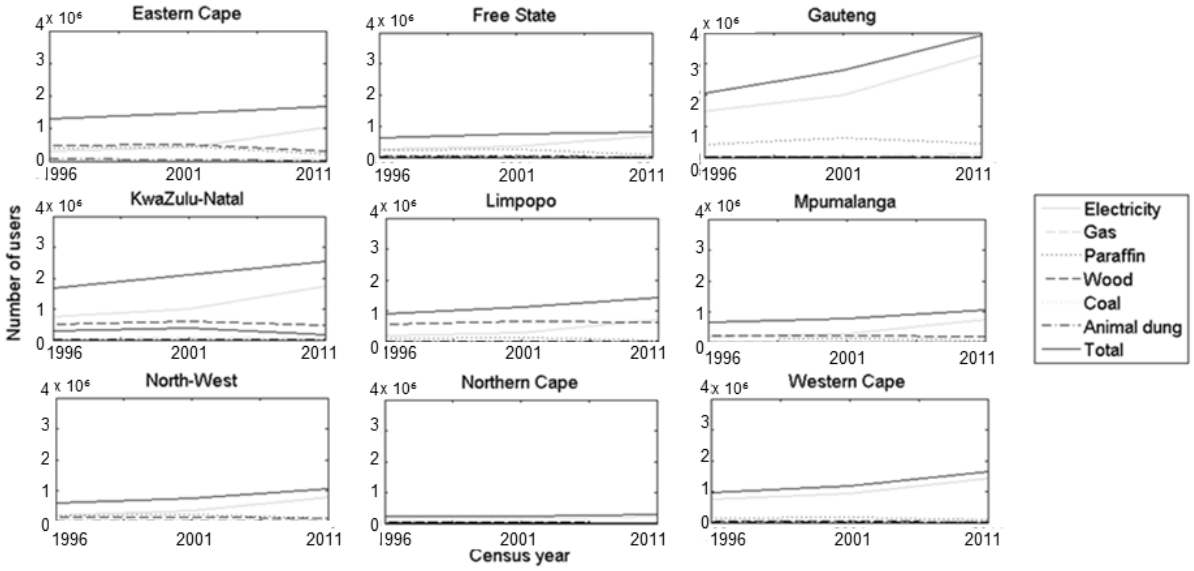

Figure 3-2: Energy usage trends for cooking in South African provinces.

The combustion of fuels for heating and cooking purposes showed similar patterns throughout the different provinces. The only difference between these trends was that the electrification rate for heating purposes was slower. In order to manage fuel combustion in SA the reason behind fuel choice should be explained.

3.4. Methodology

The South African census data (2011) was analysed on the SAL scale for the purpose of this study. The spatial information of coal mines, temperature and vegetation data were obtained from GIS datasets. A correction factor was used to take the people that used alternative energy types as secondary or tertiary fuels into consideration. This additional calculation was conducted because the census questionnaire only allowed the selection of primary fuel types used, which masked the other fuel types. Household use different fuels types to suit their energy needs (Barnes et al., 2009).

Regression modelling was conducted to compare the determinants of fuel choice to the number of houses that used a certain type of fuel on a SAL scale. This analysis were undertaken to determine the weighted influence of every factor. Determinants of both the heating and cooking fuels were considered. The parameters used in this statistical analyses were the number of people living in a household, the gender of the household-head, annual household incomes, the number of people with chronic diseases, appliances used for cooking, enumeration type of the residence, as well as the temperatures, vegetation and coal mines. The factors which illustrated the best correlation were further investigated.

Firstly, the spatial distribution of fuel burning houses was examined. The percentage of coal, wood, animal dung, gas and paraffin users were calculated by dividing the number of households that used a specific fuel type by the total number of houses in that SAL. These results were spatially illustrated to indicate the location of fuel burning settlements. Consequently, household income levels as determining factor were studied. Annual household income information was categorized in classes that range from households with no income up to income classes higher than R2 457 600. Every fuel was separately categorized in order to link income levels to a certain fuel use. The

percentage of users in an income group was distinctly calculated for every fuel type. A different approach was then used to determine the influence of family sizes on the fuel type preferred. The number of people living in a household was classified into classes that range from one person in a house, up to family sizes larger than ten people. Then, the percentage of users was obtained by dividing the number of households using a specific fuel by the total number of households in that category.

Lastly, number of people exposed to domestic fuel burning emissions was calculated. People affected by indoor and outdoor air pollution were distinctly determined. Houses that rely on fuels as primary energy source was multiplied by the average number of people living in a house. This estimation showed the number of residents whom are exposed to indoor air pollution. Settlements (small area layers) where more than 30% of the households burn energy alternatives were classified as a domestic fuel burning settlement. The fraction of people exposed to outdoor air pollution was calculated within these SAL's. These indoor and outdoor pollution results gave an indication of the lower limit of people that is exposed. The upper limit was calculated by multiplying these values by a correction factor of two. This calculation was conducted to include the users that depend on fuel as a secondary energy source.

3.5. Results

The following map (Figure 3-3) displays all the small area layers where 80% or more people relied either on wood, coal, paraffin or dung for cooking and heating purposes. Areas that could be linked to high fuel usages include the north-western part of North-West, South-east in Mpumalanga, patches in Kwazulu-Natal and the East Coast. More people used fuels for heating than for cooking. Referring to the Northern-Cape, it is clearly illustrated that more people used energy carries for heating purposes. The Kwazulu-Natal had the largest fraction of settlements that depends on dirty fuels. These settlements could be classified areas with high air pollution levels.

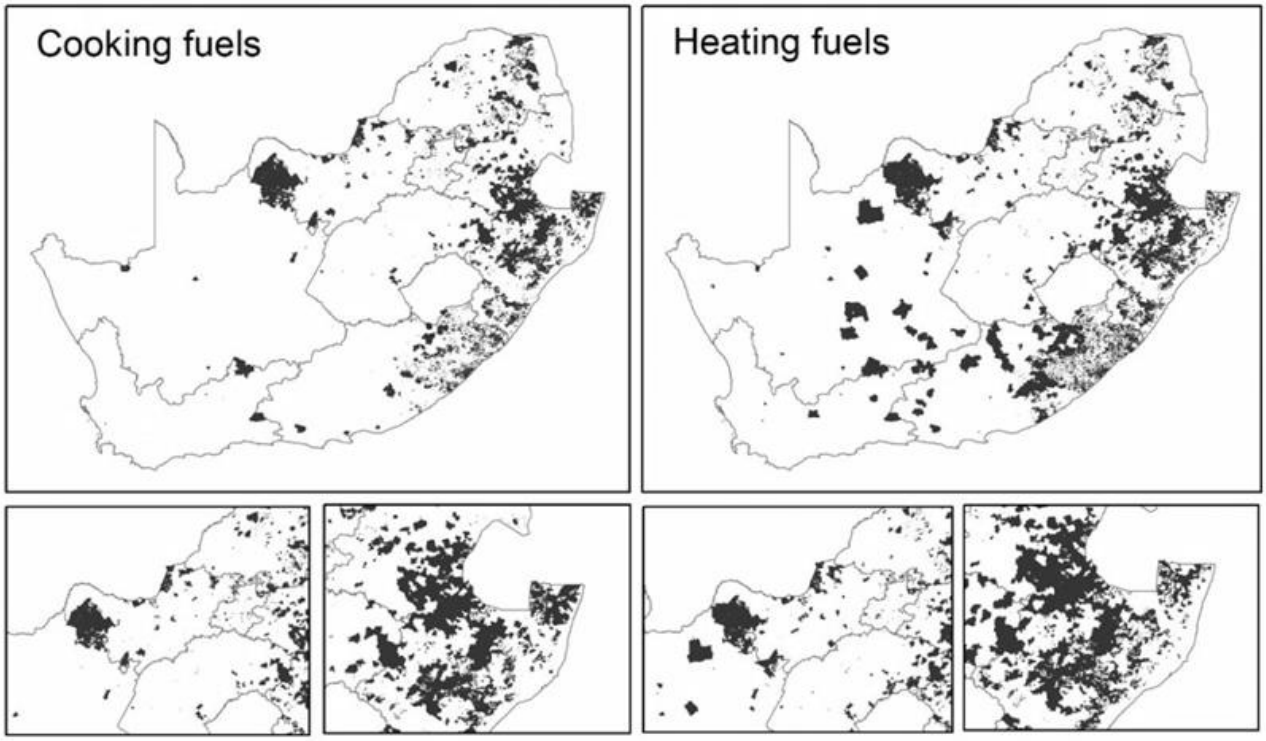

Figure 3-3: Areas where more than 80% of the population combust dirty fuels for cooking and heating.

Houses that use energy alternatives for cooking purposes are spatially displayed in Figure 3-4. This graph also represented the fuels combusted for heating purposes due to the similarities between the results. The percentage of fuel users (coal, wood, gas, paraffin and animal dung) was calculated for every individual fuel. People in the Western-Cape are associated with fewer combustion occurrences, compared to the rest of the provinces. Coal was mostly used in the Highveld region and in the Northern Cape. Wood was the most popular fuel type burnt across the South African extend. Gas was the preferred fuel choice in the Northern Cape and was also very abundant in a small area of Kwazulu-Natal. The distribution of paraffin indicated that it was widely applied for cooking purposes in the eastern areas of South Africa. Animal dung was the least popular fuel type and was burnt in parts of the North-West, Mpumalanga, Free State and the Eastern Cape. This fuel type was a secondary choice and only used when other fuels were inaccessible, or too expensive.

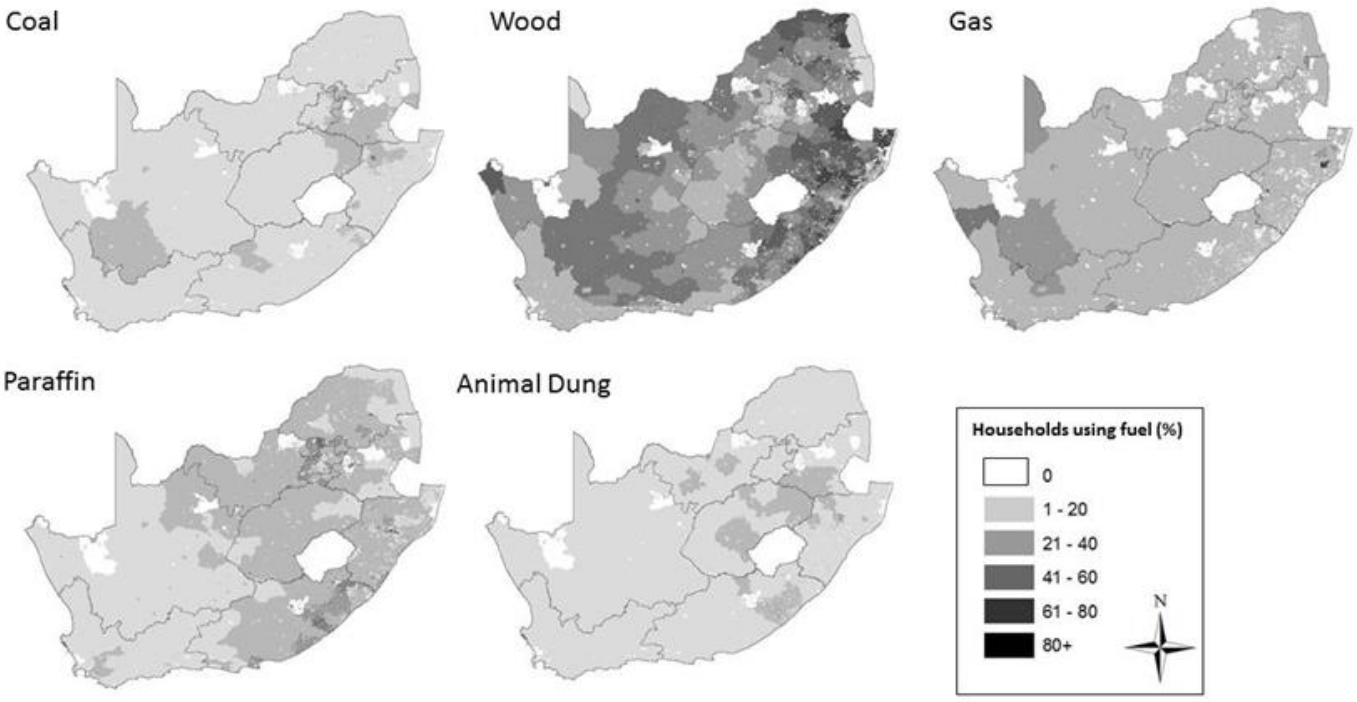

Figure 3-4: The percentage of cooking fuels used in South Africa for the different fuels types.

Income levels were used in developing countries to explain fuel use tendencies (Ouedraogo, 2006, Pauw et al., 2008, Sood & Mitchell, 2011; Ogwuche & Asobo, 2013, Mushtaq et al., 2014). Most of the people that burnt dirty fuels in South Africa were poor (Figure 3-5). The usage of fuels for cooking and heating applications illustrated similar tendencies, but the general income levels of heating users were higher. Having fuels for heating is not a necessity, but a luxury. This cumulative chart shows the percentage of fuel users versus the annual household incomes. Animal dung and wood were chosen by the poorest fraction of the population. The top 80% of wood, paraffin and animal dung users were between the income classes of R14 600 and R28 900. Furthermore, 80% of coal users were found in income classes up to R28 900 per year, the electricity users in the income class of R115 100 and gas in R230 700.

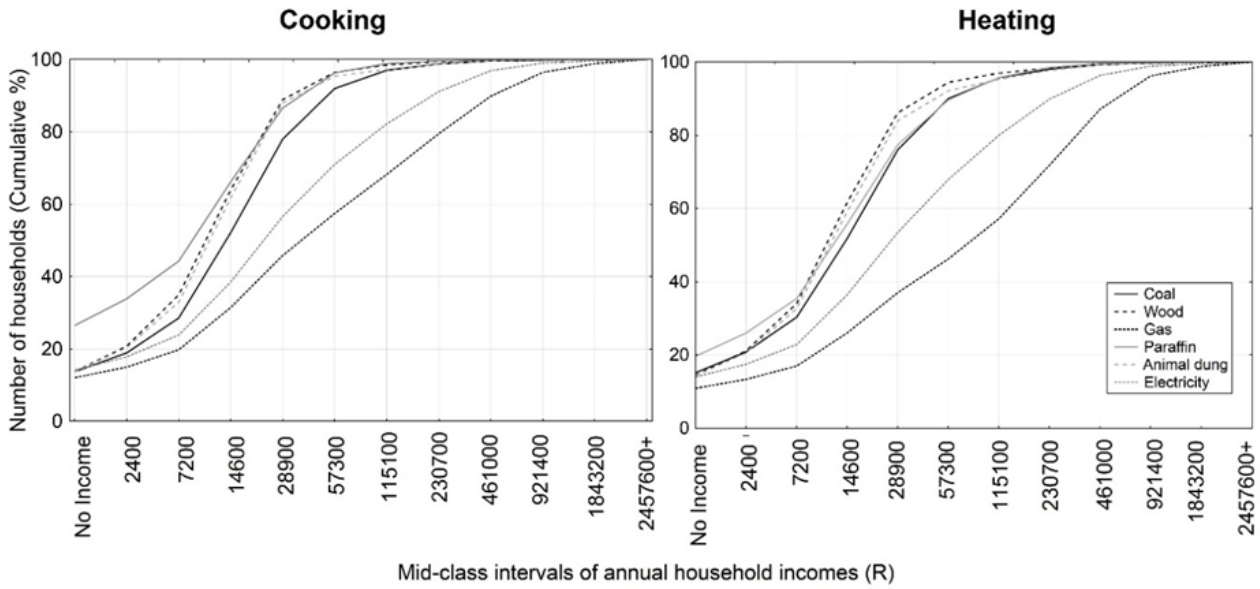

Figure 3-5: The percentage of solid fuel users per income class in South Africa for; a) Cooking purposes and b) Heating purposes.

Significant differences in the fuel user rates were found as the family sizes increased (Figure 3-6). The following graph displays the percentage of households that uses a certain fuel type versus the family size in one household. The number of households using coal, wood and animal dung for cooking increased as the family sizes increased. Paraffin and gas were more commonly combusted in households with fewer members. The trends of heating fuels only differed from cooking fuels by the number of users.

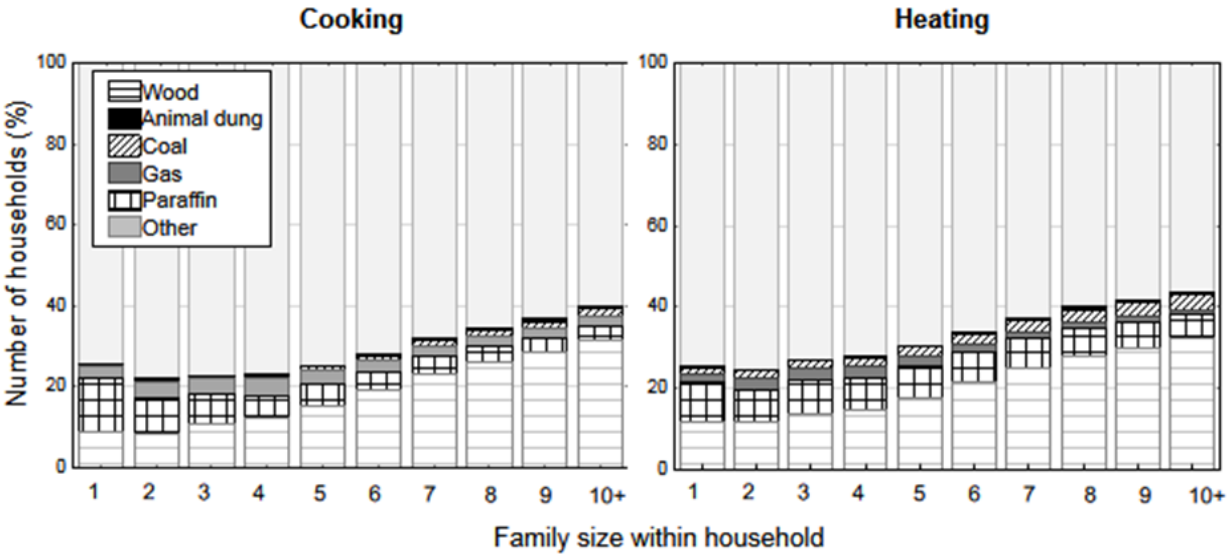

Figure 3-6: The number of households (%) that uses a certain fuel type versus the family size within house.

South African fuel determinants for cooking and heating practices illustrated a clear pattern versus the number of users. The r-square results showed that the most important characteristic of typical fuel users were the type of dwelling (informal and traditional houses). Fuel burning settlements had a positive correlation against the family sizes of the households, appliances used, the gender of the household head, annual household incomes as well as the number of chronic disease cases. The r-square value of these factors differed for every fuel type, which means certain fuels were more influenced by particular factors than others.

Health impacts that result from domestic fuel burning are a serious problem in South Africa. A high number of 3 683 961 households burned dirty fuels for cooking purposes and 4 123 251 houses for heating. Thus, the average number of people exposed to indoor air pollution is 12 570 994 for cooking purposes and 14 199 261 for heating. These results showed that in 2011 approximately 27.4% of the population was exposed to indoor air pollution. When looking at settlements (SAL's) where more than 30% of the households burnt dirty fuels, 29 562 were identified for cooking and 34 985 for heating. The number of people living in these areas whom were exposed to outdoor air pollution is 15 833 055 for cooking and 19 148 085 for heating. More than 28 398 522 people are influenced by indoor air pollution from heating purposes which is 54.8% of the population according to the upper limit calculations. The upper limit of people exposed to outdoor air pollution from heating practices was 38 296 170 persons which was 73.9% of the total population.

3.6. Discussion and concluding comments

Domestic fuel burning is a serious air quality problem in South Africa which needs to be addressed. The spatial distribution showed clear patterns in the types of fuel used. This result refers to the first research question. Numerous people are continuously exposed to indoor as well as outdoor air pollution, with serious health consequences. The second research question was directed at the exposure rates. The high need for addressing these emissions are emphasized by the high number of people affected. Certain predictor factors could be used to identify typical fuel burning households, and applied in mitigation strategies. A large fraction of people whom used dirty fuels were found in

informal or traditional settlements with low annual incomes. People that used fuels for heating purposes were generally richer than people relying on fuels for cooking. The number of people living in a household tended to increase as the number of residential fuel burners increased. Furthermore, the spatial illustration of fuel types used clarified that locality of communities' influences the fuel type preferred. The availability of resources was an important factor affecting choice. Certain areas were identified as high residential combustion zones whereas other areas had fewer combustion cases. All these aspects are important for the offset policy towards developing new standards and strategies for air quality management.

CHAPTER 4:

SOURCE APPORTIONMENT OF AMBIENT PARTICULATE MATTER IN KWADELA, MPUMALANGA

This chapter provides an overview of the ambient air quality situation in Kwadela. The method of aerosol sampling is described and the characteristics of particulate matter are given. Sources contributing to the air pollution situation are discussed.

4.1. Background

Developing countries often struggle with air quality problems because of residential fuel burning-practices. The South African industrial and low-income residential areas regularly experience atmospheric aerosol levels that exceed the national ambient air quality standards (Engelbrecht et al., 2002). These elevated aerosol loadings are mainly caused by power stations, mining and domestic fuel combustion emissions. The Department of Environmental Affairs (DEA) aims to maintain ambient PM_{10} and $PM_{2.5}$ levels that are below the limits of 40 µg/m^3 and 25 µg/m^3 per year (DEAT, 2009). Air quality in overpopulated residential areas is in such a poor state that citizens, particularly those in poor communities, might be exposed to PM levels that are harmful to their health and well-being (DEA, 2013). The vulnerability of these communities lies in their low living standards and lack of proper medical treatment (Matooane et al., 2004). The health impacts caused by PM have highlighted the need for proper mitigation strategies. The NEM:AQA (DEA, 2004) states in Chapter 3, (16)(iv) that emissions from fossil fuels in residential applications should be addressed. Other evidence illustrates the need for such measures, for example the high recorded GHG levels. South Africa is ranked fourth in the world with regards to the per capita CO_2 levels in the household combustion sector (Nation Master, 2010). In order to effectively address air quality problems in South Africa, analyses must be conducted at specified temporal, societal and spatial scales. Air

pollution from source emissions differs spatially, with varying contributions from each source. For the purpose of this research the ambient air quality of a small low-income community was analysed in order to identify the main sources to air pollution in the area.

4.2. Methodology and data

Ambient air quality sampling was undertaken in a low income community, Kwadela to determine the main air polluter on a local scale. This residential is notorious for the coal combustion practices that take place. In order to identify the influence of domestic fuel-burning on local air quality compared to the nearby emissions from coal mines and Power Stations, source apportionment techniques were used. This low income community was selected due to the number of people depended on coal sources. Coal is the main fuel type used for cooking and heating purposes. 48.5% of Kwadela's residents use coal for cooking and 45.6% uses electricity (Piketh and Burger, 2013). Coal is selected by 52.9% of the residents for heating purposes and 39.1% of the people prefer electricity. Emissions from coal-burning will thus influence the air quality in Kwadela.

Ambient PM was sampled during a winter and summer campaign at the Kwadela Secondary School monitoring station (26.463290° S, 29.663457° E). Data was collected during the winter from 21/07/2014 to 29/07/2014 and from 05/08/2014 to 12/08/2014. The summer sampling extended from 27/03/2015 to 01/04/2015 and 07/04/2015 to 13/04/2015. Sampling was conducted in a small township, Kwadela. The meteorological conditions of Kwadela were analysed to explain differences found in the amount of PM and the different chemical compositions. The weather parameters analysed in this study included wind speed and wind direction, atmospheric pressure, temperature as well as the amount of rainfall experienced. A mobile monitoring station equipped with all the required meteorological instruments was employed during this research. Instruments used included the Vaisala HMP60 for temperature and humidity measurements, the RM Young 61302 for atmospheric pressure, the RM Young 52203 for rainfall and the RM Young 05103 for wind measurements. Sampling was conducted on one minute time intervals. Synoptic weather maps were evaluated to determine whether the climate patterns had an influence on the amount of suspended PM.

Ambient particulates were sampled by means of the Maenhaut (Figure 4-1). A SFU contained two Nuclepore filters; the 0.4 µm and the 0.8 µm. The 0.4 µm filter was used to capture fine particles and the 0.8 µm collected coarse particles. Sampling was undertaken for a twelve hour period, and collected every day at 06:00 am and 06:00 pm respectively.

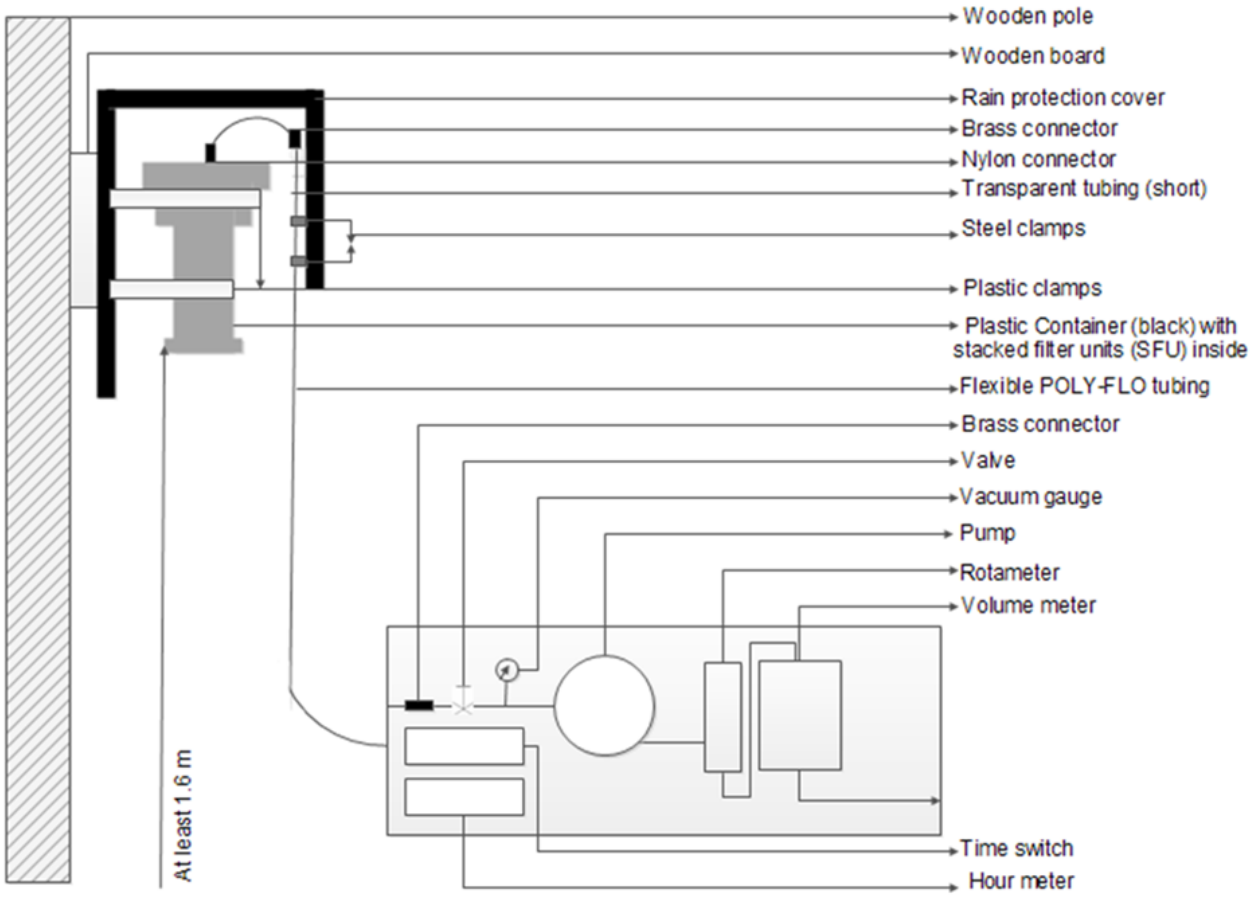

Figure 4-1: Schematic diagram of sampling coarse and fine PM using the SFU approach.

Power outages occurred regularly in Kwadela throughout the winter sampling period. All the power outages were recorded and the influence on the volume of air sampled was adjusted. The longest power failure occurred during sample 12, with an approximate two hour electricity deficiency. Other samples with an estimated hour electricity shortage included samples 1, 7, and 32. All the power shortages were taken into account during data evaluation.

The chemical and elemental profiles of potential sources in Kwadela were obtianed from either previous analyses (Engelbrecht et al., 2002; Walton et al., 2013) or from the USEPA database. Thirteen sources were identified that could influence the air quality of Kwadela. The sources identified were agricultural soil, emissions from gasoline (petrol), light duty diesel and heavy-duty diesel motor vehicles, marine aerosols, biomass burning, paved and unpaved road dust, coal dust, residential coal combustion, refuse/wood combustion, power plant fly ash and secondary particulates. The reference and representativeness of these profiles were comprehensively explained in Chapter 2.

The Nucelopore filters were weighed prior and subsequently to the sampling period to determine the gravimetric weights of every sample. The masses were used in the CMB to determine the amount of species from distinct polluting sources. The filters were then sent for chemical and elemental analysis to determine the chemical and elemental species present in the samples as well as the concentration of each individual species.

The elemental components of the samples were obtained using either ICP-MS or XRF and ionic elements using the ICS method. A pre-requisite of the ICP-MS/ICS methods was to extract all the chemical species from the filter into a solution. Prior to the preparation procedure the solutions were analysed with ICP-MS and ICS equipment. All these procedures were conducted in a stabilised room to prevent any contamination. The results of particle composition analyses were necessary for identifying all the polluting sources involved (Begum et al., 2007). The CMB was used to apportion the pollutants in ambient PM to their sources. The main source that contributed to ambient concentrations was shown by means of the SCE values. Differences that were found in results were explained by the meteorological conditions, emission rates and varying chemical compositions.

4.3. Results and discussion

4.3.1 Meteorology overview

The meteorological conditions for the winter and summer sampling campaigns are given below. The measured mean, minimum and maximum values were provided for wind speed, temperature, relative humidity and pressure. The average diurnal wind speeds for

the winter varied between 3 m/s and 4 m/s (Figure 4-2). A minimum temperature of 0° C and a maximum of 30°C were recorded during the sampling period. The average relative humidity ranged from 20% to 70%. No pressure data were recorded during this period.

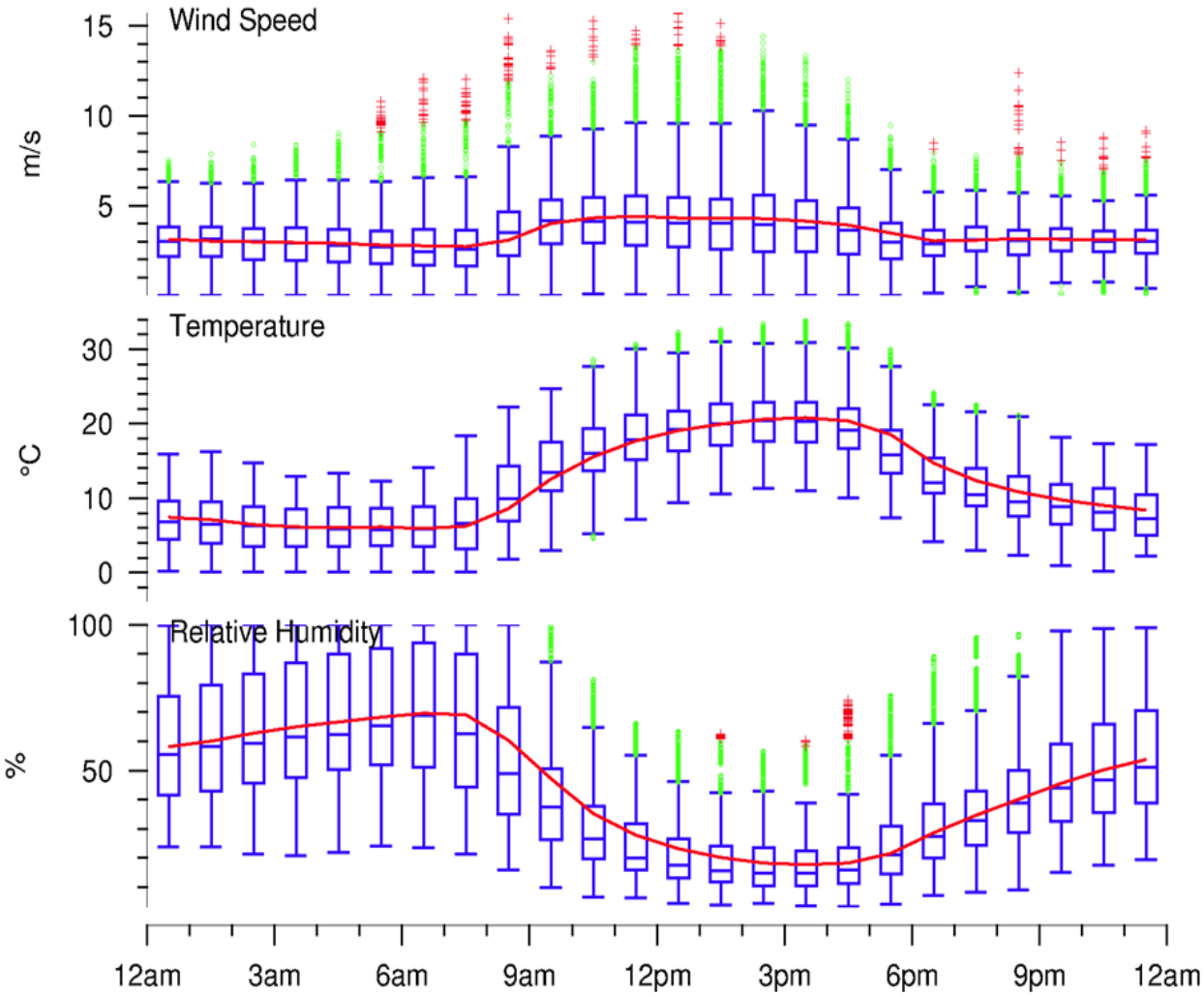

Figure 4-2: The meteorological conditions of Kwadela experienced during the winter 2014 (Burger and Piketh, 2015). Averages are indicated by the red line, a box and whisker plot is indicated by blue and the outliers by green.

Similar to the winter, recordings of the mean wind speed in the summer was between 3 m/s and 4 m/s (Figure 4-3). Temperatures were much higher during the summer with a minimum of 4°C and a maximum of 32°C. The average relative humidity varied between 45% and 85% and the ambient pressure between 832 hPa and 834 hPa.

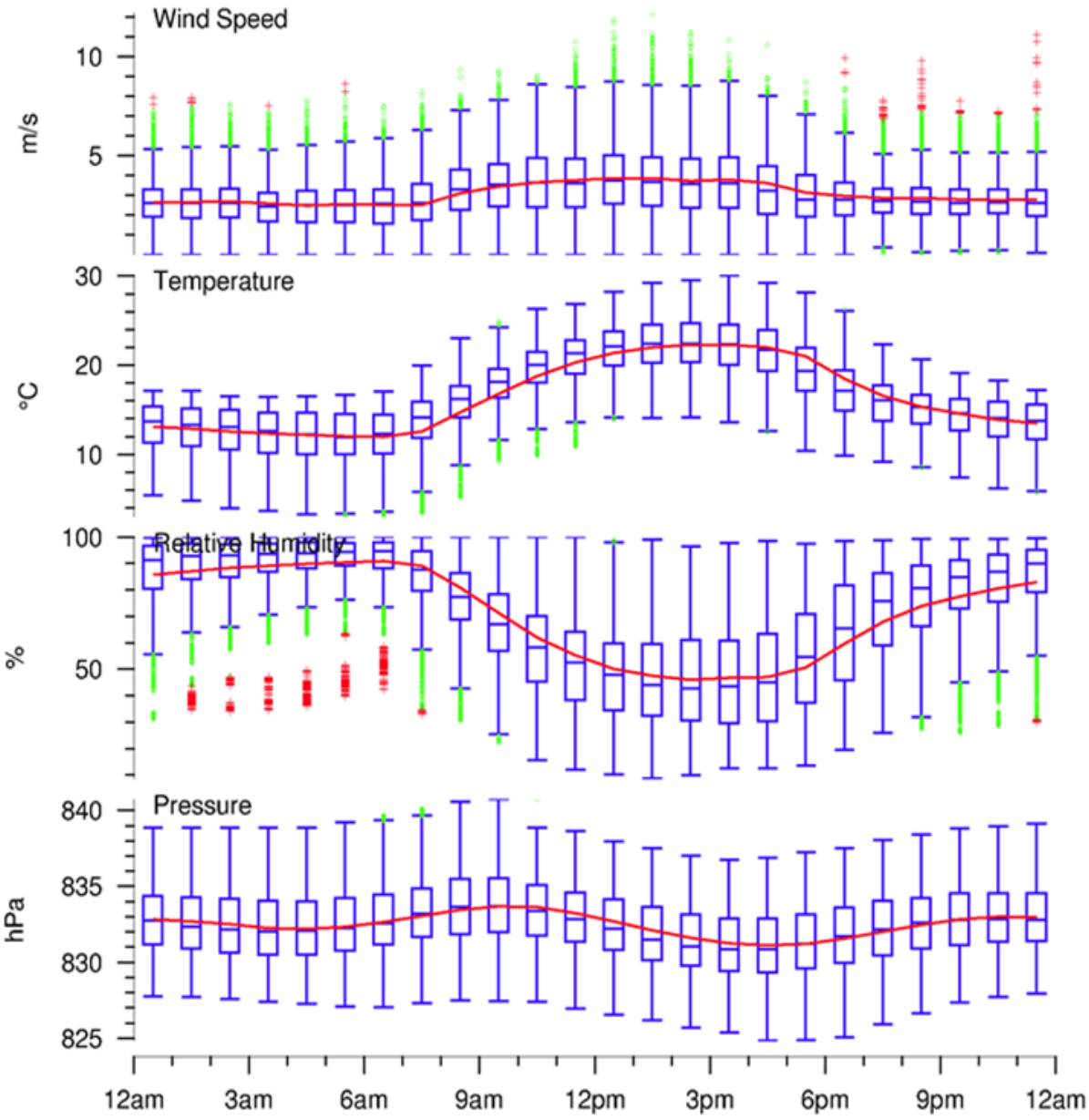

Figure 4-3: The meteorological conditions of Kwadela experienced during the summer 2014 (Burger and Piketh, 2015). Averages are indicated by the red line, a box and whisker plot is indicated by blue and the outliers by green.

The wind speed and direction are illustrated for both the sampling periods. The main wind direction is illustrated on a map of Kwadela, to show the possible influences of the different polluting sources. Wind was predominantly experienced from a north-western and west-north-western direction during the winter (Figure 4-4). This figure shows that emissions from Syferfontein and Forzando mines had an influence on Kwadela's air quality.

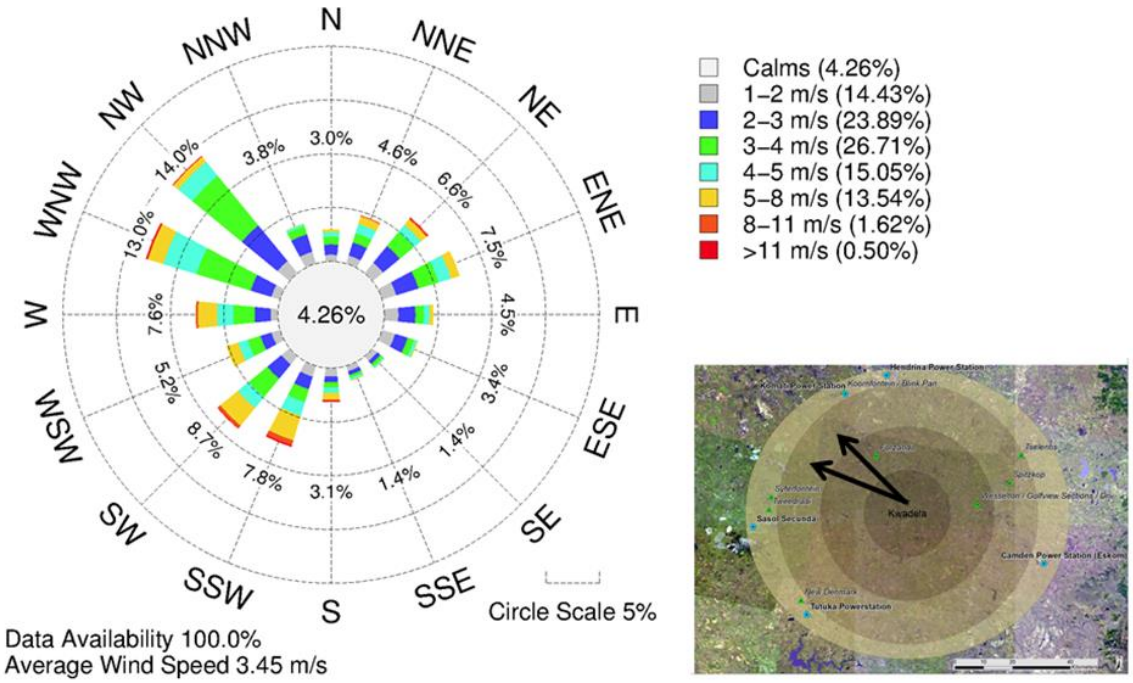

Figure 4-4: a) Wind direction and speed recorded during the winter 2014 at Kwadela (Burger and Piketh, 2015). b) The insertion indicates were the wind is from in accordance to Kwadela and the coal mines and power stations in proximity.

North-westerly winds occurred 12% of the campaign and easterly winds dominated with an occurrence of 13.1% (Figure 4-5). The possible air pollution sources to consider were Syferfontein, Spitzkop and Tselentis mines.

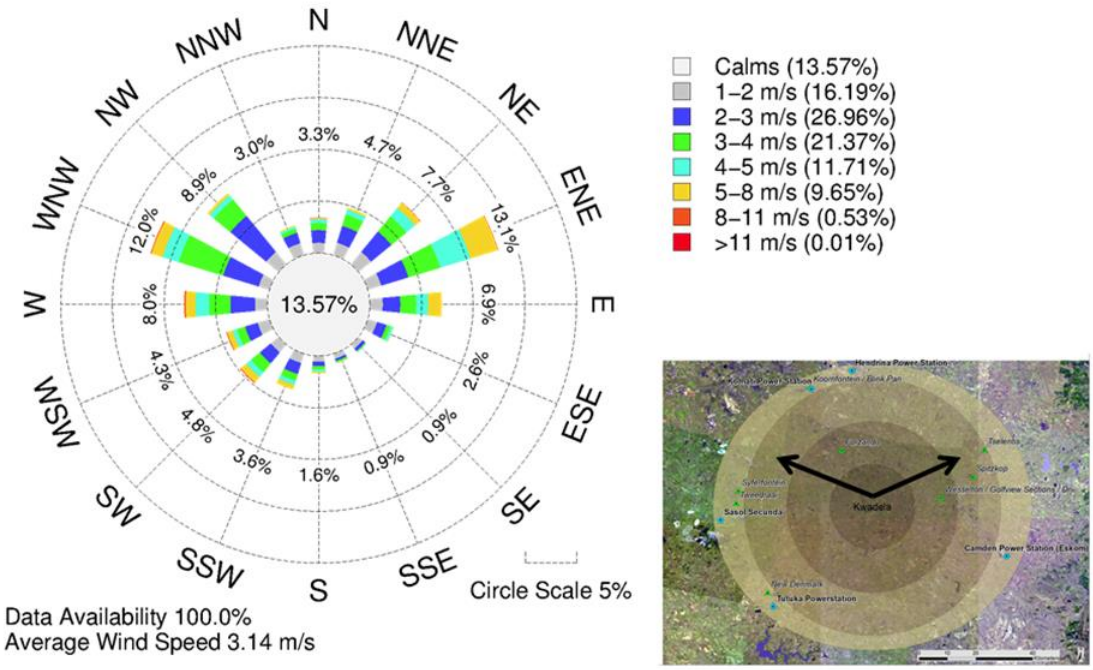

Figure 4-5: a) Wind direction and speed recorded during the summer 2014 at Kwadela (Burger and Piketh, 2015). b) The insertion indicates were the wind is from in accordance to Kwadela and the coal mines and power stations in proximity.

The synoptic weather conditions were obtained from the South African Weather Service for the winter sampling period in 2014 (07/21 to 07/29 and 08/05 to 08/12) and the summer sampling period in 2015 period (03/27 to 04/01 and 04/07 to 04/13). A High pressure system dominated the weather on 21/07 with sunny, cool to warm conditions. A surface trough with sunny, cool to warm conditions was experienced from07/22– 07/23. A cut of low with an upper air trough was situated over the country on 08/06 with cool conditions. A high ridge associated with a cold front moved west over the country from 07/25 to 07/27. A high pressure system with cool to warm air, cloudy conditions and light rain was experienced on 07/28 to 07 -29. An upper air cut off dominated the climate patterns from 08/05 to 08/08 and a surface trough brought clouds and light rain from 08/09 to 08/12. Similarly, a surface trough was seen on the synoptic maps from 03/27 to 03/31 during the summer. An upper air low and high ridge associated with cloudy and warm conditions were occurred over the country on 04/01. A surface trough with an upper air trough brought rain to the Mpumalanga area from 04/07 to 04/10. Warm conditions were recorded on the 04/12 were a surface trough with a High pressure cell moved over the South African extend.

4.3.2 Gravimetric contribution

The gravimetric measurements of coarse particulates and fine particulates were determined for both the sampling campaigns (Figure 4-6). Data was systematically sampled for the winter campaign using time-based intervals. Left of the dash the gravimetric results are given for data captured from 21/07/2014 to 29/07/2014 and on the right the sampling period from 05/08/2014 to 12/08/2014 is illustrated. The amount of coarse particles sampled dominated the aerosol loadings during the winter period.

The coarse/fine ratio were 0.56 which shows that much more coarse particulates were sampled. The maximum gravimetric concentrations measured for the winter were 86.56 µg/m^3 for fine PM and 170.19 µg/m^3 for coarse PM. The lowest concentration was 2.41 µg/m^3 for fine aerosols and 5.28 µg/m^3 for the coarse fraction. The highest coarse sample corresponds to one of the lowest fine samples.

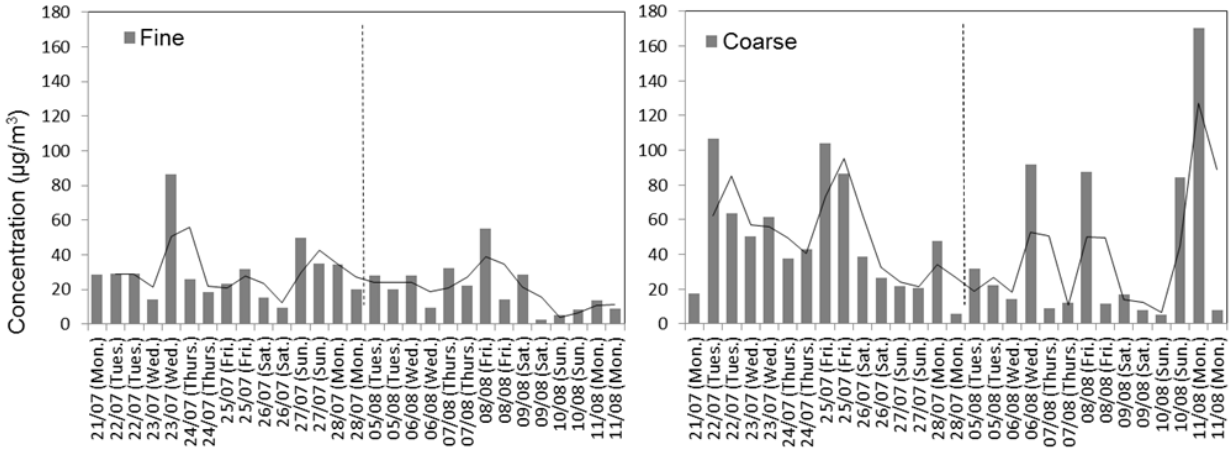

Figure 4-6: Gravimetric results for coarse and fine PM collected during the winter.

The summer profiles were collected on a continuous time-based interval (Figure 4-7). The overall gravimetric results showed that PM concentrations were much higher during the winter compared to the summer. Both fractions of the winter dataset were approximately two times higher than the summer samples The fine/coarse ratio of the summer samples was 0.60 which shows that the coarse PM dominated the aerosol loadings. The highest coarse concentration was 106.06 µg/m^3 and the lowest was 5.9 µg/m^3. The fine concentrations were less that the coarse concentrations with a maximum concentration of

25.86 µg/m^3 and a minimum of 2.55 µg/m^3. The individual gravimetric fractions had less variations compared to the winter fluctuations. This statement can be supported by the standard deviation of 21.46 for the summer and 32.15 for the winter data.

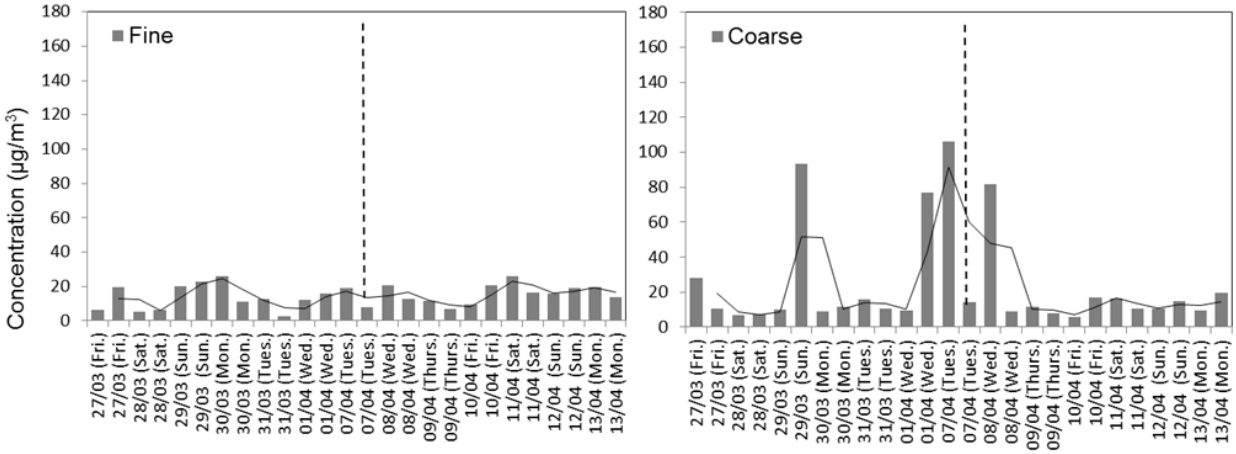

Figure 4-7: Gravimetric results for coarse and fine PM collected during the summer.

A comparative evaluation was then conducted to explain the variations in the gravimetric results of winter and summer periods. The total concentration of aerosols sampled during the day (06:00 am – 06:00 pm) and night (06:00 pm to 06:00 am) were displayed with wind speed and temperature data. A synopsis of meteorological data was constructed in a box-whiskers plot format to represent data recorded at an interval of one-second per sample. Non-consecutive data are separated by means of a dash.

A gravimetric concentrations and meteorological data showed that wind speed had a superior impact on amount of suspended aerosols during the winter (Figure 4-8). The factor coordinates of wind speeds and PM were slightly higher than temperatures and PM with 0.816 compared to 0.781. Consequently, higher concentrations were found in the diurnal samples alongside higher wind speeds and temperatures. The highest wind speed recorded during the day (08/08) at Kwadela was associated with a sudden increase in coarse PM. The nocturnal/diurnal ratio of wind speed was 0.79 and explains the occurrence of more coarse PM during the day. It was noted that a field fire occurred during the night on the 23/07 which caused the highest amount of fine PM. On the day with the lowest sampled coarse PM (10/08) a light rainfall occurred. A power outage of 2 hours occurred on 21/07 and 26/07 and can be seen in the lower concentrations sampled. The surface trough that moved over South Africa from 22/07 to 23/07 is associated with

colder conditions. This cold front could have caused that more coal combustion occurred for thermal heating practices. Higher fine PM concentrations were sampled on the 23/07.

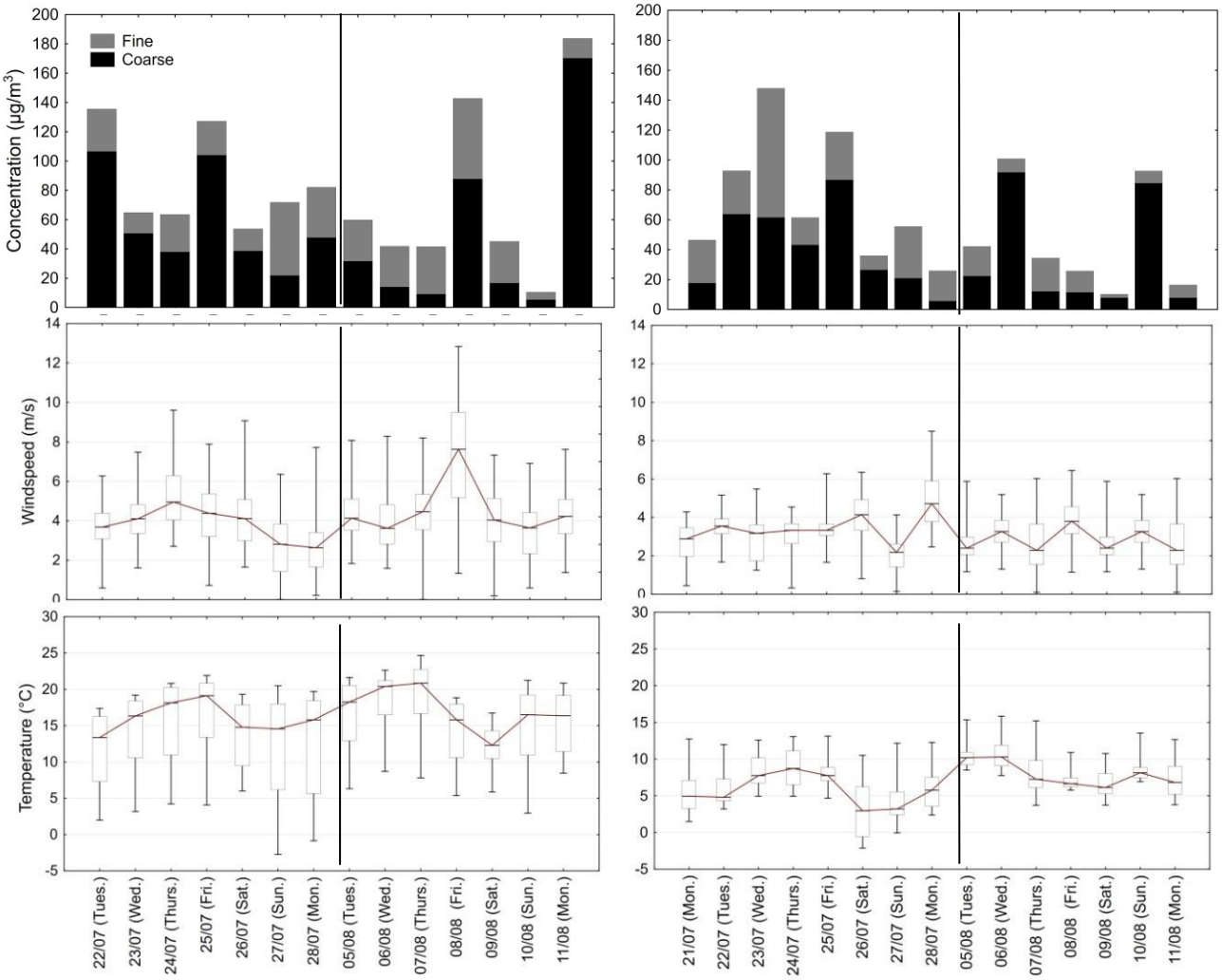

Figure 4-8: A time series of the total gravimetric concentrations of winter a) diurnal and b) nocturnal samples. The wind speed and temperature data are also provided.

The relationship between meteorological data and gravimetric concentrations were lower in the summer dataset (Figure 4-9). Factor coordinates from PCA were 0.738 between PM concentration and wind speed and 0.720 between temperature and particulate concentrations. The highest wind speed median (4.12 m/s) was recorded on the 6th of April and could explain the abundant amount of coarse species found on the 7th of April. Overcast and rainy conditions were experienced on the 28/03 which explains the decrease in the amount of coarse PM sampled.

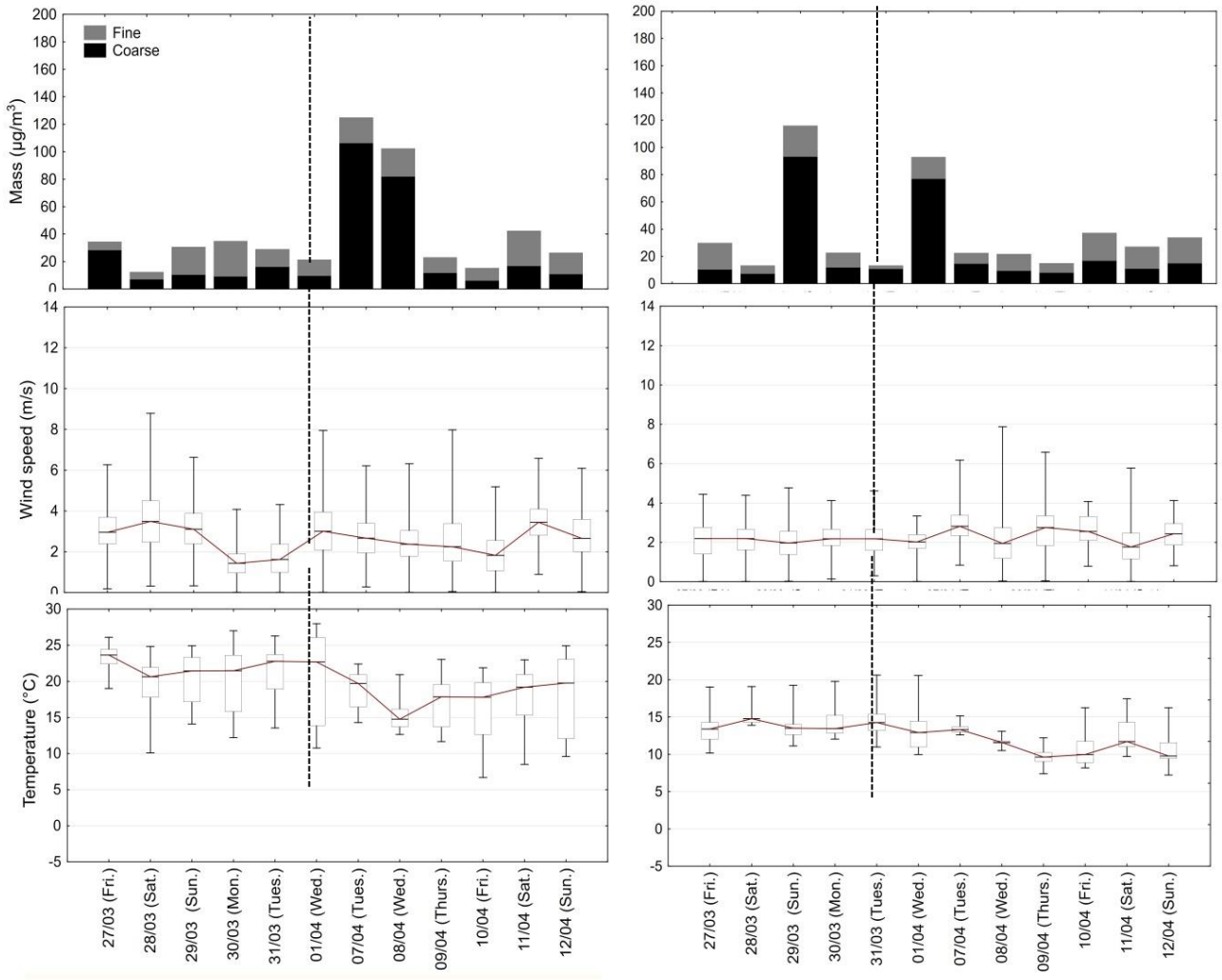

Figure 4-9: A time series of the total gravimetric concentrations of summer a) diurnal and b) nocturnal samples. The wind speed and temperature data are also provided.

Outdoor activities also play an important role in the quantity of suspended aerosols in the atmosphere. People are more active during the day playing soccer and moving livestock adjacent to the SFU which contributes to coarse particulates.

4.3.2.1 Ambient profiles

The chemical and elemental composition for both the fine PM and the coarse PM samples were obtained from ICP-MS, XRF and ICS analyses. The species with low S/N ratios were omitted from both datasets. The elemental acquisition methods differed for the winter and summer datasets which led to variations in the identified species.

The median atmospheric concentrations in coarse and fine PM are presented in Table 4-1 for both the sampling campaigns. The ambient coarse fractions of the winter dataset mainly comprised of ionic elements such as $SO_4^{2-,}$ sodium (Na), NO_3^-, Ca, and NH_4^+ species. The most abundant elements found in the fine fractions were SO_4^{2-}, Na, NO_3^-, NH_4^+, and chlorine (Cl). Important species that were not included in the winter samples' analyses were S, Si and total dissolved organic acids (DOC). The fine PM of the summer dataset illustrated a strong presence of copper (Cu), Si, Fe, cadmium (Cd) and SO_4^{2-} species.

Secondary aerosol SO_4^{2-} dominated the PM concentrations in the winter, with a total contribution of 45.65% to fine PM and 43.15% to coarse particulates. The presence of other secondary species in the winter period were also significantly high where fine aerosols comprised 14.81% of NH_4^+, 8.68% of NO_3^- and coarse fractions of 6.67% of NH_4^+ and 14.26% of NO_3^-. Similarly, the summer samples had plentiful of $SO4^{2-}$ in the coarse fractions (18.29%) and fine fractions (2.14%) along with NH_4^+ that contributed a total of 7.56%. The SO_4^{2-}, NO_3^- and NH_4^+ species found in the fine aerosols have a secondary origin due to atmospheric gas-to-particle conversion processes that occurred from S and N species. NO_3 results from the oxidation of NO_x products. Acidic gasses (sulphuric acid (H_2SO_4) and HNO_3) could have reacted with coarse PM to procedure secondary particles (Schlesinger & Cassee, 2003; Walton, 2013). The presence of anthropogenic source emissions was illustrated by the Cu/Ni, Zn and Pb/As elements (Nguyen et al., 2013). Elements Cu and Ni are characteristics of metal source were higher in the summer samples (Heidam et al., 2004; Nguyen et al., 2013). The Cu/Ni ratio was higher in both the summer fine and coarse fractions. The summer coarse particulates thus had the highest presence metal species where the median of Cu was 3.93 µg/m^3 and Ni 0.0059 µg/m^3. Pb, Zn and As that are linked to combustion practices were present during both the sampling periods (Heidam, 2004). Higher Zn concentrations was noted in the summer samples with a total contribution of 0.18% to coarse PM and 0.21% to fine PM.

Ratios of Pb/As were generally high throughout the sampling campaigns with 4.68 and 2.23 in fine and coarse factions of the winter samples and 2.78 and 38.01 in the summer samples. High concentrations of S elements measured in the summer fine (6.94%) and coarse (10.59%) aerosols indicate the dominance of coal combustion species. Even though S species were not included in the winter sampling analyses, the high occurrences of NO_3^- in coarse and fine particulates as well as SO_4^{2-} illustrate the prevalence of coal combustion emissions. Significantly low Se concentrations were measured in both campaigns which are linked to oil combustions processes (Nguyen et al., 2013). Biomass burning emissions contributed to the amount of PM in the summer as indicated by the presence of K species.

A total contribution of 1.68% was detected in the fine samples and 0.54% in the coarse samples. High percentages of Al (0.6%) and Si (1.5%) found in the coarse summer samples illustrate the involvement of crustal dust in ambient PM. The summer fine particulates also contained high concentrations of Al (5.40%) and Si (13.36%) species which are associated with coal combustion emissions. Gasoline motor vehicle emissions contributed to atmospheric pollution in the summer as seen by the high percentage of PM in the fine fractions. Elemental Pb is a constituent of petrol and act as an 'antiknock' chemical, in the form of a tetra-alkyl Pb (Lucarelli & Mando, 2004; Walton 2013). Ubiquitous metal vanadium is mainly is a result of anthropogenic and natural sources and mainly ascribed to the steel industry production (Visschedijk et al., 2013). The vanadium content were very low in the winter and summer samples. Vanadium metal can travel over long distances since it is mainly in submicron particle fraction size. The species associated with wood burning can clearly be seen in the summer samples. These include Cl with 0.37%, K with 1.68 and SO_4^{2-} with 2.14% in the fine fraction. The compilation of coarse particles collected in the summer also displays the presence of wood combustion emissions, with 0.22% Cl, 0.54% K, 10.59% S and 1.46% of SO_4^{2-}.

Table 4-1: Median concentrations for the different chemical species in the fine and coarse fractions sampled.

	Fine fraction (µg/m^3)		Coarse fraction (µg/m^3)	
Chemical specie	Winter 2014 N=29	Summer 2015 N=26	Winter 2014 N=29	Summer 2015 N=26
Ag	2.59E-05		8.77E-06	
Al	0.003921	0.213805	0.000657	0.051349
As	7.69E-06	0.001305	2.39E-06	0.025253
Au	1.45E-05		1.6E-06	
B	0.002669		0.001431	
Ba	3.51E-05	0.002246	4.13E-06	0.003395
Be	1.57E-07		1.64E-08	
Ca	0.004617	0.162593	0.001138	0.067269
Cd	1.42E-06	0.147483	2.8E-06	0.535904
Cl		0.014972		0.017729
Co	2.4E-06	0.002677	3.21E-07	0.006691
Cr	0.001882	0.006687	0.00075	0.006962
Cu	2.12E-05	1.860018	1.3E-05	3.936899
Fe	0.002181	0.318	0.000268	0.073073
Hg	0.000231		5.16E-06	
K	0.000778	0.066536	0.000134	0.043904
Mg	0.000909	0.030697	0.000161	0.007197
Mn	8.94E-05	0.004177	7.52E-06	0.001305
Mo	2.75E-06		1.04E-06	
Na	0.004678	0.031855	0.004626	0.014372
Nd		0.008875		0.018282
Ni	1.81E-05	0.005939	1.18E-05	0.005935
P	0.0005	0.009068	0.000108	0.003111
Pb	1.72E-05	0.049605	1.12E-05	0.069663
Pd	6.7E-06		1.93E-06	
Pt	8.46E-07		1.32E-07	
S		0.275103		0.848798
Sb	7.04E-06		9.51E-07	
Se	3.07E-06		1.08E-06	
Si		0.528985		0.121492
Sr	1.25E-05	0.014995	1.52E-06	0.034157
Ti	8.87E-05	0.028728	1.2E-05	0.006213
Tl	4.89E-08		3.59E-08	
U	3.51E-07		1.57E-07	
V	7.95E-05	0.011793	7.46E-06	0.007185
Zn	0.000182	0.008562	0.00012	0.015068
Na^+	0.510746	0.005427	0.926596	0.0087
NH_4^+	0.53068	0.031565	1.154096	0.60553
K^+	0.251338	0.001805	0.376942	0.012761
Mg^{2+}	0.231997	0.001784	0.042283	0.003469
Ca^{2+}	1.00901	0.010513	0.200996	0.019612
F^-	0.021103	0.000127	0.005956	0.000154
Cl^-	0.379284	0.006366	0.841525	0.01493
NO_3^-	0.972038	0.010184	0.676588	0.017944
SO_4^{2-}	2.983193	0.084881	3.557493	1.465063

Major differences were found between the elemental datasets of the sampling periods. This finding can be ascribed to different analyses techniques that were used. Controversial ratios were found between the elemental species' fine/coarse ratios of the sampling periods compared to the gravimetric mass ratios. The total concentration of the summer's elemental coarse fraction was higher than the winter samples by a factor of 257.63 and the fine fraction was 401.05 times higher. Ionic compositions displayed conflicting results. The coarse fraction of the winter ionic species was higher by a factor of 3.2 and the fine particulates with 50.98. The ionic species and gravimetric mass of the winter samples were higher than the summer which was conflicted by the elemental masses. The elemental concentrations obtained from ICP-MS analyses for the winter samples were inaccurate. The winter samples were thus excluded from the CMB analyses. The fine-to-coarse chemical and elemental concentrations were much higher in the summer dataset than the winter. This is illustrated in Figure 4-10, which compared the median of fine/coarse ratios. The median $PM_{2.5}/PM_{10}$ ratio was 0.48 for the summer PM and 0.41 for the winter. The summer PM thus comprised of more fine particulates than the winter. Diurnal $PM_{2.5}/PM_{10}$ ratios were lower with 0.41 in the winter but higher in the summer PM with 0.55 compared to the nocturnal samples. The nocturnal $PM_{2.5}/PM_{10}$ ratios were 0.47 for the both sampling periods respectively. The summer samples exhibited the highest fine/coarse ratios such as Mg with 4.26, Si with 4.35 and 4.62 in Ti. Mg is typically associated with anthropogenic emissions such as industrial pollution. Ti is a product from residential combustion processes. The maximum ratios for species found in the winter samples ranged between 1.1 – 2.2 for Cd, Na^+, NH_4^+, K^+, Cl^- and SO_4^{2-}. The ratio for summer samples was 35.36 times higher for Fe, 34 for Ti, 24 for Al and Mg, 38 for Mn and 17 for F. The K^+ ratio was much lower in the summer than the winter. More sea salt species (Mg, Na, Sr, V) were found in the summer with an average ratio of 2.14. A high median ratio for the Al, Ca, Fe, Mn, Si, Ti species in the winter and the samples show the presence of mineral dust species.

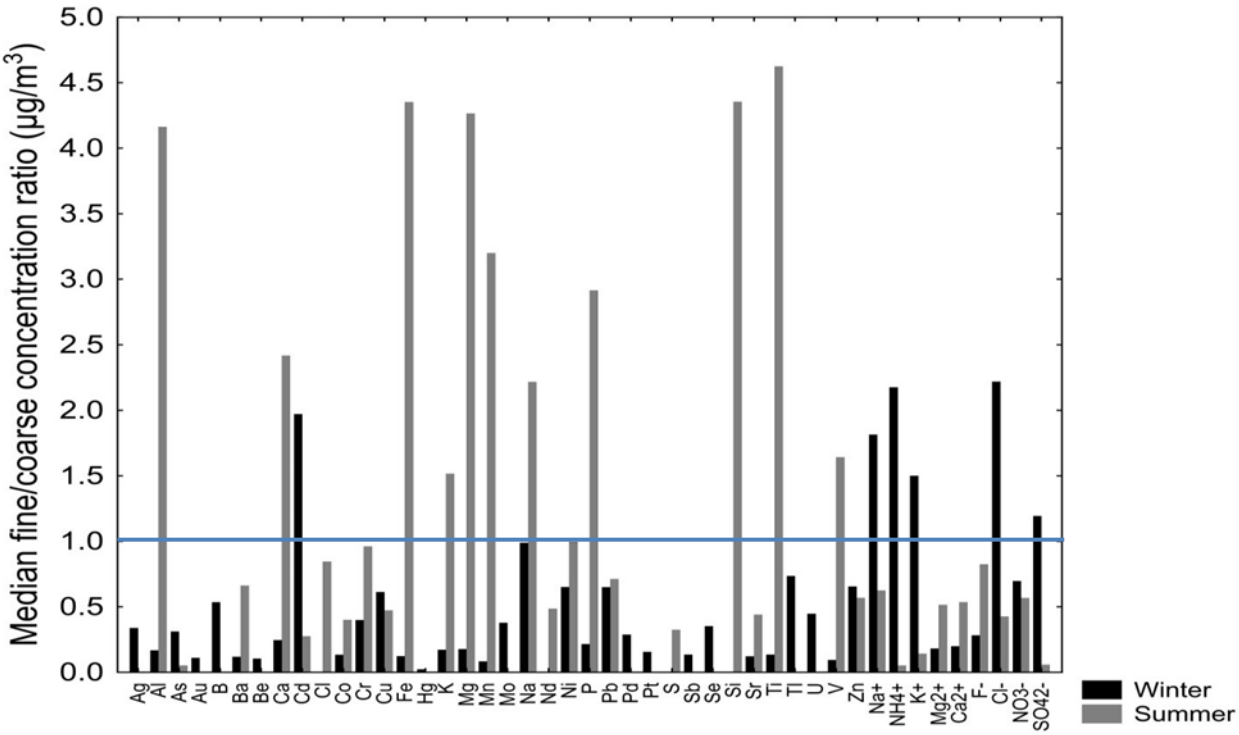

Figure 4-10: Graph shows the fine/coarse ratios of the median of samples collected in the winter and summer. Blue line inserted at 1 to clearly show the f/c ratio.

The time series of ionic species found in both the sampling campaigns are illustrated for coarse and fine fractions (Figure 4-11). The Ionic species concentrations were higher in the winter coarse particulates. The most abundant species detected in all the samples were secondary SO_4^{2-} with a median of 3.84 µg/m^3 and 3.54 µg/m^3 for the winter coarse and fine particulates and 1.46 µg/m^3 and 0.09 µg/m^3 for the summer samples. SO_4^{2-} also displayed the highest standard deviation in all the samples. The lowest ion detected (F^-) in all the samples is recognized as a species from industrial emissions. Cl^- and Mg^{2+} had the best correlation matric with a correlation matrix of 0.95 in the coarse aerosols measured in the winter. NO^{3-} and NH_4^+ secondary species were abundant with a maximum concentration of 2.88 µg/m^3 and 2.26 µg/m^3 in the winter fine samples. Fewer correlations were detected in the winter fine species with the best fit found between SO_4^{2-} and K^+ (R= 0.78). The correlation matrix of summer fine fractions between SO_4^{2-} and NH_4^+ displayed a good fit of 0.95. A variety species had a good correlation in the summer coarse fractions of approximately 0.99 such as Mg^{2+}, Ca^{2+} Na^+, Cl^- ions and between the SO_4^{2-} and NH_4^+ species. High concentrations of SO_4^{2-}, NO^3 and NH_4^+ species that were detected in all the samples represents secondary pollutants that are a result of NH_3, SO_2

and NO_x transformations (Han et al., 2015). The overall concentrations of NH_4^+, Cl^- and SO_4^{2-} were higher in the winter samples. Crustal sources comprised of Na^+, Mg^{2+} and Ca^{2+} species (Wang et al., 2005) was more abundant in the winter samples. K^+ that result from biomass burning (Duan et al., 2004) was the highest in the summer fine particulates (median = 0.37 µg/m^3).

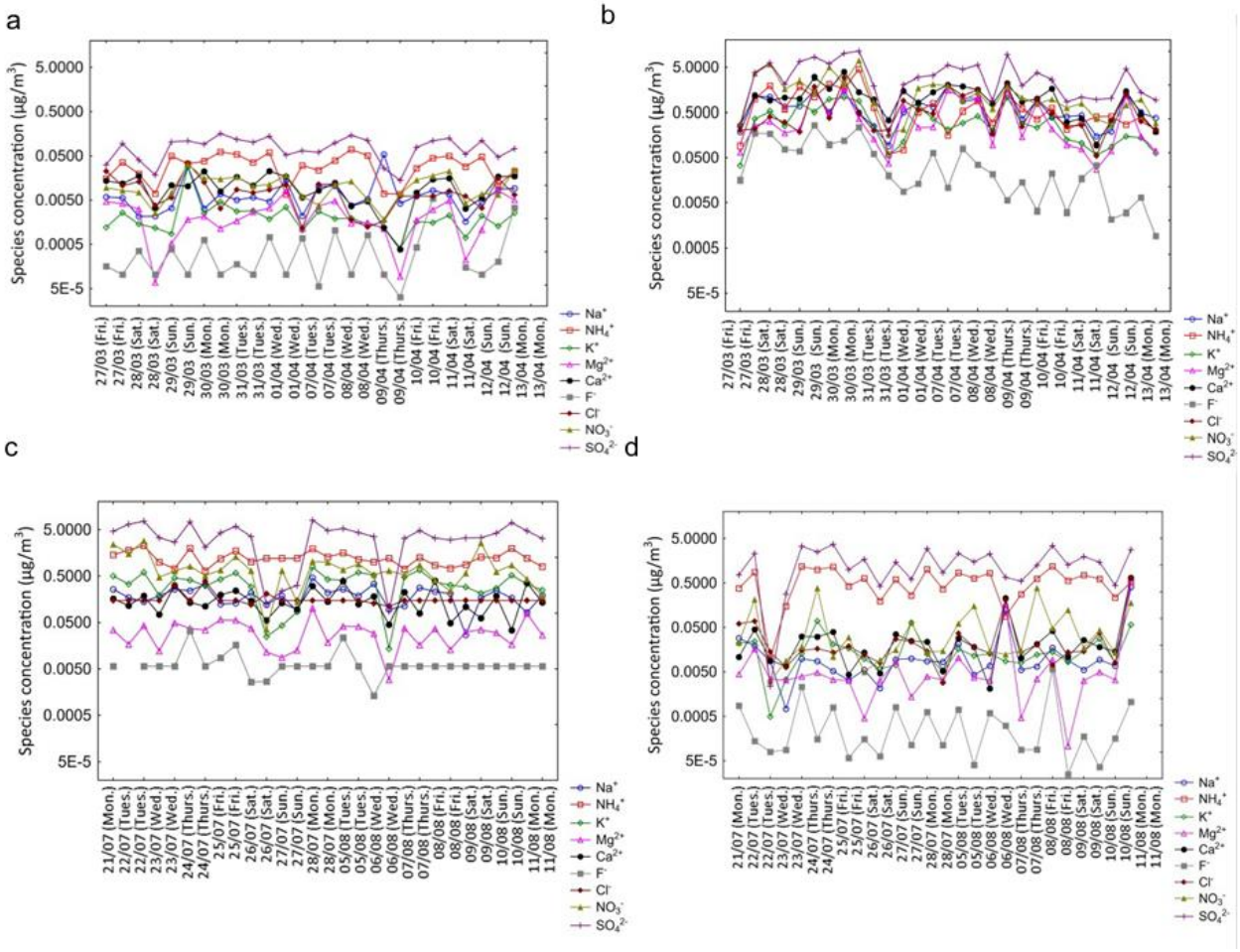

Figure 4-11: Time series of the ionic species found in a) winter fine fractions, b) summer fine fractions, c) winter coarse fractions and d) summer coarse fractions.

4.3.3 Chemical mass balance model

The primary purpose of this investigation was to identify the main source responsible for air pollution in Kwadela. The winter samples obtained high Chi-square values and low mass percentages and R-square values in the CMB. This dataset could thus not be

resolved in the CMB. Twenty summer samples were analysed through the CMB to identify the main polluter in Kwadela.

The CMB targets as are given for the different performance measures along the results of the summer campaign (Table 4-2). The summer dataset showed good R-square, mass percentage, Df and Tstat values. The high mass percentages indicate that the number of source species corresponded to the number of measured masses. High R-square values are used to show that inconsistencies in the SCE's corresponds to inconsistencies in measured species The chi-square numbers which indicates the fit of SCE, were higher than the targets.

Table 4-2: Averages of the performance measures obtained from the CMB.

	R-Square	Mass %	DF	Chi-square	Tstat
CMB targets	0.8 to 1.0	100% ± 20%	>5	0.0 to 4.0	>2
Summer					
$PM_{2.5}$	0.90	89.25	5.84	4.89	5.89
PM_{10}	0.819	86.64	4.85	7.7	4.54

The SCE shows the omnipresence of coal combustion emissions. Coal combustion was identified to be the leading cause of ambient coarse PM concentrations during summer with vehicle emissions as the next most significant source. Coal combustion emissions contributed to a total of 173.49 µg/m^3 (of the twenty of the selected samples) and diesel motor vehicles to a total of 88.32 µg/m^3 to the coarse aerosols. The average concentration was 8.67 µg/m^3 and 4.41 µg/m^3 for domestic coal burning and diesel vehicle emissions respectively. The total concentration of biomass species of 13.44 µg/m^3 and paved road dust 5.96 µg/m^3 were measured. The fine fraction sampled during summer showed that residential coal combustion and refuse/wood burning were the main contributing sources to the aerosol loadings. The high average PM (6.46 µg/m^3) and total contribution (122.78 µg/m^3) of household coal combustion indicated the prevalence of this source. The average SCEs of refuse/wood combustion was 1.58 µg/m^3, 1.49 µg/m^3 for paved road dust, 1.13 µg/m^3 for coal dust and 0.72 µg/m^3 for diesel vehicle emissions. A high average of the paved road dust source profile was detected. The TM indicated that biomass burning (30.05 µg/m^3), paved road dust

(28.35 µg/m^3) and coal dust (21.61 µg/m^3) sources had smaller effect on the ambient air quality, compared to household burning emissions.

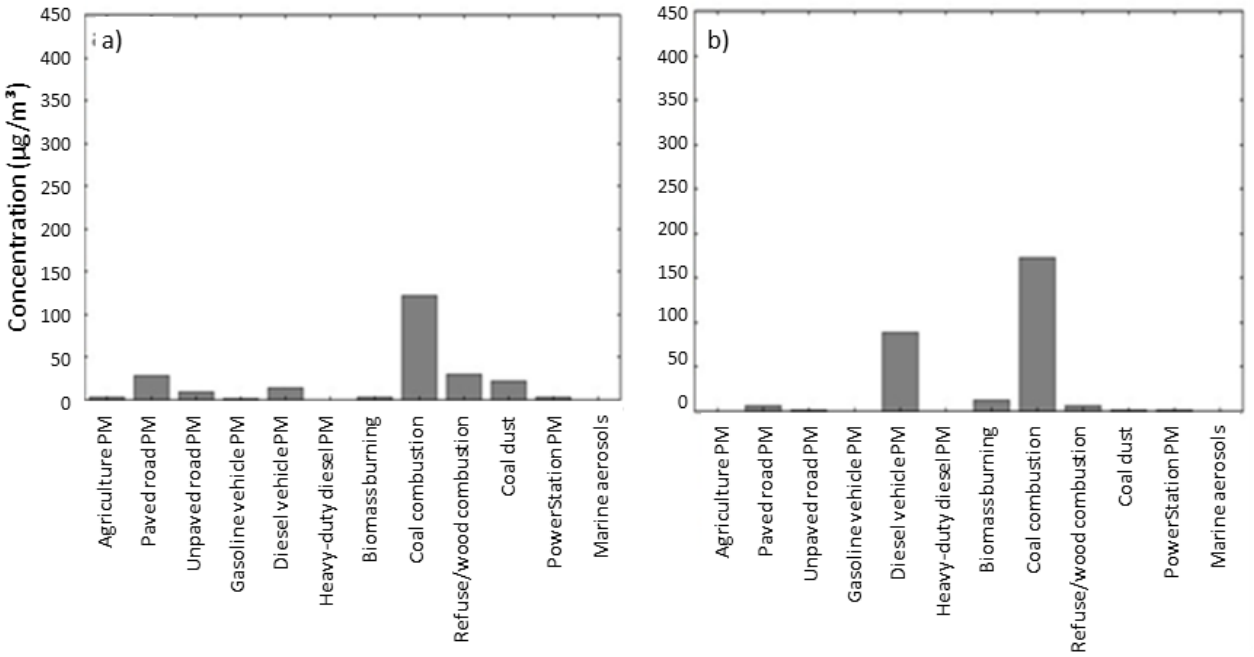

Figure 4-12: The SCE to ambient coarse concentrations for the a) fine concentrations and the b) coarse fractions

The contribution percentages of the polluting sources are illustrated in Table 4-3. The household combustion SCE consist of wood combustion and coal combustion estimates, the soil SCE is a compilation of all the soil profiles and the vehicle emissions include PM emitted from diesel motor vehicles, gasoline vehicles and heavy-duty trucks. The household combustion emissions were the leading cause of PM in both; the coarse (59.89%) and fine (58.67%) fractions. More soil particulates (17.81%) were detected in the fine fractions with the second highest SCE. Pollution from vehicles was significantly high in the coarse PM with a contribution of 11.45%. Particulates from power stations and refuse burning were very low.

Table 4-3: Source contribution estimates for coarse and fine fractions

Source contribution	% Detected mass in Kwadela	
	Fine fraction	Coarse fraction
Domestic combustion	58.67	59.89
Refuse burning	1.39	4.31
Pollutant of soil	17.81	1.95
Vehicle emissions	5.28	11.45
Coal dust	8.36	1.41
Power stations	2.55	0.66
Unexplained	5.94	20.33

A time series of SCE's on a linear and logarithmic graph are displayed for the diurnal and nocturnal samples (Figure 4-13). Wind speed and temperature data are given to illustrate influence on the amount of PM. Variances in the diurnal SCE's were lower with a standard deviation of 4.35 compared to the 7.28 of the nocturnal samples. The logarithmic graph clearly shows the omnipresence of coal combustion emissions, whereas the contribution of other sources varied over the sampling period. In the diurnal samples certain days showed a high presence of (01/04) gasoline vehicle emissions, biomass burning (08/04) and coal dust (11/04). The high occurrence of biomass burning pollutants was accompanied by an increase in wind speed and a temperature drop. The high coal dust SCE was measured on the same day as the highest median wind speed. The nocturnal SCE's shows three occurrences of high wood combustion pollutants. Wood combustion that occurred on Saturday (28/03) and Sunday (29/03) could be a result of social activities over the weekend such as cooking meat over a wood lit fire.

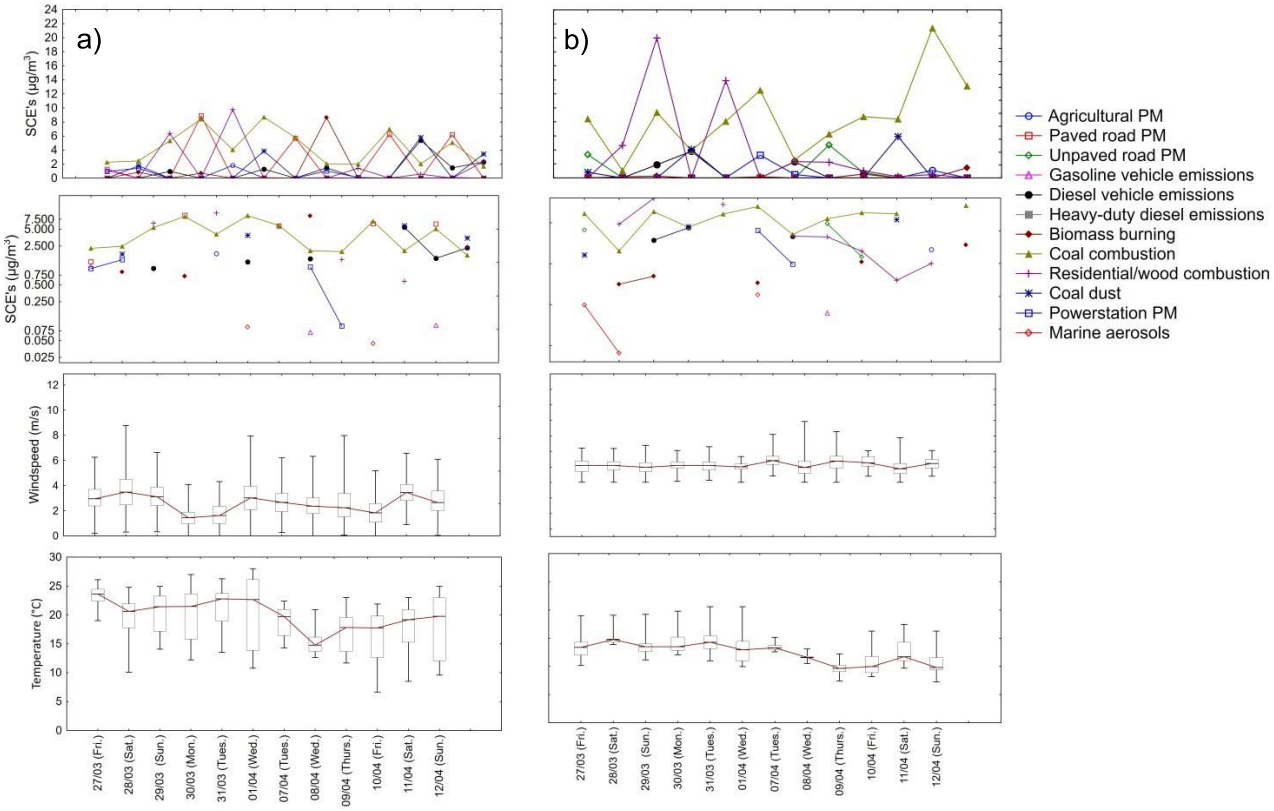

Figure 4-13: A time series of a) diurnal and b) nocturnal SCE's of the fine fractions are displayed on a linear and logarithmic graph along with the associated wind speeds and temperatures.

The mass of polluting source species is 1.65 times higher in the coarse fractions than the fine fractions (Figure 4 -14). More sources contributed to the amount of PM during the day. This can be ascribed to more activity such as driving, sport and movement that causes excessive dust. Factors other than meteorological conditions influenced the residential coal combustion practices. High diesel vehicle emissions and paved road dust were found on the same day (01/04). No correlations were found between the meteorological conditions and amount of suspended PM.

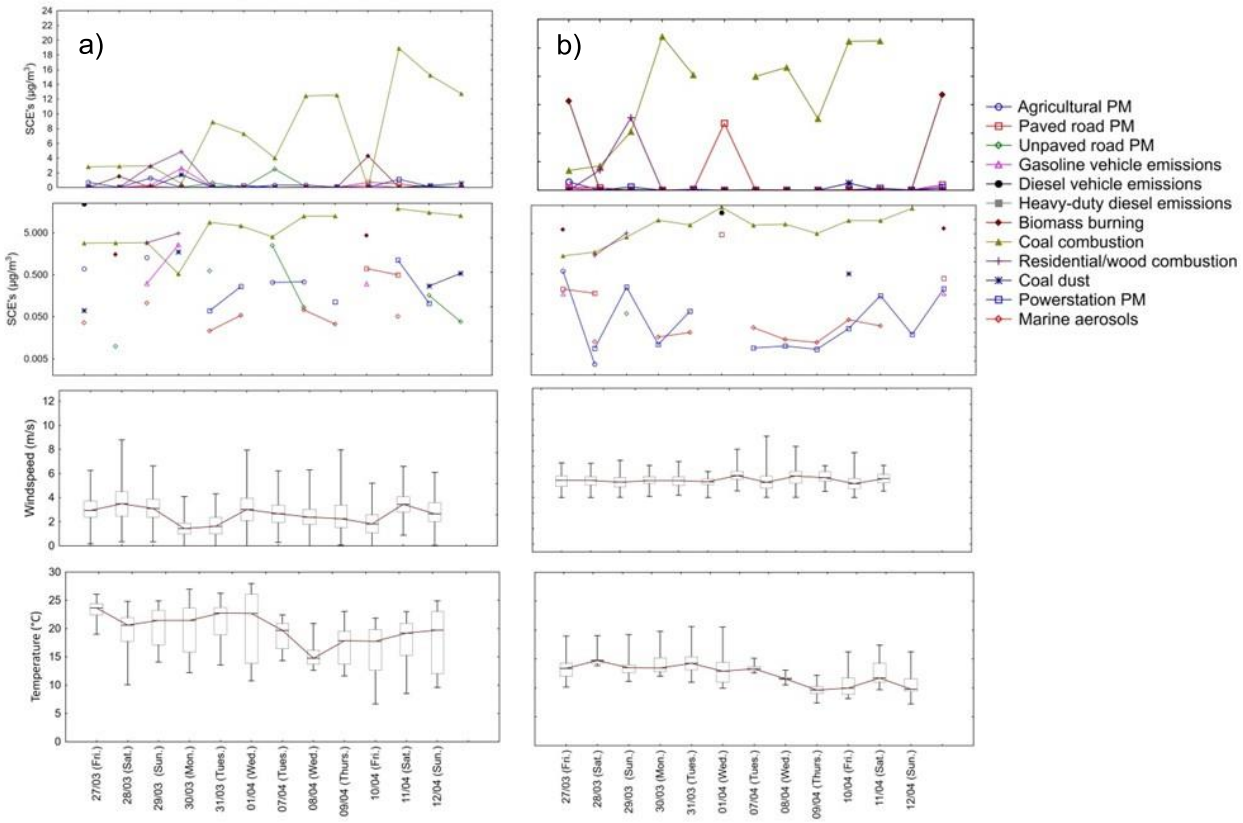

Figure 4-14: A time series of a) diurnal and b) nocturnal SCE's of the coarse fractions are displayed on a linear and logarithmic graph along with the associated wind speeds and temperatures.

4.4. Discussion and conclusion

PCA showed that meteorological conditions slightly had a higher impact on the amount of PM in the winter compared to the summer. The nocturnal/diurnal ratio of wind speed indicates that higher wind speeds were measured during the day alongside more coarse particles. Other than meteorology are the amount of PM influenced by social activities that took place. Activities such as soccer and movement of livestock (such as goats and cattle) could cause more dust in the area. Sampling in the summer was conducted during school holidays. Kids could have organized soccer tournaments in the field adjacent to the sampling instrument which could have contributed to the amount of coarse PM.

Variances were found between the chemical and elemental composition of the

winter and summer PM samples. This can partially be ascribed to the different methodologies followed for the detection of elements. Winter samples did not include S and Si species that were vital for this study. SO_4^{2-} was the dominant species found in the winter samples and Cu in the summer. The elemental fine/coarse fraction ratios displayed that the concentrations of summer samples were higher. Controversially, the gravimetric masses and ionic concentrations illustrated that much more PM were sampled during the winter. The approach of IC analyses thus delivered inaccurate results for the elemental species in the winter fractions. Differences found in chemical and elemental species and gravimetric mass can also be explained by the meteorological conditions. The lower temperatures in the winter led to higher fuel combustion practices and thus higher occurrences those species associated with coal combustion. The prevalent wind direction of the winter campaign was from a north-westerly direction. Wind directions recorded in the summer varied between north-easterly and north westerly winds. Pollutants from different sources were thus present in the ambient samples.

Secondary species (SO_4^{2-}, NH_4^+, NO_3^-) dominated the aerosol loadings of the winter campaign. High concentrations of SO_4^{2-} and NH_4^+ were also found in the summer samples. The presence of anthropogenic source pollutants was clearly illustrated through secondary pollutants and the high Cu/Ni, Zn and Pb/As species. The summer samples contained more species from a metal origin than the winter samples. Household combustion practices had a major influence on the PM loading of Kwadela. High occurrences of Pb, Zn and As species were found in both sampling campaigns with a high prevalence of S species. The S species of the summer samples shows that residential coal combustion was a major polluting source. The high occurrences of NO_3 and SO_4^{2-} concentrations of the winter samples also show that residential coal combustion species were dominant. The presence of biomass burning, wood burning and vehicle emissions in fine PM and crustal dust in coarse PM were found.

The winter samples were not analysed by means of the CMB due to poor results found through IC analyses. The summer samples that were analysed were within the required range of the CMB performance measures. The summer samples could thus be used to represent Kwadela's PM compilation, with the exception of higher coal combustion contributions in the winter. The dominant cause of the high ambient

PM loadings in Kwadela can be assigned to their residential fuel-burning practices. Coal combustions profiles are comprised mainly of S, Cl^-, SO_4^{2-} and NO_3^- species. The high amount of secondary species found in the ambient samples corresponds to the residential coal combustion profiles. None of the other polluting sources can compare to the high PM values associated with residential coal combustion. The local air quality management plans should thus include mitigation measures for household fuel burning in order to achieve acceptable ambient air quality.

CHAPTER 5:

SUMMARY AND CONCLUSIONS

The section provides the results of the research aim, and all the research questions. The domestic fuel use patterns in South Africa are illustrated and all the factors that influenced fuel choice are discussed. The characteristics of Kwadela's particulate matter are provided and all the polluting sources are identified. The proportional contribution of every polluting source is discussed and the main findings are summarised.

This research explored the significance of domestic fuel burning practices in South Africa. Firstly, fuel combustion tendencies were investigated along with the factors that influence fuel choice. The number of people exposed to this indoor and outdoor air pollution was determined. The second part of this study was based on a local scale where people relied on coal as alternative energy source. The ambient air quality of a low income community were analysed to compare household combustion emissions to other polluting sources such as power stations. The fuel use patterns were explained by the spatial distribution of fuel uses, the temporal scale of use and the factors that influences these tendencies.

5.1 Domestic fuel burning in South African low income settlements

Domestic fuel combustion practices contribute significantly to the amount of ambient PM in South Africa. Residents depend on a mixture of energies to support their energy needs, such as wood, coal, gas, paraffin and animal dung. The South African 2011 census data were used to explain fuel use patterns on a SAL scale.

- The following areas (SALs) were had more than 80% of residential fuel burning households.
    - The north-western parts of the North-West.

  - The south-east of Mpumalanga.
  - Small patches in the east coast.
  - Almost no SALs were found in the Western-Cape.
- The spatial distribution of fuel use differed throughout the South African extend.
  - Coal combustion occurred in the Highveld and in the Northern-Cape.
  - Paraffin energy was preferred in the eastern areas of South Africa.
  - Residential areas of the Northern Cape and Kwazulu-Natal relied on gas as energy source.
  - Wood use was spread over the South African extent.
  - Animal dung use could be linked to people that lived in the North-West, Mpumalanga, Free State and Eastern Cape.
- Income levels were one of the main factors that influenced choice.
  - Animal dung and wood sources were used by the poorest fraction of the population.
  - The household income levels of coal users extended up to the category of R28900 per year.
  - Gas was used by a richer fraction of the population, up to the household income class of R230700.
- The number of residents in a household indicated the type of fuel preferred:
  - Larger families that lived in one household preferred coal, wood and animal dung.
  - Smaller households were associated with paraffin and gas use.
- R-square calculation showed that people living in traditional and informal settlements generally used energy alternatives.
  - Factors such as family sizes, appliances, the gender of the household head, annual household incomes and the number of chronic diseases illustrated a positive correlation to the fuel burning settlements.
- Proximity analyses based on the accessibility of fuel types illustrated that coal users were located in the Highveld area and wood users were distributed across the South African extend.
- Seasonal changes influenced the combustion practices.
  - Solid fuels were more frequently burnt during the winter periods for heating purposes.

  - Colder areas were associated with higher emissions due to more fuels that were burnt.
- The number of people that were affected by air pollution from residential fuel burning are listed below:
  - Indoor air pollution from cooking practices affected 12 570 994 people.
  - 14 199 261 people were affected by indoor pollution that occurs for heating.
  - In 2011 27.4% of the population were affected by indoor air pollution.
  - 29 562 settlements were identified where more than 30% of the residents depend on dirty fuels for cooking.
  - More than 30% of the residents found in 34 985 of the low income settlements combusted fuels for thermal heating.
  - Outdoor air pollution from cooking practices affected 15 833 055 people and 19 148 085 from fuels burnt for heating.
  - The upper limit calculations showed that more than 28 398 522 of the population could have been exposed to indoor air pollution which were approximately 54.8% of the South African population.
  - 38 296 170 persons were exposed to outdoor population according to the upper limit calculations which were 73.9% of the total population.

These estimates highlighted the importance of domestic fuel burning inventories in South Africa and the need for proper management measures.

5.2. Source apportionment of ambient particulate matter in Kwadela, Mpumalanga

Ambient PM were sampled during a winter and summer period according to the Maenhaut method. The gravimetric masses measured showed that the ambient air quality conditions in Kwadela are poor. Residents are exposed to particulate loadings above the national standards, which could be harmful to their health and well-being. Sources that contributed to the PM were agricultural soil, emissions from gasoline, diesel and heavy-duty motor vehicles, marine aerosols, biomass burning, paved and unpaved road dust, coal dust, coal combustion, refuse/wood combustion, power plant fly ash and secondary particulates. The main results of the PM quantification procedure are discussed below:

- Gravimetric masses illustrated the amount of PM during the winter and summer:
    - More fine PM was sampled during the winter which shows that combustion practices was the foremost polluter and illustrated the importance of secondary species
    - The fine/coarse ratio indicates that coarse particulates were more abundant during the summer
        - The amount of PM sampled during the winter was much higher that the summer
    - Meteorological conditions had a slightly higher correlation compared to the winter samples
    - PCA showed that wind speed had a larger influence than temperature
    - Other activities such as soccer, family get-togethers or movement of livestock could have influenced the amount of PM
- The XRF and ICS methods were effectively applied to define the characteristics of ambient aerosols.
- ICP-MS used for elemental analyses of the winter dataset delivered poor results. The following chemicals were found:
    - Secondary species are important on a local scale. Secondary species dominated in the winter samples and had a major impact on the chemical composition of the summer samples.
    - SO_4^{2-}, Na, NO_{3-}, Ca and NH_4^+ were the prevalent chemicals in the $PM_{2.5}$ winter samples.
    - Ambient PM_{10} consisted out of SO_4^{2-}, Na, NO_{3-}, NH_4^+ and Cl chemicals.
    - The detection methods applied to the winter samples did not include S and Si elements.
    - Cu, Si, Fe, Cd and SO_4^{2-} dominated in the fine aerosol loadings of the summer samples.
    - The coarse particles of the summer datasets mainly contained Cu, SO_4^{2-}, S and Cd chemicals.
    - The Cu/Ni, Zn and Pb/As species showed the importance of anthropogenic sources.
    - Metal species Cu/Ni were higher in the summer samples
    - Pb, Zn and As that are associated with combustion practices were high in all the samples.

  - The high occurences of S, SO_4^{2-}, NO_3^- and Cl^- illustrated the dominace of coal combustion emissions in the summer samples and SO_4^{2-}, NO_3^- of the winter samples.
  - The K species showed that biomass burning contributed to the amount of PP in Kwadela, Al and Si showed the involvement of crustal dust, Pb of petrol pollutants.
- The CMB average performance measures were calculated the show the goodness and fit of the data.
  - The winter samples were not analysed with the CMB model.
  - Good R-square values that were obtained for the summer samples were 0.9 for PM_{10} and 0.81 for $PM_{2.5.}$ and shows a good correlation between the measured species and SCE's
  - The mass percentage was 89.25% for the $PM_{2.5}$ and 86.64% for $PM_{10.}$ The mass indicates the number of species in the sources that correspond to the measured species.
  - The average DF was 5.84 for the fine fractions.
  - The PM_{10} DF was below the standard with 4.85.
  - The following samples obtained high Chi-square values; 4.89 ($PM_{2.5}$) and 7.7 (PM_{10}) for the summer samples.
  - All the samples were in accordance to the required Chi-square standard. The SCE shows the fit of SCE to the measured species.
- The influence of agriculture activities, paved road PM, unpaved road PM, gasoline and diesel vehicle emissions, heavy-duty diesel vehicles, biomass burning, coal combustion, refuse/wood combustion, coal dust, power stations and marine aerosols were researched in this study.
- The SCE show the proportional contribution of all the polluting sources. The aim of this research was to determine the main polluter on a local scale. The average SCE for the main sources were as follow:
  - The summer coarse particles showed the dominance of coal species with 8.67 µg/m^3 and followed by diesel pollutants with 4.41 µg/m^3.
  - The main cause of fine particles in the summer was coal combustion (6.46 µg/m^3), refuse/wood burning (1.58 µg/m^3) and unpaved road dust (0.46 µg/m^3).

- The TM of twenty samples selected from summer datasets showed the overall contribution of the polluting sources:
    - The summer $PM_{2.5}$ samples showed that coal combustion (122.78 µg/m^3) were the main cause of air pollution and then refuse/wood burning (30.06 µg/m^3) and unpaved road dust (21.61 µg/m^3).
    - Coal combustion dominated the summer PM_{10} aerosols with 173.49 µg/m^3, and followed by 88.32 µg/m^3 from diesel vehicle emissions and 13.45 µg/m^3 from biomass burning.
- The SCE's for the source profiles showed that:
    - Domestic combustion activities were the highest polluter which caused 58.67% and 59.89% PM in the fine and coarse fractions
    - The second highest contributors were soil which contributed 17.81% to fine PM and vehicle emissions caused 11.45% of the coarse PM
- Evaluations of the SCE's of coal combustion showed that:
    - Temperature had a better correlation versus the amount of fine PM that result from coal combustion than wind speed

The purpose of this research was to determine the main source of air pollution. These SCEs clearly identified coal combustion practices as the leading polluter.

Higher gravimetric masses of fine PM shows that coal combustions practices were higher during the winter. People generally burn more solid fuels for heating purposes. In order to manage the air quality of Kwadela and to improve the livelihoods of local residents, coal combustion practices must be controlled.

5.3. Summary of the main results

Domestic fuel burning practices in South Africa are the primary source of air pollution in certain areas. This statement can be supported by the main findings of this research:

• Fuel use patterns vary over a spatial extend and can be ascribed to a number of determinant factors, such as location, income levels, type of household, number of people living in a house etc.

- An estimated 27.4% of the population is affected by indoor air pollution from fuel combustion practices. The number of 191 480 085 people are exposed to outdoor air pollution. Upper limits calculations showed that an approximated 54.8% of the population is exposed to indoor air pollution and 73.9% are exposed to outdoor air pollution.

- Ambient PM sampled in a low income community, Kwadela exceeds the national air quality standards. The air quality is thus poor. Winter samples had a high abundance of SO_4^{2-}, NH_4^+ and NO_3^- species and the summer samples was compiled of Cu, SO_4^{2-} and S chemicals.

- The sources that influenced air quality in Kwadela includes agricultural dust, paved and unpaved road PM, gasoline and diesel vehicle emissions, heavy-duty diesel vehicle emissions, biomass burning, coal combustion, refuse/wood combustion, coal dust, power stations and marine aerosols.

- Residential coal emissions contributed a total mass of 403.11 µg/m^3 to the $PM_{2.5}$ of twenty samples collected in the winter period. Heavy-duty diesel vehicles were the second largest source of air pollution, with a total mass of 4.188 µg/m^3. Household coal combustion is thus the greatest cause of air pollution in Kwadela.

5.4. Recommendations and future work

The chemical compositions of ambient particulate matter were determined by means of ICP-MS or XRF and ICS techniques. The winter species were detected through ICP-MS and ICS analyses. Summer samples were analysed through XRF and ICS measures. Different elements were also researched. Consistency in analyses procedures and the type of elements identified are required to effectively compare the datasets.

The CMB model was used to link the pollutants found in ambient samples to their sources. Source profiles were collected from the USEPA database and previous samplings undertaken in South Africa. Where possible, the local source profiles were used. However, not all the source profiles were completely representative of the local conditions in Kwadela. Future work will thus be directed to the sampling of local source profiles.

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