$PM_{2.5}$ CHEMICAL COMPOSITION, SOURCE APPORTIONMENT AND GEOGRAPHICAL ORIGIN OF AIR MASSES IN PRETORIA, SOUTH AFRICA.

by

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Submitted in fulfillment of the requirement for the degree

Doctor of Philosophy (Environmental Health)

at the

School of Health System and Public Health

FACULTY OF HEALTH SCIENCES

UNIVERSITY OF PRETORIA

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February 2020

DECLARATION

I, Adewale Adekunle ADEYEMI, declare that, this thesis which is titled "***$PM_{2.5}$ chemical composition, source apportionment and geographical origin of air masses in Pretoria, South Africa***," submitted for the degree of Philosophiae Doctor (Environmental Health) at the School of Health Systems and Public Health, University of Pretoria, is my own work and has not been submitted by me for any other degree or examination at any institution of higher learning.

.....
Adewale Adekunle Adeyemi

Signed on the ……. day of ……..in …….

ETHICS STATEMENT

The author, whose name appears on the title page of this thesis, has obtained, for the research described in this work, the applicable research ethics approval.

The author declares that he/she has observed the ethical standards required by the University of Pretoria's code of ethics for researchers and the policy guidelines for responsible research.

MANUSCRIPT SUBMITTED FOR PUBLICATION

Adewale A. Adeyemi, Peter Molnar, Janine Wichmann. Association between air pollution and respiratory disease hospital admissions in Pretoria, South Africa. International Journal of Environmental Health Research. (Under review, resubmitted after revision made on 31 July 2019)

DEDICATION

This research work is dedicated to the Almighty God, the Alpha and Omega to whom I ascribe all the glory for my success and achievement in life.

I also dedicate this piece of work to my wonderful parents. Chief Albert Adebisi Adeyemi, and late (Mrs.) Margaret Adeyemi, who have been a source of inspiration and encouragement to me all through my life.

ACKNOWLEDGMENTS

It is fulfilling and amazing to see the work of these years coming to completion. A though the journey was not always smooth, there were times that I thought that it would never be completed. For this achievement, it is important to express my gratitude to the following persons and institutions.

First, to my Creator, God Almighty, for giving me life, courage, strength and perseverance to complete my studies. I will never be ungrateful Lord. Thank you for accompanying me in this journey; I would not have made it this far without you.

I wish to express my profound gratitude to my supervisor, Assoc Prof. Janine Wichmann, for her excellent supervision, tireless patience and encouragement throughout my research work and whose repository of knowledge added value to this research. Her monumental patience encouraged me greatly. I am also grateful to her for ensuring that scholarships were secured to make this journey stress free. "Dankie Ma'am." To my co-supervisor, Assoc. Prof. Peter Molnár, thank you for your excellent supervision, comments, advice and training on the model used for this study, you are indeed an embodiment of knowledge. Many thanks to Dr Nico Classen for the training and assistance provided throughout the project. To other project team members, John Williams and Novela Rhiranhzu, thank you for making this bigger project a success, Mbunu Nandi, Nwase Maxine, thank you for your support during sampling period.

The financial assistance of the National Research Foundation (NRF) towards this research is hereby acknowledged.

I am truly grateful to the following Institutions/organizations for the financial support/funding:

- Forestry Research Institute of Nigeria for the opportunity given me to pursue my dream in PhD research and for granting me a study leave.
- The University of Pretoria for the postgraduate research support bursaries.

I am grateful to the NOAA Air Resources Laboratory (ARL) for the provision of the HYSPLIT transport and dispersion model and READY website used in this thesis.

No words of appreciation can sufficiently express my thanks to my beloved parents. Chief and Mrs. Albert Adeyemi for their love, encouragement, financial support and understanding they showed me during the course of this programme. What kind of word is there to describe the '*sacrifice of a mother*' I enjoyed in my mother, late Atinuke Margaret Adeyemi, your early

departure shortly before my departure to start this study was indeed a great loss to me, continue to rest in the bosom of the Lord.

Family: my wonderful siblings, Oluwatoyin Olorunisomo, Seun Adyemi, Nike Adeyemi and Adeoye Adeyemi for all your unrelenting encouragements, various sacrifices and invaluable support through this journey, I am so grateful. The pastors, Pastor Austen Ayo, Pastor 'Toye Abioye the leadership and members of the Redeem Christian Church of God Jesus House for all Nations, Pretoria, where I found a home away from home, thank you for spurring me on to success and for your prayers.

I would like to thank Mr. and Mrs. Yemi Ogun, (my in-laws), Pst. & Pst (Mrs.) Oloyede Ojo, and Mr. Bisi Ojo for their love and prayers. I am also indebted to Prof. and Dr. (Mrs.) Solomon Badejo, former Executive Director of Forestry Research Institute of Nigeria. Dr. Aderoke Adio, your kind assistance and support during my study leave application is highly appreciated.

I am particularly thankful to my wonderful, loving, caring and God given wife, Dr. Modupeoreoluwa Abosede Adeyemi for the great sacrifices you have made for the success of this noble endeavour, and for your patience, support and prayers, thank you, thank you, thank you.

I could not forget to thank my children, Emmanuel Adebayo and Nathaniel Modadeoluwa, your arrival during our PhD studies was indeed a blessing to the family. I love you boys

Lastly, to all those whose names are not mentioned, your love will not be forgotten by the Father in heaven and may He bless us all Amen.

RESEARCH FUNDING

The financial assistance of the National Research Foundation (NRF) towards this research is hereby acknowledged. Opinions expressed and conclusions arrived at, are those of the author and not necessarily to be attributed to NRF

FINANCIAL ASSISTANCE

3-year doctoral scholarship NRF - Collaborative Postgraduate Training Programme grant (CPT160424162937).

3- year University of Pretoria- UP Doctoral Bursary.

LIST OF ABBREVIATIONS

AEL's	Atmospheric Emission Licenses
APPA	Atmospheric Pollution Prevention Act
AQMP	Air Quality Management Plan
CAPCO	Chief Air Pollution Control Officer
CBD	Cerebrovascular Diseases
CI	Confidence Interval
CO	Carbon Dioxide
COPD	Chronic Obstructive Pulmonary Diseases
CVD	Cardiovascular Diseases
DEFF	Department of Environment, Forestry and Fisheries
GHG	Green House Gases
HRV	Heart Rate Variability
ICD10	10th Version of the International Classification of Diseases
IQR	Inter-quartile Range
LRT	Long range transport
NAAQS	National Ambient Air Quality Standards
NEMAQA	National Environmental Management: Air Quality Act
NO_2	Nitrogen Dioxide
NO_x	Nitrogen Oxides
O_3	Ozone
PM10	Particles smaller than 10μm
Pb	Lead
ppb	Parts per Billion
RD	Respiratory Diseases
SAAQIS	South African Air Quality Information System
SO_2	Sulphur Dioxide
UNEP	United Nations Environment Programme
USA	United States of America
VOC	Volatile Organic Compound
WHO	World Health Organization
$\mu g/m^3$	Micrograms Per Cubic Meter

ABSTRACT

Background: Air pollution is one of the major problems being faced by most of the big and industrial cities of the world and has become a major environmental threat over the last few years. This environmental threat has gained more attention because of its increased health effects on humans which includes morbidity and mortality. It has various adverse effects, such as increased pulmonary infections, respiratory diseases, acute illnesses and hospitalizations and can eventually lead to death. The need for source identification and abatement strategies for air pollution is crucial in order to meet the SDG goal to reduce pollution by 2030. Therefore, this study investigated the characterisation of $PM_{2.5}$, source apportionment, origin of air masses into Pretoria alongside the association between air pollutant and hospital admissions due to respiratory diseases.

Method: This study was divided into primary and secondary data collection phases. For the primary data collection, daily 24-hour $PM_{2.5}$ samples were collected every third day between 18 April 2017 and 17 April 2018 at an urban background site in Pretoria. A total of 122 $PM_{2.5}$ samples and 25 duplicate $PM_{2.5}$ samples were collected and analysed for particulate mass, soot, black carbon (BC), organic carbon (OC) and 18 trace elements. Source apportionment analysis was conducted on this dataset using the positive matrix factorisation method. Air mass trajectories, as a surrogate for distant sources of $PM_{2.5}$, were estimated using the HYSPLIT model (version 4.9). The daily average trajectories were calculated backwards for 72 h and used for cluster analysis. The clustering algorithm coupled in HYSPLIT was based on the distance between a trajectory endpoint and the corresponding cluster mean endpoint.

The secondary data, daily hospital admissions, PM_{10}, NO_2 and SO_2 data, used for this project were obtained from a private hospital group and the South African Air Quality Information System, managed by the South African Weather Services. The time-stratified case-crossover epidemiology study design and conditional logistic regression models were applied to investigate the association between PM_{10}, NO_2, SO_2 and respiratory disease (RD) hospital admissions during the study period 1 January 2011 to 30 November 2014.

Results: The annual mean for $PM_{2.5}$ (n =122 days) was 21.1µg/m^3 (range 0.7 - 66.8 µg/m^3). The highest $PM_{2.5}$ mean value was recorded during winter, which was significantly higher than autumn, spring, and summer ($p<0.0001$). No significant difference between weekdays and weekend ($P>0.9567$) was observed. Most exceedances of $PM_{2.5}$, when compared with daily

World Health Organization (WHO) guidelines and South African standards were observed in mid-autumn and winter. Soot, BC and OC followed the same trend as $PM_{2.5}$ concentration. Average S (1480 ng/m^3) concentration was the highest among elements detected, followed by Si, Fe, K and Ca, in that order. Seven sources and their contributions to the total $PM_{2.5}$ were identified and quantified. These included vehicle exhaust – 8.6%, and base metal/ pyrometallurgical - 0%, soil dust -13.2%, secondary Sulphur – 31.4%, vehicle exhaust – 12.5%, road traffic – 7.3%, coal burning -27.2%, while the percentage of $PM_{2.5}$ specie in the base metal/ pyrometallurgical factor was 0%. The identified source factors exhibited seasonal variations, coal burning and secondary Sulphur being the highest during winter while soil dust and road traffic were lowest during summer.

Five transport clusters were identified during the 1-year study period: National Limpopo (Nat-LP), transboundary (TB), Easterly-Indian Ocean, South Easterly-Indian Ocean and South Westerly-Atlantic Ocean. In addition to this, 85% of the transport clusters were of local and transboundary origin, 15% were long-range transport, while cluster 1 had the highest $PM_{2.5}$ concentration. Cluster 1 can be attributed to main source of pollution contributing to the PM level at the sampling site due to the activities going on in the region, such as biomass burning, coal mining.

Of the 17,647 hospital admissions in Pretoria, 51.8% (n=9,147) were women and 61.6% (n=10,870) were 0-14-year old. In the unstratified analysis, a 10 μg/m^3 increase in PM_{10} was associated with statistically insignificant increase of 0.2% (-0.7%; 1.2%) in RD hospital admissions; no significant association was observed for NO_2 during cold days. Significant association between SO_2 and RD hospital admissions was observed for females and male patients during warm and cold days.

Conclusion: This project contributes to the very few source apportionments studies of $PM_{2.5}$ in Africa and specifically South Africa. Coal burning remains one of the main sources that should be addressed. Late autumn and winter season recorded the highest concentration. The risks of RD hospital admission due to PM_{10} exposure in Pretoria were higher on warm days than on cold days. The apportioned sources and the origin of air masses from this study align with the known existing sources in the country. Oceanic influences, local and transboundary sources (Southern African countries) contribute to the air masses passing over Pretoria, therefore, abatement strategies are paramount to reduce the level of pollution during this time.

The findings of the study can be of help to the government in the formulation of air pollution guidelines as a measure to mitigate the effect of air pollution on the environment. In addition to this, if there is strict compliance to the already formulated regulation on the identified sources, this will significantly reduce the effect of air pollution in our cities. Lastly, the outcome of this project will help the South African government in their air quality management plan that are reviewed regularly.

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CHAPTER ONE: INTRODUCTION

1.1 Air pollution: the world's worst environmental health risk

Developed and developing countries of the world continue to be faced with the problems of air pollution both outdoors (ambient) and indoors (household) which is a major environmental health risk. For a few decades, air pollution has been a growing concern with an increase of acute episodes of its occurrence in many cities of the world. According to a report by WHO in 2014, it was estimated that over 7 million people die prematurely due to air pollution every year, i.e. one in every eight deaths globally, which is far more than the combined deaths recorded as a result of HIV, tuberculosis and malaria. With respect to the above, air pollution has been tagged as a major risk factor in Africa.[1,2]

The alarming rate at which air pollution is rising invariably impacts negatively on the economy and the quality of life thereby making it a public health emergency.[3] The WHO (2015) estimated that more than 36 million deaths are recorded each year as a result of NCD, and nearly 80% of these are from low to middle-income countries, such as South Africa.[4]

However, exposure to outdoor air pollutants is beyond what individuals can regulate and therefore needs attention and urgent action by public authorities at various levels e.g. national, regional and international. The WHO, in 2015, passed a global road map to address health effects due to air pollution.[3] The road map urges "*.....Member States to develop air quality monitoring systems and health registries to improve surveillance for all illnesses related to air pollution; promote clean cooking, heating and lighting technologies and fuels; and strengthen international transfer of expertise, technologies and scientific data in the field of air pollution*". In recent years, and despite the tremendous effort being put in place to clean up the environment, air pollution has remained a major health problem to the public globally and the occurrences are greatest in developing countries.

For example, the National Environmental Management: Air quality Act (AQA Act No.39 of 2004) of South Africa was put into effect in 2005 as a method to manage the air quality of the country, by introducing air quality management (AQM) as the control strategy. In 2009, national ambient air quality standards were established for seven different pollutants for sulfur dioxide (SO_2), nitrogen dioxide (NO_2), particulate matter (PM_{10}), ozone (O_3), benzene (C_6H_6), lead (Pb), and carbon monoxide (CO) Despite the promulgation of this act, currently the

country has no ambient metal standards in place except for lead, making it difficult to protect the general population from the adverse effect of these pollutants[5].

The existence of foreign substances in the air, which affects the health and well-being of living things, is termed air pollution[6]. Air pollution is a complex mixture of particles, vapours, and gases, which have their origin from either natural or anthropogenic sources. Anthropogenic sources include emissions from transport and industrial sectors, burning of biomass fuel, re-suspended dust, and inadequate waste burning and management. According to a review by Scorgie et al[7], it was reported that the main anthropogenic sources of particulate matter emission into the atmosphere in South Africa includes motor vehicles, industries burning fossils fuels in equipment or machines that come without emission regulator devices, and residential areas where the use of coal, firewood and kerosene is high. i.e. areas without electricity supplies.

Sulphur dioxide (SO_2), nitrogen dioxide (NO_2), nitrogen oxides (NOx), carbon monoxide (CO), volatile organic compounds (VOCs), POPs, ozone, lead and particulate matter (ambient PM_{10} and $PM_{2.5}$) are most commonly monitored and require management on local and regional levels. Particulate matter (PM) is an important atmospheric pollutant due to its impact on local and regional air quality[8], and adverse effect on visibility[9], climate system[10] and human health. Most trace metals constitute only about 1% of the total PM but play a critical role in the identification source. Therefore, sources can be precisely and clearly classified using specific trace elements.[11]

In addition, the investigation of these sources can be achieved by source apportionment. Source apportionment studies are used to determine the contributing sources to ambient PM measured at a particular monitoring site. Chemical compounds determined in the measured PM defines some "chemical fingerprint," which hints of the attribution of the PM to sectors as sources of ambient PM[12]**.** The positive matrix factorisation (PMF) is one of the methods used to determine the source profile and source contribution to the total $PM_{2.5}$ based on the knowledge of the variation of the elemental concentration, their periodicity, correlation with other elements and meteorological parameters[13].

Aerosol particles concentrations in an urban area are affected by several factors such as exhaust, non-exhaust and suspension emissions. Beside these factors, industrial emission and long-range transport can affect the PM number and mass concentration[14].

Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model is widely used in determining the origin or fate of air parcels in the atmosphere[15]. The use of air quality data with back trajectory analysis can be used to determine the potential source area for the LRT.[16-19]

Furthermore, there has been significant interest in recent years in the health effects of exposure to both short fluctuations and long-term levels of air pollution, especially the common environmental pollutants which includes PM, ozone, carbon monoxide, nitrogen dioxide and sulphur dioxide[20].

Brunekreef and Holgate[21] reported that increased mortality and morbidity worldwide has been traced to temporary or acute exposure of ambient air pollution in which particulate matter is of great concern. Epidemiological studies such as cohort and time series studies have associated higher concentrations of PM to increased morbidity and mortality[22-25]. Most of these studies have evaluated the health effects of particles expressed as the risk per unit mass/m^3 of PM_{10} or $PM_{2.5}$. However, the cause-effect chain is thought to be very complex, including issues such as chemical composition and physical characteristics of the inhaled particles, and it is not yet clear which causative agents and underlying mechanisms are responsible for the adverse health effects. Therefore, identification of sources and characteristics of particulate matter is of utmost important because it is responsible for the documented health burden to the public.

1.2 Rationale for the study

Outdoor and indoor air pollution in South Africa have been perceived to be a serious problem with the emissions of different air pollutants such as SO_2, NO_2, particulate matter VOC etc., and their corresponding concentrations in the atmosphere being of great concern. Emission of pollutants from various sources in different parts of the country have greatly imparted the air quality, thus creating a serious threat to the vital roles the atmosphere plays in protecting and supporting life through the absorption of dangerous ultraviolet radiation, warming the surface and regulating the earth temperature.[26] Outdoor air quality remains a key issue in South Africa, especially in cities such as Cape Town, Durban, Johannesburg and Pretoria[27-29]. Considering South Africa as one of the middle-income countries, it is difficult to quantify the impacts of air pollution around the cities due to limited availability of information on exposure to air pollution and its effects on health of the people.

Thus far, there is no air quality programme for the WHOs Africa region, but they exist in other WHO regions. This is of the strategic importance to the study. At the United Nations Sustainable Development Summit on 25 September 2015, world leaders adopted the 2030 Agenda for Sustainable Development. The 17 Sustainable Development Goals (SDGs) (valid 2016-2030) included the vision to end poverty, fight inequality and injustice, and address environmental risk factors such as air pollution and climate change by 2030. They replaced the Millennium Development Goals that were valid from 2000-2015. One of them also addresses air pollution explicitly, namely Goal 3.9, which reads as follows: By 2030, substantially reduce the number of deaths and illnesses from hazardous chemicals and air, water and soil pollution and contamination.[30]

Therefore, it is important to take cognizance of the report released by WHO[3] in 2016 regarding the update of the global urban ambient air pollution database. Despite this overall progress in formulating progressive environmental legislation in South Africa, the breath of possible health impacts of air quality in South Africa has not been comprehensively investigated in epidemiological studies thus constituting a major research gap. Hence, this study characterises $PM_{2.5}$, apportion sources and determines the origin of air masses into Pretoria. Case cross-over design was used to investigate the association between air pollutant and respiratory hospital admissions. Lastly, the results of this project will contribute immensely to the City of Tshwane AQMP, to ensure effective mitigation strategies are implemented.

1.3 Research question

The main research question addressed in this project was:

1. What are the likely sources of ambient $PM_{2.5}$ in Pretoria?
2. Are there any associations between air pollution, respiratory disease and hospital admissions in Pretoria, South Africa?

1.3.1 Aim

This project aimed to investigate the characterisation of $PM_{2.5}$, source apportionment, origin of air masses into Pretoria from 18 April 2017 to 17 April 2018, and the association between hospital admissions due to respiratory diseases and air pollutants (PM_{10}, NO_2 and SO_2) over the period of January 2011 to November 2014.

1.3.2 Objectives

This project had five objectives, namely:

1. Collection of $PM_{2.5}$ filter samples at one site in Pretoria over a period of one (1) year
2. To determine the soot, BC, OC and trace elements content of the collected $PM_{2.5}$ samples
3. To identify the sources of pollution that contributes to $PM_{2.5}$ in Pretoria using Positive Matrix Factorization (PMF).
4. To identify the geographical origin of air masses (surrogate for distant sources of $PM_{2.5}$) that pass Pretoria by applying the Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model.
5. To determine cases of the associations between air pollutants (PM_{10}, NO_2 and SO_2) and hospital admissions for respiratory diseases in Pretoria South Africa over the period of January 2011 to November 2014.

1.4 Outline of thesis

In the current chapter, the general introduction to the research topic is given, the problem statement, significance of the study, the research aims and objectives, as well as the research questions were addressed.

Chapter 2: Literature review

This chapter focused on air pollution, indoor and outdoor air pollution, characteristics of air pollution, air pollutants, Short term effect of air pollution on hospital admission, Epidemiological studies in South Africa**,** South Africa's policy and laws on air pollution, Air quality management plan in South Africa, state of air pollution monitoring in Africa and source apportionment.

Chapter 3: Research methodology

This chapter focused on the exposure assessment study i.e. study location air sampling monitoring, chemical analysis of $PM_{2.5}$ filters, source apportionment, origin of air masses in Pretoria and statistical analysis. The epidemiology study, i.e. study design, data analysis, statistical analysis (calculation of the associations between air pollutants and RD hospital

admissions), were presented. Lastly, ethics application was submitted to the Faculty Research Ethic Committee for approval.

Chapter 4: Overview of $PM_{2.5}$, soot, BC, OC and trace elements

This chapter informs the reader about the findings of the study as it related to objective one two and four, i.e. collection of $PM_{2.5}$ filter samples in Pretoria, determination of soot, BC, OC and trace elements contents of the collected sample. Lastly, there is a section involving origin of air masses in Pretoria. (i.e. surrogate for distant sources of $PM_{2.5}$)

Chapter 5: $PM_{2.5}$ source apportionment

This chapter presented the findings on the identified sources of $PM_{2.5}$ in Pretoria alongside the origin of air masses (transport clusters).

Chapter 6: Epidemiological study

This chapter focused on the results of the association between air pollutants and RD hospital admissions, which is written in article format.

Chapter 7: Conclusion and recommendations

This chapter summarised the main findings of the project, its strengths and limitations, as well as recommendations.

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CHAPTER TWO: LITERATURE REVIEW

2.1 Air pollution

Air pollution, namely outdoor and indoor, originates from different sources both natural and man-made (anthropogenic) including industrial processes, road traffic, combustion of biomass, windblown sand and volcanoes. Air pollution can be classified as either primary, i.e. direct emission from a process, or secondary, formed due to a chemical reaction between the primary pollutants in the air (e.g. ground level ozone). In South Africa, the major sources of pollution include power generation, industrial processes, waste disposal, transportation, biomass burning, landfill sites, wastewater treatment, agriculture and burning dirty fossil fuels in appliances that are outdoors and domestic use of coal, wood and paraffin which are mostly indoor[1]. In addition, biomass burning is also a significant source of gases and particulate matter emissions to the atmosphere. Pollutants associated with biomass burning include greenhouse gases (carbon dioxide (CO_2), methane and nitrous oxide (NO), carbon monoxide (CO), and Volatile Organic Compounds (VOCs) especially in tropical and subtropical regions. South Africa has been reported as the ninth highest atmospheric sulphur emitting country and biomass burning emissions from this region are known to have a global effect.[2,3,4]

2.1.1 Indoor air pollution

Indoor air pollution can be referred to as pollution inside or near the household. Cooking, lighting and space heating are the major contributors to household air pollution. Generation of electricity in South Africa significantly increased after the apartheid era, resulting in the increase of household using electricity for lighting and cooking from 57% in 1996 to 84% in 2011[5]. Furthermore, according to the census conducted by Statistics South Africa in 2011, around 26% of household still utilise fuels other than electricity for daily cooking[6]. Sources of indoor air pollutants include inefficient heating stoves, cooking stoves, and open fire from burning of coal or biomass. Indoor air pollution, black carbon and emissions can be significantly reduced by adopting cleaner fuels and more efficient stoves, this in turn improves health conditions decreases demand for fuel, and provides economic and other health benefits[7,8]. The contribution of household air pollution to ambient air pollution has been estimated to be responsible for about 12% of outdoor combustion-derived particulate matter $PM_{2.5}$ (mass of particles smaller than 2.5 μm with 50% cut-off point) globally[9]. Exposure to household air pollution is known to be a leading environmental risk factor for ill health. Bonjour *et.al*[10] reported that globally, nearly 2.8 billion people use solid fuels primarily for

cooking and the WHO reported that approximately 4.3 million premature deaths are attributed to household air pollution exposures (Figure 2.1).

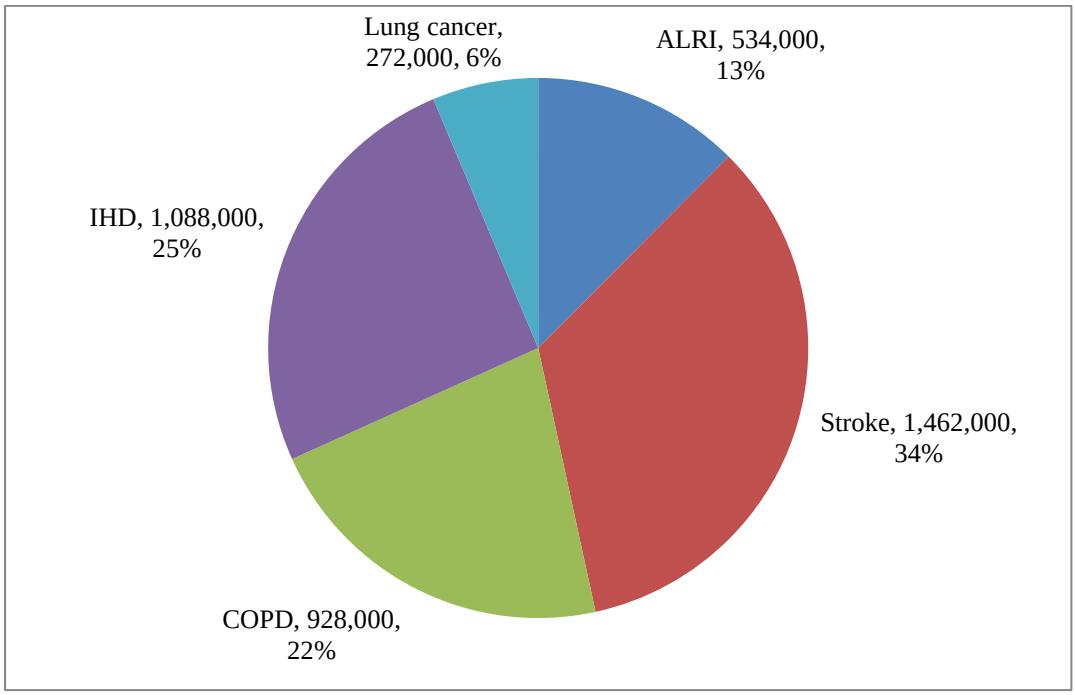

Figure 2.1: Deaths attributable to household air pollution, 2012. ALRI = acute lower respiratory infections; IHD = ischemic heart disease; COPD = chronic obstructive pulmonary disease. Source: WHO, 2014[11]

2.1.2 Outdoor air pollution

Outdoor air pollution is known to be a worldwide problem associated with serious effect on human health. In 2011, approximately 80% of the world's population were exposed to air pollution level that exceeded the WHO air quality guidelines[12]. Air quality globally is affected by pollutants emitted by numerous sources while the corresponding concentrations in the atmosphere cause a great concern. The major contributors to these pollutants in South Africa are mining, automobiles, biomass burning and heavy industries using biomass fuel for energy production. Outdoor air pollution is known for large emission of various pollutants such as SO_x, NO_x PM and CO. Most recent studies on outdoor air pollution have focused on $PM_{2.5}$ particles due to its capability to penetrate the lung tissue and induce local and systemic effects[13]. It is noteworthy that the International Agency for Research on Cancer (IARC) has recently classified outdoor air pollution as carcinogenic to humans (Group 1)[14]. In addition, some studies have reported the association that exists between outdoor air pollution and various health conditions such as asthma, cardiovascular diseases, respiratory infections, adverse birth outcomes and additional cancers, such as leukemia.[15-17].

2.2 Characteristics of air pollution

According to the WHO,[18] "A*ir pollution is contamination of the indoor or outdoor environment by any chemical, physical or biological agent that modifies the natural characteristics of the atmosphere"*. Air pollution is a complex mixture of many components, which complicates the linking of health effects to air pollution. Six common air pollutants have been identified and prioritised as a concern globally[19]. Particulate matter e.g. $PM_{2.5}$ and PM_{10} (mass of particles smaller than 10 μm with 50% cut-off point), ground level ozone, carbon monoxide, sulphur dioxide, NO_x (NO_2 and NO combined) and lead (Pb).

These six air pollutants are referred to as "criteria" pollutants because they are regulated in different countries. The correlation that exists between these pollutants makes it difficult to know which pollutants (or combination of pollutants) are causally associated with the negative effect.

2.2.1 Air pollutants and sources

Air pollutants measured in the urban atmosphere originate largely from human activities. Source of pollution can be stationary emission or mobile emission.[20]

- **Stationary sources**: this can be grouped into (a) Rural area sources such as agricultural production, mining and quarrying, (b) Industrial point and area sources such as manufacturing of chemicals, non-metallic mineral products, basic metal industries, power generation, and (c) Community sources, e.g. heating of homes and buildings, municipal waste and sewage sludge incinerators, fireplaces, cooking facilities etc.

- **Mobile sources:** this includes any form of combustion-engine vehicles, e.g. light duty gasoline powered cars, light and heavy-duty diesel-powered vehicles, motorcycles, aircraft, and even line sources such as fugitive dusts from vehicle traffic.

Air pollutants can be classified as either primary, i.e. direct emission from a process, or secondary, formed due to chemical reactions between the primary pollutants in the atmosphere (e.g. ground-level ozone). The characteristics of the main chemical pollutants and their sources are summarised in Table 2.1

Table 2.1: Summary description of the major air pollutants. Adapted from Ballester, 2005[21]

Pollutant	Formation	Physical state	Sources
Suspended particulates (PM_{10})	Primary and secondary	Solid, liquid	Vehicles (mainly diesel), industrial process, black smoke, tobacco smoke
Sulphur dioxide (SO_2)	Primary	Gas	Industrial processes, vehicles
Nitrogen dioxide (NO_2)	Primary	Gas	Vehicles Gas heater and cookers
Carbon monoxide (CO)	Primary	Gas	Vehicles (mainly Petrol), Tobacco smoke
Volatile organic Compounds (VOCs)	Primary and secondary compounds	Gas	Vehicle, industry, tobacco smoke
Lead (Pb)	Primary	Solid (fine particulates)	Vehicles, industry
Ozone (O_3)	Secondary	Gas	Vehicles (secondary to photo- oxidat NO and volatile organic compounds)

2.3 Criteria pollutants

Criteria pollutants include air pollutants that are known to be harmful to public health and the environment. Currently, seven criteria pollutants have been tagged globally and in South Africa. In addition, biomass burning has been identified as an important source of pollution in Southern Africa. In South Africa, a primary health-based National Ambient Air Quality Standards (NAAQS) under National Environmental Management Air Quality Act (NEMAQA) has sets the safe amount of the pollutants that can be present in the atmosphere[22]. The most common air pollutants in South Africa are shown in Table 2.2. The criteria air pollutants are a good indicator of air quality in general.

Table 2.2: Pollutants of concern in South Africa Adapted from DEFF, 2017[23]

Criteria pollutants declared in terms of section 9 of AQA	Priority Pollutants declared in terms of section 29 of AQA	Possible future pollutants	
		National pollutants	**Local pollutants**
Sulphur dioxide Nitrogen dioxide Ozone Carbon monoxide Lead (Pb) Particulate matter ($PM_{2.5}$, PM_{10}) Benzene	Carbon dioxide (CO_2) Methane (CH_4) Nitrous oxide (N_2O) Hydrofluorocarbons (HFCs) Perfluorocarbons (PFCs) Sulphur hexafluoride (SF_6)	Mercury Dioxins Furans POPs Other VOCs N_2O	Chrome (Cr_6) Fluoride (particulate and gas) Manganese (Mn) Hydrogen Sulphide Asbestos Black carbon

2.3.1 Nitrogen oxides

The main anthropogenic sources of NO_x are mobile and stationary combustion processes. Nitrogen oxide is emitted as nitrogen monoxide during the combustion of fossil fuels used in transport, heating and energy generation. Nitrogen dioxide is formed in the atmosphere by the oxidation reaction of nitrogen monoxide and ozone. In its free radical state, NO_2 has been found to cause inflammation and injury to the airway and has been implicated in number of airways disorders[24]. NO_2 absorbs visible solar radiation and contributes to impaired atmospheric visibility.

2.3.2 Carbon oxides

Carbon monoxide and carbon dioxide are the major oxides of carbon and they are released into the atmosphere by either complete or incomplete combustion of fuels. The major ambient source of CO is from incomplete combustion of fossil fuels from automobiles exhaust systems. CO_2 is known to be the main pollutant that is responsible for the greenhouse effect. CO is known to be toxic and fatal when inhaled, this is due to the reaction with hemoglobin thereby reducing the supply of oxygen to the body.[24]

2.3.4 Sulphur dioxide

The anthropogenic source of SO_2 is from combustion of sulphur containing fossil fuels such as coal and heavy oils, smelting of sulphur containing ores, volcanoes and ocean are the major natural sources. Sulphate particles are formed due to atmospheric oxidation of SO_2. There has been a major reduction in the emission of SO_2 due to the changes in the type of fuels used in Western Europe. The exposure to SO_2 has been found to be a major cause of respiratory problems.[19,25-27]

2.3.5 Ground-level ozone

Ozone is a secondary pollutant. The formation of ozone in the urban areas involves the atmospheric reaction between NO_x, oxygen, hydrocarbon and ultraviolet. Light concentration of ozone in the atmosphere is associated with increased temperature. O_3 is created by chemical reactions between air pollutants from vehicle exhausts, gasoline vapours, and other emissions. Induced inflammation in the respiratory tract, impaired pulmonary functions, and increased risk of cardiovascular problems are health effects associated with exposure to ozone[19]. O_3 has been found to trigger health problems in people of all ages, especially children, and the elderly who have lung diseases such as asthma[28]

2.3.6 Volatile organic compounds

VOCs are compounds of hydrocarbons present in the atmosphere, which includes alkanes, alkenes, aromatics, aldehydes, ketones, alcohols, esters, and some chlorinated compounds. These compounds are formed during the incomplete combustion of carbonaceous materials. Many VOCs are carcinogenic and mutagenic in nature, e.g. benzene has been a major concern due to its carcinogenicity. Biogenic volatile organic compounds are emitted from the terrestrial ecosystems into the atmosphere where they influence the chemistry and composition of the atmosphere including aerosols and oxidants.

2.3.7 Lead

Lead is one of the metals found in the environment. Leaded petrol contributes to the amount of Pb found in the air, other sources involve anthropogenic sources which includes fuel combustion, industrial process and solid waste combustion.

2.3.8 Polycyclic aromatic hydrocarbons

Polycyclic aromatic hydrocarbons (PAHs) are widespread environmental pollutants with two or more fuse aromatic (e.g. benzene) rings that are formed as a result of incomplete combustion of carbonaceous materials at high temperarute[29]. PAHs (2 and 3 rings) occur in the atmosphere largely in the vapour phase, whereas multi-ringed PAHs (5 rings or more) are largely bound to particles. Particle-bound PAHs are known to be hazardous to human health. Benzo(a)pyrene is often used as a marker for total exposure to carcinogenic PAHs, as the contribution of Benzo(a)pyrene to the total carcinogenic potential is high[30]. Emissions from traffic have been found to be the main outdoor source for the indoor PAH concentration in urban and suburban

locations in many industrialised countries[31]. Vehicular emissions of PAHs account for about 46–90% of the mass of individual PAHs in ambient air particles in urban areas[32]. The burning of fossil fuel, solid fuel and biomass has been documented as a significant source of airborne PAHs as it releases a wide range of air pollutants, including PAHs, which are emitted to the indoor atmosphere in unvented or flueless combustion and to the outdoor air[33], this study reported that about 500 PAHs and related compounds are detected in the air, but most measurements have been carried out on Benzo(a)pyrene[34].

2.3.9 Particulate matter

Ambient particulate matter (PM) is a complex and varying mixtures of solid and liquid particles that are suspended in the air. The particles vary in size and composition and are produced due to natural and anthropogenic activities.[35] Sulphates, nitrates, ammonia, sodium chloride, black carbon, hydrocarbons, mineral dust and water are found to be the major components of particulate matter. Particulate matter can be derived from variety of sources. PM emitted directly from its source is known as primary PM while those generated through atmospheric chemical reaction of gases are termed secondary PM. PM has both natural and anthropogenic sources such as industry, power plants, refuse incinerator, motor vehicles, construction activity, fires and natural windblown dust. Particulate matter can be suspended in air for hours or days and can travel over a considerable distance, hence PM is a long-range transported pollutant that depends on size, chemical composition, and other physical and biological properties of particles. The PM measured at the ground level is largely form dust, industrial secondary sulphate, and secondary organics but significant attention has been focused on the impact of biomass burning on particulates in the country.

PM is categorised into ultrafine particles (UFP), (particles that are smaller than 0.1 µm, (i.e. 100 nm), fine particles, particles that are smaller than 2.5 µm and coarse particles, which are particles between 2.5 and 10 µm. While the measures of PM vary across the globe based on local sources, $PM_{2.5}$ generally represents 40-70% of the total PM mass.[19] Some studies have shown that $PM_{2.5}$ arising mainly from man-made sources are more harmful than coarse particles[36-40] and, therefore, the measurement of PM in health effect studies has currently focused on $PM_{2.5}$ rather than on coarse particles ($PM_{10-2.5}$). However, some studies have also detected adverse health effects related to coarse particles[39,41-42] highlighting the importance of controlling the concentration and studying the potential health effects of all PM size fractions.

2.3.10 Biomass burning in Southern Africa

Southern Africa has been the focal point of intensive environmental research, which is directed towards aerosol and trace gas emissions, transport, owing to its different sources of aerosols and gases[42-46]. During the Atmosphere Research Initiative SAFARI campaign programme1992-2000, biomass burning was documented as a common phenomenon occurring on the savannah plains located to the south of the Equator[42, 47]. The SAFARI campaign of 1992 and 2000 investigated the emissions from savannah fires incidences in Southern Africa, their transportation across the continent, and the relationship between fires and savanna ecology. The global biogeochemical processes reportedly been influenced by emissions such as CO_2, NO_X and CH_4, and other compounds originating from these fires [48].

Globally, Southern Africa is an important source region of atmospheric pollutants, for example a prominent NO_2 hotspot was seen on global maps of NO_2 satellite retrievals over the South African Highveld; Statistics South Africa in 1990 reported that South Africa was the ninth highest atmospheric sulphur emitting country and biomass burning emissions from this region are known to have a global effect[49-51]. Regional biomass burning is reported to occur between June to October[52]

Biomass burning remains the source of PM, inorganic species, gases and thousands of organic compounds that are emitted to the atmospheres. Green houses gases (carbon dioxide, methane and nitrous oxide), carbon monoxide, and VOCs especially in the tropical and subtropical regions, are pollutants associated with biomass burning. The emissions of these pollutants react with the hydroxyl radicals in the atmosphere, which results in the formation of ozone and other photochemical oxidants[53]. Almost 90% of all biomass-burning emissions are thought to be anthropogenic[53]. Veld fires are a persistent problem in South Africa as they pose a risk to life, and cause damage to property and the environment. According to the National Veldfire Risk Assessment[54], there is a marked trend in fire incidence from the eastern to western parts of the country and, to a lesser extent from northern to southern parts (Figure 2.2)

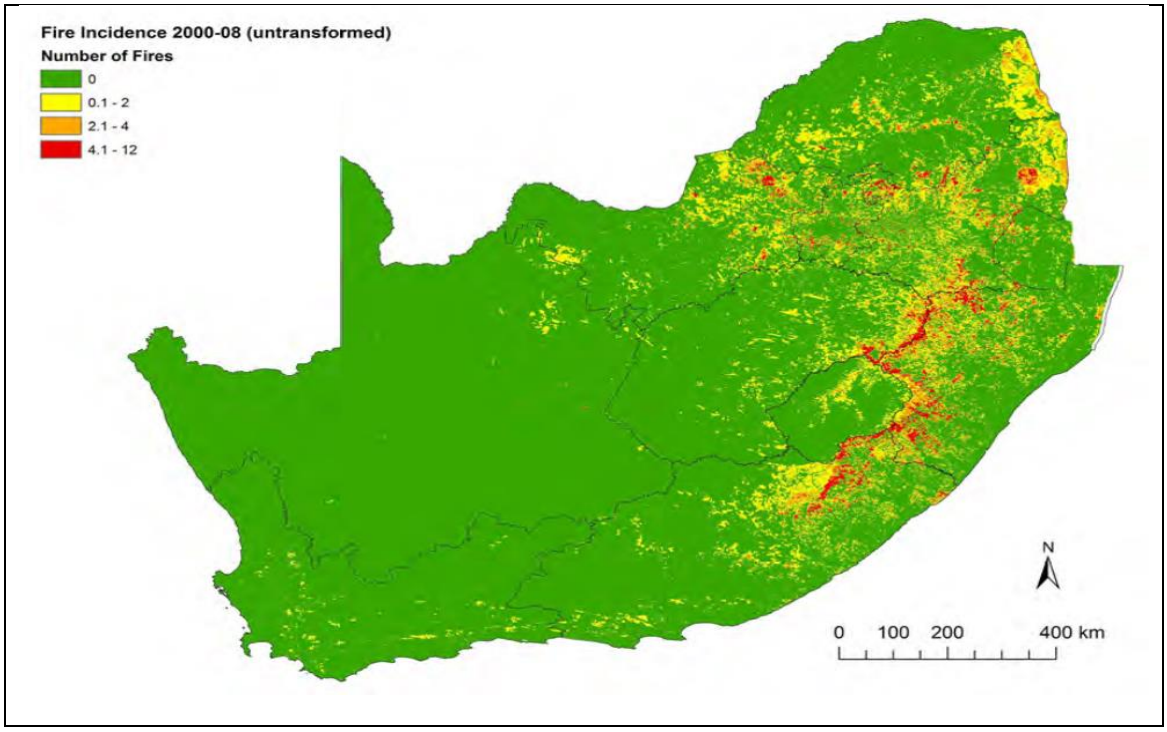

Figure 2.2: Map of fire incidences in South Africa for January 2000 to December 2008 Source: Forsyth et. al[54]

South Africa is not the major culprit for the contributor to atmospheric pollution through biomass burning, but other countries in the sub-region are[55]. Air masses from the north and northeastern parts of the subcontinent transport a substantial amount of products of biomass burning into the country. Andreae *et al*[56] have indicated that the amount of gases emitted from biomass burning may even be comparable to that released from global industrial activities. During the burning months of August-October, air reaching South Africa is most likely to be dominated by biomass burning products[57-58].

Andreae[59] reported that biomass burning accounts for roughly one-half of the atmospheric sources of hydrocarbons in Southern Africa. Piketh *et al*.[60] also reported that biomass burning products, together with aeolian dust, marine aerosols and industrial sulphur are the major contributors to total aerosol loading of the lower tropospheric haze layer throughout the year. During the spring season, however, products of biomass burning contribute significantly to the African haze layer between 10° and 20°S[55, 61-62]. However, products of biomass burning have the least contribution to total inorganic-component aerosol loading over the country[60], the highest contributor to total aerosol loading being industrial sulphur, especially during the summer season. Studies by Hersey *et al*[63] and Magi *et al*[64] reported that biomass burning that

significantly impacts on South Africa originates from neighbouring countries particularly from Zimbabwe and Mozambique.

2.4 Effects of air pollution on health

It is well known that exposure to air pollution can adversely affect human health in a variety of ways, ranging from subtle biochemical and physiological changes to severe illness and death. A comprehensive review is beyond the scope of this research proposal and the reader is referred to the WHO REVIHAAP report.[65] Air pollution is also one of the nine health and environmental risks that are highlighted as potential key risks in the National Climate Change and Health Adaptation Plan (NCCHAP) of the South African National Department of Health. This adds to the NCCRP by expanding further on the potential health impacts of climate change.[66]

Air pollution can result in both acute and chronic effects on human's health by affecting various numbers of systems and organs in the body. This health effect ranges from minor upper respiratory irritation to chronic respiratory and heart disease, lung cancer, acute respiratory infections in children and chronic bronchitis in adults. It also increases the pre-existing heart and lung disease or asthma attacks.[67]

The level or concentration at which pollutants can be detrimental or lead to variety of health problems depends on the type of pollutant, amount of the pollutant one is exposed to, duration and frequency of exposure, and associated toxicity of the specific pollutant. Acute or chronic health effects ranging from sub-clinical effects to premature mortality are associated with the exposure to this pollutant[68].

NO_2 as a free radical has been reported by the WHO[24] to cause inflammation and injury to the airways and is responsible for various airway disorders[24] while CO has been found to be toxic, and sometimes deadly in enclosed spaces. If inhaled, it combines with the haemoglobin therefore reducing the amount of oxygen being transported by the red blood cells[19]. The part of the respiratory tract where the particle will be deposited is a function of its size. The fine and ultrafine particles get to the lung alveoli while PM_{10} is mostly deposited in the upper respiratory tract. However, the size and surface of particles, frequency, period, degree of exposure and their composition are among the important factors that helps in studying the health effects of the particles on humans.[69]

Association between the exposure to fine particles and health effects has been reported in various epidemiological studies in terms of increased mortality, cardiovascular diseases and respiratory illness[15,36,70]. However, a report by the WHO[71] stated that long term exposure to PM is more important than the short-term effects in terms of public health significance and should be of high priority, while the acute effects are also important to study.

The threshold level regarding the relationship between exposure and health outcomes has not been documented despite the risk of various health effects that are associated with increased exposure to PM[19]. The World Health Organization air quality guideline suggested 10 μg/m^3 as an annual mean and 25 μg/m^3 as a 24 h mean for $PM_{2.5}$[19]. Both short- and long-term exposure to air pollutants have been associated with health impacts[72]. In a study by Cohen *et.al*[73], it was reported that for measurable assessment of health outcomes, $PM_{2.5}$ and PM_{10} have been widely used as exposure indicators for most epidemiological studies being carried out on the effect of exposure to air pollution. The health hazards associated with particulate matter are said to be dependent on its physical (such as size, shape and surface area), chemical properties (solubility, chemical species, etc.), toxicological and biological properties as well as the oxidative potentials. The response to health system is determined by the particles ability and where to deposit in the human respiratory tracts[74-75]. The deposition of particles in the lung is determined by particle characteristics (size, shape, composition, electrical discharge density and hygroscopicity), anatomy of the respiratory tract, tidal volume and breathing pattern

Exposure to environmental pollution has been ascertained to be important risk factors for many diseases. Short and long-term increases in mortality rates in relation to air pollution, particularly airborne concentrations of PM, have been well established on the basis of large multi-city studies conducted in the USA[70,38] and in Europe.[37-38] Health effects as a result of exposure to air pollutants depend on the type of pollutant, amount of the pollutant exposed to, duration and frequency of exposure, and associated toxicity of the specific pollutant. The health effects associated with the exposure could either be acute or chronic health effects varying from sub-clinical effects to premature mortality[75].

Health outcomes, such as chronic obstructive respiratory disease, nasopharyngeal cancer, ischaemic heart disease, tuberculosis, lung cancer and eye defects in adults, low birth weight and most importantly acute lower respiratory infection (ALRI) such as pneumonia in children less than age five, are some of the health effects associated with long-term exposure to air pollutants.[76-77] In addition, Brook *et al*[78], confirmed there was a causal relationship between

exposure to $PM_{2.5}$ and cardiovascular death and morbidity. There is increasing evidence that exposure to PM and birth outcomes results in low birth weight and preterm birth as reported by Shah *et al*.[79] The relationship between exposure to fine particle and health effects such as increased mortality, cardiovascular diseases and respiratory illness, has been confirmed through various epidemiological studies.[70,36,15]

The WHO[71], reported a significant reduction in the life expectancy of the average population due to the long-term exposure to high concentration of particulate air pollution. Additionally, in the report, long-term effects of exposure to PM prevail over short-term effects when it comes to matters of public health significance and should be of great concern when considering the acute effects of this problem.[71]

2.5 Sustainable Development Goals on air pollution

This study addresses the United Nations Sustainable Development Goals (SGDs) (valid 2016-2030), specifically with the following three SDGs:

1. Goal 3.9: "By 2030, substantially reduce the number of deaths and illnesses from hazardous chemicals and air, water and soil pollution and contamination."
2. Goal 11.6: "By 2030, reduce the adverse per capita environmental impact of cities, including by paying special attention to air quality and municipal and other waste management" where 11.6.2: specifies "Annual mean levels of fine particulate matter (e.g. $PM_{2.5}$ and PM_{10}) in cities (population weighted)."
3. Goal 13: "By 2030, take urgent action to combat climate change and its impacts."

Many human activities that contribute to air pollution emissions also contribute to climate change. Climate change in turn may influence the health effects from air pollution, by altering levels, chemical composition and transboundary movement[80], which is associated with additional health impacts, e.g. black carbon (or soot), a primary product of incomplete combustion sources, which is a strong climate-altering pollutant and health damaging[81].

South Africa launched its National Climate Change Response Plan white paper (NCCRP) in 2011, prior to hosting the 17th session of the conference of the parties (COP17) to the United Nations framework convention on climate change (UNFCCC) in Durban. The NCCRP calls for increased data collection and research on links between climate and health. Climate change in turn may influence the health effects from air pollution, by altering levels, chemical

composition and trans-boundary movement. Meteorological factors (e.g. temperature, rain, wind speed and direction) influence atmospheric chemical processes, (e.g. rainfall washes away air pollution)[82]. Air pollution is also one of the nine health and environmental risks that are highlighted as potential key risks in the National Climate Change and Health Adaptation Plan (NCCHAP) of the South African National Department of Health, which adds to the NCCRP by expanding further on the potential health impacts of climate change[82].

The SDGS replaced the Millennium Development Goals (valid 2000-2015) and will guide the global development agenda until 2030. The South African Government will have to report on the progress to achieve these SDG goals to the United Nations until 2030. In order to take actions to reduce exposure to air pollution (i.e. intervention step in an AQMP) and its associated health effects, it is essential to know the sources and activities contributing to air pollution. A recent review indicated that only two out of 419 global quantitative air pollution source apportionment studies have been conducted in Africa[66], two more studies were identified through a literature search.

2.6 State of air pollution monitoring in Africa

In a report by WHO, air pollution was reported to be a growing challenge for Africa, this was because of the increased death rate due to outdoor pollution from 164 000 in 1990 to 258 000 in 2017 i.e. a growth of nearly 60%. Factors such as population growth, industrial growth and consumption growth have been reported to increase the levels of pollution in Africa. Africa's population (1.1 billion people) has been forecasted to double by 2050, and more than 80% of this growth will happen in the cities[83]. This growth will impact the continent greatly in the area of traffic volumes and increase the demands for importation of old second-hand vehicles[84]. As the use of fossil fuel increases, air pollution also increases, and this will continue to get worse unless there is a transition to a more sustainable path of growth. Efforts are being put in place by various governments to invest in renewable energies. However, the knowledge of air pollution continues to grow but we do not know the comprehensive full extent of the health impacts and epidemiology, especially in Africa. This is due to considerable data gaps in reliable ground level monitoring of air pollution where there are large and growing populations of children (Figure 2.3).

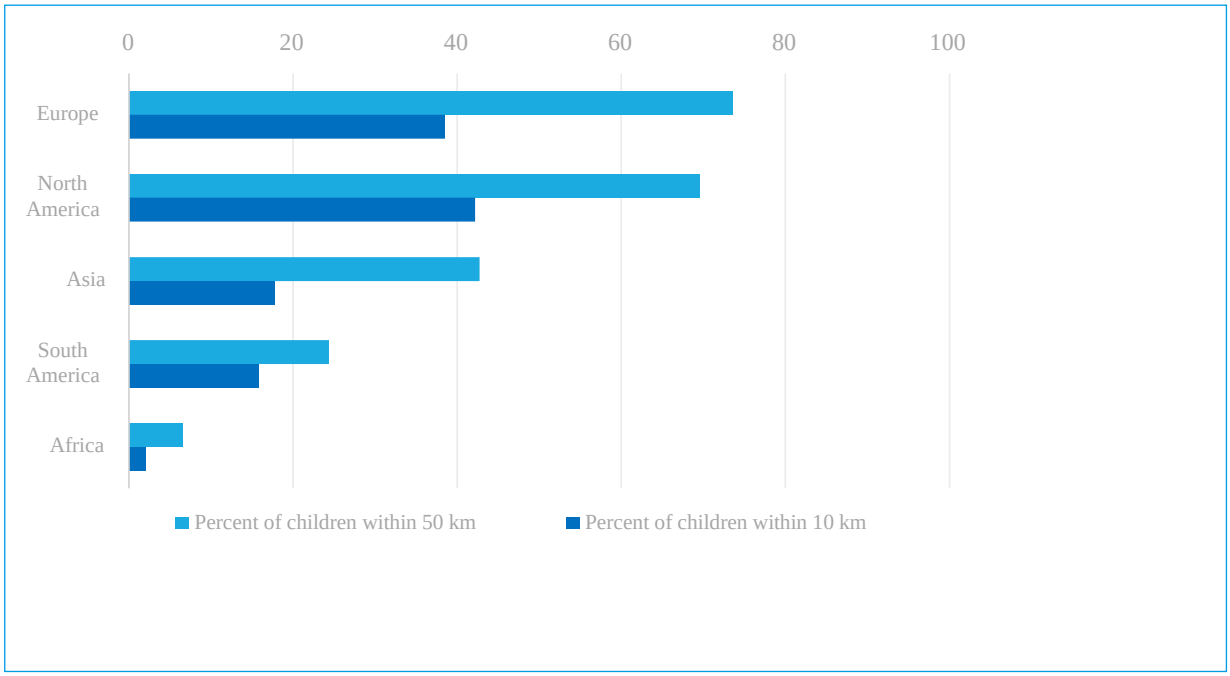

Figure 2.3: Percentage of children living within 10 and 50 km of air monitoring stations (by continent), 2019. (Source WHO 2019)[85]

The overview of ground-level monitoring stations in Africa showed very few stations were available, compared to other regions of the world, the number of African countries that have reliable, real time air pollution monitoring is significantly low, i.e. only seven out of 54 countries (Figure 2.4). This difference is alarming.

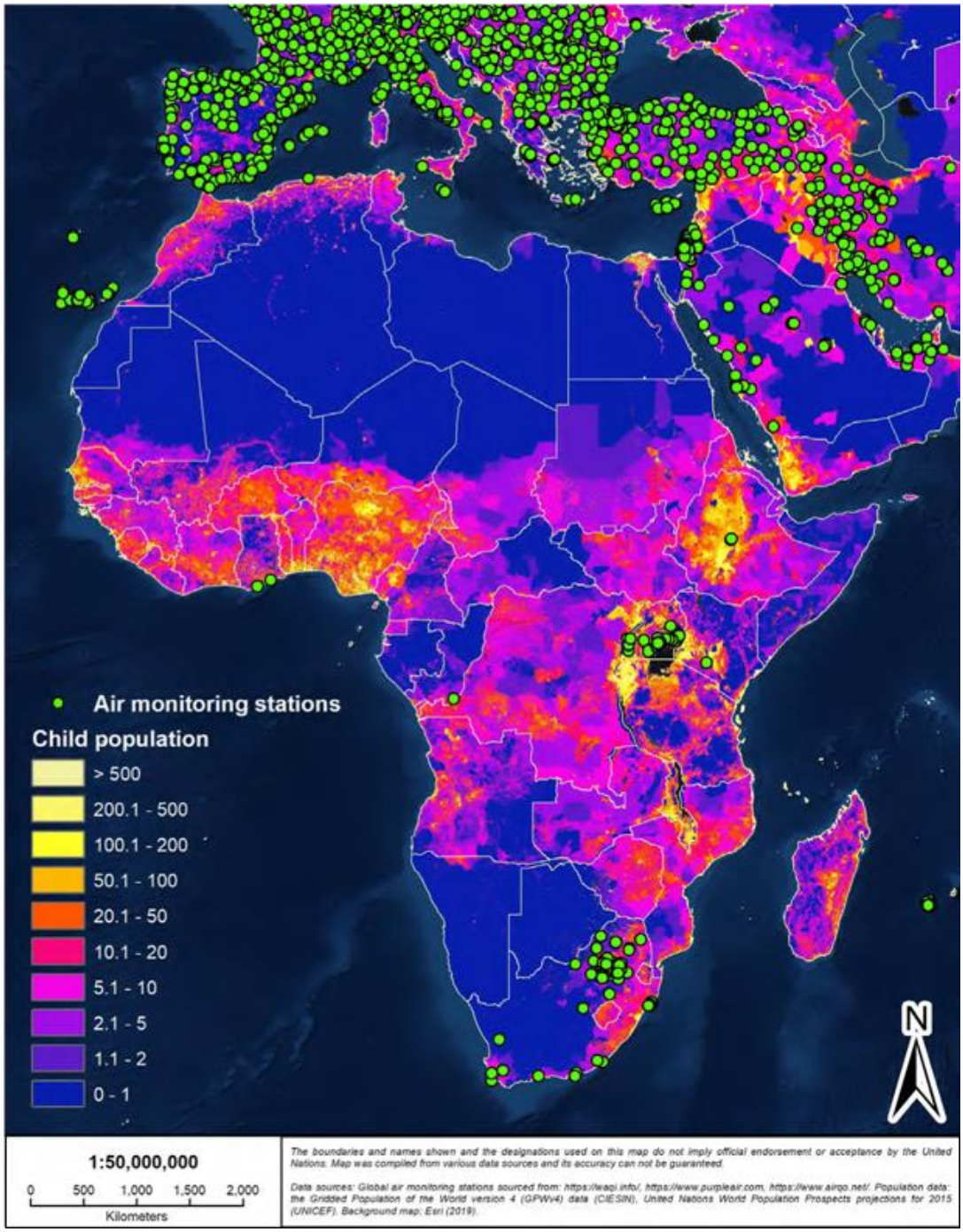

Figure 2.4: Overview of air pollution monitoring in Africa. (Source: WHO, 2019)[85]

The advantage of ground level real time data is that it helps to capture daily even hourly fluctuation that occurs during sampling. It improves public awareness, which helps people tailor their behaviours and action to both reduce air pollution and exposure to it. Ground-level monitoring is very important for identifying sources of pollution, shaping public health policy and informing community level action and intervention that target the most affected, in addition it provides data for epidemiological studies tailored towards how pollutants are impacting human health. The use of satellite based remote sensing helps to address the gaps that cannot

be addressed by local air quality monitoring. Satellite imagery is gaining more attention for tracking long-distance air pollutant transport, filling in data gaps between monitors, and cross-checking ground-based emissions estimates and measurements. However, satellite-based remote sensing does not replace ground-based monitors, and in fact, it is often highly dependent on reference grade ground-level measurements of air pollution to improve the accuracy of their estimates (Figure 2.5).

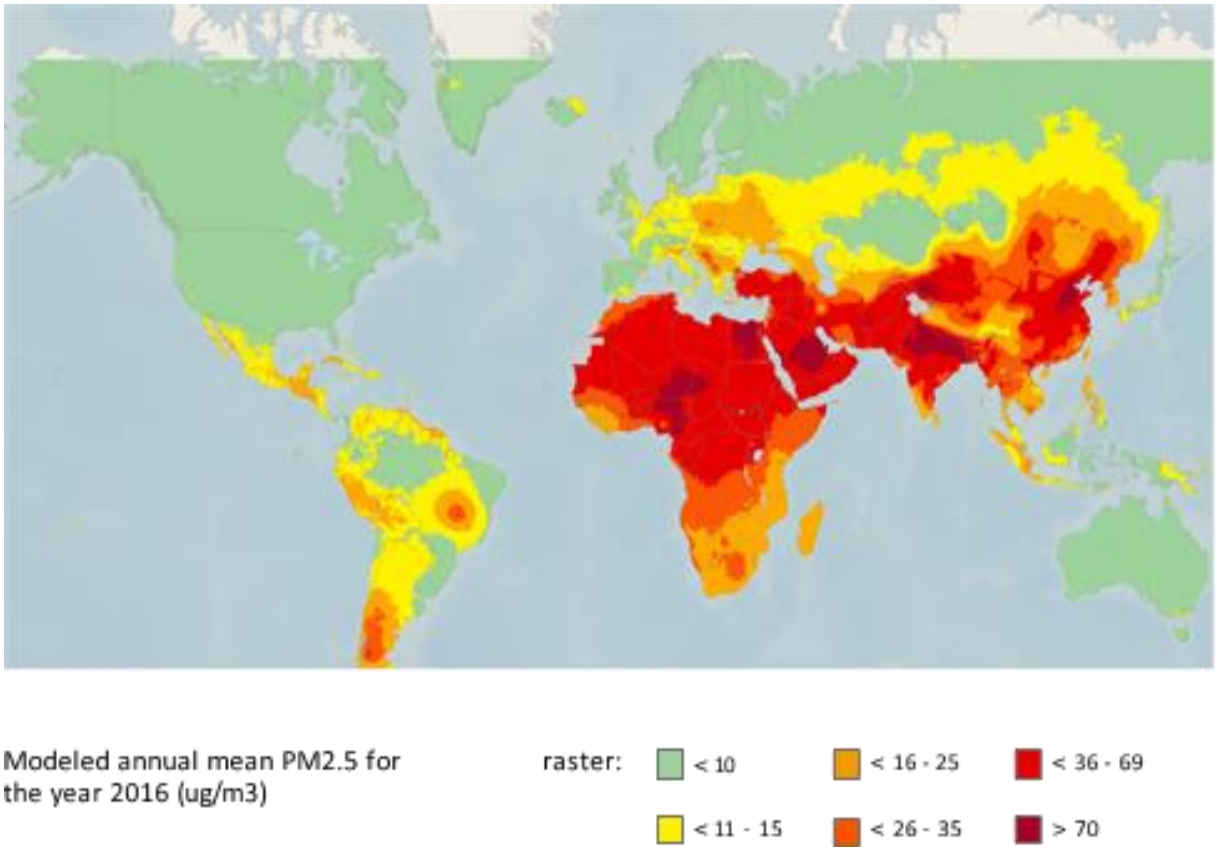

Figure 2.5: Overview of annual mean of $PM_{2.5}$ modeled for the year 2016 (Source: WHO, 2019)[85]

2.7 Air pollution in South Africa

Air pollution in South Africa is not so different to what is being experienced in other developing countries. Both direct and indirect effects of air pollution are experienced in the entire country most especially in areas of high industrial activities such as the South Durban Industrial Basin and the Vaal Triangle and where there is high dependency on non-electric sources of energy[86]. Indoor air pollution is also problematic in South African rural areas where accessibility to forms of clean energy sources are less or inadequate[87].

Air quality management and control in South Africa is complicated due to inadequate air pollution statistics and information[72]. The difference between indoor and outdoor pollution and the source contribution to each is necessary for effective monitoring of air quality in South Africa[66]. South African air quality standards are lenient and has been regarded as fairly good because of the low levels of SO_2, ozone and nitric oxide, and these levels fall within the South African air quality standard[88]. However, there are exceptions for some areas due to high industrial development such as the Vaal Triangle[89-90], the South Durban Industrial Basin[91] and Cape Town[92], where exceedances of the recommended WHO guidelines and the South African National guidelines sometimes occur[91-93].

Numerous studies in South Africa have reported that people who are exposed to air pollution come down with health problems such as respiratory conditions (e.g. wheeze, shortness of breath, blocked and running nose, hay fever, coughs, pneumonia)[94-96] and high blood lead levels in children.[97]

2.7.1 South Africa policy and laws on air pollution

The problem of urban industrial pollution and air quality has characterised the state of air quality in South Africa from 1960 to the late 1990s. Since then, the evolution of democratic rule in South Africa, policy and legislative changes brought about the sweeping and incorporation of best practices and international norms into the approach to environmental management. Locations where high concentration of industrial activities exist in South Africa has been identified as "hotspots" of air pollution, and a few epidemiological and anecdotal evidence has supported this identification of hotspots[98-99].

The "hotspots" identified include the Vaal Triangle and South Durban[100-101]. There exists a problem of developing control for pollution due to the various sources of pollution. The Atmospheric Pollution Prevention Act (APPA), has been found to be inadequate to tackle the problem of air pollution effectively and is generally thought as having caused the developments of the "hotspots"[98,102] The necessity to update air pollution control to allow the principles contained in broader policy, such as the South African Constitution was due to the fundamental policy changes. APPA in South Africa was passed in 1965 to address primarily industrial sources with limited influence over noise, dust and vehicle emission control.

Emission standards, as part of the "best practicable means" approach to air pollution were employed with little success[103]. Several criticisms have been levelled at APPA which includes

the out-dated approach and lack of proactive management of air pollution to secure desirable environmental quality[102]. Other shortcomings of APPA, such as the "best practicable means" strategy, was found to be biased towards industry, the penalty system was criticised as inadequate as a deterrent to polluting behaviour[103] and the use of guideline values that cannot be enforced is a major problem.

2.7.2 National Environmental Management*: Air Quality Act*

The change in legislation governing air pollution in South Africa in the past 12 years has resulted in the phasing out of the Atmospheric Pollution Prevention Act (Act No. 45 of 1965) and enacting the revised National Environmental Management: Air Quality Act (Act No.39 of 2004)[101]. The National Environmental Management: Air Quality Act (AQA) was promulgated in 2005 as the updated measures to air pollution control by introducing air quality management (AQM) as the control strategy[99]. One of the measures missing in APPA but included in the AQA was the ambient air quality standards for the criteria air pollutants as well as emission standards that are focused on regulating emissions from industries. In addition, the local government has been delegated with responsibility to implement the majority of the control measures in the AQA, which includes the completion of an Air Quality Management Plan (AQMP) and the mid-term and long-term review of the AQMP. The United Kingdom, United States and New Zealand are some of the countries that have adopted this best practicable means approach to the use of ambient air quality standards in AQM[104].

The National Environmental Management Act, (NEMA) (1998), provides principles for environmental management which also includes the framework and mechanism for implementation. The new legislation under NEMA supports sustainable development through principles such as waste avoidance and minimisation, pollution prevention, integration including implementation of the best practicable environmental option, and environmental justice[100]. Some of the key features of the new legislation include;

- Decentralising air quality management responsibilities
- Requiring significant emission sources to be identified, quantified, and addressed
- Setting ambient air quality targets as goals for driving emission reductions
- Stipulating air quality management planning by authorities, and emission reduction and management planning by sources
- Providing access to information and public consultation.

The South African National Atmospheric Emissions Inventory System (NAEIS) was established in 2015, with the sole objective to provide all stakeholders with relevant, up to date and accurate information on South Africa's emissions profile for informed decision making[105]. NAEIS offers a new innovative approach to reports emission as it is required by the AQA.

2.7.3 Air quality management plan

The AQA of 2005 brought about the update of air pollution control and a shift to the AQM strategy. The ambient air quality standards gave an indication of the environmental quality that is to be achieved, which was lacking under the APPA[89]. To achieve this environmental quality under the AQM, there are tools that have to be in place, such as ambient monitoring, emission inventories, dispersion modelling and emission reduction measures[106-108] The tools are part of what makes up the air quality management plan which explains the process of implementation and management activities in a specified timeline.

The AQA summarises the AQMP requirements and details on the structure and contents which is prepared by the Department of Environment, Forestry and Fisheries[109]. Impact identification of poor air quality means of addressing various pollutant sources and international obligations, consideration of best practice in AQM and the overall objective of air quality improvements are some of the content of the AQMP[110].

The main idea behind the AQMP is to assists government departments in planning AQA implementation, control measures, and financial provision. The development and implementation of an AQMP is a dynamic process involving the following six steps[110];

1. Establish stakeholder groups and the baseline air quality i.e. goal setting legislation
2. Undertake a gap and problem analysis i.e. baseline air quality assessment
3. Develop air quality vision and goals i.e. air quality management system (AQMS)
4. Develop an implementation plan (intervention strategies and rules for implementation)
5. Action plans implementation
6. Monitoring reporting and evaluation

This PhD project will inform the City of Tshwane AQMP report[111] about step 2 of the AQMP, which was published in 2005 and needs to be reviewed every 5 years according to the AQA of

2005. This will be achieved by making available the findings of this study to the City of Tshwane, via the Department of Environment, Forestry and Fisheries.

2.7.4 National ambient air quality standards

Authorisation was given to the National Ambient Air Quality Standards (NAAQS) by the NEMA air quality act to monitor the pollutant level in the atmosphere to protect the citizens of South Africa. This was achieved by establishing and enforcing some standards for some of the pollutants, which include PM_{10}, $PM_{2.5}$ (since 2012), NO_2, SO_2, CO, O_3, lead and benzene. The standards relate to specific chemical elements or compounds except for those for suspended particles. The summary of WHO guidelines and South African standards for these pollutants are shown in Table 2.3 and 2.4.

Table 2.3: World Health Organization air quality guidelines of 2005[112]

Pollutant	Averaging Period	Concentration
PM_{10}	24 h	50 µg/m^3
	1 Year	20 µg/m^3
$PM_{2.5}$	24 h	25 µg/m^3
	1 Year	10 µg/m^3
NO_2	1 h	200 µg/m^3
	1 Year	40 µg/m^3
SO_2	10 Minutes	500 µg/m^3
	1 hr	Not applicable
	24 h	20 µg/m^3
	1 Year	Not applicable
Ground-level O_3	8 h	0.075 ppm
CO	1 h	30 mg/m^3
	8 h	10 mg/m^3
Benzene	1 year	No safe level of exposure can be recommended
Lead (Pb)	1 year	0.5 µg/m^3

Table 2.4: South African National Ambient Air Quality Standards[113]

Pollutant	Averaging Period	Concentration	Frequency of Exceedences	Compliance Date
PM_{10}	24 h	75 µg/m^3	4	1 January 2015
	1 Year	40 µg/m^3	0	1 January 2015
The reference method for the determination of the PM_{10} fraction of suspended particulate matter shall be EN 12341				
$PM_{2.5}$ (since 2012)	24 h	40 µg/m^3	4	1 January 2016 - 31 December 2029
	1 Year	20 µg/m^3	0	1 January 2016 - 31 December 2029
	1 Year	15 µg/m^3	0	1 January 2030
The reference method for the determination of the $PM_{2.5}$ fraction of suspended particulate matter shall be EN 14907				
NO_2	1 h	200 µg/m^3 (106 ppb)	88	Immediate
	1 Year	40 µg/m^3 (21 ppb)	0	Immediate
The reference method for the analysis of NO_2 shall be ISO 7996				
SO_2	10 Minutes	500 µg/m^3 (191 ppb)	526	Immediate
	1 h	350 µg/m^3 (134 ppb)	88	Immediate
	24 h	125 µg/m^3 (48 ppb)	4	Immediate
	1 Year	50 µg/m^3 (19 ppb)	0	Immediate
The reference method for the analysis of SO_2 shall be ISO 6767				
Ground-level O_3	8 h (Running)	120 µg/m^3 (61 ppb)	11	Immediate
The reference method for the analysis of ground-level O_3 shall be UV photometric as described in SANS 13964				
CO	1 h	30 mg/m^3 (26 ppb)	88	Immediate
	8 h	10 mg/m^3 (8.7 ppb)	11	Immediate
The reference method for the analysis of CO shall be ISO 4224				
Lead	1 year	0.5 µg/m^3	0	Immediate
The reference method for the analysis of lead shall be ISO 9855				
Benzene	1 year	5 µg/m^3 (1.6 ppb)	0	1 January 2015

2.8 Source apportionment of particulate matter

Source apportionment are local studies determining the contributing sources to ambient PM measured at representative monitoring sites[114]. This can be achieved by using different methods: (1) emission inventories, (2) source-oriented models and (3) receptor-oriented models (RMs). RMs are commonly used to apportion PM levels based on chemical composition, e.g. major ions, carbonaceous fractions, trace elements and organic markers. From the physiochemical analysis of PM components, it is possible to estimate the pollution sources attributed to the total PM_{10} and $PM_{2.5}$ mass, along with associated uncertainties[115].

The atmospheric PM consists of particles that have their origin from several sources and processes, which includes direct emissions from mobile sources, space heating, metal processing, refuse incineration, erosion and re-suspension, gas-to-particle conversion and long-range transport[116]. Source apportionment of PM is the quantitative estimation of the contributions from different source categories to the concentrations of the measured PM in the atmosphere, based on chemical and physical characteristics of the PM and temporal co-variation of PM components. Source apportionment involves the process of identification of aerosols emission sources and quantification of the contribution of these sources to the aerosol mass and composition. The possibility of revised or new regulation as a control strategy can be followed after the characterisation of the source's emission rate and emission inventory. Different methods have been identified for source apportionment but in the last few years, receptor models have been the most widely used for source apportionment[117].

The chemical composition of the PM measured at a particular location "receptor" is used in the source apportionment models to resolve the main sources of PM at that site[118]. Source apportionment methods include chemical mass balance methods (CMB), mass reconstruction, mass closure and several types of multivariate methods such as multiple linear regression (MLR), factor analysis (FA), principal component analysis (PCA), target transformation factor analysis (TTFA), PMF, and combinations of the above. PMF is a new development in the class of data analysis techniques called factor analysis[119], in which the underlying principle is to resolve the identities and contributions of components in an unknown mixture.

The identification of pollution sources is part of the many tasks concerning air quality legislation. RMs have been used to accomplish source apportionment by analyzing the chemical and physical parameters measured at one or more specific sites (receptors). RMs are most commonly used to apportion PM based on chemical composition. Major ions (e.g. nitrates and sulphates) carbonaceous fraction (organic and elemental carbon), trace elements and organic markers, VOCs, PAHs, inorganic gases and aerosols size distribution have been used for source apportionment with RMs. The application of wind speed and direction or backward trajectories are included in the analysis, RMs are suitable to study medium to long-range transport[120-121].

PMF is a multivariate factor analysis method that utilises two matrices: source profiles and source contributions[122]. $PM_{2.5}$ composition is used as input in the PMF model in order to determine which sources contributed to the ambient samples[122]. PMF is also based on the

principle that a relationship between sources and receptor exists when mass conservation can be assumed. 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 of measured data and standard deviations are used with the benefit of introducing the limitation of non-negativity[121]

The advantage of PMF analysis is the ability to identify factors without strong priors or knowledge that is required for methods such as CMB[123].The application of PMF depends on the estimated uncertainties for each of the data values. The uncertainty estimates provide a useful tool to decrease the weight of the missing data and values below the detection limit data, in the solution. Values below the detection limit will be replaced by half of the detection limit value and their overall uncertainties were set at 5/6 of the detection limit values.

In South Africa, a study conducted by Van Zyl *et al*[124] at the western Bushveld Igneous complex of North west in 2013 used PCA to identify four emission sources of PM. These sources included crustal, pyrometallurgical, base metal and windblown dust. This study reportedly is the most comprehensive atmospheric trace metal concentration study conducted in South Africa and published in the peer-reviewed public domain. Source identification of $PM_{2.5}$ and PM_{10} has been conducted by several authors in South Africa using different methods, such as PCA, PMF and CMB. In South Africa, the application of these models has been used in Kruger National Park using the multivariate analysis[125], Soweto using the PCA[126] for apportionment of aerosols (particles of aerodynamic diameter<15µm), coal smoke and road dust, and at different sites such as the Vaal Triangle, Amersfoort, Skukuza and Louis Trichardt[127].

RMs and emission inventories have been used for source apportionment for PM_{10}, SO_2 and NO_x in the Vaal Triangle[90]**,** mass concentration have been applied for apportionment of PM (PM_1, $PM_{2.5}$ and PM_{10}) in Rustenburg[128-132] and lastly, a study in Limpopo[133] used the PMF model for source identification of PM. The sources identified in this study included industrial coal, vehicular emission of air pollutants, agriculture/wood burning, Ferrochrome smelter and road dust.

The recent review by Mathuthu *et al*.[134] identified some published peer-reviewed journal article work on source apportionment in South Africa. The authors categorised the sources of PM contributing to pollution levels in South Africa as mining, vehicular emissions[135-136], biomass

burning[129,136-137], coal burning[115] and dust forms. The source categories vary depending on the sites where the study was conducted in South Africa. Pretoria as an example of an urban and industrial area, the likely sources of pollutants will be influenced by anthropogenic and industrial activities as well as mineral dust. Studies have shown that Cape Town's major source of pollution is vehicle emission contributing about 65% of brown haze from diesel[136]**.** Traffic emissions are associated with particulate air pollution in megacities such as Pretoria. The contributions of different sources to ambient particulate matter ($PM_{2.5}$) for example biomass burning have been reported to exhibit seasonal variations[60,137-139]. The dry season (August - October) is associated with increase fire activities which contributes significantly to the concentration of particulate air[128,139-141]. Mine tailings and waste dumps from gold mining are reported to be prevalent in Gauteng, while emissions are dependent on wind speed. Petroleum refineries are also a major source in Gauteng, the High veld air pollution priority area (HPA) and Durban, with some presence in Cape Town. It is also known that coal-fired plants are located in the three priority areas. Overall, Gauteng and HPA have the highest density of anthropogenic emissions from all sources including road emissions.[63] This study will contribute to the knowledge gap in PM source apportionment by using PMF to apportion sources in this region

Source apportionment studies are important to identify and understand the sources of pollution and the implementation of health risk assessment. The source apportionment studies also help in the designing of effective emission control strategies to abate particulate air pollution Many trace metal studies performed in South Africa have been carried out by local authorities or certain industries and are not available in the public domain. Also, it is noteworthy to mention that few source apportionment studies done in South Africa are published mostly in "grey" literature.

2.9 Hybrid Single-Particle Lagrangian Integrated Trajectory model

Hybrid Single-Particle Lagrangian Integrated Trajectory model (HYSPLIT) is a comprehensive system for computing simple air parcel trajectories, complex transport, dispersion, chemical transformation, and deposition simulations.

HYSPLIT is a popularly and widely used atmospheric transport and dispersion models by the atmospheric science community[142-143]. Back-trajectory analysis is one of the popularly used model applications to determine the origin of air masses and establish source–receptor

relationships[144]. Also, HYSPLIT has gain a lot of importance due to its applications in series of simulations describing the atmospheric transport, dispersion, and deposition of pollutants and harmful materials. The application of HYSPLIT has been described by various authors and it includes tracking and forecasting the release of radioactive material[145-147], wildfire smoke[148], wind-blown dust[149-150], pollutants from several stationary and mobile emission sources[151], allergens[152], and volcanic ash[153].

The model calculation method is a fusion between the Lagrangian approach, which involves the use of a moving frame of reference for the advection, and diffusion calculations as the trajectories or air parcels move from their original location, and the Eulerian methodology, which involves the use of a fixed three-dimensional grid as a frame of reference in order to compute pollutant air concentrations. The Back-trajectory calculations feature contained in HYSPLIT makes this model very attractive and useful, which is why it has been extensively used in many studies[142-143].

Even though trajectories offer a simple assessment of source–receptor relationships, a single trajectory may not sufficiently depict the turbulent mixing processes that air parcels experience during transport. Nevertheless, the combination of back-trajectory calculation with Lagrangian dispersion component can give a more practical picture of the link between the concentrations at the receptor and the sources impacting it[154-155]. A wide selection of meteorological model data sets can be used in HYSPLIT calculations, extending from mesoscale to global scales. The calculation of forward and backward trajectories helps determine airflow patterns for easy interpretation of transport of pollutants over diverse spatial and temporal ranges.

Personal computers, Mac, or Linux platforms using a single processor allows smooth running of the HYSPLIT model. Multiple processor parallelised environment calculations using a message passing interface (MPI) implementation can be used on both Mac and Linux. Also, Real-Time Environmental Applications and Display System (READY) allows public access to meteorological data and allows the running of HYSPLIT trajectory and dispersion simulations[156.]

Commonly, trajectories are used to trail the air mass history or to predict air mass movement, and to account for the uncertainty in the associated wind patterns. Merging trajectories that exhibit some commonalities in space and time makes easy their analysis and interpretation and decrease the uncertainty in the determination of the atmospheric transport pathways[144].

2.10 Types of epidemiological studies

There are three "classic" epidemiology study designs, namely the cohort, case-control and cross-sectional study designs.[65] The cohort design is considered the ideal design to establish a causal relationship between a risk factor (e.g. air pollution exposures) and health outcomes. However, very few researchers worldwide and even fewer in Africa have access to already existing cohort study data. There is also no funding to start a cohort epidemiological study and follow participants 10 to 30 years. Another option is to consider a case-control study design, however, to recall bias is a major limitation in this design (i.e. study participants may not remember all their air pollution exposures and lifestyle risk factors). Another option is to apply the cross-sectional study design; this design however has many limitations, the biggest being that it is not feasible for rare chronic health outcomes (e.g. cancer) unless the sample size is on the scale of a national census. Information on exposure and disease status is also collected at the same time, hence it is not sure whether exposure took place before the health outcome.

Time-series studies measure the effects of short-term changes in air pollution on acute health effects by assessing associations between day-to-day variations in both air pollution and in mortality and morbidity counts. Daily measures of the number of health events (e.g., daily mortality count), concentrations of PM and other pollutants (e.g., 24 h average PM_{10}), and weather variables (e.g., daily temperature) for a given area are the data required for the time series analysis[160].

The case-crossover epidemiology study design is used to investigate the effect of short-term exposures on the risk of onset acute events. The application of this study has been used in different fields of epidemiology which includes injury, drug adverse events, air pollution etc. the situation where each case serve as its own control remains the main feature of this study design, i.e. each case has a matched control[157,160] (more information in chapter 6)

2.11 Dearth of epidemiological studies on air pollution in South Africa

Despite formulating progressive air quality legislation in South Africa, the breath of possible health impacts associated with air quality has not been comprehensively researched in local epidemiological studies and this creates a major research gap[158-160]. Many of the

epidemiological studies were conducted in North America and Western Europe, with more from Asia and South America since 2004.

According to Coker and Kizito[161], there is a dearth of ambient air pollution epidemiological studies in sub-Saharan Africa, but most of the studies are focused on indoor air pollution. Therefore, most of the literature regarding health effects of ambient air pollutions are from North America and Western Europe. In Coker and Kizito's[161] review, and according to their criteria for inclusion, 12 studies were identified and three-quarters (25%) of these studies were carried out in South Africa. Lastly, most epidemiological studies in South Africa used cross-sectional design while, only few used case-crossover designs

In a prospective study by Mentz *et al*[162], in Durban South Africa, school-age children were recruited to investigate the effect of ambient air pollutant on acute respiratory outcomes. This study measured PM_{10}, SO_2, and CO at the participant's school, while NO_2, O_3 and NO were obtained from the government ambient air pollution monitoring sites. Acute symptoms observed include cough, wheezing, shortness of breath, chest tightness or heaviness. There was significant association with each of the air pollutant, with all but one resulting in an adverse effect.

The interaction between daily pollutants levels (PM_{10}, SO_2, NO_2, and NO) and CD14 cell genetic polymorphisms within-day changes in lung function among children (7-9 years of age) was carried out as a prospective cohort study in Durban, South Africa by Makamure *et al*[163]. Continuous measurement of the ambient air pollutants was conducted in the school except for some cases where it was done out of the school ground. No significant association was observed between the pollutants and lung function alone when stratified by CD14 polymorphism status. Prospective study design and consideration of individual level susceptibility using a biologically plausible polymorphism in a gene related to cellular immunity and asthmatic symptoms were the most important contribution of the study.

Naidoo *et al*[164] in their study in Durban, South Africa tested single pollutant associations between ambient air pollutants (PM_{10}, SO_2, NO_2) and several chronic lower and upper respiratory outcomes using cross-sectional design. School-aged children were recruited for the study; PM_{10} and SO_2 were sampled at the participants' school and NO_2, was monitored by the Government at the eight sites. The study observed no association between air pollutants and study outcomes except for SO_2 which was significantly associated with increased odds of

airway hyper reactivity. This study is the only study that investigated biologic measures of airway hyper reactivity.

The case-crossover study conducted in South Africa by Wichmann *et al*[165] considered a year-round and seasonal association between daily NO_2, SO_2 and PM_{10} with respiratory disease (RD) mortality, cardiovascular disease (CVD) mortality, and cerebrovascular disease (CBD) mortality between 2001 and 2006. Every hour daily pollutants data was obtained from government air monitoring sites while the daily mortality data were acquired from City of Cape Town mortality records. The findings of the study revealed that statistically significant positive year-round association for CVD and CBD mortality with NO_2, PM_{10}, and SO_2. For RD mortality, there was a significant positive association with NO_2 and PM_{10} exposures during the warmer periods of the year.

Thabethe[166] reported in her MSc dissertation, excess mortality risks of 0.4% (-0.4%; 1.1%) and 1.0% (0.3%; 1.7%) for RD and CVD mortality, respectively following a 10 $\mu g/m^3$ increase in PM_{10} using case-crossover study design. These results were the combined estimates of Cape Town, Durban and Johannesburg in the study period 2006-2010, after a meta-analysis was conducted.

The study by Shirinde *et al*.[167] investigated the association of wheeze with air pollution among children using cross-sectional study in Ekurhuleni Metropolitan Municipality (Tembisa and Kempton park) of Gauteng province South Africa. The study revealed that children living in one of the air pollution priority areas had an increased risk of wheezing due to exposure to air pollution sources.

Nkosi[168] reported in his thesis that children residing in exposed communities had an increased likelihood of current wheeze OR 1.38 (95% CI: 1.10 1.71), rhinoconjunctivitis OR 1.54 (95% CI: 1.29 1.82). Also, exposed elderly persons had a significantly higher prevalence of chronic respiratory symptoms and diseases than those who were unexposed using short-term longitudinal study design. Findings from this study also indicated that living close to mine dumps was significantly associated with asthma (OR = 1.57; 95% CI: 1.20 2.05), chronic bronchitis (OR = 1.74; 95 CI: 1.25 2.39), emphysema, pneumonia and wheeze (OR = 2.01; 95% CI: 1.73 2.54). He concluded that community proximity to mine dumps is associated with increased respiratory symptoms and diseases among the children and the elderly in south Africa.

The finding of the study on the respiratory status of 10 year old children in the Vaal Triangle by Mundackal[169] using a cross-sectional inter-comparative study reported that the 6-month prevalence of sinusitis, bronchitis, and pneumonia in 2010 was lower when compared to the yearly prevalence of these illnesses in 1990 the study, while the use of gas heater acted as a risk factor for pneumonia (lower respiratory illness) while environmental tobacco smoke within the household was protective of hay fever and sinusitis (upper respiratory illness).

The cross-sectional study by Swiegers[170] reported that the prevalence of chronic obstructive pulmonary disease (COPD) and small airways diseases was small but significantly higher in exposed mine workers. Also, the risk of developing COPD was 2.7 times higher for exposed workers. The risk for non-smokers was small but significant.

The baseline assessment of child respiratory health in the Highveld Priority Area by Albers[171] using a cross-sectional study reported that air quality in this area was of great concern. The health outcome with the highest prevalence was hay fever accounting for 32%. Also, the use of non-electric heating sources, parental smoking, and mould in the house were risk factors of most of the respiratory health problems.

A similar study by Maluleke and Worku[172] to ascertain the key predictors of asthma among children living in Polokwane concluded that ambient air pollution was an important risk factor for asthma in children.

Studies by various authors have also reported that the inhalation of particulate matter may result in immune-suppression, nausea, cardiovascular infection, lung cancer and asthma, and may even result in premature death[36,68,173]. Noteworthy to mention that most of the epidemiological studies were conducted in North America and Western Europe while others are from Asia and South America since 2004. Only few studies have been conducted in Africa as a whole, thereby constituting a research gap in establishing the association between air pollution and various health outcomes. while the study will contribute to the already available information

2.12 Short term effect of air pollution on hospital admission

There is increasing evidence that mortality and/or hospital admission for air pollution for some health outcomes such as cardiovascular and respiratory disease are due to the duration in which an individual is being exposed to short-term fluctuations and long-term levels of air pollution.

Numerous studies concluded in Europe, USA etc. mostly reported the association between air pollution levels and human health in terms of hospital admission[174-175]. Several published meta-analysis studies[176-180] have also found that ambient air pollutants in the atmosphere are associated with higher incidence, prevalence, hospitalisation or worsening of symptoms of asthma, heart failure[181-182] hypertension[183], and pneumonia[184]. Air pollution has been found to be positively associated with hospital admission for cardiovascular[185], respiratory disease[186] and gastrointestinal disease[187].

Outdoor air pollution has shown an increased association with acute lower respiratory infection (ALRI) mortality and with increased symptoms, admissions to hospital and emergency respiratory (ER) visits. Studies have reported that mortality and hospital admission from pneumonia have resulted from short-term exposure to lower concentration of outdoor air pollution in developed countries[188-189]. ALRI has been reported to account for nearly one-fifth of the mortality recorded among young children under 5 years worldwide. Records show that 90% of ALRI deaths have been attributed to pneumonia, the burden of this problem being felt mostly in Asia and Africa and is responsible for 134 and 131 million cases of lower respiratory infections ravaging Asia and Africa respectively out of the 429.2 million cases for all ages. The three-year trend analysis for selected main groups of underlying causes of deaths between 2014 to 2016 as reported by statistics South Africa is shown in Figure 2.6

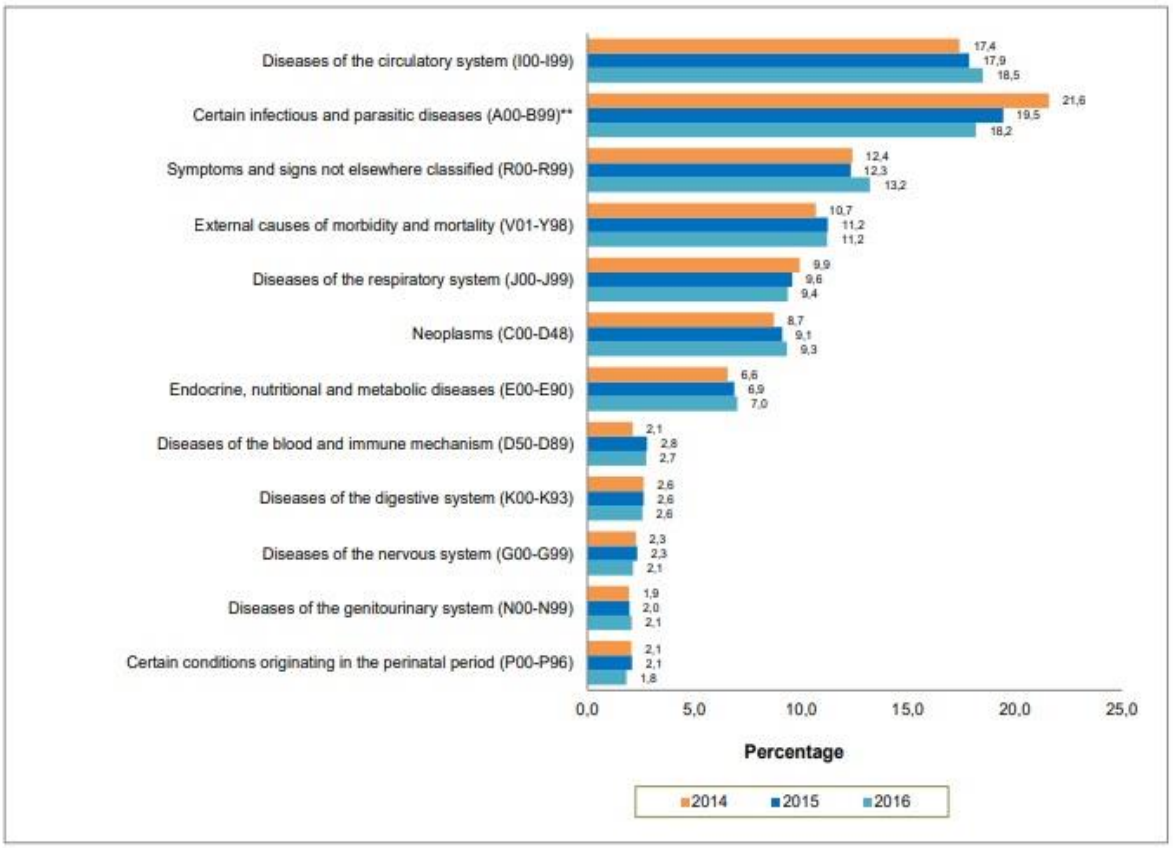

Figure 2.6: Percentage distribution of deaths by selected main groups of causes of death, 2014-2016 (source Statistics SA, 2019[190]).

In addition to ambient air pollutants, temperature has been linked to an increase in the risk of hospital admission for a variety of causes among children[191-193], high temperatures in summer and low temperatures in winter and large diurnal temperature ranges may have link to the cause of number of children admitted to hospital for respiratory diseases[194-196]. However, there are few studies that have investigated the effect of ambient temperature in children and fewer studies have been conducted in developing countries[162-163].

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CHAPTER THREE: RESEARCH METHODOLOGY

3.1 Exposure assessment in Pretoria

3.1.1 Study location/site

The study site was located at the School of Health Systems and Public Health (SHSPH), University of Pretoria. Samples were collected on the roof of HW (6th floor) Snyman South Building, Prinshof Campus (Figure 3.1). The sampling site (coordinates: -25.7S; 28.2E) was located close to the central business district (Pretoria CBD). This was an urban background site i.e. a "cleaner" option than industrial site. The SHSPH is approximately 1 km to 2 km away from a major road (Steve Biko/Gezina Road) and about 5 -10 km away from the Pretoria CBD. The choice of this location was due to the safety of the GilAir 5 pump and constant supply of electricity. Urban background sites are commonly used to depict the typical exposure to the population without a dominating influence of nearby point sources.

(a)

(b)

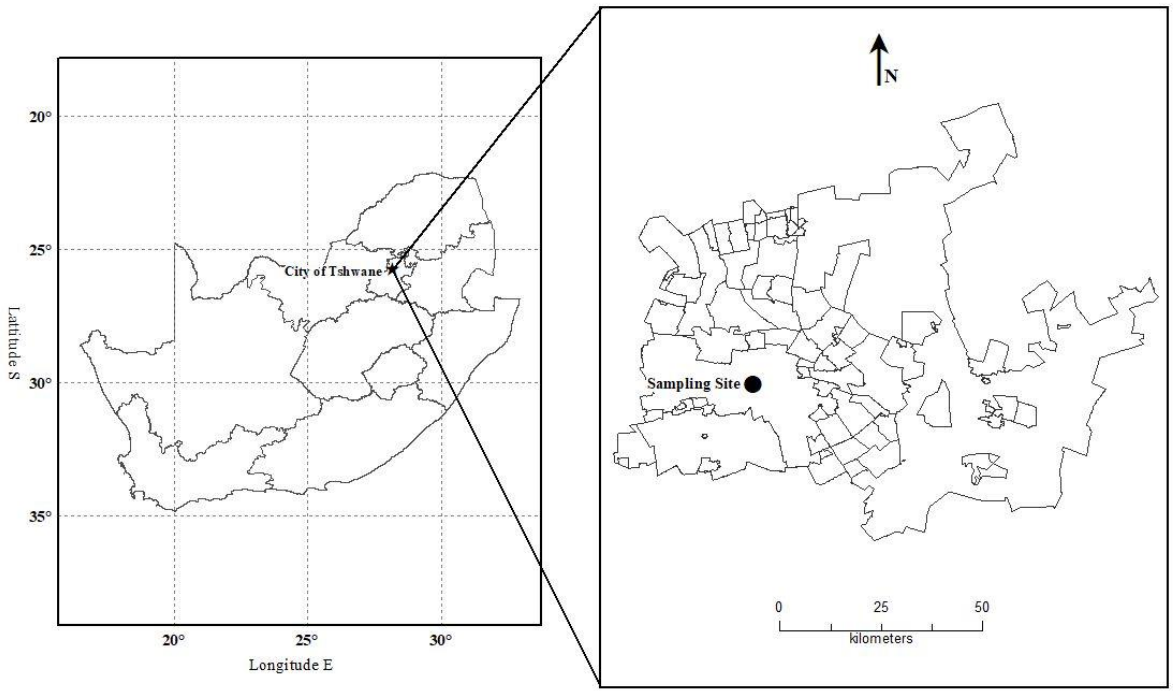


Figure 3.1: (a) Google Earth image and (b) sketch map depicting the sampling location

3.1.2 Sampling procedure and equipment

$PM_{2.5}$ filter samples were collected over 24 h and every third day. The sampling period was between 18 April 2017 and 17 April 2018. $PM_{2.5}$ samples were collected on 2.0 µm pore sized PFTE, PTFE supports (Zefluor), 37 mm membrane filters (Zefon International, Inc Ocala, FL34474 USA). Two sets of equipment (two GilAir-5 personal air samplers) were used for the samples and duplicates, which comprised of a GK 2.05 (KTL) cyclone, for $PM_{2.5}$ connected to a small Gilian GilAir-5 personal sampling pumps with flow rate of 4l min^{-1}.

The sampling time was 24 hours (09:00 AM to 09:00 AM the following day, UTC + 2 hours). The reason for the choice of time was due to accessibility to the roof top. Also, the sampling set up is manually started and stopped, so starting and stopping at midnight not practical. Sampling was carried out on 122 days, during which 122 samples and 25 duplicates samples were successfully collected. Other equipment and materials used were petri dishes (diameter of at least 55mm), flat point tweezers (to insert filter in the filter holder in the laboratory), and

micro-balance for weighing filters. Field and laboratory forms were used to record data in the field, such as weighing condition, weight of the filters, and rotameter calibration (Table 3.1).

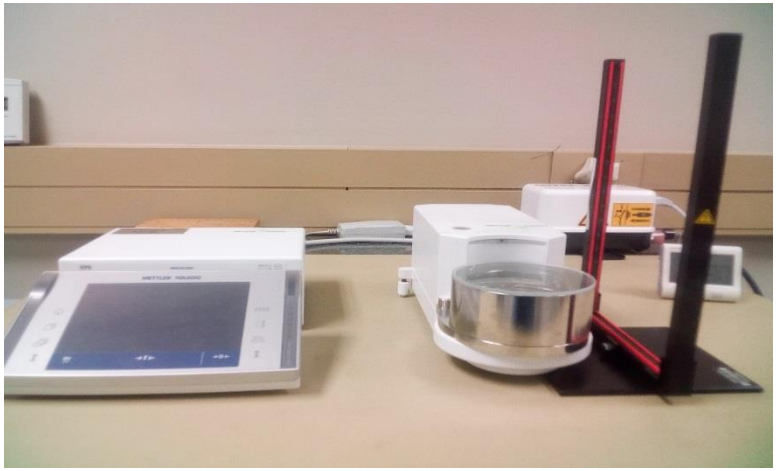
The sampling station in Figure 3.4 was adopted for $PM_{2.5}$ collection. This was to ensure that air flowed from all directions to the air sampler (i.e. no obstruction to hinder the flow of air to the sampling point). All necessary precautions were put in place to ensure the station was not blown off by strong wind and rainfall. This intervention was a success for the entire study period.

Table 3.1: Sampling equipment and materials used for $PM_{2.5}$ sampling

S/N	Item	Type	Manufacturer	Part number
1	Primary calibrator	Gilibrator-2	Sensidyne	850190-1
2	Cyclone	Aluminium	Sensidyne	GK 2.69
3	Personal air samplers	GilAir-5	Sensidyne	800883-171
4	Air sampling cassette	Polystyrene (37 mm)	Casella	P101026
5	Cassette tubing adapter	Polystyrene (6.4 mm)	Zefon International	ZA0005
6	Filters	PTFE	Zefon International	FPTPT237
7	Filter support	PTFE	Zefon International	FPTPT237
8	Tweezers	Flat point		

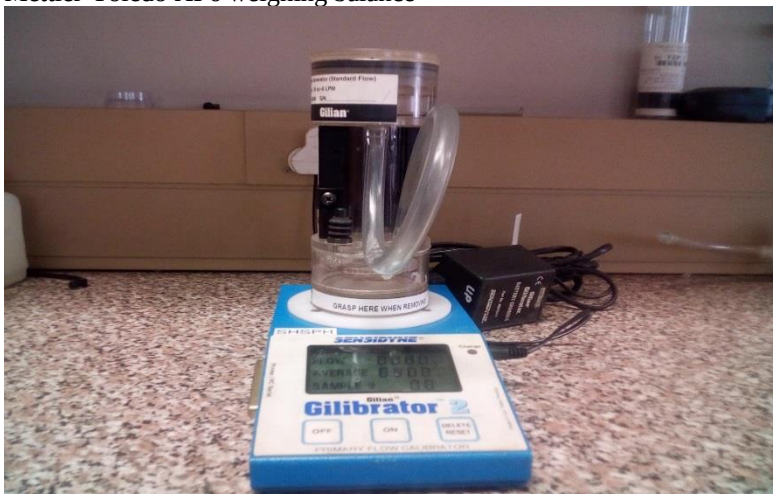
$PM_{2.5}$ levels were compared with an Aeroqual instrument (Figure 3.5) which is also located next to the sampling point on the 6th Floor of the School of Health Systems and Public Health. The Aeroqual records continuous monitoring data $PM_{2.5}$. The $PM_{2.5}$ levels from the aeroqual was compared with levels recorded via gravimetric analysis as a measure of reliability. The aeroqaul was scheduled for maintenance during my sampling campaign but was available during the next campaign which is also part of the bigger project.

Hourly air pollution data was obtained, then averaged into daily averages (9 am to 9 am, i.e. same time as the manual sampling). The Aeroqual had missing data between 19 April 2018 and 15 May 2018, thus the comparison was from 16 May 2018 to 20 April 2019.



Mettler-Toledo XP6 weighing balance

M43D smoke stain reflectometer



Sensidyne primary calibrator

Gilair5 pump

Figure 3.2: A set of sampling equipment used during the project

Model OT21 Optical Transmissometer

Sample of filter before and after use

Hobo data logger (for temperature and relative humidity)

Cyclone

Figure 3.3: Additional set of sampling equipment and materials used during the sampling period.

Figure 3.4: Sampling station on top of School of Health Systems and Public and Health building showing the sampling set up.

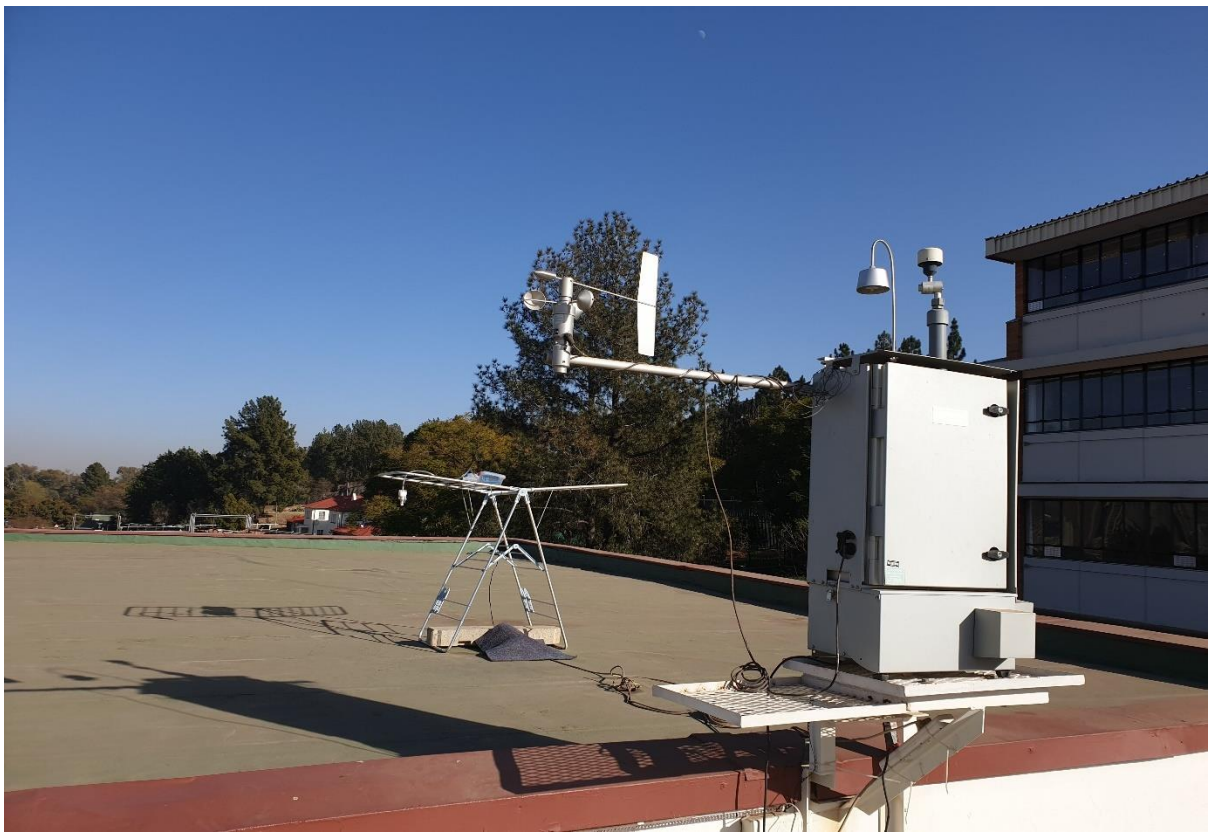


Figure 3.5: Image of Areoqual next to GilAir pump measurements on roof top of School of Health Systems and Public and Health building

3.2 Chemical analyses of $PM_{2.5}$ filters

3.2.1 Gravimetric analysis

Gravimetric analysis was carried out at the Air Quality Laboratory, SHSPH, as it has a state-of-the-art microbalance. The Teflon filters were conditioned or equilibrated at a constant relative humidity and temperature for 48 h before weighing in the Air Quality Laboratory, SHSPH. The temperature and relative humidity were maintained at 21 ± 0.5°C and 50 ± 5%, respectively in the weighing room (i.e. a temperature and relative humidity-controlled weighing and filter storage room). For the gravimetric analysis, a 1 µg sensitivity microbalance (Mettler Toledo, XP6) was used for the weighing process. Tweezers were used to handle the filters, which were stored in the labelled petri-dishes.

An initial calibration of the balance was performed to ensure that the weighing balance was functioning properly. Temperature and relative humidity of the weighing room was documented. The Teflon filters were weighed before and after sampling in order to determine the mass of the collected material. The weighing followed a standard operating procedure, (SOP) where three field blanks were used for a batch of 20 filters. The SOP used for the weighing procedure was a modified version of the SOP used in the ULTRA study.[1] Before weighing, the filters were deionised on both sides using an alpha radiation source (Po-210) in order to remove static charge. Filters were pre-weighed a maximum of 2 months before being used for sampling and post-weighed up to 2 months after sampling (i.e. in batches of 20). (Appendix 7).

The SOP states that each of the filters must be weighed twice so that if the result differs by more than 2 µg, weighing of a new pair of filters must be carried out. This was continued until the requirement was met. Additionally, the average field blank mass increase (or decrease) was subtracted from each sampled filter mass in each of the batches. The limit of detection (LoD) was evaluated by weighing batches of blank filters according ISO/CD 15767.[2] Pre- and post-weighed filters were stored in a fridge at 4°C to limit weight losses due to volatilisation of ammonium nitrate from Teflon filters which have been documented to occur in one week. (Appendix 7)

Collected particle mass was calculated as the difference in weight of the filter before and after sampling. The average filed blank is subtracted from the mass of the PM collected of the filter:

$$M = W_2 - W_1 - B$$

Where;

W_1 = adjusted filter weight before sampling (µg)

W_2 = adjusted filter weight after sampling (µg)

B = mean adjusted filter weight change of field blank filters (µg)

Filter weights (W_2 and W_1) are the average of the two weight readings of the filter during each weighing session. Filter weights was adjusted for the deviation of the control filter weights on a weighing day from the nominal value. For blank filters, the average deviation (B) of the three blank Teflon filters from the nominal value was subtracted. For exposed filters, the average deviation of the two exposed Teflon filters from the nominal value of the two exposed filters was subtracted. The nominal value is defined as the average of the previous 10 weighing sessions.

3.2.2 Soot measurements

The $PM_{2.5}$ collected on the filters were analysed for soot using a M43D EEL smoke stain reflectometer (Diffusion Systems Ltd., London UK) in batches of 20 filters. Soot measurements were also conducted in the Air Quality Lab, SHSPH. The procedure used also followed a SOP similar to the ULTRA study.[1,3] The reflectometer is an instrument that provide an estimation of the black carbon concentration in collected air particulate matter on the sampled filters by reflectance techniques. The measurement was done based on the smoke stain on the filters. A steady light was emitted onto the smoke stain by photo-electric reflectometer. The light was reflected back from the smoke stain to a photo-sensitive element. The reading was made by amplifying the electrical response. Each filter was measured for reflectance five times on different locations (Figure 3.6), according to the five-point method (i.e. in the center and in each quadrant) and the average reflectance derived from the five measurements were used in the calculations (Appendix 8).

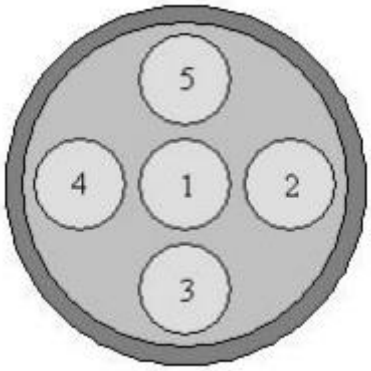


Figure 3.6: Areas (1-5) shows where the light beam should approximately strike during reflectance measurements (Source: SOP 4.0, 2002)[4]

After every 25 filters, three filters were selected and measured a second time to ensure the two measurements differed only by a maximum of 3%. Correction due filter was adjusted for by measuring the blank filter. The absorption coefficient, *a*, was used to express the reflectance according to the formula ISO 9835[5] as follows:

$$a = (A/2V) * In (Ro/Rs)$$

Where;

A is the loaded filter area (m^2).

V is the sampled air volume (m^3)

Ro is the average reflectance of field blank filters

Rs is the average reflectance of the sampled filter

The absorption coefficient (*a*) is expressed in 10^{-5} m^{-1}

3.2.3 Black carbon and organic carbon

Black carbon (BC) and organic carbon (OC) were performed using a Model OT21 Optical Transmissometer (Magee Scientific Corp., Berkeley, CA USA). The additional absorption in the UV light, at 370 nm, due to the organics indicate the presence of biomass burning.[6-8]. The transmissometer comprises three parts, namely- display unit, keyboard and tray aperture. The filter trays hold the sample and blank filters in place, permitting accurate absorbance measurements to be made. Before inserting the filter in the slide, the correct adapter size (25mm, 37mm or 47mm diameter) for the filter has to be selected. The blank reference filter has to be placed in the circular slide pocket at position 1 (outer tray position) and the sample filter into the circular slide pocket at position 2 (inner tray position), the slide by the handle has to be grasped and the slot inserted until it clicks (and beeps) into position 1, and lastly the

ENT button pressed to start the measurement cycle while the instruction on the LCD is followed to complete the analysis. The slide is further pushed to position 2 and the optical transmission of the sample filter is measured at both wavelengths. Calibration is performed each time a different type of blank filter or tray adapter size is used.

3.2.4 XRF analysis

A XEPOS 5 energy-dispersive X-ray fluorescence (EDXRF) spectrometer (Spectro analytical instruments GmbH, Germany) at the Department of Chemistry and Molecular Biology, Atmospheric Science division, University of Gothenburg, Sweden was used to analyse the elemental composition of the aerosol particles on all filters. The analysis was done under four separate operating conditions, two of them using a He atmosphere, to optimise the detection of trace quantities of elements in the analytical range. The Spectro XRF Analyzer Pro software processed and quantified the EDXRF spectra. All samples were analysed using a total time of 3000 seconds, automatically divided between the four analytical setup conditions. Calibration of the EDXRF spectrometer was performed by the manufacturer and the calibration was verified using the standard reference filter, NIST SRM2783. The concentrations of the following 18 elements were analysed for: S, Cl, Si, K, Ca, Ti, V, Fe, Ni, Cu, Zn, As, Se, Br, Sb, Ba, Pb, U. Field blanks were analysed in the same way as the samples, and concentrations detected above the limit of detection (LoD) were subtracted from the concentrations of the same elements in the samples.

The calibration was carried out daily over the period of analysis to check for the stability of the instrument using the standard reference filters. The standards were run, fitted and evaluated for element's net peak area (P_A) and associated background area (B_A). The known concentration CA and the obtained P_A and B_A were used for the calculation of detection limit (DL_A) using the IUPAC equation below[9].

$$DL_A = 3 \times C_A \times \frac{\sqrt{B_A}}{P_A}$$

To correct for positive artifacts from organic carbon (OC), measured concentration values from the field blanks were subtracted from the measured sample concentration values[10]. For filters of low concentrations, a narrower and more fine-tuned spectrum fit was obtained to improve the data recovery of the lighter elements. The mean analytical precision was 5%, as calculated from repeated analysis (N =5) of two randomly selected filters, one having a low and the other,

a high mass loading. If the elemental concentration was below the limit of detection (LoD), the LoD was divided by the square root of two which was used in the calculations.[11]

3.3 Source apportionment by positive matrix factorization

Source apportionment analysis depends on the results obtained from soot and trace element content of the filters. PMF is performed by installing the software on the computer by using soot and trace element input variables. The order of operation within the EPA PMF base model is shown in Figure 3.7. The first set of analysis to be performed on the data set includes concentration/uncertainty, concentration scatter plots, concentration time series and data exception screens. This is then followed by Base Model Runs and Base Model Results. The characterisation of particulate matter sources was achieved using the PMF technique applying the multilinear engine (ME) technique using the ME-2 programme.[12] The Environmental Protection Agency software EPA-PMF 5.0 was used.[12]

PMF is a multivariate receptor model concept that estimate the source profiles and their contributions based on a weighted least square approach.[13-14] The task of the PMF model in the equation below was to obtain the unknown matrices, G and F, by the iterative treatment of a least square method.

Considering the below equations;

$$X = GF + E$$

Where;

X = the data matrix (size m x n) consisting of n chemical components analysed in m samples

G = the source contribution to each sample (size m x p) for p factors

F = the matrix of source profile (size p x n)

E = the residual

The main mission of the iteration was to minimise the Q value, which is defined in the equation

$$Q(E) = \sum_{i=1}^{m} \sum_{j=1}^{n} \left(\frac{e_{ij}}{s_{ij}} \right)^2$$

Where: -

(e_{ij}) = squares of residual

(s_{ij}) = error estimates of data points

Q = goodness of fit parameter.

Therefore, for the PMF analysis, BC, OC and the following elements (such as S, Cl, Si, K, Ca, Ti, V, Fe, Ni, Cu, Zn, As, Se, Br, Sb, Ba, Pb, U) determined by the XRF analysis were included in the PMF analysis. The signal to noise ratio of a given specie is calculated by using the sum of the concentration values divided by the sum of the uncertainty values. Only concentration values that exceed the uncertainty contribute to the signal portion of the signal to noise ratio (S/N) calculation, this is because the concentration value is essentially equal to the sum of signal and noise, while signal is the difference between concentration and uncertainty.

To determine the S/N, two calculations are performed.

1. Concentrations below uncertainty are determined to have no signal.
2. Concentration above uncertainty

The below equation was used for calculating S/N

$$\left(\frac{S}{N}\right)_j = 1/n \sum_{i=1}^{n} d_{ij}$$

The signal to noise ratio can be categorised as follow;
Bad: S/N ratio less than 0.5
Weak: S/N ratio greater than 0.5 but less than 1
Strong: S/N ratio greater than 1

Lastly, S/N was used to select the species for further analysis. If the S/N was below 2 for a variable in the model, the variable would be deemed weak (and the uncertainty would be tripled) and excluded from further analysis. In all the analyses, the elemental mass concentrations have been recalculated to their mean oxidised mass concentrations, when applicable according to Beuck *et al.*[15] The data below the detection limit (DL) were substituted with one-half of the respective DL, and uncertainty was set to ±5/6 times the DL[16]. The input variables were classified using the S/N criteria.[17] The PMF model was run with four to seven source profiles; the results were compared and tested for stability of the solution (using bootstrap and displacement analysis) and examining the resulting source profiles for consistency with known source profiles.

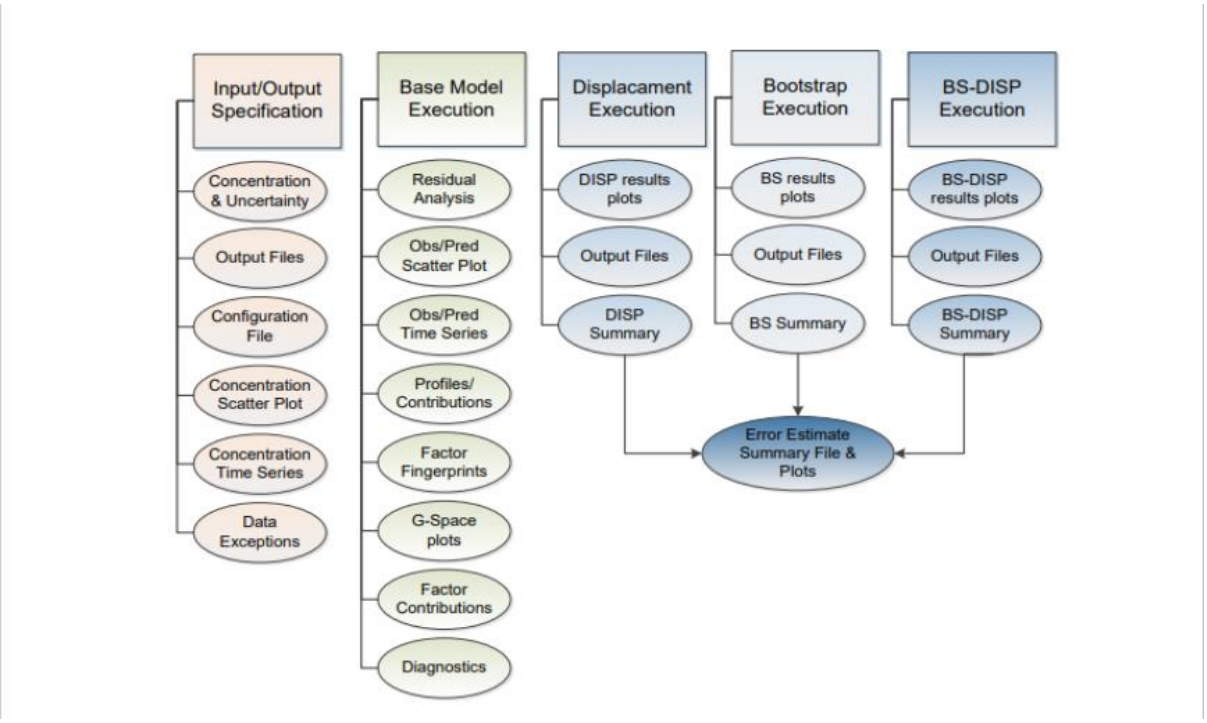


Figure 3.7: Flow chart of operations within EPA PMF – Base Model (Source: Norris et al, 2014)[18]

3.3.1 Source profiling by PMF

The number of factors that corresponds to potential PM sources remains the important step in PMF analysis. To reduce exposure to air pollution or mitigate the effect on human health and environmental quality, source apportionment is necessary to identify the sources and activities contributing to local air pollution. To evaluate air quality and determination of PM quantities in a region, it is important to identify all sources contributing pollutants to the atmosphere. Noteworthy to mention, the amount and characteristics of every source emission differ over temporal and geographic scales due to a number of influences.[19] In a review paper on source apportionment or air particulates in South Africa, by Mathuthu *et.al*[20] mining, vehicle emissions, biomass burning, coal burning, and dust storm were identified as sources of PM responsible for air pollution by various studies. Pretoria being an urban/industrial area, is likely to have source categories influenced by anthropogenic and industrial activities. Based on the known emission sources in Pretoria, the following eight sources were identified, biomass burning, vehicle exhaust systems, industrial emission, road/soil dust, waste burning, coal combustion, domestic fuel burning, mining and emission from power generation station. Source profiles were obtained from previous source apportionment studies[20]

Local speciation profiles were developed for various sources in Qalabotha (Free state)[21-22] and Soweto in Gauteng.[23] These studies also made use of the USEPA speciate database. All identified sources have chemical and elemental profile which are used in source apportionment analyses. In the PMF model, after the application of bootstrap and displacement tests on the data set, five to seven factors were deemed appropriate for a seven-source solution. The profile graph displays the mass of each species apportioned to the factor (blue bar) and the percentage of each species apportioned to the factor (red square). The factors were identified according to the type of elements dominating in percentage in that factor.

3.4 Application of HYSPLIT model

3.4.1 Transport clusters

The backward trajectories were produced for all days from 18 April 2017 (i.e. start of $PM_{2.5}$ sampling) to 17 April 2018 (i.e. end of $PM_{2.5}$ sampling) by the Hybrid Single Particle Lagrangian Integrated Trajectory (HYSPLIT) software[24-25]. The model was driven by the NCEP/NCAR (National Center for Environmental Prediction/National Center for Atmospheric Research) Global Reanalysis Meteorological Data at the web server of the National Oceanic and Atmospheric Administration Air Resources Laboratory (NOAA ARL). An analysis field (resolution 2.5° x 2.5° and 17 vertical levels) was provided every 6 h and the wind field was interpolated linearly between each analysis. The settings applied in the HYSPLIT set up for generation of backward trajectories are shown in the Table 3.2

Since a single backward trajectory has a large uncertainty and is of limited significance, an ensemble of trajectories with 500 m starting height and a fixed offset grid factor of 250 m were used in this study (i.e. 250 m and 750 m were used) as in another study[25]. The daily average trajectories were calculated backwards for 72 h and used for cluster analysis. 4380 backward trajectories were generated for the study period (i.e. 24 h/6 h = 4, 4 x 3 heights x 365 days = 4380 trajectories)

The clustering algorithm coupled in HYSPLIT was based on the distance between a trajectory endpoint and the corresponding cluster mean endpoint[26]. The cluster analysis was conducted seasonally (autumn, winter, spring and summer) due to the limitation of using very large sample sizes in the clustering function of the HYSPLIT software, as in other studies[25-27].

For the study, the seasons were categorised into the following dates: Autumn: 18 April – 31 May 2017 and 1 March – 17 April 2018; Winter (1 Jun – 31 Aug 2017); Spring (1 September – 30 November 2017) and Summer (1 December 2017 – 28 February 2018). Five transport clusters were determined according to their mean pathways in the four seasons: Autumn, Winter, Spring and Summer.The HYSPLIT method used in Molnar *et al*[25] and Wichmann *et al*[27] were applied in this study.

Daily mean temperature, relative humidity, and wind speed were calculated from the hourly data obtained from SAWS. Quality control were performed to check for missing data and days where no observations were recorded. After checking the data for completeness, and daily average calculated, 33% of daily average for wind speed and wind direction data were missing while no rainfall observations were recorded against the sampling date

Table 3.2: HYSPLIT user settings for backward trajectory cluster plots

S/N	Parameter	Setting
1	Starting date and time	17 04 18: 00
2	Location	-25.731683S; 28.200333E
3	Total run time (hrs)	-72
4	Meteorology data model	CDC-1
5	Vertical motion	Model vertical velocity
6	Level 1 height (elevation, above ground level)	250m,500m, 750m
7	Trajectory interval	6 hours
8	Plot projection	Default
9	Zoom factor	30%
10	Analysis field (resolution)	2.5^{o} x 2.5^{o}

3.5 Statistical analysis

Statistical analyses were performed with the SAS System for Windows, version 9.3.[28] Regarding the exposure assessment part of the PhD project, descriptive statistics for $PM_{2.5}$, soot, BC, OC and the trace elements were reported. Non-parametric tests were applied since the exposure levels and the PMF sources did not have normal Gaussian distributions. Kruskal-Wallis test was conducted to test whether $PM_{2.5}$ $PM_{2.5}$, soot, BC, OC and the trace elements and PMF sources differed significantly across seasons and the trajectories. Wilcoxon's rank-sum test was applied to test whether $PM_{2.5}$ levels and PMF sources differed significantly between weekdays and weekends and between the local dispersion conditions. Regarding the

epidemiology study part of the PhD project, the statistical analyses performed can be seen in section 6.2.2.4 in chapter 6.

3.6 Epidemiology study

Please refer to the methods section 6.2.2 in chapter 6

3.7 Ethical considerations

The study was approved by the Research Ethics Committee, Faculty of Health Sciences, University of Pretoria, Ethics Number 469/2017 (Appendix 1-6). This project made use of secondary hospital admissions register data and involved an exposure assessment study. No human participants were recruited or included in the entire project.

3.8 Quality assurance

Adequate training was provided on the handling and operating of the equipment needed for sampling and analysis of the $PM_{2.5}$samples collected. This was to ensure that errors were eradicated before, during and after the experiments. A record sheet was designed to ensure accurate and efficient collection and recording of all necessary data. All equipment used for the sampling and analysis was calibrated before usage in order to ensure correctness of measurements and result. Strict compliance with standard operating procedures (SOPs) was ensured during the analysis of the samples collected and their storage; data entry was checked for any errors before performing statistical analysis.

Secondary data from SAWS were checked for outliers and missing data. The quality of the data, especially the meteorological and air pollution data was not of high quality based on the missing data that was prominent. Also, outliers for NO_2 could not be explained. Several factors, such as faulty equipment might be responsible for this missing air pollution data.

Temperature, relative humidity and wind speed obtained from the South African Weather Services (SAWS) were converted into 24 h (daily) averages. At least 85% data recovery was required for both hourly and 24 h averages to represent the local meteorological conditions accurately. Data capture for primary collected $PM_{2.5}$ was 100%. Amongst the receptor models that are commonly used, PMF is able to provide reliable uncertainty analysis for modeling by bootstrapping. This method randomly replaces a few samples from the data set to create new data set and a new execution of model will be based on this new data set[29] This process is also important in modelling results for the purpose of quality assurance and quality control.

A recent study by Liu *et al*[30] reported ambient air pollution and daily mortality in 652 cities across 24 countries including South Africa. Six cities from South Africa were included in the study. Air quality data for the City of Johannesburg and City of Tshwane (including Pretoria) are from their respective municipality government monitoring stations which are coordinated by SAWS, these data were obtained and used in their analyses. This study also reported 15.3% missing data from the air quality data.

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CHAPTER FOUR: OVERVIEW OF $PM_{2.5}$ MASS, SOOT, BC AND OC LEVELS

Summary

The highest mean concertation of $PM_{2.5}$ for the study was observed during winter and mid-autumn, while the lowest concentration was observed during summer.

4.1 Overview of $PM_{2.5}$ mass, soot, BC and OC levels

The normality test (Shapiro-Wilks test) for the entire $PM_{2.5}$, BC, OC and soot levels revealed they are not normally distributed, however, the normality test showed that $PM_{2.5}$ concentrations for winter ($p > 0.173$), spring ($p > 0.313$) and summer ($p > 0.752$) had normal Gaussian distribution while autumn had non-Gaussian distribution. Also, normality test for soot showed that only winter ($p > 0.201$) and summer ($p > 0.105$) data had normal Gaussian distribution while autumn and spring had non-gaussian distribution. There were significant differences when seasonal $PM_{2.5}$ and soot were subjected to a Kruskal-Wallis test. ($p < 0.000$). Also, no significant difference was found for $PM_{2.5}$ and soot between weekdays and weekends. ($p > 0.957$ and $p > 0.268$ respectively).

The daily $PM_{2.5}$ concentration in Pretoria for the entire study period (18 April 2017 to 17 April 2018) is shown in Figure 4.1. The highest daily concentrations were observed from late April 2017 to mid-August 2017 and started to drop late August. The highest daily concentration (66.8 $\mu g/m^3$) was recorded on 2 of June 2017 (winter season) while the lowest concentration (0.7 $\mu g/m^3$) was on 1 October 2017 (spring season). The $PM_{2.5}$ concentrations in spring and summer were the lowest, and there were a few days with fairly good air quality. However, the $PM_{2.5}$ pollution was much worse in late autumn and winter season. Both the mean concentration and frequency of pollution episodes were greater in winter and autumn than the other two seasons. Similarly, the daily soot levels recorded in the study also followed the same pattern as $PM_{2.5}$ levels (Figure 4.2). Highest daily soot level (8.6 $x10^{-5}m^{-1}$) was recorded 20 July 2017 while the lowest value (0.1 $x10^{-5}m^{-1}$) on the 23 December 2017 (summer season).

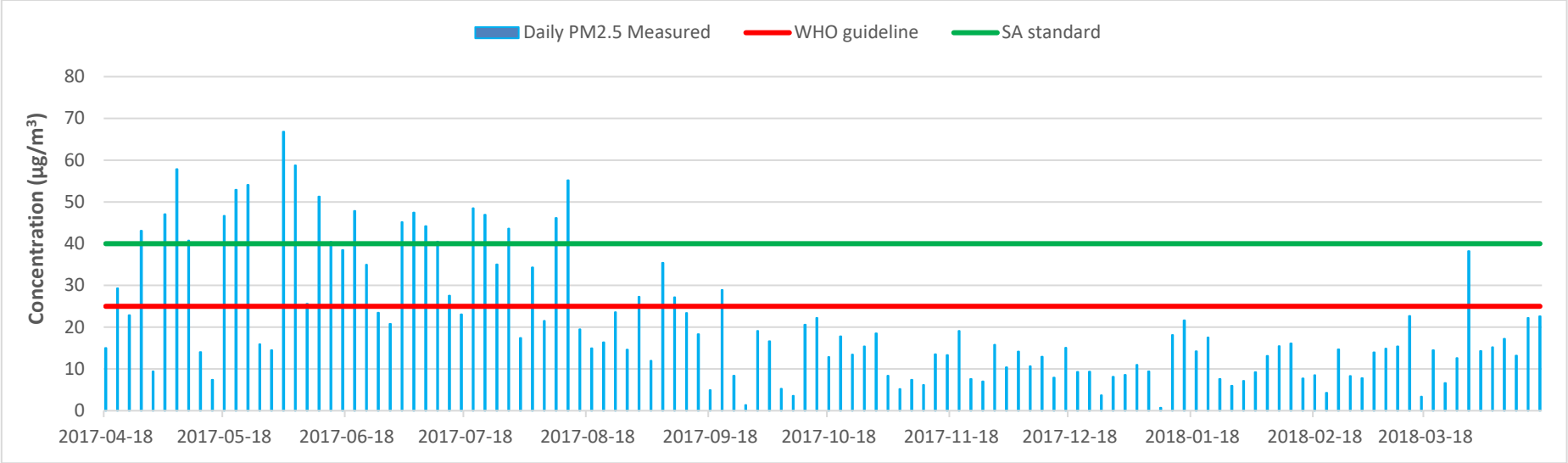


Figure 4.1: Daily PM2.5 levels measured at the School of Health System and Public Health, University of Pretoria from 18 April 2017 to 17 April 2018

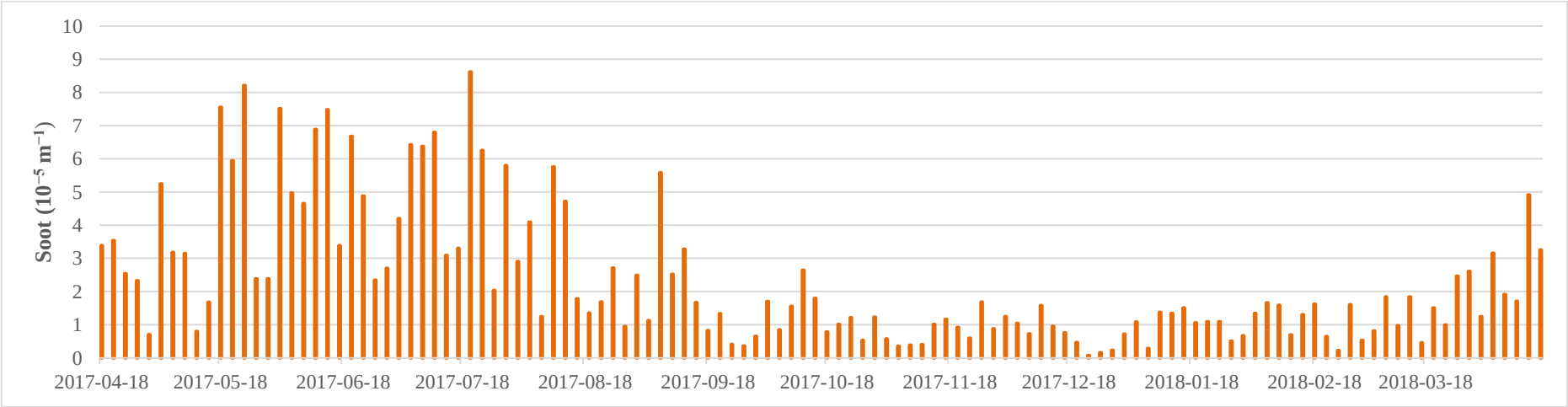


Figure 4.2: Daily soot levels measured at the School of Health System and Public Health, University of Pretoria from 18 April 2017 to 17 April 2018

The graph plots for BC and OC also showed similar seasonal trend to $PM_{2.5,}$ where the highest daily values were recorded during the cold seasons and the lowest daily values observed during the warm seasons (Figures 4.5 and 4.6). The mean concentrations for $PM_{2.5}$, BC, OC and soot level for the entire sampling period are summarised in Table 4.1 while the summary for the $PM_{2.5}$ duplicate samples are presented in Table 4.2.

The scatter plot of $PM_{2.5}$ concentrations and its associated duplicate sample is shown in Figure 4.3; this shows that the samples and duplicates are well correlated. Results showed there was no significant difference between the mean of the samples and the duplicates. Furthermore, the paired t-test confirmed that no significant difference ($p>0.05$) existed between the mean concentration of both the sample and duplicates.

The mean concentration for $PM_{2.5}$ (n =122 days) is 21.1 $\mu g/m^3$, which was higher than the annual WHO guideline and South African standard (2015) of 10 $\mu g/m^3$ and 20 $\mu g/m^3$, respectively, while the range of $PM_{2.5}$ for the study was 0.7 – 66.8 $\mu g/m^3$. Out of the 31 days of sampling during winter, 21 days and 14 days exceeded the daily WHO guideline and South African standard, respectively (Table 4.3).

In autumn, 29% (9 days) and 23% (7 days) of the 31 days were found to exceed the daily WHO guideline and South African standard respectively, while 10% (3 days) of the spring period exceeded the WHO daily guideline (Table 4.3). It is speculated that pollution levels in Pretoria was high due increased $PM_{2.5}$ concentration recorded on these days. The mean monthly $PM_{2.5}$ concentrations are shown in Figure 4.7. June and December had the highest and lowest concentrations with 40.0$\mu g/m^3$ and 10.2 $\mu g/m^3$, respectively.

The highest mean value for $PM_{2.5}$ was recorded in winter season (35.5 $\mu g/m^3$), followed by autumn (23.4 $\mu g/m^3$), spring (14.3 $\mu g/m^3$) and summer (10.7 $\mu g/m^3$), and they differed significantly across the seasons ($p<0.0001$) (Table 4.1). The box and whisker plot of seasonal concentration of $PM_{2.5}$ is shown in Figure 4.8, which clearly shows that seasonal variation exists.

Noteworthy to mention that the $PM_{2.5}$ measurements were continued at the same location (16 May 2018 to 23 April 2019) after my own sampling campaign period (as part of a long-term research project)[1]. The results for the $PM_{2.5}$ levels during this period showed a good ($r = 0.75$) correlation with real-time continuous measurements collected with an aeroqual that is located

directly next to the GilAir sampling site. (Figure 4.4) No aeroqual measurements during my sampling campaign as the instrument was serviced.

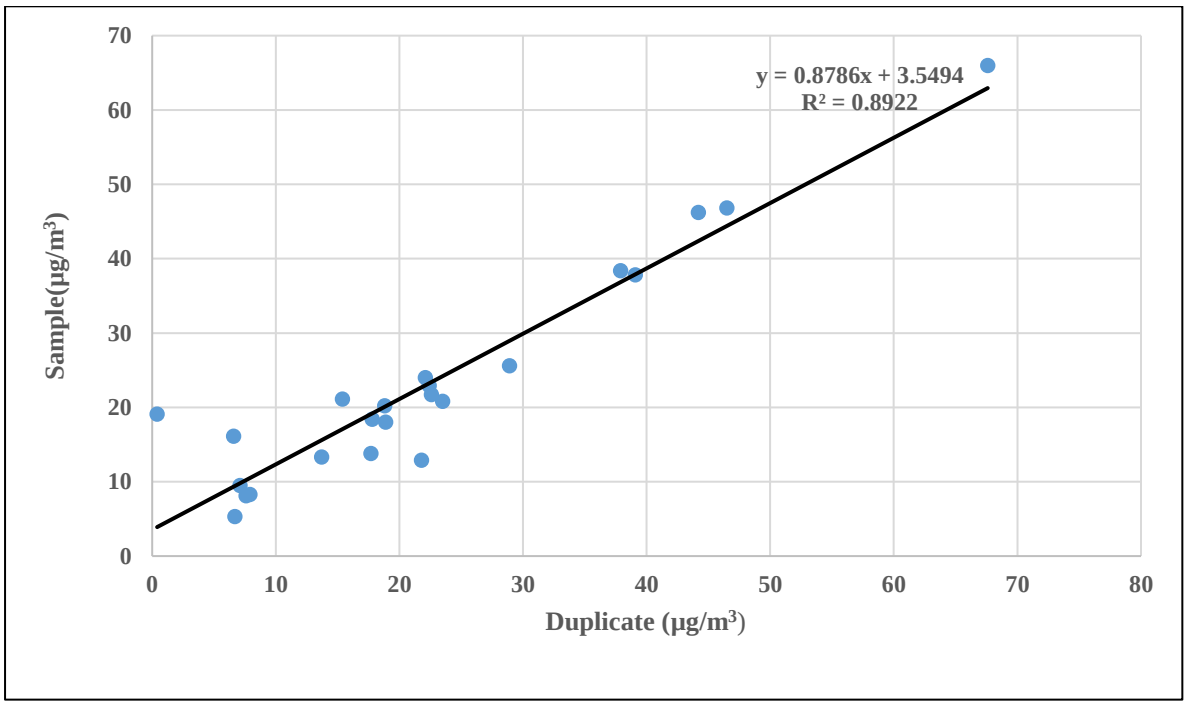


Figure 4.3: Relationship between $PM_{2.5}$ samples and duplicates measured at School of Health System and Public Health, University of Pretoria during 18 April 2017 and 17 April 2018

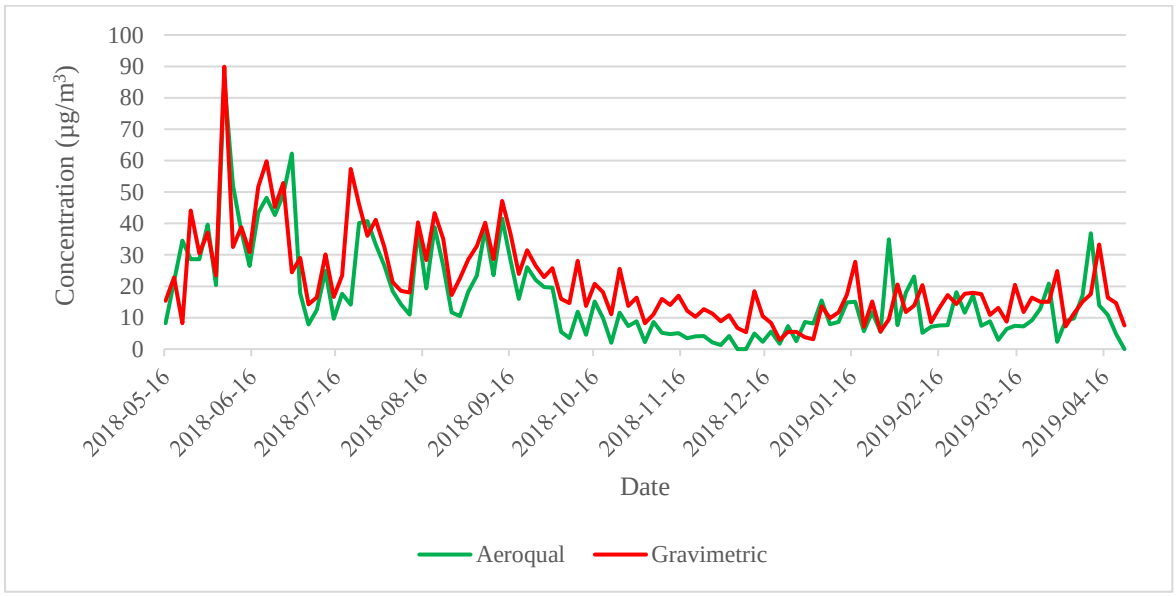


Figure 4.4: Gravimetric concentrations against the aeroqual concentration measured at School of Health System and Public Health, University of Pretoria during 16 May 2018 to 23 April 2019. (Source Mwase, 2020)[1]

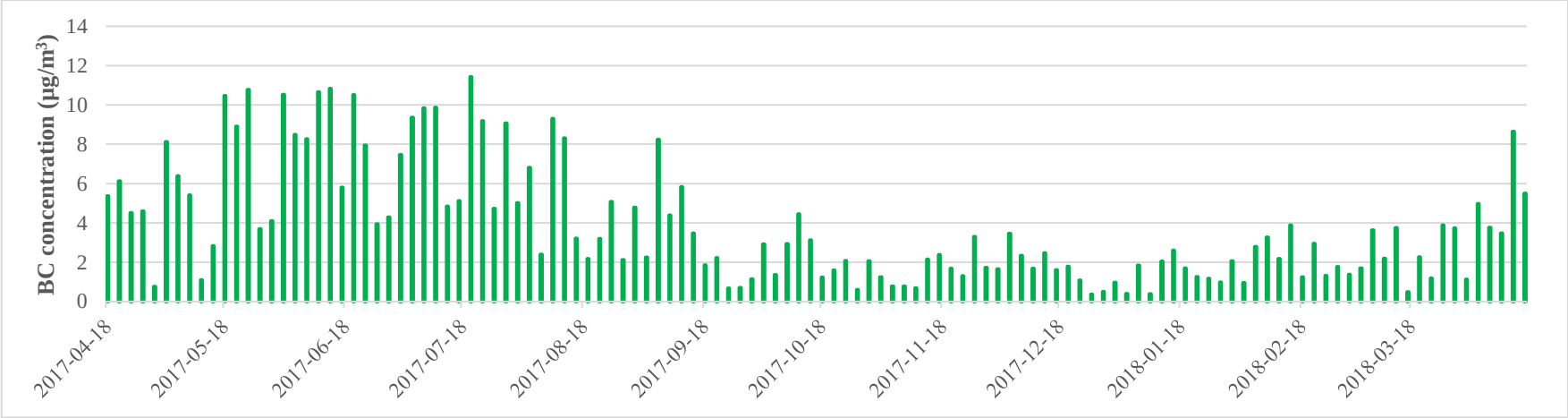


Figure 4.5: Daily black carbon levels measured at the School of Health System and Public Health, University of Pretoria from 18 April 2017 to 17 April 2018

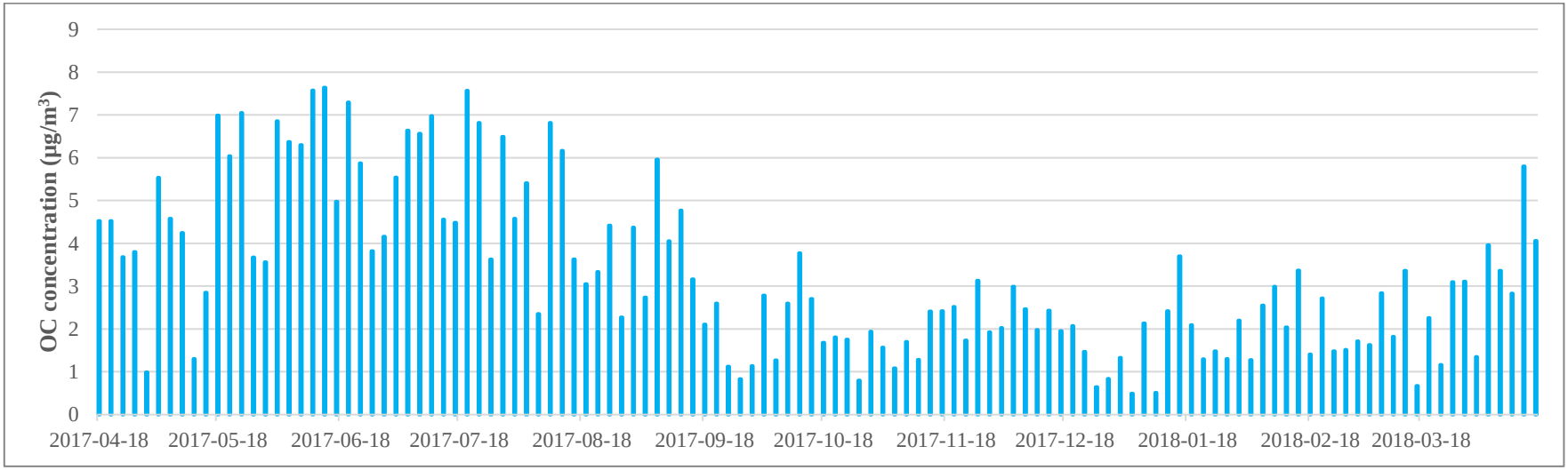


Figure 4.6: Daily organic carbon levels measured at the School of Health System and Public Health, University of Pretoria from 18 April 2017 to 17 April 2018

Table 4.1:Summary of $PM_{2.5}$, black carbon, organic carbon concentration and soot levels for full study and by seasons, weekdays and weekends measured at the School of Health System and Public Health, University of Pretoria during 18 April 2017 and 17 April 2018

		$PM_{2.5}$ (µg/m^3)				Soot (10^{-5} m^{-1})			
	N	**mean**	**SD**	**median**	**range**	**mean**	**SD**	**Median**	**range**
Full study	122	21.1	15.0	15.6	0.7 – 66.8	2.3	2.0	1.0	0.1 – 8.6
Autumn	31	23.4	15.7	15.4	3.4– 57.9	2.7	1.9	2.4	0.5– 8.2
Winter	31	35.5	14.2	35.0	14.6 -66.8	4.3	2.2	4.2	0.9 – 8.6
Spring	30	14.3	8.2	13.5	1.3 – 35.4	1.3	1.1	1.0	0.4 – 5.6
Summer	30	10.7	4.6	9.3	0.7 – 21.6	1.0	0.5	1.1	0.1 – 1.7
Weekdays	88	20.7	14.3	16.0	0.7 – 66.8	2.4	2.1	1.7	0.2 – 8.6
Weekends	34	22.1	17.0	15.3	3.4 – 57.9	2.2	2.1	1.3	0.1 – 6.9
		BC (µg/m^3)				**OC (µg/m^3)**			
	N	**mean**	**SD**	**median**	**range**	**mean**	**SD**	**Median**	**range**
Full study	122	3.9	3.0	2.9	0.4 – 11.4	3.3	1.9	2.8	0.5 – 7.6
Autumn	31	4.3	2.9	7.5	2.1 – 11.4	3.4	1.6	5.5	2.3 – 7.6
Winter	31	6.9	2.8	3.8	0.5 – 10.8	5.4	1.7	3.4	0.7 – 7.0
Spring	30	2.3	1.7	2.0	0.6 – 8.3	2.3	1.2	2.0	0.8 – 5.6
Summer	30	1.8	0.9	1.7	0.4 – 3.9	1.9	0.8	2.0	0.5 – 3.4
Weekdays	88	4.0	3.0	3.2	0.4 – 11.4	3.4	1.9	2.9	0.5 – 7.6
Weekends	34	3.5	3.2	2.1	0.5 – 10.7	3.1	2.1	2.3	0.7 – 7.6

Table 4.2: Summary of $PM_{2.5}$, black carbon, organic carbon concentration and soot levels measured at the School of Health System and Public Health, University of Pretoria during 18 April 2017 and 17 April 2018 (n=25 for duplicate samples)

Species	Mean	SD	Median	Range
$PM_{2.5}$ (µg/m^3)	24.2	15.2	20.5	6.7 – 66.0
BC (µg/m^3)	4.4	2.9	3.4	0.4 – 10.5
OC (µg/m^3)	3.6	1.7	3.4	0.8 – 6.9
Soot ($10^{-5}m^{-1}$)	2.7	2.1	2.0	0.2 -7.4

Table 4.3: Number of times the recorded $PM_{2.5}$ exceeded WHO daily guideline and South Africa air quality standard

Seasons	No of exceedance	
	WHO daily guideline for $PM_{2.5}$ (25.0 µg/m^3)	**Daily South Africa standard for $PM_{2.5}$ (40.0 µg/m^3)**
Summer	0	0
Spring	3	0
Autumn	9	7
Winter	21	14

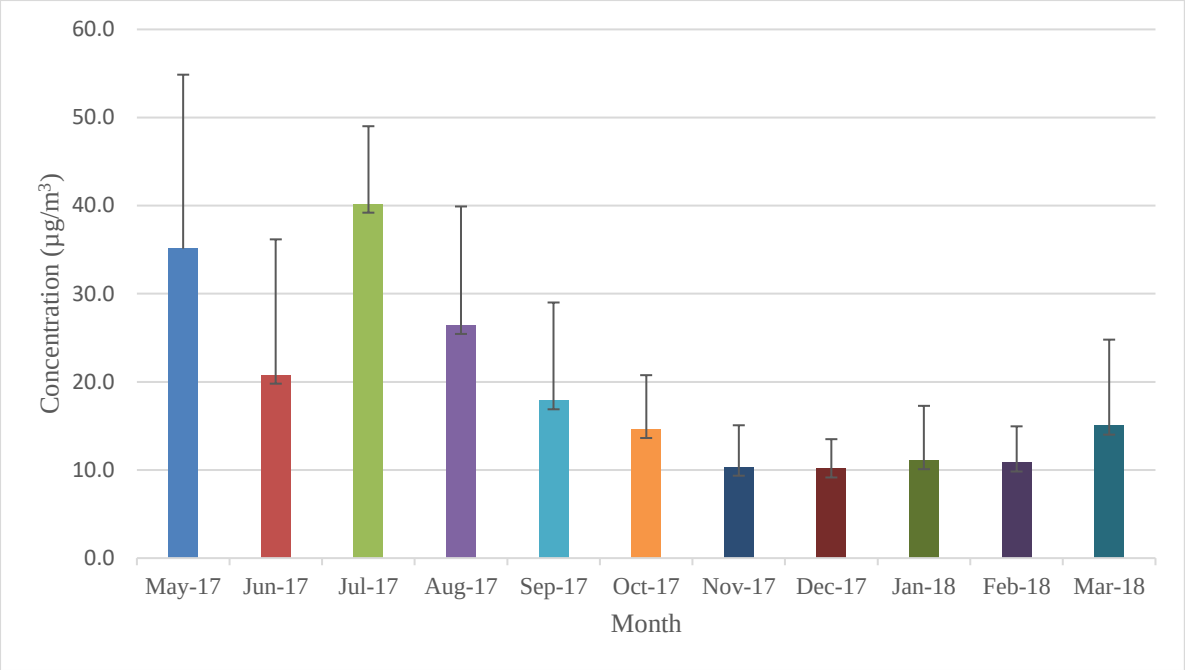


Figure 4.7: Monthly $PM_{2.5}$ levels measured at the School of Health System and Public Health, University of Pretoria from May 2017 to March 2018 (mean and standard deviation for each bar)

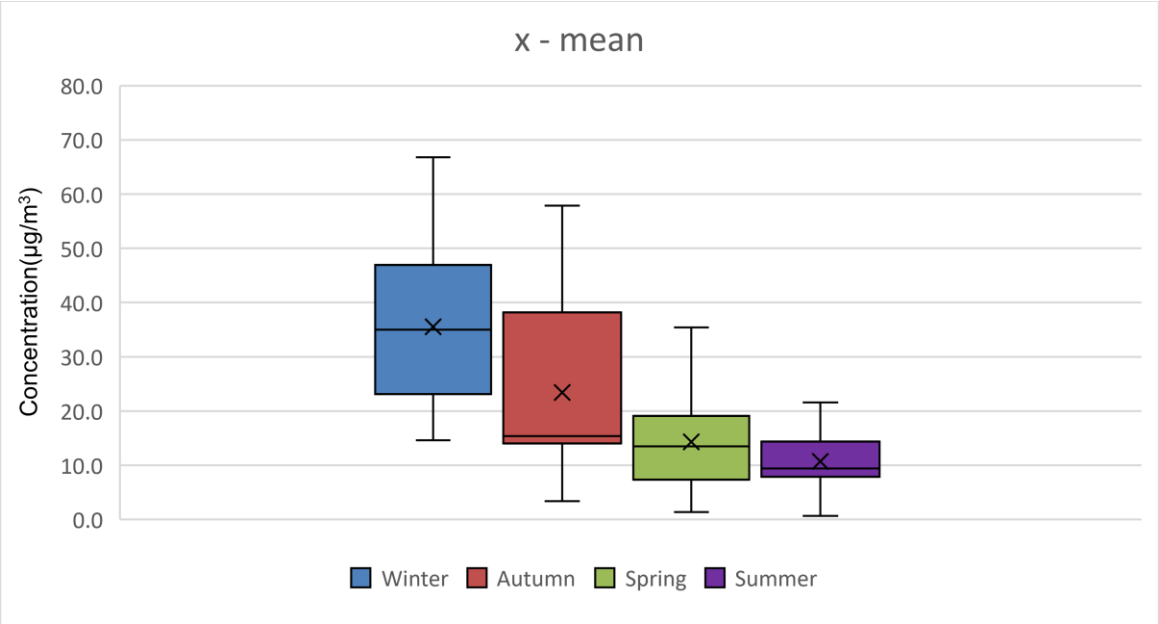


Figure 4.8: Box and whisker plots of seasonal $PM_{2.5}$ levels measured at the School of Health System and Public Health, University of Pretoria from 18 April 2017 to 17 April 2018.

The number of weekends recorded during the entire study period was 34 days with $PM_{2.5}$ mean value of 22.1 µg/m^3, while 88 days were weekdays with mean value of 20.7 µg/m^3 (Table 4.1) No significant difference between weekdays and weekends (p>0.9567) was observed.

The annual mean for soot (n =122 days) was 2.3x10^{-5} m^{-1} (range 0.1x10^{-5} m^{-1} – 8.6x10^{-5} m^{-1}) (Table 4.1). Soot also followed the same seasonal trend than $PM_{2.5}$. Overall, the soot mean value in winter, autumn, spring and summer was 4.3 x10^{-5} m^{-1}, 2.7 x10^{-5} m^{-1}, 1.3 x10^{-5} m^{-1}, 1.0 x10^{-} m^{-1}, respectively. The soot mean value for weekdays (2.4x 10^{-5} m^{-1}) was slightly higher than weekends (2.2x10^{-5} m^{-1}) (Table 4.1). There was no significant difference between weekdays and weekend (p>0.2680) for soot.

The mean monthly soot levels are shown in Figure 4.9. June and December had the highest and lowest mean levels with 5.2 x10^{-5} m^{-1} and 0.7 x10^{-5} m^{-1}, respectively. Soot levels had a consistent downward trend over a five-month period (August-December).

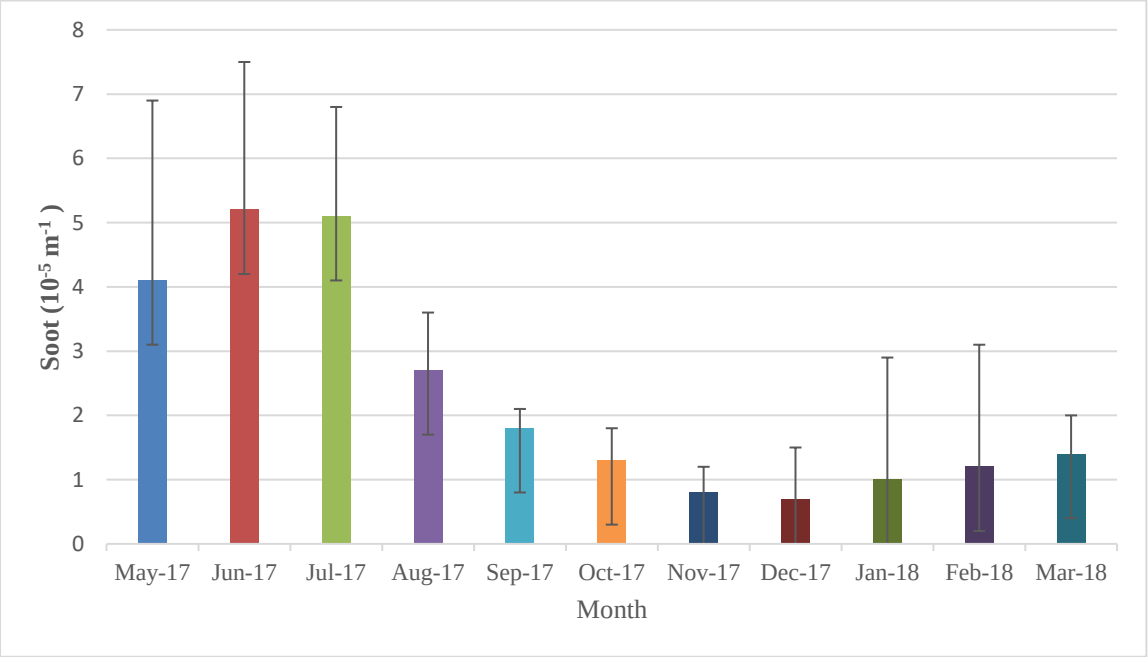


Figure 4.9: Monthly soot levels measured at the School of Health System and Public Health, University of Pretoria during May 2017 and March 2018 (mean and standard deviation for each bar)

$PM_{2.5}$ concentration and soot had a strong significant positive correlation (r = 0.86; p<0.000) (Figure 4.10), indicating a strong relationship between these two parameters in the study area. The contributions of other components such as ionic and elemental components is indicated by the positive intercept[51].The seasonal plot of $PM_{2.5}$ and soot showed a strong significant positive

correlation for winter, (r = 0.82; p<0.000), autumn (r = 0.77; p<0.000) and spring (r = 0.72; p<0.000), but a weak significant correlation for summer (r = 0.44; p<0.015) (Appendix 9).

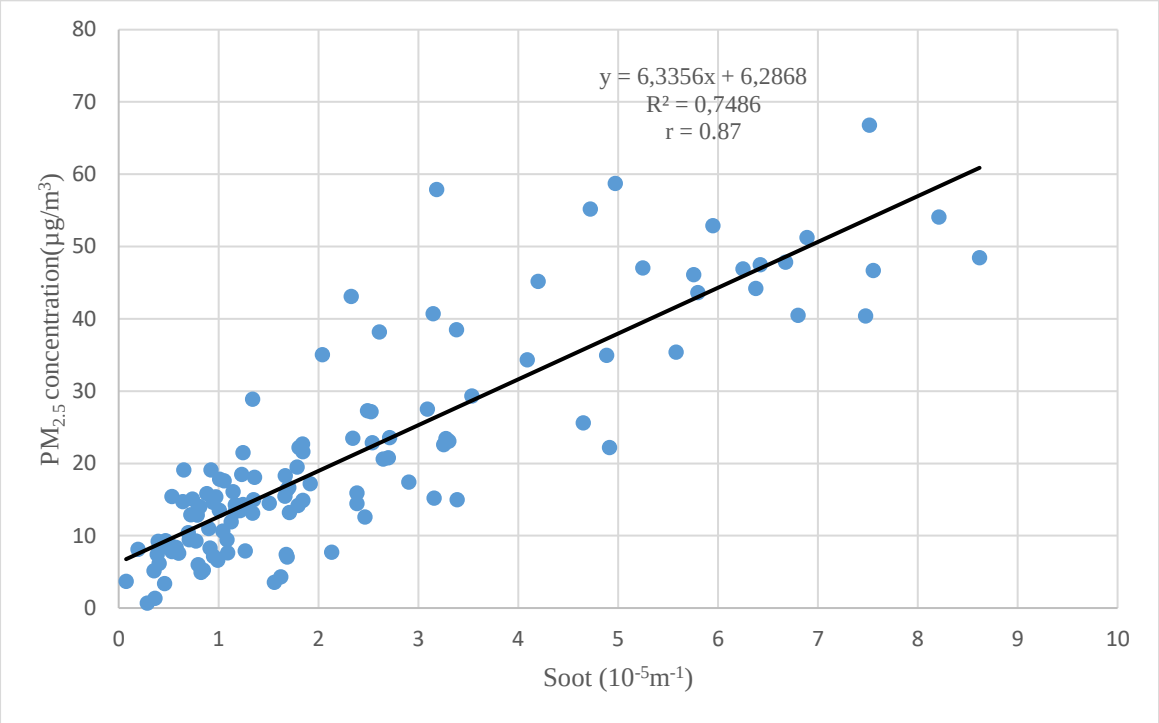


Figure 4.10: The relationship between $PM_{2.5}$ and soot levels measured at the School of Health System and Public Health, University of Pretoria during 18 April 2017 and 17 April 2018.

Weekly daily average graphs of $PM_{2.5}$ concentration and soot levels over the period of measurement are shown in Figure 4.11 and 4.12, respectively. The highest mean levels were recorded on a Thursday, while Wednesday and weekends had the same concentration for $PM_{2.5}$. Soot recorded the highest mean levels on Friday followed by Monday

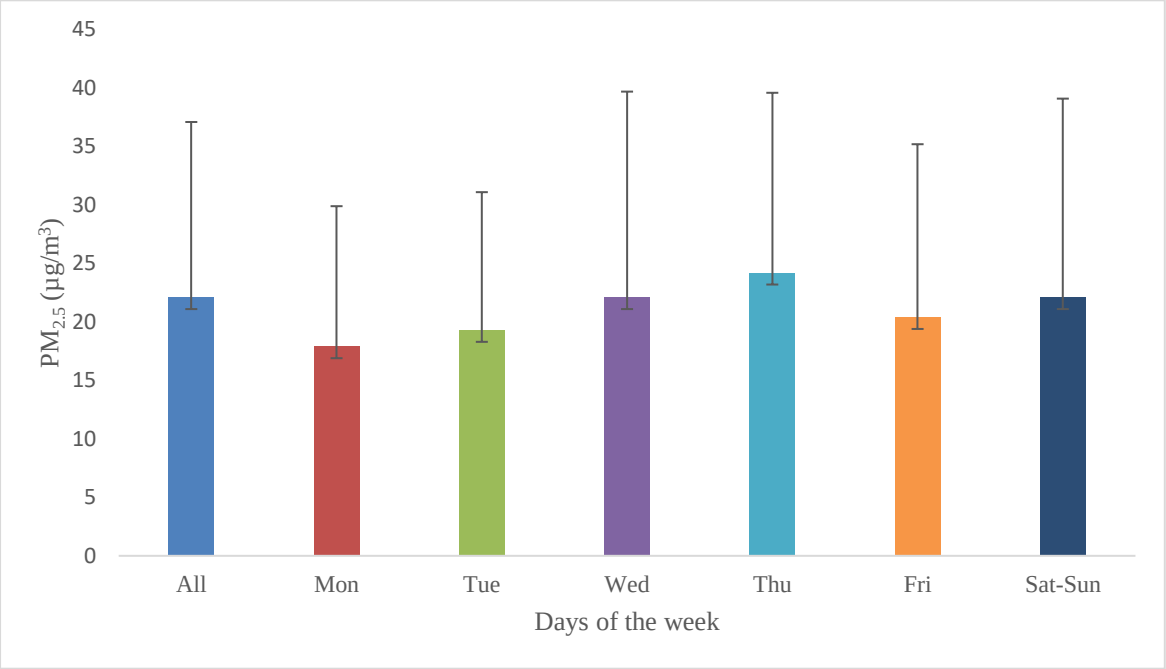


Figure 4.11: Weekly-daily average of $PM_{2.5}$ levels measured at the School of Health System and Public Health, University of Pretoria during 18 April 2017 and 17 April 2018 (mean and standard deviation for each bar)

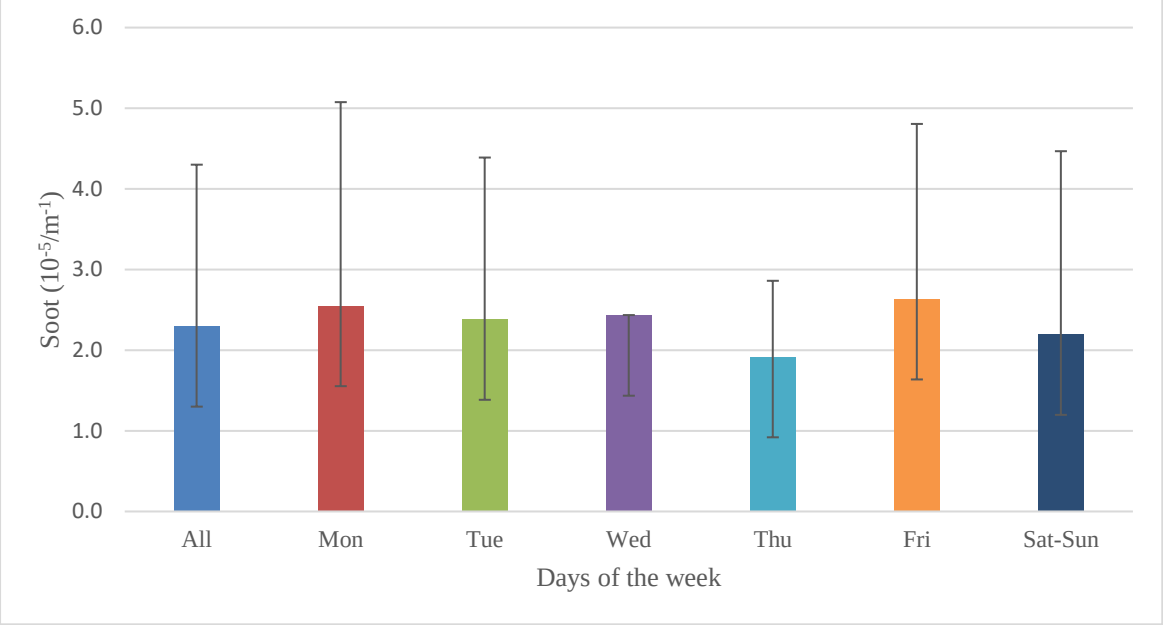


Figure 4.12: Weekly-daily average of soot levels measured at the School of Health System and Public Health, University of Pretoria during 18 April 2017 and 17 April 2018 (mean and standard deviation for each bar)

The annual mean for BC and OC was 3.9 µg/m^3 and 3.3 µg/m^3, respectively (Table 4.1). The seasonal mean concentration for BC and OC is summarised in Table 4.1. BC and OC followed the same seasonal trend as $PM_{2.5}$ levels, where winter and autumn had the highest levels of BC and OC and spring and summer experienced the lowest levels. There was significant difference across the seasons for both BC and OC ($p<0.0001$ and $p<0.0001$, respectively). The mean levels of BC was higher than that of OC for all the days of the week (Figure 4.13), while the highest mean levels for BC and OC was recorded on a Thursday.

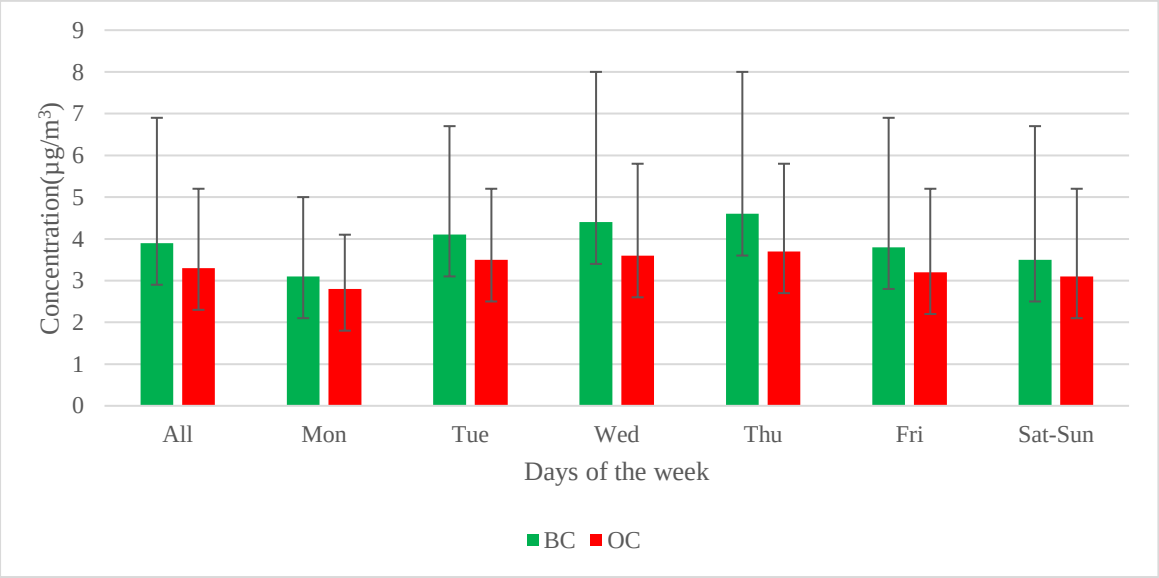


Figure 4.13: Weekly-daily average of black and organic carbon levels measured at the School of Health System and Public Health, University of Pretoria during 18 April 2017 and 17 April 2018 (mean and standard deviation for each bar)

Similarly, the BC mean levels was higher than that of OC during weekdays and weekends (Table 4.3); no significant difference was observed during weekdays and weekends for both BC and OC ($p>0.131$ and $p>0.211$ respectively). During the monthly observation, an increase was recorded for both BC and OC in May and later decreased at the end of autumn before increasing again. Highest peak was recorded in July and again decreased at the end of winter (Figure 4.14). Significant differences were observed for the monthly BC and OC levels ($p<0.0001$ for BC and OC).

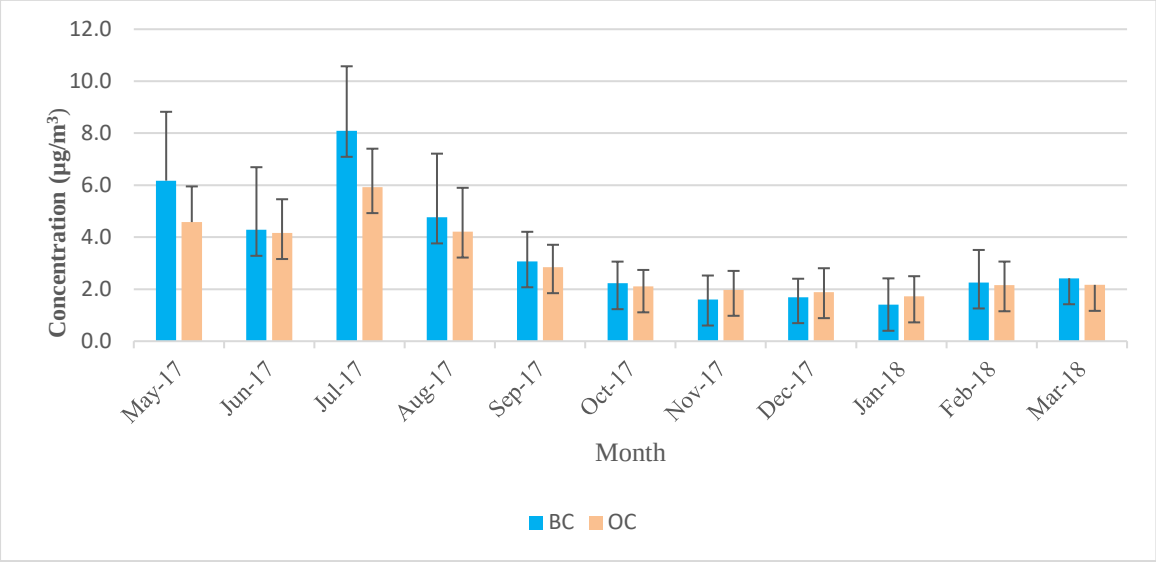
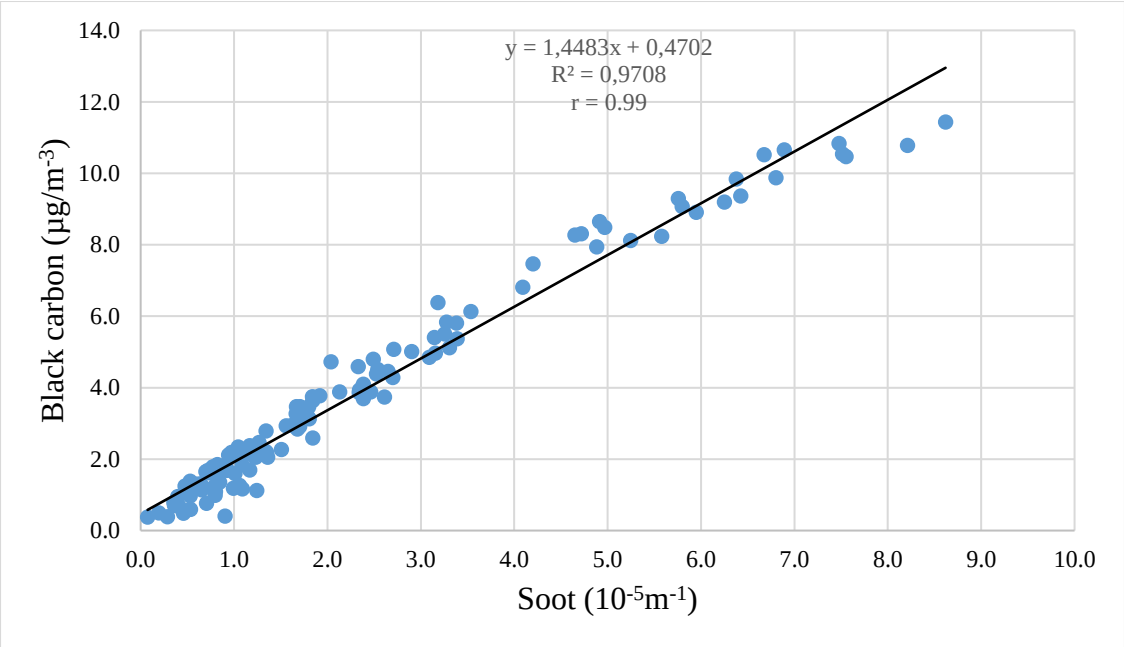


Figure 4.14: Monthly BC and OC levels measured at the School of Health System and Public Health, University of Pretoria in Pretoria from 18 April 2017 to 17 April 2018 (mean and standard deviation for each bar).

4.3 Relationship between BC, OC and soot

BC and OC levels were highly correlated with soot. ($r = 0.99$, $p<0.0000$ and $r=0.96$ $p<0.0000$ respectively) (Figure 4.15a and 4.15b), over the entire study period.

(a)



(b)

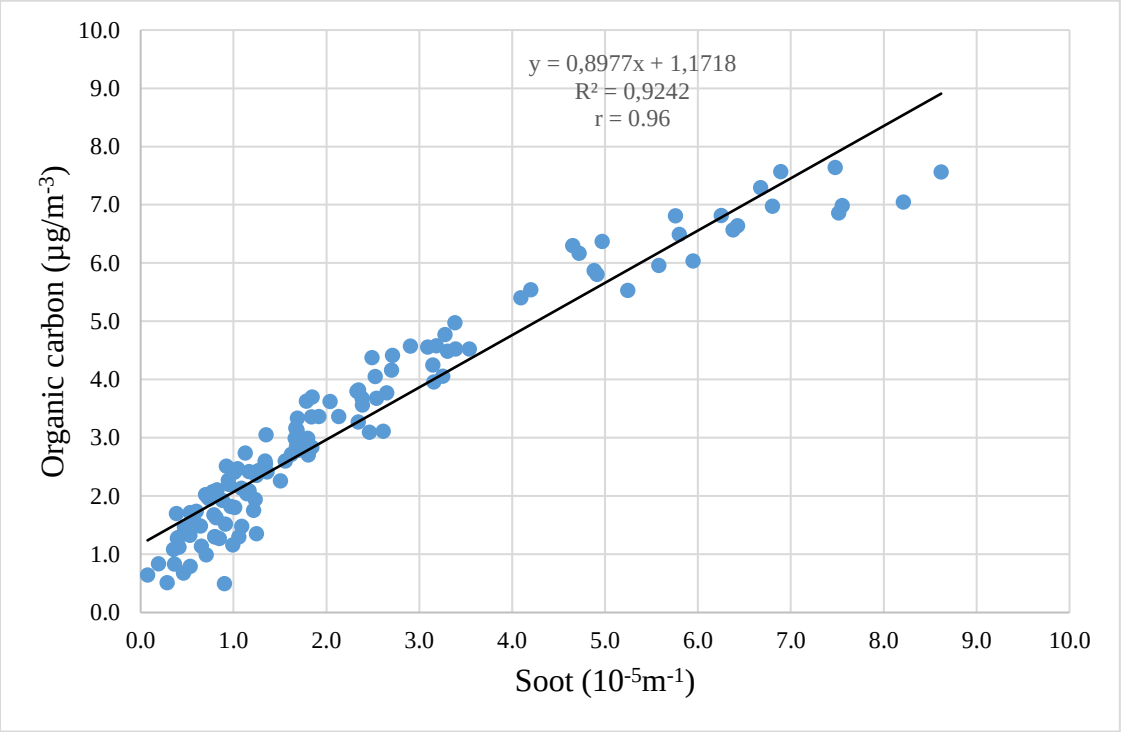


Figure 4.15: Black carbon (µg/m^3) versus soot (10^{-5}/m^{-1}) (a) and organic carbon (µg/m^3) versus soot (10^{-5}/m^{-1}) levels(b) measured at the SHSPH, University of Pretoria during 18 April 2017 and 17 April 2018.

4.4 Spearman correlation coefficients between air pollution and weather variables

The spearman correlation between the air pollution and weather variables are shown in Table 4.4. $PM_{2.5}$ and soot had a significant positive correlation and a significant negative correlation with temperature and relative humidity. Also, BC and OC had a significant negative correlation with temperature, but a significant negative correlation with relative humidity. Wind speed had a weak positive significant correlation with $PM_{2.5}$ and OC, but a significant negative correlation with temperature.

Temperature, RH and wind speed have been found to have significant impact on the ambient particulate matter. The mean temperature, RH and wind speed for the full study are 17.6 °C, 54.3% and 1.6 m/s respectively. Mean temperature and wind speed were highest for summer with 20.4°C, and 1.6 m/s (n= 30 days) respectively (Table 4.5). Winter had the lowest mean temperature and wind speed with 14.4°C and 2.11 m/s (n= 21 days) respectively. Autumn had the highest mean RH and wind speed with 64.1% and 1.2m/s, respectively. Daily temperature and RH during the study period is shown in Figure 4.16

Table 4.4: Correlation between air pollution and weather variables measured at the School of Health System and Public Health, University of Pretoria during 18 April 2017 and 17 April 2018.

Variable	$PM_{2.5}$	Soot	BC	OC	Temp	RH	WS
$PM_{2.5}$							
Soot	0.9629* 0.0000						
BC	0.7598* 0.0000	0.8744* 0.0000					
OC	0.7639* 0.0000	0.8548* 0.0000	0.9805* 0.0000				
Temp	-0.3181* 0.0016	-0.4914* 0.0000	-0.4945* 0.0000	-0.4591* 0.0000			
RH	-0.2238* 0.0284	-0.0272 0.7927	-0.0604 0.5587	-0.1208 0.2410	-0.3949* 0.0001		
WS	0.2090* 0.0410	0.1020 0.3227	0.1829 0.0745	0.2114* 0.0387	-0.3929* 0.0001	-0.0437 0.6725	

Abbreviations: $PM_{2.5}$: particulate matter with an aerodynamic diameter of less than 2.5 μm, BC- Black carbon Temp-temperature; RH- relative humidity; OC-Organic carbon; WS-wind speed
* significant ($p < 0.05$)

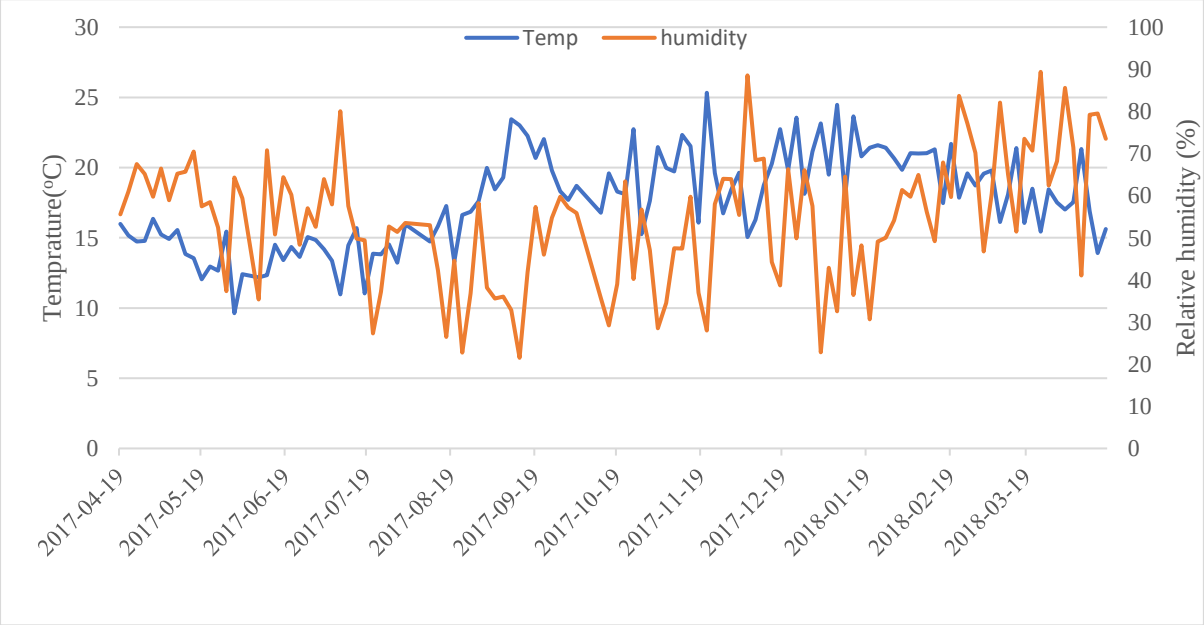


Figure 4.16: Time series plot of temperature and relative humidity levels in Pretoria during 18 April 2017 and 17 April 2018 by month.

Table 4.5: Summary of temperature, relative humidity, wind speed and rainfall in Pretoria during 18 April 2017 and 17 April 2018

Variables	Autumn	Winter	Spring	Summer	Full study
Mean temperature (ºC)	16.2	14.4	19.4	20.4	17.6
Min temperature (ºC)	9.6	11.0	11.5	16.5	9.6
Max temperature (ºC)	21.6	17.6	26.2	24.7	26.2
Mean RH (%)	64.1	49.9	45.1	57.3	54.3
Min RH (%)	37.4	22.7	14.1	30.1	14.1
Max RH (%)	81.9	80.0	73.2	84.1	84.1
Mean wind speed (m/s)	1.2	2.11	1.5	1.6	1.6
Max wind speed (m/s)	2.1	2.3	3.1	2.4	3.1
Rainfall (mm)	0	0	0	0	0

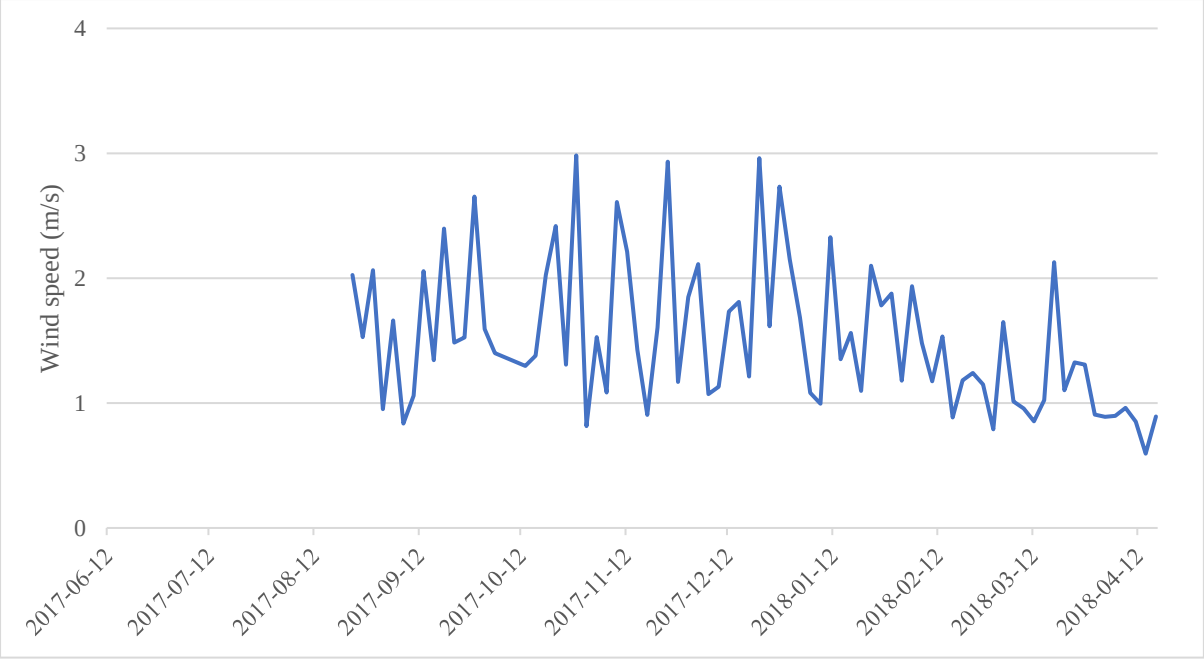


Figure 4.17: Time series of wind speed in Pretoria during 18 April 2017 and 17 April 2018 by month.

Among the meteorological variables that influences air pollution is wind speed. This influences the amount of pollutants in the atmosphere at any particular time and area. The maximum wind speed was observed in the months of October and December Figure 4.17. During the study

period, four wind directions dominated namely, W (27.4%), WSW (16.4%), E (13.7%) and ENE (13.7%). The frequencies for the E and W directions differ from each other with percentage frequencies of 34.2% and 41.1% respectively while the combined N and S directions account for 24.7%. (Appendix 10). No data were available for the monthly and seasonal wind direction for the months of April and May 2017 during the study period (Appendix 10). The plot of seasonal wind direction showed that W directions dominated in autumn with 81%, while it was lowest during winter (10%). Spring and summer W wind directions were virtually identical with 41% and 40%, respectively. E wind was highest during spring (38%) and lowest during winter (5%). Changes in $PM_{2.5}$, soot, BC and OC with temperature, relative humidity and wind speed are shown in Appendix 11 and Figures 4.18 and 4.19

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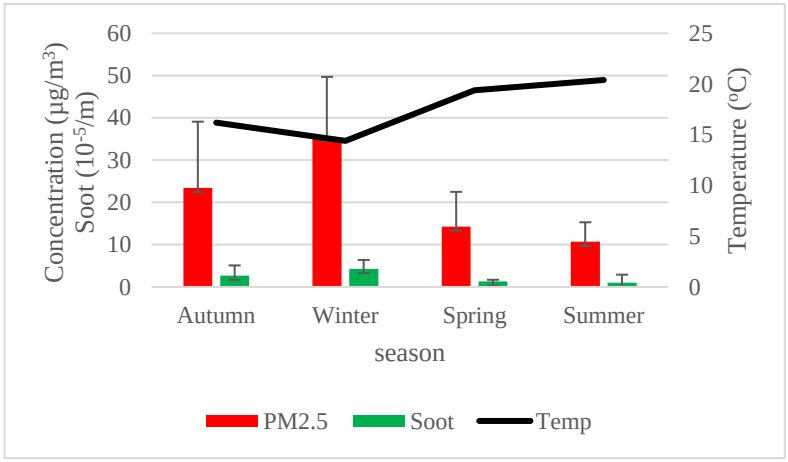
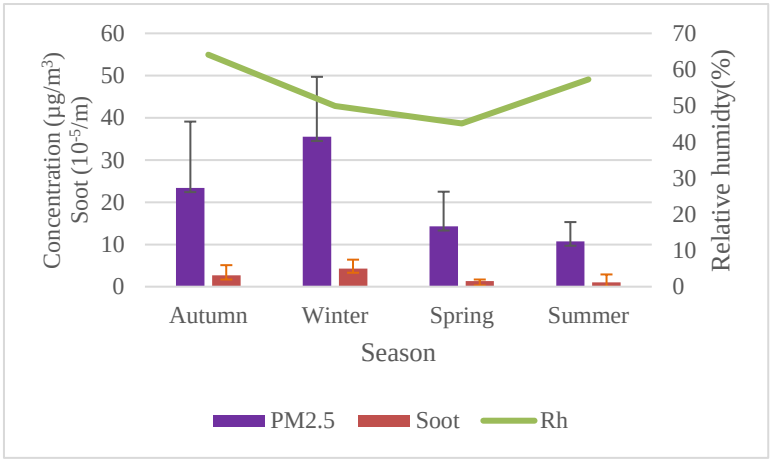




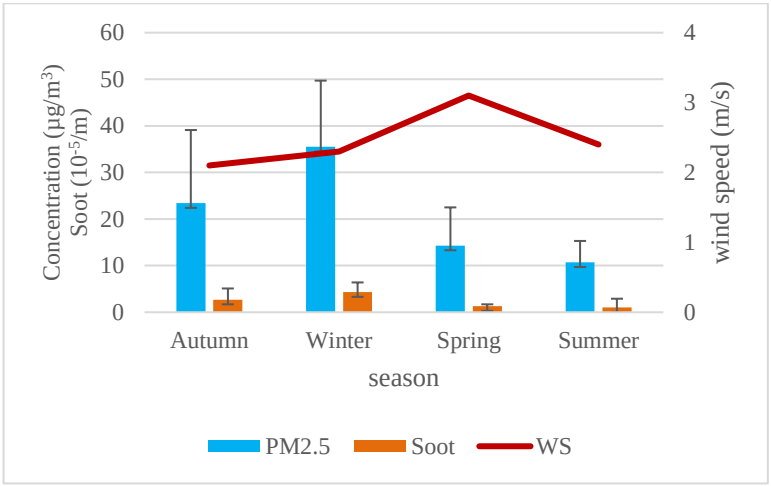
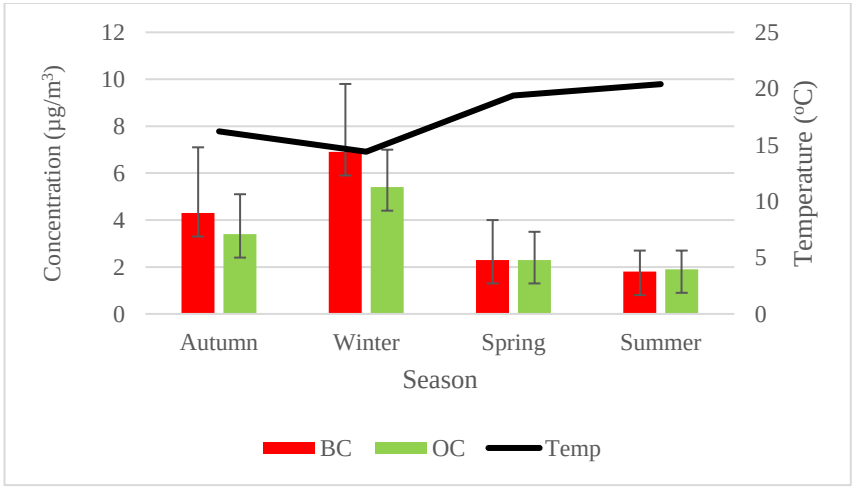
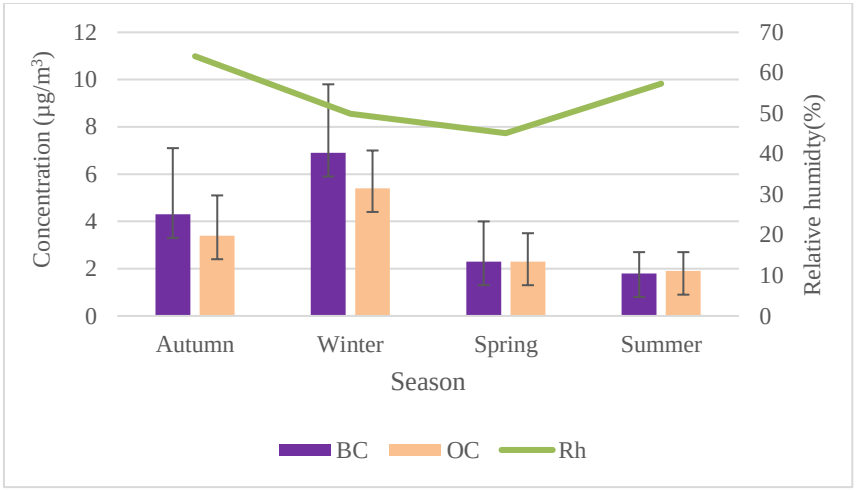


Figure 4.18: Seasonal concentration of $PM_{2.5}$ and soot versus temperature, relative humidity and wind speed in Pretoria from 18 April 2017 and 17 April 2018.





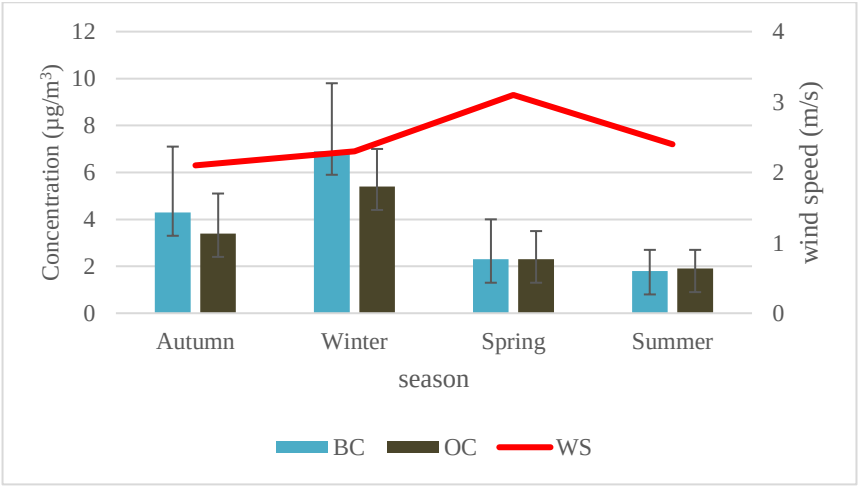


Figure 4.19: Seasonal concentration of BC and OC versus temperature, relative humidity and wind speed in Pretoria from 18 April 2017 and 17 April 2018.

4.5 Trace elemental composition.

The annual and seasonal mean concentration of the trace elements are summarised in Tables 4.6 and 4.7. Average S concentration was the highest among elements detected, followed by Si, Fe, K and Ca, in that order. The majority of the species concentration followed the seasonal trend observed for $PM_{2.5}$. Major elements including S, Si, Fe, Ca and K had the highest concentration in winter rather than summer while V and Ba did not follow any particular pattern. The daily concentrations of S, Ba, Ni, V, Fe, Si, K and Ca are presented in Figure 4.20 to 4.21 and others in Appendix 12-14. The daily concentration of elements, e.g. S, K, Ca, Fe, Ni and Si were present in appreciable amounts, these are also used as marker for relevant sources. The daily average S concentration recorded was highest during winter and early autumn of year 2018. S also recorded the highest concentration among the elements analysed for in the $PM_{2.5}$, winter being the highest with mean concentration of 1982.9 ng/m^3.

4.5.1 Weekly variation of some selected elements

The weekly variation of average concentration of K, Ca, Pb, Cl, S, and Fe are presented in Figure 4.22 to 4.23 One significant observation from this weekly variation is that the highest concentration of the elements was recorded on Thursdays, lowest on Mondays for (K, Pb, and Cl), Tuesday for (S) and weekends for (Fe, Ca). The concentrations of Pb and Cl displays variation during weekdays indicating variability in their sources, while the concentration of K, Ca, Fe and S do not display significant variation within the weekdays, and this indicates some consistency for their possible sources.

Table 4.6: Trace elemental composition of $PM_{2.5}$ levels measured at the School of Health System and Public Health, University of Pretoria during 18 April 2017 to 17 April 2018 (in ng/m^3)

Element	Detection limit	Mean	SD	median	Range
S	114	1476.2	1302.3	1094.8	8.0 – 6127.5
Cl	115	69.5	146.8	17.6	0.4 – 1223.5
K	115	357.0	373.7	1176.8	7.7 – 1527.1
Ca	115	174.3	168.9	117	3.7 – 834.9
Ti	115	31.2	23.7	25.5	1.1 – 129.8
Fe	115	368.1	197.6	335.9	47.9 – 1102.8
Ni	115	146.1	90.9	190.4	0.3 – 266.3
Cu	100	7.9	8.8	3.8	0.0 – 58.6
Zn	115	55.6	83.0	17.6	0.1 – 462.5
Si	115	565.5	517.1	405.9	4.1 – 2503.8
U	114	1.6	1.3	1.1	0.1 – 8.2
Pb	42	10.3	25.6	2.1	0.1 – 194.9
Ba	115	11.9	10.9	5.3	1.0 – 40.8
As	31	1.9	4.1	0.4	0.4 – 23.3
Br	113	18.1	28.4	8.3	0.4 – 253.7
Se	54	1.2	2.0	0.4	0.0 – 14.6
Sb	115	11.1	10.7	5.3	0.6 – 49.5
V	13	1.5	5.3	0.6	0.6 – 46.8

Table 4.7: Trace elemental composition of $PM_{2.5}$ levels measured at the School of Health System and Public Health, University of Pretoria during 18 April 2017 to 17 April 2018 by season (in ng/m^3)

Elements	Autumn	Winter	Spring	Summer
Si	424.2 ± 365.0	1039.4 ± 629.7	408.8 ± 414.2	374.1 ± 269.8
S	1599.2 ± 1650.2	1982.9 ± 1543.5	861.7 ± 707.7	1406.9 ± 773.5
Cl	62.2 ± 119.2	177.7 ± 230.7	19.3 ± 26.0	16.8 ± 37.2
K	201.5 ± 201.1	792.9 ± 379.8	308.8 ± 303.0	118.0 ± 92.7
Ca	160.2 ± 112.3	346.4 ± 204.5	95.4 ± 117.3	87.8 ± 54.0
Ti	26.1 ± 15.9	51.5 ± 30.1	21.7 ± 17.7	25.0 ± 14.9
V	1.7 ± 4.2	0.6 ±0.0	2.2 ± 8.4	1.6 ± 5.3
Fe	329.1 ± 185.8	549.6 ± 202.9	354.7 ± 137.9	236.6 ± 102.7
Ni	105.6 ± 107.1	171.2 ± 85.6	205.3 ± 35.4	106.4 ± 76.1
Cu	9.8 ± 10.8	10.7 ± 6.9	5.1 ± 4.7	6.1 ± 10.3
Zn	85.2 ± 97.2	105.2 ± 103.1	20.9 ± 31.5	9.9 ± 10.9
As	1.9 ± 4.0	4.9 ± 6.1	0.5 ± 0.6	0.4 ± 0.0
Se	1.0 ± 1.4	2.2 ± 3.4	0.7 ± 0.9	0.6 ± 0.4
Br	24.9 ± 46.9	34.8 ± 19.3	8.0 ± 6.2	3.9 ± 2.5
Sb	12.0 ± 11.5	8.3 ± 6.6	12.1 ± 12.5	12.1 ± 11.3
Ba	11.5 ± 10.7	14.4 ± 12.5	11.9 ± 11.2	10.1 ± 9.2
Pb	12.4 ± 23.6	23.6 ± 42.1	2.5 ± 1.5	2.7 ± 3.2
U	1.2 ± 0.7	2.0 ±1.8	1.5 ± 1.2	1.6 ± 1.2

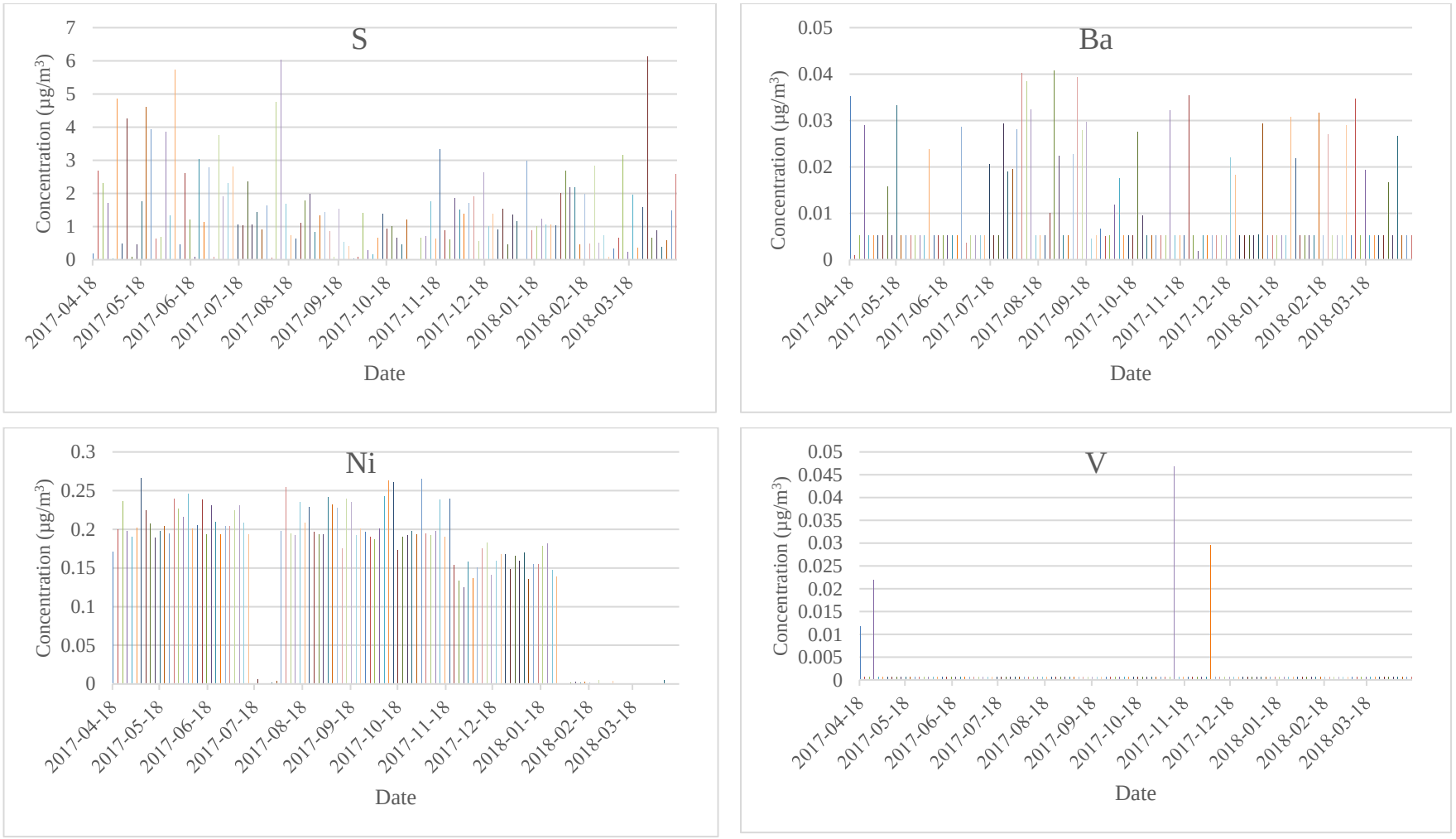


Figure 4.20: Graph of S, Ba, Ni and V content in $PM_{2.5}$ measured at the School of Health System and Public Health, University of Pretoria from 18 April 2017 to 17 April 2018.

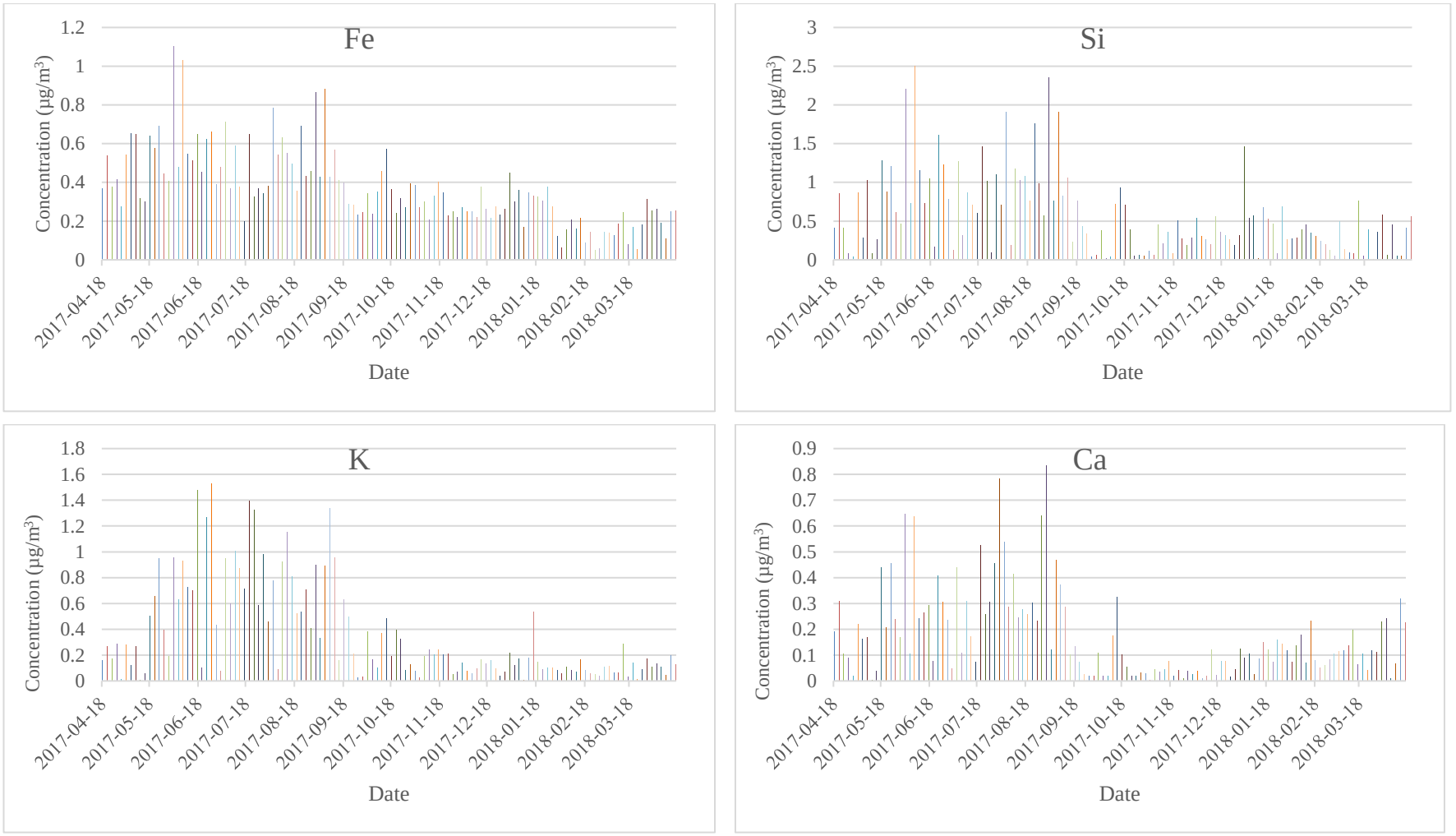


Figure 4.21: Graph of Fe, Si, K and Ca content in $PM_{2.5}$ measured at the School of Health System and Public Health, University of Pretoria from 18 April 2017 to 17 April 2018.

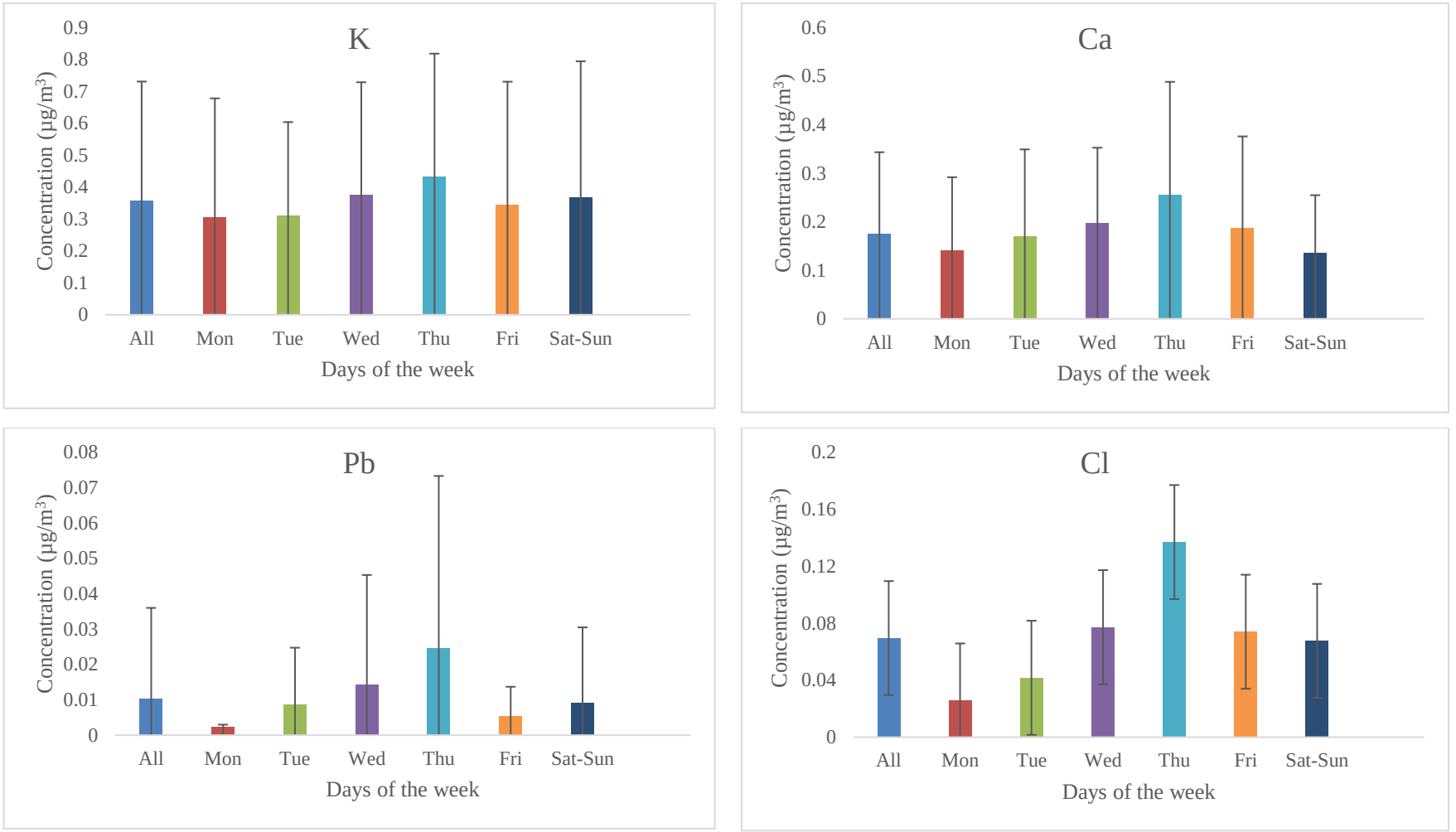


Figure 4.22: Weekly average of K, Ca, Pb and Cl content of $PM_{2.5}$ measured at the School of Health System and Public Health, University of Pretoria during 18 April 2017 to 17 April 2018 by days of the week.

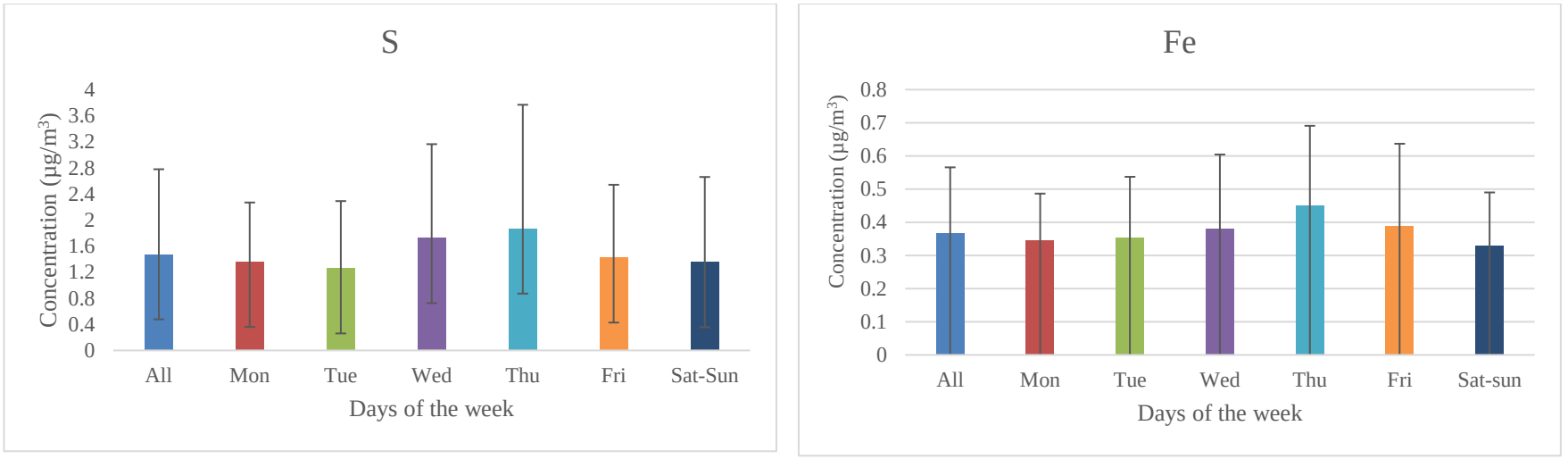


Figure 4.23: Weekly average of S and Fe content of $PM_{2.5}$ measured at the School of Health System and Public Health, University of Pretoria during 18 April 2017 to 17 April 2018 by days of the week.

4.6 Summary of $PM_{2.5}$ mean concentration and transport clusters

The mean levels of temperature, relative humidity and wind speed by transport clusters is shown in Table 4.8. The $PM_{2.5}$ mean concentration per cluster is summarised in Table 4.9. Five transport clusters were used in this study as the total spatial variance (TSV) value indicated that this is the optimum number of clusters (Figure 4.24).

During the $PM_{2.5}$ sampling period cluster 1 originated locally from North Limpopo Province (NLP), while cluster 2 can be referred to as Eastern inland (EI), though it has its root from the Indian Ocean and passes through Mozambique and Mpumalanga, marine influence on cluster 2 is minimal. Clusters 3 and 4 are short and long Indian Ocean (SIO and LIO) respectively. These clusters pass over Kwazulu-Natal Province before reaching the site. Lastly, cluster 5 is Southwest inland (SWI), which loses its marine influence as it travels from Western Cape to the sampling site.

Table 4.8: Mean levels of temperature, relative humidity and wind speed measured at the School of Health System and Public Health, University of Pretoria during 18 April 2017 to 17 April 2018 by transport cluster

Cluster		No of days	Mean	SD	Min	Max
Temperature (ºC)						
1	30%	38	15.9	3.1	11.9	24.7
2	50%	57	19.2	3.3	11.0	26.2
3	11%	16	16.5	2.9	9.6	21.1
4	4%	3	15.6	3.7	13.3	19.9
5	6%	8	17.3	4.1	11.0	22.9
Relative humidity (%)						
1	30%	38	54.9	12.9	22.7	74.6
2	50%	57	53.4	15.7	14.1	81.9
3	11%	16	60.7	13.9	31.4	84.1
4	4%	3	51.3	16.9	38.8	70.5
5	6%	8	46.5	11.9	34.8	65.2
Wind speed (m/s)						
1	30%	38	1.9	0.6	0.7	2.5
2	50%	57	1.5	0.5	0.7	2.8
3	11%	16	1.4	0.8	0	2.7
4	4%	3	1.1	-	1.1	1.1
5	6%	8	2.0	0.7	1.0	3.1

Table 4.9: Mean concentration of $PM_{2.5}$ Soot, BC and OC levels measured at the School of Health System and Public Health, University of Pretoria during 18 April 2017 to 17 April 2018 by transport cluster

Cluster			$PM_{2.5}$ (µg/m^3)				Soot (10^{-5} m^{-1})			
		No of days	Mean	SD	Min	Max	Mean	SD	Min	Max
NLP	30%	38	31.0	16.7	8.4	66.8	3.6	2.4	0.1	8.6
EI	50%	57	17.2	11.8	3.4	47.9	1.6	1.4	0.2	6.7
SIO	11%	16	16.7	13.5	0.7	51.3	2.3	2.0	0.2	6.9
LIO	4%	3	11.9	4.0	7.4	15.0	1.4	0.3	1.2	1.7
SWI	6%	8	14.2	8.8	1.3	25.6	1.8	1.5	0.4	4.7
			BC (µg/m^3)				OC (µg/m^3)			
		No of days	Mean	SD	Min	Max	Mean	SD	Min	Max
NLP	30%	38	5.7	3.3	0.6	11.4	4.4	2.0	0.8	7.6
EI	50%	57	2.8	2.2	0.5	10.5	2.7	1.5	0.7	7.3
SIO	11%	16	3.8	3.1	0.4	10.7	3.2	2.1	0.5	7.6
LIO	4%	3	2.5	0.3	2.2	2.8	2.8	0.3	2.4	3.1
SWI	6%	8	3.0	2.8	0.4	8.3	2.7	2.1	0.5	6.3

1: NLP- North Limpopo, 2: EI- Eastern Inland, 3: SIO- Short Indian Ocean, 4: LIO- Long Indian Ocean, 5: SWI-South-west Inland

4.6.1 Seasonal summary of $PM_{2.5}$ mean concentration and transport clusters

The seasonal mean of $PM_{2.5}$ and seasonal transport cluster is summarised in Table 4.10. Five transport clusters were used for each of the seasons as the TSV value indicated that this is the optimum number of clusters (Figure 4.24). These are Atlantic Ocean, Indian Ocean, Regional, National and Transboundary. The Atlantic-Ocean trajectory was characterised by air masses from southerly and south-westerly parts of the Atlantic Ocean, Indian-Ocean by air masses emanating from the south-easterly or easterly, regional by air masses emanating from within Gauteng; National (Nat) by air masses emanating from any of the provinces, and Transboundary (TB) by air masses emanating from neighbouring countries such as Mozambique, Zambia, Botswana and Namibia.

Table 4.10: Seasonal mean concentration of $PM_{2.5}$ per transport cluster

Cluster		No of days	Mean	SD	Min	Max
Winter						
WNW	34%	16	39.3	14.7	16.4	66.8
NLP	18%	8	34.1	12.5	14.6	47.9
EKZN	40%	3	37.4	17.8	17.4	51.3
LAO	5%	1	15.0	-	-	15.0
LIO	2%	3	24.1	1.3	23.1	25.6
Autumn						
SEC	11%	16	28.2	17.0	12.6	57.8
EMP	51%	11	20.2	13.4	3.4	43.1
SIO	9%	3	14.4	7.8	6.6	22.9
NLP	25%	1	7.4	-	-	7.4
LIO	4%	0	-	-	-	-
Spring						
SIO	33%	4	19.3	13.7	8.4	35.4
LIO	26%	18	13.9	8.2	3.6	28.9
NLP	8%	5	14.8	5.4	7.0	20.6
SAO	8%	1	13.3	-	-	13.3
LIO	25%	2	7.1	8.1	1.3	12.8
Summer						
EIO	58%	2	9.4	0.1	9.3	9.4
NLP	12%	20	11.9	4.7	4.3	21.6
WCI	13%	5	7.5	5.3	0.7	14.2
LIO	8%	0	-	-	-	-
SIO	9%	3	9.1	1.6	7.9	11.0

WNP-Western North west, NLP-North Limpopo, EKZN-Eastern Kwazulu-Natal, LAO-Long Atlantic Ocean, LIO- Long Indian ocean, SEC-Southern Eastern cape, SIO-short Indian Ocean, EMP- Eastern Mpumalanga, SAO-South Atlantic Ocean, EI- Eastern Inland, WCI- Western cape inland

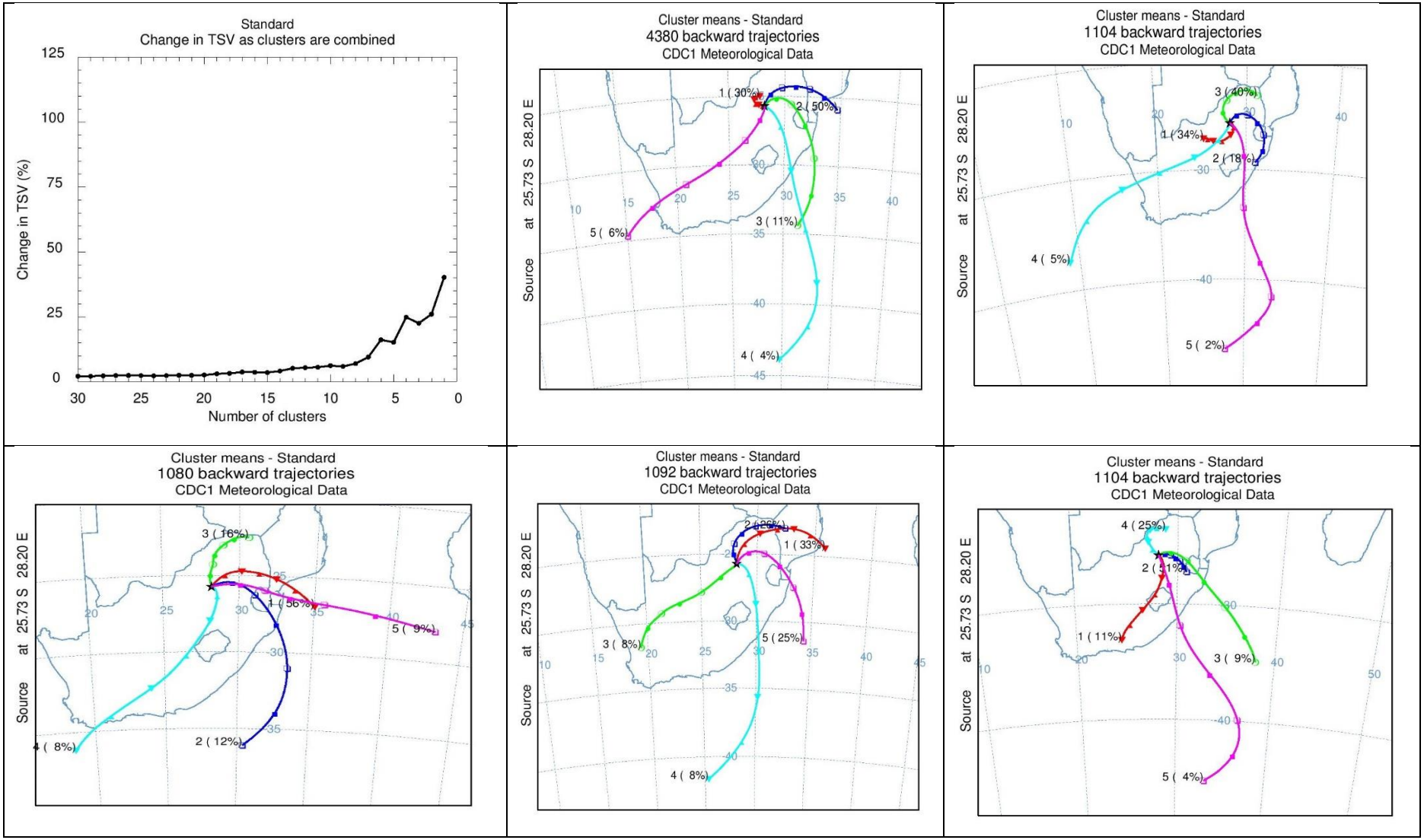


Figure 4.24: Five transport pathways (cluster plots) arriving at the sampling site during 18 April 2017 and 18 April 2018 by seasons, clockwise from top left, TSV, all year, winter, autumn, spring and summer.

4.7 Discussion

This section discusses the findings of the exposure assessment part of this study, which includes $PM_{2.5}$ characterisation, trace elemental composition, and origin of air masses into Pretoria during the study period 18 April 2017 to 17 April 2018. It is noteworthy to mention that studies on $PM_{2.5}$ and its chemical composition are scarce in this region; however, findings of this study will be discussed based on available information and other studies in different part of the country. Sampling days and methods used in other studies differ from what was used in this study, which may not be a good basis for comparison.

4.7.1 Relationship between $PM_{2.5}$ concentration and soot

During this study period, 33 days and 21 days had daily $PM_{2.5}$ that exceeded the daily WHO and South African standard of 25 µg/m^3 and 40 µg/m^3, respectively. The highest daily mean of 66.8 µg/m^3 was observed during winter (2 June 2017), when the origin of air masses for the day was locally originated via cluster 1. This cluster falls within Waterberg Bonjala Priority Area of Limpopo and North West. The high concentration of PM in this area has been attributed to existing mining and metallurgical activities in the western arm of the Bushveld Ingenious Complex (BIC)[1]. Venter *et al*[2], also reported significant exceedances of PM and ozone in this area. In comparison with other studies, the mean concentration of $PM_{2.5}$ in this study contrasted with the findings of the study conducted by Tshehla and Djolov[3] in Limpopo, which found the mean concentration of $PM_{2.5}$ to be lower than the WHO guideline and South African ambient standard. In contrast to this study, a two - and half-year study by Feig *et al*[4], conducted in the Waterberg area in Limpopo reported the mean annual $PM_{2.5}$ concentration of three monitoring stations to be below the South Africa standard.

In summer, the daily WHO guideline and South African standard for $PM_{2.5}$ were not exceeded, while the highest numbers of exceedance were recorded during the winter season. The winter season recorded the highest average amount of $PM_{2.5}$, which may partly be due to the low mixing atmospheric layers, biomass burning and other anthropogenic sources. The observed increased PM during winter can be supported by the findings of several authors who reported that the higher concentration of $PM_{2.5}$ observed during winter could be associated with biomass burning, as this and agricultural activities have been reported as major contributors to $PM_{2,5}$ concentration in the atmosphere[5-9]. The most significant areas for biomass burning in Southern Africa (veld fires which occur late early autumn and late winter), informal settlements

(primarily domestic burning throughout winter), mine tailings and waste dumps, which depend on wind speed as reported by Hersey *et al*.[9] in Gauteng, could partly be responsible for the high concentration of $PM_{2.5}$ recorded in this study. Other significant sources in the province that could lead to the elevated concentration of $PM_{2.5}$ during winter are three coal fire plants, two petrochemical refineries in the south, five steel manufacturing facilities and three cement factories in the south and the central part of the region.

The day when the highest daily value for $PM_{2.5}$ was observed (2 June 2017), the mean meteorological variables (temperature, relative humidity and wind speed) were low while no rainfall was recorded for the day, i.e. 12.4^{o}C, 59.4% and 0.2 m/s respectively. The higher values observed may be attributed to these weather conditions, as these hinder the dispersion of air pollutants. It is known that temperature has as an important meteorological factor that correlates with air quality. Some authors have reported that temperature correlates positively with $PM_{2.5}$ but other studies[10-12] reported a negative correlation with temperature which was consistent with the findings of this study. Low relative humidity also allows pollutants to stay longer in the air, contributing to the high concentration observed. The lowest daily $PM_{2.5}$ recorded (10 January 2018) during summer was attributed to the high temperature, relative humidity and wind speed.

The highest daily soot level was recorded on 20 July 2017 did not match with the day the highest daily $PM_{2.5}$ was recorded. Based on a personal observation during the sampling campaign, this may be attributed to waste burning in the incinerator around the sampling area, otherwise all other values were well correlated with $PM_{2.5}$. There was a significant difference across the seasons, the highest value was recorded during winter (June to August), which can be supported by some studies[5,13-15] that reported that the contributions of different sources to the PM, for instance biomass burning showed seasonal variation. In South Africa, the highest concentrations of PM_{10} and $PM_{2.5}$ are recorded in townships and informal settlements during winter, this is when domestic burning is most prevalent for heating and cooking, and when emissions are confined in a shallow boundary layer[16-19]. Soot and $PM_{2.5}$ showed a high correlated relationship with each other. The mean concentrations for weekdays and during weekends did not differ significantly from each other ($p > 0.05$).

4.7.2 Effect of meteorological conditions on ambient PM concentrations

Important to mention is the quality of the meteorological parameters obtained for the study from SAWS, i.e. not of good quality. As stated in the result section, Pretoria has a moderate subtropical climate. The effect of meteorological conditions significantly influences air quality. Temperature was categorised into three ranges to see if the concentration of $PM_{2.5}$ and other variables differed from what was reported in literature for seasonal variations. In this study, the mean temperature for winter was lower with increased amount of $PM_{2.5}$, while in summer (high temperature) low concentrations of $PM_{2.5}$ were recorded. This is supported by studies of Cavanagh *et. al*[20] and Tyson *et al*[21], who reported that air pollutants tend to increase with calm winds and stable atmospheric conditions while the concentration of air pollutant decreases with higher wind speeds and unstable atmospheric conditions. The sources of pollutants also play an important role in the concentration of ambient PM. The increased conventional uplift and turbulence facilitating the dilution and dispersion of pollutants as well as reduced domestic burning activities can reduce the concentration of pollutants during warm season[20, 22].

The spearman correlation showed that $PM_{2.5}$ concentration had significantly low negative correlation with ambient temperature for this study. Ambient PM is directly proportional to ambient temperatures, but most times negatively correlated with each other. Studies conducted in Japan[23] and China[24] respectively reported that temperature can either be positively or negatively correlated with ambient particulate matter depending on the geographical location, meteorological variable and other factors. A personal observation during campaign was that more inversions were observed during late autumn and winter which may also contribute to the high concentration of $PM_{2.5}$. When relative humidity and wind speed were categorised into three ranges, the highest concentration of $PM_{2.5}$ and OC were recorded when the relative humidity was lower and vice versa when the relative humidity was high.

The soot level was the same when the relative humidity were higher and lower. However, the highest concentration of $PM_{2.5}$ was recorded when the wind speed was moderate. This change could be due to the missing data for wind speed. Low temperature and wind speed can cause increased PM particles due to lack of dispersion, and high anthropogenic activities which includes biomass burning and space heating, these have been attributed to the high $PM_{2.5}$ concentration observed during the winter period.[25-27] Precipitation, wet surface during summer could be responsible for the lower concentration of $PM_{2.5}$ during summer. This reduces the re-

suspension of road and crustal dust which could have increased the concentration of PM in the atmosphere.[28-30]

4.7.3 Black carbon and organic carbon

In the entire study period, the mean concentration of BC was found to be higher that OC. Seasonally, the mean concentration for BC was highest during winter followed by autumn, spring and summer, the same trend was also observed with OC. The concentration of BC was higher than OC for days of the week while the highest was recorded on a Thursday. Also, no significant difference was found between weekdays and weekends for both BC and OC. This observation during weekdays and weekends can be attributed to the high traffic during weekdays. The monthly mean concentration also showed that the concentration for both BC and OC were highest mid-July before starting to decrease gradually until spring and summer. There exists a strong relationship between BC, OC and soot for the study period, and a strong correlation between each other.

Seasonal trends observed for BC and OC in this study is consistent with the findings of *Zhang et al.*[31] where the maximum concentration was observed during winter and lower concentration during summer. The seasonality observed may be governed by the variability in emission strength and meteorological parameters. For example, Yassaa *et.al.*[32], reported that low molecular weight semi-volatile organic compounds are mostly in gaseous phases at high temperatures in summer. This might also be responsible for the low concentration observed in summer during our study. The relatively higher concentration of BC and OC during spring compared to summer can be attributed to open biomass burning source which is seen with the high concentration of K during spring in our study. Our results can be supported by the findings of Cao *et al.*[33] who reported that aerosols from open biomass are generally characterised by elevated OC and BC.

4.7.4 Trace elemental composition

Most of the elements had the same seasonal trends with $PM_{2.5}$, i.e. the highest mean concentrations of these elements were recorded during winter followed by autumn, next to that is spring and then summer. The graph plots of the elements depict the effect of seasons on the availability of these element in the atmosphere. For weekly concentrations, it was observed

that the highest mean concentration was mostly on Thursdays, but the reason for this was not known.

Ca origin has been attributed to either soil or mineral dust while K may be from soil or biomass burning. The moderate correlation between Ca and K during autumn may suggest the source of K to come from biomass burning and soil dust. The correlation between K and Ca during spring may suggest the origin of K to be from soil, no correlation was observed during winter and summer. In addition, concentration of K appeared to be highest during winter, which could be due to biomass or coal burning for heating processes.[34]

Studies have shown that S is present in aerosols in the form of sulphate and is generally assigned to secondary aerosols[35]. Secondary aerosols are generated due to the reaction and transformation that occurs between the pollutants in the atmosphere. The high concentration of S in winter season (Table 4.5) in this study contrast with other studies that recorded highest value during summer when photochemical activities were highest.[36-37] High sulphate concentration has also been reported by Zhang *et al*[38] and Cesari *et al*[39] and high S concentration in $PM_{2.5}$ was reported by Molnar *et al*[40]. Two factors namely, local sources (e. g. domestic heating supplied by coal burning) and meteorological conditions may be considered to have influenced the high S concentration obtained during winter compared to what literature has reported.

Chloride (Cl) is attributed to sea salt in coastal cities[41] Since Pretoria is an inland city, and studies have shown from chemical analysis of $PM_{2.5}$ samples[42] and source profiles from the laboratory[43] that Cl is an elemental tracer for coal combustion. This study also reveals that the concentration was higher during autumn and winter which happens to be the cold period when coal is utilised for heating. The drastic drop in Cl concentration in spring and summer further confirms that its origin is from coal burning as less or none is being used during warm seasons for heating.

Pb concentration also showed a similar seasonal trend as Cl, i.e. high concentration during the cold season. The contributing factors may include coal and wood burning for heating process as it has been mentioned that Pb is present in wood[44] and lead is a by-product of coal ash. In South Africa, Pb is listed in the ambient air quality standards, hence, there is an annual average target of 0.5 $\mu g/m^3$. The annual $PM_{2.5}$ Pb concentration recorded (Table 4.4) was found to be below the targeted concentration and well below the WHO[45] annual guideline concentration

of 500 ng/m^3. The level was also found to be within the range of what was recorded in some western European cities such as Rotterdam, Netherlands[46], 8.9 ± 12 ng/m^3; Nice, France[47], 11.4 ± 5.2 ng/m^3, and higher than the value obtained in Gothenburg, Sweden[48] 3.0 ng/m^3. The low concentration of Pb recorded could probably be due to the phasing out of leaded gasoline in South Africa since 2006.

4.7.5 Transport clusters and potential source areas

The sources of air pollution reaching the sampling site originates from Gauteng province itself, other provinces such as Mpumalanga, Limpopo, Western Cape, and KwaZulu-Natal, and neighbouring countries such as Zimbabwe and Mozambique, which also contribute to the air pollution reaching South Africa. The sources of air pollution in these areas includes natural and anthropogenic sources, namely power generation activities, industrial processes, waste disposal, biomass burning, domestic fuel burning, agricultural activities, vehicle emissions, landfill sites, and mining activities. Five transport clusters per season were used to determine the origin of air masses reaching the sampling site. All the seasons showed similar patterns with varying trajectory percentages except for the winter season.

Hersey *et al*.[9] reported in their study that autumn and summer are characterised by air arriving from the east and southeast; during spring, air is transported from the north and east with some contribution from southwest and southeast, lastly, air in winter is directionally distributed in Gauteng. For the entire study, cluster 1 was associated with a high concentration of $PM_{2.5}$ originating from the direction of north Limpopo before arriving in the sampling area. The activities that contributes to the sources of air pollution in this area includes coal power stations, mining, domestic fuel burning, agriculture and veld fires. For example, Maenhaut *et al*[49] reported that wildfires represent significant sources of combustion related emissions within the province of Limpopo. This happens between the month of July and September, as this coincides with the dry season. Although cluster 2 has its root from the Indian Ocean with minimal marine influence, the direction of air parcels passes over the Mozambique region before reaching the sampling area. This region is associated with air pollution from biomass burning, waste burning, emissions from vehicles and industrial activities. This can be supported by the study of Hersey *et al*.[9] which reported that emissions from biomass burning in Mozambique and Zimbabwe reaches South Africa.

Clusters 3 and 4 are oceanic and associated with low concentration when compared to cluster 1, this may be due to the oceanic effect. The time air masses spend on the ocean may likely determine the amount of pollutants that will reach the sampling area. DEA[50] pointed that air parcels that reach South Africa include Indian, Atlantic and African plume among others, while Indian Ocean is reported to transport low concentration of pollutants. In addition, clusters 3 and 4 pass over the region of Kwazulu-Natal before reaching the site, this region is associated with pollution from industrial and traffic emissions, domestic burning and veld fires. Lastly, cluster 5 originates from the Indian Ocean but loses it marine influence as it travels over the Western Cape region before reaching the site. The Western Cape region's sources of pollution include, industrial activities, mining and quarrying, transportation (inland and water), veld fires waste disposal and agriculture. The length of the clusters also impacts on the amount of pollutants that reaches the site. The impact of local pollution in the province (i.e. Gauteng) will also impact the amount of pollutants reaching the site. The longer the time the air masses spend over the ocean, the less the amount of $PM_{2.5}$ that reaches the site, i.e. emission are less over the oceans when compared to the inland clusters. Also, the longer the length of the cluster, the lower the concentration of the $PM_{2.5}$ that reaches the site and vice versa.

Seasonally, during winter, clusters 1, 2 and 3 are known to be regional in origin (i.e. inter province). They differ in direction in which they travel and source contribution. For instance, cluster 1 passes over the region of North-west. The source of air pollution in this area includes vehicle emission, veld fires, mining and industrial activities, agriculture and landfill sites, while the sources of pollution for other regions (Kwazulu-Natal and Limpopo) have been discussed earlier. The high mean concentration of $PM_{2.5}$ recorded via these clusters can be attributed to the anthropogenic activities (Figure 4.25). Clusters 4 and 5, with low percentage of trajectories originate from the South Atlantic Ocean and Indian Ocean, respectively. These are regarded as LRT that are affected by meteorological parameters thereby reducing the amount of PM reaching the site. In autumn, cluster 2 has 51% of the trajectories originating from Mpumalanga region to the receptor site. The possibilities of the air masses from Mozambique and local sources of Gauteng cannot be overlooked as contributing sources to the pollutant reaching the site. Clusters 3 and 5 originate from the Indian Ocean and travel over the Kwazulu-Natal region.

The air masses for cluster 1 originates from the Eastern Cape Province. The source of pollution in this area includes biomass burning, waste burning, brick making, wood and coal use[51].

Summer and spring showed similarities with all the clusters showing all trajectories originating either from the South Atlantic Ocean or the Indian Ocean except for cluster 3 during summer, which has its origin from Limpopo, while clusters 2 and 3 originate from Mozambique and Western Cape respectively during spring. Cluster 1 for both summer and spring with 56% and 33% of the trajectories respectively originates from the Indian Ocean passing over Mozambique and Mpumalanga before reaching the receptor site, while clusters 4 and 5 for spring and 2 and 5 for summer also originate from the Indian Ocean passing over Durban and Swaziland to the receptor site. However, it should be noted that not all trajectories (e.g. cluster 1 and 5 in spring, 1 and 3 in summer) passed over industrial areas, but they can assist in transporting a number of pollutants from various sources, which in turn contributes to the concentration of the $PM_{2.5}$ observed during the campaign measurement.

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CHAPTER FIVE: POSITIVE MATRIX FACTORISATION MODELLING OF $PM_{2.5}$ SAMPLES IN PRETORIA

This chapter summarises the modelling process and results by PMF on the $PM_{2.5}$ samples collected in Pretoria using trace metals as markers. Five, six and seven factor models were run. In the end, seven factor model was chosen and the sources and their contributions to the total $PM_{2.5}$ were identified and quantified. These include vehicle exhaust/fossil fuel combustion – 22%, soil dust - 8%, secondary sulphur – 15%, vehicle exhaust – 14%, road traffic – 14%, coal burning - 18%. The percentage of $PM_{2.5}$ specie in base metal/pyrometallurgical was 9%. It is important to note that Sulphur, coal burning and emission from vehicle exhausts arises from fossil fuel combustion. However, some sulphur may be due to biomass burning. The sources identified in this study are consistent with what has been reported in the few available studies conducted in South Africa, for instance, Tshehla and Djolov.[1]

5.1 Model parameters setting

The chemical species concentration data and the corresponding uncertainty concentration data are essential for the PMF model to run. Preliminary data checks were done as well as the noise to signal ratio. The uncertainty is calculated using equation below[2],

$$\text{Uncertainty} = (0.05 * X_{ij}) + DL_{ij}$$

where X_{ij} and DL_{ij} are the concentration and detection limit of the j^{th} species in the i^{th} sample, respectively. The signal-to-noise ratio $(S/N)_j$ is interpreted as the relationship between a certain portion of the concentration that exceeds the uncertainty. The signal-to-noise ratio is one of the important references used to decide which species (or variable) should be included in the model run[3]. Most often the species with a signal-to-noise ratio below 0.2 were excluded and the species with the signal-to-noise ratio between 0.2 to ~2 were marked as weak. Therefore, 21 species were used in the model run including $PM_{2.5}$ mass, which is shown in Table 5.1. The species $PM_{2.5}$ mass is set to be a "Total Variable" and therefore given a higher uncertainty. It was automatically set to "Weak", because a total variable should not have a large influence on the solution.

Table 5.1: Input Data Statistics for positive matrix factorisation modelling

No	Species	Category	Signal-to noise ration
1	$PM_{2.5}$	Weak*	5.7
2	BC	Strong	4.7
3	OC	Strong	4.7
4	Si	Strong	9.0
5	S	Strong	9.0
6	Cl	Strong	8.7
7	K	Strong	9.0
8	Ca	Strong	8.5
9	Ti	Strong	8.8
10	V	Weak#	0.3
11	Fe	Strong	9.0
12	Ni	Strong	7.6
13	Cu	Strong	6.0
14	Zn	Strong	8.9
15	As	Weak	2.1
16	Se	Strong	3.8
17	Br	Strong	8.8
18	Sb	Strong	4.1
19	Ba	Strong	3.4
20	Pb	Weak	2.9
21	U	Strong	3.5

*$PM_{2.5}$ is set to be a "Total Variable" and therefore automatically set to "Weak".
#Vanadium (V) was used in run as test for fuel combustion. This was only observed in the 7 factors profile.

Following the setting of the signal to noise ratio, base model runs were carried out for the species. Base model runs can be defined as the process by which the primary factor profiles and factor contribution are produced[4]. Three to seven factors were used in the analysis to explore which factor had the most practical physical meaning. In this practice, the number of runs was set to 100 and the seed was set as 12 as the starting point. Later it was changed to "random" to assess if the solution found was local or global[4].

5.2 Sources of $PM_{2.5}$ identified by PMF

Pretoria is known to be an urban/industrial area of the country and it is likely to have source categories influenced by anthropogenic and industrial activities, which may include mineral dust. However, the $PM_{2.5}$ elemental composition dataset for the full year period is enough to apply the PMF techniques to determine the sources and their contribution to the total $PM_{2.5}$ mass. In this study, the researcher run the model with five to seven factors. The identified sources are shown in Figures 5.1 to 5.3 for the models, while the time series for the estimated daily contributions from each source to the $PM_{2.5}$ mass concentration for the seven factors model is shown in Figure 5.4. The sources and its relative contributions are presented in the following section.

5.3 Modelling results

In the PMF model run, the number of runs and seed number remained unchanged as well as the species selection and its corresponding signal to noise ratio. The factors were only changed to evaluate if more sources would be identified. The PMF model output for five, six and seven factors are summarised in Table 5.2. The seven factors used for apportionment are discussed below.

Table 5.2: Summary of positive matrix factorisation outputs for the different number of factors showing the probable sources and main markers

Factor	Number	Probably sources	Main marker/Analysed chemi
5	1	Diesel/vehicle exhaust	Sb, S, U, Pb
	2	Coal/biomass burning	As, Pb, Br, Cl
	3	Mineral dust	Si, K, Ca, Ti
		Soil, resuspended	
	4	Vehicle exhaust/fossil fuel combustion	Ba, Cu, Ca, Se
	5	Residual/diesel/road traffic	Ni, Fe
6	1	Mineral dust	Si, K, Ca, Ti
		Soil, resuspended dust	
	2	Coal burning/biomass burning	Br, As, K, Cl, BC, S
	3	Residual oil /diesel	S, Sb, U
	4	Residual oil/ domestic heating/industrial	Ni, Fe
	5	Vehicle exhaust/ road traffic	Cu, Zn
	6	Industrial/ road traffic	Cu, Se, Ba, Pb
7	1	Vehicle exhaust/ fossil fuel combustion	Cu, Se, Pb, U
	2	Mineral dust/ soil dust	S, Cl, K, Ca, Ti, Fe
	3	Secondary sulphur/ vehicle exhaust	S, BC
	4	Vehicle exhaust	Zn, Cl, Cu, BC
	5	Road traffic	V, Sb, Ba, Ca, Se
	6	Base metal/Pyrometallurgical	Ni, Fe, V, U
	7	Coal burning	BC, K, As, Pb, Cl

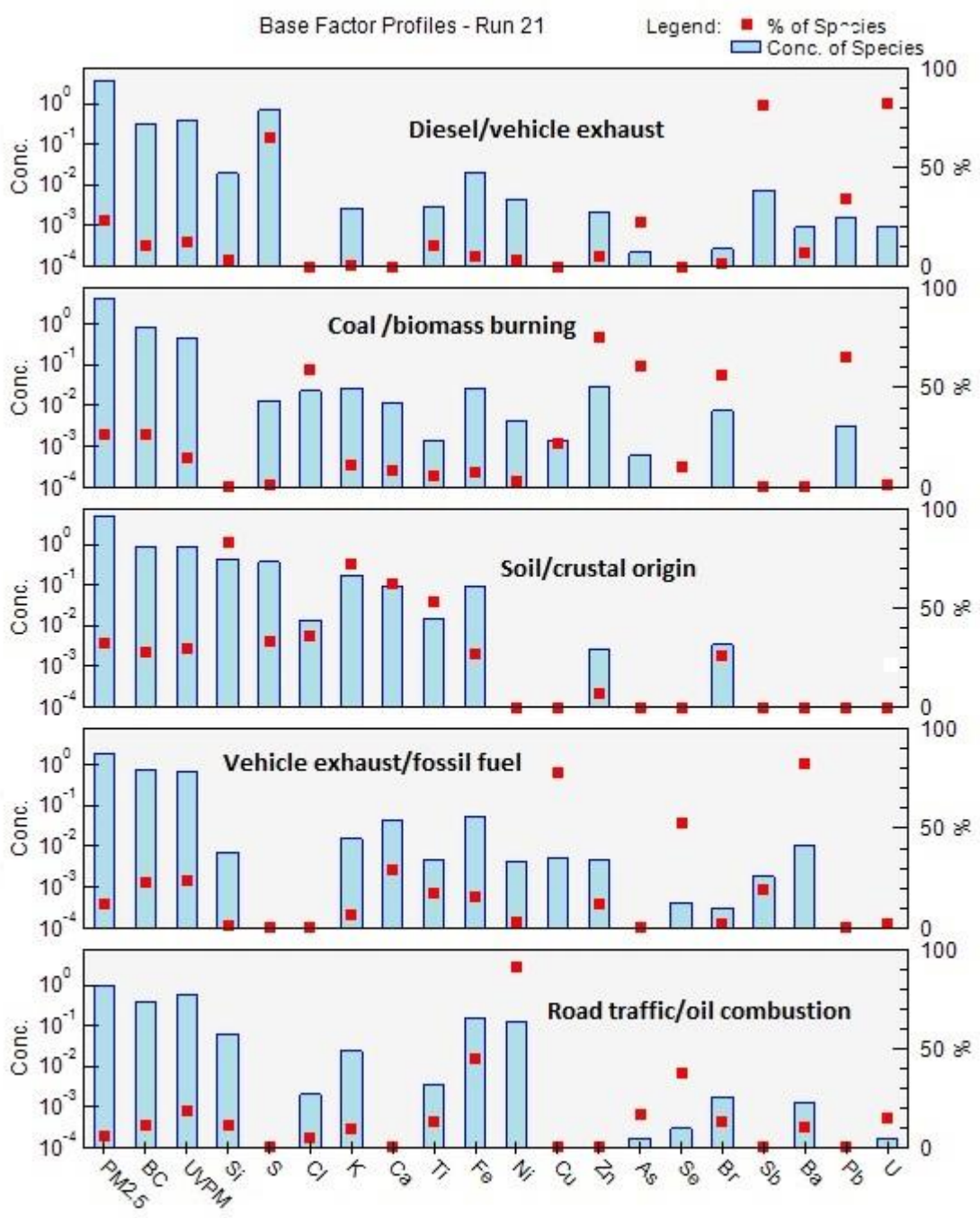

Figure 5.1: Positive matrix factorisation sources and mean contributions (concentrations are in µg/m^3) of $PM_{2.5}$ measured at the School of Health System and Public Health, University of Pretoria between 18 April 2017 and 17 April 2018 for five factors.

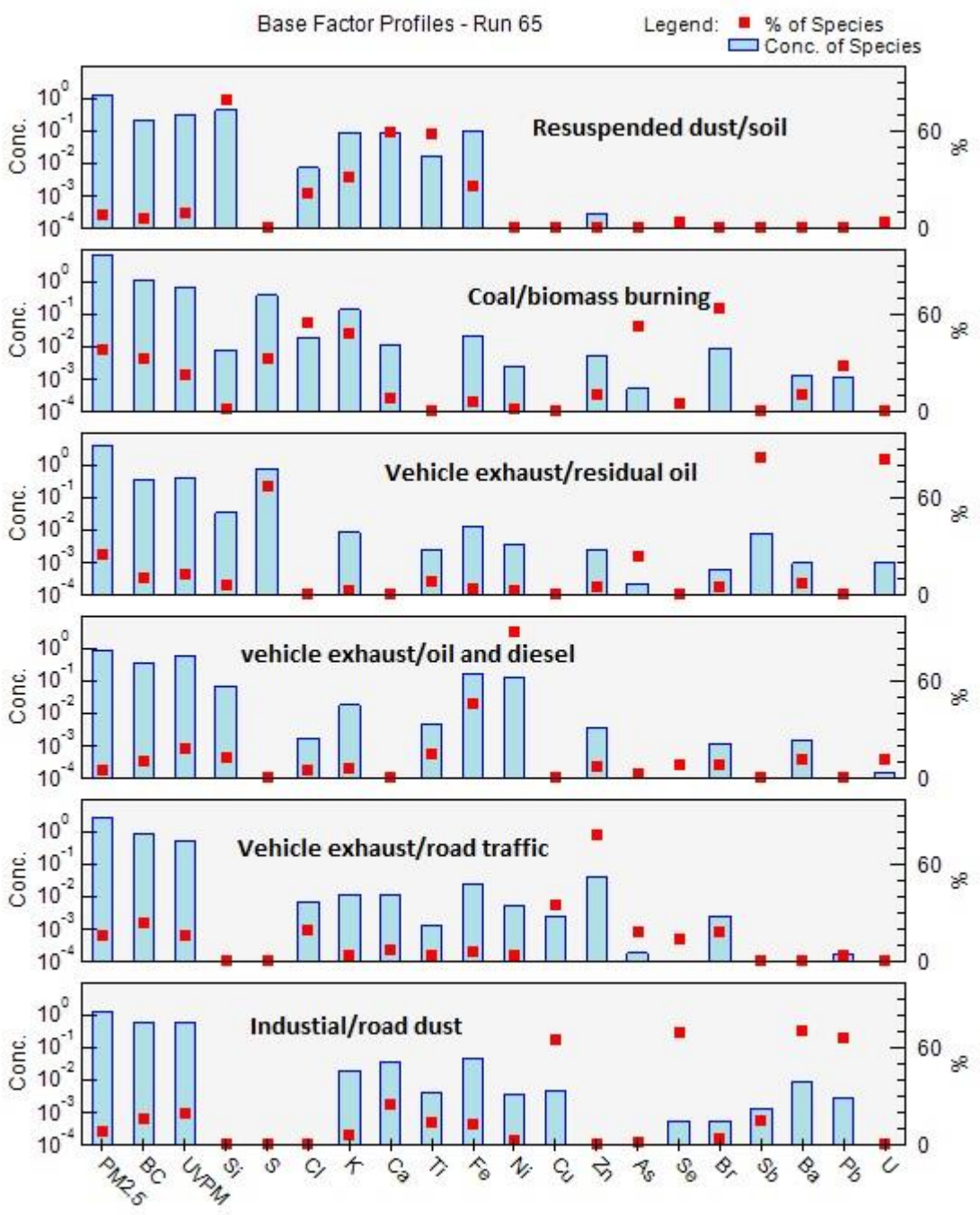

Figure 5.2: Positive matrix factorisation sources and mean contributions (concentrations are in µg/m^3) of $PM_{2.5}$ measured at the School of Health System and Public Health, University of Pretoria between 18 April 2017 and 17 April 2018 for six factors.

5.3.1 Seven factors

Factor 1 – Vehicle exhaust/fossil fuel combustion

This factor is characterised by Cu, Se, Pb and U. The presence of Se, Pb and U are present in this factor as fossil fuel combustion and account for 4.1 µg/m^3 (22%) of the mass concentration. Se and Pb with other elements (such as Ni and As) have been reported by Jayasekher[5] to increase in concentration near a coal-fired thermal plant when compared with an urban background.

Our findings also revealed increased concentration during winter which can be attributed to the use of coal for heating, this is consistent with the findings of Wåhlin *et al.*[6] and U has been found to be a by-product of coal ash. However, the presence of Cu and Pb in this source could be partly attributed to vehicle exhaust. Possible source of lead in this factor may be from wear (tires, brake linings, clutch) rather than fuel since lead has been banned in fuel since 2006. This situation has also been reported by Smichowski *et al.*[7], who reported that vehicular Pb emission originates mainly from wear rather than fuel combustion, hence this source can be termed as vehicle exhaust/fossil fuel combustion.

Factor 2 – Soil dust

The soil dust usually represents wind-blown dust from the surrounding areas. This factor is characterised by Si, Cl, K, Ca, Ti and Fe. This factor accounted for 75, 32, 45, 64, 57 and 27% of these elements respectively, hence this factor is categorised as soil dust. The contribution of this source to $PM_{2.5}$ was 1.4 µg/m^3 or 8% on annual average. This finding is consistent with the study by Tshehla,and Djolov[1] and Zíková *et al.*[8] where mineral dust which includes soil, and road re-suspended dust is characterised by the presence of Al, Si, Ti and Ca. Gaita *et al.*[9] reported that the absence of S, Zn and Pb in this factor differentiates the mineral dust from the re-suspended road dust. The study of Van Zyl *et. Al*[10]. identified the presence of K and Fe as metal species that are considered to be from windblown dust, therefore, indicative of crustal factor.

Factor 3 - Secondary sulphur

In this study, this factor was found to contribute 2.7 µg/m^3 (15%) of total $PM_{2.5}$ mass concentrations. This factor from the PMF analysis is mainly characterised by the single element S (80%) with traces of BC (17%). Other trace elements found in this factor, i.e. secondary sulphur or exhaust traffic emission were due to noise-signal ratio in the analysis. Secondary sulphur is in line with some studies[11-13] that identified this factor in most urban PMF studies.

In addition, the presence of S and BC has been identified as exhaust traffic emissions by different studies[14-15] but in these studies leaded petrol was still in use. Recent findings indicate that sulphate concentrations over Southern Africa, previously attributed only to South African industrial emissions, have a contribution from the Zambian Copperbelt.[16]

Factor 4 – Vehicle exhaust

This factor is mainly characterised by Zn, Cl and Cu with a small amount of BC (22%). This is a mixed source of vehicle exhaust and industry emissions which contributes 2.5 μg/m^3, or 14% of $PM_{2.5}$ on an annual basis. It is important to know that Cu and Zn are major additives to lubricating oils. Carbonaceous species are generated from the vehicle exhaust tail pipe while the presence of Cu may be produced from both fuel/lubricant combustion and brake abrasions.[17] The presence of Cl in this factor may originate from coal burning used in the industrial activities[18]. Hence this factor can also be termed vehicular/industrial emissions.

Factor 5 – Road traffic

This source category comprises various kinds of emission deriving from different vehicle types and associated processes. The Road traffic represented 14% of the total $PM_{2.5}$ mass. This source is characterised by the presence of V, Sb, Ba, Ca and Se**.** Ba, among other elements (i.e. Cu and Zn), have been reported to be associated with tail pipe emissions, brake and tyre wear and Sb is known as a tracer of brake wear dusts[19-20]

Factor 6 – Base metal/ pyrometallurgical-related factors

This factor is characterised by Ni, Fe, V and U (9%) which are attributed to a mixture of pyrometallurgical-related factors and base metal processing. The trace elements identified in this factor except for Cr is similar to the study by Venter *et al*.[21] where it was identified as pyrometallurgical-related factors. Fe and Cr are associated with ferrochromium smelters in the Bushveld Igneous Complex, while Ni was attributed to base metal smelters that refines base metals from the PGM production processes. The presence of Ni (88%) and V (33%) in this factor agrees with previous studies, which reported that Ni and V were used as markers for residual oil combustion[22]. However, the combination of these metals can be traced to metal processing origin. Wang *et al*.[23] also described a similar factor in the source apportionment of $PM_{2.5}$ in Beijing.

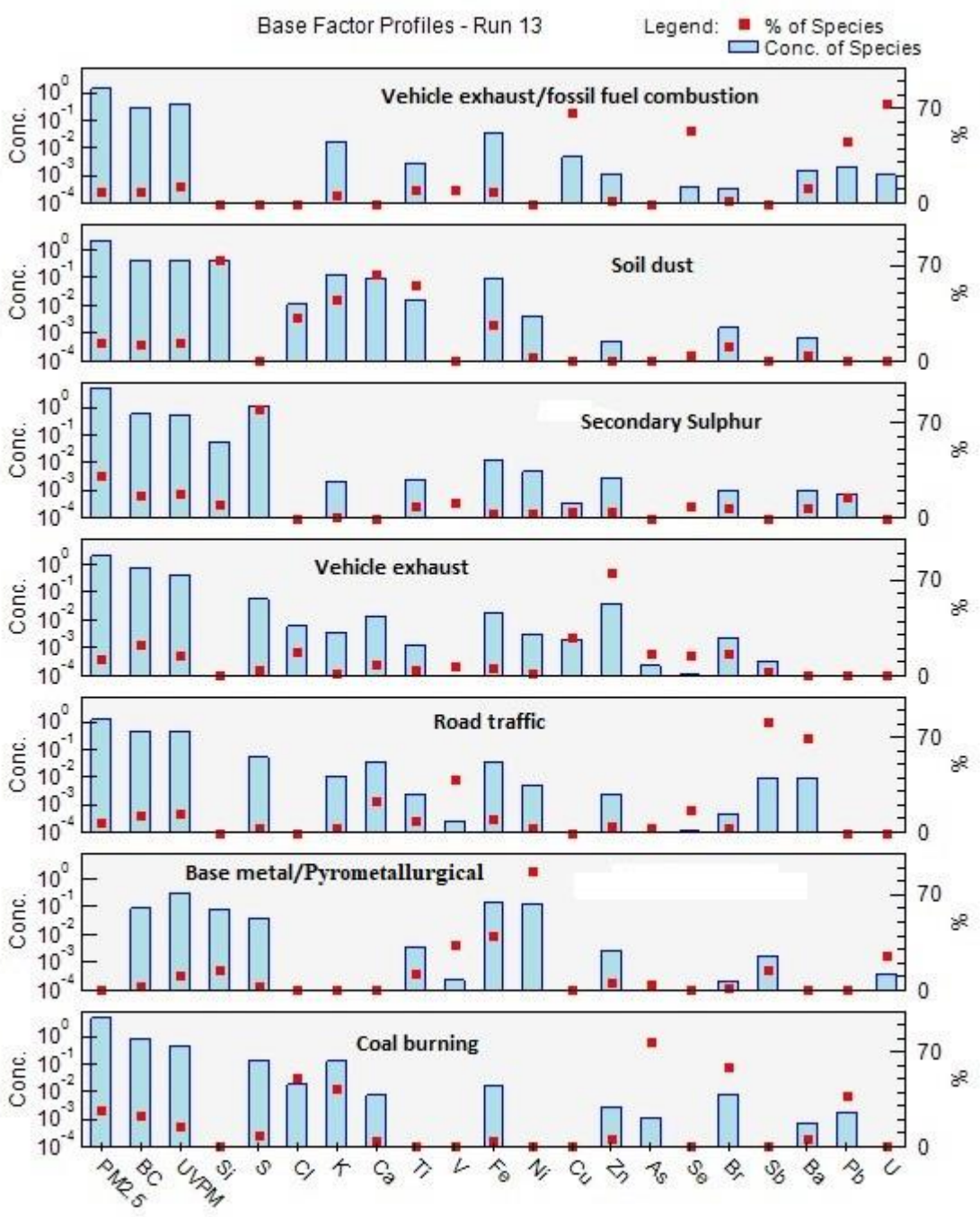

Figure 5.3: Positive matrix factorisation sources and mean contributions (concentrations are in µg/m^3) of $PM_{2.5}$ measured at the School of Health System and Public Health, University of Pretoria between 18 April 2017 and 17 April 2018 for seven factors.

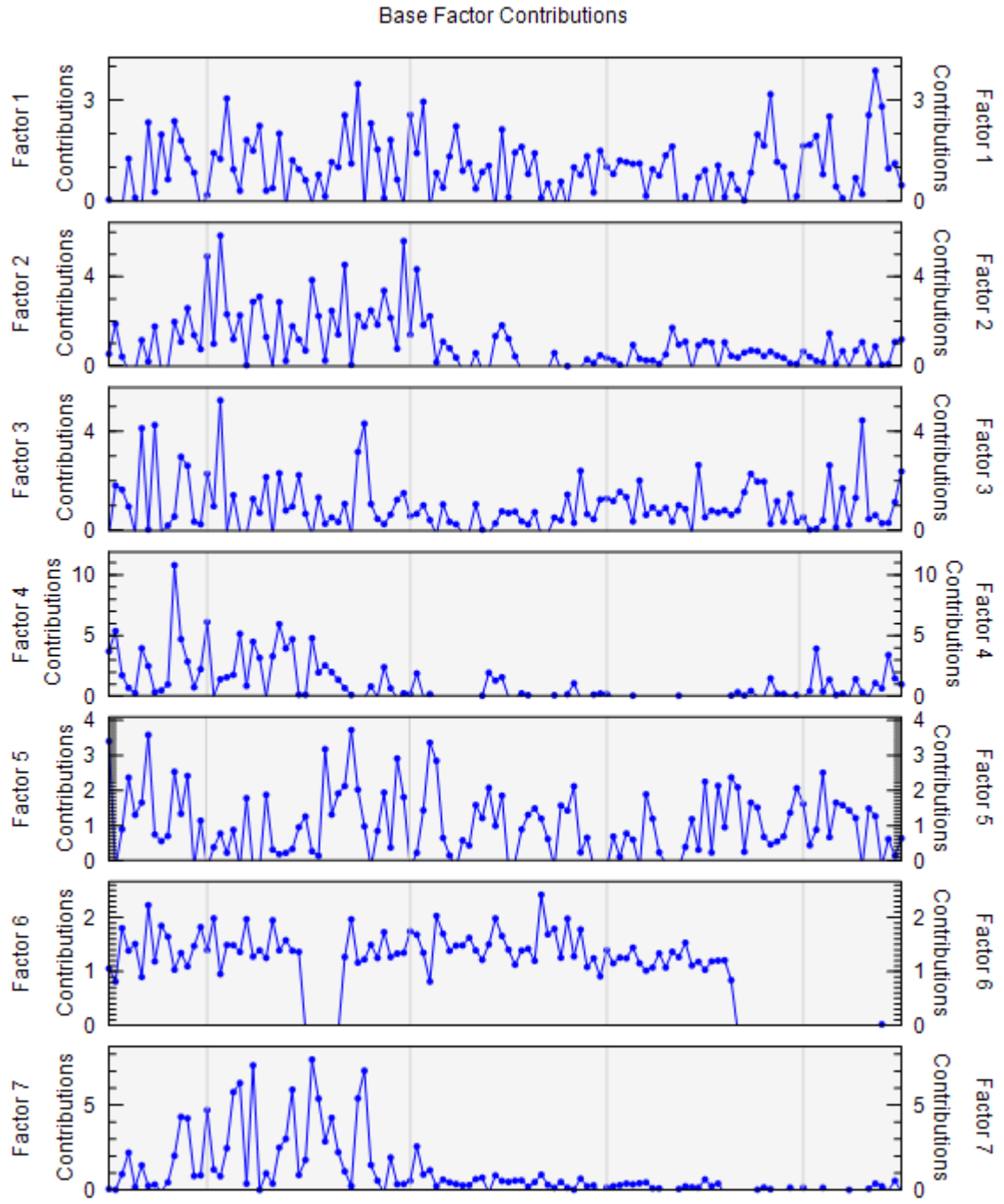

Figure 5.4: Positive matrix factorisation factor contributions (concentrations are in µg/m^3) of $PM_{2.5}$ measured at the School of Health System and Public Health, University of Pretoria between 18 April 2017 and 17 April 2018.

Factor 7- Coal burning

Coal burning also contributed significant amount (3.3µg/m^3 or 18%) to the total $PM_{2.5}$ mass. This factor is mainly characterised by BC, K, As, Br, Pb and Cl and can be categorized as coal burning. The amount of S present in this factor also helps in the identification of the factor as coal source. The presence of Cl serves as an elemental marker for coal combustion. The presence of As and Pb in this source is consistent with the finding of Wåhlin *et al*.[6] who reports that As, Se and Pb are typical anthropogenic sources of coal combustion.

As stated above the PMF model generated seven sources which were identified and shown in Figure 5.3, with information on mass contribution and percentage of the variables within each source. The mean $PM_{2.5}$ concentration of the seven sources can be found in Table 5.3 (Vehicle/Fossil fuel, Soil dust, Secondary Sulphur, Vehicle exhaust, Road traffic, Base metal/pyrometallurgical, Coal burning) were 4.1, 1.4, 2.7, 2.5, 2.6, 1.7 and 3.3µg/m^3 respectively (Figure 5.5). Vehicle/fossil fuel was found to be the major contributor with 22% of elemental mass in $PM_{2.5}$ concentration on a yearly average. This is followed by coal burning which accounted for 18% of the total $PM_{2.5}$ mass. The sum of soil dust and road traffic adds up to a mean contribution of 4.0 µg/m^3 (22% of the total $PM_{2.5}$).

5.4 Seasonal and weekly variations of $PM_{2.5}$ sources identified from the PMF model

The $PM_{2.5}$ sources were found to have a seasonal behaviour, as shown in Table 5.3. In winter, coal burning, and secondary sulphur were found significantly higher compared to other seasons (Figure 5.6). In autumn, $PM_{2.5}$ concentration from soil dust and road traffic factor were higher than other seasons, as was vehicle/ fossil fuel combustion.

The increased contributions from coal burning during winter were expected since residential heating and power generation would increase the emissions, also the increased sulphur observed during winter can be attributed to the soot from the exhaust pipes of vehicles using diesel or power plants using diesel as a source of fuel. The high concentration of soil dust and road traffic dust observed during the autumn could be due to the elevated wind speed that occurs during the season.

Table 5.3: Descriptive statistics of $PM_{2.5}$ mass, explained mass from the positive matrix factorisation model, and estimated source contributions for the seven sources. (All concentrations in µg/m^3)

species	Full study		Autumn		Winter		Spring		Summer	
	Mean	**Range**	**Mean**	**Range**	**Mean**	**Range**	**Mean**	**Range**	**Mean**	**Range**
$PM_{2.5}$	21.1	0.7-66.8	23.4	3.4-57.9	35.5	14.6-66.8	14.3	1.4-35.4	10.7	0.7-21.6
Vehicle/fossil fuel	4.1	0-4.7	6.3	5.1-8.2	4.3	0-14.6	2.0	0-9.1	3.7	2.4-5.1
Soil dust	1.4	0.1-4.0	2.8	0.6-3.3	1.1	0-10.1	1.0	0-3.4	0.8	0-2.5
Secondary sulphur	2.7	0.3-6.5	0.8	0-4.9	8.1	0.6-13.5	1.2	0-4.9	0.5	0-2.2
Vehicle exhaust	2.5	0-5.9	3.1	0-4.4	3.9	0-12.0	1.4	0-3.7	1.6	0.1-3.8
Road traffic	2.6	0-4.7	4.2	0-5.8	3.3	0-9.1	1.8	0-4.2	0.9	0.2-1.4
Base metal/Pyrometallurgical	1.7	0-3.5	1.5	0-4.1	2.8	0-9.4	1.4	0-6.7	0.9	0-3.0
Coal burning	3.3	0.6-6.0	2.2	0.1-5.4	8.9	1.4-14.5	0.8	0-5.1	1.41	0.2-3.5

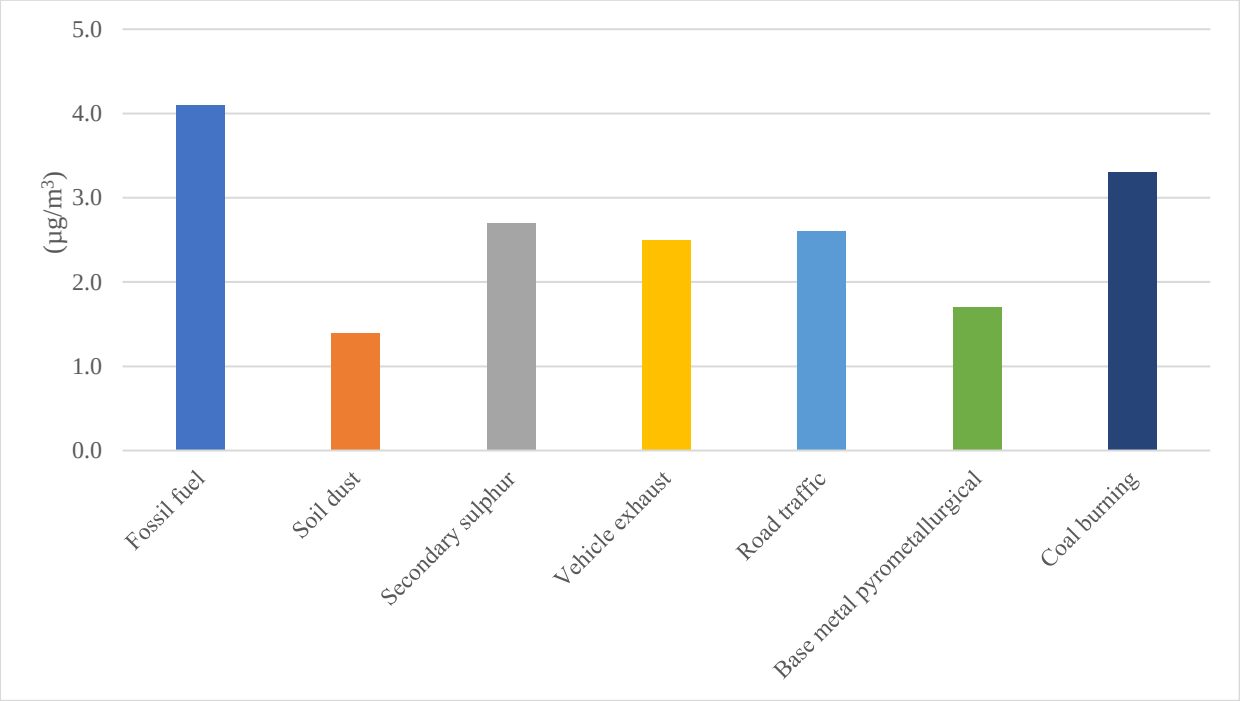

Figure 5.5: Mean source contributions from the seven-factor positive matrix factorisation model to the yearly mean $PM_{2.5}$ concentration

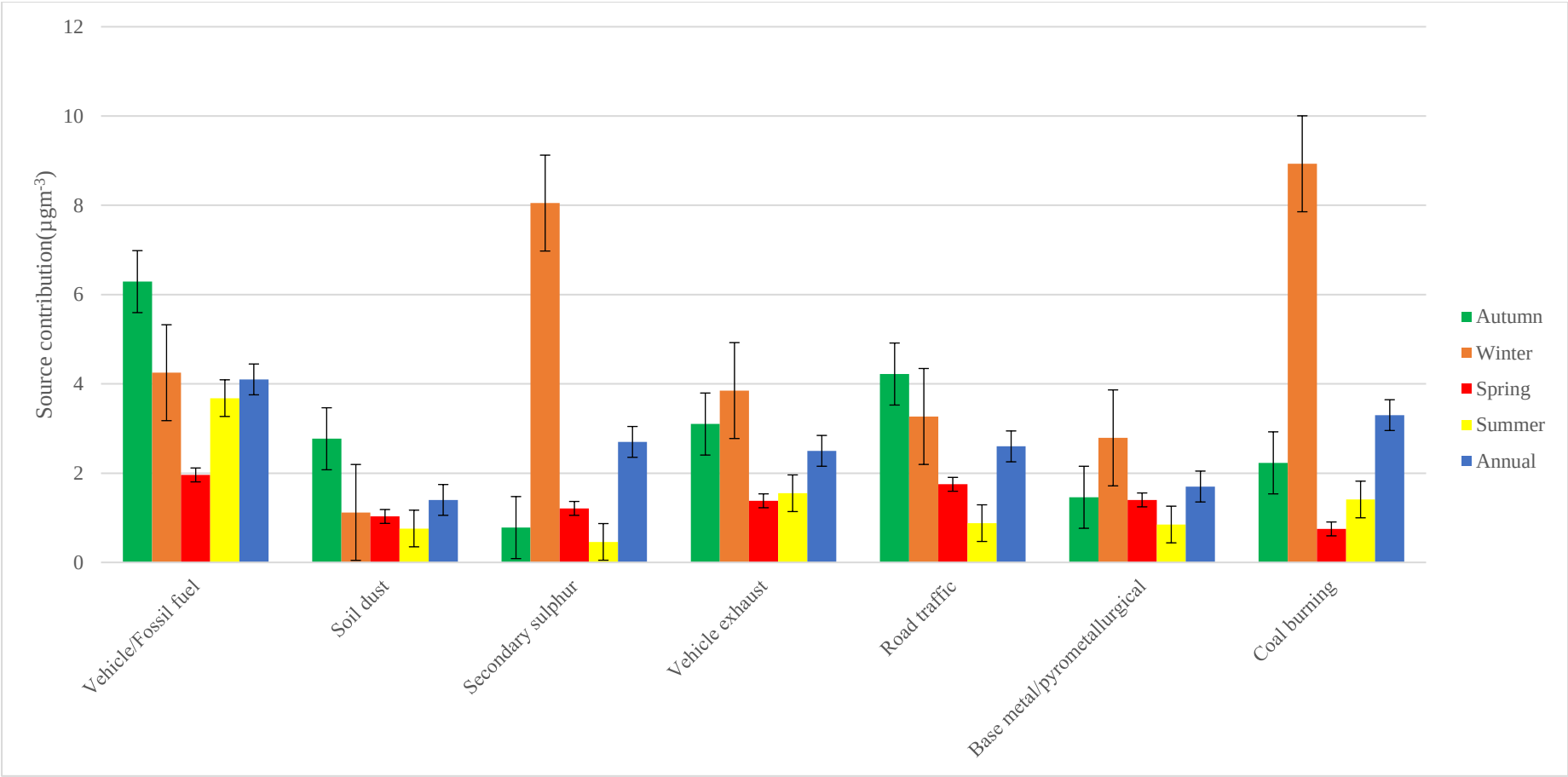

Figure 5.6: Seasonal mean concentration of the source contribution from 18 April 2017 and 17 April 2018 in Pretoria

5.5 Transport clusters and potential source areas

Five transport cluster pathways have been identified in this study. However, these pathways are not accurately following a particular direction. Long-range transport (LRT) and local sources characterised these pathways. (Figure 4.16). Cluster 1 (North-LP) is a local source and Cluster 2 (Eastern Inland) has a transboundary source origin while, 3, 4 and 5 (Short-Indian Ocean, Long Indian Ocean and SW-Inland) depicts LRT sources. For instance, the highest mean level for $PM_{2.5}$ (66.8 µg/m^3) that occurred on June 2, 2017 during this study was found to be from a local source via North-LP. Furthermore, about 78% of the days in which the WHO and SA standard were exceeded were of local source origin (cluster 1: North-LP; cluster 2: EI), while only 22% was attributed to LRT sources. The annual frequencies of the five transport clusters show that clusters 3, 4 and 5 were of long-range transport, with 11%, 4% and 6% respectively.

5.6 Source apportionment of $PM_{2.5}$ for each cluster

The combination of seven sources from the PMF model and the five transport clusters present the opportunity to compare the source strength and the different transport clusters. This is shown in Table 5.4 and Figure 5.7. Vehicle exhaust/fossil fuel source was the largest contributor irrespective of their cluster followed by coal burning and secondary sulphur. The Base metal/pyrometallurgical related factors had the highest concentration when their air masses originated nationally from Limpopo to the receptor site (cluster one; North-LP) followed by North-Easterly Indian Ocean (cluster 2; EI) and South-Westerly Atlantic Ocean (cluster 5; SW-Inland) respectively. Short-Indian Ocean (cluster 3) and South-Easterly Indian ocean (cluster 4; Long-Indian Ocean) were the lowest. The vehicle exhaust/fossil fuel peaked when the air masses originated from North-LP and cluster 3; Short-Indian Ocean which are both National and LRT.

Secondary sulphur also recorded the highest concentration when the air masses came from North-LP followed by Short and Long-Indian Ocean (clusters 3 and 4), with SW-Inland being the lowest. Soil dust was found in all transport clusters, with the highest concentration in North-LP and the lowest in NE and SE-Indian ocean. Coal burning also had a significant contribution when air masses originated from North-LP, SW-inland, Long-Indian Ocean (cluster 4), East-Indian ocean and North-East Indian Ocean respectively. Road traffic and vehicle exhaust exhibit their highest concentration when the air masses originated from SW-Inland and the

lowest concentrations were recorded in Short and Long-Indian Ocean clusters 3 and 4 respectively

5.7 $PM_{2.5}$ sources and transport clusters

In a study by Molnar *et al*[24], it was stated that the average size of the impact from different source areas and source types at a receptor point depends on the combination of source strength and frequency. The highest mean concentration recorded during winter (Figure 4.17), in this study, shows that the air masses originated form the high-polluted areas where mining and smelting process were predominant, therefore making them potential transporter of heavy metals to the receptor site. This accounts for 45% of the transport clusters bringing in pollutants to the sampling site. During autumn clusters 1 and 2 accounted for 81% of the local transport clusters responsible for the pollutants arriving at the sampling site. This is also responsible for the elevated mean concentration of $PM_{2.5}$ during the season (Figure 4.17). Other transport clusters may be classified as regional and long-range transport clusters. It should be noted that some of these trajectories may not pass over the region where industrial or mining is prominent, although they can be a potential source of these pollutants as a result of recirculation over the Southern Africa. The process whereby air is transported away from one point of origin and back in opposite direction from which it has travelled after recurved or rotated cyclonically or anticyclonically can be termed as recirculation

Table 5.4: Mean concentrations for $PM_{2.5}$ and for the five estimated positive matrix factorisation sources by the six different transport clusters in ($\mu g/m^3$)

Transport cluster	$PM_{2.5}$	Vehicle exhaust/ fossil fuel	Soil dust	Secondary Sulphur	Vehicle exhaust	Road traffic	Fossil fuel	Coal burning
NLP (31 days)	25.0	6.4	4.1	3.7	1.2	2.0	7.0	4.3
EI (40 days)	14.9	2.3	1.0	3.3	1.8	2.8	2.1	1.6
SIO (41 days)	26.2	2.7	1.0	2.1	0.5	1.2	0.7	1.6
LIO (4 days)	9.8	0.3	2.6	3.5	0.9	0.1	0.6	2.1
SWI (6 days)	15.7	2.2	2.7	1.3	2.2	3.2	1.5	2.5

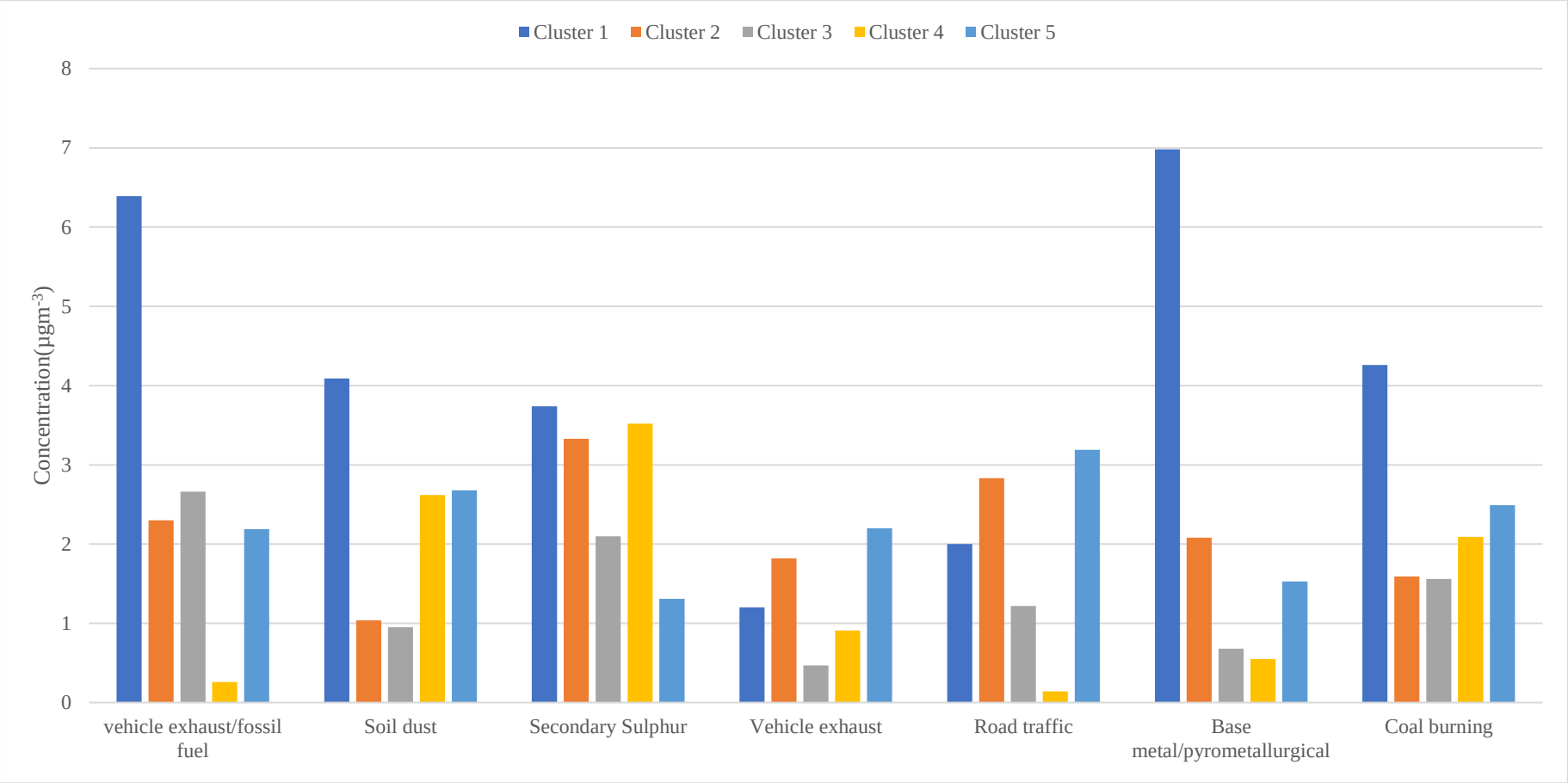

Figure 5.7: Mean contribution to the $PM_{2.5}$ concentration per source and transport cluster from 18 April 2017 to 17 April 2018 at the School of Health System and Public Health, University of Pretoria.

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CHAPTER SIX: ASSOCIATION BETWEEN AIR POLLUTION AND RESPIRATORY DISEASE HOSPITAL ADMISSIONS IN PRETORIA, SOUTH AFRICA

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6.1 Abstract

***Background*.** Air pollution is one of the major problems being faced by most of the big and industrial cities of the world and has become a major environmental threat over the last few years. This environmental threat has gained more attention because of its increased health effects on human which includes morbidity and mortality and have various adverse effects, such as increased pulmonary infections, respiratory diseases, acute illnesses and hospitalizations, and can eventually lead to death.

Objective. The aim of the present study is to determine the association between respiratory disease (RD), hospital admissions and air pollutants (PM_{10}, NO_2 and SO_2) in Pretoria, South Africa.

Method. Individual-level RD hospital admission data (International Classification of Disease, 10th version [ICD-10] (J00–J99) were obtained from a private hospital group following ethical approval. Data from 1 January 2011 to 30 November 2014 were electronically available from three hospitals located in Pretoria, South Africa. The total population of the study group was 27480. The time-stratified case-crossover epidemiology study design and conditional logistic regression models were applied to investigate the association between PM_{10}, NO_2 and SO_2 and respiratory disease (RD) hospital admissions during the study period.

***Results*.** The study included 17 647 RD hospital admissions. Daily PM_{10} levels exceeded the daily WHO air quality guideline (50 μg/m^3) on 662 days. Mean apparent temperature (Tapp) was 16.6°C; PM_{10}, NO_2 and SO_2 had negative correlation with Tapp, temperature and relative humidity. In the study period, a 10 μg/m^3 increase in PM_{10} was associated with an increase of

[1] Manuscript was first submitted to a journal and reviewed by two reviewers. Feedback was provided on 13 July 2019. Manuscript was resubmitted on 31 July 2019

0.2% (-0.7%; 1.2%) in RD hospital admissions. A 10 μg/m^3 increase in NO_2 increased RD hospital admissions by 19.0% (2.3; 38.4) on warm days, while SO_2 showed no clear effects.

Conclusion. Overall, exposure to ambient PM_{10} and NO_2 levels in Pretoria is an important risk factor of RD hospital admissions, predominantly on warm days, among young children and elderly.
Keywords: Air pollution, particulate matter, hospital admission, case-crossover, respiratory diseases, Pretoria, South Africa, warm days, cold days

6.2.1 Introduction

Respiratory diseases are documented as one of the leading causes of both morbidity and mortality globally[1]. In a report by the World Health Organization in 2014, it was estimated that over 7 million people die due to air pollution every year, i.e. one in every eight deaths globally, which is far more than the combined deaths recorded as a result of HIV, tuberculosis and malaria. In lieu of the above, air pollution has been flagged as a major risk factor in Africa.[2-3] Air pollution has been demonstrated to be linked with potential effects on human health, weather and climate, including elevated mortality hazard, increased rates of emergency department visits and hospital admissions, exacerbated chronic respiratory conditions (e.g., COPD and asthma), deteriorated lung function and changed climate[4]. The effects of air pollution on human health have been studied, both acute and long term, and reported to include bronchoconstriction and increased asthma symptoms, and chronic effects including chronic lung disease and premature mortality.[5-7]

In recent times, many epidemiological studies have investigated the association between daily variations in temperature and air pollution with respiratory disease hospital admissions and mortality in different parts of the world including North America,[8-11] Europe,[12-15] Australia,[16-17] some Asian studies including China, Taiwan, India, Iran and Vietnam[18-26] and Africa.[27] These studies have established an increase in mortality and/or hospital admissions for cardiovascular and respiratory diseases due to short- and long-term exposure to air pollutants. However, the problem of air pollution is likely to persist, even when pollutant levels are within the standards required by legislation, as the world continues to experience rapid urbanisation and industrialisation. Among the ambient pollutants studied widely, particulate matter has been found to have multiple effects on human health.

Particulate matter (PM), or respirable particles of concern, include PM_{10} (coarse particle with aerodynamic diameter from 2.5 - 10µm) and fine particles with aerodynamic diameter <2.5µm ($PM_{2.5}$) and 1µm (PM_1).[28] Other common environmental pollutants that have been reported to increase mortality and hospital admissions include nitrogen dioxide (NO_2), sulphur dioxide (SO_2), carbon monoxide (CO) and ozone (O_3).[29,6] Continued attention has been given to the health consequences of NO_2, which is an alternate for traffic-sourced air pollutants and one of the crucial environmental pollutants.[30] Regarding the short-term effects, the association between NO_2 and health effects remains in many studies after adjusting for other pollutants.

In a recent study by DeVries *et al*[31] they found a causal relation between NO_2 short-term exposure and respiratory effects. Mehta *et al*[32] assessed the effects of exposure to PM_{10} and other pollutants, such as NO_2, SO_2 and O_3, on acute lower respiratory infection hospital admissions among children aged <5 years. In that study, PM_{10} was associated with increased acute lower respiratory infection hospital admissions in the dry season, but its effects could not be distinguished from that of NO_2 due to their high correlation. However, exposure to environmental NO_2, PM_{10} and SO_2 has in recent times been linked with the worsening of symptoms and lung function degeneration during asthma exacerbations.

This study addresses the United Nations Sustainable Development Goals (SGDs) (valid 2016-2030),[33] specifically Goal 3.9, "By 2030, substantially reduce the number of deaths and illnesses from hazardous chemicals and air, water and soil pollution and contamination." In South Africa generally, studies investigating the associations between air pollutants and respiratory disease hospital admission are scarce. Therefore, this study aimed to examine the associations between air pollutants (PM_{10}, NO_2 and SO_2) and respiratory disease hospital admission in Pretoria, South Africa, over the period January 2011 to November 2014.

6.2.2 Material and methods

Ethical approval (reference 469/2017) was obtained from the Research Ethics Committee, Faculty of Health Sciences, University of Pretoria in 2017. All data were retrieved after signing data agreements with the relevant agencies. (Appendix 3-5)

6.2.2.1 Study design

The associations were investigated with the time-stratified case-crossover epidemiological design[34] The case-crossover epidemiological study design was developed as a variant of the case-control design to study the effects of transient exposures on short-term health outcomes,

comparing each person's exposure in a time-period just prior to a case-defining event with the person's exposure at other times. If the control days are chosen close to the event day, personal characteristics that vary slowly over a short period of 24 h are controlled by matching. Such characteristics may include co-morbidities (e.g. HIV status, hypertension, smoking status and so forth), nevertheless they may be potential effect modifiers, i.e. indicate susceptibility. However, information on such characteristics is not provided by hospitals. Air pollution studies using case-crossover designs allow the use of routinely monitored air pollution data and concurrently makes it possible to study the individuals rather than days as the unit of observation

The time-stratified approach was applied to select the control days, defining the day of respiratory disease hospital admission as the case day and same day of the week in the same month and year as control days[34] Figure 1 displays the map of the City of Tshwane (name for the municipality of Pretoria) showing the air pollution monitoring stations and private hospitals in Pretoria.

6.2.2.2 Hospital admission data

Individual-level respiratory disease hospital admission data (International Classification of Disease, 10th version [ICD-10] (J00–J99) and J18 were obtained from a private hospital group following ethical approval. Data from 1 January 2011 to 30 November 2014 were electronically available from three hospitals located in Pretoria, South Africa (See Appendix 6). The total population of the study group was 27 480 but one of the hospitals only opened after 31 December 2104, therefore excluding the population from the hospital and leaving us with a population of 17 647

6.2.2.3 Air pollution and weather data

Pretoria has a network of eight sites that continuously monitor air pollutants similar to the United States Environmental Protection Agency and in accordance with ISO 17025 guidelines (National Environmental Management: Air Quality Act, 2004). Air pollution data are stored in the South African Air Quality Information System, which is managed by the South African Weather Service (SAWS). Hourly air pollution data (PM_{10}, NO_2, SO_2) between 2011 and 2014 were obtained from the SAAQIS; The choice of the pollutants used were dependent on the missingness of the data obtained Temperature (°C) and relative humidity (%) data were obtained from the SAWS, and used to calculate apparent temperature (Tapp), which reflects

the physiological experience of combined humidity and temperature and is better able to capture a response on health[35]. Tapp equation is shown below while the monitoring stations are shown in Table 6.1 and Figure 6.1

Saturation vapour pressure

6.112 x 10 $^{(7.5 \text{ x temperature oC / (237.7 + temperature oC)}}$

Actual vapour pressure

= (relative humidity (%) × saturation vapour pressure)/100

Dew point temperature °C

= (−430.22 + 237.7 × ln (actual vapor pressure))/−ln (actual vapor pressure) + 19.08)

Apparent temperature °C

= −2.653 + (0.994 × temperature °C) + 0.0153 × (dew point temperature °C)

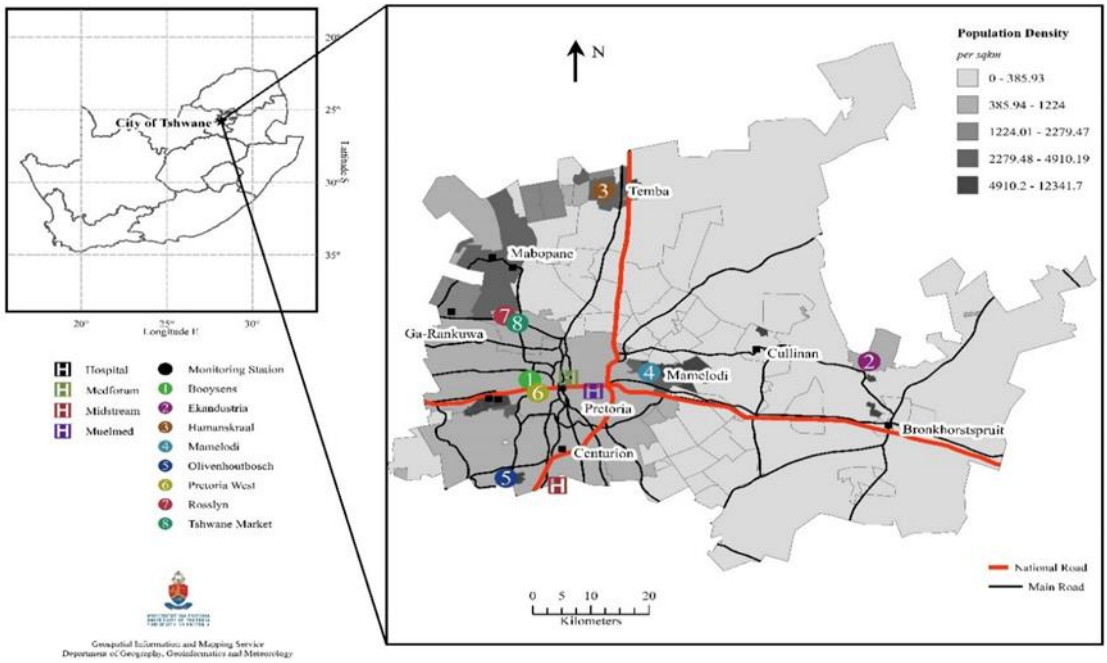


Figure 6.1: Map of City of Tshwane showing the air pollution monitoring stations and private hospitals in Pretoria.

Table 6.1: Classification and year of establishment of air pollution monitoring stations is the City of Tshwane

Station	Suburb	Classification	Coordinates	Year of Establishment	Colour code	Pollutants measured
Bodibeng	Soshanguve	Residential & traffic	25º 29’34,155” S 28º5’37,495” E	2011	Brown	SO_2, NO_2, PM_{10}, CO O_3, NO, NOx, $PM_{2.5}$
Booysens	Claremont	Residential & traffic	25°42'49,205"S 28°07'55,539" E	2009	Green	SO_2, NO_2, PM_{10}, CO, NO, NOx
Ekandustria	Bronkhorspruit	Industrial	25°41'23,617"S 28°42'47.800"E	2012	Purple	SO_2, NO, NO_2, NOx PM_{10} CO, O_3, $PM_{2.5}$
Mamelodi	Mamelodi	Residential & traffic	25°43'00,408"S 28°20'11"700 E	2009	Light blue	SO_2, NO, NO_2, NOx PM_{10} CO, O_3,
Olievenhoutbosch	Centurion West	Residential	25°54'42,035"S 28°5'34,638"E	2009	Dark blue	SO_2, NO, NO_2, NOx PM_{10} CO, O_3, $PM_{2.5}$, PM coarse
Pretoria West	Pretoria West	Industrial & traffic	25°45'19,611"S 28°8'45,922"E	2005	Lemon green	SO_2, NO, NO_2, NOx PM_{10} CO, O_3, $PM_{2.5}$,
Tshwane Market	Pretoria West	Industrial & traffic	25˚44’23,612” S 28˚9’57,773” E	2014	Green	SO_2, NO, NO_2, NOx PM_{10} CO, O_3, $PM_{2.5}$, H_2S
Rosslyn	Pretoria North	Industrial	25°37'30,528"S 28°5'41,089"E	2005	Red	SO_2, NO, NO_2, NOx PM_{10} CO, O_3, $PM_{2.5}$, PM coarse

Confounder data

Daily temperature (^{o}C) and relative humidity (%) are the 24 h changing variables obtained from the South African Weather Services (SAWS) from the air pollution monitoring site for the period 2011 to 2014. In the case-crossover epidemiological study design, only variables that change over a 24 h period need to be considered as possible confounders. Hence, confounders such as age, gender, tobacco smoke, ETS, HIV status, any medication use, etc. do not need to be considered as possible confounders. However, age, gender, tobacco smoke, ETS, HIV status and medication use may be effect modifiers seeing that these variables may be associated with RD hospital admissions.

6.2.2.4 Data analysis

Spearman correlation tests were applied to investigate if the air pollutants and Tapp were correlated. Conditional logistic regression models were applied to investigate the association between air pollutants and respiratory disease hospital admissions. The PROC PHREG procedure in SAS version 9.4 was used. One-pollutant and two-pollutant models were run. Since PM_{10} and NO_2 were significantly correlated ($p<0.01$), one-pollutant models were run.

Models were adjusted for public holiday variables (binary variable) and Tapp. There is no default method to include lags of air pollutants and Tapp in models.[36-39] As in other studies, the same lag of an air pollutant and Tapp were included in a model.[40-41] Lag0 to lag5 and the 2-cumulative to 6-cumulative averages were investigated; where lag0 is the same day as the hospital admission, lag5 is 5 days before the hospital admission, 2-cumulative average (CA2) refers to the average of lag0 and lag1 and 6-cumulative average (CA6) refers to the average of lag0 to lag5. The shape (i.e. linear or non-linear) of the association between the Tapp and respiratory disease hospital admissions was investigated using R version 3.5.0. Using log likelihood ratio tests, we compared non-linear Tapp and linear Tapp; non-linear Tapp was included in the regression models as a natural spline with three degrees of freedom.

The associations between hospital admissions and air pollutant level are presented as the percentage excess risk in hospital admissions per $10\mu/gm^3$ increase in an air pollutant level, similar to other studies.[42-43] Susceptibility of age groups (<15 years, 15-64 years and ≥65 years) and sex (male/female) on warm and cold days was investigated in stratified analyses followed by models with interaction terms. Intra-individual factors cannot be examined as effect modifiers due to the nature of the case-crossover design. However inter-individual variation,

using an interaction term between the effect modifier and an air pollutant in the conditional logistic regression model, can detect a p-value for interaction. Temperature, as an effect modifier of air pollution, is usually incorporated in regression models using short lags. In this study, we used Tapp and lag0 to 1, similar to other studies.[42-43]

Warm and cold days were defined as days when Tapp was higher than the 75th percentile of Tapp of the study period and lower than the 25th percentile of Tapp, respectively. Normal days were those equal or higher than the 25th percentile of Tapp, but lower or equal to the 75th percentile of Tapp. Other studies have used a similar approach.[42-43] Extreme warm and extreme cold days were also investigated, i.e. when Tapp was higher than the 95th percentile of Tapp of the study period and lower than the 5th percentile of Tapp, respectively. However, two-pollutant models were not run for either extremely cold or warm days due to few respiratory disease hospital admissions that occurred on such days. The majority (46%) of hospital admissions were due to bronchopneumonia (ICD10 J18), hence models were also run for this specific respiratory disease hospital admission outcome. Lastly, sensitivity analyses were run for the period 1 November 2011 to 30 November 2014 i.e. NO_2 outliers were excluded for the analysis.

6.2.3 Results

6.2.3.1 Descriptive statistics

In total, 17 647 respiratory disease hospital admissions were included in this study (Table 6.2). Approximately equal numbers of male (n=8 500) and female (n=9 147) patients were admitted (Figure 6.2). At most, 47 patients were admitted for respiratory disease in a day (Table 6.2). The mean Tapp for the study period was 16.6°C, PM_{10} peaked at 170.2 $\mu g/m^3$ (Figure 6.3). During the 1430-day study period, the PM_{10} levels exceeded the daily WHO guideline (50 $\mu g/m^3$) on 662 days, while SO_2 levels exceeded the daily WHO guideline (20$\mu g/m^3$) on three days (Figure 6.4) The time series plot of RD hospital admissions, PM_{10}, NO_2 and SO_2 level for the study period are also shown in Figure 6.2 to 6.5. PM_{10} and NO_2 had a significant positive correlation, and PM_{10} and NO_2 had a negative correlation with Tapp, temperature and relative humidity. Also, SO_2 had significantly positive correlation with NO_2 (Table 6.3). On warm and cold days, the mean (range) of Tapp was 21.2 (21.2 - 27.2) and 12.1°C (0.8 - 12.1), respectively.

There were 7 483 and 10 164 respiratory disease hospital admissions, respectively on warm and cold days. During the 1430-day study period, there were 418 cold days and 337 warm days

with 25th percentile temp (12.1°C) and 75th percentile temp (21.1°C), respectively. Also, there were 48 extreme cold days and 67 extreme warm days with 5th percentile temp (7.9°C) and 95th percentile temp (23.7°C), respectively. On warm and cold days, the mean (range) of PM_{10} levels was 43.2 (10.5-150.6) and 69.6 (3.5-170.2) $\mu g/m^3$, respectively. For NO_2, the mean (range) was 11.4 (0.9-94.5) and 20.8 (2.0- 185.3) $\mu g/m^3$, respectively on warm and cold days. There were 4803 and 3483 respiratory disease hospital admission during cold (<25th percentile of Tapp for lag2) and warm (>75th percentile of Tapp for lag2) days respectively. Most of the respiratory disease hospital admissions (8421) occurred during normal days. On extreme cold and extreme warm days, 934 and 665 respiratory disease hospital admissions occurred.

Table 6.2: Summary of daily statistics of the health outcome, air pollutants and meteorological conditions in Pretoria, South Africa, 1 January 2011 - 30 November 2014 (1430 days)

Variable	Mean	Min	Percentile (%)			Max
			25	50	75	
RD hospital admissions						
All ages and both sexes (n=17 647)	12.3	1.0	6.0	12.0	17.0	47.0
Females (n=9 147)	6.6	1.0	3.0	6.0	9.0	30.0
Males (n=8 500)	6.2	1.0	3.0	6.0	8.0	24.0
0-14 year olds (n=10 870)	7.6	0.0	4.0	7.0	11.0	34.0
15-64 year olds (n=6 070)	4.2	0.0	2.0	4.0	6.0	17.0
≥65 year olds (n=707)	0.5	0.0	0.0	0.0	1.0	5.0
PM_{10} ($\mu g.m^{-3}$)	60.0	3.5	32.5	47.9	75.9	170.2
NO_2 ($\mu g.m^{-3}$)	16.5	0.9	9.5	13.8	20.6	185.3
SO_2 ($\mu g/m^3$)	5.9	0.7	3.7	5.4	7.4	26.1
Tapp (°C)	16.6	0.8	12.1	17.4	21.2	27.2
Temperature (°C)	17.8	3.1	14.3	18.5	21.4	27.2
Relative humidity (%)	52.3	9.6	39.5	52.4	65.1	94.5

Abbreviations: PM_{10}: particulate matter with an aerodynamic diameter of less than 10 μm; NO_2: nitrogen dioxide, Tapp: apparent temperature; Max: maximum value; Min: minimum value, RD: Respiratory disease

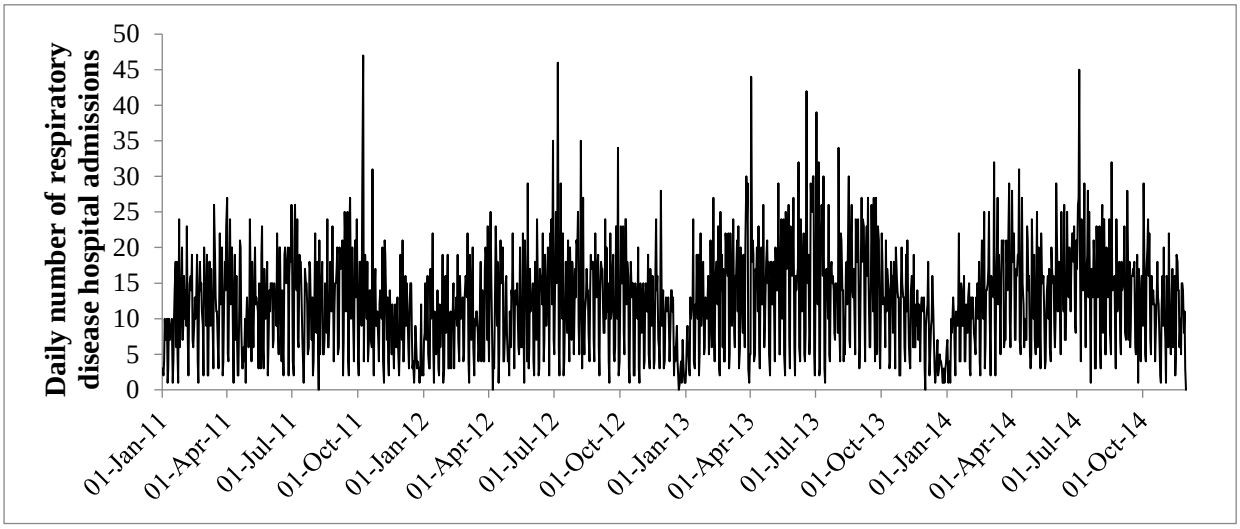


Figure 6.2: Daily number of respiratory disease hospital admissions for the period January 2011 to November 2014

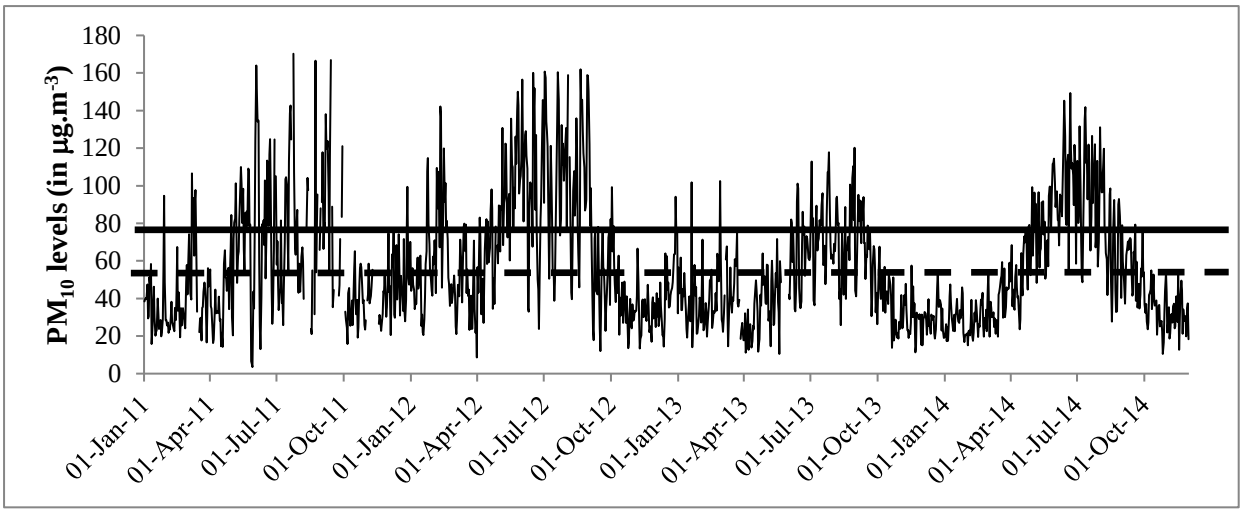


Figure 6.3: Daily PM_{10} levels in Pretoria for the period January 2011 to November 2014

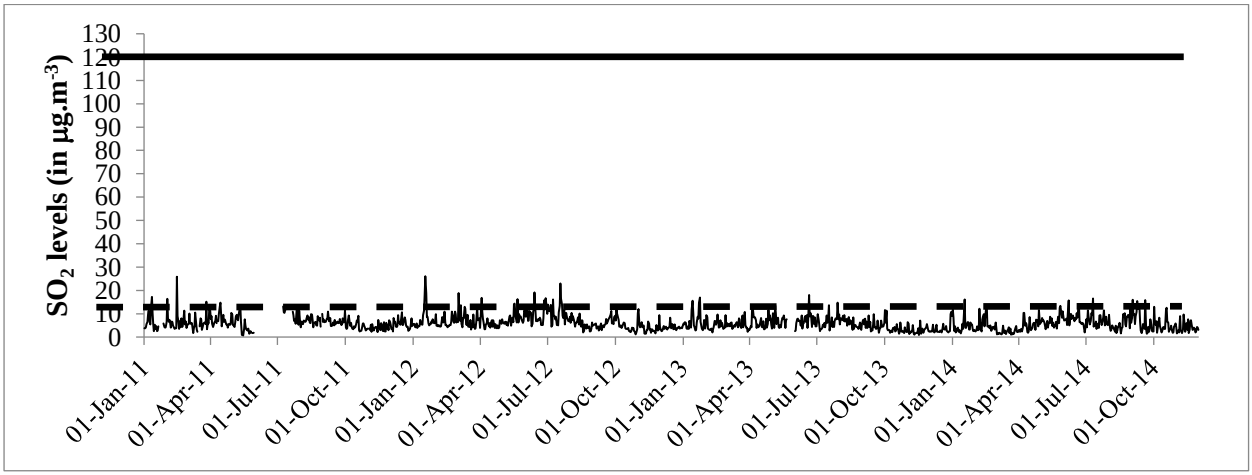


Figure 6.4: Daily SO_2 levels in Pretoria for the period January 2011 to November 2014

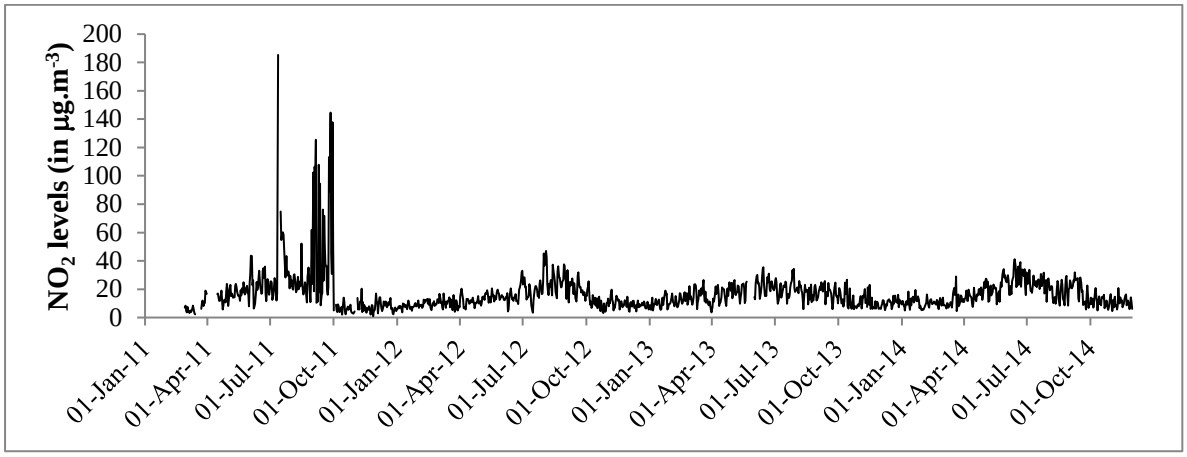


Figure 6.5: Daily NO_2 levels in Pretoria for the period January 2011 to November 2014

Table 6.3: Spearman correlation coefficients between air pollution and weather variables

Variable	NO_2	SO_2	Tapp	Temperature	RH
PM_{10}	0.608	0.379	-0.396	-0.324	-0.496
NO_2		0.485	-0.544	-0.459	-0.529
SO_2			-0.316	-0.261	-0.375
Tapp				0.970	0.281
Temperature					0.100

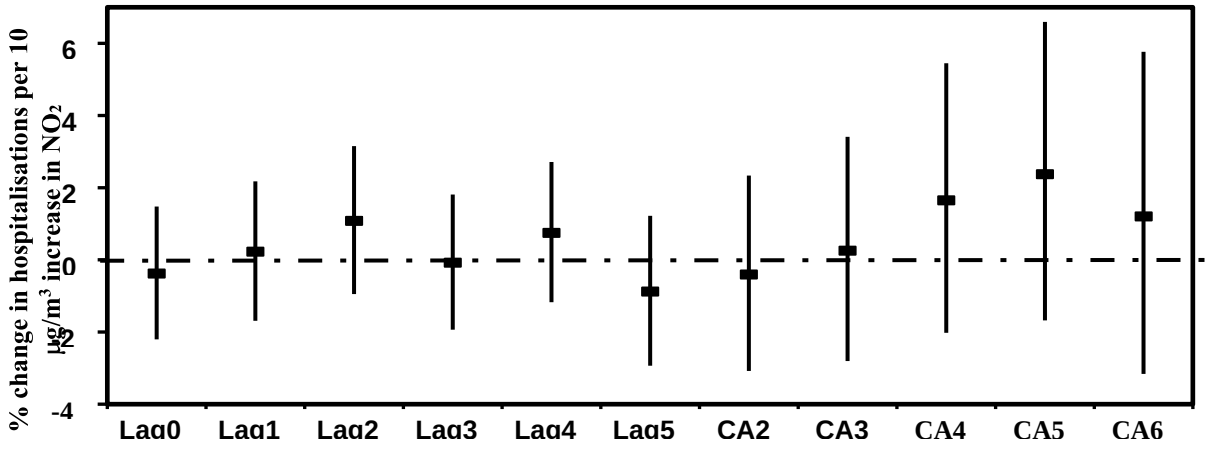
Abbreviations: PM_{10}: particulate matter with an aerodynamic diameter of less than 10 µm; NO_2: nitrogen dioxide; Tapp: apparent temperature; RH: relative humidity. All correlations were significant ($p < 0.01$)

6.2.3.2 Exposure - response estimates

Figure 6.5 shows the percentage changes in admission visits by a single model. No significant effects on hospitalisation were observed for PM_{10} and NO_2. Both warm and extreme warm days for PM_{10} follow the same pattern, but differ from cold and extreme cold days, where a 10 µg/m^3 increase in PM_{10} led to a decrease in respiratory disease hospital admissions across all ages and both sexes. The unstratified analysis shows that (entire Tapp range) a 10 µg/m^3 increase in PM_{10} was associated with an insignificant decrease of 0.2% (-0.7%; 1.2%) in respiratory disease hospital admissions. On warm days, a 10 µg/m^3 increase in PM_{10} led to more respiratory disease hospital admissions 1.5% (-1.5%; 4.6%) (Table 6.4). A 10 µg/m^3 increase in PM_{10} increased hospital admission by 1.9% (-2.0%; 6.0%) for 0-14 years old (n=10870) and higher admissions of female patients by 1.5% (-2.8%; 5.9%). The association between PM_{10} and respiratory disease hospital admissions was stronger for women than men on warm days.

Overall, the most marked association between hospital admissions and PM_{10} air pollution level occurred in the age group ≥65 years old, with an increase of 11.0% (-5.3%; 30.1%) on warm days.

(A)



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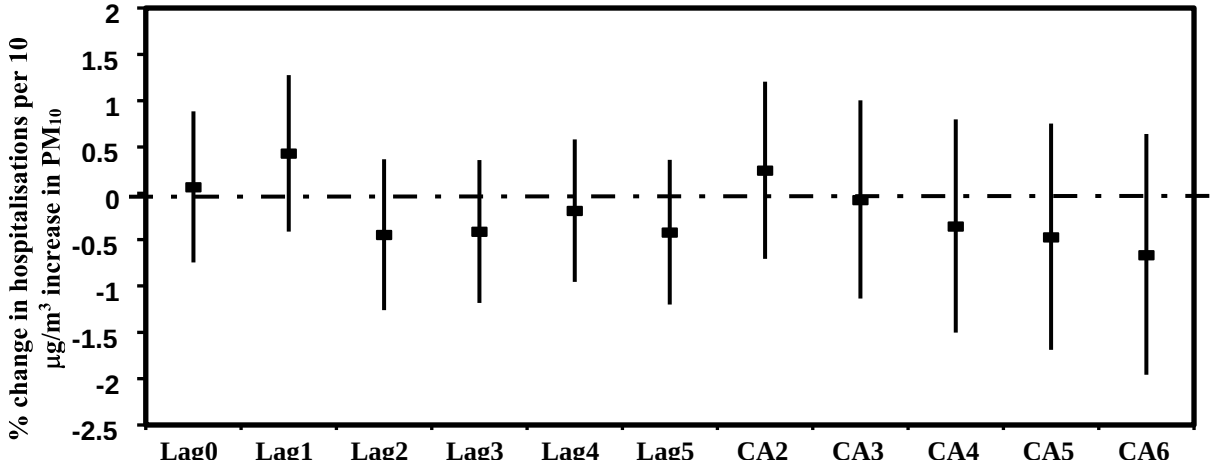


(C)

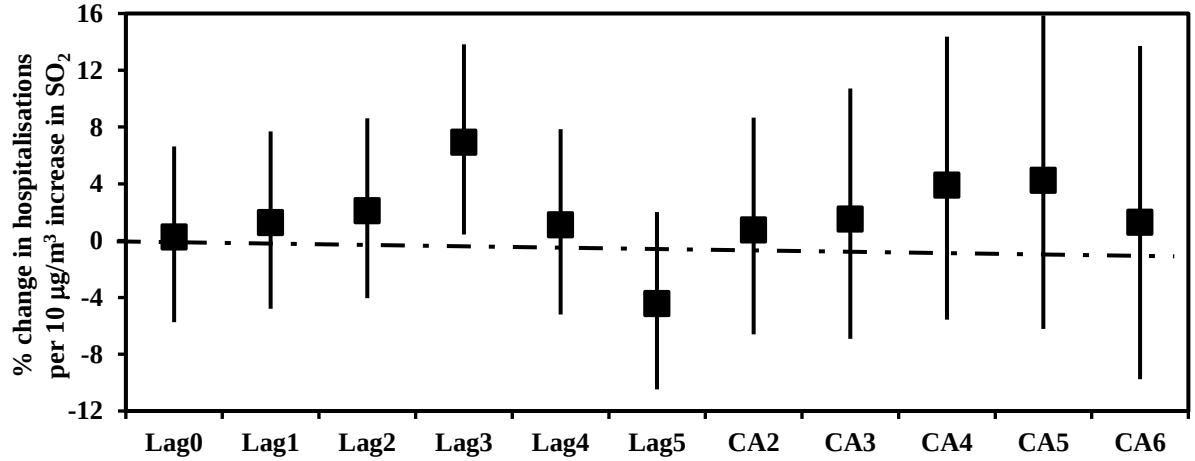


Figure 6.6: Percentage changes (95% CIs) in RD hospitalisation per 10µg/m3 increase in (a) NO_2 (b) PM_{10} (c) SO_2

On cold days a 10 μg/m^3 increase in PM_{10} led to a 3.0% increase in respiratory disease hospital admissions for the age group ≥65 years (-3.9%; 10.5%) and an insignificant increase in respiratory disease hospital admissions by 0.8% (-1.6%; 3.3%) for the 15 - 64 year old group. During extreme warm days, a 10 μg/m^3 increase in PM_{10} did not influence respiratory disease hospital admissions. On extreme cold days there was a significant decrease in respiratory disease hospital admission by -5.0% (-9.7%; 0.0%) and -7.6% (-13.6%; -1.2%) for all and the 0 – 14 years old group, respectively (Table 6.8). On warm days, a 10 μg/m^3 increase in PM_{10} led to a decrease in bronchopneumonia hospital admissions for all groups except the ≥65 years old and female patients (Table 6.9).

An insignificant increase of 6.7% (-22.0%; 45.9%) and 3.4% (-8.1%; 16.2%) in pneumonia hospital admissions were recorded for the ≥65year old group on warm and cold days, respectively. Conversely, a 10 μg/m^3 increase in PM_{10} led to a decrease in bronchopneumonia hospital admissions for the 0 – 14 years group during warm and cold days. Figure 6.6 shows the percentage change in admissions in a single pollutant model. Although no significant associations were observed between PM_{10} and bronchopneumonia hospital admissions for the 0-14 and ≥65 years old groups on cold days, ≥65 years olds were significantly more vulnerable than 0-14 years olds to increasing PM_{10} levels (interaction term p-value < 0.05); the same was not observed for respiratory disease hospital admission (ICD10 J00-J99 codes). After excluding NO_2 outliers (12 July 2011), none of the sub groups analysis differed significantly from each other (Table 6.5).

No significant association was observed for NO_2 (-0.4% (-3.1%; 2.3%)) in the unstratified analysis (entire Tapp range). On warm days, there was a significant increase of 19.0% (2.3%; 38.4%) in respiratory disease hospital admissions (Table 6.4). No significant association between NO_2 and respiratory disease hospital admissions was observed for females and male patients during warm and cold days. For NO_2, a 10 μg/m^3 increase for all groups led to a -3.0% decrease in respiratory disease hospital admissions (-6.9%; 1.1%), but there was a 2.2% increase in hospital admission for ≥65 years old (-13.9%; 21.3%). The results showed that males are more at risk of respiratory disease hospital admissions compared to females, -5.5% (-11.1%; -0.4%) and -0.7% (-6.1%; 5.1%) respectively. Lastly, no sign of interaction between hospital admissions and NO_2 levels was observed among the age groups on warm days.

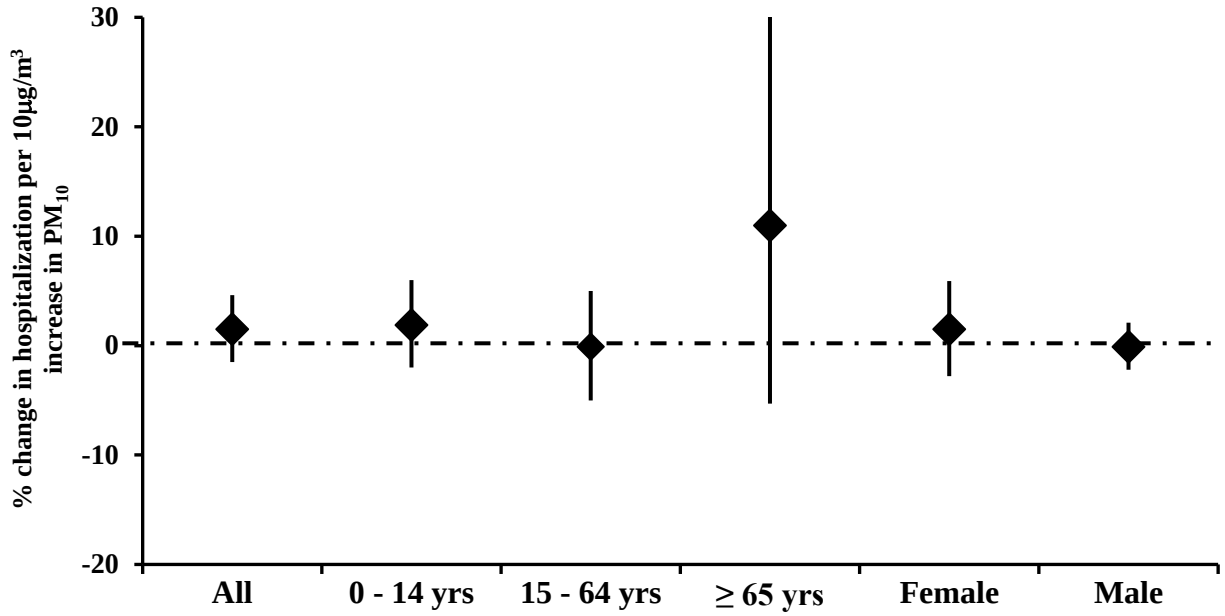
In Table 6.8, elevated NO_2 levels did not influence RD hospital admissions for both extreme warm and cold days. However, an insignificant increase in RD hospital admissions was observed for≥65 years old by 29.2% (-89%; 1475.8%) for extreme warm days.

Elevated NO_2 levels did not influence pneumonia hospital admissions during the warm and cold days, but an insignificant increase of 29.0% (-82.6%; 855.2%) for pneumonia hospital admission was recorded during warm days for age >65years. (Table 6.9). Elevated NO_2 levels did influence hospital admissions for male patients during the extreme warm days when compared to the warm days where no influence on hospital admission was observed. Although no significant associations between NO_2 and J18 hospital admission were observed for males and females on cold days, males were less vulnerable than females to increasing NO_2 levels (interaction term p-value < 0.05), the same was observed for RD hospital admission (ICD10 J00-J99 codes).

No significant difference detected for any of the effect modifiers (Figure 6.8). The results from the two-pollutant model showed no significant associations between NO_2 and RD hospital admission for males and females on cold days after adjusting for PM_{10}. However, males were less vulnerable than females to increasing NO_2 levels (interaction term p-value < 0.05) after adjusting for PM_{10}, so the same was observed as in the 1-pollutant models. Also, no sign interaction for NO_2 when adjusted for SO_2 (Table 6 5)

The high levels of NO_2 observed during the period of July 2011 to October 2011 cannot be explained. However, we did run a sensitivity analysis where the period (1 January 2011 to 31 October 2011 of high levels of NO_2 was excluded in the analysis. The results from the sensitivity analysis (1 November 2011 – 30 November 2014) for the one pollutant model showed that significant associations were obtained but no significant difference exists between males and females on cold days (Table 6.6). Also, the sensitivity analysis for the two-pollutant model showed that none of the subgroups differ significantly from each other after excluding outliers for NO_2 and controlled for PM_{10}. In addition, the sensitivity analysis showed that none of the subgroup analyses differ from each other after excluding outliers for NO_2 and controlled for SO_2 in two pollutants model. Interaction term p-value >0.05. (Table 6.7). Also, the strength of association after excluding outliers for NO_2 increase across all age group and sex except for 15-64 year olds and males on warm days while on cold days, there was a decrease in the association except for 15-64 year olds and males.

(A)



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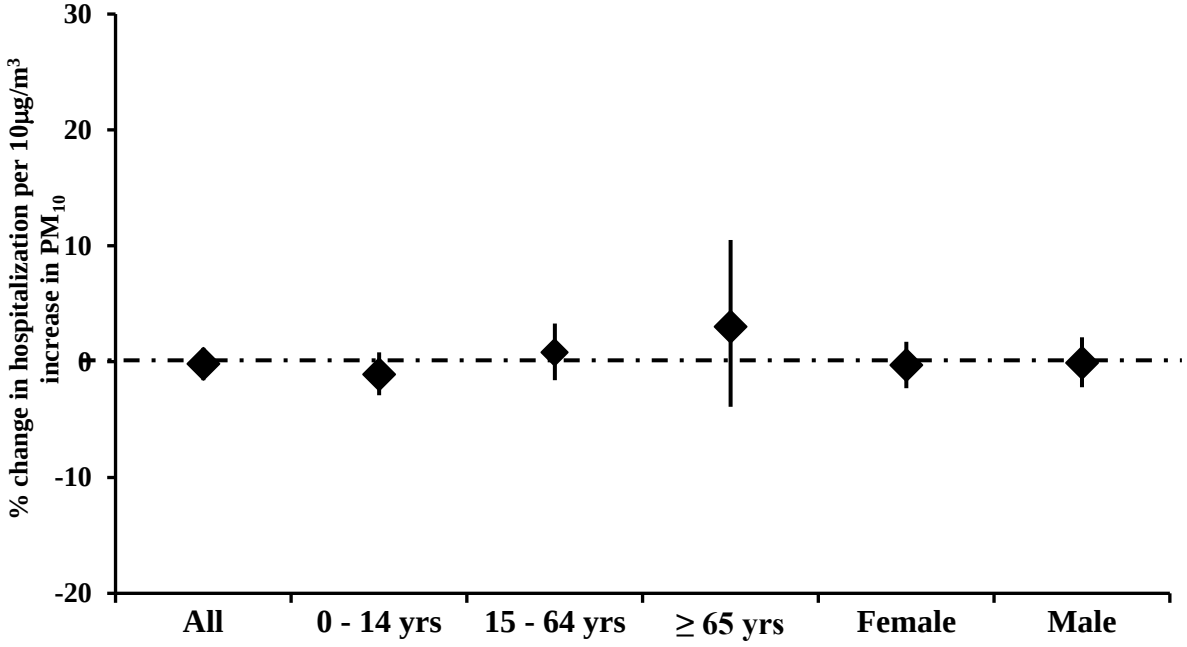
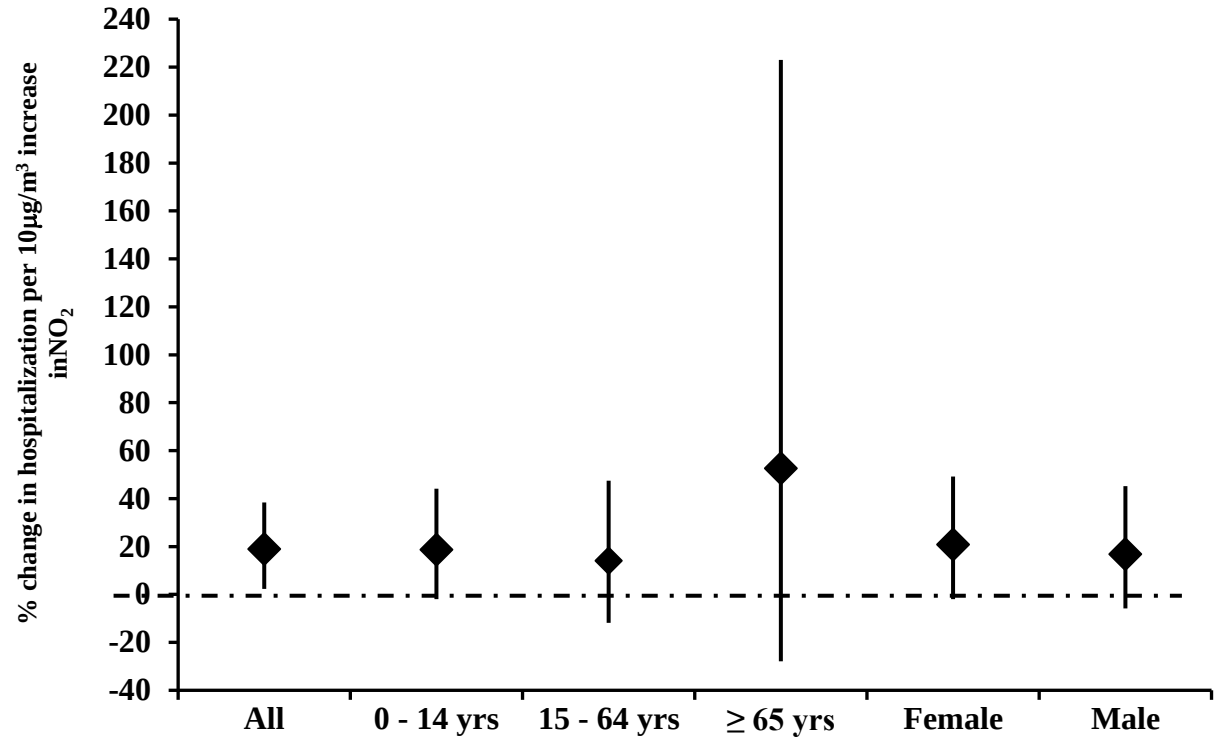


Figure 6.7: Effect modification by warm and cold apparent temperature (A) PM_{10} warm (B) PM_{10} cold

(A)



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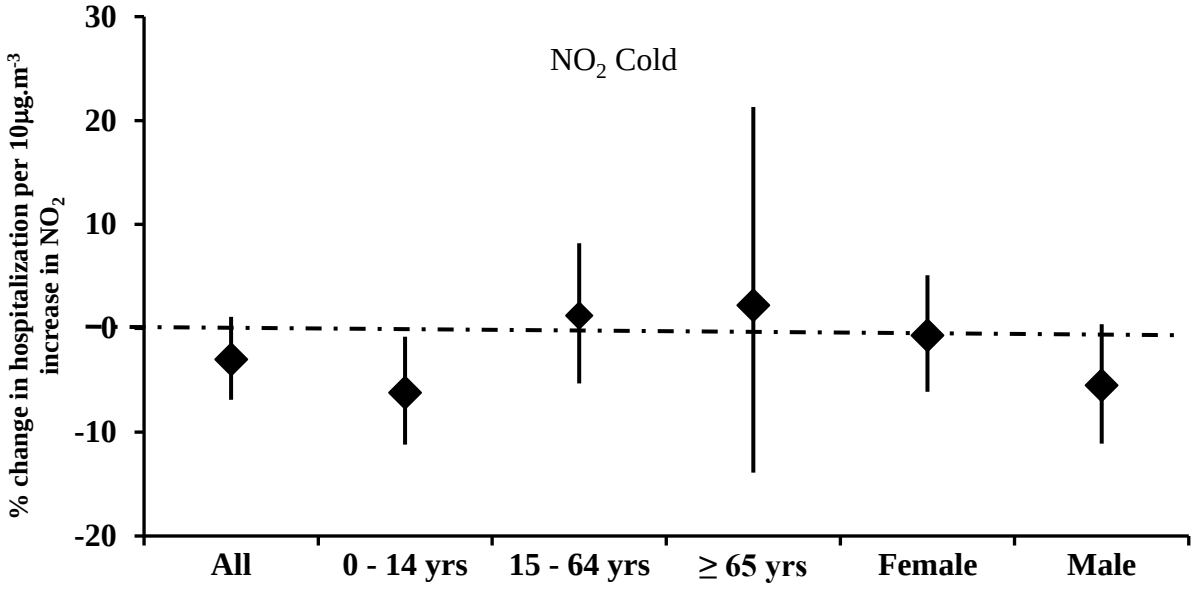
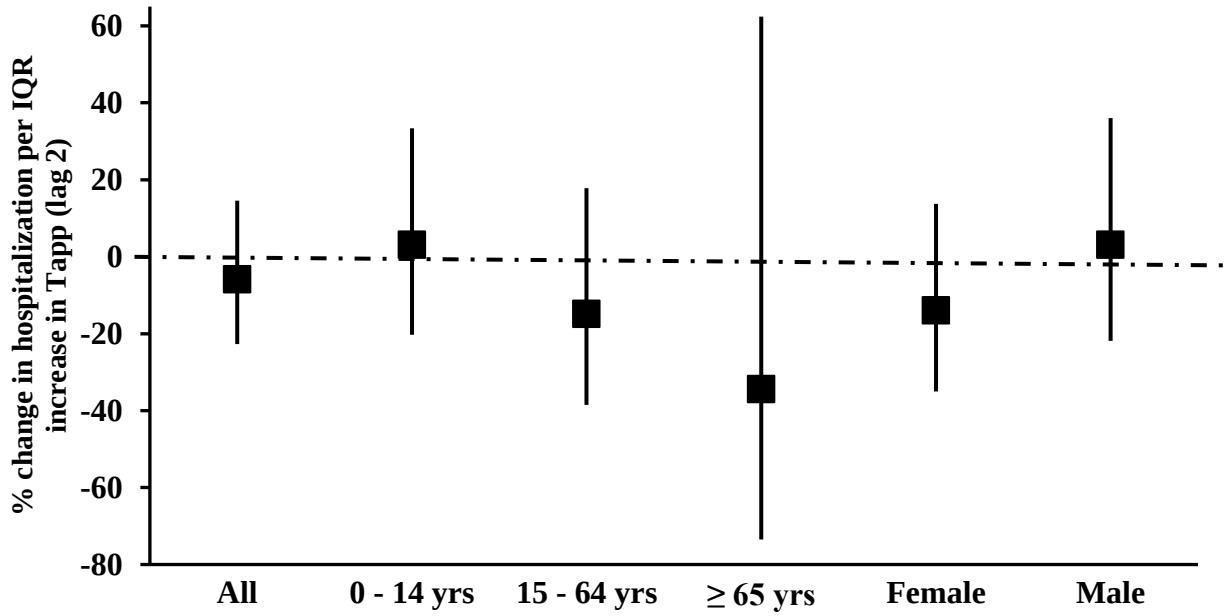


Figure 6.8: Effect modification by warm and cold apparent temperature (A) NO_2 warm (B) NO_2 cold

(A)



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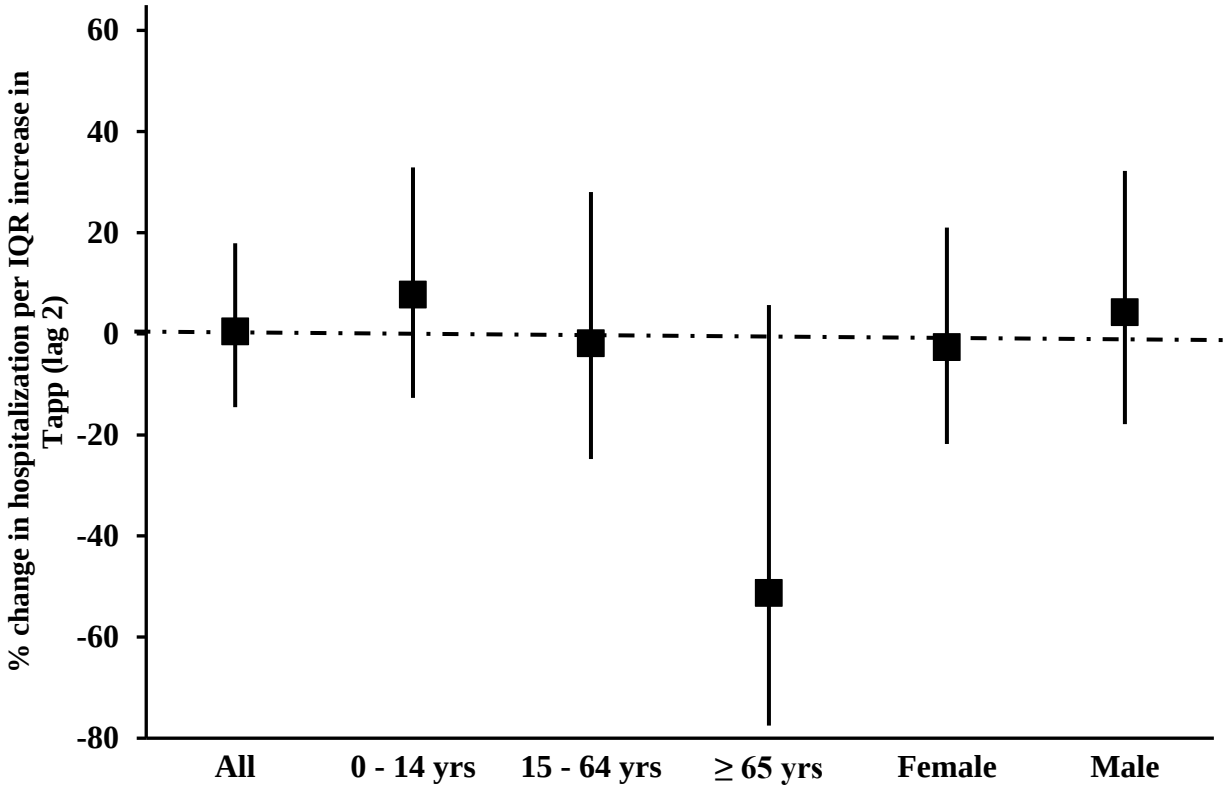

Figure 6.9: Effect modification by warm and cold apparent temperature (A) SO_2 warm (B) SO_2 cold

No significant association was observed for SO_2 0.7% (-6.6%; 8.7%) in the unstratified analysis. A 10 $\mu g.m^{-3}$ increase in SO_2 led to increased RD hospital admissions for 0-14 years 3.1% (-20.3%; 33.4%) and male 3.1% (-21.9%; 36.0%) on warm days (Table6.4), while there was an increase hospital admission for all 0.4% (-14.5%; 17.9%), 0-14 years 7.7% (-12.7%; 32.9%) and male 4.2% (-17.9%; 32.2%) on cold days. Significant association between SO_2 and RD hospital admissions was observed for females and male patients during warm and cold days. Lastly, the most noticeable decrease between hospital admissions and SO_2 air pollution level occurred in age group of ≥65 years old by -34.4 (-73.5; 62.4) and -51.3 (-77.5; 5.7) for both warm and cold days respectively.

In Table 6.5, elevated NO_2 levels did not influence respiratory disease hospital admissions for both extreme warm or cold days. Elevated NO_2 levels did not influence bronchopneumonia hospital admissions during the warm or cold days (Table 6.8). Although no significant associations were observed between NO_2 and bronchopneumonia hospital admission for males and females on cold days, males were less vulnerable than females to increasing NO_2 levels (interaction term p-value < 0.05); the same was observed for respiratory disease hospital admission (ICD10 J00-J99 codes).

The results from the two-pollutant models showed no significant associations between NO_2 and respiratory disease hospital admission for males and females on cold days after adjusting for PM_{10}. However, males were less vulnerable than females to increasing NO_2 levels (interaction term p-value < 0.05) after adjusting for PM_{10}, as observed in the one-pollutant models. No significant associations were observed for NO_2 when adjusted for SO_2 (Table 6.5)

No significant association was observed for SO_2 (0.7%: -6.6%; 8.7%) in the unstratified analysis (entire Tapp range). A 10 $\mu g/m^3$ increase in SO_2 led to a 3.1% increase in respiratory disease hospital admissions for the 0-14 years (-20.3%; 33.4%) and 3.1% for males (-21.9%; 36.0%) on warm days (Table 6.4), while there was an increase of 0.4% in hospital admission for all (-14.5%; 17.9%), 7.7% in 0-14 years (-12.7%; 32.9%) and 4.2% in males (-17.9%; 32.2%) on cold days. No significant association between SO_2 and respiratory disease hospital admissions was observed on warm and cold days.

In Table 6.8, elevated SO_2 levels did not influence respiratory disease hospital admissions for both extreme warm and cold days. Elevated SO_2 levels did not influence bronchopneumonia hospital admissions during warm and cold days (Table 6.9). No significant interactions were

observed for SO_2 and hospital admissions in the subgroups. No significant difference was detected for any of the effect modifiers (Figure 6.9). Lastly, after controlling NO_2 outliers, none of the subgroup analysis differed significantly from each other (interaction terms p-value >0.05).

Table 6.4: Percentage change (95% CI) in daily respiratory disease hospital admissions per 10 μg/m^3 increase in an air pollutant level (lag0-1) on warm and cold days by age groups and sex

Air pollutant	Tapp	All	0-14 year olds	15-64 year olds	≥65 year olds	Females	Males
PM_{10}	**Warm**	1.5 (-1.5; 4.6)	1.9 (-2.0; 6.0)	-0.1 (-5.0; 5.0)	11.0 (-5.3; 30.1)	1.5 (-2.8; 5.9)	1.6 (-2.7; 6.0)
	Normal	0.5 (-1.3; 2.3)	0.0 (-2.3; 2.4)	0.8 (-2.3; 4.0)	3.6 (-5.3; 13.2)	-0.2 (-2.7; 2.4)	1.2 (-1.4; 3.9)
	Cold	-0.2 (-1.6; 1.2)	-1.1 (-2.9; 0.8)	0.8 (-1.6; 3.3)	3.0 (-3.9; 10.5)	-0.3 (-2.3; 1.7)	-0.1 (-2.2; 2.1)
NO_2	**Warm**	19.0 (2.3; 38.4)	18.8 (-2.0; 44.1)	14.0 (-11.9; 47.5)	52.6 (-27.9; 223.0)	20.9 (-2.0; 49.2)	16.9 (-5.8; 45.2)
	Normal	-0.6 (-4.7; 3.6)	1.2 (-4.2; 6.9)	-4.4 (-10.8; 2.4)	6.4 (-11.2; 27.6)	0.4 (-5.2; 6.2)	-1.7 (-7.5; 4.5)
	Cold	-3.0 (-6.9;1.1)	-6.2 (-11.2; -0.8)	1.2 (-5.3; 8.2)	2.2 (-13.9; 21.3)	-0.7 (-6.1; 5.1)*	-5.5 (-11.1; 0.4)*
SO_2	**Warm**	-5.9 (-22.7; 14.6)	3.1 (-20.3; 33.4)	-14.9 (-38.5; 17.8)	-34.4 (-73.5; 62.4)	-14.0 (-35.0; 13.7)	3.1 (-21.9; 36.0)
	Normal	8.2 (-3.9; 21.9)	11.0 (-4.3; 28.8)	3.0 (-16.4; 26.8)	8.7 (-42.0; 103.7)	13.9 (-3.6; 34.6)	2.8 (-13.2; 21.8)
	Cold	0.4 (-14.5; 17.9)	7.7 (-12.7; 32.9)	-1.9 (-24.8; 28.0)	-51.3 (-77.5; 5.7)	-2.7 (-21.8; 21.0)	4.2 (-17.9; 32.2)

Abbreviations: PM_{10}: particulate matter with an aerodynamic diameter of less than 10 μm; NO_2: nitrogen dioxide; Tapp: apparent temperature; RH: relative humidity; Warm: Apparent temperature > 75th percentile; Cold: Apparent temperature < 25th percentile; Normal: Apparent temperature >= 25th and <= 75th percentile
All the interaction term p-values > 0.05, except for males/females on cold days*

Table 6.5: Percentage change (95% CI) in daily respiratory disease hospital admissions per 10 μg/m^3 increase in an air pollutant level (lag0-1) on warm and cold days by age groups and sex

Air pollutant	Day type	All	0-14 year olds	15-64 year olds	≥65 year olds	Females	Males
PM_{10} controlled for NO_2	**Warm**	1.2 (-1.9; 4.4)	1.2 (-2.9; 5.4)	0.1 (-4.9; 5.4)	12.2 (-4.8; 32.3)	1.5 (-2.9; 6.1)	0.9 (-3.5; 5.5)
	Normal	0.5 (-1.4; 2.5)	-0.2 (-2.7; 2.3)	1.2 (-2.1; 4.7)	4.9 (-4.5; 15.2)	-0.4 (-3.0; 2.4)	1.6 (-1.3; 4.5)
	Cold	0.3 (-1.4; 2.1)	-0.1 (-2.3; 2.2)	0.6 (-2.2; 3.5)	3.2 (-4.7; 11.8)	-0.5 (-2.8; 1.9)	1.3 (-1.3; 4.0)
NO_2 controlled for PM_{10}	**Warm**	16.5 (-0.7; 36.6)	19.5 (-2.6; 46.7)	9.4 (-16.4; 43.1)	33.9 (-38.2; 190.2)	14.1 (-8.6; 42.5)	19.1 (-5.3; 49.7)
	Normal	-1.0 (-5.8; 4.1)	0.6 (-5.8; 7.4)	-2.5 (-10.3; 6.0)	-5.7 (-23.4; 16.1)	-0.8 (-7.2; 6.0)	-1.2 (-8.4; 6.6)
	Cold	-2.1 (-6.9; 3.0)	-4.1 (-10.4; 2.7)	0.7 (-7.1; 9.2)	-0.7 (-19.4; 22.4)	1.8 (-4.9; 8.9)	-6.6 (-13.6; 0.9)
PM_{10} controlled for SO_2	**Warm**	1.5 (-1.5; 4.7)	1.9 (-2.1; 6.0)	-0.1 (-5.0; 5.0)	12.4 (-4.3; 31.9)	1.6 (-2.7; 6.1)	1.5 (-2.8; 6.0)
	Normal	0.3 (-1.5; 2.2)	-0.2 (-2.6; 2.2)	0.8 (-2.4; 4.0)	3.8 (-5.2; 13.7)	-0.5 (-3.0; 2.1)	1.3 (-1.4; 4.1)
	Cold	0.3 (-1.5; 2.2)	-0.9 (-3.2; 1.4)	2.0 (-1.0; 5.1)	3.1 (-5.1; 12.0)	0.5 (-1.9; 2.9)	0.1 (-2.5; 2.8)
SO_2 controlled for PM_{10}	**Warm**	-5.7 (-22.6; 14.8)	3.8 (-19.8; 34.4)	-15.2 (-38.8; 17.5)	-34.0 (-73.6; 65.0)	-14.8 (-35.6; 12.8)	4.4 (-20.9; 37.8)
	Normal	5.4 (-6.8; 19.2)	9.1 (-6.5; 27.3)	0.2 (-19.1; 24.1)	-6.6 (-51.5; 79.8)	12.1 (-5.6; 33.1)	-1.2 (-17.1; 17.8)
	Cold	0.2 (-15.5;18.7)	12.7 (-9.7; 40.5)	-8.7 (-31.2; 21.3)	-53.3 (-79.2; 4.6)	-2.7 (-22.7; 22.5)	3.7 (-19.4; 33.5)
NO_2 controlled for SO_2	**Warm**	25.6 (6.1; 48.7)	22.0 (-1.6; 51.2)	22.2 (-8.5; 63.1)	136.7 (-8.7; 514.0)	32.2 (4.4; 67.5)	19.1 (-6.5; 51.7)
	Normal	-2.5 (-7.0; 2.2)	-1.5 (-7.4; 4.9)	-5.4 (-12.3; 2.1)	7.5 (-11.9; 31.1)	-1.8 (-7.8; 4.6)	-3.3 (-9.8; 3.7)
	Cold	1.9 (-5.1; 9.5)	-5.1 (13.5; 4.2)	12.1 (-0.5; 26.2)	24.8 (-11.4; 75.8)	6.9 (-3.0; 17.8)	-3.7 (-13.4; 7.1)
SO_2 controlled for NO_2	**Warm**	15.4 (-33.2; 7.1)	-8.1 (-32.5; 25.0)	-18.8 (-45.1; 19.9)	-61.4 (-88.0; 24.8)	-24.4 (-46.0; 5.7)	-5.4 (-32.1; 31.8)
	Normal	16.0 (1.3; 32.8)	16.8 (-1.6; 38.6)	17.0 (-7.5; 48.0)	-6.4 (-53.1; 87.0)	22.0 (1.0; 47.3)	10.5 (-9.1; 34.2)
	Cold	-1.4 (17.1; 17.4)	13.1 (-9.9; 41.8)	-11.6 (-33.7; 17.9)	-61.0 (-83.4; -8.7)	-8.6 (-27.8; 15.8)	7.8 (-16.6; 39.3)

Abbreviations: PM_{10}: particulate matter with an aerodynamic diameter of less than 10 μm; NO_2: nitrogen dioxide; Tapp: apparent temperature; RH: relative humidity;
Warm: Apparent temperature >75th percentile; Cold: Apparent temperature < 5th percentile; Normal: Apparent temperature ≥25th and ≤75th percentile
All the interaction term p-values > 0.05, except for males/females on cold days*

Table 6.6: Percentage change (95% CI) in daily respiratory disease hospital admissions per 10 µg/m^3 increase in an air pollutant level (lag0-1) on warm and cold days by age groups and sex. (November 2011 November 2014)

Air pollutant	Tapp	All	0 - 14 year olds	15- 64 year olds	≥65 year olds	Females	Males
NO_2	**Warm**	19.6 (2.7; 39.3)	21.0 (-0.4; 47.1)	12.1 (-13.6; 45.5)	50.4 (-29.4; 220.4)	20.1 (-2.9; 48.6)	18.9 (-4.5; 48.0)
	Normal	2.1 (-4.9; 9.7)	0.9 (-7.8; 10.3)	4.2 (-8.2; 18.3)	3.6 (-28.1; 49.3)	-3.1 (-12.5; 7.2)	7.7 (-2.6; 19.2)
	Cold	5.4 (-2.0; 13.2)	2.6 (-6.6; 12.6)	10.4 (-2.3; 24.7)	5.4 (-23.9; 46.0)	8.4 (-1.6; 19.5)	1.6 (-8.8; 13.2)

Abbreviations: PM_{10}: particulate matter with an aerodynamic diameter of less than 10 µm; NO_2: nitrogen dioxide; Tapp: apparent temperature; RH: relative humidity;
Warm: Apparent temperature >75th percentile; Cold: Apparent temperature < 5th percentile; Normal: Apparent temperature ≥25th and ≤75th percentile
All the interaction term p-values > 0.05, except for males/females on cold days*

Table 6.7: Percentage change (95% CI) in daily respiratory disease hospital admissions per 10 µg/m^3 increase in an air pollutant level (lag0-1) on warm and cold days by age groups and sex. (November 2011 November 2014)

Air pollutant	Tapp	All	0 - 14 year olds	15- 64 year olds	≥65 year olds	Females	Males
NO_2 controlled for SO_2	**Warm**	26.2 (6.3; 49.9)	24.7 (0.2; 55.2)	19.2 (-11.1; 59.8)	138.7 (-9.5; 529.6)	31.5 (3.4; 67.2)	21.0 (-5.4; 54.8)
	Normal	-2.6 (-11.4; 7.0)	-5.4 (-16.0; 6.6)	0.8 (-14.6; 18.8)	12.0 (-31.5; 83.0)	-13.4 (-24.1; -1.2)	10.5 (-3.5; 26.6)
	Cold	6.4 (-1.7;15.1)	1.1 (-8.7; -11.9)	13.7 (-0.3; 29.7)	25.4 (-13.4; 81.5)	10.8 (-0.3; 23.2)	1.2 (-10.1; 13.8)
SO_2 controlled for NO_2	**Warm**	-15.1 (-33.1; 7.7)	-9.0 (-33.4; 24.1)	-16.2 (-43.4; 24.0)	-62.6 (-88.5; 22.1)	-24.1 (-45.9; 6.4)	-5.1 (-32.1; 32.7)
	Normal	14.1 (-3.8; 35.3)	19.1 (-3.9; 47.4)	10.0 (-18.5; 48.4)	-19.4 (-67.9; 102.1)	37.3 (8.3; 74.0)	-6.6 (-27.0; 19.5)
	Cold	-5.3(-21.0; 13.4)	8.6 (-14.1; 37.4)	-15.9 (-37.6; 13.5)	-58.6 (-82.9; 0.4)	-11.6 (-30.9; 12.9)	2.7 (-21.4; 34.2)
NO_2 controlled for PM_{10}	**Warm**	1.2 (-2.0; 4.5)	1.2 (-3.0; 5.5)	0.4 (-4.7; 5.9)	7.7 (-9.3; 28.0)	1.4 (-3.1; 6.1)	1.0 (-3.5; 5.7)
	Normal	1.9 (-0.8; 4.6)	1.5 (-1.9; 5.0)	1.6 (-3.1; 6.5)	7.3 (-5.3; 21.6)	2.0 (-1.7; 5.9)	1.8 (-2.1; 5.8)
	Cold	-0.7 (-3.1; 1.8)	0.5 (-3.7; 2.7)	-1.4 (-5.4; 2.9)	3.2 (-7.3; 14.8)	-1.9 (-5.1; 1.4)	1.0 (-2.7; 4.8)
NO_2 controlled for PM_{10}	**Warm**	17.0 (-0.5; 37.6)	21.9 (-1.0; 50.2)	7.0 (-18.5; 40.6)	36.3 (-37.5; 197.6)	13.3 (-9.6; 41.9)	21.1 (-4.0; 52.9)
	Normal	-1.8 (-10.6; 7.9)	-2.0 (-12.8; 10.1)	0.7 (-14.9; 19.1)	-14.4 (-46.7; 37.5)	-7.3 (18.8; 5.8)	4.0 (-9.0; 18.9)
	Cold	7.5(-3.0; 19.0)	3.7 (-9.3; 18.5)	16.2 (-2.0; 37.8)	-6.4 (-40.2; 46.4)	14.6 (0.0; 31.3)	-1.1(-15.3; 15.6)

Abbreviations: PM_{10}: particulate matter with an aerodynamic diameter of less than 10 µm; NO_2: nitrogen dioxide; Tapp: apparent temperature; RH: relative humidity;
Warm: Apparent temperature >75th percentile; Cold: Apparent temperature < 5th percentile; Normal: Apparent temperature ≥25th and ≤75th percentile
All the interaction term p-values > 0.05, except for males/females on cold days*

Table 6.8: Percentage change (95% CI) in daily respiratory disease hospital admissions per 10 μg/m^3 increase in an air pollutant level (lag0-1) on extremely warm and extremely cold days by age groups and sex

Air pollutant	Day type	All	0-14 year olds	15-64 year olds	≥65 year olds	Females	Males
PM_{10}	**Extreme warm**	-1.9 (-10.6; 7.7)	2.7 (-9.2; 16.3)	-8.2 (-20.9; 6.5)	14.4 (-40.9; 121.4)	4.0 (-8.6; 18.4)	-8.0 (-19.7; 5.4)
	Normal	0.7 (-0.4; 1.8)	0.1 (-1.3;1.5)	1.4 (-0.5; 3.2)	4.4 (-1.2; 10.3)	0.8 (-0.7; 2.3)	0.7 (-0.9; 2.3)
	Extreme cold	-5.0 (-9.7; 0.0)	-7.6 (-13.6; -1.2)	-1.0 (-9.0; 7.6)	-1.2 (-23.7; 27.9)	-3.5 (-9.8; 3.1)	-7.0 (-14.1; 0.7)
NO_2	**Extreme warm**	10.8 (-25.1; 64.0)	18.5 (-28.3; 96.0)	0.4 (-47.6; 92.2)	29.2 (-89.4; 1475.8)	22.2 (-30.5; 114.8)	-2.4 (-43.6; 69.0)
	Normal	1.0 (-2.2; 4.3)	2.3 (-1.9; 6.7)	-1.8 (-6.9; 3.5)	7.7 (-6.9; 24.7)	2.3 (-2.0; 6.9)	-0.4 (-5.0; 4.4)
	Extreme cold	-2.9 (-9.2; 3.7)	-3.1 (-11.3; 5.9)	-2.1 (-11.9; 8.7)	-8.6 (-34.9; 28.3)	0.4 (-8.1; 9.8)	-6.7 (-15.7;3.2)
SO_2	**Extreme warm**	34.1 (-13.5; 107.6)	51.8 (-14.4; 169.4)	33.5 (-33.6; 168.2)	-93.8 (-99.9; 156.9)	19.3(-35.3; 120.2)	48.9 (-20.6; 179.3)
	Normal	1.2 (-6.6; 9.7)	5.2 (-5.1; 16.7)	-4.6 (-16.8; 9.4)	-5.6 (-36.9; 41.2)	4.6 (-6.5; 17.1)	-2.1 (-12.8; 9.9)
	Extreme cold	634.3(-37.4; 8506.1)	776.5(-61.9; 20057.7)	869.6(-88.7; 83220.7)	-88.7(-100.0; 305056.4)	254.3(-82.5; 7068.9)	3015.3(-58.1;231676.7)

Abbreviations: PM_{10}: particulate matter with an aerodynamic diameter of less than 10 μm; NO_2: nitrogen dioxide; Tapp: apparent temperature; RH: relative humidity;

Extreme warm: Apparent temperature >95th percentile; Extreme cold: Apparent temperature <5th percentile; Normal: Apparent temperature ≥5th and ≤95th percentile

All the interaction term p-values > 0.05

Table 6.9: Percentage change (95% CI) in daily J18 (Bronchopneumonia) hospital admissions per 10 µg/m^3 increase in an air pollutant level (lag0-1) on warm and cold days by age groups and sex

Air pollutant	Tapp	All	0-14 year olds	15-64 year olds	≥65 year olds	Females	Males
PM_{10}	**Warm**	-1.0 (-5.4; 3.6)	-1.2(-6.1; 4.0)	-1.0 (-10.8; 9.8)	6.7 (-22.0; 45.9)	0.0 (-6.3; 6.7)	-1.8 (-7.9; 4.6)
	Normal	1.5 (-1.1;4.2)	1.5 (-1.4; 4.5)	0.3 (-6.1; 7.1)	6.3 (-9.6; 25.0)	-2.0 (-5.6; 1.8)	4.8 (1.1; 8.7)
	Cold	0.5 (-1.8; 2.8)	-0.8 (-3.4;1.8)	4.9 (-0.2; 10.2)	3.4 (-8.1; 16.2)	0.2 (-2.9; 3.5)	0.7 (-2.5,4.1)
NO_2	**Warm**	10.4 (-12.1; 38.6)	2.5 (-19.9; 31.3)	70.3 (-8.8; 218.2)	29.0 (-82.6; 855.2)	14.5 (-17.2; 58.4)	5.2 (-23.7;45.0)
	Normal	2.6 (-3.1; 8.7)	5.0 (-1.8; 12.2)	-5.1 (-16.3; 7.6)	3.3 (-24.8; 41.8)	0.6 (-6.8; 8.5)	5.6 (-3.4; 15.4)
	Cold	-6.3 (-12.3; 0.1)	-6.9 (-14.2; 1.0)	-5.3 (-17.1; 8.2)	-6.4 (-26.1;18.4)	-1.8 (-10.2;7.3)	-11.3 (-19.7; -2.0)
SO_2	**Warm**	3.1 (-23.2; 38.4)	0.1 (-28.1; 39.4)	57.1 (-21.2; 213.5)	-85.7 (-98.6; 42.9)	8.8 (-29.9; 69.0)	-1.6 (-33.8; 46.3)
	Normal	7.0 (-10.2; 27.4)	13.5 (-6.3; 37.5)	-23.0 (-51.3; 21.9)	-6.7 (-75.1; 250.1)	-1.3 (-23.5; 27.2)	14.6 (-10.0; 45.9)
	Cold	-12.8 (-32.3; 12.4)	-2.0 (-26.8; 31.2)	-34.7 (-62.6; 14.1)	-54.1 (-88.0; 75.4)	-22.0 (-45.7; 12.0)	-2.6 (-31.7; 38.9)

Abbreviations: PM_{10}: particulate matter with an aerodynamic diameter of less than 10 µm; NO_2: nitrogen dioxide; Tapp: apparent temperature;

Warm: Apparent temperature > 75th percentile; Cold: Apparent temperature < 25th percentile; Normal: Apparent temperature >= 25th and <= 75th percentile

6.2.4 Discussion

Studies in South Africa have used the case-crossover study design to investigate air pollutants and health outcomes. However, to the best of our knowledge, this study happens to be the first to study the association between exposure to air pollutants (PM_{10}, NO_2 and SO_2) and respiratory hospital admission using case-crossover study design.

This study revealed that out of the 1430 days study period, PM_{10} was found to exceed the WHO daily guideline of 50 $\mu g/m^3$ for almost half of the period (46.3%). On extreme days, daily concentration of PM_{10} was observed to be as high as 170.2 $\mu g/m^3$. Non-linear Tapp did not add value to the model and Tapp was then included as a linear term, similar to other studies.[42-43] The negative correlation that exists between temperature and PM_{10} and NO_2 was in line with the studies by Huang *et al*[44] and Loung *et al*.[25] In this study, PM_{10} and NO_2 had stronger detrimental effects on respiratory disease hospital admissions on warm days. The effects observed for PM_{10} over the Tapp range in this study is in contrast with the systematic and meta-analysis of Newell *et al*.[45]

The study by Sousa *et al*[46] reported that an increase in 10 $\mu g/m^3$ of PM_{10} was associated with an increase of 2% in risk of respiratory admission. Kloog *et al*[47]also indicated that exposure to each 10 $\mu g/m^3$ increase in $PM_{2.5}$ was associated with 2.2% increase in hospital admission for respiratory disease in the US. In a study by Phung *et al*,[48] it was reported that the risk of respiratory admission (for all ages) in Vietnam increased by 0.7% for each 10 $\mu g/m^3$ increase in PM_{10}.

The effect of PM_{10} observed in this study on warm days (1.5%: -1.5%; 4.6%) falls into the range of 1–2% per 10$\mu g/m^3$ increase in PM_{10} level for excess risk of respiratory disease hospitalisation.[46,49-50] Positive association between outdoor air pollution levels and related respiratory symptom emergency visits across all age groups observed in Villeneuve *et al*[51] is similar to our result. However, some studies have found stronger effects, for instance, Rodopoulou *et al*[10] and Tramuto *et al*[12] established strong effects of PM_{10} on risk of respiratory admissions on warm days with estimated increases of 3.2% and 3.9% for each 10 $\mu g/m^3$ increase in concentration, while other studies reported no increased risk with PM_{10}.[52-53] These varying results may be due to the source and composition of PM, which is found to vary with ambient temperature with greater proportion of its toxic forms present at high temperature, which could invariably increase respiratory disease hospital admissions.[54-56]

Therefore, synergistic effects on humans could occur if high levels of PM and extreme temperatures occur concurrently. Studies conducted in the European region using time series and case-crossover design have demonstrated indirect evidence of interaction between air pollution and heat, which is consistent with this study's results, i.e. effect are stronger in the warm season than the cold season.[57-58] Increased risk of child hospitalisation for respiratory diseases due to high temperatures have been reported by some authors,[59-61] which is also consistent with this study's findings for NO_2. The findings of the meta-analysis by Chen *et al*[42] reveals that moderate evidence exists that temperature modifies the effects of PM_{10} on mortality. For respiratory death, effect of PM_{10} on mortality is greatly boosted with high temperature and smallest with low temperature. Li *et al*[43] in their study reported that extreme temperature significantly increased the effect of PM_{10} on non-accidental and cardiovascular mortality, while the effect of PM_{10} on cardiovascular mortality was reduced with low temperature. This study further reveals that no interaction was observed between SO_2 and NO_2 at either high or low temperature. The study conducted by Liu *et al*[62] focused on air pollution and mortality in 652 cities across 24 countries and reported a positive and significant association between PM_{10} and $PM_{2.5}$ for all cause-mortality, which includes daily respiratory mortality. Also, a 10µg/m^3 of PM_{10} concentration was associated with an increase of 0.44% (0.39% - 0.50%) in a pooled estimate of daily all-cause mortality. Furthermore, an increase of 10µg/m^3 of PM_{10} for cause-specific analyses was associated with an increase of 0.47% in daily respiratory mortality.

Increased susceptibility to respiratory infections has also been traced to NO_2. Significant association between NO_2 and respiratory disease hospital admissions in this study was observed during warm days. This finding is in line with the study by Gao *et al*[40] where NO_2 and PM_{10} were associated with increased respiratory disease hospitalisation. Significant association between NO_2 concentrations in the ambient air and hospital admissions for chronic obstructive pulmonary disease have also been reported in studies conducted in India[22] and Hong Kong.[63]

The findings of the studies in Italy,[12] France[64] and Rome[52] established a positive association between NO_2 and respiratory conditions during warm seasons, which is consistent with the current study but in contrast with the results of Atkinson *et al*[65] and Villeneuve *et al*[51] where no significant association was reported. In line with the above, the reason for the inconsistency in the various studies could be as result of the quality and distribution of the air pollutants, which

may be affected by human activities, geographically characteristics and climatic conditions over a region.

In this study, no significant associations were observed for SO_2. The WHO guideline for SO_2 was only exceeded on three days. The results in this study are consistent with studies by Chen *et al*[20] and Shahi *et al*[66] conducted in China and Iran respectively; no association between respiratory hospital admission was found, despite the higher mean values for SO_2 reported, which is higher than the mean value for this study. However, the short study period of 4 years, small sample size and low mean value for SO_2 may contribute to the lack of association. Conversely, this study contrasts with others that reported an association between SO_2 and respiratory hospital admission. For instance, studies conducted in different provinces of China[49,67-68] reported that an increase of 10μg/m^3 of SO_2 led to increased respiratory disease hospitalisation. Dastoorpoor *et al*[69] also reported a significant relation between SO_2 and respiratory admission of suppurative and necrotic conditions of lower respiratory tract, which includes abscesses of the lung and mediastinum and pyothorax.

Findings have shown that children and elderly people are more susceptible to the effect of air pollution and this is consistent with the effect observed among children and elderly in the current study for PM_{10} and NO_2. The plausible explanation for this could be that children harbour undeveloped lung growth and host-defence capacity, and in elderly individuals, air pollution increases inflammatory responses of remodelled airways.[70-72] Furthermore, it was observed that the effect of NO_2 on hospital admission was higher than observed in PM_{10} during the warm season. This contrasts with a review study by Ab_Manan *et al*[73], who reported that PM (either $PM_{2.5}$ or PM_{10}) had a higher influence on cardiovascular or respiratory disease hospital admissions than other pollutants. In addition, when comparing the risk estimates between warm and cold season, this study found that stronger risk effect was observed during warm seasons, which is consistent with results from other studies.[74-76] Before the removal of NO_2 outliers, results showed that there was significant difference between males and females on cold days, after the sensitivity analysis (i.e. NO_2 outliers excluded) results showed no significant difference between males and females on cold days.

This study presents a clear seasonality of air pollution effects on human respiratory health. In relation to studies by Tramuto *et al*,[12] this can somewhat be explained because the warm seasons present the period when people devote their time largely to outdoor physical activities, resulting in higher respiratory volumes and exposure to ambient pollution, whereas during high polluted

days in winter, exposure is lower since outdoor activity time reduces and because of the application of air purifiers in homes and the workplace. In the meta-analysis by Zheng *et al*,[38] strong association between air pollutants and asthma-related hospital admissions for warm seasons was also reported, which also supports the findings of this study.

6.2.5 Strength and limitations

The contribution to existing knowledge of the association between air pollutants and hospital admissions in developing countries of the world, as a research gap identified in literature, was one of the strengths of this study.[77-78] Susceptibility by age and gender was also investigated. The data obtained from the group hospitals and the South African Weather Services also made it possible for us to apply models to investigate the association between hospital admissions and air pollutants (NO_2 and PM_{10}). The reliance on fixed-site environmental measurement could possibly introduce exposure misclassification. The influence of missing data on our estimates seems not to be significant[62]

The limitations of this study were as follows: Respiratory disease hospitalisation from only one city was investigated, which may limit the generalisability of the results to other cities or regions in South Africa. The researchers were not able to investigate data on effect modifiers or individual behaviours, which were possible confounders (such as tobacco use or environmental tobacco smoke). This information was not made available in the hospital admissions register.

The assumption that air pollutants and meteorological variables measured at a few sites were representative across the whole city introduces non-differential measurement error, which leads to associations that are biased towards being statistically insignificant.[79]

The study used data from three private hospitals and therefore the results cannot be generalised to the entire population in Pretoria. Private hospitals in South Africa are mainly attended by middle to upper socio-economic groups, which is around 17% of the population,[80] the vast majority of the population attends public hospitals. It is postulated that investigating data from public hospitals would include people from the lower socio-economic status and could potentially indicate stronger effects due to air pollution levels in Pretoria. Poverty and malnutrition may make those in the lower socio-economic status more susceptible to air pollution effects.

The hospitals used in this study belong to a group of private hospitals with many located across the country. The private hospital group manages 3 hospitals in Pretoria. As mentioned in the manuscript, public hospital data were not available for the study period and are still not available currently.

The form of selection bias introduced in this the study resulted from using private hospital and not public hospitals, i.e. bias which is introduced to how the study population was selected. However, the middle and higher SES classes have the privilege to acquire private health insurance in South Africa in order to access private treatment in the hospitals. It should be noted that not all these people were born into middle or higher SES families or homes, hence they also could have been born and bred in poor households and only moved into the middle and higher SES classes when they got jobs that included benefits such as private health insurance. It has been established that growing up in poverty predisposes people to health risks, even much later in life[81-83].

Another limitation was that the study period was short and could have contributed to the lack of observation of any significant associations.

6.2.6 Conclusions

In conclusion, no clear significant association was observed between air pollutants and hospital admissions for different age groups and genders on warm or cold days, except for NO_2, which had a strong association with hospital admissions on warm days.

6.2.7 Financial interests' declaration

None declared.

6.2.8 Acknowledgements

The authors would like to thank the South African Weather Services for the air pollution and meteorological data, the private hospital group for providing the hospital admission data, University of Pretoria and the National Research Foundation for the doctoral scholarship of A.A.A

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CHAPTER SEVEN: CONCLUSION AND RECOMMENDATIONS

7.1 Introduction

The author carried out this PhD project in Pretoria as part of a bigger project, which was also conducted in two other locations, namely; Cape Town[1] and Thohoyandou[2], from 18 April 2017 to 17 April 2018. Overall, this project aimed to investigate the characterisation of $PM_{2.5}$, source apportionment, origin of air masses into Pretoria during 18 April 2017 and 17 April 2018 alongside the association between hospital admissions due to respiratory diseases (RD), and air pollutants (PM_{10} and NO_2 add SO_2) over the period of January 2011 to November 2014.

This project had five objectives, namely:

1. Collection of $PM_{2.5}$ filter samples at one site in Pretoria over a period of one (1) year
2. To determine the soot, BC, OC and trace elements content of the collected $PM_{2.5}$ samples
3. To identify the sources of pollution that contributes to $PM_{2.5}$ in Pretoria using Positive Matrix Factorization (PMF).
4. To identify the geographical origin of air masses (surrogate for distant sources of $PM_{2.5}$) that pass Pretoria by applying the Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model.
5. To determine cases of the associations between air pollutants (PM_{10}, NO_2 and SO_2) and hospital admissions for respiratory diseases in Pretoria South Africa over the period of January 2011 to November 2014.

7.2 Main Findings

7.2.1 Collection of $PM_{2.5}$ filter samples in Pretoria over a period of one year

One hundred and forty-seven filters samples (122 samples and 25 duplicate samples) were collected. The mean annual concentration for $PM_{2.5}$ (21.1 ± 15.0 μg/m^3) for the study period was higher than the South African standard and doubled the WHO's annual limits concentration. Most daily exceedances were observed during winter and late autumn. The mean concentrations of the duplicate samples were higher than the samples for $PM_{2.5}$, soot, BC and OC respectively. No significant difference between the samples and the corresponding duplicate samples. There exists a significant difference for $PM_{2.5}$ levels observed across the

seasons, while no significant difference was observed between weekdays and weekend. The influence of meteorological data was noticed during the sampling period. Low temperature and wind speed have been associated with increased PM. Biomass burning, and other anthropogenic sources can be attributed to high $PM_{2.5}$ recorded during the winter.

7.2.2 To determine the soot, BC, OC and trace elements content of the collected $PM_{2.5}$ samples

Mean annual soot level was $2.3 \pm 2.0 \times 10^{-5} m^{-1}$. $PM_{2.5}$ and soot were well correlated as they followed the annual pattern. No significant difference was observed for soot levels by seasons, also no significant difference was observed between weekdays and weekend. The influence of meteorological data was noticed during the sampling period with wind speed taking the lead, followed by temperature. Mean annual of BC and OC were 3.9 ± 3.0 µg/m^3 and 3.3 ± 1.9 µg/m^3 respectively. BC and OC also followed the same pattern as $PM_{2.5,}$ where highest concentrations were recorded during winter, followed by autumn, and winter and lastly, summer. No significant difference was found between weekdays and weekends for both BC and OC.

Most of the trace elements also had the same seasonal trend as $PM_{2.5.}$ Elements such as S, Si, Fe, Ca and K had the highest concentration in winter than summer. Of the elements analysed in the aerosol samples, atmospheric S had the highest concentration while Si was the second most abundant species. Fe and K were third and fourth most abundant respectively. Relatively higher levels were also observed for Ca and Ni. Pb was found to be lower than the promulgated (0.25 µg/m^3) South Africa standard and WHO guideline (0.5 µg/m^3). Phasing in of unleaded petrol in South Africa could possibly be partially responsible for the low concentration recorded in this study. Variability of sources was observed during weekdays and weekends as there are significant differences. The basis for comparison with other studies in the region was a major limitation for $PM_{2.5}$ and other species as most study researched on PM_{10}.

7.2.3 To identify the sources of pollution that contribute to $PM_{2.5}$ in Pretoria using PMF

Pretoria is known to be an urban/industrial area and it is likely to have source categories influenced by anthropogenic and industrial activities. Sources of pollution that contribute to the $PM_{2.5}$ in Pretoria was achieved using the PMF model.

Seven sources of $PM_{2.5}$ were identified based on the variables used in the PMF model. The leading factor identified was secondary sulphur (31.4%) followed by coal burning (27.2%), soil dust (13.2%), vehicle exhaust (12.5%), vehicle exhaust and fossil fuel (8.6%), road traffic

(7.3%) and base metal/pyrometallurgical in that order. Secondary sulphur was found to be the major contributor with 30% of the total $PM_{2.5}$ level on a yearly average. $PM_{2.5}$ sources were found to have seasonal behaviour, where coal burning and secondary sulphur were significantly higher in winter compared to other seasons. Conversely, soil dust and road traffic were higher in autumn. The use of PMF in this study helped identified seven sources, which corroborate the known sources that have been identified in South Africa by other studies using different methods. However, these studies were conducted in different region of the country.

7.2.4 To identify the geographical origin of air masses that pass Pretoria by applying the HYSPLIT model

The origin of air masses that passed through Pretoria during 18 April 2017 and 17 April 2018, which is surrogate for distance sources of $PM_{2.5}$, was investigated using backward trajectories in the HYSPLIT model.

Five transport clusters pathways were identified where long-range transport (LRT) and local sources characterised these pathways. The cluster analysis showed that clusters 1 and 2 were local source origin, (i.e. North Limpopo and Eastern inland respectively) while 3 and 4 (Short and Long Indian Ocean respectively). Cluster 5 originated from the Atlantic Ocean which but loses it marine effect while passing through Western Cape region. These clusters depict LRT sources. For instance, the highest mean level for $PM_{2.5}$ (66.8 µg/m^3) which occurred on 2 June 2017, was found to be local source via which travel in the direction of cluster 1, while this region has been attributed to main sources of pollution into Pretoria apart from the influence of local sources. Furthermore, about 85% of the days in which the daily $PM_{2.5}$ WHO guideline and daily $PM_{2.5}$ South African standard were exceeded were of local source origin (clusters 1 and 2), while only 15% was attributed to LRT sources. Seasonally, cluster 1 was the prominent of the clusters during winter and autumn while cluster 2 were predominant in spring and summer. The average size of the impact from different source areas and source types at a receptor sites depends on the combination of source strength and frequency[3]. The possibilities of the air masses from Mozambique and local sources of Gauteng cannot be overlooked as contributing sources to the pollutant reaching the site.

The source apportionment results and the origin of air masses reaching the region can be applied in the review of the current 2008 City of Tshwane Air Quality Management Plan (AQMP). AQMPs need to be reviewed every 5 years according to the National Environmental Management: Air Quality Act of 2005.

7.2.5 To examine the associations between air pollutants and hospital admissions for respiratory diseases in Pretoria South Africa over the period January 2011 to November 2014

In total, 17 647 RD hospital admissions were included in this study. About equal numbers of male (n=8 500) and female (n=9 147) patients were admitted, and at most, 47 patients were admitted for RD in a day. The mean apparent temperature (Tapp) for the study period was 16.6°C. PM_{10} peaked at 170.2 $\mu g/m^3$. During the 1430-day study period the PM_{10} levels exceeded the WHO daily guideline value (50 $\mu g/m^3$) on 662 days.

No significant association between RD hospital admissions and air pollution (PM_{10}, NO_2 and SO_2) was found across the entire range of apparent temperature (Tapp) (unstratified analysis).

However, on warm days, there was a significant increase of 19.0% (2.3%; 38.4%) for RD hospital admissions for NO_2. The association between PM_{10} and RD hospital admissions was stronger for women than men on warm days, while the most marked association for PM_{10} was observed in the age group >65 years. No significant association between NO_2 and RD hospital admissions was observed for females and male patients during warm and cold days.

Although no significant associations were observed between NO_2 and RD hospital admission for males and females on cold days, after adjusting for PM_{10}, males were significantly less vulnerable than females to increasing NO_2 levels (interaction term p-value < 0.05). The same was observed in the one pollution models. No vulnerable groups (i.e. no significant interactions) were observed for SO_2 or PM_{10} in the two pollution models.

This study is one of the few studies in Africa that investigated the interaction between temperature and air pollution and identifying vulnerable groups in the population.

7.3 Potential bias and limitations

The limitations of this study were as follow; firstly, RD hospitalisation from only one city was investigated, which may limit the generalisability of the results to other cities or regions in South Africa.

Secondly, the researcher was not able to investigate data on effect modifiers or individual behaviours which are possible confounders (such as tobacco use or environmental tobacco smoke). This information was not made available in hospital admissions register. Thirdly, the assumption that air pollutants and meteorological variables measured at a few sites are representative across the whole city which leads to nondifferential measurement error and

associations that are biased towards being statistically insignificant[4]. Also, error due to exposure measurement cannot be overruled, i.e. deficiencies that may occur during ambient monitoring. Since personal exposure monitoring will be different from data in a systematic way.

The form of selection bias introduced in this the study resulted from using private hospital and not public hospitals, i.e. bias which is introduced to how the study population was selected. However, the middle and higher SES classes have the privilege to acquire private health insurance in South Africa in order to access private treatment in the hospitals. It should be noted that not all these people were born into middle or higher SES families or homes, hence they also could have been born and bred in poor households and only moved into the middle and higher SES classes when they got jobs that included benefits such as private health insurance. It has been established that growing up in poverty predisposes people to health risks[81-83]

Another limitation was that the study period was short and could have contributed to the lack of observation of any significant associations. The study used data from three private hospitals and therefore the results cannot be generalised to the entire population in Pretoria. Private hospitals in South Africa are mainly attended by middle to upper socio-economic groups, which is around 17% of the population [8] Most of the population attends public hospitals. It is postulated that investigating data from public hospitals would include people from the lower socio-economic class and could potentially indicate stronger effects due to air pollution levels in Pretoria. Poverty and malnutrition may make the lower socio-economic groups more susceptible to air pollution effects[9].

No significant association was found with PM_{10} and SO_2 except for NO_2, this may be due to the short study period as it depended on the availability of air pollution data.

There is an assumption that the meteorological parameters measured at the government monitoring stations are the same across the locations might have resulted in measurement error. Missing data from these monitoring station also pose some limitation to the study. Inadequate local studies to compare with this study also pose a limitation for the interpretation of the findings.

7.4 Strengths

The contribution to existing knowledge of association between air pollutants and RD hospital admissions in developing countries of the world, as a research gap identified in literature constitute one of the strengths of this study[10-11]. Also, susceptibility by age and sex was investigated. The data obtained from the group hospitals and the South African Weather Services also made it possible for us to apply models to investigate the association between hospital admissions and air pollutants (NO_2, SO_2 and PM_{10}).

7.5 General recommendations

The South African Government at all levels should look into policies and investments that will support cleaner transport, energy-efficient housing, power generation, industry and better municipal waste management to reduce key sources of urban outdoor air pollution based on the identified contributing sources to air pollution in this study (SDG Goal 11.6).[12]

Government should incorporate enforceable stringent rules for all stakeholders in the new act to mitigate air pollution as the day-to-day human activities contribute to air pollution emissions thereby contributing to climate change.

Government at all levels should increase the public awareness campaign programme on climate change and its adverse effects on human health effects (SDG Goal 13).

Although there was no significant interaction between air pollutants and RD hospital admissions probably due to the short period of this study and private hospitals used, further research that includes a longer study period and both private and public hospitals should be encouraged.

Government should encourage and provide proper data capturing facilities to all public hospitals, this would allow robust epidemiological studies that would investigate the association between air pollution and human health.

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2. Novela, RJ. Quantification of fine particulate matter, soot in ambient air and the geographical origin of the air masses passing through Thohoyandou. MSc (Environmental Sciences). University of Venda, South Africa, 2019
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Appendix

Appendix 1: Letter of approval from the Academic Advisory Committee

UNIVERSITEIT VAN PRETORIA
UNIVERSITY OF PRETORIA
YUNIBESITHI YA PRETORIA

Faculty of Health Sciences

29 September 2017

MR ADEWALE ADEKUNLE ADEYEMI
(17299340)
PhD (Environmental Health)

Dear Mr Adeyemi

Approval Academic Advisory Committee: Protocol

This letter confirms that your PhD protocol was served and approved by the Research Committee on 26 September 2017 after a successful protocol defence. One internal and two external reviewers attended your PhD protocol defence on 13 June 2017 and also reviewed your PhD protocol. All three reviewers approved the corrections that were made.

Your PhD protocol was subsequently served and approved also by the Academic Advisory Committee on 29 September 2017.

Your PhD project title is: **$PM_{2.5}$ chemical composition, source apportionment and geographical origin of air masses in Pretoria, South Africa**

You can now submit to the Faculty of Health Sciences Research Ethics Committee.

Sincerely

Prof Brendan Girdler-Brown
Acting chairperson
SHSPH Academic Advisory Committee

cc Prof Janine Wichmann (supervisor), Dr Peter Molnàr (co-supervisor)

AAC members: Prof Debashis Basu, Prof Stephen Hendricks, Dr Neo Ledibane, Prof Halina Rollin, Dr Flavia Senkubuge, Mrs Annette Welman, Prof Janine Wichmann (Chairperson), Mrs Kathy Pieterse

Email address shsph@up.ac.za
http://shsph.up.ac.za
www.up.ac.za
http://www.facebook.com/SHSPH

Appendix 2: Letter of approval (Amendment) from the Academic Advisory Committee

UNIVERSITEIT VAN PRETORIA
UNIVERSITY OF PRETORIA
YUNIBESITHI YA PRETORIA

Faculty of Health Sciences

16 April 2019

A A Adeyemi
17299340
PhD Environmental Health

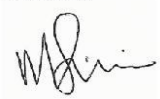
Dear Mr Adeyemi

SHSPH Academic Advisory Committee: Protocol

Your protocol titled "*PM2.5 chemical composition, source apportionment and geographical origin of air masses in Pretoria, South Africa*" *with minor changes* served at the Academic Advisory Committee (AAC) via e-mail on 11 April 2019 and was approved.

You can now submit to ethics.

Sincerely



Dr J Shirinde
Chairperson
SHSPH Academic Advisory Committee

cc Prof J Wichmann

Appendix 3: Letter of approval (New Application) from Research Ethics Committee

The Research Ethics Committee, Faculty Health Sciences, University of Pretoria complies with ICH-GCP guidelines and has US Federal wide Assurance.
- FWA 00002567, Approved dd 22 May 2002 and Expires 03/20/2022.
- IRB 0000 2235 IORG0001762 Approved dd 22/04/2014 and Expires 03/14/2020.

UNIVERSITEIT VAN PRETORIA
UNIVERSITY OF PRETORIA
YUNIBESITHI YA PRETORIA

Faculty of Health Sciences Research Ethics Committee

26/10/2017

Approval Certificate
New Application

Ethics Reference No: 469/2017

Title: PM2.5 chemical composition, source apportionment and geographical origin of air masses in Pretoria, South Africa

Dear Mr Adewale Adeyemi

The **New Application** as supported by documents specified in your cover letter dated 28/09/2017 for your research received on the 28/09/2017, was approved by the Faculty of Health Sciences Research Ethics Committee on its quorate meeting of 25/10/2017.

Please note the following about your ethics approval:
- Ethics Approval is valid for 3 years
- Please remember to use your protocol number (**469/2017**) on any documents or correspondence with the Research Ethics Committee regarding your research.
- Please note that the Research Ethics Committee may ask further questions, seek additional information, require further modification, or monitor the conduct of your research.

Ethics approval is subject to the following:
- The ethics approval is conditional on the receipt of **6 monthly written Progress Reports**, and
- The ethics approval is conditional on the research being conducted as stipulated by the details of all documents submitted to the Committee. In the event that a further need arises to change who the investigators are, the methods or any other aspect, such changes must be submitted as an Amendment for approval by the Committee.

Additional Conditions:
- Approval is conditional upon the Research Ethics Committee receiving approval letter from the Scientific Committee.

We wish you the best with your research.

Yours sincerely

Dr R Sommers; MBChB; MMed (Int); MPharMed,PhD
Deputy Chairperson of the Faculty of Health Sciences Research Ethics Committee, University of Pretoria

The Faculty of Health Sciences Research Ethics Committee complies with the SA National Act 61 of 2003 as it pertains to health research and the United States Code of Federal Regulations Title 45 and 46. This committee abides by the ethical norms and principles for research, established by the Declaration of Helsinki, the South African Medical Research Council Guidelines as well as the Guidelines for Ethical Research: Principles Structures and Processes, Second Edition 2015 (Department of Health).

☎ 012 356 3084 deepeka.behari@up.ac.za / fhsethics@up.ac.za http://www.up.ac.za/healthethics
✉ Private Bag X323, Arcadia, 0007 - Tswelopele Building, Level 4, Room 60, Gezina, Pretoria

Appendix 4: Letter of approval (Annual Renewal) from Research Ethics Committee

UNIVERSITEIT VAN PRETORIA
UNIVERSITY OF PRETORIA
YUNIBESITHI YA PRETORIA

Faculty of Health Sciences

The Research Ethics Committee, Faculty Health Sciences, University of Pretoria complies with ICH-GCP guidelines and has US Federal wide Assurance.

- FWA 00002567, Approved dd 22 May 2002 and Expires 03/20/2022.
- IRB 0000 2235 IORG0001762 Approved dd 22/04/2014 and Expires 03/14/2020.

10 April 2019

Approval Certificate
Annual Renewal

Ethics Reference No.: 469/2017
Title: PM2.5 chemical composition, source apportionment and geographical origin of air masses in Pretoria, South Africa.

Dear Mr AA Adeyemi

The **Annual Renewal** as supported by documents received between 2019-03-12 and 2019-04-10 for your research, was approved by the Faculty of Health Sciences Research Ethics Committee on its quorate meeting of 2019-04-10.

Please note the following about your ethics approval:

- Renewal of ethics approval is valid for 1 year, subsequent annual renewal will become due on 2020-04-10.
- Please remember to use your protocol number (469/2017) on any documents or correspondence with the Research Ethics Committee regarding your research.
- Please note that the Research Ethics Committee may ask further questions, seek additional information, require further modification, monitor the conduct of your research, or suspend or withdraw ethics approval.

Ethics approval is subject to the following:

- The ethics approval is conditional on the research being conducted as stipulated by the details of all documents submitted to the Committee. In the event that a further need arises to change who the investigators are, the methods or any other aspect, such changes must be submitted as an Amendment for approval by the Committee.

We wish you the best with your research.

Yours sincerely

Dr R Sommers
MBChB MMed (Int) MPharmMed PhD
Deputy Chairperson of the Faculty of Health Sciences Research Ethics Committee, University of Pretoria

The Faculty of Health Sciences Research Ethics Committee complies with the SA National Act 61 of 2003 as it pertains to health research and the United States Code of Federal Regulations Title 45 and 46. This committee abides by the ethical norms and principles for research, established by the Declaration of Helsinki, the South African Medical Research Council Guidelines as well as the Guidelines for Ethical Research: Principles Structures and Processes, Second Edition 2015 (Department of Health)

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Fakulteit Gesondheidswetenskappe
Lefapha la Disaense tša Maphelo

Appendix 5: Letter of approval (Amendment) from Research Ethics Committee

UNIVERSITEIT VAN PRETORIA
UNIVERSITY OF PRETORIA
YUNIBESITHI YA PRETORIA

Faculty of Health Sciences

The Research Ethics Committee, Faculty Health Sciences, University of Pretoria complies with ICH-GCP guidelines and has US Federal wide Assurance.

- FWA 00002567, Approved dd 22 May 2002 and Expires 03/20/2022.
- IRB 0000 2235 IORG0001762 Approved dd 22/04/2014 and Exoires 03/14/2020.

17 May 2019

Approval Certificate
Amendment

Ethics Reference No.: 469/2017
Title: PM2.5 chemical composition, source apportionment and geographical origin of air masses in Pretoria, South Africa.

Dear Mr AA Adeyemi

The **Amendment** as supported by documents received between 2019-05-07 and 2019-05-17 for your research, was approved by the Faculty of Health Sciences Research Ethics Committee on its quorate meeting of 2019-05-15.

Please note the following about your ethics approval:

- Please remember to use your protocol number (469/2017) on any documents or correspondence with the Research Ethics Committee regarding your research.
- Please note that the Research Ethics Committee may ask further questions, seek additional information, require further modification, monitor the conduct of your research, or suspend or withdraw ethics approval.

Ethics approval is subject to the following:

- The ethics approval is conditional on the research being conducted as stipulated by the details of all documents submitted to the Committee. In the event that a further need arises to change who the investigators are, the methods or any other aspect, such changes must be submitted as an Amendment for approval by the Committee.

We wish you the best with your research.

Yours sincerely

Dr R Sommers
MBChB MMed (Int) MPharmMed PhD
Deputy Chairperson of the Faculty of Health Sciences Research Ethics Committee, University of Pretoria

The Faculty of Health Sciences Research Ethics Committee complies with the SA National Act 61 of 2003 as it pertains to health research and the United States Code of Federal Regulations Title 45 and 46. This committee abides by the ethical norms and principles for research, established by the Declaration of Helsinki, the South African Medical Research Council Guidelines as well as the Guidelines for Ethical Research: Principles Structures and Processes, Second Edition 2015 (Department of Health).

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Appendix 6: Letter of approval from Mediclinic hospital

MEDICLINIC

MEDICLINIC OFFICES
STRAND ROAD
STELLENBOSCH
7600

PO BOX 456
STELLENBOSCH
7599

T +27 21 809 6500
F +27 21 809 6756
ETHICS LINE 0800 005 316

www.mediclinic.co.za

18 September 2017

To whom it may concern

Re: **DATA FOR RESEARCH PURPOSES**

This letter confirms that Mediclinic Southern Africa provided data (from 1 January 2011 to 31 December 2016) from our hospitals to Professor Janine Wichmann for analysis in her study on the effects of temperature on cardiovascular and respiratory disease.

Kind regards

Dr Chris du Plessis
General Manager Clinical Services
MEDICLINIC SOUTHERN AFRICA

MEDICLINIC (PTY) LTD
REG. NO. 1969/009218/07

Appendix 7: Standard operating procedure for weighing filter

FILTER WEIGHING PROCEDURE

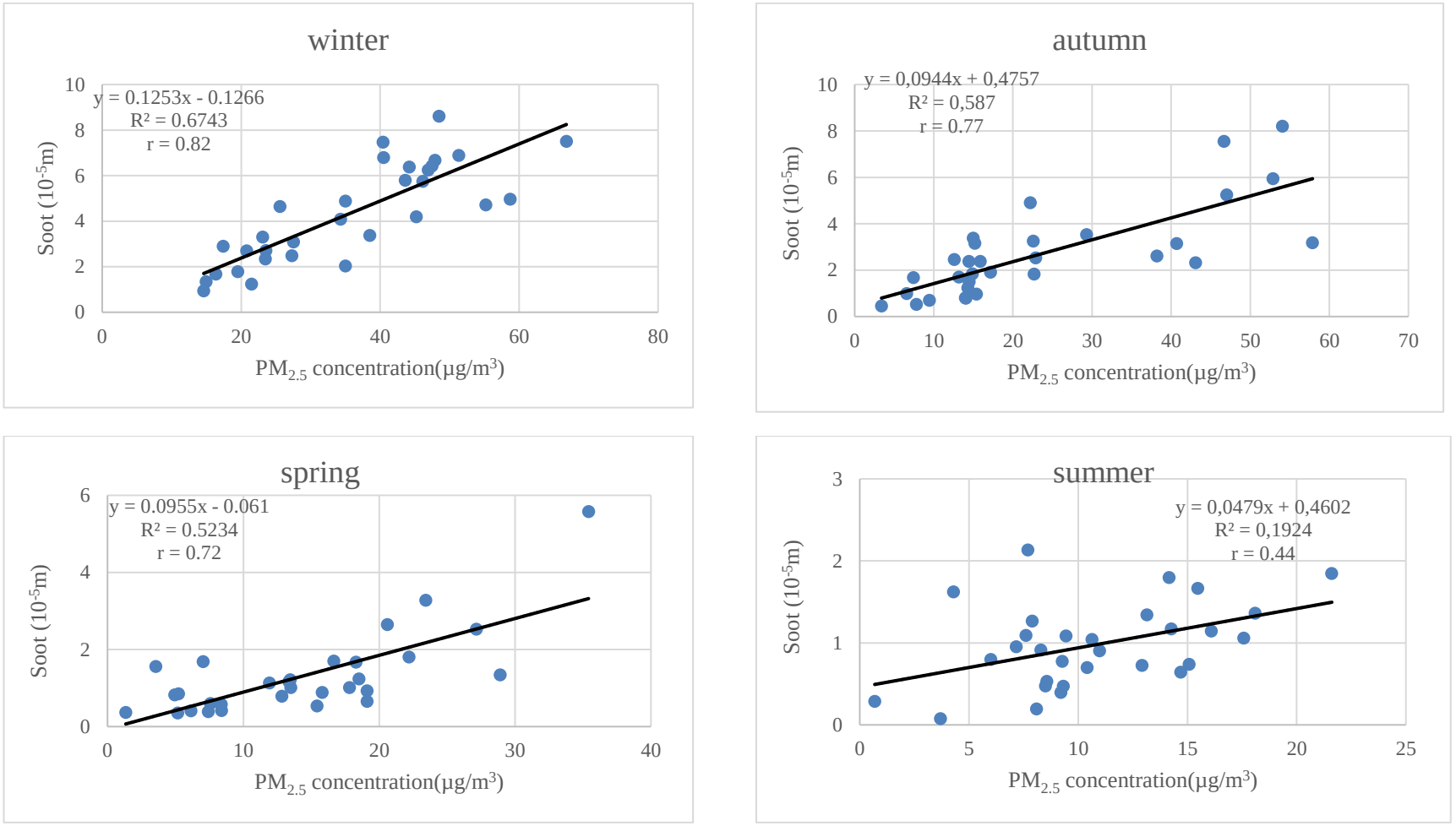
Mettler-Toledo ultra-microbalance weighing procedure: Reference (control) and sample filters

1. Capture data from laboratory environmental monitor
2. Check if environmental conditions in the laboratory were maintained for the previous 24 hours within prescribed limits
    - Dry air temperature: 21 ± 1.0 °C
    - Relative humidity: 50 ± 5 %
3. Capture environmental conditions immediately prior to weighing reference (control) filters
4. Ensure balance is level
5. Tare balance
6. Weigh a 2 gram weight (certified weight set) repeatedly until repeatability is reached. Repeatability is when the min. and max. readings of three (3) consecutive measurements do not differ from the mean by more than one (1) percent
7. When repeatability is reached, remove weight
8. Close weighing chamber door and tare balance
9. Open balance and place three (3) reference filters on the weighing grid.
10. Close balance and start 30 second countdown timer. Allow 30 seconds for balance to stabilise. Note: Record reading on balance immediately when settling time is reached
11. Record three (3) consecutive readings for the reference filters
12. Remove reference filters from the weighing chamber and hold it next to the chamber with a flat-nosed tweezers. Note: Do not breathe over the filters
13. Close weighing chamber door and wait for balance to return to zero (0)
14. If balance does not return to zero (0), do the following:
    - Reject all prior mass measurements
    - Inspect balance pan for dust or any other obstacles
    - Reweigh filters
15. Repeat steps 9-14 for 10 individual sample filters. Note: Reference filters are reweighed after every 10 samples
16. Record environmental conditions immediately after the final reference filters are weighed. Note: If min. and max. readings of reference filters differ by > 10-3 (0.001). percent from the mean – reject all readings preceding it and reweigh samples
17. Place weighed filter samples on clearly labelled support pads in clean petri slides

Appendix 8: Standard operating procedure for soot measurement

DETERMINATION OF SOOT (ABSORPTION COEFFICIENT) USING REFLECTROMETRIC METHOD (SOP 4.0)

1. Clean the measuring head, mask and standard plate with an alcohol swab.
2. Connect measuring head to the reflectometer and switch it on. Note: Allow lamp to warm up for 30 minutes
3. After 30 minutes, disconnect measuring head and adjust reading to 0.0 using the zero knob
4. Tightly attach the mask to the measuring head and reconnect it to the reflectometer
5. Place measuring head over white standard of standard plate and adjust reading to 100.0 using the coarse and fine knobs.
6. Once completed, move the measuring head to the grey standard (for the EEL model 43 D it should read 33.5 ± 1.5). Repeat Step.5 if out-of-limits
7. Perform reflectance measurements on five (5) reference (control) filters. Each filter is measured in quintuplicate (5x) using the five-point method. Note: Do not readjust the reflectometer between filters
8. Calculate the mean (μ) of each reference filter. The filter with the median mean (M) is selected as the primary control filter. Note: This step is only performed in the first measurement session. The primary control filter must be stored safely and used is to adjust the reflectometer to 100.0 in subsequent sessions
9. Proceed with filter sample measurements
10. Using a flat-nosed tweezers, remove the sample from petri slide and place it centrally over the white standard of standard plate
11. Carefully place the measuring head on the sample and record reading. Repeat measurements using five-point method and record readings
12. Repeat reflectometer calibration using the primary control filter after every 25 sample measurements. Record reading of control filter before adjustments (if required). Note: Control filter should read 98-102
13. Clean mask, standard plate and tweezers after every 25 samples (with recalibration)
14. At the end of each measurement session, repeat the measurement of 10 % of the samples. Note: If the mean of the replicate measurements deviates by more than ± three (3) percent from the original readings, all samples in measured during the corresponding session must be measured again
15. If new boxes of filters are opened and used, record lot numbers and check reflectance of blank filter. Note: The mean of the blank filter must be within ± two (2) units of the primary control filter. If not, a separate mean needs to be calculated.



Appendix 9: The relationship between $PM_{2.5}$ and soot levels measured at the School of Health Systems and Public Health, University of Pretoria in Pretoria during 18 April 2017 and 17 April 2018 for, clockwise from top left, winter, autumn, spring and summer

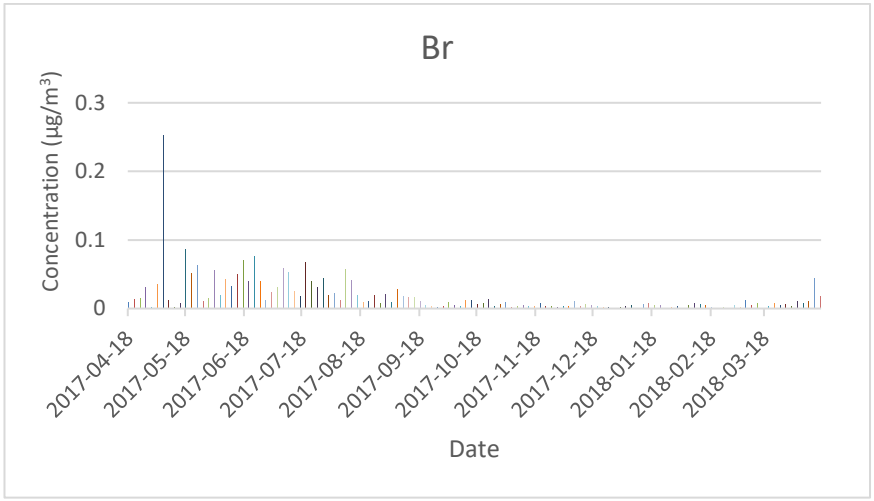
Appendix 10: Month and seasonal wind direction information in Pretoria during 18 April 2017 and 17 April 2018

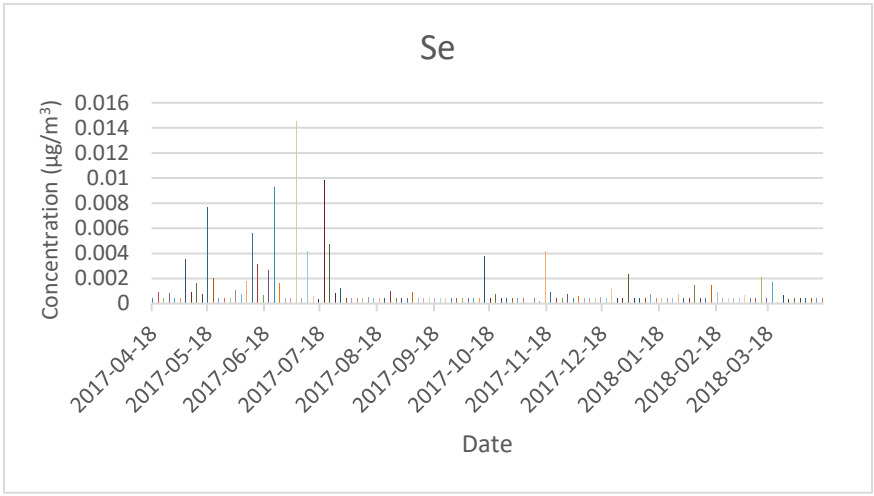
Mnth	N	NNE	NE	ENE	E	ESE	SE	SSE	S	SSW	SW	WSW	W	WNW	NW	NNW	T/O	E/D	W/D	%E	%W	O/D	%O/D	S/D	Wknd	%Wknd	Wkdys	%Wkdys
Apr-17	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0
May	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0
Jun	0	0	0	0	0	0	0	7	0	0	0	0	0	0	0	0	7	0	0	0	0	7	100	1	2	2	5	5
Jul	0	0	0	0	0	0	0	9	0	0	0	0	0	0	0	0	9	0	0	0	0	9	100	1	3	3	6	6
Aug	0	0	0	0	1	0	1	0	0	0	0	1	1	0	0	0	4	1	2	25	50	1	25	4	0	0	4	4
Sep	0	0	0	1	0	3	3	0	0	0	2	1	0	0	0	0	10	4	1	40	10	5	50	5	4	4	6	6
Oct	0	0	0	3	1	0	0	0	0	0	1	2	2	0	0	0	9	4	4	44	44	1	12	5	2	2	7	7
Nov	0	0	0	1	2	0	0	0	0	0	0	3	4	0	0	0	10	3	7	30	70	0	0	4	3	3	7	7
Dec	0	0	0	1	4	0	0	0	0	0	0	2	3	0	0	0	10	5	5	50	50	0	0	4	3	3	7	7
Jan-18	0	0	0	1	5	0	1	0	0	0	0	1	3	0	0	0	11	6	4	55	36	1	9	5	3	3	8	8
Feb	0	0	0	2	3	1	0	0	0	0	0	1	2	0	0	0	9	6	3	67	33	0	0	5	3	3	6	6
Mar	0	0	0	0	1	0	0	0	0	0	0	3	6	0	0	0	10	1	9	10	90	0	0	3	2	2	8	8
Apr	0	0	0	2	0	0	0	0	0	0	0	1	3	0	0	0	6	2	4	33	67	0	0	3	2	2	4	4
mean	0	0	0	0.8	1	0.3	0	1	0	0	0	1.2	2	0	0	0	7.3	2.5	3	27.2	34.6	1.8	22.8	3.1	2.1	2.2	5.2	5.5
Season	N	NNE	NE	ENE	E	ESE	SE	SSE	S	SSW	SW	WSW	W	WNW	NW	NNW	T/O	E/D	W/D	%E	%W	O/D	%O/D	S/D	Wknd	%Wknd	Wkdys	%Wkdys
Autumn	0	0	0	2	1	0	0	0	0	0	0	4	9	0	0	0	16	3	13	19	81	0	0	4	4	4	12	13
Winter	0	0	0	0	1	0	1	16	0	0	0	1	1	0	0	0	20	1	2	5	10	17	85	5	5	5	15	16
Spring	0	0	0	5	3	3	3	0	0	0	3	5	7	0	0	0	29	11	12	38	41	6	21	7	9	10	20	21
Summer	0	0	0	4	0	1	1	0	0	0	0	4	8	0	0	0	30	5	12	17	40	13	43	6	9	10	21	22
mean	0	0	0	2.8	1	1	1	4	0	0	1	3.5	6	0	0	0	24	5	10	20	43	9	37	6	6.8	7.1	17	17.9

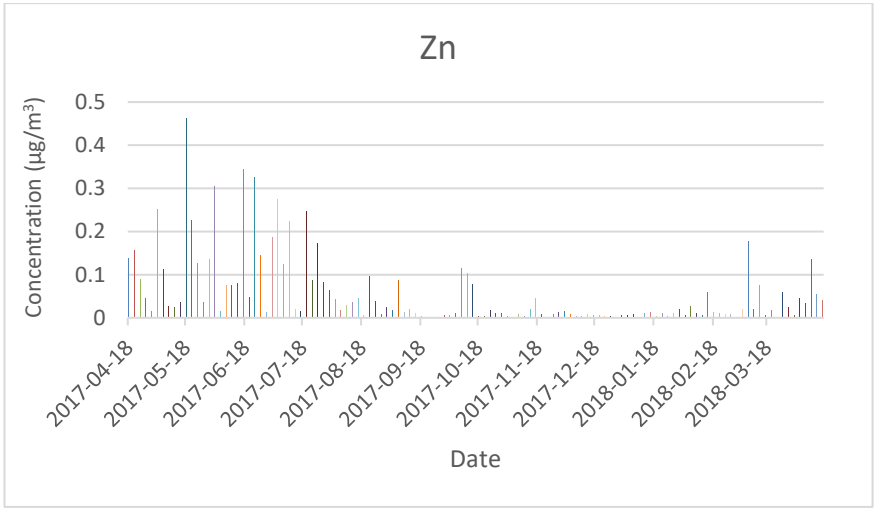
Appendix 11: Mean $PM_{2.5}$, soot, BC and OC levels for the three categories of temperature, relative humidity and wind speed in Pretoria during 18 April 2017 and 17 April 2018

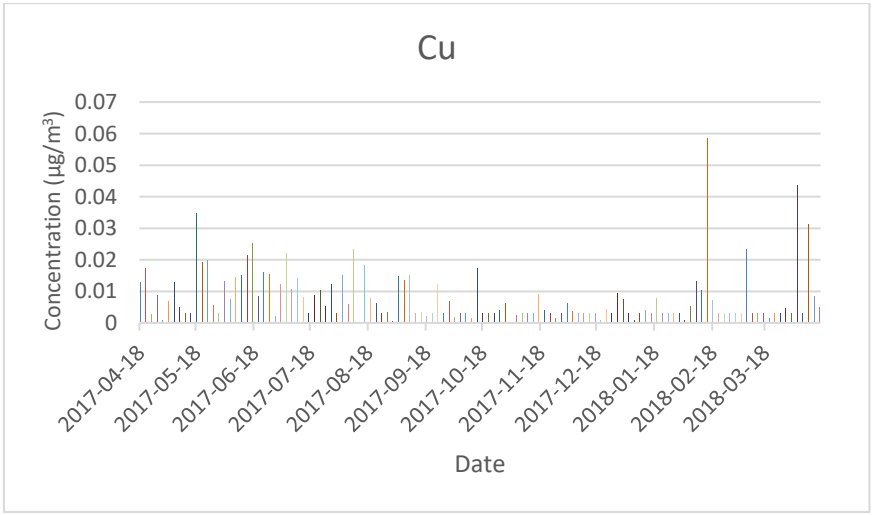
Temperature (ºC)	**<16**	**16 – 20**	**>20**
$PM_{2.5}$ concentration (µg/m^3)	32.6	13.1	14.6
Soot (10^{-5}m^{-1})	4.1	1.3	1.2
BC (µg/m^3)	6.5	2.3	2.3
OC (µg/m^3)	4.9	2.2	2.3
Number of samples	44	40	34
Relative humidity (%)	**<65**	**65 - 75**	**>75**
$PM_{2.5}$ concentration (µg/m^3)	21.9	18.3	16.8
Soot **(**10^{-5}m^{-1})	2.4	2.0	2.4
BC (µg/m^3)	4.0	3.3	4.1
OC (µg/m^3)	3.4	2.8	3.2
Number of samples	86	26	6
Wind speed (m/s)	**<1.5**	**1.5 – 2.5**	**>2.5**
$PM_{2.5}$ concentration (µg/m^3)	15.1	21.2	9.1
Soot (10^{-5}m^{-1})	1.6	2.3	1.3
BC (µg/m^3)	2.7	3.9	2.3
OC (µg/m^3)	2.5	3.4	2.4
Number of samples	41	51	4

Appendix 12: Cl, Pb, As and Sb content in $PM_{2.5}$ levels measured at the School of Health Systems and Public Health, University of Pretoria during 18 April 2017 to 17 April 2018.

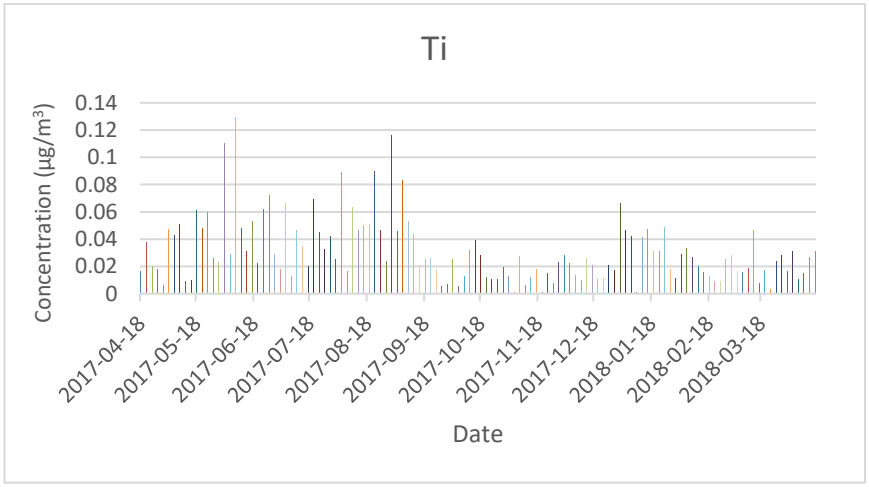


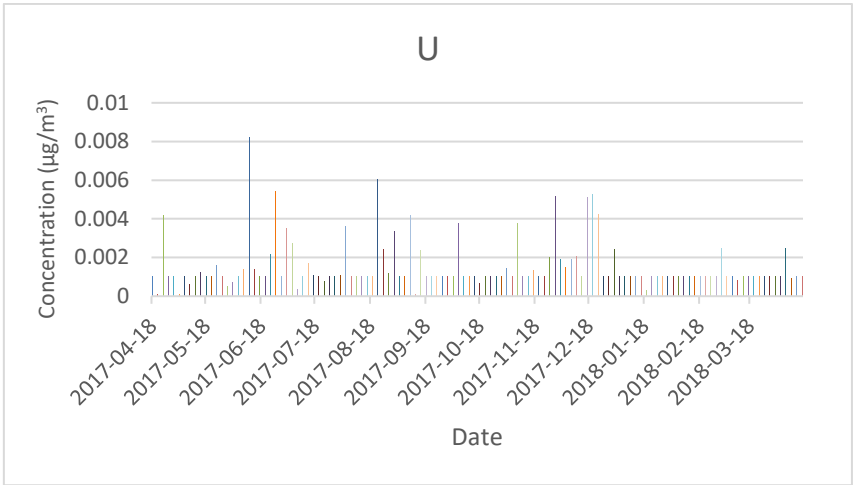




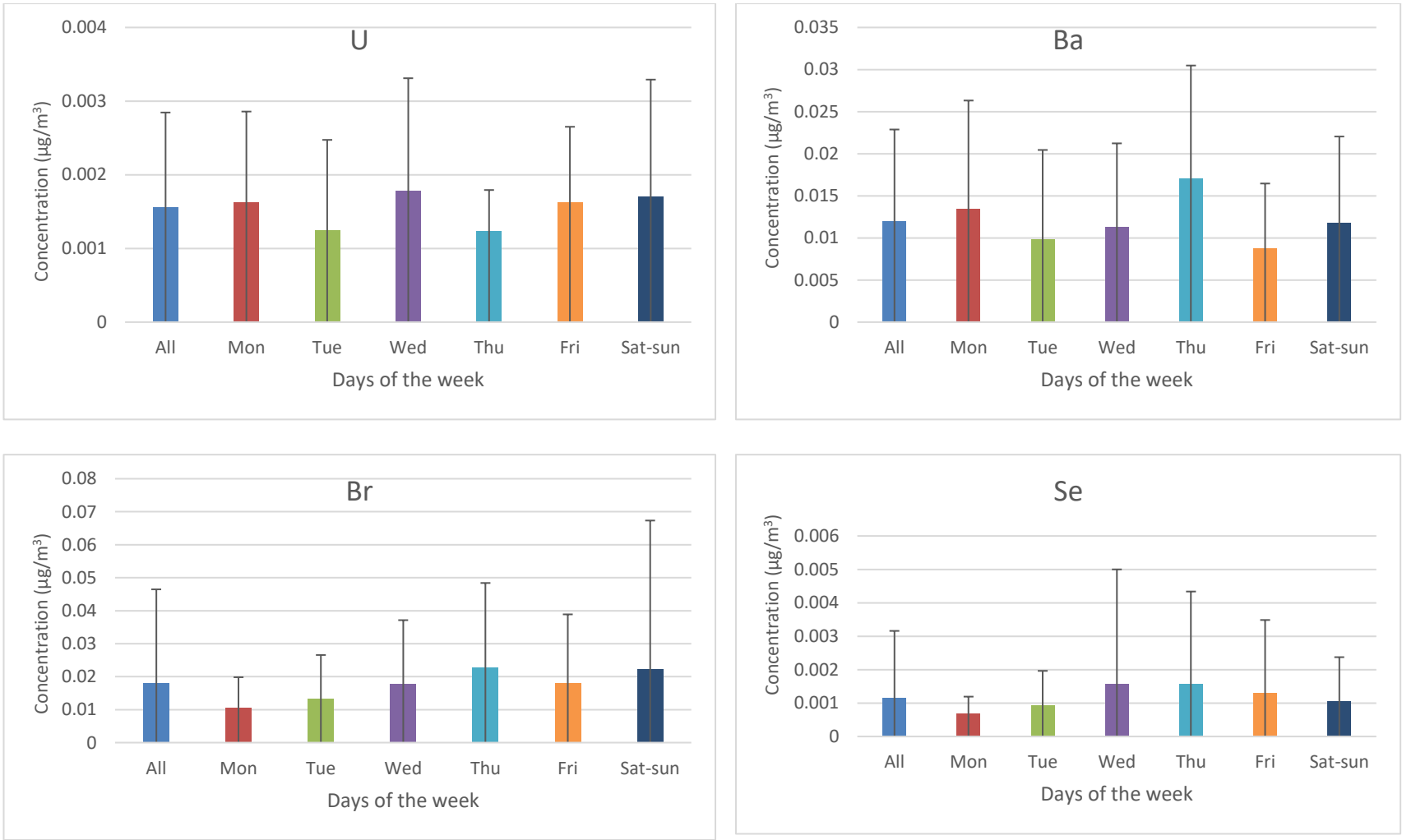


Appendix 13: Br, Se, Zn and Cu content in $PM_{2.5}$ levels measured at the School of Health Systems and Public Health, University of Pretoria during 18 April 2017 to 17 April 2018.

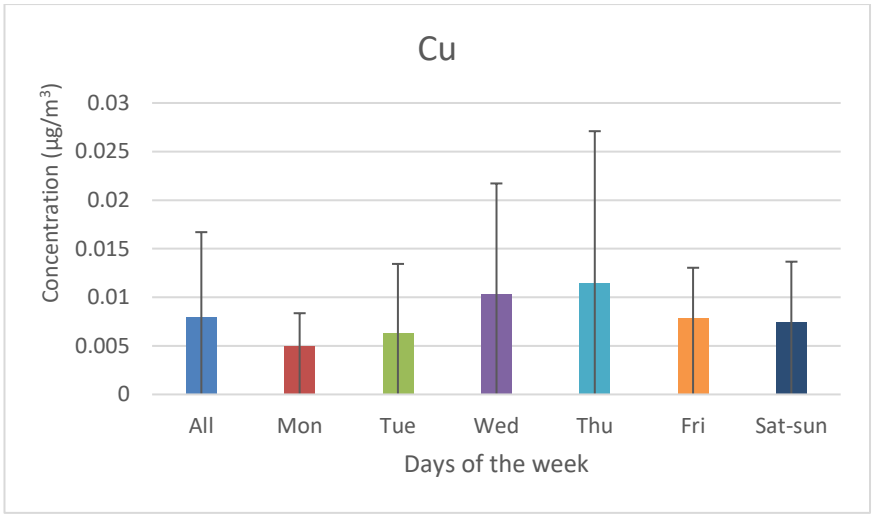


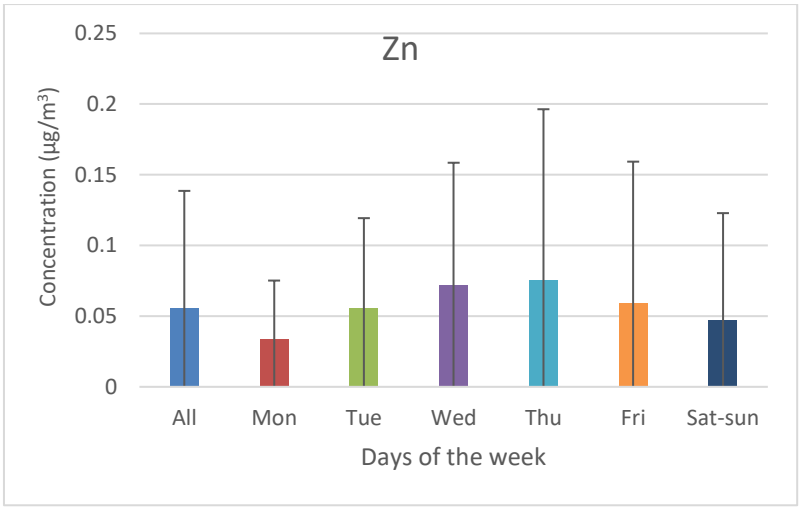


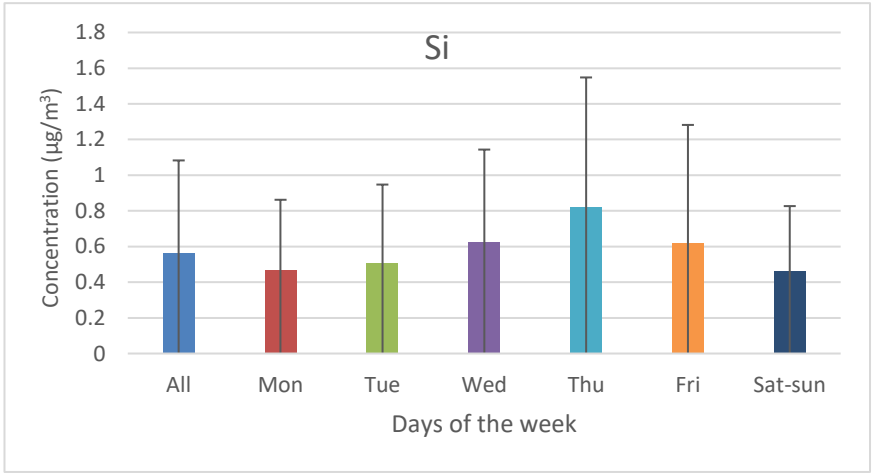
Appendix 14: Graph of Ti and U content in $PM_{2.5}$ levels measured at the School of Health Systems and Public Health, University of Pretoria during 18 April 2017 to 17 April 2018.

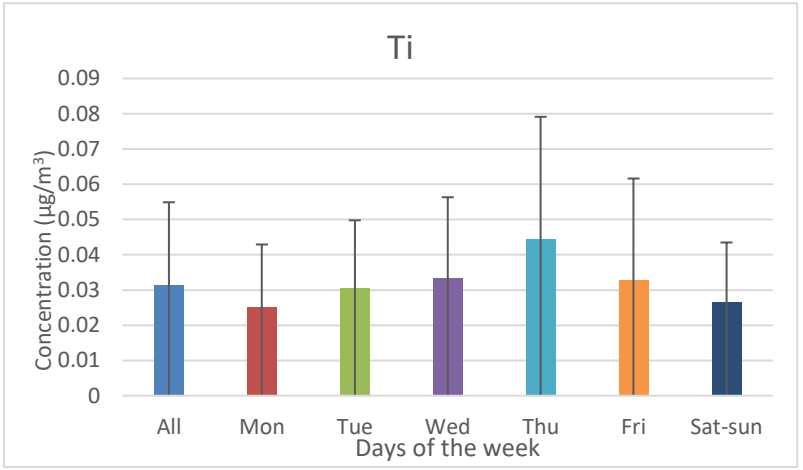


Appendix 15: U, Ba, Br and Se content in $PM_{2.5}$ levels measured at the School of Health Systems and Public Health, University of Pretoria during 18 April 2017 to 17 April 2018.

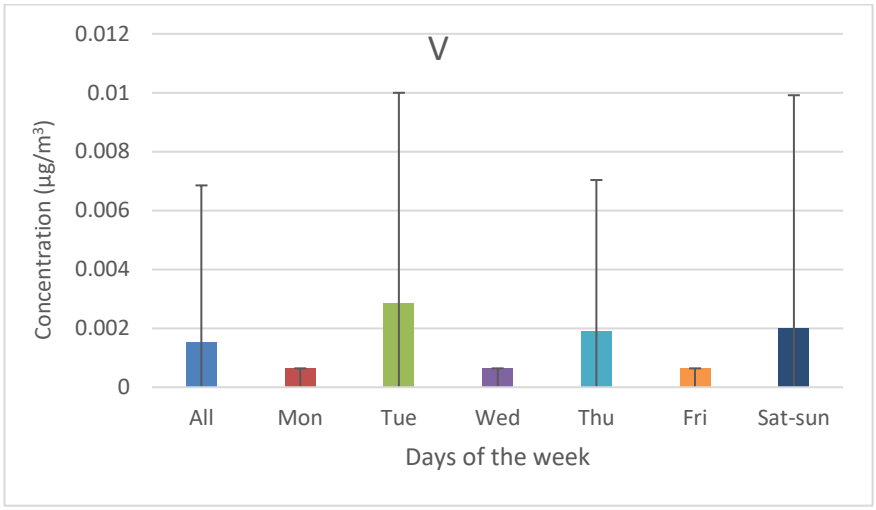


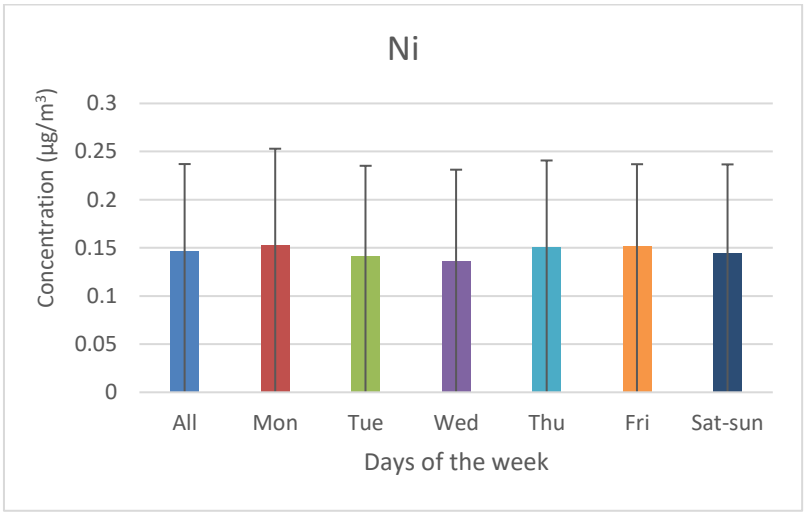




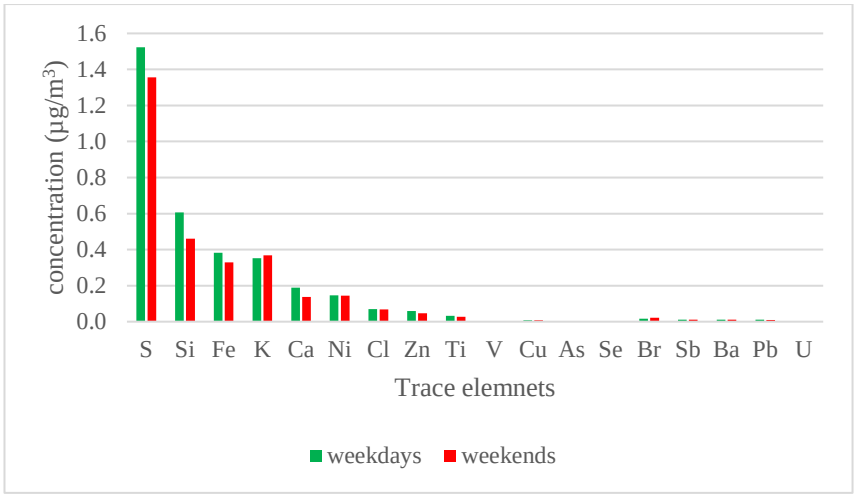


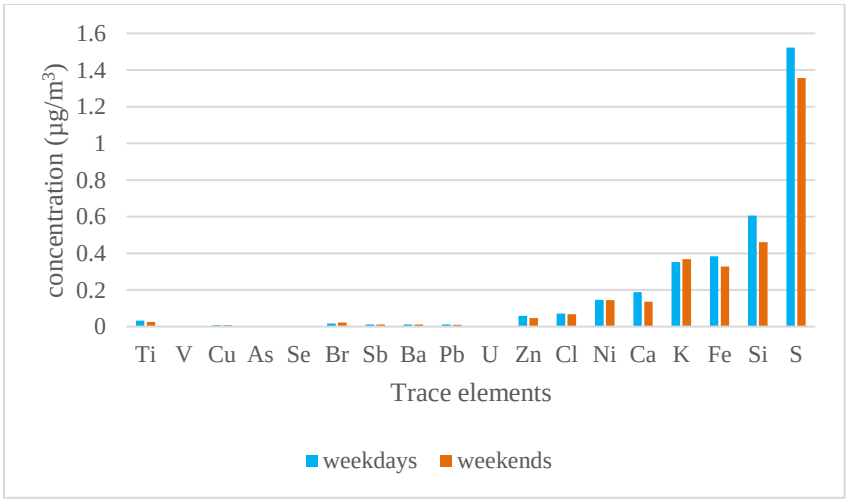
Appendix 16: U, Ba, Br and Se content in $PM_{2.5}$ levels measured at the School of Health Systems and Public Health, University of Pretoria during 18 April 2017 to 17 April 2018.

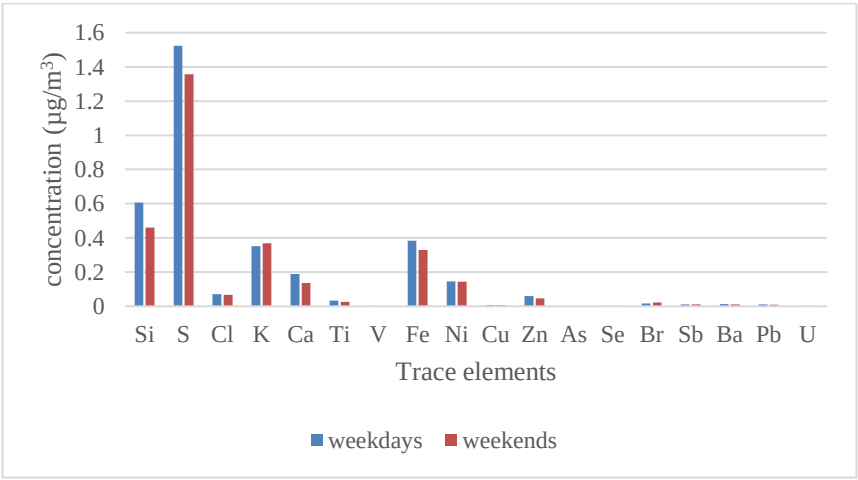




Appendix 17: U, Ba, Br and Se content in $PM_{2.5}$ levels measured at the School of Health Systems and Public Health, University of Pretoria during 18 April 2017 to 17 April 2018.







Appendix 18: Trace elemental content of PM2.5 levels measured at the School of Health Systems and Public Health, University of Pretoria during 18 April 2017 to 17 April 2018 by weekdays and weekends

Appendix 19: Certificate from the Language Editor I

Gill Smithies

Proofreading & Language Editing Services

59, Lewis Drive, Amanzimtoti, 4126, Kwazulu Natal

Cell: 071 352 5410 E-mail: g-tech@mweb.co.za

Work Certificate

To	Mr A. Adeyemi
Address	Faculty of Health Sciences, School of Health Systems and Public Health, University of Pretoria
Date	20/12/2019
Subject	Thesis: **$PM_{2.5}$ chemical composition, source apportionment and geographical origin of air masses in Pretoria, South Africa**
Ref	AA/GS/02

I, Gill Smithies, certify that I have edited the following for language, grammar and style,

Thesis: Chapters 1 to 5 & 7 – **$PM_{2.5}$ chemical composition, source apportionment and geographical origin of air masses in Pretoria, South Africa,**

to the standard as required by the University of Pretoria.

Gill Smithies

Appendix 20: Certificate from the Language Editor II

Gill Smithies

Proofreading & Language Editing Services

59, Lewis Drive, Amanzimtoti, 4126, Kwazulu Natal

Cell: 071 352 5410 E-mail: moramist@vodamail.co.za

Work Certificate

To	A. Adeyemi
Address	School of Health Systems and Public Health, Faculty of Health Sciences, University of Pretoria
Date	27/09/2019
Subject	Article: Association between Air Pollution and Respiratory Disease Hospital Admissions in Pretoria, South Africa
Ref	AA/GS/01

I, Gill Smithies, certify that I have edited the following for language, grammar and style,

Article: Pages 1 – 13, Association between Air Pollution and Respiratory Disease Hospital Admissions in Pretoria, South Africa

to the standard as required by the University of Pretoria.

Gill Smithies

Appendix 21 Proof of Submission of article to International Journal of Environmental Health Research

Adewale Adeyemi <u17299340@tuks.co.za>

International Journal Of Environmental Health Research - Account Created in Manuscript Central

International Journal Of Environmental Health Research <onbehalfof@manuscriptcentral.com> 5 June 2019 at 19:39
Reply-To: ijehr.cem@btinternet.com
To: abifaz@yahoo.com

05-Jun-2019

Dear Mr. Adewale Adeyemi:

A manuscript titled Association between air pollution and respiratory disease hospital admissions in Pretoria, South Africa (CIJE-2019-0225) has been submitted by Mr. Adewale Adeyemi to International Journal Of Environmental Health Research.

You are listed as a co-author for this manuscript. The online peer-review system, Manuscript Central, has automatically created a user account for you.

The site URL and your USER ID for your account is as follows:

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USER ID: abifaz@yahoo.com
https://mc.manuscriptcentral.com/cije?URL_MASK=c0b5deeef414475faaa5191c62a67305
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Thank you for your participation.

Sincerely,
International Journal Of Environmental Health Research Editorial Office

Appendix 22 Proof of Submission of article to Journal of Pollution and Health

UNIVERSITEIT VAN PRETORIA
UNIVERSITY OF PRETORIA
YUNIBESITHI YA PRETORIA

Adewale Adeyemi <u17299340@tuks.co.za>

Submission Confirmation for Association between air pollution and respiratory disease hospital admissions in Pretoria, South Africa

1 message

JH&P <em@editorialmanager.com> 1 October 2019 at 14:47
Reply-To: JH&P <sandy@journalhealthpollution.org>
To: Adewale Adekunle Adeyemi <abifaz@yahoo.com>

Dear Mr Adeyemi,

Your submission entitled "Association between air pollution and respiratory disease hospital admissions in Pretoria, South Africa" has been received by journal Journal of Health and Pollution

You will be able to check on the progress of your paper by logging on to https://www.editorialmanager.com/jhealthpollution/.

Your manuscript will be given a reference number once an Editor has been assigned.

Thank you for submitting your work to this journal.

Kind regards,

Journal of Health and Pollution

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