University of Nairobi

College of Architecture and Engineering

Institute of Nuclear Science and Technology

Calibration of Low-Cost Optical Particle Counters and Application for Atmospheric Particulate Matter Measurement in Urban Nairobi and Rural Nanyuki, Kenya

by

David Ng'ang'a

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BSc. Chemistry

Signature _____ Date _____

A thesis submitted in partial fulfilment for the degree of Master of Science in Nuclear Science at the Institute of Nuclear Science and Technology in the University of Nairobi

Declaration

This thesis is my original work and has not been presented for a degree in any other university.

Signature ……. Date…….

David Ng'ang'a

S56/82840/2015

Supervisors' Approval

This thesis has been submitted with our knowledge as university supervisors

Prof. Michael J. Gatari Sign …….....……

Institute of Nuclear Science and Technology Date …….....

University of Nairobi

Dr. Francis D. Pope Sign…….....……

School of Geography, Earth and Environmental Sciences Date …….....…….

University of Birmingham

Dedication

I dedicate this work to my late grandmother Salome, my parents, siblings, schoolmates and all who made the completion of this research a success. Above all, I would like to thank the Almighty God for the gift of life, knowledge and will to pursue my studies amidst the challenges.

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List of Abbreviations

APM-Atmospheric particulate matter

OPC-Optical Particle Counter

WHO-World Health Organization

USEPA-United States Environmental Protection Agency

EEA-European Environmental Agency

EDXRF-Energy Dispersive X-ray Fluorescence

PMF-Positive Matrix Factorization

HYSPLIT-Hybrid Single Particle Langragian Integrated Trajectory Model

SSA-Sub-Saharan Africa

ASAP- A Systems Approach to Air Pollution in East Africa

Abstract

The study of atmospheric particulate matter (APM) is of great significance for both air quality and monitoring studies. Understanding the sources and concentration of these particulates depend on the presence of long-term data, which is largely missing in the Sub-Saharan Africa (SSA). This study aimed at calibrating and using low-cost Optical Particle Counters (OPC-N2) sensors, collocated with an Andersen dichotomous impactor in the provision of the much-needed temporal resolution data for APM monitoring in Nairobi. Three sensors ran by a raspberry pi low-cost microprocessor packaged in weather-proof boxes were deployed in three sites in Kenya; two in Nairobi (urban background and roadside site) and one in Nanyuki, upwind of Nairobi representing the rural background site. Comparison of APM mass concentration obtained by the sensors and gravimetric means aided in the calibration process before deploying them in the three sites for a period of two months (February-March 2017). From the comparisons, differences of concentrations were observed and correction of the OPC data was implemented using factors, 1.63 and 1.70 for $PM_{2.5}$ and PM_{10} species respectively, obtained from the collocated calibration sampling. The mean daily concentration observed for $PM_{2.5}$ using the OPC-N2 at the rural background, urban background and roadside sites were 13 ± 7 μgm^{-3}, 25 ± 14 μgm^{-3} and 37 ± 26 μgm^{-3} respectively while those of PM_{10} were 20 ± 9 μgm^{-3}, 53 ± 34 μgm^{-3} and 94 ± 45 μgm^{-3} at the same sites respectively. The concentrations of APM observed at the two urban sites regularly exceeded the WHO limits in both the $PM_{2.5}$ and PM_{10} size fractions. A "Lenschow" approach showed a clear urban and roadside increment in the city of Nairobi which was estimated to 7 μgm^{-3} and 18 μgm^{-3} for $PM_{2.5}$ respectively while the same increments for PM_{10} were 33 μgm^{-3} and 43 μgm^{-3} respectively. A clear diurnal pattern in APM mass concentration was noted in the peak traffic periods which was consistent with the roadside increment, spatial variation and source apportionment findings that implicated traffic emissions as a dominant source of air pollution in Nairobi. Analysis of gravimetric samples by Energy Dispersive X-ray Fluorescence Spectroscopy (EDXRF) obtained during the collocation measurements of $PM_{2.5}$ at the urban background site showed the presence of Fe, Ca, Zn, Mn and Cr. Their mean concentrations were 496±32 ngm^{-3}, 407 ± 106 ngm^{-3}, 42.7 ± 5 ngm^{-3}, 8 ± 1 ngm^{-3} and 2 ± 1 ngm^{-3} respectively. The presence of mineral dust elements (Fe and Ca) affirmed the contribution of mineral dust as a significant source of APM in Nairobi while Mn, Cr and Zn indicated the contribution of mixed sources such as biomass burning, industrial and traffic-related emissions. This was consistent with source apportionment findings obtained using the PMF model. Results obtained from this study show that the use of low-cost and calibrated OPC-N2 sensors collocated alongside standard gravimetric reference methods can provide cheap and reliable air quality measurements in Kenya urban centers.

Chapter 1: Introduction

1.1. Background

Atmospheric particulate matter (APM) in urban centers, particularly those in the developing world have been a subject of great concern. Much concern has focused on the increased level of these particles in air because of their role in affecting the health of the urban populace (**Amegah and Agyei-Mensah, 2017**). A strong correlation exists between high levels of APM and the exacerbation of morbidity and mortality rates (**Pope and Dockery, 2006; Evans et al., 2013**). In addition, these particulates contribute significantly to climate modulations in the world leading to more problems for the global community. The scourge of atmospheric pollution has claimed many lives with over 7 million deaths attributed to outdoor air pollution (**WHO, 2016**) and over 92% of these cases occur in the developing world (**Landrigan et al., 2018**). Despite strong evidence linking air pollution, public health and climate change, the topic is under-researched in the developing world (**Ngo et al., 2015**).

Lack of long-term air quality research in many parts of the developing world has resulted in poor understanding of the concentrations and sources of air pollutants in those countries. These countries, particularly those in SSA, still lag behind in conducting long-term monitoring studies on air pollution. This makes it difficult to understand the long-term effects of these pollutants to public health (**Gatari et al., 2009; Gaita et al., 2014**). Besides, people in these parts of the world are at risk of succumbing to respiratory and cardiovascular diseases due to lack of proper air quality guidelines to mitigate the effects of APM pollution. Therefore, there is a need to conduct more studies in the continent if mitigation strategies on air pollution are to be adopted (**Mkoma et al., 2014; Petkova et al., 2013**). This will help in dealing with the air pollution problem in a continent

that is experiencing high economic growth and continued use of biomass fuel as its main source of energy.

Various environmental bodies such as USEPA, EEA, and health regulatory bodies such WHO among other have come up with standards on air pollution. These standards are based on mass concentration measurements of $PM_{2.5}$ (particles with an aerodynamic diameter <2.5μm) and PM_{10} (particles with an aerodynamic diameter of <10 μm) (**Meister et al., 2012; WHO, 2016**). For African cities such as Nairobi, the cost of monitoring and measuring the concentration of these particulates is a barrier to efforts aimed at mitigating the impact of air pollution. This implies that even in nations where air quality standards exist, there is a poor network of monitoring systems to ensure compliance with the set standards. The high cost of instrumentation and required personnel to setup reliable networks dictates the adoption of cheap technologies that will aid in long-term air quality monitoring in these parts of the world (**Roy, 2016**). Low cost sensors based on light scattering principles such as OPC-N2 have the potential to reduce the cost of research on air quality measurements by several magnitudes (**Pope et al., 2018**). While theses sensors offer a cheap alternative, their results are only reliable if they are properly calibrated by accurate collocation with standard reference gravimetric instruments. This has the prospective of making air quality monitoring and measurement accessible and attainable in the developing countries like Kenya.

1.2. Problem Statement

The rapid economic growth of urban centers in Low and Middle Income Countries (LMICs) coupled with an increase in population growth, poor transport systems and urban planning has resulted in high concentrations of APM beyond the acceptable limits. In particular, the increase in vehicular transport in the developing nations is a major contributor to air pollution (**Colvile et al., 2001).** Nairobi, the Kenyan capital with a population of 4 Million which is projected to increase

to 7.14 Million by 2030, is showing similar trends in vehicular transport increase and air pollution (**Raje et al., 2018**). This has resulted in the creation of pollution hotspots in the city, where the urban population is exposed to APM levels beyond the limits set by the WHO and other regulatory bodies.

Atmospheric particulate matter (APM) contain harmful amounts of potentially toxic anthropogenic constituents associated with health risks, such as respiratory diseases, lung cancer and cardiovascular diseases among others (**Katanoda et al., 2011; Lin et al., 2015**). Therefore, there is a need to draft regulations and policies that will mitigate the effects of air pollution in public health. These regulations and policies need to be backed by quality, accurate and long-term measurements of these particulates. At present, there exist little information on the levels of air pollutants in urban centers of African cities such as Nairobi, and in instances where this information exists, it is mostly from short-term studies or satellite information, which is unreliable unless backed by sound ground measurements (**Gaita et al., 2014; Kinney et al., 2011**). High cost of instrumentation impedes studies on air pollution in the Kenyan capital thus leading to lack of long-term and accurate real-time data on APM pollution. As a result, understanding the effects of APM pollution in public health and climate change is difficult since the sources and concentration of these particulates are not understood in the developing world.

1.3. Objectives

1.3.1. Main Objective

- The main objective of this research is to demonstrate that calibrated low-cost OPCs can be applied in atmospheric particulate matter measurements and in turn help in creating affordable and reliable networks in air quality monitoring in urban and rural sites like Nairobi and Nanyuki respectively.

1.3.2. Specific Objectives

The specific objectives of the study are as follows:

- To calibrate and apply the OPCs by comparing APM measurements obtained using optical methods and those obtained through gravimetric means
- To measure and compare the mass concentration of $PM_{2.5}$ and PM_{10} in three selected sites in Kenya
- To compare the temporal and spatial variations in APM mass concentrations at the three selected sites
- To use the "Lenschow approach type" in estimation of the urban and rural increment of APM mass measurements
- To determine the concentration of elements in APM samples using EDXRF and conduct source apportionment of $PM_{2.5}$ using PMF

1.4. Justification and Significance

Emission levels from anthropogenic activities in urban centers of LMICs, such as Kenya have deteriorated the quality of air in these areas. The emissions have had a significant impact on the quality of health of the urban populace, with more deaths and health effects attributed to APM exposures in these areas (WHO, 2016). Despite existing studies showing a high correlation between exposure to particulate matter and human health effects, there exist a poor understanding and knowledge on the pollutant levels and their sources within LMICs compared to other parts of the world. **Gatari et al., (2009**) observed a dearth in air pollution and monitoring data in the continent. In instances where these data exist, it is mostly from short-term studies. With few long-term studies on air quality and monitoring systems existing in urban centers of the developing world, this study aims at providing adequate and quality temporal resolution data needed to

mitigate the impacts of APM pollution in Nairobi, Kenya. Successful application of calibrated low-cost OPC-N2 sensors in Nairobi and Nanyuki demonstrated that these types of sensors can help in the creation of cheap and reliable air pollution monitoring networks in the urban centers of the developing countries. In addition, data obtained will help in the improvement of our understanding of APM concentrations, types and their probable sources in the region. As a result, policy makers and health practitioners will find it easier to assess, monitor and predict the impact of air pollution on public health using locally derived data from the low-cost sensors.

Chapter 2: Literature Review

2.0. Introduction

The recent rise in cases of cancer, respiratory, and cardiovascular diseases among other adverse health effects on the urban population in SSA has raised much concern over the quality of air in the cities. **WHO (2016**), revealed that over 90% of people in the world breath low quality air. Majority of exposed people to APM are those in the urban centers, particularly those in the South-Eastern parts of Asia and the SSA (**Lelieveld et al., 2015**). Most epidemiological and toxicological research show a strong correlation between low quality of air in urban centers and the exacerbation of acute respiratory diseases among the populace (**Pope and Dockery, 2006**; **Kelly and Fussel, 2011**). **Brugha and Grigg.** (2014) revealed a strong link between air pollution in urban centers of the developing world and the use of morbidity and mortality reported in those countries.

Developed nations have made great strides in dealing and mitigating the impact of air pollution on public health. However, SSA nations still lag behind in overcoming the challenge of air pollution. As a result, the associated health risks of APM pollution to public health in these countries are even higher. 92% of all global pollution-related deaths are reported from these nations (**Bachmann, 2015). Mabogunje, (1995)** reported that most cities in SSA face severe environmental challenges from: atmospheric pollution from burning of waste and emission from transportation; poor solid waste management and the inability to provide safe water and sanitation services. While these challenges remain high for SSA cities, outdoor and indoor air pollution remains the biggest environmental threat facing the urban populace in these cities (WHO, 2016). Their citizens are exposed to high concentrations of APM exceeding the limits set by WHO. This poses great danger to their health status. Efforts to mitigate these effects require long-term air quality measurements in those regions to help in the assessment and measurements of the impact

of air pollution on public health. However, there are few air monitoring networks in the continent and this results in the shortage of data. As part of addressing this need, electronic miniaturization of air quality monitors commonly known as Low-Cost Sensors (LCSs) have been proposed. **Lewis (2018)** endorses the use of these devices in air pollution studies in areas with poor air quality networks because of their low cost, small size and low energy consumption compared to the established analytical reference methods that rely on gravimetry.

The African continent contributes littles towards air pollution studies and with the popularity gained in the use of LCSs in air quality monitoring, this chapter looks at the principle of operation of these sensors, the challenges of using them as far as uncertainties and errors are concerned, the ways to minimize errors and the literature review on the successful use of these sensors in real-world APM monitoring and that of previous studies conducted on measurement of particulate matter in Nairobi.

2.1. Principle of Light Scattering and Low-Cost Optical Particle Counters

Three different techniques are used in the determination of the mass concentration of particulate matter in a given place. They include techniques such as gravimetry, B-attenuation and optical methods. The gravimetric method relies on the measurement of mass loadings of PM in air trapped in a pre-weighed filter membrane and the concentration in a given volume of air identified. Unlike the gravimetric method which relies on the difference in mass of filter before and after sampling, the B-attenuation method exploits the Beer-Lambert's law to determine the mass concentration of PM. The difference between the baseline beta count before and after a filter has been exposed to particles is proportional to the mass of PM sampled in a given volume of air. It is effective in the measurement of particles with a size less than 0.3 micrometer as smaller particles do not scatter light effectively **(Costello et al., 2007).** The third method, optical method, is based on the principle of light scattering. According to this principle, when a beam of light is shone through particles, scattering occurs. The intensity of light scattered by the particle is a function of the incident light

intensity, wavelength of the incident light, scattering angle, shape, size and refractive index of that particle.

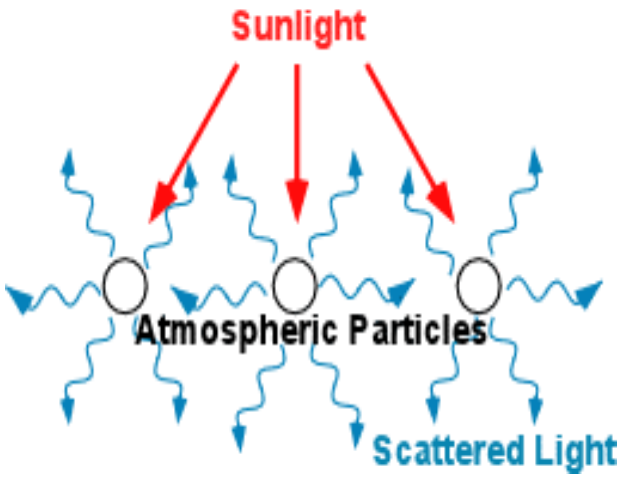

Figure 2. 1: The light scattering phenomenon by atmospheric particles adopted from WW2010 (2010)

Two theories are used in explaining the concept of light scattering and its application in PM measurement. The theories; Rayleigh and Mie are used in the quantification of the amount of light scattered from the incident light interacting with particulate matter. This allows one to quantify and approximate the size of the particles interacting with light. Unlike Mie's theory, Rayleigh's theory describes the elastic scattering at molecular level where particles whose diameter is less than about a tenth of the size of wavelength of incident light. This causes re-radiation of the scattered light in all direction thus ensuring that the intensity of scattered light is not a function of the angle of incident light (**Xu, 2015**). This makes it not an effective theory in the quantification of PM. The latter theory describes the elastic scattering of light by large particles whose diameter is larger than the wavelength of the incident light. This makes the scattering a strong function of the angle of incident beam. Besides, the scattered light is a function of the wavelength of incident light, the size and refractive index of particles as well as the angle made with respect to the incident light beam. Non-uniform distribution of the electromagnetic field by the particles scattering the light makes it possible to use the Mie's theory in the quantification of the size and distribution of particles such as $PM_{2.5}$ and PM_{10}.

Equipment based on the light scattering phenomenon have a light source (laser beam), which irradiates particles entering the optical counter through the inlet. The particles scatter the light in different directions and angles. A relation between the angle of scattering, incident light, wavelength, particle size and refractive index of the particle is estimated based on the predictions of the Mie's theory. This is because small particles at low intensities scatter light through large angles while the large particles scatter light at low angles. Therefore, the angle of scatter is inversely proportional to the spherical particle size of an object scattering the light. This implies that the diameter of airborne particulate matter can be estimated by detecting the amount of light scattered when light photons interact with particles entering the counters. However, airborne particulate matter are not spherical and hence large uncertainties exist in the measurement of PM across the different size range. The measurement involves a conversion of the amount of scattered light into an electrical signal whose peak amplitude is proportional to the amount of scattered light. The information is then fed into an algorithm that uses the scattered light intensity into a size distribution pattern and ultimately a number concentration based on the Mie's theory. Figure 2.2 shows the schematic flow of how light scattering equipment is used in the particle size distribution and concentration measurement.

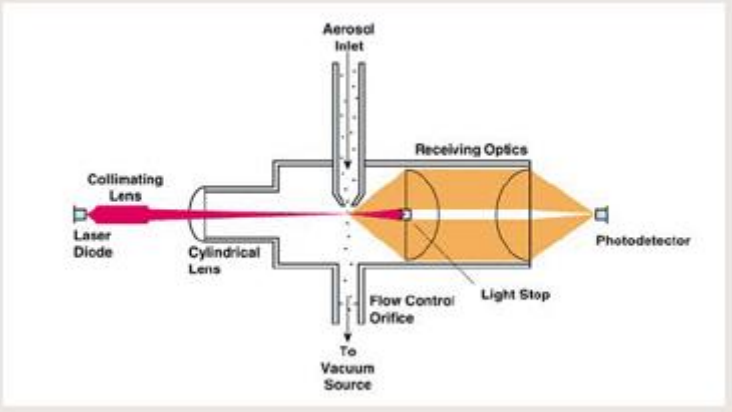

Figure 2. 2: The schematic flow of particles in an optical counter. Adopted from Dustmonitors (2009)

2.3. Uncertainties in Using Optical Counters in Aerosol Studies

The use of low-cost OPCs in areas with limited network of air monitoring instruments provide a number of advantages ranging from the low cost, small size and low weight as well as the reduced power consumption. However, despite the fast and easier way of accessing air quality information

from these devices, there are uncertainty issues that come with the information on the particle size and number concentration obtained using these sensors.

Steinle et al., (2013) showed that the quality of data in both field and lab obtained from the LCSs is highly variable even when counters from the same designers are used. This implies that there are a number of uncertainties involved in using these devices for air quality monitoring studies. Some of these uncertainties are based on the number of assumptions made when aerosol particles enter the optical devices. One of these assumptions is that the OPC measures the size of the particle. This is not possible as the counter measures the optical scattering cross-section, which is later translated into a particle size. However, for one to estimate the size of the particle, one must make other assumptions such as the shape and refractive index of the particles entering the counter. It is assumed that the particles are spherical in nature, and have equal refractive index and density. These assumptions create high level of uncertainties and questions on the accuracy and reliability of these devices are often raised as aerosol particles are complex in nature and vary in shape, density and refractive index (**Manikonda et al., 2016**).

Other factors affecting the accuracy and reliability of these sensors is the effect of atmospheric factors such as relative humidity, which results to high level of uncertainty in the size of a dry particle when these devices are used. **Hojaiji et al. (2017)** showed that the particle mass concentration of pollutants measured using instruments based on light scattering increases with increased humidity due to coagulation. In addition, hygroscopic growth occurs when the relative humidity exceeds the deliquescence point of a substance (**Petters et al., 2008)**. This leads to the formation of fog droplets or mists which are detected as particles leading to high uncertainty in measurements conducted using these sensors. Therefore, the high levels of uncertainties in the use of low-cost optical counters imply that there is need to conduct proper calibration through

comparisons of readings obtained from these devices with those from standard gravimetric reference instruments. The comparison of the readings lowers the uncertainties in the LCSs by a given factor hence making the results obtained reliable and useful in air pollution studies.

2.4 Review on the Use of Low-Cost Sensors in Air Pollution Studies

Questions concerning the reliability and accuracy of low-cost optical counters have prompted the conduction of numerous studies across the globe. These studies aim at checking on the accuracy of these sensors against results obtained from collocated standard reference instruments in air pollution studies. **Zikova et al., (2017)** collocated 66 new low-cost sensors with a Grimm particle spectrometer to ascertain the reliability of these sensors to measure the APM mass concentrations under real working conditions. From the study, the level of bias and precision of these sensors was determined. The correlation between concentrations obtained from the sensors and the GRIMM standard instruments was low. In addition, there was high variability in the results obtained from the different sensors despite being from the same manufacture with levels of bias reaching as high as 100% in some units. Therefore, the reliability and stability of these sensors in air quality monitoring studies is subject to assessment before the instrument can be used in real-case studies. There is need to conduct proper calibration to lower the level of uncertainty in each of the instrument before any study can be conducted using low-cost sensors.

A similar study conducted by **Liu et al., (2017)** showed that varying some factors affect the calibration performance of low-cost sensors. In this study, four low-cost optical counters were tested in the lab and the size distribution and composition varied. It was observed the calibration data had a good linear relationship, under steady conditions. However, varying the size and composition resulted in a different calibration dataset. This implied that there was a need to conduct calibration at steady particle mass concentration such as in an urban background or

laboratory set up instead of transient concentrations. This helps in the reduction of the level of uncertainties that come with the correction factors obtained during calibration.

The accuracy of low-cost optical particle counters depends on proper calibration using standard reference equipment. **Steinle et al., (2015)** conducted personal exposure monitoring of $PM_{2.5}$ using light-scattering based instruments, the Dylos 1700 at an urban background site. The instruments were calibrated using results obtained from collocated TEOM-FDMS at the two monitoring sites; an urban background and a rural background. The use of two sites was to help in the validation of the results obtained by the Dylos 1700 in the field. A good correlation between the instruments after calibration was obtained with the urban background site recording an R^2=07 and rural background site recording an R^2=09. This implied that with proper calibration, results obtained from low-cost sensors can be relied upon in real-time studies.

In addition to the inter-instrument comparison studies conducted on the reliability of low-cost sensors, it is prudent to calibrate these devices for relative humidity dependence. This is because of the uncertainty created by high level of humidity. **Crilley et al., (2018)** observed that the accuracy of measurements made by the OPCs is affected by the RH levels exceeding 85%. The RH dependence on measured particle mass is due to the hygroscopic nature of aerosol particles which make them take up water. This results in discrepancies between measurements made by the low-cost sensors and other reference materials. Therefore, for studies conducted in areas with high RH levels, there is need to check for RH dependence before using the equipment.

deSouza et al., (2017) examined the strength and technical limitation of using LCSs in air quality studies. It was found out that these sensors have the potential to address the challenges of poor air quality and inadequacy of data in SSA cities. However, the strength of these sensors hinges on the

ability to conduct proper calibration using collocated gravimetric instruments. This will help in the reduction of some of the uncertainties that come with assumptions made in the design of the devices. It is for this reason that this study aims at calibrating the low-cost Alpha-Sense OPC-N2 devices before deploying them in particulate matter studies in Nairobi. The calibration will help in reducing the uncertainties in the measurements obtained and hence provide a reliable temporal resolution data for APM measurement.

2.5. Review of Selected Studies on Air Pollution in Nairobi

Gaita et al., (2014) reported concentration levels of $PM_{2.5}$ in the range of 3 µg $m^{-3}$and 53 µg $m^{-3}$and an overall mean of 21 µg m^{-3}. The measurements were conducted at an urban background site in Nairobi between April 2009 and March 2010. These concentrations exceeded the yearly limits set by the WHO of 10 µg m^{-3} by a factor of two.

Heavy local traffic and dense population in Nairobi plays a significant role in contributing to APM pollution. **Kinney et al., (2011)** reported $PM_{2.5}$ mass concentration levels monitored over an 11-hour timeframe ranging between 58 µg m^{-3} and 98 µg m^{-3}. In addition, a decrease from 129 µgm^{-3} to 19 µgm^{-3} in the horizontal dispersion of fine particulate matter was observed at the downwind of one of the major junctions in Nairobi. A further decrease from 120 µgm^{-3} to 43 µgm^{-3} in the fine particulate matter was observed at the vertical dispersion from a roadside site to a rooftop within the CBD of Nairobi. Further studies by Ngo et al., (2015) highlighted the significant role played by anthropogenic activities in influencing the air quality in Nairobi. According to the study, motorists playing their trade in the city were exposed to concentrations levels as high as 103 µgm^{-3}. In addition, those in informal sectors like Mathare were exposed to concentrations of about 63 µgm^{-3}.

The severity of APM pollution in urban centers of the developing world is even higher in the informal settlements. These areas have the highest diagnoses of respiratory problems such as asthma and bronchitis. **Egondi et al., (2016**) reported high concentration levels of fine particulate matter in Viwandani and Korogocho slums of Nairobi. Viwandani slums, located in the Northern part of Nairobi had a mean mass concentration of PM of 67 μgm^{-3} with bimodal peaks observed in the morning and evening at 76 μgm^{-3} and 82 μgm^{-3} respectively. Korogocho slums, at the western side of Dandora, had a mean concentration of 166 μgm^{-3} and bimodal daily peaks of 214 μgm^{-3} and 164 μgm^{-3} for both morning and evening respectively.

Chapter 3: Methodology

3.0. Introduction

This chapter describes the steps that were used in the process of data collection for the study. It also offers a systematic procedure and description of the process used in data collection, instrumentation and analysis of the data collection.

3.1. Study Area

The study focused on three sites in Kenya as shown in Figures 3.1 and Figures 3.2 below. Two of the selected sites were in Nairobi, Kenya's capital city. The city covers an area of 696 Km^2 and had a population of 3.5 million people as at 2016 (**WPR, 2017**). In addition, Nairobi acts as the communication, diplomatic and financial capital of the East African bloc because of its popularity among tourists and diplomats (**Rajé et al., 2017**). Geographically, Nairobi is located at approximately latitude of 1.3^{o}S and longitude of 36.0^{o}E. Its highest elevation point is at an altitude of 1700m above sea level. The rapid increase in the city's population and motorization coupled with the absence of proper regulation and mitigation policies against air pollution are likely to increase the levels of APM pollution in Nairobi. There were two field sites within Nairobi; one representing the urban background and the other the roadside site. The third field site was in Nanyuki town, located north-north East of Nairobi at an approximate aerial distance of 147 km and 240 km by road, and served to represent the rural background site.

3.1.1. Nairobi urban background site

The urban background site in Nairobi was located at American Wing, University of Nairobi (UoN). This site stood at an elevation of 17 meters above the ground level. The site is free from obstruction hence free flow of air thus making it a good urban background site. It is closer to Harry Thuku road, which has few on-road vehicles with little or no heavy commercial vehicles. According to **Kinney et al., (2011),** the American wing site is an ideal background site because of the little influence from local sources. Its height level ensures that dispersed pollutants in the regional air mass are collected at the site.

3.1.2. Nairobi urban roadside

The urban roadside site located at the fire station situated in Tom Mboya street of Nairobi is characterized by high traffic density. This includes the PSVs popularly referred to as "matatus." In addition, this site is located next to an urban street canyon with high vehicular emissions from the idling vehicles. This exposes the site to the urban heat island effects. It also neighbors the site where vertical dispersion measurements of $PM_{2.5}$ were conducted by **Kinney et al., (2011).**

3.1.3. Nanyuki rural background site

One of the requirements of the Lenschow approach in air pollution studies in an urban center is the need to have a rural background site for comparative purposes (**Lenschow, 2001).** For this study, Nanyuki town, situated at the equator and geographically located at a latitude of 1.3° S and 36.0° E. According to the last conducted census, Nanyuki town is home to over 50,000 residents (LC, 2015). There are significant agricultural activities taking place in the area thus making it an ideal rural background site. This is also enhanced by the strategic positioning of the site at the slopes of Mt. Kenya, which allows the free flow of regional air mass within the site thus minimizing the influence of local sources at the site where the OPCs were placed.

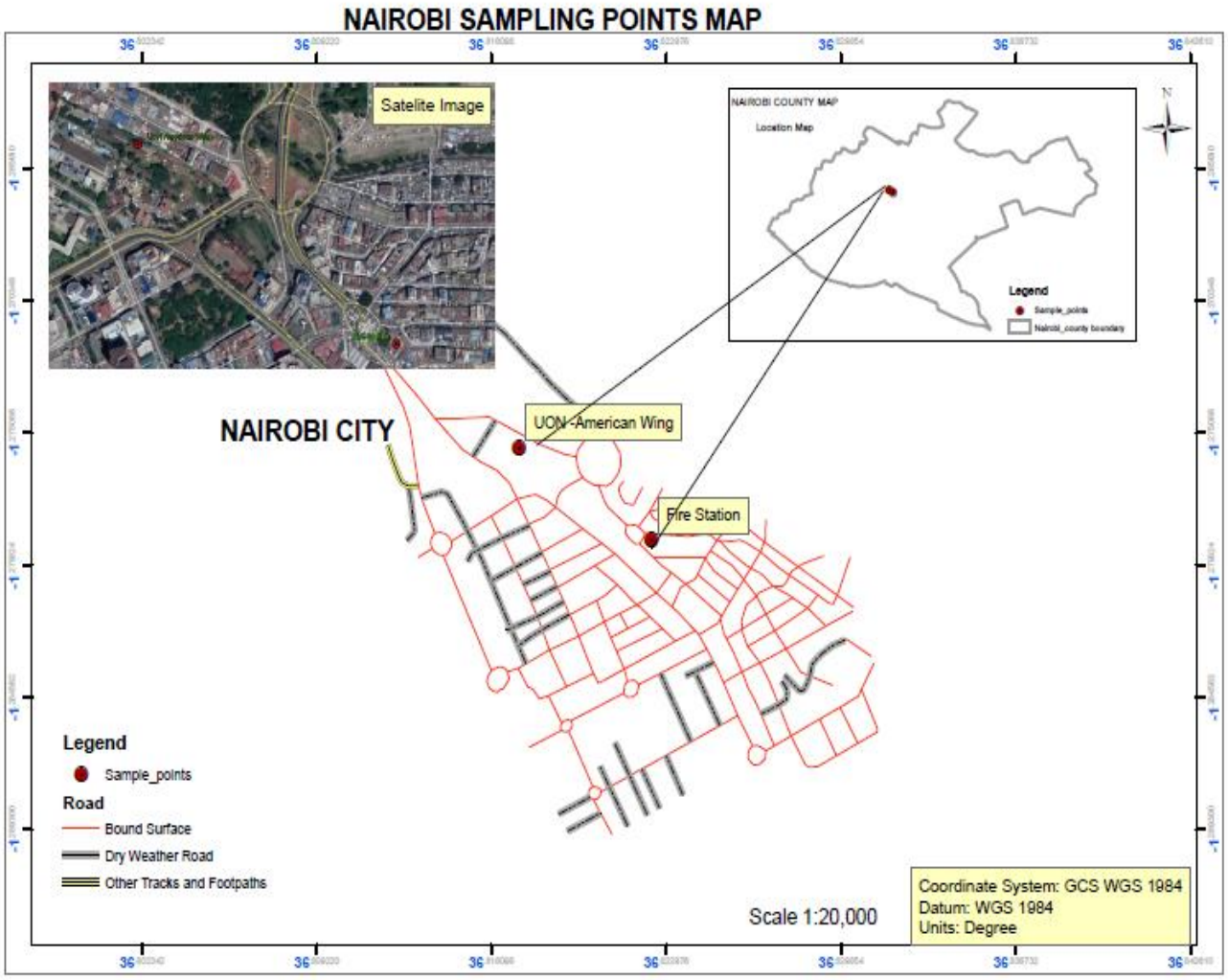

Figure 3. 1: Map showing the sampling sites in Nairobi: UoN-American Wing Site representing the urban background site, Fire Station representing the urban roadside site in Nairobi

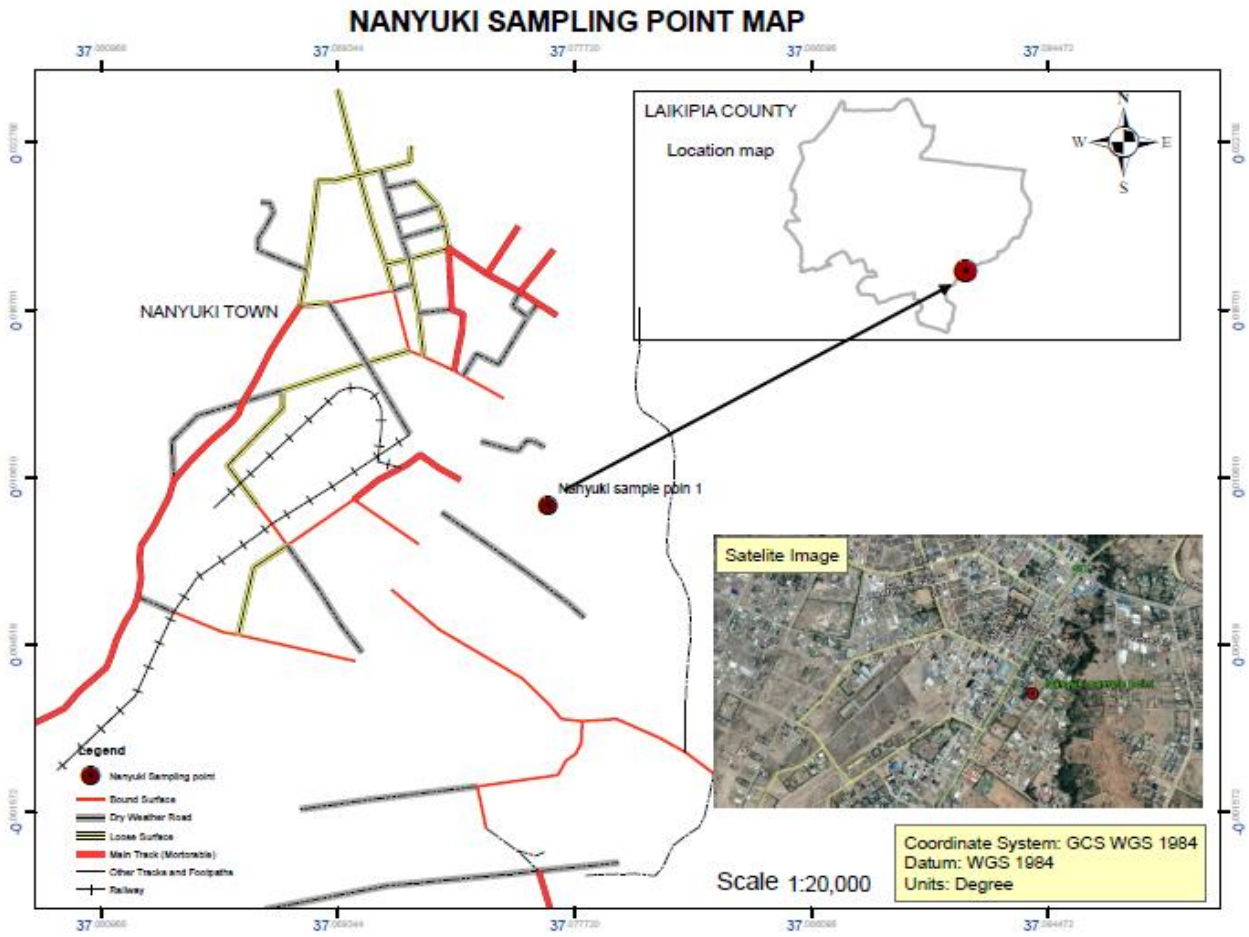

Figure 3. 2: Map showing the rural background site in Nanyuki

3.2. Study Design

The study was mainly field work and lab-based, with the field work involving the collection of data from the three selected sites using the low-cost OPC-N2 sensors. An Andersen dichotomous impactor was used at the urban background site to collect particulate matter on filters for the calibration purpose. Additionally, the samples collected using the filters were subjected to a lab-analysis process where they were analyzed for trace metals and subsequent source apportionment conducted to predict the probable sources of the pollutants.

3.3. Particulate Matter Sampling

Sampling of atmospheric particulate matter for this study was done using low-cost optical methods and gravimetrically using an Andersen dichotomous impactor sampler. For optical and real-time sampling, miniaturized low-cost optical particle counters (Alphasense OPC-N2, firmware 18) were used in the measurement of APM mass concentration at the three sites. The use of gravimetric reference equipment in APM measurements was to help in the calibration of the optical instruments. In addition, the equipment was used to allow for the collection of samples loaded in filters at the American wing site, which were to be analyzed for trace metal composition and source apportionment purposes.

3.3.1. Alphasense OPC-N2

In the measurement of particulate matter concentration, small low-cost optical particle counters (Alphasense OPC-N2, version 18) were used. The OPC-N2 are miniaturized sensors weighing less than 105 g and with dimensions of 75 mm by 60 mm by 65 mm. In addition, at a price of approximately Ksh. 25,000, these sensors cost 30 to 50 times less than the conventional reference OPCs thus making them a cheaper alternative. Gravimetric reference grade instruments are even more expensive. Therefore, the Alphasense OPC-N2 provided an opportunity for simultaneous measurements of PM at the three sites. These sensors measure PM in the size ranges of between 0.38 µm and 17 µm distributed across 16 different size bins (**Hagan et al., 2018).** The maximum particle count of the OPC-N2 was 10,000 particles per second. Assuming an average and uniform density of 1.65 gcm^{-3}, the number concentration is then converted to PM mass concentration by an on-board factory algorithm based on the EN481 European Standard (OPC-N2 Alphasense manual).

3.3.2. Anderson dichotomous impactor

For gravimetric analysis, the Andersen dichotomous impactor (Sierra Instruments Inc., USA) was used. It has a flow-rate of 16.7 L/Min and a sampler that segregates a stream of air that passes through the 10 μm into two portions that are filtered separately. The sampler cuts the 0 to 10 μm total sampler into 0 to 2.5 μm and 2.5 to 10 μm fractions that are collected on separate 37 mm Teflon filters.

Figure 3. 3: Andersen dichotomous air sampler deployed on the roof of the American Wing Site at the UoN.

3.4. Meteorological Station

For this study, the local weather conditions of Nairobi were monitored at a local meteorological station at the American Wing site. This was conducted using the Vaisala instrument (WXT510)

shown in Figure 3.4. The following variables were part of the measurements; Wind direction, wind speed, local temperature, barometric pressure, relative humidity and rainfall level. Measurements were taken at a resolution of five-minute interval. The nearness of the local meteorological station to the fire station site made the measurements representative and applicable to the sites in Nairobi. Continuous monitoring of the weather parameters was conducted from 2nd February 2017 to 6th April 2017 and this covered the entire APM measurement period.

Figure 3. 4: Vaissala weather instrument deployed at the American Wing Site.

3.5. Alphasense OPC-N2 Calibration

In-situ calibrations of the OPC-N2 are necessary before they are deployed for PM mass concentration at any given site **(Crilley et al., 2018).** This requires the collocation of the OPCs and a gravimetric reference instrument. For this study, an Andersen dichotomous impactor at the American Wing site was used for calibration purposes. The three OPCs were all collocated at the site on 9th February 2017 and the average mass concentrations of $PM_{2.5}$ and PM_{10} obtained over a

24-h period used for the calibration. For calibration purpose, PM_{10} sampled is the sum of $PM_{2.5}$ and $PM_{2.5-10}$ and the date chosen was one where no rain was recorded. It also had a similar ambient temperature and RH profile compared to most of the days in the campaign period. The following procedure was used in the calibration of the OPC-N2 before deploying them to other sites:

- Three OPC-N2 devices were collocated with an Andersen Dichotomous impactor loaded with pre-weighed Teflon filters (37-mm diameter) as from 1700Hrs on 9th February and all instruments were left to sample for a period of 24 h.
- After a period of 24-h, the average mass concentration of both the fine and coarse particulate mass fraction obtained from the OPC-N2 was calculated and its uncertainty taken as the standard deviation.
- For the Andersen dichotomous sampler, mass concentration was obtained gravimetrically by finding the difference in the masses of the loaded and empty filters and dividing the difference by the volume of air sampled during that period at the flow rate of 1 m^3 h^{-1} for 24 h.
- Uncertainties in the mass concentrations of both the fine and coarse PM mass fraction were estimated from instrument error of 10%, weighing 25% and sampling error of 7%.
- Having obtained the average mass concentrations using both the gravimetric and optical methods, comparisons were made to obtain the scaling or correction factor for the OPC-N2 data before deployment. Equation 1. below was used:

  Correction factor (scaling factor) = $\frac{Average\ Gravimetric\ mass\ concentration}{Average\ Optical\ mass\ concentration}$…Equation 1.

3.5.1. Atmospheric Particulate Matter Measurement

After obtaining the correction factor for the two-sized fractions obtained using the OPC-N2 sensors, the OPCs were deployed in the three sites; American Wing, Fire Station and the site in Nanyuki. They were deployed as from 2nd February 2017 to 24th March 2017 and $PM_{2.5}$ and PM_{10} were sampled during that period. At times, the OPC-N2 were collocated at the American wing site to help in conducting the inter-instrument precision assessment. This was done only once for the American Wing site between 16th February and 18th February 2017. A different collocation at the Fire Station site was conducted towards the end of the campaign period; 4th March to 27th March 2017. All this was done to check whether the monitors were comparable over the sampling period. It was also conducted to check the sampling variation of the equipment over the local RH conditions. The image shown below shows one of the OPC-N2 at the site:

Figure 3. 5:Shows one of the OPC-N2 deployed at the urban roadside site in Nairobi.

For the purpose of elemental analysis and subsequent source apportionment of the particulate matter, an Andersen dichotomous impactor collocated alongside the OPC at the urban background site at the American Wing building was used to collect PM samples on Teflon filters. The sampling of $PM_{10-2.5}$ and $PM_{2.5}$ was conducted between February and March from 9th February 2017 for 24-h each day. The samples were collected from 1700 Hrs to 1700 Hrs the following day. The choice of the timing was based on the fact that 1700 Hrs is the time when most people are leaving from their workplace and other daily activities. Therefore, most of the vehicles are on the road. This was to ensure that much of the emissions are factored in the collection. The 24-h sampling period using the dichotomous sampler helped in accounting for both the morning and evening traffic session in Nairobi. Extending the sampling period using gravimetric method as compared to the sampling of particulate matter using the OPC-N2 was to ensure that there were enough samples for conducting elemental analysis and the subsequent source apportionment.

3.6. Data Analysis

Particulate mass concentrations ($PM_{2.5}$ and PM_{10}) obtained using the Alphasense OPC-N2 were recorded in their respective size bins. This data was logged in the bins after every 10 seconds for the whole campaign period. For the analysis, the PM mass concentrations obtained were aggregated into one-hour time bins and the mean averages multiplied by the correction factors obtained during the calibration. Statistical visualization and analysis of the data obtained via OPC-N2 was done using the openair package R-programming language (version 3.4.2) as recommended by **Carslaw and Ropkins (2012**).

After sampling for the coarse ($PM_{2.5-10}$) and fine particulate ($PM_{2.5}$) sized-fractions using the Andersen dichotomous impactor, each loaded filter was kept in its own petri-dish to avoid contamination and weighed in the laboratory using a digital mass balance at the Institute of Nuclear

Science and Technology with a lower limit of detection at 0.1ug at a sensitivity drift of ± 1 ppm/^{o}C. The first phase of analysis of this data began by determining the mass concentration of both the coarse and fine size fractions using gravimetric methods. This was done by determining the mass difference between the loaded and empty filters. A total of 40 PM loaded filters were analyzed for mass concentration and elemental composition.

3.7. Elemental Analysis

For the elemental analysis of Fe, Ca, Zn, Mn and Cr, Energy Dispersive X-ray Fluorescence (EDXRF) spectroscopy was used. The choice of the method is based on the fact that it is a non-destructive analytical method. It is also fast and easy to operate. The EDXRF spectrometer used in the study is located at the Institute of Nuclear Science and Technology, University of Nairobi. It has a Tungsten Anode X-ray tube and a silver secondary target. The general X-ray fluorescence principles apply, where a beam of electron emitted from the Tungsten anode is accelerated towards the secondary target. The beam excites the secondary target, which in turn emit characteristic X-rays that excite the elements in the sample analyzed. For characterization, a Si (Li) detector is used in the detection of the characteristic spectral lines, an indication of the elemental composition of the sample. Each of the filter samples was run for a period of 1000 seconds at a current and voltage of 80 µA and 30 KV respectively. Quantification of the elements identified in the qualitative analysis process was done using the AXIL software donated by the International Atomic Energy Agency (IAEA). The software works by using the iterative least square method in the quantification of all elements identified using the k_{α} and L_{α} lines in the spectra. In this case, the elemental concentrations from the samples were obtained in units of $\mu gm^{-2.}$ This was later converted to units of mass by multiplying by the area of filter exposed to the sample. The final

value was then converted to a concentration unit by factoring in the volume of air that was sampled during the collection period using the Teflon filters.

Validation of the qualitative and quantitative analysis was conducted by analyzing thin standard reference sample filters from the IAEA. The standards were analyzed in a similar manner as the samples collected using the dichotomous impactor. Standard spectra from certified reference materials were used in the determination of the detection limit (DL) of the analytical method using the International Union of Physical and Applied Chemistry (IUPAC) equation (van Grieken and Markowicz 2002).

$$\mathbf{DL = 3C\ \sqrt{N_b}/\ N_p} \quad \textbf{.....Equation 2}$$

In the equation above, C is the certified concentration based on the standard reference material used in the validation while N_b and N_P represent the background area and element peak areas respectively.

An advanced positive matrix factorization (PMF version 5.0) software by the US Environmental Protection Agency (USEPA) was used in apportioning the elements and the APM sampled at the American Wing site. The software, a multivariate statistical model works by decomposing a matrix of speciated sample data into two matrices; factor contributions and factor profile. This helps in the prediction of the probable sources of trace elements in the APM at the site. The software was chosen over others such as Principal Component Analysis (PCA) because of its robustness and guarantee for non-negative results **(USEPA, 2014**).

Chapter 4: Results and Discussions

4.0 Overview

In this chapter, results for the site meteorology and those used in the calibration of the OPC-N2 before being deployed in the three sites will be presented. In addition, results on the analysis of the mass concentration of atmospheric particulate matter (APM) obtained from the three sites using OPC-N2 will form part of this chapter. This will entail a discussion on the mean mass concentration, daily WHO exceedance of the APM mass concentration at the three sites, diurnal variation, and the urban and rural increment of APM mass concentration. The chapter will end with a discussion of the results obtained from the elemental analysis of the samples collected using the Andersen dichotomous sampler collocated with the OPC-N2 sensor at the urban background site and subsequent source apportionment findings from the analysis of the speciated particulate matter data using PMF.

4.1. Site Meteorology

For this study, meteorological variables were measured before and during the study period at the urban background site in Nairobi. Table 4.1 below depicts the statistical summary of the different weather parameters measured at the American Wing Site during the study:

From the summarized statistics in table 4.1, the mean average speed of the wind was 1.9ms^{-1}. The winds were North-Easterly as shown in the wind rose in Figure 6. This direction conforms well to back trajectory analysis of air mass reaching Nairobi during the campaign period using the HYSPLIT model from NOAA (**Stein et al., 2015; Appendix 1**). The rural background site in Nanyuki is in the north of Nairobi and the weather parameters were used in assessing the rural

APM mass loadings reaching the sites in Nairobi. The measurements of these weather parameter coincided with the hot and dry season in Kenya that begins from mid-January to mid-March. Despite being a hot and dry season with the air temperature ranging between 15^{o}C and 31^{o}C and a mean of 22^{o}C, there were episodes of precipitation during the campaign period. This was noted on 17th, 19th and 24th day of February 2017. Other rain events were noted during the 17th and 22nd day of March 2017. The relative humidity at the urban background site ranged between 15% and 90% with a mean average of 51%, which was lower than the 85% pointed by **Crilley et al., (2017)** on the use of OPC-N2 in PM measurements. The 85% mark was passed on less than 10% of all the nights hence results obtained in this study were not affected by the RH.

Table 4. 1: The meteorological results observed at the weather station in the Nairobi urban background site during the sampling period

	Wind Speed (ms^{-1})	**Relative Humidity (RH) (%)**	**Temperature (°C)**	**Pressure (mbar at 1680 m a.s.l)**
Minimum	0.1	15.0	15.2	827.3
1st Quartile	1.0	37.0	18.9	831.4
Median	1.6	51.0	21.5	832.4
Mean	1.9	51.4	22.1	832.4
3rd Quartile	2.5	66.0	25.2	832.4
Maximum	10.5	90.0	30.7	836.4

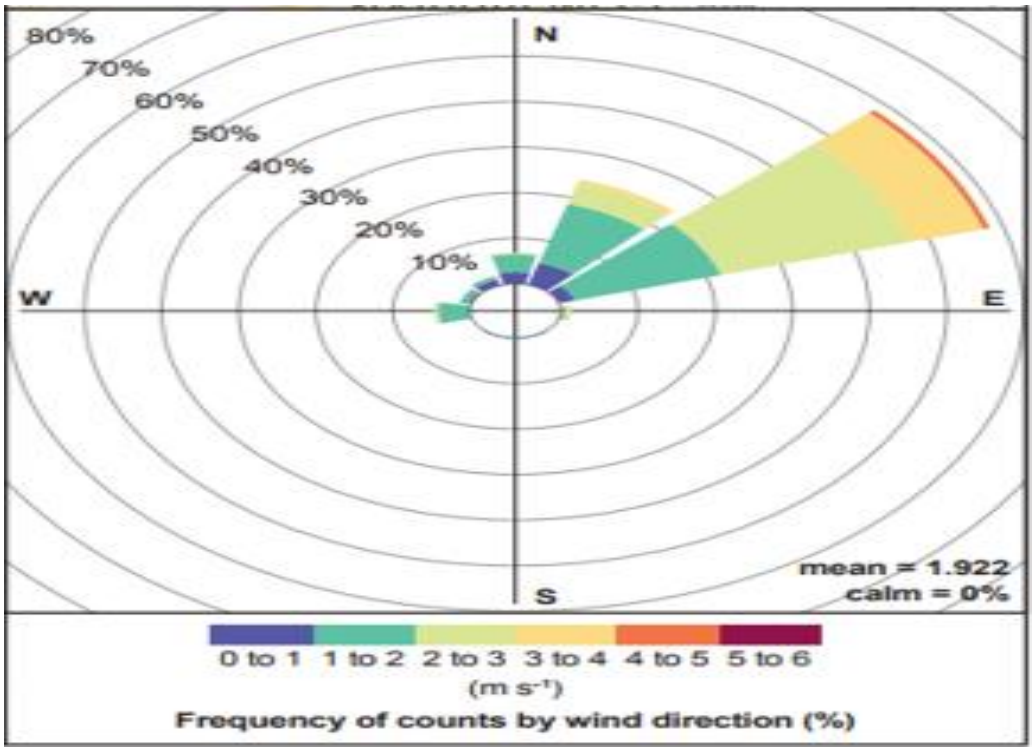

Figure 4. 1: A wind rose for Nairobi during the campaign period

4.2. Calibration Concentration for OPC-N2 Data

Table 4.2 shows the mass concentrations of $PM_{2.5}$ and PM_{10} obtained using gravimetric methods. The average mass concentrations were used in the determination of the correction factor for the OPCs. The weighing balance values were multiplied by a factor of 0.83, which was derived from a comparison of mass values of a sample weighed using the mass balance at the Institute of Nuclear Science and Technology and those obtained for the same sample at the Lamont-Doherty Earth Observatory, Columbian University, New York, USA.

By comparing the mass concentrations obtained from the gravimetric analysis and those from the OPC-N2 using equation 1 correction factors for the low-cost sensors were 1.63 and 1.70 for $PM_{2.5}$ and PM_{10} respectively.

Table 4. 2: Shows the calibration results from the gravimetric instrument used in the calibration of OPC-N2

Particle mass fraction	**Empty filter weight (mg)**	**Filter+ Sample weight (mg)**	**Particulate Matter (µg)**	**Atmospheric Conc. (µgm-^{3})**	**Corrected Conc. (µgm-^{3})**	**Average Mass Conc. From OPC-N2**
Fine $PM_{2.5}$	122.3	123.1	800	33.3	27.6	16.9
Coarse $PM_{10\text{-}2.5}$	123.3	124.0	700	29.2	24.2	30.6
Respirable PM_{10} ($PM_{10\text{-}2.5}$+ $PM_{2.5}$)			1500	62.5	51.8	

The correction factors obtained in this study were high compared to those obtained by **Sousan et al., (2016)** where mass concentration of PM obtained using OPC-N2 against those from gravimetrically corrected instruments; (SMPS + APS) showed an almost linear relationship. The differences could be attributed to the fact that this study tested the performance of the OPC-N2 in a real-case set-up while the Sousan et al. (2016) used laboratory reference aerosol dust from the Arizona. Real cases involving complexation of aerosols, and variations of RH and temperature in the atmosphere have an impact on the PM sampled using OPC-N2 instruments. While this study made direct comparison of the PM measurements obtained by the OPC-N2 and a gravimetric reference instrument, **Crilley et al., (2017)** used an indirect approach where gravimetrically corrected TEOM air samples were used in the comparison of the mass of $PM_{2.5}$ and PM_{10} obtained by the OPC-N2. The correction factors in this study were found to be between 3 and 4 times higher than those sampled using the TEOM.

4.3. Particulate Matter Mass Concentration

Figure 4.2 provides a time series for $PM_{2.5}$ and PM_{10} sampled at the three sites using OPC-N2 sensors. There are instances where missing gaps were observed and this is presented by the grey lines in the figure. This included a period when two or three of the sensors were collocated at a given site for inter-instrument cross comparison. Other reasons for the gaps include when either the OPC-N2 malfunctioned or power failed thus requiring the restart of the equipment.

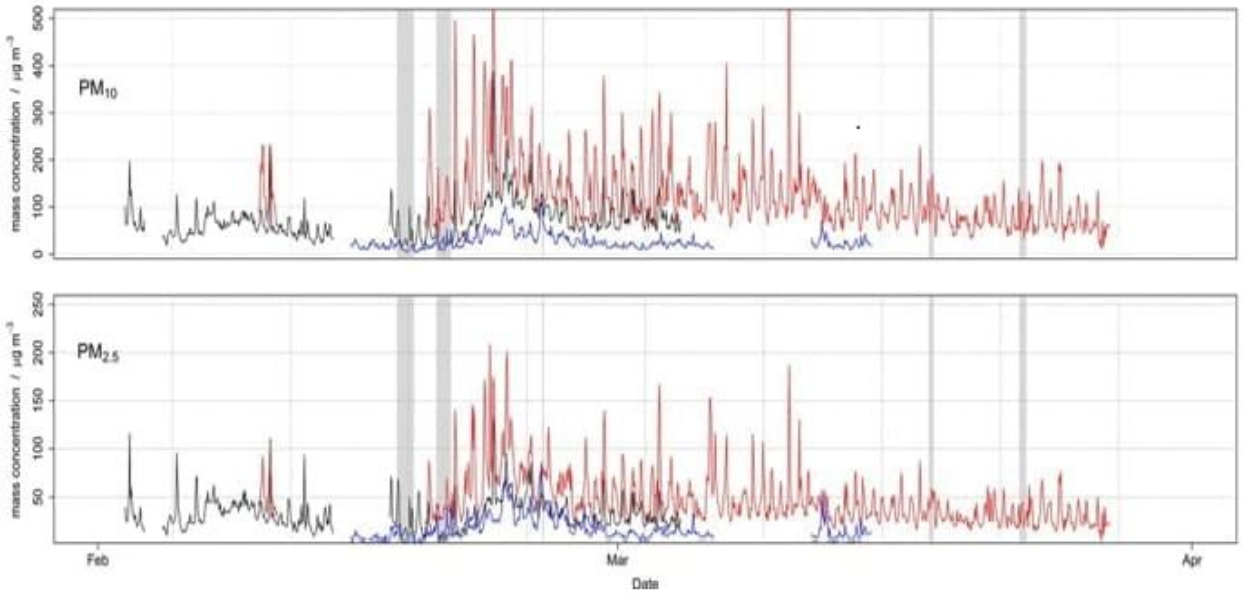

Figure 4. 2: The time series of PM mass concentration measurement at the three sites.

The time series show a good incremental pattern in the three sites across the campaign period.

A distinct peak of PM mass concentration in the two size fractions was observed in the three sites on 23rd day of February 2017. The peak is likely an indication of a long-range pollution during the study. For the two size fractions; $PM_{2.5}$, and PM_{10} measured using the OPC-N2, the rural background site represented by the blue lines in the time series had the lowest mean daily concentration followed by the urban background sites represented by the black lines in the graph. The highest mean daily concentration was noted in the urban roadside site, represented by a red

line in the graph. The incremental pattern from the rural to urban background and urban roadside site can be explained by the difference in the economic activities in the three sites. Therefore, having few economic activities such as transport and industrial activities in the rural areas explains the low concentration of the PM in the site as compared to the urban background and urban roadside site. The urban roadside site had the highest concentration of PM in all size fractions as shown in the graph because of the presence of local sources such as high vehicular emission compared to the urban background site, where the influence of local sources is minimal.

From the study, the mean daily concentrations of fine $PM_{2.5}$ were 13 ± 7, 25 ± 14 and 37 ± 26 μgm^{-3} while those of the respirable PM_{10} were 20 ± 9, 53 ± 34 and 94 ± 50 μgm^{-3} for the three sites respectively. The WHO limits for both PM_{10} and $PM_{2.5}$ were exceeded in the urban roadside site in Nairobi during the study.

The results are comparable to those obtained by Gaita et al. (2014) who observed an overall mean of 21μgm^{-3} for $PM_{2.5}$ at the urban background site thus an indication of the growing levels of APM pollution in Nairobi. Findings by Kinney et al., (2012) showed a decrease in $PM_{2.5}$ mass concentration from a vertical dispersion site on the roadside, 120 μgm^{-3} to 43μgm^{-3}, thus affirming the significance of local traffic in Nairobi as observed in this study.

4.4. Percentage Daily Exceedances of PM

In assessing the quality of air exposed to a given population, standardized limits set by environmental bodies such as WHO are used. The recommended daily limits for fine particulates ($PM_{2.5}$) and respirable PM_{10} are 25 and 50μgm^{-3} respectively (**Kumar et al., 2015**). For this study, the status of air quality was compared against limits set by the WHO. Percentage daily exceedances of the WHO limits were calculated. Table 4.3 below provides the mean daily APM mass concentrations and the daily particulate matter exceedances of $PM_{2.5}$ and PM_{10} as per the guidelines

set by the WHO. It also shows the daily exceedances of WHO limit for both fine and coarse particulates in all the three sites. The urban roadside site had the highest number of days exceeding the limits while the rural site had the lowest. The low percentage of level of exceedances in the urban background site compared to the urban roadside site is due to the elevation of the site. The elevation removes the direct impact of local sources of particulate matter pollution in that site. Therefore, it can be assumed that the particulate mass concentration observed in this site represents the lowest limit for the ground level APM in Nairobi County as most of the emissions in the city is due to ground level sources such as vehicular emissions, biomass burning, local industries and others.

Table 4. 3: The % daily exceedances of particulate matter concentration in the three sites. The recommended WHO daily limits for fine particulates (PM2.5) and respirable PM10 are 25 and 50µgm-3 respectively.

Sampling site	No of days measurements taken	Average $PM_{2.5}$ mass concentration (μgm^{-3})	Average PM_{10} mass concentration (μgm^{-3})	% daily $PM_{2.5}$ exceedances	% daily PM_{10} exceedances
Urban background	29	25	53	32	40
Urban roadside	40	37	94	85	90
Rural background	25	13	20	12	0

For this study, there was inadequate temporal data to provide an annual average value for PM mass concentrations. Therefore, the yearly average values for Nairobi can be estimated from the data using the mean average values in Table 4.3.

Since the study was conducted when there was little precipitation and thus minimal wet deposition of APM mass loadings during the campaign period, the average values are likely to be the upper estimates for the annual values. The short-rainy season in Nairobi (October-December) reduces

the particulate matter concentrations at the American Wing site by almost two times (**Gaita et al., 2014).** The average measurements observed in this study for $PM_{2.5}$ and PM_{10} at the American wing site exceeded the annual WHO limits by factors of 3 for both fine and coarse while for the roadside site, the factors were 4 and 5 for the $PM_{2.5}$ and PM_{10} respectively. The high levels of exceedances for both the urban background and roadside site suggested that majority of the urban populace in Nairobi will be subjected to poor air quality in excess of the annual limits set by WHO for particulate matter. Figure 4.3 shows a box and whisker plot depicting the hourly averaged data for $PM_{2.5}$ and PM_{10} mass concentration observed at the three sites:

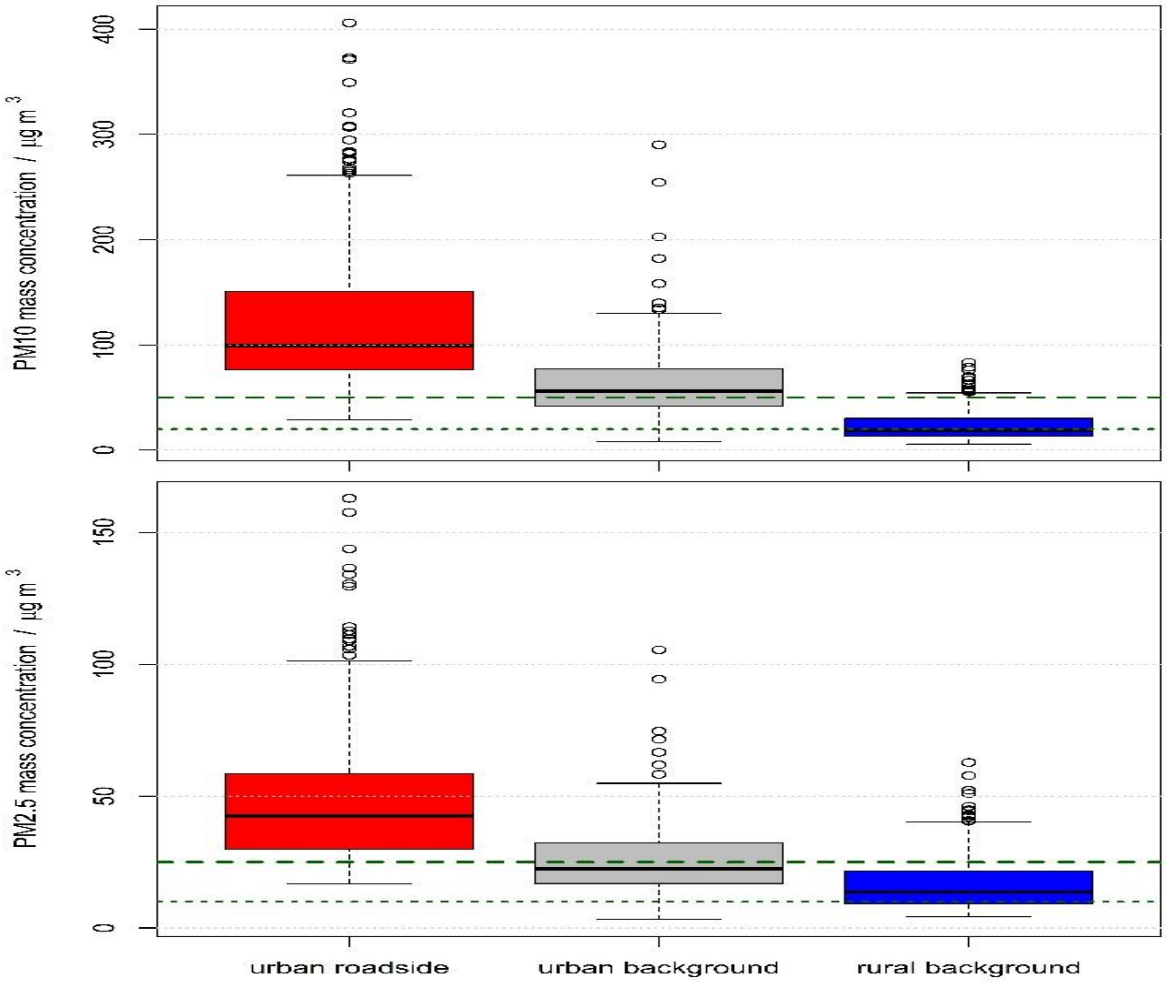

Figure 4. 3: Box and whisker plot showing hourly averaged data for PM mass concentration observed at the three sites.

Other information provided in the figure 4.3 includes the daily and annual limits set by WHO for particulate matter. This information is represented by green dashes and dotted lines for annual and daily PM concentration limits respectively.

4.5. Temporal Variation in PM Measurements

The estimation of the extent of the impact of air pollution in any given urban center requires a proper understanding of the temporal trends in APM mass concentrations (**Jhun et al., 2015; Liu et al., 2018**). One such trend is the use of diurnal variations to check the difference in these concentrations at different times of the day. Figure 4.4 below shows the diurnal variation in the three size fractions observed during the campaign period, in the three sites:

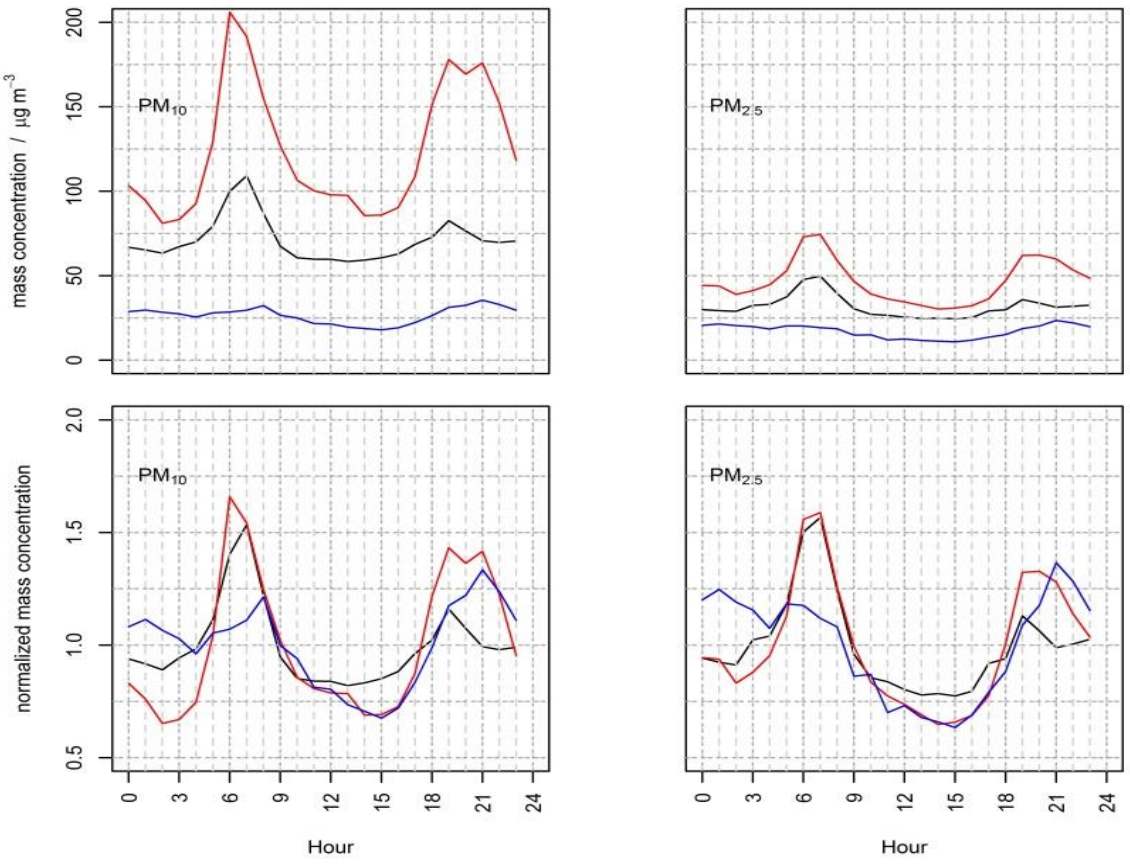

Figure 4. 4: The mean average diurnal profiles for the two size fractions measured using the OPC-N2 at the three sites during the campaign period

From Figure 4.5, the top and bottom panels show the measured mass concentrations and their normalized concentrations at the three sites. Normalization was done by subtracting the mean concentration from the concentrations obtained and dividing the final value by the variance. A clear diurnal variation with distinct peaks was observed in both the urban road side site represented by a read line and the urban background site represented by a black line. The observed peaks coincided with the peak hours during the morning hours, 5:00 A.M to 10:00 A.M and in the evening peak, 6:00 P.M to 12:00 A.M. These periods correspond to the times when traffic in Nairobi is at its peak. For the rural background site; represented by blue lines in the profile, a distinct diurnal variation pattern was observed. The patterns show similar traffic-related signals but more pronounced in the fine particulate size fraction. However, the overall diurnal variation pattern for the rural site corresponds to the solar insolation between rural and urban sites. This suggests that the major factor affecting the APM mass concentration in Nanyuki site is the height of the local boundary layer, which decreases during the nighttime and increase in high solar insolation.

4.6. Lenschow Incremental Analysis of PM Measurements

The comparison of APM data collected in rural background, urban background and roadside sites helps in estimating the level of urban and roadside increments in urban centers. For this study, a Lenschow-type approach was used in estimating the increments. They were calculated for all the three size-fractions. Urban increments were estimated by finding the difference in the hourly average values of APM mass concentrations observed during the study at both the urban and rural backgrounds. Back trajectory analyses of air mass flow showed that winds reaching Nairobi were dominantly northeasterly and this made the Nanyuki site a good representative of a rural background influencing the quality of air in Nairobi. For the roadside increment, hourly average

values of APM mass concentrations in the urban background site were subtracted from those observed at the urban roadside site. This site has high fleet density compared to other streets in Nairobi as it serves as a terminal point for the popular "matatus" or public service vehicles. Therefore, the roadside increment observed here is likely a representative of a value closer to the upper limit for most of the roads in Nairobi. Table 4.4 below depicts summary statistics for the urban and roadside increment of the three size-fractions studied in this campaign:

Table 4. 4: A summarized statistic of the urban and roadside increment of PM size fractions measured during the campaign period

	Urban site Increment (μgm^{-3})		Roadside site Increment (μgm^{-3})	
	$PM_{2.5}$	PM_{10}	$PM_{2.5}$	PM_{10}
Minimum	-27	-24	-6	-32
1st Quartile	2	20	11	22
Median	7	33	18	43
Mean	8	37	23	58
3rd Quartile	13	48	30	83
Maximum	86	258	124	293

From the results shown in the table 4.4 the mean roadside enhancement over the urban background was found to be 23 and 58 μgm^{-3} respectively for the two size fractions measured using the OPC-2. In addition, the hourly average urban background concentration was found to exceed the rural background concentration by 8 and 37 μgm^{-3} for the two size fractions. The results are in agreement

with the Lenschow model that predicts the impact of regional sources of particulate matter pollution on urban background sites, which are representatives of the lower limits in PM mass concentration in urban centers (**Lenschow et al., 2001).** Additionally, the large difference between the urban and roadside sites increments affirm the significance of traffic-related pollution in an urban center.

The roadside increment values in this study are comparable to those obtained by Kinney et al., (2011) where vertical dispersion measurement revealed a decrease in $PM_{2.5}$ concentrations from 119.5µgm^{-3} at a street level to 42.8µgm^{-3} on a third-floor rooftop in the CBD of Nairobi. The difference in the two studies can be explained by the difference in sampling times. Kinney et al., (2011) sampled for 11-h periods subdivided into two shifts while this study was taken continuously hence the slight variation.

4.7. Spatial Variation of PM Measurements

Atmospheric factors such as wind direction and wind speed affect the ways in which particulate matter mass concentration is dispersed in a given geographical area (**Jhun et al., 2015).** In this study, spatial variation of PM mass concentration at the urban background and road side site was determined using bivariate polar plots from the Open-air package in R. The input data for the plots were the wind parameters and PM mass concentrations at the two sites. The applicability of the wind data from the urban background site to the two sites (urban background and urban roadside sites) was because of their proximity to each other at an aerial distance of less than 0.5 km apart. The rural site was not included in the spatial analysis as it was not possible to get wind direction data from that site.

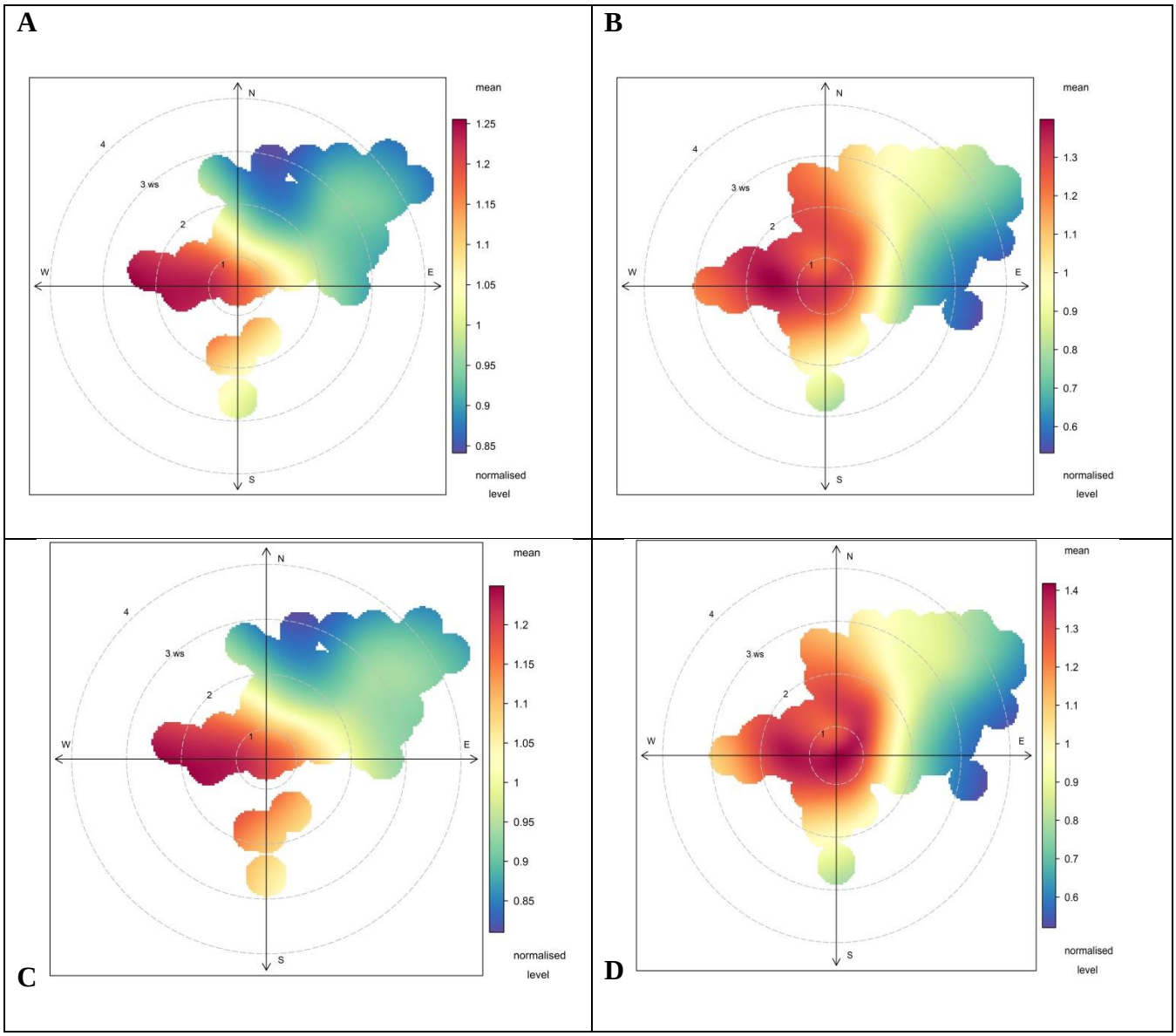

Figure 4. 5: Bivariate polar plots: A and B are the polar plots for the spatial variation of PM2.5 and PM10 respectively in the Urban Background (UB) site. Plots C and D show the polar plots for the spatial variation of PM2.5 and PM10 respectively in Urban Roadside (UB) site.

Normalization was done by subtracting the mean concentrations from the concentration measured and dividing the final value by the variance of the concentration. The normalization was done to ease comparison between the PM size fractions studied at the different sites.

From the bivariate polar plots in figure 4.5, a significant variation between APM mass concentrations at the two sites in Nairobi is observed. For instance, at the UB site, the spatial variation for the two size-fractions was observed. High levels of concentrations of APM were observed at low speeds and when the winds were blowing from the south-west direction. These directions coincide with the proximity of the UB site to the Mombasa-Nairobi-Malaba highway. In addition, the roadside increment and diurnal variations discussed previously coupled with the general wind direction observed highlights the significant role played by roads in contributing to APM pollution in Nairobi.

At the UR site, a distinct spatial variation in APM concentration was observed. For instance, the highest concentration of PM_{10} observed when the winds were blowing from the north west and least when blowing from the north east direction. A steady reduction in the concentrations was observed between the two extremes. In the case of $PM_{2.5}$, the highest concentrations were observed at low speeds and when the wind was blowing from the west and north directions. The high concentration of $PM_{2.5}$ at the roadside was likely due to the heavy congestion of vehicles at the UR site. Most of the vehicles “matatus” idle at the site thus emitting fumes, which are mostly in the range of $PM_{2.5.}$

4.8. Ratio Distribution between Coarse and Fine PM

In the determination of how long particles stay afloat in the atmosphere and where they get deposited in the human respiratory system requires an understanding of their sizes. Therefore,

analyzing the ratio distribution between coarse and fine particulate matter size fraction is an important parameter in air pollution and monitoring studies. For this study, the ratio of such distribution at the three sites is shown in figure 4.6 below. There is a distinct variation in each of the sites as evident in the reported distribution ratio between coarse and fine PM. Variations in the median ratio at each of the three sites were observed. For the rural background, American Wing and road sites, the median ratios were 0.5, 1.3 and 1.6 respectively. Although there is no universal constant median ratio for coarse to fine PM that applies to all sites, a ratio of above 0.73 highlights the impact of anthropogenic sources at the site (**Marcazzan et al., 2002**). This explains the high difference between the median ratios at the roadside site compared to the rural background which has a lower ratio. Small ratios are indicative of natural sources such as mineral dust as the dominant source of pollution at the site (**Sugimoto et al., 2016).** The attribution of PM to vehicular emissions reported in this study from the high median ratio of coarse to fine coincides well with the findings observed by **Gaita et al., (2014)** where dominant sources of PM in Nairobi were found to be mineral dust resuspension by non-exhaust vehicular emissions. In addition, the ratio observed at the background site is an intermediate between both the rural and roadside site thus an indication that it is likely affected by both local and non-local sources. The ratios were consistent with the urban and roadside increments discussed in table 4.4.

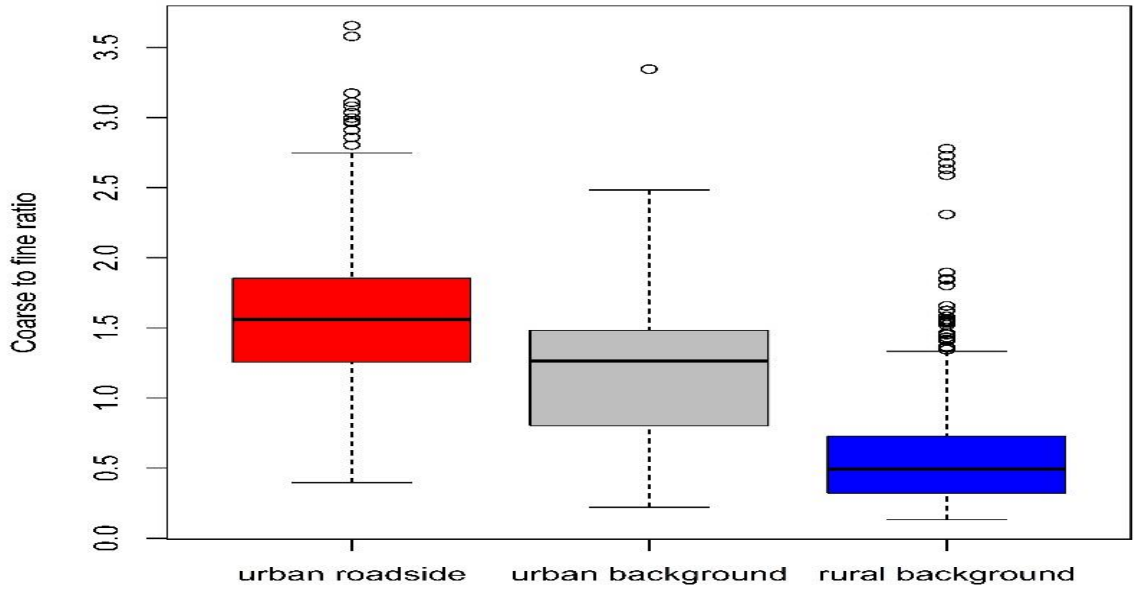

Figure 4. 6: Box and whisker plot depicting the ratios of coarse to fine fraction of PM

4.9. Elemental Analysis of PM

In the analysis of the elemental composition of the $PM_{2.5}$ sampled at the American Wing site, loaded filters were analyzed using EDXRF spectrometry method. This method was first validated using a set of certified reference materials, ICP multi-elements for thin samples from the International Atomic Energy Agency (IAEA, 1997). The aim of the validation was to establish whether the values obtained experimentally using the EDXRF were in statistical agreement with the certified values. Results of the validation analysis of the certified reference materials are shown in the table 4.5 below:

Table 4. 5: The experimental values against certified values as mass loadings per filter in units of ug per filters

	Experimental concentrations (µg/filter)	Certified concentrations (µg/filter)
Ca	14.6 ± 2.1	13.2 ± 2.3
Cr	<0.15	
Mn	0.3 ± 0.1	0.3 ± 0.1
Fe	27.3 ± 1.6	26.5 ± 1.6
Zn	1.8 ± 0.1	1.8 ± 0.1

The validation results agreed well with the certified values and within acceptable margins of uncertainty thus proving the accuracy of the concentration results obtained by EDXRF method. The data from the certified reference materials was further used in the calculation of the detection limits based on the IUPAC equation described by van Grieken and Markowicz and (2002).

The elements Ca, Cr, Mn, Fe and Zn in $PM_{2.5,}$ sampled at the American Wing site, were determined above the detection limits in most of the filters. The detected trace metals accounted for less than 15% of the total $PM_{2.5}$. The remaining 85% of the $PM_{2.5}$ constituents could be components such as black and elemental carbon, persistent organic pollutants (POPs), nitrates, sulphates, metal oxides, silicates and other chemicals that could not be effectively detected using EDXRF analysis. Example of such chemicals includes all the low Z-elements such as Na and those below it in the periodic table. The highest concentration of trace elements was observed in Fe and

Ca with average concentrations of 496 ± 32 ngm^{-3} and 408 ± 106 ngm^{-3} respectively. The high concentrations of Fe and Ca observed highlighted the significance of the impact of resuspension of mineral dust as a source of particulate matter in Nairobi. The mean concentrations of Zn, Mn and Cr observed in this study were 43 ± 5 ngm^{-3}, 8 ± 1 ngm^{-3} and 2 ± 1 ngm^{-3} respectively. The presence of Mn, Cr and Zn could be attributed to the contribution of mixed sources such as biomass combustion, industrial and traffic emissions (Dai et al., 2015). Zn and Cr are identified with the presence of industrial activities (**Tchounwou et al., 2012**), and this is likely an indication of the significance of industrial activities as contributors towards ambient PM in Nairobi.

Table 4. 6: A comparison of the concentration of the elemental components of PM2.5 in this study with those from other studies conducted in the different cities in the region

	This study (ngm^{-3})	**Gaita et al., (2014) in Nairobi** (ngm^{-3})	**Bennet et al., (2005) in Dar es Salaam** (ngm^{-3})	**Ezzeh et al., (2015) in Lagos** (ngm^{-3})
Fe	496 ± 32	730 ± 340	44 ± 15	1579 ± 22
Ca	408 ± 106	340 ± 270	270 ± 89	2309 ± 8
Zn	43 ± 5	120 ± 12	3 ± 1	7 ± 3
Mn	8 ± 1	53 ± 23	91 ± 69	27 ± 2
Cr	2 ± 1	Not reported	Not reported	97 ± 2

Elemental concentration results in this study are comparable to those obtained in different studies conducted in Nairobi and other cities in the region as shown in Table 4.6. Gaita et al., (2014) observed higher concentrations of elemental components in $PM_{2.5}$ compared to what was reported in this study as shown in table 4.7. The higher concentrations observed by **Gaita et al. (2014)**

could likely be explained by the difference in the collection period. This study focused on sampling of PM in two months while the latter sampled between 2009 and 2010 when Kenya experienced a severe drought hence high levels of pollutants associated with the resuspension of dust and other PM sources.

The lower concentration of elemental components in PM observed by **Bennet et al., (2005)** could be attributed to the low traffic density and economic activities in Dar es Salaam compared to Nairobi, which is the regional economic hub of East Africa. Similarly, the findings in this study can be compared to those obtained by **Ezeh et al., (2015)** where higher concentrations were recorded in the elements in $PM_{2.5.}$ The large size and population of Lagos makes it prone to high level of emission from traffic and other anthropogenic sources compared to the city of Nairobi. Besides, the samples in **Ezeh et al., (2015)** were collected at a roadside site while those in Nairobi were collected at an urban background site.

4.10. Positive Matrix Factorization Analysis and Source Apportionment

The development of proper and effective control strategies to mitigate the impacts of air pollution requires adequate understanding of the concentrations as well as the sources of pollutants. This requires the use of multivariate statistical receptors models such as positive matrix factorization (PMF) analysis. In this study, PMF version 5.0 by EPA was used for analysis (**USEPA, 2014**). The software had 20 parameters, which were all retained in the analysis as part of minimizing the model uncertainties. The inputs of the model were the total concentrations of $PM_{2.5}$ and the speciated elemental concentrations together with their respective uncertainties. In instances of missing values, the median was used instead and its uncertainty estimated as four times the median (**Polissar et al., 1998; Reff et al., 2007**). Chromium was treated as a weak signal because of its low signal-to-noise ratio, which was <2 (EPA version 5.0). The total $PM_{2.5}$ was considered a weak

signal by default by the software. Factors were varied from one to 9 and based on the signal output, only two gave meaningful results for the probable sources. The first factor was ascribed to mineral dust sources as shown in Figure 4.6. The factor is dominated by elements associated with crustal rocks (Fe, Ca and Mn). The factor had a similar pattern to the mineral dust cluster obtained for trace elements in soil samples from different parts of SSA (**Towett et al., 2015**). This highlights the significant contribution of mineral dust to $PM_{2.5}$ in Nairobi. The presence of Manganese in urban air in Nairobi could be attributed to the windblown erosion of mineral dusts, resuspension by vehicles and soil.

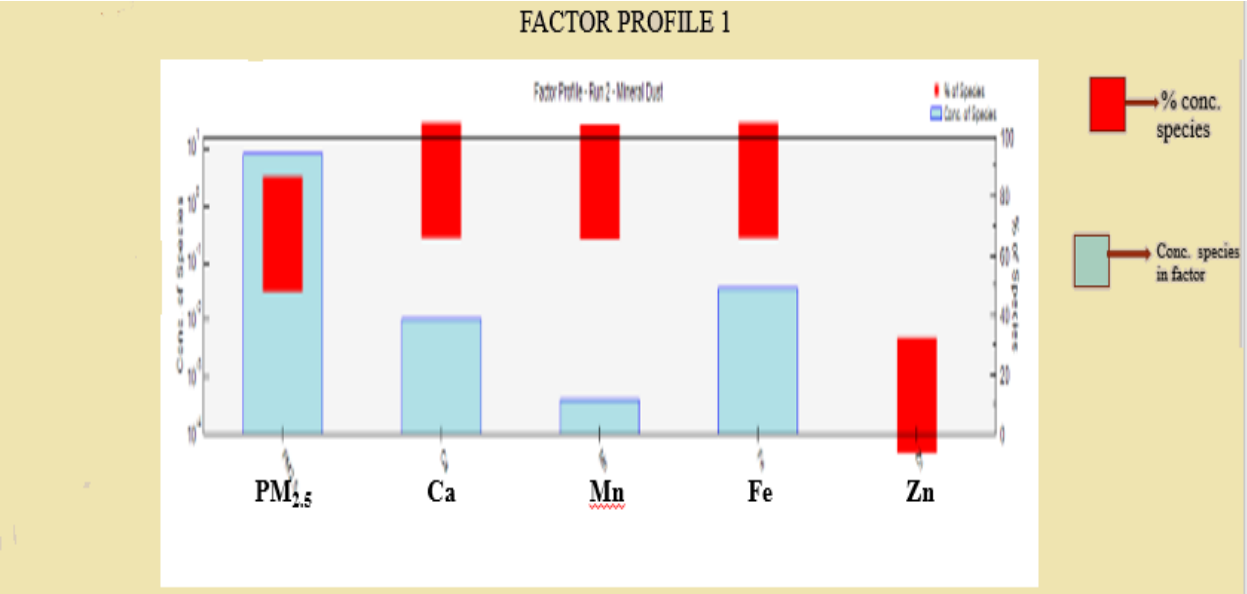

Figure 4. 7: Mineral dust factor from PMF analysis of trace metal concentrations in PM2.5 sampled at the UB site in Nairobi.

The second factor was attributed to a mixture of sources ranging from industrial and traffic related emissions as shown in Figure 4.7. These sources account for 33% of the mass concentration analyzed. The presence of Zn in the factor profile is indicative of the presence of industrial sources in the city. Within the city, there are metal smelters commonly known as “Jua kali” and this could be the probable source of the metal in urban air in Nairobi. Another probable source is the

resuspension of dust containing deposits of zinc released during the abrasion of tires and tail-pipe exhausts from vehicles (**Norrström and Jacks, 1998**).

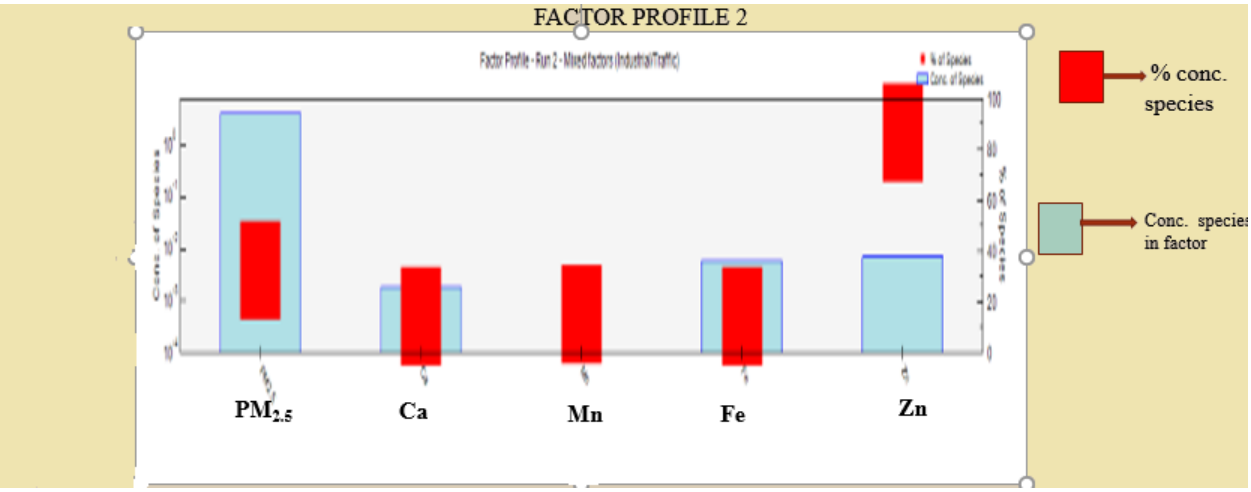

Figure 4. 8: Shows mixed factor (traffic and industrial emissions) from PMF analysis of trace metal concentrations in PM2.5 sampled at the UB site in Nairobi.

The PMF analysis conducted was limited by the lack of adequate data on speciated elemental concentration from the $PM_{2.5}$ sampled at the urban background site and only five elements were reported in this study. Compared to thirteen elements observed in **Gaita et al. (2014),** the lack of enough data resulted to a smaller number of factor profiles after PMF analysis. Hence, sources such as biomass combustion, sea salt and secondary aerosol particles were not accounted for in this study. The low number of elements observed and a smaller number of factors in this study could be attributed to the limitation of the instrument used in the quantification and characterization of elements below the limits of detection.

Chapter 5: Findings and Recommendations

5.1. Findings

The findings of this study show that residents of Nairobi are exposed to low air quality. This is evident in the regular exceedance of the WHO daily limits for both $PM_{2.5}$ and PM_{10} at the two urban sites.

A clear diurnal pattern in APM mass concentration was noted in the peak traffic periods which was consistent with the roadside increment, spatial variation and source apportionment findings that implicated traffic emissions as a dominant source of air pollution in Nairobi.

The presence of mineral dust elements (Fe and Ca) affirmed the contribution of mineral dust as a significant source of APM in Nairobi while Mn, Cr and Zn indicated the contribution of mixed sources such as biomass burning, industrial and traffic-related emissions. This was consistent with source apportionment findings obtained using the PMF model.

Results obtained from this study show that the use of low-cost and calibrated OPC-N2 sensors collocated alongside standard gravimetric reference methods can provide cheap and reliable air quality measurements in urban centers in the developing world.

5.2. Recommendations

The use of low-cost calibrated OPC-N2 sensors collocated with a gravimetric reference proved effective in the measurement and monitoring of PM and their elemental composition in Nairobi. However, owing to the limited number of calibration points, it was difficult to obtain a linear relationship between data obtained from optical and gravimetric methods. Therefore, it is recommended that future studies on the same have more than one calibration points to increase the reliability of the results obtained. Besides, a broad scope of the study should be conducted with many sensors deployed in many sites across the city for a long time will help in the observation of the seasonal variation of PM in the areas sampled.

Whilst this study focused on PM and elemental composition, studies have shown that there are other pollutants which cause even greater health risks apart from those studied. These pollutants include NO_2, ground level O_3, SO_2 and PAHs among others. There is a need to increase the scope of analysis of these other pollutants to understand the health risks associated with airborne pollutants in Nairobi and other cities in SSA.

Based on the findings of this study, traffic-related emissions play a significant role in contributing towards PM pollution in Nairobi. Therefore, there is need to conduct more studies that will back mitigation policies aimed at reducing traffic-related emissions in not only Nairobi but also in other cities across SSA. Some of these policies should aim at decongesting the cities and introduction of proper maintenance of vehicles.

In addition, the promotion of citizen science on the proper use of low-cost sensors in air pollution measurements is a recommended plan that will aid in dealing with the shortage of data on the state of air quality in Nairobi and other cities across SSA.

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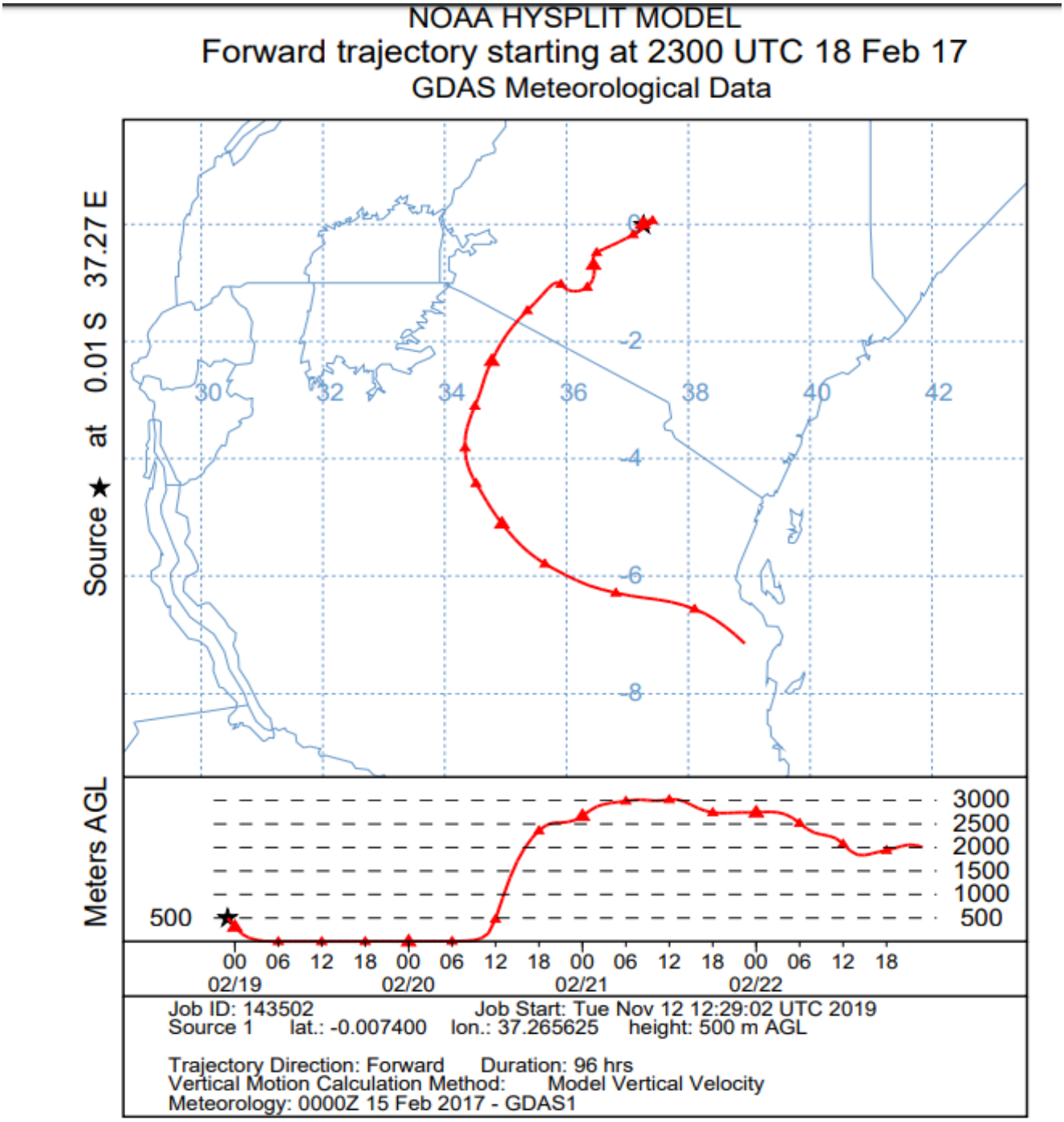
7.Appendix

Appendix 1. Back trajectory results for four days showing influence of Wind from Nanyuki in Nairobi



Appendix 2. Concentration of elements in particulate matter analyzed using EDXRF

	Average mass Concentrations of elements in PM2.5 (ngm-3)	Uncertainties in the mass concentration (ngm-3)
Ca	408	106
Mn	8	1
Fe	496	32
Zn	43	5
Cr	2	1

Appendix 3: Daily Aggregate Data for Particulate matter ($PM_{2.5}$ and PM_{10}) sampled at the Urban Roadside site in Nairobi

Date	Corrected PM2.5	PM10
2/18/2017	22.7787	77.19944
2/19/2017	17.12635	43.11679
2/20/2017	25.87133	65.24994
2/21/2017	36.46343	92.11923
2/22/2017	46.61609	119.0858
2/23/2017	45.5688	100.1889
2/24/2017	34.22143	73.95148
2/25/2017	32.16147	61.83067
2/26/2017	25.22132	53.20495
2/27/2017	25.55613	62.60877
2/28/2017	26.8168	64.03514
3/1/2017	24.33544	61.12615
3/2/2017	22.29009	63.47977
3/3/2017	28.65696	73.72633
3/4/2017	20.67127	54.1747
3/5/2017	21.81996	55.2673
3/6/2017	31.38831	72.4139
3/7/2017	18.29083	53.29386
3/8/2017	22.85108	61.79992
3/9/2017	19.50581	53.49177
3/10/2017	25.39367	80.1667
3/11/2017	21.81913	55.69724

3/12/2017	17.63508	38.52658
3/13/2017	19.006	48.48105
3/14/2017	18.02189	45.57676
3/15/2017	17.95534	40.1977
3/16/2017	18.64323	43.42164
3/17/2017	18.48036	45.18886
3/18/2017	17.49347	39.94181
3/19/2017	13.36784	28.76606
3/20/2017	15.77762	33.5341
3/21/2017	17.41859	38.12979
3/22/2017	15.44679	30.17158
3/23/2017	18.41629	41.08645
3/24/2017	17.7532	40.4519
3/25/2017	14.95946	31.38262
3/26/2017	13.43032	27.36449
3/27/2017	14.1954	30.44082
3/28/2017	11.75146	23.31414
3/29/2017	12.95752	24.70257
3/30/2017	15.94738	31.99421
3/31/2017	20.58648	47.05997
4/1/2017	21.41114	45.21082
4/2/2017	14.35106	31.16085
4/3/2017	17.73746	35.00572

Appendix 4: Daily Aggregate data for Particulate matter ($PM_{2.5}$ and PM_{10}) sampled using OPC-N2 in the Urban background

Date	Corrected PM.2.5	Corrected PM.10
2/4/2017	10.00437	16.64758
2/5/2017	13.22293	19.24988
2/6/2017	16.49496	27.32825
2/7/2017	19.97639	32.01806
2/8/2017	20.42581	32.93638
2/9/2017	17.94007	31.06605
2/10/2017	15.43821	27.96907

2/11/2017	12.16782	21.87235
2/12/2017	11.37193	18.6076
2/13/2017	12.27813	18.93375
2/14/2017	NA	NA
2/15/2017	NA	NA
2/16/2017	23.54657	39.57896
2/17/2017	15.97145	20.539
2/18/2017	18.16405	40.24752
2/19/2017	17.12393	43.06382
2/20/2017	25.89919	65.3138
2/21/2017	36.47564	92.18582
2/22/2017	46.66745	119.1179
2/23/2017	45.50816	100.0916
2/24/2017	34.23392	73.9186
2/25/2017	32.11037	61.78532
2/26/2017	25.19969	53.14171
2/27/2017	25.5707	62.6682
2/28/2017	26.83137	64.04837
3/1/2017	24.29066	60.95573
3/2/2017	22.23778	63.4166
3/3/2017	28.72356	73.97746
3/4/2017	20.00396	49.00852

Appendix 5: Daily Aggregate data for Particulate matter ($PM_{2.5}$ and PM_{10}) sampled at the rural background site in Nanyuki.

Date	Corrected PM.2.5	Corrected PM.10
2/14/2017	4.6305	10.26516
2/15/2017	3.295384	7.684739
2/16/2017	4.207759	7.468842
2/17/2017	4.944492	6.740909
2/18/2017	3.88233	5.161764
2/19/2017	7.149931	8.663315
2/20/2017	8.053257	10.42202

2/21/2017	10.59745	16.06035
2/22/2017	16.91073	24.75721
2/23/2017	18.39578	24.19221
2/24/2017	16.7372	21.91322
2/25/2017	15.20841	18.50708
2/26/2017	10.52643	13.33986
2/27/2017	8.481963	11.22893
2/28/2017	6.393353	8.815042
3/1/2017	5.471445	8.114812
3/2/2017	4.450125	6.806517
3/3/2017	6.702686	10.16199
3/4/2017	6.612215	9.966465
3/5/2017	6.058904	9.024875
3/6/2017	4.519543	6.293264