SPATIAL-TEMPORAL VARIABILITY OF TROPOSPHERICAL OZONE IN CITY OF NAIROBI, KENYA

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DECLARATION

I understand that plagiarism is an offence and therefore declare that this thesis is my original work and has not been presented to any other institution for any other award.

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DEDICATION

I would like to dedicate the research study to my wife, Stella Mueand my children Kelvin and Ndanu

TABLE OF CONTENTS

LIST OF TABLES

LIST OF FIGURES

ACRONYMS AND ABBREVIATIONS

AMSL	Above Mean Sea Level
CBD	Central Business District
DJF	December, January and February
GHG	Greenhouse Gas
GIS	Geographic Information Systems
HYSPLIP	Hybrid Single - Particle Lagrangian Integrated Trajectory
IPCC	Intergovernmental Panel of Climate Change
JJA	June, July and August
KMD	Kenya Meteorological Department
MAM	March, April and May
MKNGAW	Mount Kenya Global Atmosphere Watch
NCEP-NCAR	National Centre for Environmental Prediction -National Centre for Atmospheric Research
NOAA	National Oceanographic and Atmospheric Administration
O_3	Ozone
SON	September, October and November
TOCs	Tropospheric Ozone Columns
UNEP	United Nations Environmental Programme
UV	Ultra violet
VOCs	Volatile Organic Compounds
WDCGG	World Data Centre for Greenhouse Gases
WHO	World Health Organization
WMO	World Meteorological Organization

ABSTRACT

Nairobi is an urban city whoseair pollution is increasingly being recognizedas a major publichealthandenvironmentalissue.This is due to poor or deterioratingair quality which is as aresultsof high levels of energy consumption by industries, transport and domestic use. Thenatureofairpollutionisdependentonthesourceprofile of thecity and the presence ofsunlight topromote production ofsecondary pollutants, such as ozone, through photochemical reactions. The study sought to analyze the spatial-temporal variability of surface ozone over the city of Nairobi, and identify the source region. Surface ozone data for Nairobi was obtained from Kenya Meteorological Department ranging from the year 2011 to 2014. Another set of data was collected fromfour sampling sites for a period of ten days to determine the special variability and source of the surface ozone over the city of Nairobi. . Analysis of meteorological field from National Centre for Environmental Prediction -National Centre for Atmospheric Research (NCEP- NCAR) was used in running Hybrid Single Particle Lagrangian Integrated (HYSPLIT)model. From the analysis it was found out that June, July and August experienced the highest ozone levels compared to the other months of the year in both lower and upper levels. This was due to incursion from the south according to the backward trajectories from the HYSPLIT model, which has been proven to have high ozone concentration during this season due to high biomass burning. On the other hand, the diurnal variation of ozone in the foursampling sites namely Industrial Area,Nakumatt Junction, Landhies Road and Pangani Roundabout, showed low ozone levels in the early morning and at night hours, with the peak realized during the day. The peak in midday could be due to the fact that surface ozone was produced by photochemical oxidation of precursor gases that are produced by motor vehicle and industries. The highest eight-hour mean was 20.2 ppb from industrial area site, which is below the WHO mean of 51 ppb. Therefore, no much health effects are expected due to the exposure to surface ozone. It is recommended that there should be a continuous monitoring of surface ozone and other gases that are harmful to human health for better understanding and advice to the citizen.

CHAPTER ONE

1.0 Introduction

1.1 Background of the study

Ozone (O_3) is one of the most important trace gases in the Earth's atmosphere. It is naturallypresent in our atmosphere and is a critical atmospheric trace species in the stratosphere andtroposphere. Most O_3resides in the stratosphere where it traps the Ultra Violet (UV) raysmaking it a Green House Gas (GHG). The region between 20 and 40 km, where most atmospheric $O_3$is located,is commonly referred to as the ozone layer (Chalita *et al.,* 1996).

Ozone is one of the most important chemical species involved in atmospheric chemistry, owing to its role in oxidizing many atmospheric trace species and its potential impact on the environment (chemical oxidation capacity, health, greenhouse effect and vegetation) (Chameides, *et al.,* 1999;Mickley *et al.,* 2001). Great concern has always been attached to the elevated tropospheric ozone concentrations, particularly in densely populated urban and suburban areas. Photochemical ozone formation involving its chemical precursors and ozone transport from upwind air are the two major anthropogenic sources of ozone in rural and urban areas (Logan, 1985; Liu *et.al.,*1987). Ozone at ground level, poses adverse impacts on plants and animal even at low concentration (National Research Council, 1991). Unfortunately, ozone distributions within the troposphere are not sufficiently well researched and increasing trends in average tropospheric ozone concentrations remain a matter of discussion (Sarkar, 2012; Bojkov, 1988; Logan, 1994). The budget, and therefore the trend of tropospheric ozone are highly dependent on precursor emissions into the atmosphere of carbonaceous and nitrogenous precursors (Crutzen, 1988; Finlayson-Pitts and Pitts, 2000) occurring mainly in the boundary layer. Human activities contribute significantly to increased levels of certain precursors in relation to natural sources.

Over the years, GHGs have increased. World Meteorological Organization (WMO) estimates thattropospheric O_3 has increased by 36% since 1750 (WMO, 2012). Carbon dioxide(CO_2) has grown by about 40% of its pre-industrial period; when its concentration was about 280 parts per million volume (ppmv) of dry air to a concentration of 389 ppmv by 2010(World Data Centre for Greenhouse Gases (WDCGG), 2012). On the other hand, Methane (CH_4)has grown by about 150% to about 1780 parts per billion volumes (ppbv) of dry air. These increases have been as a result of increase in human activities; fossil-fuel combustion, cement manufacturing, transportation and deforestation.

Emission of precursors like carbon monoxide, methane and hydrocarbons during biomass burning activities contribute to the formation of O_3, through photochemical oxidation. Other biomass combustion by-products include primary and secondary aerosols that result in intense regional haze. Over the months of July to October, biomass burning in Africa, believed to be largely human initiated progresses southward from tropical West Africa towards Congo and Angola and ultimately south-eastward across Zambia, Zimbabwe and Mozambique, to ~25°S (Cahoon*et al*., 1992). Smoke and excess tropospheric O_3 emitted by this biomass burning have been observed over large regions of the tropics with satellites, aircraft, balloons and ground-based instrumentation (Fishman *et al*., 2003; Thompson *et al*., 2011; Kirchhoff *et al.,* 1991).

High smoke aerosol and enhanced tropospheric O_3 concentrations have been observed to have an atmospheric lifetime of a few weeks (IPCC 2013), but variable in terms of space and time (Sonkaew and Macatangay 2015). Tropospheric Ozone has also been observed over the adjacent Atlantic Ocean (Fishman *et al.,* 2003; Thompson *et al*., 1996, 2002),where strong ozone-biomass burning links have been confirmed by airborne and ship-based measurements (Andreae*et al.,* 1994; Jacob *et al.,* 1996; Weller *et al.,* 1996). It is evident that the highest values of tropospheric ozone were over the Atlantic Ocean, adjacent to the heavy biomass burning areas of Africa and Brazil. Vertical O_3 measurements during SAFARI- 92/TRACE-A showed that this ozone enhancement over

the mid-Atlantic occurred in an elevated mid-tropospheric layer, between 2 and 10 km altitude (Thomson *et al*., 1996).

Studies of vertical ozone profiles using ozonesonde measurements over Lusaka have shown elevated ozone in the boundary layer and mid-troposphere, thus providing an indication of enhanced ozone levels in the vicinity of biomass burning over the continent (Thomson *et al.,* 2002).

Ozone is the most important photochemical oxidant in the troposphere. It can have both good and bad impacts on human health and the environment. In the troposphere O_3 is a harmful pollutant that causes damage to plants and animal lung tissue. Itis a major constituent of smog (Rajab *et al*., 2010). Though it occurs naturally in the stratosphere to provide a protective layer high above the earth, at ground-level it is the prime ingredient of smog. When inhaled, even at very low levels, ozone can: cause acute respiratory problems, aggravate asthma, causes significant temporary decreases in lung capacity of 15% to over 20% in some healthy adults, cause inflammation of lung tissue, impair the body's immune system defenses, making people more susceptible to respiratory illnesses, and also interferes with the ability of plants to produce and store food, so that growth, reproduction and overall plant health is compromised (WHO, 2014; Lippmann, 1989).

It is formed by photochemical reactions in the presence of precursor pollutants such as NO_x and volatile organic compounds. In the vicinity of strong NO_x emission sources, where there is an abundance of NO, O_3 is "scavenged" and as a result its concentration is often low in busy urban centres andhigher in sub-urban and adjacent rural areas. On the other hand, O_3 is also subject to long-range atmospheric transport and is therefore considered as a trans-boundary problem (Thompson et al., 2002).

Tropospheric ozone acts both to control the oxidizing capacity of the atmosphere and as a greenhouse gas (Logan and Kirchhoff, 1986; Crutzen, 1988). Most of the oxidation of long-lived gases by hydroxyl radicals (OH) takes place in the tropics, where high UV and

humidity promote the formation of OH from the photolysis of ozone (Logan *et al.,* 1981; Thompson, 1992).As a result of its photochemical origin, O_3 displays strong seasonal and diurnal patterns, withhigher concentrations in summer and in the afternoons. The correlation of $O_3$with other pollutantsvaries by season and locations.

Satellite observations offer the possibility to measure the distribution of tropospheric ozone over large areas and to study its large-scale temporal and spatial variability. This is of great importance, since ozone, which is formed over regions where large amounts of ozone precursors are emitted can be transported over long distances and affect areas far from the source (Thompson et al., 2000).

Kenya Meteorological Department (KMD) monitors the concentration of greenhouse gases (GHG) at various sites in Kenya as one of its functions. Themonitored GHGs include O3, carbon dioxide (CO), carbon monoxide (CO), aerosols and particulatematter of different sizes. For almost six years, KMD only monitored vertical profiles and total column ozone using ozonesonde soundings and a Dobson spectrophotometer instrument. Since July 2012KMD installed surface ozone analysers specifically to measure ozone values at 10 m above the ground.

In 1999 the Mount Kenya Global Atmosphere Watch (MKN GAW) station was installed on north-western slope of Mt Kenya to automatically monitor the atmospheric concentrations of ozone and other gases. This location was chosen after a feasibility study in the East African region recommended the area as the most suitable for a background air pollution monitoring station. Here the measurement of other GHGs, along with the meteorological parameters, is done continuously. This automated program produces one-minute averages which are archived.

Generally, surface ozone has not been much recorded for a long period of time and only one station in the city of Nairobi monitors surface ozone. There is need for more stations,

since an increasing number of people in Kenya will be living in urban environments which is within the tropics, and there is possibility of increased negative health effects caused by the increased surface ozone.

1.2 Statement of the Research problem

In both developed and developing nations urban air pollution is increasingly being recognised as a major public health and environmental issue (Salviet al., 1999; WHO, 2001; Brunekreefand Holgate, 2002). Poor or deteriorating air quality in many cities results from high levels of energy consumption by industries, transport and domestic use (WHO, 2014).The nature of air pollution is dependent on the source profile of the city, the presence of sunlight to promote production of secondary pollutants, such as ozone, through photochemical reactions and the altitude, which affects combustion processes and global air circulation patterns.

The two sources of air pollutants in most urban areas are transportation and fuel combustion by stationary sources, including industrial heating (WHO, 2001). However, motor vehicle emissionsseemto be the dominant source of air pollutants especially in areas with high traffic densities and industrial activities (Seinfeld, 2006). In recent years, the public concern is being aroused due to the wide publicity on the damage to human health from the inhaling of gaseous pollutants and fine particulates. It has also been suggested that high incidence of respiratory health in urban areas may be associated with inhaling noxious gases and particulates in the air (Pope III, 2004).

The City of Nairobi is one of the fastest growing industrial and economic hubs in East Africa. This is likely to increase the population living in the City of Nairobi and hence, contribute to increase industrial/garbage waste production and transport activities, which are therefore, expected to increase the surface O3 and also likely to cause a lot of negative effects to both plants and human beingsSurface O3 is poisonous to plants and the breathing system of humans, and causes asthma attacks, among other effects.

Industrialisation and associated transport activities produce the O3 precursor gases like nitrogen dioxide (NO2), nitric oxide (NO); oxides of nitrogen (NO), Carbon monoxide among many others, that lead to the production and increase of O3 near the ground surface. This O3 may also be transported by airflows from other source regions into another area which is also need to be understood very well and caution given to the publicon the time and area where increase surface ozone is expected.

Pollution in the city of Nairobi is worsened by its increasing population, second hand cars, roadside rubbish fires, diesel generators, charcoal, paraffin and indoor cooking stoves which are predicted to cause urban health crisis leading to death of about 1.5 million people within a generation (UNEP; WHO 2001).Air pollution will have a huge economic and health impact with more cancers, heart, asthma and respiratory diseases predicted to increase. Despite existence of laws and policies on air pollution, Kenya is yet to start tests on emissions (KNBS; UNEP; World Bank 2016). There is evidence from controlled human and animal exposure studies of the potential for O3   to cause adverse health effects. Epidemiological studies have also addressed the effects of short and long-term exposures to O3 and provided important results. However, the health effects of O3 have been less studied than those of Particulate Matter (PM) and thus more research is needed, especially addressing the spatial, temporal and seasonal patternsof individual exposure in association with health outcomes.

1.3 Objectives of the study

The main objective of the study was to assesses spatial - temporal variation of surface Ozone and identify the source regions over the City of Nairobi.

The specific objectives of the study were to:-
- (i)   Investigate the spatial variation of surface Ozone over City of Nairobi.
- (ii)   Investigate the temporal Surface Ozone distribution over City of Nairobi.
- (iii)   Determine the source regions of surface Ozone observed over City of Nairobi.

1.4 Research questions

(i) How does surface Ozone vary spatiallyoverthe City ofNairobi?

(ii) How does surface Ozonevary intime over the City of Nairobi (Diurnal and Seasonal)?

(iii)What are the source regions of the observed surface ozone in City of Nairobi?

1.5 Significance of the study

Few studies have been made in the region regarding the levels of emissions of the Ozone as compared to the other air pollutants in the City of Nairobi (UNEP, 2017). These studies have been mainly for the distribution of Ozone (Thiong'o, 1997; Bundi, 2004; Barasa, 2007). The City of Nairobi, like many cities in Africa is experiencing rapid population growth. Taking into account, the fact that the city is also the economic hub of Kenya and indeed of the East and Central African region; this means, the city hasa higher number of inhabitants with lifestyles that demand high energy, more land for the built environment, and increasing consumption of natural resources. In order to mitigate the negative impacts of such growth, there is need for surface ozone monitoring and analysis so as to reduce its impacts on environment and human health.

The research intended to provide insight into variation of O_3 distribution in the City of Nairobi. It was therefore designed to increase our understanding of O_3 behavior and distribution in urban situation. Nairobi is build up area and the increase in motor vehicle and use of charcoal in slum area provide precursor gases for the formation of O_3. Therefore, Nairobi is an Ideal for the study of tropospheric Ozone for an urban city in a developing country.The understanding of the surface ozone over the city of Nairobi will greatly reduce the negative effects on human health and the general City of environment. The research was carried out with a view to assisting regulatory agencies optimize future monitoring networks, and to help identify locations and time where human health and natural resources could be at risk, now and in the future. Although the research was limited to the City of Nairobi, the results are valuable for improving the conceptual

understanding of formation of high ozone concentrations in a more general sense in the Kenya and East African region.

1.6 Scope of the study

For the purposes of this study The City of Nairobi refers to Nairobi County which constitutes the present Nairobi County boundaries. The study investigated the spatial variation of surface Ozone, temporal surface ozone distribution and the source regions of surface Ozone observed over the City of Nairobi.

CHAPTER TWO

2.0 Literature Review

2.1   Introduction

Ozone (O_3) gas exists at various heights throughout the atmosphere. Studies (Parrish*et al*., 2009; Sitch*et al*., 2007; Vingarzan, 2004) observe that tropospheric O_3 concentration is increasing at a high rate; which is attributed to increase in anthropogenic precursor emissions. According toIntergovernmental Panel on Climate Change(IPCC2013) stratospheric ozone is key in climate studies since it influences the climate in terms of climate feedback and subsequent temperature modulation both within the atmosphere and at the earth's surface. The following sections highlight some of literature on source of ozone in the tropics, trends of tropospheric ozone, its impacts and transportation.

2.2 Source tropospheric ozone

Tropospheric ozone (O_3) is a short-lived trace gas that is formed in situ by reactive gases and solar radiation or that is transported from the stratosphere. In the tropics, tropospheric ozone sources due to stratospheric intrusion is excluded (Homeyer et al. 2011) by the nature of the Brewer Dobson circulation. However, stratospheric air can also influence ozone levels at high altitudes by lofting air masses over the Himalayas (Dupont *et al.* 2012).

Ground-based O_3 is created by the photo-chemical and chemical reactions between oxides of carbon (CO), nitrogen (NO_x), and volatile organic compounds (VOCs) (e.g. Finlayson-Pitts and Pitts 1997, Reid *et al.* 2005, Lamarque *et al.* 2010, Sahu 2012) wherein the cellulosic material and aromatic compounds from open biomass burning are one of the causes of the production. Anthropogenic activities, such as biomass burning are one of the main sources of ozone. Tropospheric ozone is also an important greenhouse gas. Its ambient concentration has increased due to increases in precursor

trace gas emissions (e.g. CO, NO_x, VOCs) rom anthropo- genic activities. Differences in tropospheric ozone burden have been observed in numerous emissions data mainly due to biomass burning (Lamarque et al 2010, Cionni *et al.* 2011, Young *et al.* 2013). Jaffe and Wigder (2012) presented a critical review of O_3 production from wild fires focusing on its influence on O_3 production. According to this study, wild fires produced 170 Tg per year of O3(3.5%) globally. The majority of O_3 production was high in tropical and equatorial biomes and lower in the other regions.

Tropospheric ozone acts both to control the oxidizing capacity of the atmosphere and as a greenhouse gas (Lin *et al,* 2012a,2012b; Miller, 2014). Most of the oxidation of long-lived gases by hydroxyl radicals (OH) takes place in the tropics, where high UV and humidity promote the formation of OH from the photolysis of ozone (Logan et al., 1985; Thompson, 1996).

O3 is produced within the troposphere by the photochemical oxidation of hydrocarbons, methane (CH) and carbon monoxide (CO) in the presence of nitrogen oxides (NO) (Crutzen, 1988; Fishman et al., 1991) A rapid formation from reactive hydrocarbons (urban pollution, biomass burning) close to their source, followed by its mixing within the troposphere, and a slow and delayed formation (2 or 3 weeks) from less reactive precursors, such as CO and CH4, during or after their redistribution in the troposphere by horizontal and vertical atmospheric motions (Logan, 1985). A very slow process of production in the uppermost troposphere which results from background tropospheric carbon species and nitrogen oxides. The latter process can be accelerated by lightning, NO as the dry season gives way to thunderstorms of the wet season (Edwards et al., 2003; Chatfield et al., 2004). While this appears to be a non-pollution process, the organic radical producers may indeed be increased by pollution.

A second source of tropospheric O3 is transport from the stratosphere. In the lower troposphere the major source of O3is in situ production whereas in the upper troposphere

transept of stratospheric O3 is more important. The O3 production is balanced by reactions in which O3 is destroyed, as shown in the series of equations represented by equation 1 below. In the stratosphere, a catalytic O3destruction chain plays the most important role (Bates and Nicolet, 1950).

$X + O_3 \longrightarrow XO + O_2$

$XO + O \longrightarrow X + O_2$

net: $O + O \longrightarrow O_2 + O_2$ Eq. (1)
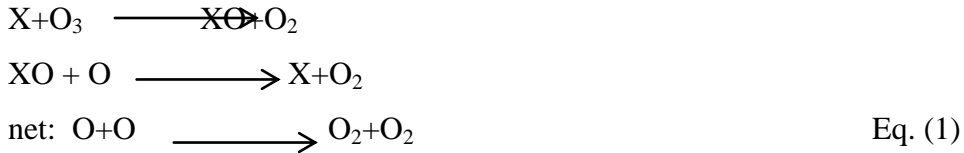
Where, X is the catalyst species. The species X is not destroyed in this reaction, so only small amount of X is needed to destroy a large amount of O_3. Several species have been suggested for the catalytic X in the atmosphere. The most important of these in the natural stratosphere are Hydrogen (H), Hydroxyl radical (OH), Nitric Oxide (NO), and Chlorine (Cl), (Wayne, 1991).

However, a catalytic reaction chain can lead to the production of tropospheric O_3. The last two steps of this reaction chain are shown in equations 2 and 3 below:

$NO_2 + hv \longrightarrow NO + O$ Eq. (2)

$O_2 + O \longrightarrow O_3$ Eq. (3)

Here, the photo-dissociation of NO_2, provides an oxygen atom which in the next step reacts with molecular oxygen to produce O_3. NO and NO_2 are designated as NO_x or oxides of nitrogen. These are the precursors of surface (tropospheric) ozone. The presence of CO, CH_4 and other hydrocarbons allows the recycling of NO to NO_2.

2.3   Trend of Troposphere Ozone and Impacts

With the increased consumption of energy resources, particularly fossil fuel and bio-fuel, over the globe, there has been considerable increase in the emission of surface ozone

precursor gases with the passage of time. Volz and Kley (1988), have pointed out that, in the northern hemisphere, the tropospheric O_3 concentration has more than doubled over the past century, which is attributed basically to the anthropogenic emissions of O_3 precursor gases (Kelly and Scar, 1982; Crutzen 1988).

There is concern that the concentration of ozone in the troposphere may have been increasing in recent times. During the last decade, an increase of approximately 10% (1% per year) in ozone throughout the height of the atmosphere has been demonstrated over Europe (WMO, 2014). Ozone measurements made in rural sites of western US indicate 1 - 2% annual increase in average concentrations over the past 30 years (Jaffe, 2007). There is equally significant trends being reported, which consistently show increases of about 1% per year over the past decade or two at northern mid-latitudes (Logan, 1985; Oltmans and Levy, 1994; Bojkov, 1988).

In Africa as a whole, the forest and the savannas occupy areas of 2x 106and 1x 107 km2, respectively. The annually burnt dry matter biomass which is primary caused by human activities, is estimated at 1.3x 108tons yr-1(Delmas et al., 1999). Biomass burning has long been recognized as a significant source of reactive species such as CO, hydrocarbons and NOx, which play an important role in the chemistry and the radiative budget of the troposphere and they are precursor gases for ozone (Crutzen et al., 1998; Crutzen and Zimmermann,1991), therefore increasing the tropospheric ozone.

With the advent of industrialization and increased use of coal and petroleum products, emissions of Volatile Organic Compounds (VOCs) resulting from fossil fuel combustion has resulted in increased photochemical smog levels in most of urban areas of the world. These events have produced a situation in which large numbers of people are exposed to high levels of ozone resulting in increased risk of both acute (short-term) and chronic (long-term) health effects (National Research Council, 1991).

As human exposure to ozone has increased, there has also been an increase in exposure and damage to agricultural crops, forests and ecosystems, as well as to materials such as rubber, paints and dyes. The USA's National Acid Precipitation Assessment Program (NAPAP) found that air pollutants could be ranked in the following order of importance, in terms of their potential to negatively impact the growth, yield or quality of agricultural crops: $O_3 > SO_2 >$ acidic deposition $> NO_2$ (Shriner et al., 1990). More specifically, ozone was found to be a significant stress factor in agricultural production. This shows that surface ozone must be always monitored to avoid negative impact on agricultural productivity.

It is well documented that temperature has direct impact on the O_3 production rate (Vukovich, 1995; Sillman and Samson, 1995). Basically, higher temperatures increase the emission rate of surface ozone precursors (Steiner et al., 2010) which in turn accelerate the photochemical ozone production in the lower troposphere. However, the temperature itself may not always be the direct cause for the increase in O_3 production. It may be a surrogate for other meteorological effects such as relative humidity, cloud coverage, lightning, atmospheric stability, stagnation, wind speed and solar intensity.

There have been a few observational and model studies on the surface ozone in the recent past to understand relative importance of its various sources and sinks mechanisms over the East African region and to delineate its spatial-temporal distribution over the region. However, in other regions, studies of this kind investigating the impact of various meteorological and chemical parameters on the production of surface ozone have been done (e.g. Vukovich, 1995; Rasmussen et al., 2012; Steiner et al., 2010; Jacobson, 1999)., Therefore, there is need for similar studies over the East Africa (For instance, trend, diurnal and seasonal analyses) and the role of meteorological and chemical parameters in the photochemistry of surface ozone.

2.4 Transportation of Ozone

According to the studies by Ongoma *et al.* (2013), Ongoma *et al.* (2014) and Opija *et al.* (2007), the predominant wind direction over the City of Nairobi is generally easterly, with the direction ranging from north easterly in December January February (DJF) to south easterly in June July August (JJA), while the wind speed ranges from 4 - 11 Knots, However, urbanization of the City of Nairobi is likely to reduce wind speed over the city hence a threat to air quality (Shepherd 2005).

The lifetime of ozone is an important factor when considering its transport. The abundance of water vapor and solar radiation in the tropical boundary layer shortens the lifetime of ozone to only 2 - 5 days (Fishman *et al.,* 1991). Thus, ozone in the lowest levels will not survive long enough to be transported long distances (James*et al*., 2003). The lifetimes in the middle troposphere are much longer, approximately 90 days. This means that ozone can travel much farther if it rises out of the boundary layer to where its lifetime is longer and winds are stronger. Vertical motion on the convective scale is a mechanism by which burning by-products are lifted to higher levels where they may contribute significantly to enhanced ozone production in the upper troposphere (Chatfield and Delany, 1998).

The vertical transport is controlled by the stability structure of the atmosphere and horizontal transport by the local topographic winds near the surface or by large scale circulation in changing synoptic flow fields (Tyson *et al*., 1996; Sillman, 1999).

The understanding of the ways in which ozone is formed, accumulates and moves through space and time is very important especially in the tropics, where relatively less research has been done due to scarcity of data (Kirkman*et al*., 2000). Ozone pollution may not entirely be confined to the anthropogenic source region but can spread to a large geographical extent due to different scales of transportation of pollution plumes from the source region. Its impact has previously been reported to reach a regional and continental scale in Asia, Europe and North America (Parrish *et al*., 1993). The most critical aspect

of the tropospheric ozone problem is its formation downwind of large urban and sub-urban areas, where under certain meteorological conditions, emissions of NO and VOCs can result in ozone concentrations greater than100 ppbv.

CHAPTER THREE

3.0 Methodology

3.1   Study area

The study was done in Nairobi (Figure 3.1), the capital city of Kenya which lies some 200 km south of the Equator and is located at approximately 1°9'S, 1°28'S and 36°4'E, 37°10'E. It occupies an area of about 696km^{2}and the altitude varies between 1,600 and 1,850 meters Above Mean Sea Level (AMSL) (CBS, 2001). The western part of Nairobi is on high ground (approximately 1700–1800 AMSL)with rugged topography and the eastern side is generally low (approximately 1600 AMSL) and flat (Saggerson 1991). Key physical features include the Nairobi National Park, Ngong hills and Mathare River and the indigenous Karura forest in the north side.

3.2. Climate of the study area

Nairobi experiences a moderate climate even though it is located along the Equator. At a height of about 1795 m AMSL, the city has a subtropical high climate according to the Köppen climate classification (Peel *et al.,* 2007), as opposed to the expected tropical climate. The regional climate of East Africa is influenced by the seasonal displacement of the Inter-tropical Convergence Zone (ITCZ). The north–south–north annual shift of the ITCZ is driven by the apparent annual motion of the sun and results in monsoon trade winds, which give rise to well-defined wet and dry seasons (Camberlin and Philippon, 2002). Severe changes in this cycle usually result in droughts or flooding in the East African region (WMO, 2013). The average annual rainfall is 900 mm but varies from less than 500 mm to more than 1500 mm between years. The average daily temperature varies from 17° C in July and August to 20 $^\circ$C in March (KMD, 2013). The winds are generally easterlies and range between 4 -11 knots (Ongoma, 2014)
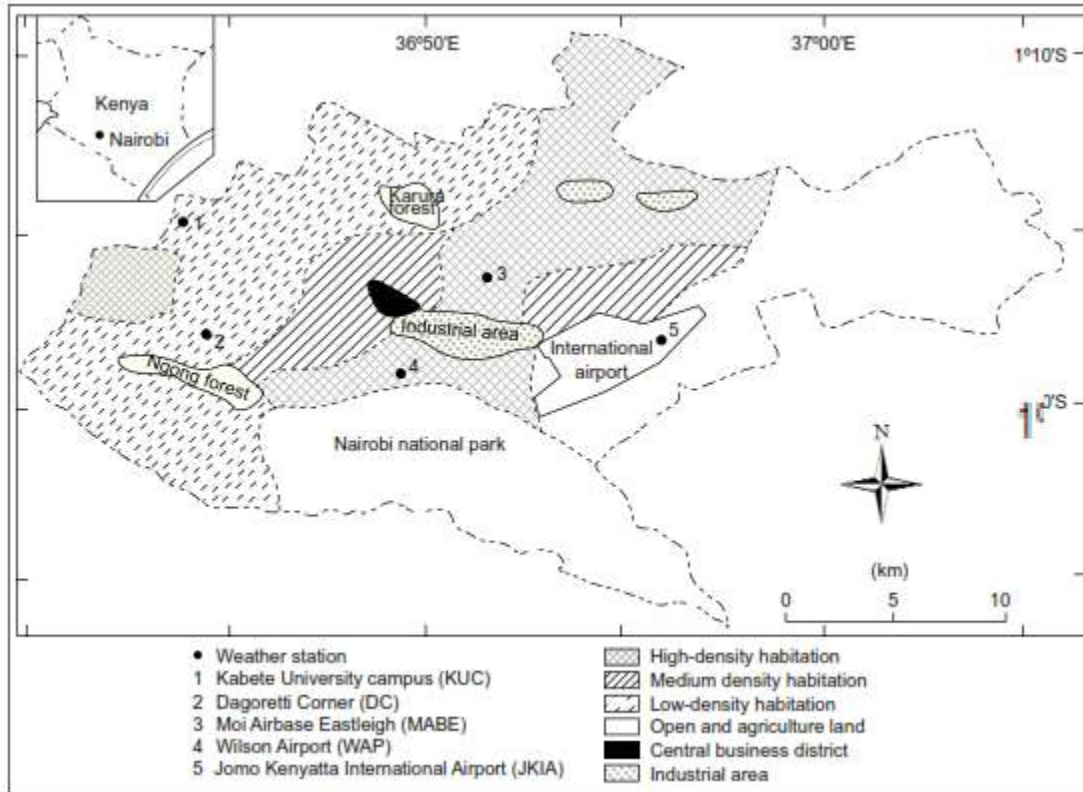
Figure 3.1: Map of Nairobi and its environs showing the study area. (Source: Makokha and Shisanya, 2010)

3.3 Demographics of the City of Nairobi

Like many other cities in developing countries, Nairobi has experienced very rapid population growth in the last 30-40 years. At a population growth rate of 4.7-4.8% annually, the population of Nairobi grew from about 0.8 million in 1979, to 2.1 million in 1999 and 3.2 million in 2009, with a daytime population of 4 million, Kenya National Bureau of Statistics KNBS, (2009).This is indeed a very high rate of population growth rate compared to an average of 3.4% annually for cities in developing countries and 1.8% for the world urban growth rate.

This increase in population has resulted in an increase in commercial, motor vehicle and industrial activities; especially within the Central Business District (CBD). Nairobi is

registering rapid growth in motor vehicle population and ownership. The country had about 600, 000 vehicle units in 2000, 950,000 in 2008 but this has risen to about 1.2 million in 2010/11(KNBS, 2012) The peri-urban areas of the city continue to experience both controlled and uncontrolled development in buildings and infrastructure, especially mushrooming of informal settlements (Karanja and Makau, 1998) In these settlements, solid waste is usually burned in open air and the majority of the households use kerosene and biomass-based fuels (charcoal, wood and vegetation) for domestic cooking (Egondi*et al.,* 2013). The city's industrial area is located to the East and Southeast and is dominated by small and medium-sized industries, which include food processing, power generation, chemical processing industries, battery manufacturing and scrap metal recycling.

The city's commercial hub, the CBD, has been growing, and currently, the main CBD has extended to Westland's, Upper hill and Ngara areas. Other commercial centers are situated at Eastleigh, Eastland's, Buruburu, Kayole, Karen, Dagoretti Corner, Kawangware and Kangemi. The city however, is faced with physicaldevelopment, health and environmental management challenges for both the old and new commercial developments and services, which are inadequate for the fast-increasing population (MoNMD, 2008).

The city's area used for industrial purposes has grown phenomenally in real terms, but has decreased slightly in relative terms. Industrial use entails both heavy manufacturing as well as light industries such as warehouses, workshops, "jua kali" workshops, garages and stores. Since the 1970s, the industrial area has expanded to Dandora, Kariobangi, off Mombasa Road up to North Airport road and off Outer Ring road. Some of these extensions have been uncoordinated, leading to incompatible mixed land uses that have merged with or encroached into residential use.

The City of Nairobi is affected by high level of air pollution and environmental degradation caused by the poor transport system. Available statistics show that air

pollution in Nairobi is high with mean daytime concentrations of fine particles ranging from 10.7µg/m^3 at the rural edge of the city to 98.1µg/m^3 on a sidewalk in the CBD (Shilenje, 2014). This implies that there are very high levels of suspended particulate matter within the inhalable range. Air quality studies indicates that there is a strong correlation between fine particulates and motor vehicles (r = 0.93), indicating that vehicular exhaust is the main source of fine particles in the air (Odhiambo*et al*. 2010). This has led to the deterioration of the quality of urban health and environment. Indeed, most of the leading disease complaints in Nairobi relate to vehicle exhaust fumes which are source of ozone precursor gases, therefore, there is need to monitor and develop policies that will likely reduce exposure to surface ozone.

3.4   Data collection

In this study, data was collected from City of Nairobi in collaboration with Kenya Meteorological Department (KMD) and the sampling was done in four different sites. The sites were designed to be at busy intersections or roundabouts (Ngong road, Landhies road and Pangani Roundabout) and a site in Industrial Area (Table.3.1). The selection was done after a feasibility assessment that mapped out busy and potential monitoring sites considering witnessed traffic jams, possible elevated emissions and assessing the general prevailing meteorological conditions that influences pollution levels. The sampling was done in the month of December 2015 for about ten days, 24 hours a day in each site. The data was collected every one minute using EcotechSerinus 10 gas analyzer for Ozone from Kenya Meteorological Department (KMD) and the meteorological parameters were measured by an automated weather observing stations.

Observed ozone data at KMD headquarter was also used, whichincluded; daily total column Ozone, measured by Dobson Spectrophotometer for a period of five years from 2011 to 2014. These were the years with complete data; weekly vertical Ozone profiles measured by Ozonesonde flights for the same period and Meteorological data (rainfall and temperature) forDagorettistation forthe five years period.

In addition, back trajectories of air masses arriving at 100m above the sampling sites were generated using the HYSPLIT_4 model (Draxler and Hess, 2010). Daily and back trajectories were run for 180h, clustered on a monthly basis and compared with monthly averages of measured hourly wind speeds and directions. To further analyze the seasonal zonal wind speed and direction, wind data was obtained from NCEP Reanalysis data provided by the NOAA/OAR/ESRL PSD, Boulder, Colorado, USA, from their Web site at https://www.esrl.noaa.gov/psd/data/gridded/data.ncep.reanalysis.pressure.html for further information on the data can be obtained from Kalnay *et al.* (1996).

Table 3.1: Location of the monitoring sites

Road	Site	Latitude	Longitude	Elevation (M)
Ngong Road	Nakumatt Junction	S. 01°17. 958'	E. 036° 45. 649'	1790
Landhies Road	Muthurwa Market near Machakos bus stage	S. 01°17. 210'	E. 036° 501'	1673
Juja road	Pangani round about	S. 01°16. 238'	E. 036° 50. 187'	1637
Industrial Area	Met Station (TX)	S. 01°18'	E. 036° 36'	1626

3.4 Data analysis

In analyzing the collected data, the following methods were applied so as to meet the objectives of the study: time series analysis was used to analyze the temporal variation of

surface Ozone, GraDS was used to analyzezonal wind and trajectories, and finally HYSPLIT model was used to determine the source region.

3.4.1 Objective One: Investigate the spatial variation of Ozone in Nairobi.

Data collected from the foursample point was analyzed, to determine the spatial distribution of surface Ozone over Nairobi. The four stations represented different locations with different intensity of human activities.

3.4.2Objective Two: Investigate the temporal Surface Ozone over Nairobi city

The time series analysis involved a graphical plot of the element which is of interest against time. The graphical representation shows the spread of the data over time. Any gaps of missing data become apparent; unless a smoothing function is applied. Seasonal cycles may also be deduced in the plotted data.The Ozone data from KMD was subjected to time series analysis and as a result seasonal, monthly and diurnal graphs were reanalyzed.

3.4.3   Objective Three: Identify the source regions of surface Ozone

A trajectory is a calculated transport pathway of an infinite small air parcel. Trajectories provide a useful tool for understanding the three dimensional transport of airborne material in the atmosphere (D'Abreton and Tyson, 1996). Backward trajectories are particularly useful in tracing aerosols and trace gases back in time and space to ascertain their origins. On the other hand, forward trajectories provide a guide on the locations where pollutants are likely to be deposited. Thus, trajectories corresponding to individual transport events provide an indication of the mean motion of an advected air parcel.

Different models for air trajectories can be used depending on the availability of input data. Each model requires gridded fields of meteorological variables at regular temporal intervals, and for this research HYSPLIT ((Hybrid Single-Particle Lagrangian Integrated Trajectory) Model was used from National Centre for Environmental Prediction -

National Centre for Atmospheric Research. The HYSPLIT model is a complete system for computing single air parcel trajectories to complex dispersion and deposition simulations. The HYSPLIT model computes the advection of a single pollutant particle, or simply its trajectory.The model was run interactively on the Web through the READY system on NOAA's site http://ready.arl.noaa.gov/. (Stein et al. 2015.

The National Center for Environmental Prediction (NCEP, $1^{o}{\times}1^{o}$) reanalysis meteorological data were inputted for model calculation. The vertical motion method in the calculations is the default model selection; it uses the meteorological model's vertical velocity fields and is terrain following. The height of the endpoint is set at 500mabove ground level. The three-day backward trajectories were calculated four times (00:00, 06:00, 12:00, and 18:00 UTC) per day. After calculation, the trajectories were clustered into several types using the HYSPLITsoftware.

The data sets used are the monthly reanalysis fields available from the National Centre for Environmental Prediction- National Centre for Atmospheric Research (NCEP-NCAR) global re-analyses. The start or end of the trajectories was determined from the time series plots of the surface ozone concentrations for Nairobi. A sudden sustained rise or fall in the ozone concentration could be indicative of changed air source characteristics or path. For a rise or fall in the ozone concentration, the timing is supposed to coincide with the start (of a backwards trajectory to indicate the path and source region of airflow. Forward air trajectories merely indicate the outflow from the station and in this case will be Nairobi.

CHAPTER FOUR

4.0 Results and Discussion

4.1   Trend analysis for tropospheric ozone columns

The Tropospheric Ozone Columns (TOCs) measured by ozonesonde at the KMD station was averaged for the period of 2011to 2014. The ozonesonde profiles were integrated from the ground to the 200 hPa pressure level and then the monthly mean were calculated: there were between 2 to 6ozonesonde measurement ascents each month as shown in Table 4.1. The monthly average values of the TOC is also shown in Table 4.2

Table 4.1: Monthly ascents

Months	2011	2012	2013	2014
Jan	4	4	5	3
Feb	4	5	4	4
Mar	5	4	4	4
Apr	4	4	4	5
May	2	4	5	3
Jun	4	5	5	4
Jul	4	4	5	5
Aug	5	6	3	4
Sep	3	5	4	4
Oct	4	4	5	5
Nov	5	4	6	3
Dec	4	4	2	5
Total	48	53	42	46

Ozonesonde ascents were done on Wednesdays of every week at KMD resulting to the number of ascent as shown in the table 4.1, which is different from one month to another.

Table 4.2: Monthly averaged ozonesonde measured data, based on three years of data (2011-2014)

	Jan	Feb	Mar	Apr	May	Jun	Jul	Aug	Sep	Oct	Nov	Dec	Annual mean	Std.dev	COV %
Ozone (ppb)	17.2	17.9	17.3	16.3	16.9	19.6	22.9	21.5	22.5	19.4	17.6	16.3	18.78	2.37	12.64

Table 4.3 Seasonal variation of Ozone

	MAM	JJAS	OND	JF
Mean Ozone (PPb)	16.83	21.63	17.77	17.55

Table 4.2, and 4.3, shows the mean monthly variation of surface ozone over Nairobi. It shows that, the ozone was lowest in the months of March to May (MAM) and November to January. Also the standard deviation and coefficient of variation is quite low (12.64%), which means that, the monthly variability is quite low. The seasonal variation in the surface ozone column is explained by the migration of the ITCZ over Kenya, when it passes twice a year, between March-May and October-December (Black *et al.,* 2003) which are the wet seasons over the study area. In both wet seasons, the tropospheric ozone columns are fairly low, due to convection of humid and ozone poor air. The increase in the tropospheric ozone values during the cold season (July-September) over Nairobi can be explained by incursion of ozone from the Southern Africa especially from Zambia which has high biomass burning during thisseason (Bundi, 2004).

4.2 Trend analysis for the surface Ozone

Figure 4.1 depicts the diurnal variation of ozone from the four sampling sites. It generally shows low amount in the early morning and at night with high amount of ozone during the day. This can be due to the presence of precursor gases that are produced during the morning traffic hours which in presence of electromagnetic energy from the sun react in the atmosphere leading to ozone formation. At the Industrial area site, the pick of surface ozone, is earlier as compared to other site, this because of the availability of the precursor gases from the night industrial activity, while the other sites which their source is mainly from motor vehicle traffic, and during the night most of these roads have low or no traffic. From the results vehicular flow in Landhies were lower compared to Nakumatt and Pangani at the time of data collection.

The 8 hour mean (8 to 17 hrs) of surface ozone at the four sites show that, Industrial area site relatively high mean ozone concentration (20.2 ppb) compared to other sites that had 10 ppb and less concentration (Table 4.4). This may mean the precursor gases at the Industrial area site are higher than in other sites considered in the study. This may be attributed to emissions from industrial activity in industrial area, while in other sites the emissions are mainly from motor vehicle traffic.The Landhies road had lower mean surface ozone (7.8 ppb), this might be due to the contribution from domestic combustion from the residential areas around Pangani and Nakumatt which contributed to higher precursor gases.in the two sites than the Landhies road.
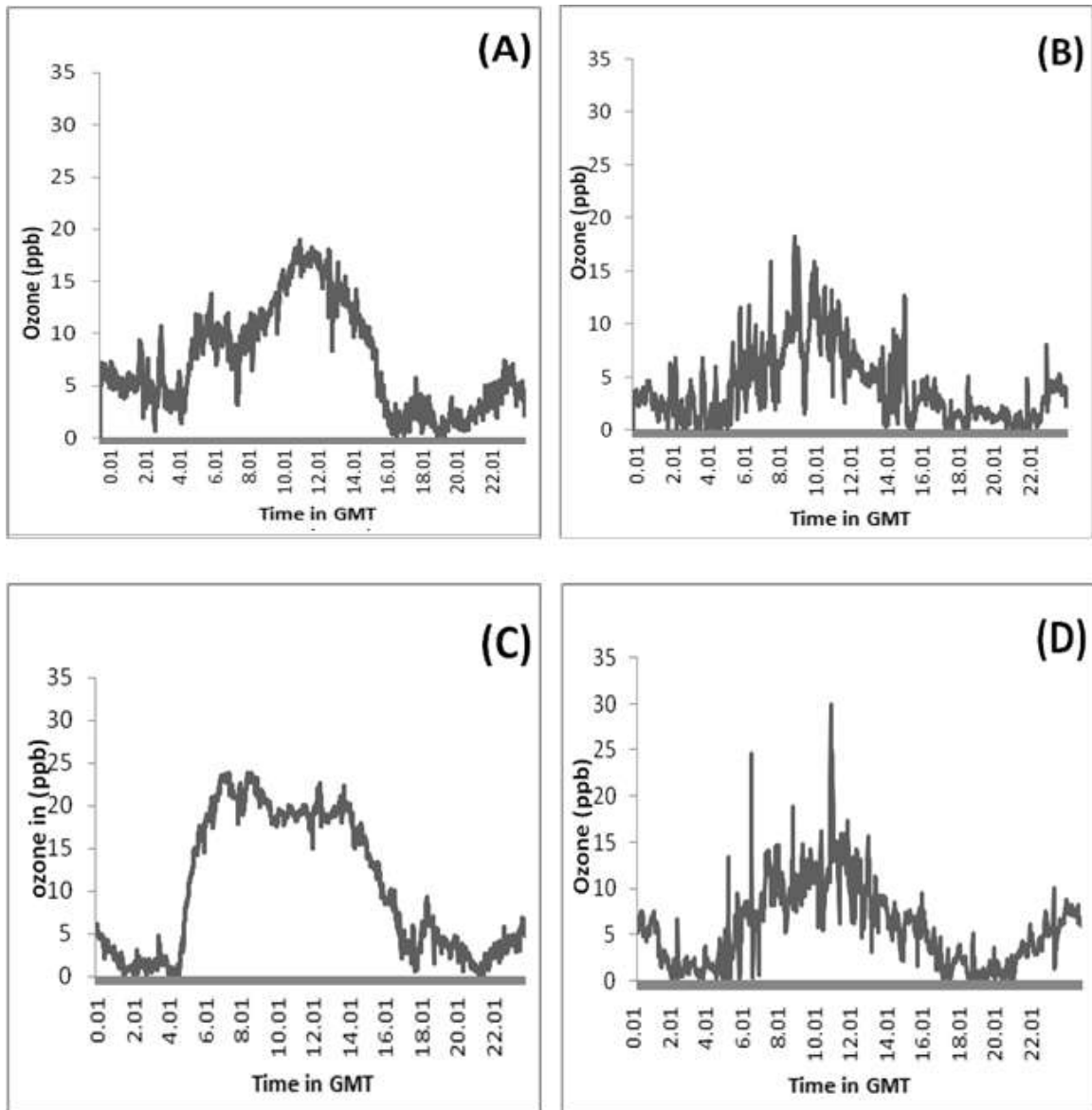
Figure 4.1: Diurnal variation of Ozone concentration at; (A) Nakumatt Junction, (B) Landhies, (C) Industrial Area and (D) Pangani.

Table 4.4: Ozone 8 hour mean values

| Site	Ozone 8 Hour Mean (ppb)
Nakumatt Junction	10.3
Landhies	7.8
Pangani	10.1
Industrial Area	20.2

4.3 Analysis of Vertical profile of Ozone

Figure 4.2 shows temperature and ozone vertical profile. The analysis, (Figure 4.2a), shows a decreasing temperature trend within the troposphere up to 16.3km (110 mb) height where it becomes constant up to 17.6 km (88 mb) height.

The average flight data is in agreement with observations made by Thompson *et al.,*2011. On the average level of tropical tropopause being located at close to16.3 km level with a height ranging from 12 - 19 km. The observed layer between 16.3 km level and 17.6 km level represents the tropopause. Although Pidwirny (2006) found out that the average depth of the troposphere is approximately 11km within the tropics, this study reveals, on average, over Nairobi it extends to about 16 km. Figure 4.2a further shows temperature increases within the lower stratosphere.

The ozone concentration showed small values, with negative trend upwards within the troposphere, up until the tropopause (Fig. 4.2b). Within the tropopause, there was a gentle increase in the ozone concentration and a sharp increase in the lower stratosphere,

peaking in the mid stratosphere. The maximum ozone concentration value of 13.04ppb is found at a pressure of about 27km. The ozone layer, with the highest concentration of ozone, is within about 23 – 30 km. The ozone layer is therefore, about 23km and covers a depth of about 7km, located in the mid stratosphere over Nairobi. The ozone concentration measured during this study are in agreement with Sauvage *et al.,* (2005)
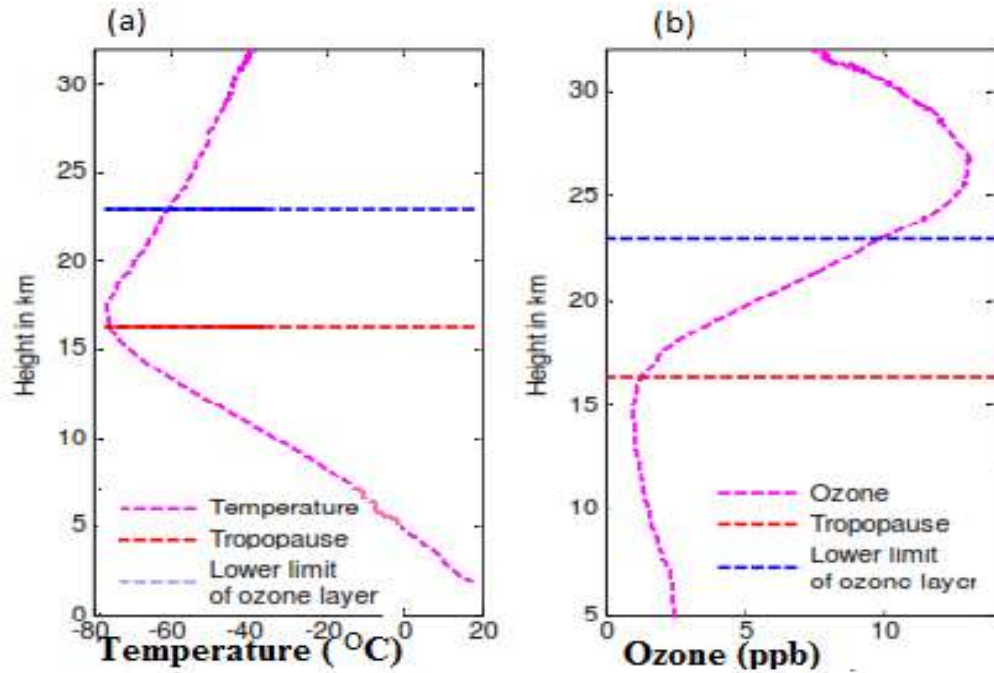
Figure 4.2: Mean Vertical variation; (a) temperature, (b) Ozone over Nairobi 2010 to 2014

The ozone mean concentration values were decomposed into 3 months interval to correspond with different weather seasons over Kenya and Nairobi in particular and are presented in Figure 4.3.
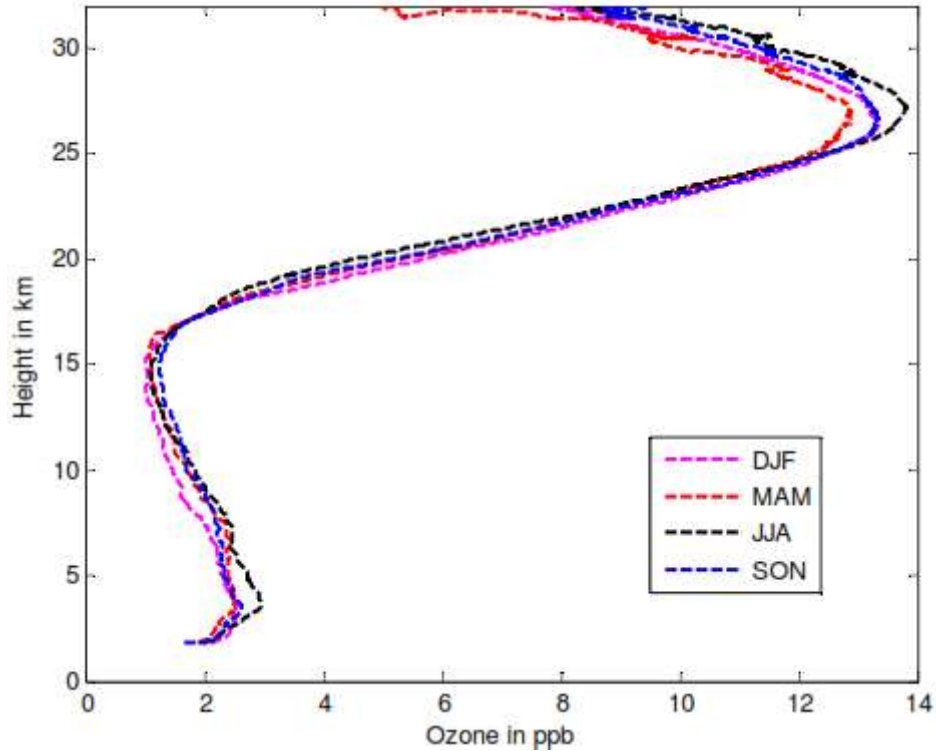
Figure 4.3: Mean Vertical seasonal variation of Ozone over Nairobi, 2011 to 2014

Generally, the June, July and August (JJA) season experienced the highest ozone concentration in both the low and upper levels as compared to the other seasons. This is partly in agreement with Pandey*et al.,* (1992); who reported that there exist a significant positive correlation between O_3 concentrations, solar radiation and temperature. However, there was an exception at the tropopause where higher values of ozone are reported during September, October, November(SON) months.

4.4 Analysis of Wind system over Nairobi

The wind analysis is to help find the source region of the surface ozone in the City of Nairobi and it distribution, it also show the circulation of wind, which affect the ozone concentration in the City of Nairobi.

Figure 4.4 shows the mean zonal wind distribution over Nairobi. Easterlies are predominant in the lower troposphere, up to about 500mb, westerlies in the mid troposphere and again, easterlies in upper troposphere, extending into the lower stratosphere, commonly known 'steering winds' in this region.
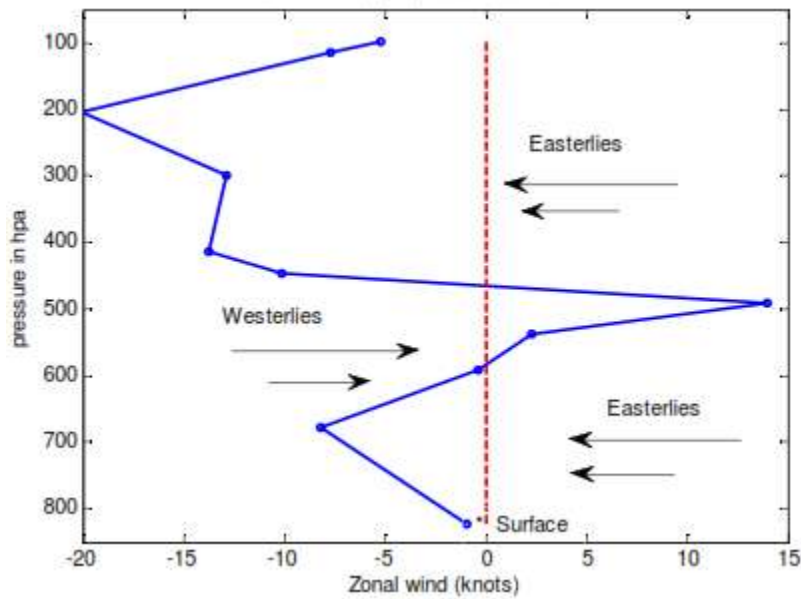
Figure 4.4: Profile of Zonal wind Component for Nairobi, 2011 to 2014

This is because of their effect of 'steering' or advecting weather systems such as convective cells or moisture in and out of the region of East Africa. The observed predominant easterlies in the lower troposphere were observed by other studies (Ongoma*et al.,* 2013; 2014).

Ozone is depleted by dry and wet deposition on the ground and long-term transport. Figure 4.5 displays mean seasonal wind speed, direction at 850mb pressure level (boundary layer) over East Africa. Meteorological conditions at pressure level 850mb are used to consider lower atmospheric conditions rather than surface conditions normally 2-10m above the ground.

During the March April May (MAM) period, there is predominantly easterly flow (Fig. 4.5b) from Indian Ocean contributing to high moisture content over East Africa. High moisture content implies presence of a humid atmosphere that is negatively correlated with ozone; similar observation was made by other studies (Ahamad *et al.,* 2014; Wang *et al.,* 2013). The winds penetrate into Nairobi during MAM, JJA and DJF but the wind vectors reverses to dry north easterly flow during DJF.
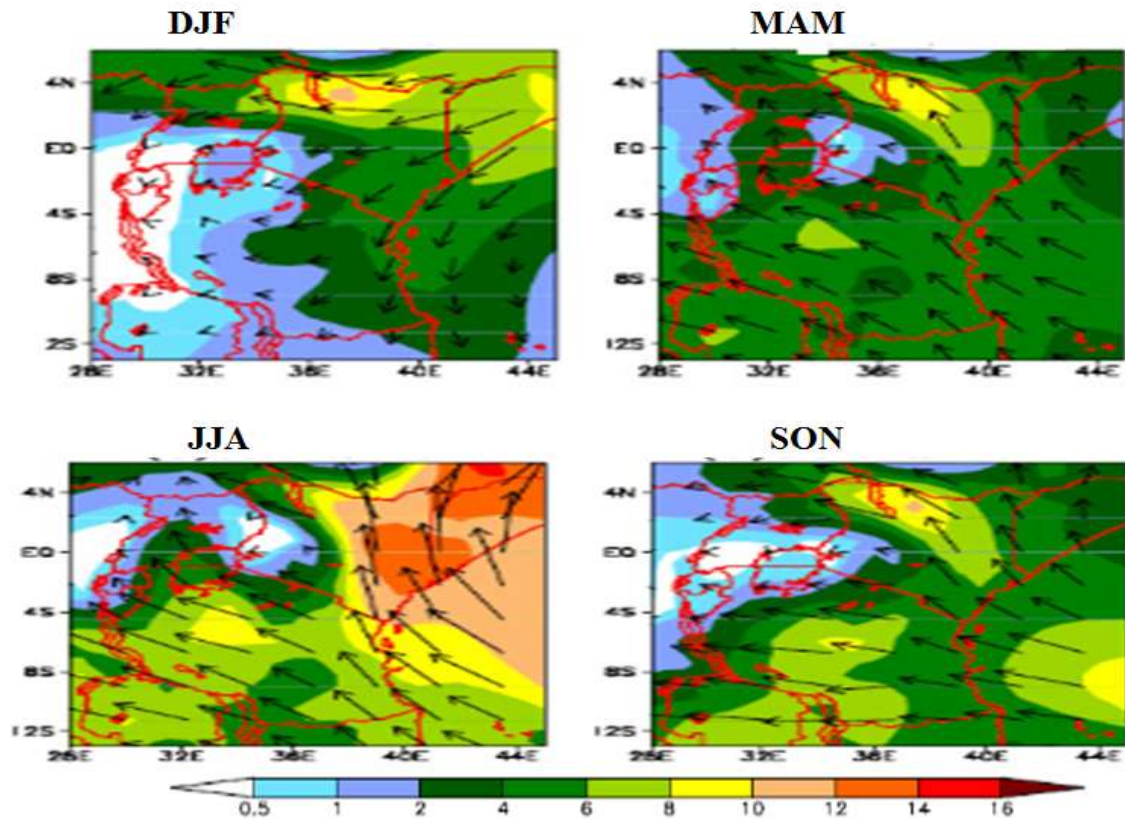
Figure 4.5: The mean seasonal wind speed (m/s) and direction (a) December, January and February (DJF) (b) March, April and May (MAM) (c) June, July and August (JJA) (d) September, October and November (SON)

A comparison of wind speeds during different seasons shows that weak north-easterlies are observed during DJF as compared to other seasons that generally experience strong

easterlies (Figure 4.5a). The observed high concentration of ozone especially in the upper levels during DJF (Figure 4.5) can partly be explained by the observed weak winds. This is supported by observations made earlier by Odhiambo *et al.,* (2010) in a study on motor vehicles air pollution in Nairobi.

4.5 Analysis of Source region for TOCs

The origin of ozone in Nairobi was analyzed using HYSPLIT backward trajectories. It was run in different months when there is high and low an ozone concentration. Each HYSPLIT backward trajectory was calculated for 180 hours with Global Data Assimilation system (GDAS) meteorological datasets. A starting time of 00:00 UTC was set to simulate the trajectories from the selected starting location (Nairobi). Figures 4.6 (a-d) demonstrate the typical trajectory of the backward trajectory of Nairobi for the months of January, May, July and November, respectively. Therefore, the source region of Nairobi tropospheric ozone will definitely be defined by the source of wind system. The transport corridors or plumes arriving over Kenya are clearly bounded and well defined (Figure 4.6).

North-easterly and south-easterly transport from the north-western and south-western Indian Ocean corresponds to their seasonal monsoon airflow counterparts. Anti-cyclonicSahara transport is a prominent feature and westerly transport mode is evident (Gatebe*et al.,* 1999). Through backtrack trajectory (Figure 4.6); the source of the ozone during the July to September is from the Southern Africa countries. This finding concurs with Thompson *et al.*, (2002) and Bundi, (2004) who found out that Lusaka had the highest concentration of tropospheric ozone at 100ppbv at $2 - 5$ km stable layer. This was due to biomass burning in Zambia during this period.
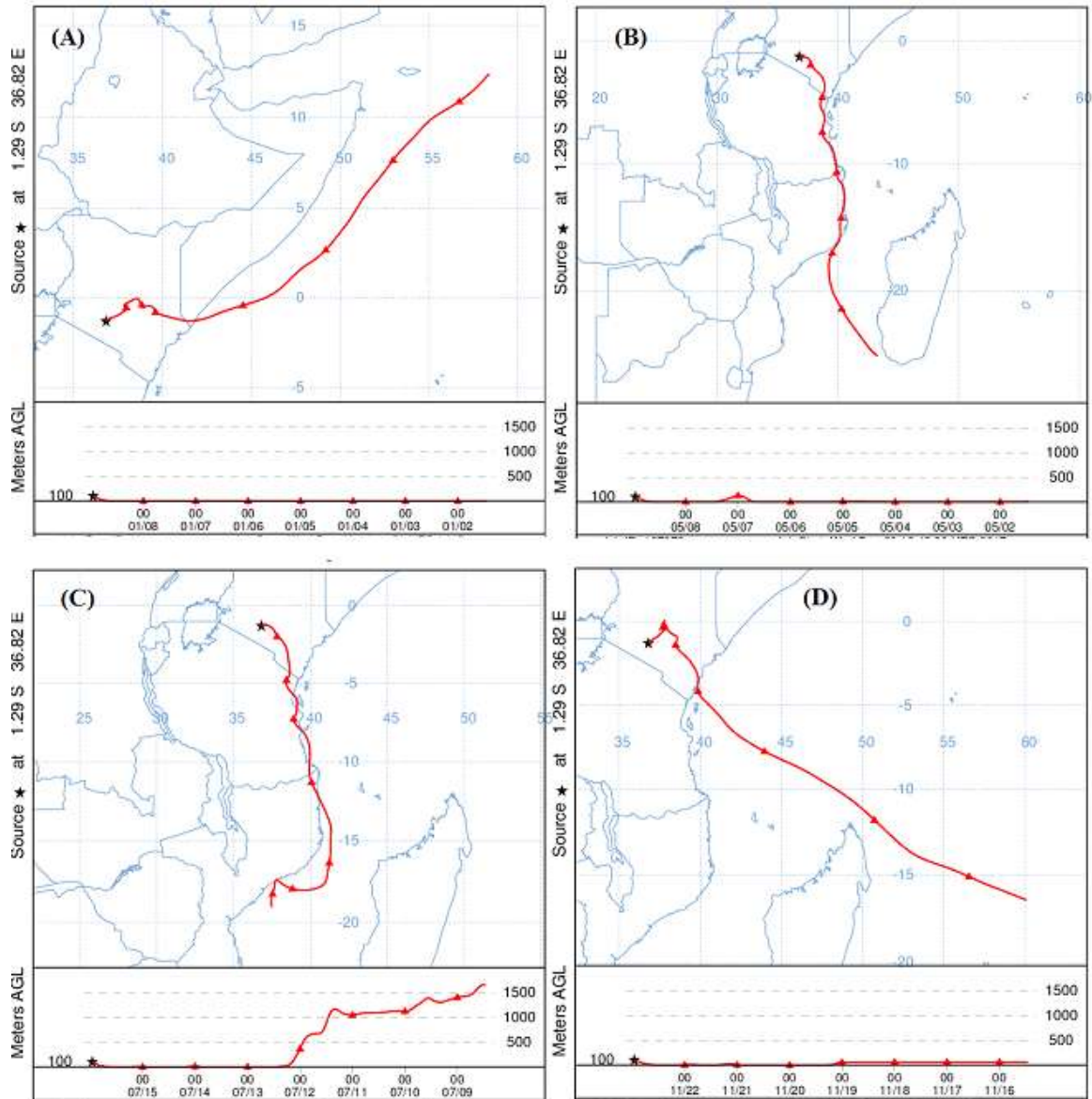
Figure 4.6: Mean backward trajectories ending at Nairobi. The ending point height is 100m for (A) January (B) May (C) July (D) November

CHAPTER FIVE

5.0 Conclusion and Recommendation

5.1 Conclusion

The main objective of this study was to assess spatial and temporal variation and identify the source regions of surface Ozone over Nairobi city. To achieve it, the following specific objectives were formulated to investigate: the spatial variation of surface ozone, the temporal variation and the find out the source region of the observed surface ozone. On the investigation of the spatial surface ozone, sampling was done at four sites within Nairobi for a period of 10 days, it was found out that the diurnal variation of Ozone in Nakumatt Junction, Landhies road and Pangani Roundabout showed low amount in the early morning and at night with the peak being realized during the day. This was due to the fact that surface ozone is produced by photochemical oxidation of CO, CH_4 and non-methane volatile organic carbons (VOCs) in the presence of NOx and UV light and these component are present due to the motor vehicle activities. However, the eight hour means for the three sites were below WHO mean of 51 ppb. In Industrial Area, the same low amount of ozone were realized in early morning and at night with high amount observed throughout the day. The high values of ozone show that there is high concentration of ozone precursors in this site which is caused by industrial activities throughout the day. The eight-hour mean was 20.2 ppb which is below the WHO mean of 51 ppb. However, this is the highest recorded amount of ozone in the four sites.

On the Temporal variability, it was found that June to August season experiences the highest ozone levels as compared to the other seasons in both lower and upper levels. This is due to incursion from the south according to the backward trajectories, which has been proven to have high ozone concentration during this season due to biomass burning. The vertical profile of ozone shows that approximately 80% of ozone is found in the stratosphere. And finally on the Source region, the analysis done by running the backward trajectory using the HYSPLIT model found out the during the peak Season of

ozone, the source region is southern Africa which has high Biomass burning during these season, then is transported to Nairobi.

5.2 Recommendations

Kenya seeks to be industrialized by the year 2030, at a time when world over, policymakers and the general public are concerned with the degradation of air quality, especially in urban centers. From this study it has been found that, the site around Industrial area had a high amount of ozone concentration with the peak of this ozone concentration being throughout the day time, which increased outdoor pollution. The governmentought to come up with adequate strategies of tackling air pollution, which has direct health impacts upon the increasing population. Therefore, concerted efforts have to be made to find a sustainable balance between industry, human health and environmental protection. The study recommends that;

1. Further monitoring of air pollution to be conducted along major roads in Nairobi and other major cities, especially the precursor gases to map the spatial ozone and other air pollutant concentration.
2. Further analysis on the impact of air pollution on health need to be conducted.
3. Develop an atlas of air pollution levels in major cities in Kenya
4. Enhancement of ad hoc air pollution monitoring in different counties in order to profile pollution levels within the country.

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