



Clean Air Journal

ISSN 1017 - 1703

Vol 31 No 2

November / December 2021

Official publication of the
National Association for Clean Air

CLEAN AIR JOURNAL

ISSN 1017-1703
November / December 2021
Volume 31, No. 2

Published twice yearly by the National Association for Clean Air, Republic of South Africa

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Contents

Editorial

- 3 The new WHO Global Air Quality Guidelines: What do they mean for South Africa?
- 6 Introduction to the special issue: Air quality on the South African Highveld

News

- 9 Clean Air Journal celebrated this year's International Day of Clean Air for Blue Skies
- 11 NACA conference 2021 - Evidence-based pathways to clean air in South Africa

Research brief

- 14 Estimating lightning NO_x production over South Africa

In memoriam

- 17 Remembering the late Benton Pillay

Research articles

- 19 Modeling tropospheric ozone and particulate matter in Tunis, Tunisia using generalized additive model

Special issue

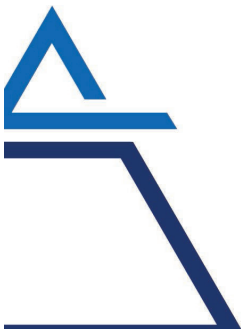
Air quality on the South African Highveld

- 36 Does apparent temperature modify the effects of air pollution on respiratory disease hospital admissions in an industrial area of South Africa?
- 48 Quantifying potential particulate matter intake dose in a low-income community in South Africa
- 56 The quality of the first and second Vaal Triangle Airshed Priority Area Air Quality Management Plans
- 71 Source apportionment of ambient fine and coarse aerosols in Embalenhle and Kinross, South Africa

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-  **Noise Modelling**
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Editorial

The new WHO Global Air Quality Guidelines: What do they mean for South Africa?

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<https://doi.org/10.17159/caj/2020/31/2.12915>

Ambient air quality standards are a key policy lever in air quality management. In South Africa, the introduction of the National Ambient Air Quality Standards (NAAQS) highlighted the shift in the focus of air quality management from source to receptor that was initiated with the introduction of the NEM:AQA. NAAQS were developed considering health impacts, ambient levels at the time and South Africa's developing economy. There is currently a process starting to review these standards, and this process aligns with the recent release of the new World Health Organization (WHO) Global Air Quality Guidelines (AQG) in September 2021 (World Health Organization, 2021). This is the first update of WHO's AQG since 2005. The WHO's guidelines take into account recent evidence of the effect of air pollution on human health, and many of the guidelines are substantially

lower than the previous guidelines (Table 1). In this editorial, we ask what the implications of the new WHO Guidelines are for air quality management and compliance in South Africa.

Overview of new guidelines

The WHO's new guidelines recommend air quality levels for six pollutants based on their health effects, these are: particulate matter with an aerodynamic diameter smaller than 10 µm and 2.5 µm (PM₁₀ and PM_{2.5}, respectively), ozone (O₃), sulphur dioxide (SO₂), nitrogen dioxide (NO₂) and carbon monoxide (CO). In addition to the guideline values, four interim target values are set as "incremental steps in a progressive reduction of air pollution." Interim targets 1-3 are the same as the interim targets

Table 1: A comparison between the South African National Ambient Air Quality Standards (2009 and 2012) and the WHO Air Quality Guidelines (WHO, 2006, WHO, 2021).

Pollutant	Averaging time	Unit	South African National Ambient Air Quality Standard		WHO Air Quality Guideline 2005	WHO Interim Targets (IT) and Air Quality Guideline (AQG) 2021				
			Current	1 Jan 2030		IT-1	IT-2	IT-3	IT-4	AQG
PM _{2.5}	24 hours ^a	µg/m ³	40	25	25	75	50	37.5	25	15
	1 year	µg/m ³	20	15	10	35	25	15	10	5
PM ₁₀	24 hours ^a	µg/m ³	75	n.c.	50	150	100	75	50	45
	1 year	µg/m ³	40	n.c.	20	70	50	30	20	15
O ₃	8 hours ^b	µg/m ³	120	n.c.	100	160	120	-	-	100
	Peak season ^c	µg/m ³	-	-	-	100	70	-	-	60
SO ₂	24 hours ^a	µg/m ³	125	n.c.	20	125	50	-	-	40
NO ₂	24 hours ^{a,d}	µg/m ³	-	-	-	120	50	-	-	25
	1 year	µg/m ³	40	n.c.	40	40	30	20	-	10

^a At the 99th percentile i.e. 4 allowed exceedance days per year
^b At the 99th percentile i.e. 11 allowed exceedance 8-hour periods per year
^c Average of daily maximum 8-hour mean O₃ concentration in six consecutive months with the highest 6-month running average O₃ concentration.
^d South Africa has a 1-hr NO₂ standard and not a 24-hour standard
n.c. = no change
- = no value

in the 2005 WHO guidelines, and the fourth interim target is the 2005 air quality guideline (Table 1). The South African NAAQS for $PM_{2.5}$, PM_{10} and O_3 fall between the WHO interim targets 2 and 3, while the South African NAAQS for NO_2 and SO_2 align with the WHO interim target 1.

Effective air quality management needs achievable goals

The new AQGs for PM and NO_2 are substantially lower than previous AQGs. This highlights that there is a risk to health at almost all exposure to these pollutants. We think this emphasizes the seriousness of poor air quality, and thus can be a motivation for focused swift action to improve air quality.

However, it has to be acknowledged that the WHO AQG levels are not attainable in many areas of the world, including many parts of South Africa. This is due in part to the many strong and varied natural sources of pollution (including dust, biomass burning, biogenic and marine sources) in South Africa. The impact of natural sources on air pollution levels is a key research gap. Long term observations in background sites, as initially envisaged in the 2012 Framework for Air Quality Management (RSA, 2012), are important to better understand these sources. Robust modelling experiments to estimate the contribution of natural sources to background levels across South Africa are also a critical piece of the puzzle that must be considered in the review of the NAAQS and setting of standards.

The great difference between the current pollution levels in South Africa and the NAAQS limits on the one hand, and the new WHO AQGs on the other hand, can be disheartening, as these targets may seem unachievable. However, urgent action to reduce air pollution levels is needed, as there are many places in South Africa where the ambient air pollution concentrations do not meet the NAAQS (e.g., Feig et al., 2016; Feig et al., 2019; Govender and Sivakumar, 2019; Hersey et al., 2015; Venter et al., 2012). Any improvement in air quality will have a positive impact on health.

South Africa's NAAQS will continue to be our country's benchmark in air quality management and compliance. The WHO guidelines and interim targets are one of many aspects that need to inform our national standards. Effective air quality management needs achievable goals, and thus, the WHO's interim targets can play an important role in South Africa's standards setting.

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Editorial for special issue

Introduction to the special issue: Air quality on the South African Highveld

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<https://doi.org/10.17159/caj/2020/31/2.12877>

The eastern Highveld of South Africa creates the ‘perfect storm’ for poor air quality. A variety of surface and elevated sources of air pollution emit into an atmosphere with frequent unfavourable conditions for the dispersion of the pollutants. Such conditions are especially dominant in winter.

Atmospheric emissions derive from eleven very large coal fired power stations, each accompanied by large scale ash disposal sites, that form the backbone of the country’s energy supply. Feeding the power stations are multiple open cast coal mines with drilling, blasting, coal and waste rock transport, crushing facilities and long unpaved haul roads. In addition to supplying the power stations, coal mines surround and supply the Sasol operation at Secunda, which is a large coal gasification, synthetic fuel production and wide-range hydrocarbon based chemical manufacturing operation. More than 140 million tonnes of coal are mined each year to supply the two key energy hubs of electrical power and liquid fuels.

Abandoned mines (some which are burning underground) discard and waste coal dumps that are prone to spontaneous combustion and sources of wind-blown dust scar the landscape. The area is also home to ferrochrome smelters and other large industrial processes near Emalahleni and Middelburg.

Air quality in the Highveld is also affected by veld fires. In summer, the entire Highveld area is green. This greenery is either large cattle pastures, seemingly endless expanses of maize and other crops and ever decreasing outcrops of natural vegetation. In winter however, the area transforms into a dry brown and khaki landscape with frequent fires that extend over large areas, transforming the once green vegetation into hundreds of tonnes of ash and semi-volatile hydrocarbons that are emitted to atmosphere.

Some 15 million people live in this area too. From individual farmsteads, through the many small towns, the larger urban centers and finally, but perhaps most importantly, the low-income, dense settlements. Multiple informal settlements append to these larger dense settlements or exist as islands with very limited services. Unemployment is rife and so is poverty in these areas with high levels of crime. The generally poor socio-economic circumstances mean that even where electricity

connections are available, domestic fuels such as low-quality coal, wood and litter are widely used in rudimentary appliances for cooking, space heating and hot water. In addition, the virtual absence of domestic waste collection services compels continual burning of such waste, often in close proximity to dwellings.

Above this highly complex array of land uses and socio-economic circumstances is the Highveld atmosphere. The quintessential feature of this atmosphere is its stability. The large continental anti-cyclone creates a highly stable atmosphere in both summer and winter, but it is in the winter that the stability really comes to the fore. The same anticyclone that drives that stability also prevents the inward flow of maritime moist air, making winters bone dry and removing all obstacles to the escape of heat from the surface to space. Diurnal temperature profiles see moderate temperatures in the middle of the day, but as the sun starts to dip, the temperatures drop reaching their lowest point just before dawn the following day and then recovering again as the sun rises. The cycle is repeated daily.

Strong, deep surface inversions are an almost everyday occurrence in the Highveld atmosphere and above that are elevated inversions too. Layers of highly stable air stack one above the other, each preventing the movement of air. A winter’s night in the Highveld sees pollution trapped below the inversions become progressively more concentrated resulting in alarming deteriorations in air quality, especially in respect of PM but also NO₂ and periodically SO₂. Compliance with the NAAQS is unacceptably infrequent.

Unsurprisingly, the people who live in that air show that they do through the manifestation of abnormally high occurrences of upper respiratory tract diseases including asthma attacks. It has been shown (*inter alia* by the seminal publication referred to in the next paragraph) that it is the children that suffer the worst of the effects. It is a veritable tragedy that the situation has remained largely unchanged for several decades, despite several efforts by government to manage air quality. The most important of these was the development of an air quality management plan for the Highveld Priority Area (DEA, 2012). The plan outlines measures that should be implemented by government, industry, and NGOs to improve air quality in the Priority Area.

It has been over 8 years since the plan was developed and implemented but there is no significant improvement in ambient air quality. Is the lack of improvement simply imperfect implementation of those interventions or is it due to an inadequate characterization of the multiple emission sources and/or the atmospheric dispersion processes that drive air quality, meaning that the management interventions are not up to the task?

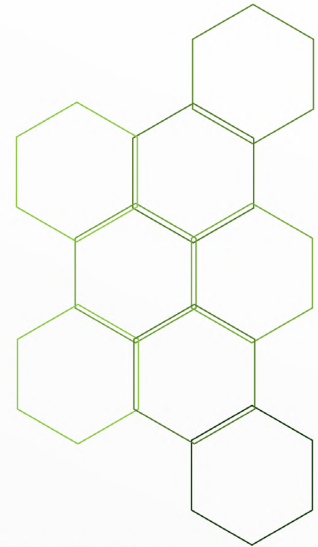
In 1987 Professor Peter Tyson, Dr Fred Kruger and Dr Wynand Louw published what was to become a seminal assessment of the air quality in this region (Tyson, Kruger and Louw, 1987). Since that time there have been many additional investigations including a *status quo* assessment for the air quality management plan described above. Perhaps most importantly, the network of monitoring stations established in the early 2000s, provides a growing database of continuous ambient air quality monitoring under a wide range of meteorological conditions, spanning more than a decade.

This call for papers sought to consolidate the most recent air quality research undertaken in the Highveld to build on the foundation established by Tyson, Kruger and Louw (1987). Our hope is that through consolidating the investigations, assessments, and ambient air quality monitoring, that management interventions can be refined, improved, and more effectively implemented, with the effect of materially reducing the prevailing disease burden and ultimately giving effect to the constitutional right of people living on the Highveld, to an environment that is not harmful to their health or welfare.

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News

Clean Air Journal celebrated this year's International Day of Clean Air for Blue Skies

Bianca Wernecke¹

¹Environment and Health Research Unit, South African Medical Research Council, Doornfontein, Johannesburg, 2094
<https://doi.org/10.17159/caj/2020/31/2.12811>

This year's theme for #CleanAirDay, which is celebrated annually on 7 September, was "Healthy Air, Healthy Planet" and it aimed to emphasize the health effects of air pollution, particularly during the COVID-19 pandemic.

To celebrate the occasion on 7 September 2021, the Clean Air Journal partnered with the Environment and Health Research Unit of the South African Medical Research Council (SAMRC) to release a suite of video talks by African researchers in the air quality space who had previously published research findings in the Clean Air Journal. The five videos covered various topics relevant to Africa, ranging from household air pollution exposure and health risks to the importance of meeting National Ambient Air Quality Standards as well as the effectiveness of government policy at reducing pollution and saving people's lives. The theme of air pollution in Africa in the time of COVID-19 was also unpacked.

In anticipation of #CleanAirDay, the Clean Air Journal and the SAMRC released one video per day for five days on their respective social media platforms (Facebook, Twitter and LinkedIn).

Official teaser to the videos:

<https://youtu.be/cd7zoUqgTvg>

Full playlist of the videos:

https://www.youtube.com/playlist?list=PLFdK7Ly-ce5lptRBVEHvo4XF_G1Y-AfHG

More detail on the talks as well as the corresponding Clean Air Journal publications are as follows:

Bianca Wernecke

Many South African households burn dirty fuels (coal and wood) as the primary source of energy for heating and cooking purposes. Unfortunately, air pollution caused by these activities has a significant influence on public health and some of the people most affected are the poorest of the poor living in low-income communities. Ms Bianca Wernecke from the SAMRC's Environment and Health Research Unit talks to us more about this.

Video: <https://youtu.be/Z8PwigYUzyg>

CAJ article: <https://www.cleanairjournal.org.za/article/view/7016>

Nick Okello

Dr Nick Okello, an Environmental Scientist from Kenya, explains the different air pollution trends and their devastating effects on human health and the effectiveness of government policy



in reducing pollution and saving people's lives in Richards Bay.

Video: <https://youtu.be/lioMYqficQA>

CAJ article: <https://cleanairjournal.org.za/article/view/8012>

Katye Altieri

According to Dr Katye Altieri, a Senior Lecturer in the Department of Oceanography from UCT, air pollution has a negative impact on human health as well the economy of any country. She explains that in order to protect human health, there is a need for effective air quality management which relies on the attainment of air quality standards.

Video: <https://youtu.be/V7eGQaEWuiQ>

CAJ article: <https://www.cleanairjournal.org.za/article/view/7005>

Ncobile Nkosi

Coal is a main energy source used by many in low-income residential areas to meet basic needs, like for cooking and heating. However, it is a major source of a pollutant of concern, namely fine particulate matter. Ms. Ncobile Nkosi, a Geography Lecturer at the North-West University, talks about emission factors associated with residential burning of solid fuels using traditional cast-iron stoves.

Video: https://youtu.be/D0_iEzi5hw4

CAJ article: <https://cleanairjournal.org.za/article/view/6961>

Andriannah Mbandi

Dr Andriannah Mbandi, a Lecturer at the South Eastern Kenya University, talks about air pollution in Africa in the time of COVID-19: the air we breathe indoors and outdoors. Her commentary on this subject was published in the Clean Air Journal.

Video: <https://youtu.be/olEtE71NuY6o>

CAJ commentary: <https://cleanairjournal.org.za/article/view/8227>

Subtitles are available on all the videos



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News

NACA conference 2021 - Evidence-based pathways to clean air in South Africa

Gabi Mkhathshwa¹, Roelof Burger², Anzel De Lange³

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<https://doi.org/10.17159/caj/2020/31/2.12883>



The Annual Conference of the National Association for Clean Air (NACA), with the theme of “Evidence-based pathways to clean air in South Africa”, was held from 6 to 8 October 2021. The conference followed the Department of Forestry, Fisheries and the Environment’s (DFFE), Air Quality Governance Lekgotla. Even though the NACA conference had to be presented virtually (online) for the second year in a row, a complete programme of scientific presentations and technical sessions was offered. In addition, a session of 3 minute talks (or 3MTs) was presented for the first time this year and will become a permanent part of the programme for future conferences.

Keynote addresses during the conference included topics such as the development of cost-effective air quality strategies, key findings and implications for Southern Africa from the IPCC AR6, the World Bank’s Pollution Management and Environmental Program, and Flue Gas Desulphurisation (FDG) technology. In terms of the programme, sessions on air quality and human health, urban and industrial air quality, monitoring, air quality modelling, and air quality validation and uncertainty guaranteed that every interest was piqued during the three-day conference.

Three well-attended workshops also took place. The first, a multi-stakeholder workshop hosted by the DFFE and NACA, was focused on low-cost sensor technologies in ambient air quality monitoring. A technical session on mine dust and gold tailings was sponsored by Global Challenges Research Fund (GRF) Mine Dust and Health Network, and a technical session on source apportionment was hosted by Eskom and the North-West University.

The conference was attended by just over 90 paid members and sponsored access by the Mine Dust and Health Network to 65 undergraduate and honours levels students from the University of the Witwatersrand, North-West University, University of Limpopo, University of Cape Town, and the University of Johannesburg.

Feedback from conference attendees was positive. More than 70% felt that the conference was adequately marketed, only 8.7% felt otherwise. 95% believed the conference website was professional and informative. 87% felt they had easy access to the virtual conference. More than 90% were happy with the conference program and 95% believed the talks were engaging and applicable to the NACA community. All three the technical sessions, including the low-cost monitoring, mine dust and health, and the source apportionment sessions were positively received with more than 80% of participants ranking it as interesting and engaging. 68% of participants said that future conferences should include a virtual component.

The NACA awards were this year sponsored by Eskom & SACNASP and presented by NACA President, Ms Gabi Mkhathshwa. These awards are used to recognise and reward outstanding contributions towards the cause of clean air. Nominations for awards are considered annually. After adjudication by the NACA Awards Committee, 2021 awards were presented at the NACA Annual Conference. The first award was a posthumous award to Mr Benton Pillay, a former president of NACA. Benton Pillay was a true atmospheric scientist in heart and soul and was very

passionate about air quality management. The second award went to Mr Louis Kleynhans an Environmental Manager of Ergo Mining, responsible for the environmental management of 85 separate dormant, active and closed tailings facilities, covering approximately 3 500 Ha stretching from Roodepoort in the west to Nigel in the east. The third and final award was to a Director and Principal at Airshed Planning Professionals, Dr Lucian Burger. Over the past three decades Lucian has been actively involved in the development of atmospheric dispersion modelling and its applications, air pollution compliance assessments, health risk assessments, mitigation measures, development of air quality management plans, meteorological and air quality monitoring programmes, strategy and policy development, training, and expert witnessing.

Over and above the NACA awards, the conference gave out conference awards to the following:

Best Scientific paper

Bianca Wernecke: South African Medical Research Council

Fuel use, Air pollution and Health in two South African low-income communities located in the Highveld Priority Area

Authors: Wernecke, B., Mathee, A., Abdelatif, N., Seocharan, I., Rajen Naidoo, R., Jafta, N., Ramcharan, K., Phaswana, S., Pauw, C., Howard, A., Smith, H., Ndala, L., Herbst, D., McCourt, B., Wright, C.

Best Scientific paper Runner up

Farina Lindeque: University of Limpopo

Air Pollution in South African Twitterverse: Exploring user awareness over the past decade

Authors: Lindeque, F., Klopper, D., Botha, I., Burger, R., Piketh, S.

Best Student paper

Coenraad Meyer: North-West University

The influence of coal pellet properties on its emissions and thermal performance in a semi-continuous coal stove

Authors: Meyer, C.W., Neomagus, H.W.J.P., Piketh, S.J., Bunt, J.R., Conradie, F.H., Annegarn, H.J., Pemberton-Pigott, C.

Best Student paper Runner up

Faith February: University of Cape Town

Observations of Aerosol size distributions during marine and continental air masses in Simon's Town and False Bay

Authors: February, F., Altieri, K., Van Eijk, A., Piazzola, J.

Best 3 Minute Talk (3MT)

Liezl Bredenkamp: North-West University

What is the background concentration of Mercury in ambient air over South African interior?

Best 3 Minute Talk (3MT) Runner up

Zamahlase Sibisi: North-West University

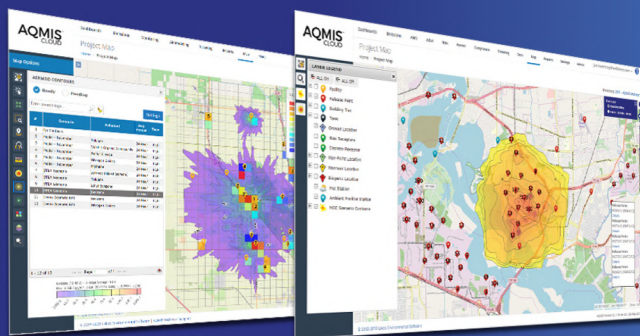
Characterisation of ambient air quality within an urban settlement using in-situ measurements: A case study for Soweto and Gauteng

3 minute Talk (3MT) Honourable mention

Faith February: University of Cape Town

What is the effect of aerosol loading on air quality in Simon's Town?

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Research brief

Estimating lightning NO_x production over South Africa

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<https://doi.org/10.17159/caj/2020/31/2.12948>

Poor air quality is a key environmental concern in South Africa, as it poses a serious threat to the well-being of the people of South Africa. Nitrogen oxides (NO_x = nitric oxide (NO) + nitrogen dioxide (NO₂)) are toxic air pollutants and play a significant role in tropospheric chemistry. Global NO_x hotspots are the industrialised regions of the USA, Europe, Middle East, East Asia and eastern parts of South Africa. Lightning is one of the many natural and anthropogenic sources of NO_x to the troposphere. The discourse on NO_x over the southern African continent has mainly focused on anthropogenic sources. However, lightning is known to be a significant source of tropospheric NO_x globally. It is therefore important to understand its contribution to the national and global NO_x budget.

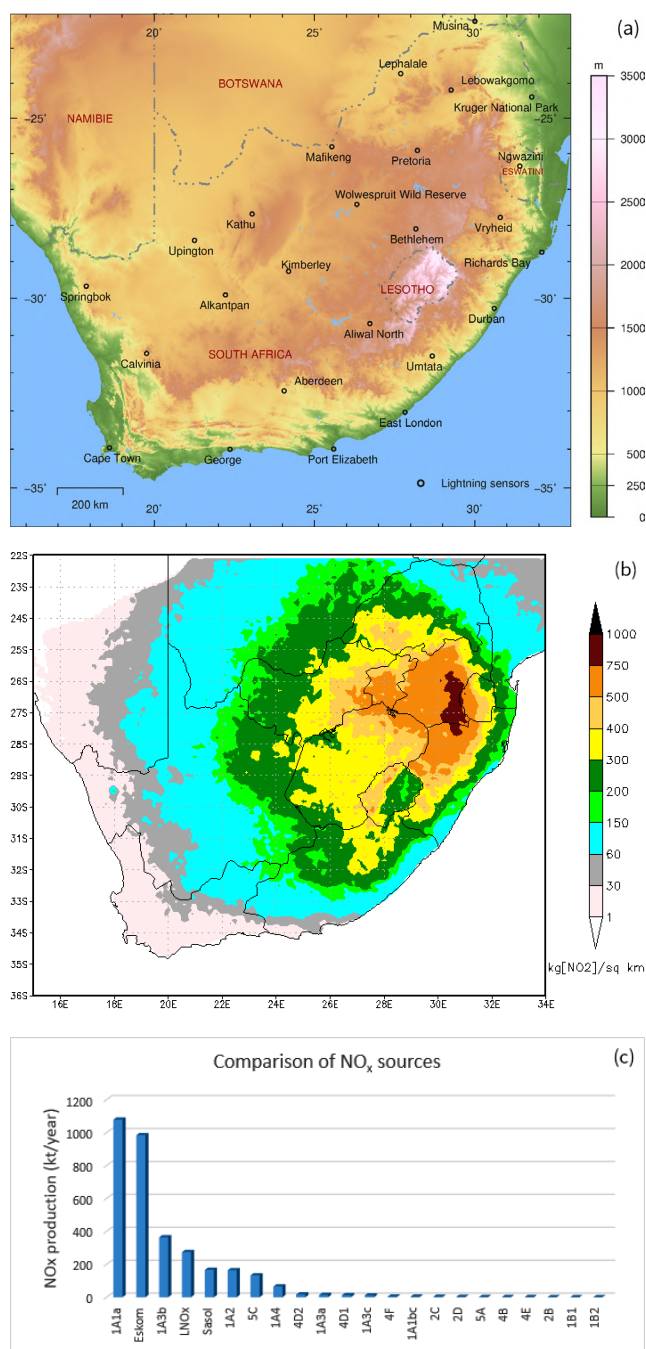
A recent paper by Maseko et al., (2021), published in the South African Journal of Science, used data from the South African Lightning Detection Network to approximate the contribution of lightning on the NO_x load over South Africa (Figure 1a), and to develop a gridded data set of lightning-produced NO_x (LNO_x) emissions for the period 2008–2015 (Figure 1b). The Network monitors cloud-to-ground lightning strikes; and theoretically has a detection efficiency of 90% and a location accuracy of 0.5 km. An emission factor of 11.5 kg NO₂/flash was employed to calculate a national LNO_x budget of ~270 kt NO₂/year. The calculated LNO_x was 14% of the total NO_x emission estimates published in the EDGAR v4.2 data set for the year 2008 (Figure 1c). The LNO_x emission inventory will improve model performance and prediction and enhance the understanding of the contribution of lightning to ambient NO₂.

References

Maseko, B., Feig, G., Burger, R., 2021. Estimating lightning NO_x production over South Africa. *South Afr. J. Sci.* 117. <https://doi.org/10.17159/sajs.2021/8035>

Schumann, U., Huntrieser, H., 2007. The global lightning-induced nitrogen oxides source. *Atmos Chem Phys* 85.

Figure 1: (a) Position of the 25 Vaisala lightning detection sensors over South Africa; (b) Average number of total LNO_x in (kg NO₂/km²/year), using the emission factor of Schumann and Huntrieser, (2007); (c) NO₂ emission from EDGAR v4.2 and lightning for the year 2008 over South Africa, in kt NO₂/year.





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In memoriam

Remembering the late Benton Pillay

<https://doi.org/10.17159/caj/2020/31/2.12914>

The Clean Air Journal regrets the untimely passing of Benton Pillay. Benton was dedicated to the improvement of Air Quality in South Africa. Benton was actively involved in NACA for more than a decade and served as President of the Association from 2017 –2018. During his time leading the association he was a staunch supporter of the Clean Air Journal. Benton passed away suddenly in July 2021

He was a qualified Chemical Engineer who worked for many years in public and private enterprise in the field of air quality management. Benton was a founding member and a director at uMoya-NILU and a senior staff member at their Gauteng office. During his time at uMoya-NILU he was influential in the development of air quality management in South Africa this includes his involvement in air quality assessments, notably the Provincial Air Quality Monitoring Plans for the Western and Eastern Cape, the development of the Air Quality management plans for the Highveld and Waterberg Bojanala Priority Areas and the development of AQMPS for South African Ports. These plans and the assessments that went into them have guided air quality management in these areas. In addition, he was instrumental in the development of the National Atmospheric Emissions Inventory System (NAEIS). From an observational aspect he was involved in the deployment and operation of a significant amount of ambient air quality observation infrastructure and stack sampling. Many of the current technicians developed under his tutelage.

We will remember Benton for his intellect and for being a great colleague and a friend, his stylish presence at the NACA conferences will be missed. He leaves behind his wife, Doreen, and his three children, Mishka, Liam and Luke, whom he loved very much. We extend our most heartfelt condolences and sympathy to Benton's family.





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Research article

Modeling tropospheric ozone and particulate matter in Tunis, Tunisia using generalized additive model

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Received: 8 October 2020 - Reviewed: 23 November 2020 - Accepted: 10 September 2021

<https://doi.org/10.17159/caj/2021/31/2.8880>

Abstract

The main purpose of this paper is to analyze the sensitivity of tropospheric ozone and particulate matter concentrations to changes in local scale meteorology with the aid of meteorological variables (wind speed, wind direction, relative humidity, solar radiation and temperature) and intensity of traffic using hourly concentration of NO_x , which are measured in three different locations in Tunis, (i.e. Gazela, Mannouba and Bab Aliwa). In order to quantify the impact of meteorological conditions and precursor concentrations on air pollution, a general model was developed where the logarithm of the hourly concentrations of O_3 and PM_{10} were modeled as a sum of non-linear functions using the framework of Generalized Additive Models (GAMs). Partial effects of each predictor are presented. We obtain a good fit with $R^2 = 85\%$ for the response variable O_3 at Bab Aliwa station. Results show the aggregate impact of meteorological variables in the models explained 29% of the variance in PM_{10} and 41% in O_3 . This indicates that local meteorological condition is an active driver of air quality in Tunis. The time variables (hour of the day, day of the week and month) also have an effect. This is especially true for the time variable “month” that contributes significantly to the description of the study area.

Keywords

Air pollution, Particulate Matter, Tropospheric Ozone, GAM, meteorology, traffic

Introduction

Nowadays, it is well known that air pollution and its impact on human health have become a primary topic in atmosphere research. A good number of epidemiological studies have demonstrated the strong link between atmospheric pollution and daily deaths and hospitalizations of pulmonary and cardiac diseases (Sinharay et al., 2017; Bourdrel et al., 2017). Tunisia is a beautiful country with diverse, complex geography and is located between the Mediterranean coast and the Saharan region. This location together with a diversity of air pollution sources (e.g. traffic, industrial, dust) leads to exceedances of air quality guideline values recommended by the World Health Organization (WHO, 2016). Tunisia reports high annual mean concentrations of $\text{PM}_{2.5}$ and PM_{10} , which should not exceed 10 and 20 $\mu\text{g}\cdot\text{m}^{-3}$, respectively (WHO, 2016). Accelerated growth in emission sources of air pollutants in most important Tunisian cities like Tunis, Sfax and Gabes (Melki, 2007; Bouchlaghem and Nsom, 2012) now cause an urgent need to adopt specific policies in managing air pollution.

Air pollution modeling is an integral part of air pollution management and policy (Karaca et al., 2006; Saffarini and Odat, 2008). Previous air quality studies conducted in Tunisia mainly

focused on the physical characteristics, correlations between pollutants, the sources of PM_{10} and forecasting air quality (Melki, 2007; Bouchlaghem et al., 2009; Ayari, Nouira and Trabelsi, 2012; Calzolari et al., 2015). A few investigations focusing on the interplay between meteorology and air quality has been done in Tunisia. The study conducted in Tunis (Melki, 2007) presents the role of the temperature inversions, which determine the majority of the highest pollution levels in the north of the country. They used multiple linear regressions to evaluate the statistic dependence between the ozone concentrations and the weather conditions. According to Bouchlaghem et al. (2009), some sea breeze events are responsible for air quality. Their result shows that under these circumstances, the nearby power plant is responsible for air quality degradation in the region of Sousse (the East central part of Tunisia). Bouchlaghem and Nsom (2012) highlighted the influence of the Saharan dust on PM_{10} concentrations. They concluded that PM_{10} concentrations on days with Saharan dust contributions are higher than the average daily value with the absence of this phenomenon. In sum, no study has as yet dealt with the relationship between particulate matter and ozone concentrations and meteorological conditions in Tunisia based on the use of a non-linear statistical approach.

Generalized Additive Model, as an extension of Generalized Linear Model, has been employed in few studies for modeling pollutant concentrations, especially PM_{10} (Taheri Shahraini et al., 2015) and O_3 (Ma et al., 2020). As a statistical tool that is able to simulate non-linear relationships by smoothing input variables (Hastie and Tibshirani, 1990), Generalized Additive Models (GAM) have been used in many environmental issues and recent studies (Ma et al., 2020; Yang et al., 2020). In the last two decades, this statistical approach has been used as a standard analytic tool in time-series studies of air pollution and human health (He, Mazumdar and Arena, 2005; Dehghan et al., 2018; Ravindra et al., 2019).

GAM models delivered good performance and can be equivalent to those of other methods such as neural networks (Schlink et al., 2003). Aldrin and Haff (2005) used meteorological predictors in order to model PM_{10} , $PM_{2.5}$ and the difference between PM_{10} and $PM_{2.5}$ mass concentrations, and their models gave a reasonably good fit in terms of the squared correlation coefficient with 72% and 80% for PM_{10} and NO_x , respectively. Pearce et al. (2011) noted the influence of local-scale meteorological conditions on air quality in Melbourne (Australia). Munir et al. (2013) offered a new GAM to predict daily concentrations of PM_{10} in Makkah using lag PM_{10} concentrations. This model showed the vital role of meteorological variables and traffic related air pollutants in describing the variations of the PM_{10} concentrations. Again based on GAM analysis, Belušić, Herceg-Bulić and Bencetić Klaić (2015) employed the novel GAM approach to quantify the influence of local meteorology on air quality in Zagreb, Croatia. This study confirmed the well-known impact of wind direction and speed in variations of air pollution.

The objective of this study is to investigate the magnitude in which pollutant concentrations respond to measures of local meteorology and temporal variables in Tunis. Statistical models were developed for hourly mean PM_{10} and O_3 concentrations for three sites of Tunis in order to quantify the impact of meteorology on PM_{10} and O_3 levels. The paper is organized as follows: The Materials and methods section provides information on our data sources and data-handling methodology. Then it presents the description of the proposed methods and a brief introduction to Generalized Additive Models. The Results and discussion section discusses the findings, highlights the most important results and details a statistical evaluation of the model. Finally, we conclude the work in the Conclusions.

Materials and methods

Site description and sample collection

The study area is located in the metropolis of the Greater Tunis region, which consists of four governorates: Tunis, Ariana, Manouba and Ben Arous. The area of the Greater Tunis is 300,000 hectares, with a population of 2.5 million. This city contributes 30% to the total pollution of the country (INS, 2014) (Fig. 1). Three urban and suburban monitoring stations (i.e. Bab Aliwa, Gazela and Mannouba) were selected for this study (Fig. 2). These

stations are located in three governorates: Tunis, Ariana and Mannouba.

Tunis City (capital of Tunisia) is located in the North part of Tunisia ($36^{\circ}49' N$, $10^{\circ}11' E$). The urban area (1 056 247 inhabitants) is about 346 km² surface. The sampling site "Bab Aliwa" is classified as urban, is located in the vicinity of one of Tunis's major traffic avenues and is near to central bus station and the largest cemetery in the country.

Ariana is also located in the North part of Tunisia ($36^{\circ} 51' N$ $10^{\circ} 11' E$). Its urban area accounts about 576 088 inhabitants. The measurement station sample "Gazela" is classified as urban and is mainly influenced by residential, traffic, and commercial activities.

Mannouba is located in the center of the northern governorates ($36^{\circ} 48' N$ $10^{\circ} 5' E$). The urban area (379 518 inhabitants) is about 1 137 km² surface. The sampling site "Mannouba" is suburban and it is known for its typically agricultural and industrial character.

The data set used consists of pollution data for the period from 01/01/2008 to 31/12/2009, with corresponding measurements of meteorological conditions provided by "Agence Nationale de Protection de l'Environnement" (ANPE). This period was chosen because it is the only one with few missing values (< 7%). At each site, air pollution is measured with standards methods used in Tunisia. PM_{10} and O_3 instruments are designed by Teledyne Advanced Pollution Instrumentation Company (<http://www.teledyneapi.com>). Levels of PM_{10} were calculated by means of automatic beta radiation attenuation monitors. For O_3 , the Teledyne model used is 400A. Data processing techniques and standard methods are described in the analyser instruction manuals. Additionally, all stations were equipped with automatic weather monitoring. All data series were collected hourly. Due to measurement errors, a few negative pollutant concentration values occasionally appeared in the raw data. These values cause problems because pollution data are modelled at log-scale (Aldrin and Haff, 2005) and have been replaced by the minimum observation in the data (1 ppb for NO_x and O_3 and $1 \mu g \cdot m^{-3}$ for PM_{10}). The limited sensitivity of the measurement instruments caused many observed zero values (about 0.05% on average), which were considered as erroneous data.

Table 1 presents a basic statistical overview of air pollution and meteorological variable values after the application of the data quality control process. Fig. 3 shows the average seasonal evolution of PM_{10} (from January 2008 to December 2009) in the studied regions. We note different behavior at the various sites with very high levels compared to the PM_{10} annual limit of the 2008 EU Air Quality Directive ($40 \mu g \cdot m^{-3}$). The right-hand plot indicates that average seasonal evolution of O_3 is around the O_3 maximum daily 8-hour mean limit (60 ppb) of the 2008 EU Air Quality Directive (Directive, 2008), except for Gazela site, an overshoot was observed. So, pollution levels can be



Figure 1: North African map displaying Tunisia and Tunis City

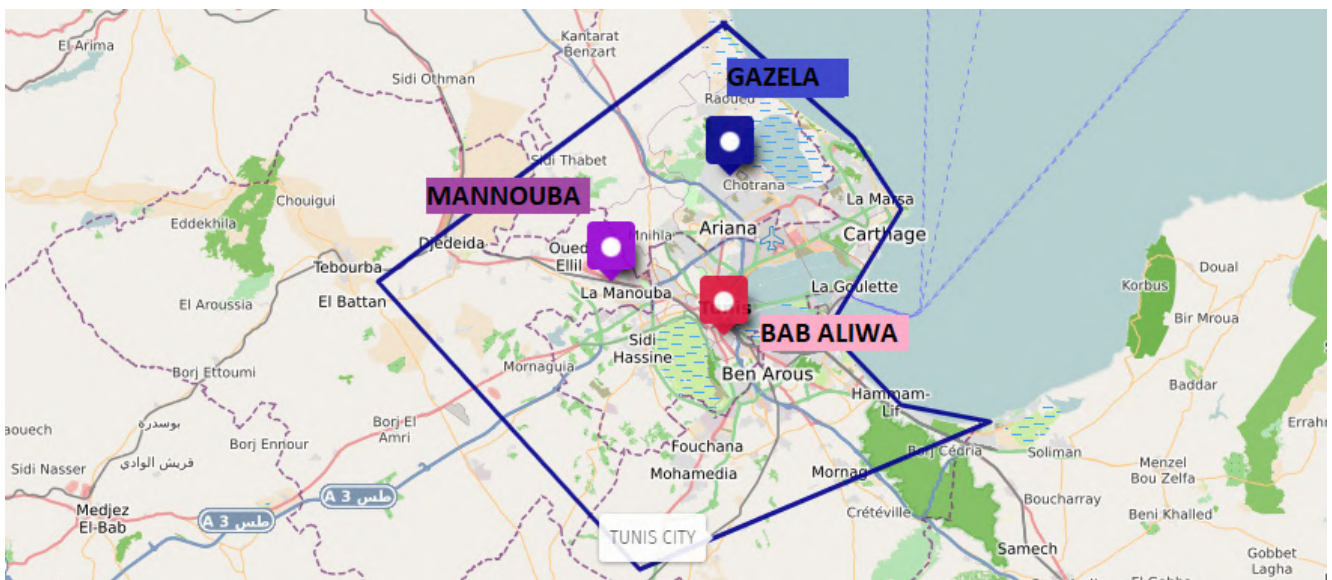


Figure 2: Map of study area showing the location of monitoring stations

Table 1: Summary statistics of data used for model development, showing the mean, median, standard deviation, minimal and maximal values of the data collected over the 3 studied stations (01/01/2008 to 31/12/2009).

Variable	Units	Mean	Median	Min	Max	SD
O ₃ (O ₃)	ppb	54.25	60	1	257	23.38
PM ₁₀ (PM ₁₀)	µg.m ⁻³	68.26	52	1	801	59.92
NO _x (NO _x)	ppb	25.96	15	1	395	28.15
Temperature (TT)	°C	18	18	3	43	6.99
Wind speed (WS)	m.s ⁻¹	1.70	1	0	8	1.19
Wind direction (WD)	deg	201.8	249	0	360	115.50
Solar radiation (SR)	W.m ⁻²	177.8	44	0	927	235.51
Relative humidity (RH)	%	61.83	63	11	100	16.83
Day of the week (DW)	Days	-	-	1	7	-
Hour of the day (HD)	Hours	-	-	1	24	-
Month (Month)	-	-	-	1	12	-

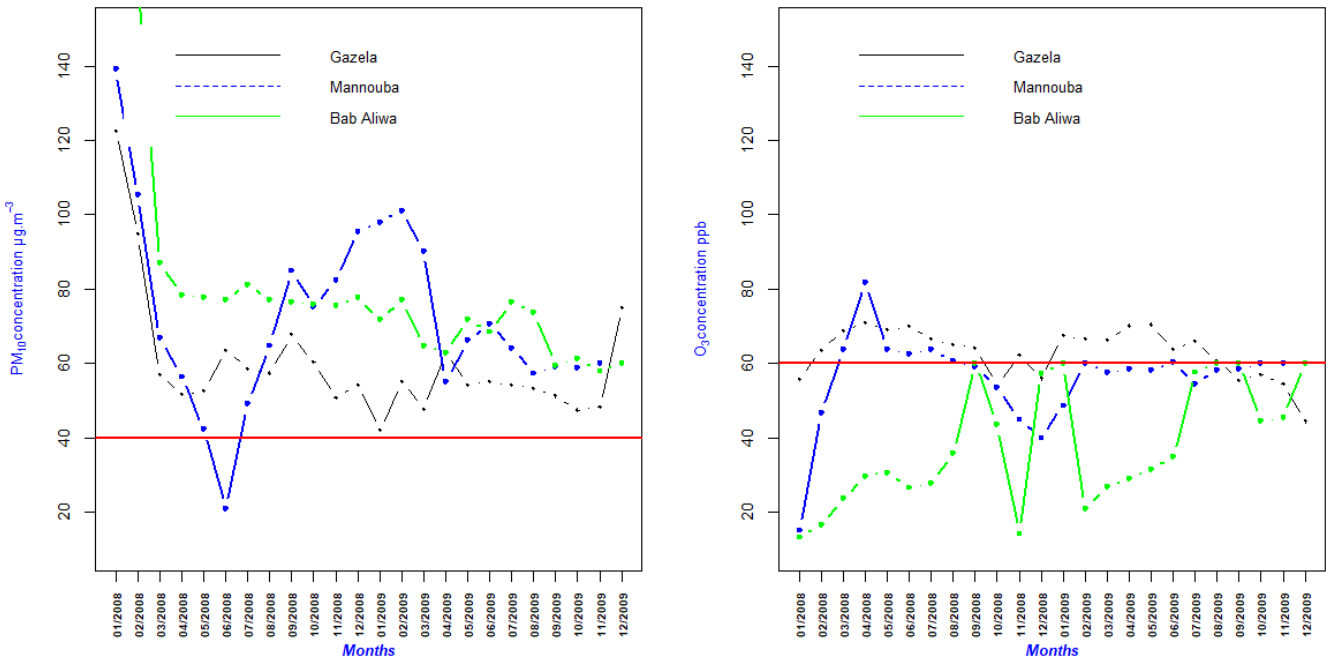


Figure 3: PM₁₀ and O₃ monthly averaged concentration recorded at all monitoring sites from January 2008 to December 2009. The horizontal red line indicates PM₁₀ and O₃ annual limit of the 2008 EU Air Quality Directive

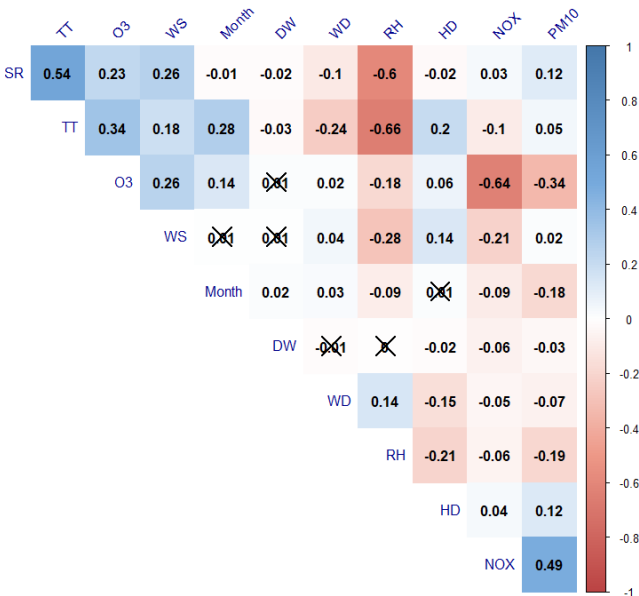


Figure 4: Pearson correlations matrix of all variables. The strikethrough coefficients were insignificant at the 0.05 significance level.

differentiated by geographical area. In Algeria, the north African country like Tunisia, the modeling results of Belhout et al. (2018) show that the Algerian annual average limit for PM₁₀ (80 µg.m⁻³) has been exceeded in some Algiers areas; by consequence, air quality guidelines fixed by the WHO (20 µg.m⁻³), (WHO, 2006) and the European Union (EU) (40 µg.m⁻³) for PM₁₀ are also exceeded. Rahal et al. (2014) found that significant pollutant releases in the study area are located at hyper-centre and at centre of the Wilaya of Algiers. Many sites in Greater Agadir Area, Morocco, have high levels of ozone and other pollutants that meet national air quality standards. The annual average of PM₁₀ is largely below the limit value on Agadir city (Chirmata, Leghrib

and Ichou, 2017) . All countries of the North Africa sub-region do not have specific legislation on air quality.

Generalized additive models

Generalized Additive Models (Hastie and Tibshirani, 1990) are used to assess the relationship between air pollution concentrations and different factors. GAMs are regression models in which linear predictor $\sum \beta_j x_j$ is replaced by a sum of smooth functions of covariates $\sum s(x_j)$. Additive models are considered as a semi-parametric extension of the generalized linear model (GLM) which automatically estimate the optimal degree of non-linearity of the model. The additive model in general form can be written as:

$$g(E(y_i)) = g(\mu_i) = s_0 + \sum_{k=1}^p s_k(x_{ki}) + \epsilon_i \tag{1}$$

where g is a link function that links the expected value to the predictor variables, μ_i is the expectation of the response variable y_i , s_0 is the overall means of the response, $s_k(x_{ki})$ is the smooth function of i^{th} value of covariate k , p is the total number of covariates, and ϵ_i is the i^{th} residual which is assumed to be normally distributed: $\epsilon_i \sim N(0, \sigma^2)$. The smooth function was used to minimize the penalized residual sum of squares (shown in equation 2):

$$RSS(f, \lambda) = \sum_{j=1}^n (y_j - s(x_j))^2 + \lambda \int s''(t)^2 dt \tag{2}$$

The term $\sum_{j=1}^n (y_j - s(x_j))^2$ evaluates the closeness to the data and $\lambda \int s''(t)^2 dt$ penalizes curvature in the function. λ is a fixed smoothing parameter. The increase of the value of λ provides a smoother function. The choice of this parameter becomes

critical given the flexibility of the GAM model and the risk of over-fitting. Generalized Cross Validation (GCV) is the most used method to fix the smoothing parameter λ . In this paper, the main purpose is to find the combination of explanatory variables which can describe a high degree of the pollutant concentration variability (R^2) in Tunis. In order to analyze the seasonality of O_3 and PM_{10} concentrations that exist in this data, we started by fitting a preliminary base model with time variables only (equation 3):

$$\log(E(y_i)) = s_0 + s(DW, k = 7) + s(HD, k = 24) + s(Month, k = 6) + \varepsilon_i \quad (3)$$

(Model with time variables only)

The variable day of the week (DW) was used to account for weekly variations. Also, the predictor hour of the day (HD) was employed with values ranging from 1 to 24. This variable is meant to take care of diurnal variation that is not explained by the other variables. Additionally, since air pollution data are known to be seasonal, k which is the maximum number of knots for each smoother. The smoothing spline for HD had 24 knots and was employed to account for processes on time scales larger than one hour. The variable DW had 7 knots one for each day. Finally, the variable Month was employed with $k = 6$. Both residuals histograms and scatter plots confirmed the adequacy of this choice of k values (see the section "Assessment of the model performance").

Tropospheric ozone O_3 and particulate matter PM_{10} concentrations were modeled separately using the model given by (equation 4), with five meteorological variables, temperature (TT°), Relative Humidity (RH %), Solar Radiation ($SR \text{ W.m}^{-2}$), Wind Speed ($WS \text{ m.s}^{-1}$), Wind Direction (WD degree from the north) applied via the GAM modeling function in the R environment for statistical computing inside the "mgcv" package (Wood, 2006). Traffic data and precipitation data were not available in the study areas. Therefore, three temporal variables and some traffic related air pollutant data were included to roughly account for traffic density and industrial emissions. Nitrogen oxides ($NO_x \text{ } \mu\text{g.m}^{-3}$) was used as explanatory variables instead traffic flow data (Pont and Fontan, 2000) and to represent a source for secondary particle matter. The predictor variables are slightly correlated (Fig. 4). For example, the correlation between the wind speed and the solar radiation is 0.26, between the temperature and hour of the day, it is 0.2. A strong negative linear relationship was detected between relative humidity and temperature (-0.66) and between relative humidity and solar radiation (-0.6). Most other correlation coefficients are 0.50 or less in absolute values. Based on these moderate correlations, we do not expect any serious problems with confounding effects between predictor variables. In this study, the Variance Inflation Factor (VIF definition in Appendix A) was used to detect the multicollinearity of variables (Belušić, Herceg-Bulić and Bencetić Klaić, 2015) and the multicollinearity is considered very important when VIF values are higher than 10 (Graham, 2003). For all variables, VIF values were lower and ranged from 1.001 for the day of the week (DW) to 2.934 for the temperature. Thus, we assumed that all variables are not collinear, and a

regression method could be applied. In order to select the final model, meteorological variables were added to the base model (equation 3) upon which Akaike's Information Criteria (AIC) was calculated. A variable remained in the final model if the fit yielded a lower AIC. Finally, the model for each pollutant can be written as:

$$\log(E(y_i)) = s_0 + s(HD, k_1) + s(DW, k_2) + s(Month, k_3) + s(TT, k_4) + s(WS, k_5) + s(WD, k_6) + s(RH, k_7) + s(SR, k_8) + s(NO_x, k_9) + \varepsilon_i \quad (4)$$

(Model with all variables)

The maximum number of knots for each smoother k must be chosen before the smoothing function is estimated. It controlled the smoothness of each function $s_k(x_{ki})$ in the final model. This particular parameter should be large enough so that the main process which governs concentrations values are included in the model. Many studies were employed forward validation which is a special form of cross-validation and is considered as the easiest method to choose optimal knots (Aldrin and Haff, 2005; Belušić, Herceg-Bulić and Bencetić Klaić, 2015). So, in this work, forward validation for each pollutant was based on hourly predictions of concentrations for Tunis, one day in advance. For each day and for the maximum number of knots, the model was re-estimated using the data up to the day before. Then, the hourly $\log PM_{10}$ and $\log O_3$ concentrations for the next day are predicted. The prediction is compared to the logarithm of the observed value and the hourly prediction errors calculated. For each day and for each of the two pollutants, this procedure was repeated. The root mean square (RMSE) of the prediction was finally calculated (RMSE definition in Appendix A). The minimum RMSE for each pollutant corresponded to $k = 15$ for (Temperature (TT°), nitrogen oxides ($NO_x \text{ } \mu\text{g.m}^{-3}$)) and $k = 10$ for (relative humidity (RH %), solar radiation ($SR \text{ W.m}^{-2}$)). The value of $k = 8$ was large enough only for wind variables.

Results and discussion

Based on the data described in Section "Site description and sample collection", the additive model with all variables was estimated for the two pollution variables PM_{10} and O_3 recorded at three different stations in Tunis.

The first two columns of Table 2 show the explained variation (squared correlation coefficients R^2) for the entire model (equation 4). The second part of the table presents the explained variation for meteorological variables only ($R^2_{m.v}$) which measured the aggregate impacts of local meteorology on each pollutant. $R^2_{m.v}$ corresponds to the explained variation of a new model given by the difference of the models with only time variables and with all variables. The highest values of R^2 were obtained for O_3 at Bab Aliwa station. We found that the explained variance for the entire model is between 0.56 and 0.85, indicating that the models explain most of the variation in pollutant concentrations, but a considerable amount of variation is still unexplained. The aggregate impact of meteorological variables was measured between 0.21 and 0.42.

Table 2: The second and third columns present the squared correlation coefficient (R^2) for each pollutant concentration modelled on log-scale with all variables (the final model). The fourth and fifth columns (R^2 m.v) show the squared correlation coefficient for only meteorological variables for each model on log-scale

Measurement site	R^2		R^2 m.v	
	PM ₁₀	O ₃	PM ₁₀	O ₃
Gazela	0.58	0.72	0.40	0.42
Mannouba	0.56	0.73	0.21	0.36
Bab Aliwa	0.59	0.85	0.29	0.41

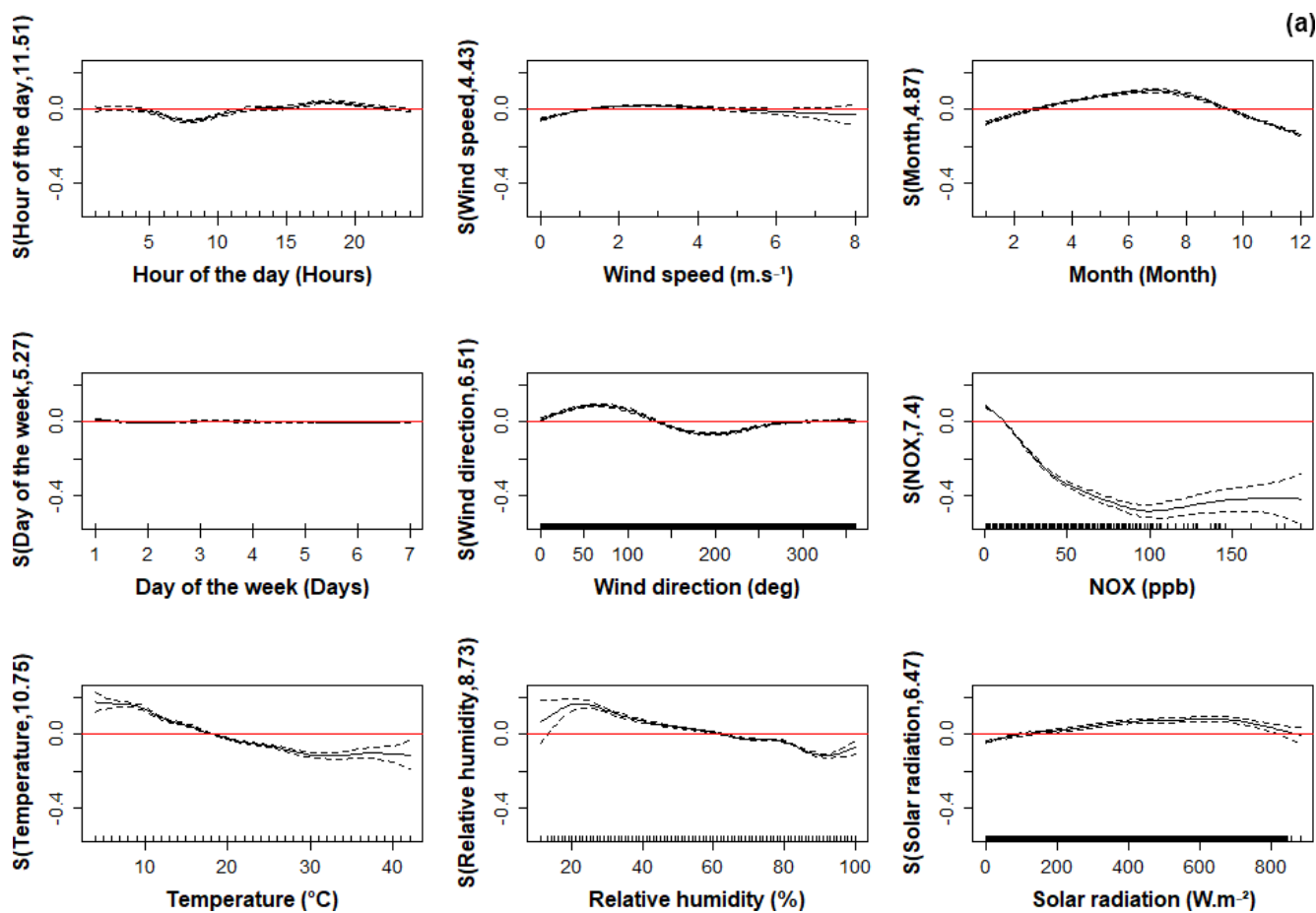


Figure 5a: GAM estimated relationships for temporal, meteorological and traffic variables on O₃ concentration for Gazela. The x-axis represents increasing variations. The y-axis indicates the contribution of the smoother to the fitted values. The region between the dashed lines represents the 95% confidence interval.

Ozone

Tropospheric ozone is considered a secondary pollutant which is formed by photochemical reactions involving the oxides of nitrogen NO and NO₂ (summed as NO_x), hydrocarbons and sunlight, particularly ultraviolet light. In urban areas, high ozone levels are observed during warm summer months when the temperature is high and the wind velocity is low. In Tunis, we found that the final model explained 85% (site of Bab Aliwa) of the variance of log-transformed O₃ concentrations (Table 2). The aggregate impact of meteorological variables explained 41% of the variance in O₃ for the same site (Bab Aliwa). The estimated effects of meteorological and temporal variables on O₃ are

shown in Fig. 5 (a), (b) and (c) for three stations in Tunis. Most meteorological, traffic and temporal factors were statistically significant in a highly non-linear way.

The influence of local meteorology on O₃

Temperature effect

For all three measurement stations, temperature (TT) was an important meteorological variable for O₃. The effect of temperature on O₃ is similar at Gazela and Bab Aliwa sites. A positive effect is seen for temperatures ranging between 5°C-20°C across only these two sites. A negative effect is noted

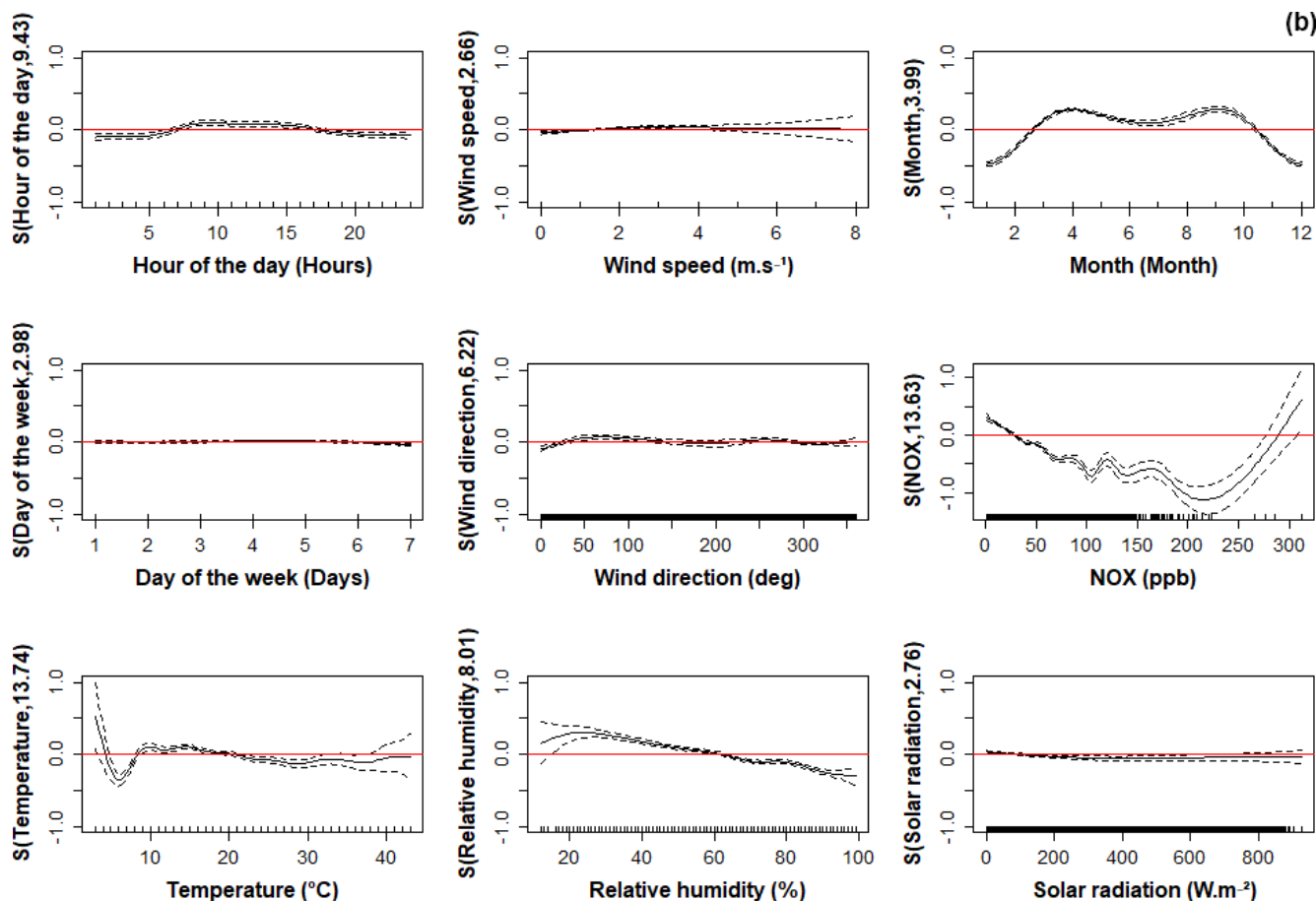


Figure 5b: GAM estimated relationships for temporal, meteorological and traffic variables on O_3 concentration for Mannouba. The x-axis represents increasing variations. The y-axis indicates the contribution of the smoother to the fitted values. The region between the dashed lines represents the 95% confidence interval.

for temperatures ranging between 20°C and 40°C for all three sites. So, if temperature increases, ozone concentrations are seen to decrease. This disagrees with common understanding of this relationship (Cheng et al., 2007; Polinsky and Shavell, 2010; Pearce et al., 2011; Ma et al., 2020), but can be due to correlations of temperature with other variables like wind direction. The formation and concentration of ground level O_3 depends on the concentrations of NO_x and VOCs, and the ratio of NO_x and VOCs. Ozone levels do not always increase with increases in temperature, such as when the ratio of VOCs to NO_x is low. As study area was surrounded by reliefs, the speeds of surface winds are low. It may be more thermal breezes than synoptic-scale winds (Melki, 2007). The high frequency of thermal breezes and calm periods may indicate stable atmospheric conditions and thus O_3 concentrations are higher during such episodes.

Wind effect

The curves in the center of Fig. 5 (a), (b) and (c) show the results obtained regarding the impact of wind direction. The estimated response for the wind direction is different for the various locations. This is as normal, since the effect of wind direction is strongly correlated on the emission locations. A non-linear relationship is observed for all stations: edf=6.51, edf=6.22 and edf=6.15 at Gazela, Mannouba and Bab Aliwa, respectively (Table

3). At the first site, O_3 exhibits maximum concentration for E-NE wind (70°-100°) and minimum concentration at around 200°. However, by examining the wind speed-direction frequencies graph of this site (Fig. 6), there is a very remarkable effect of this variable on ozone concentration. A possible explanation is the location of this measuring site which is subject to northern European pollution (i.e. O_3 is transported from Italy to Tunis). While crossing the city towards Mannouba site, the effect of wind decreases. In this station, O_3 shows secondary maxima for S-W wind (250°). The wind direction at the Bab Aliwa site seems to have a different effect on O_3 concentration. Wind direction has a positive effect on O_3 concentration for directions between 100° and 250°. This is probably associated with the cemetery effect which promotes ozone's transport. A light minimum is then observed at 270°. The effect of road traffic can explain this. In this study, increasing wind speed was found to correspond to increasing O_3 concentrations. This tendency is particularly marked for the Bab Aliwa station (Figure 5c). This agrees with previous findings of Melki, (2007). At the Gazela site, the effect of this variable is very local, so, difficult to explain. It may be possible to understand this effect on a scale larger than a city.

Solar radiation and relative humidity effects

Solar radiation had a non-linear association: edf=6.47, edf=2.75

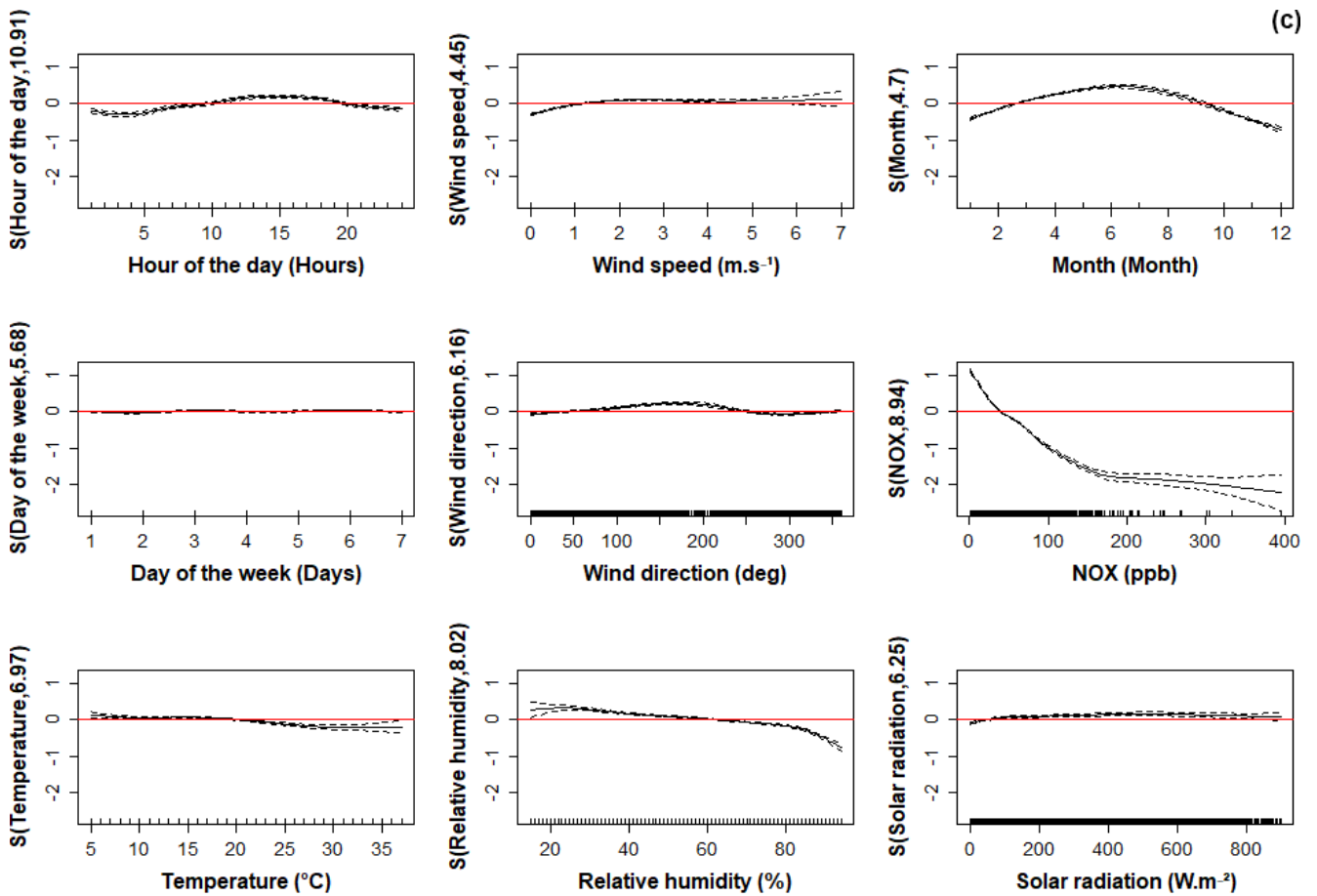


Figure 5c: GAM estimated relationships for temporal, meteorological and traffic variables on O_3 concentration for Bab Aliwa. The x-axis represents increasing variations. The y-axis indicates the contribution of the smoother to the fitted values. The region between the dashed lines represents the 95% confidence interval.

Table 3: Model estimates of the effects of predictors on O_3 (all sites). edf=effective degrees of freedom of the smooth function terms (edf>1 indicate non-linear relationships); F value is an approximate F-test, SE=asymptotic standard error. *** Significant at the 0.000 level

	Gazela Site		Mannouba Site		Bab Aliwa Site	
Smooth terms	edf	F	edf	F	edf	F
s(Hour of the Day)	11.5	30.00***	9.43	4.93***	10.91	14.20***
s(Day of the Week)	5.27	3.81**	2.97	3.27***	5.68	4.80**
s(Temperature)	10.75	66.02***	13.74	16.27***	7.01	9.51***
s(Wind Speed)	4.42	59.11***	2.66	6.23	4.45	77.38***
s(Wind Direction)	6.51	118.14***	6.22	9.07***	6.15	35.89***
s(Relative Humidity)	8.72	106.6***	8.00	33.34***	8.02	63.76***
s(Month)	4.87	436.78***	3.98	442.03***	4.70	535.48***
s(NO_x)	7.40	588.97***	13.63	72.83***	8.94	476.36***
s(Solar Radiation)	6.47	33.25***	2.75	3.94	6.25	8.42***
Linear terms	Estimate	SE	Estimate	SE	Estimate	SE
Intercept	4.2	0.001	3.94	0.004	2.95	0.004
Explained Deviance	73%	-	61.5%	-	85.6%	-
GCV score	0.01	-	0.12	-	0.08	-

and $edf= 6.25$ at Gazela, Mannouba and Bab Aliwa, respectively, (Table 3) with O_3 concentrations. These results are very clear, higher solar radiation corresponds to higher concentrations of O_3 . This positive effect was found to be strongest after values surpassed $400 W.m^{-2}$ (Gazela and Bab Aliwa station). This relationship is consistent with the literature (Pearce et al., 2011) as radiation plays a significant role in photochemistry of ozone production (Dawson, Adams and Pandis, 2007). The nature of response of O_3 to the RH showed a 10% under low RH, and then exhibited a modest negative relationship where high levels resulted in a regional decrease of up to 10% for Gazela and Mannouba, and 5% for Bab Aliwa. So, the curves go downward for increasing humidity. Generally, the results obtained in this analysis of meteorological parameters were expected, i.e. that higher ozone concentrations were associated with high temperature, low relative humidity and prolonged sunshine (Lacour et al., 2006). In this coastal region of the northern Mediterranean, at night the relative humidity of the air is important (96% on average), combined with a decline in temperature ($18^{\circ}C$ on average). This conjunction will reduce O_3 concentrations.

The impact of time and traffic variables on O_3

The upper left panel of Fig. 5 (a), (b) and (c) show how the concentrations of O_3 varies as the hour of the day (HD) changes. Each curve corresponds to one of the measurements stations. Since this variable describes the diurnal variation of O_3 in three locations, different curves are observed. The diurnal variation for Mannouba and Bab Aliwa sites shows a similar pattern with O_3 concentrations reaching the peak at around 9:00 at the Mannouba site and at around 14:00 at the Bab Aliwa site. The increase in O_3 concentrations during day time is due to the increase in solar radiation, which powers the photochemical reactions and consequently O_3 concentration (Khoder, 2009). The hour's period of negative effect is presumably due to high emissions of NO_x caused by the intensity of traffic. Monks et al. (2015) highlighted the non-linearity of the O_3 -VOC- NO_x system. VOC-limited refers to the fact that the production of O_3 is limited by the input of VOC. Indeed, high NO_x lead to lower O_3 because O_3 directly react with NO. The local production of ozone is less reduced because the NO_x react with hydroxyl radical species formed in the atmosphere. When these hydroxyl radicals do not react with NO_x (example: low emission of NO_x), they

contribute to the VOC degradation and the ozone production. Unlike other sites, Gazela is considered a residential site which is characterized by the domination of NO_x emissions (at this site, VOCs are only due to traffic, and not as much emitted as by factories like the other sites). In fact, a minimum of O_3 concentration is observed at around 8:00 and a maximum at around 19:00 when traffic is an important source of emissions and the vertical mixing is reduced. Influenced by transport of O_3 from other regions and local NO_x concentrations at night, the increase of the surface O_3 concentration during the night time was larger than that during the daytime (Lei and Wang, 2014). Day of the week at Gazela and Mannouba (Table 3) was found to have little influence on ozone, ($F=3.81, F=3.27$. respectively). For Monday to Wednesday the ozone concentrations remain more or less unchanged (Figure 5). The rise in ozone concentrations is observed on Thursday and Friday but is followed by a drop as of Saturday. This continues on Sunday when the levels of ozone then join those on Monday. This result was also found by Pont and Fontan (2000) for five large French cities: This study does not show any significant variation in ozone concentrations between weekend and week except for the strongest values where a 40% reduction in precursors would lead to a 20% increase in ozone. The weekend effect would be reversed. Due to constant of road traffic during all the days of the week in Bab Aliwa, no effect of the variable DW was observed. NO_x also has a non-linear association with O_3 concentration, with $edf=7.40$ and $edf=8.94$ at Gazela and Bab Aliwa, respectively (Table 3). Increased NO_x for these two sites was found to have a negative effect on O_3 . This finding is in agreement with other work since the chemical coupling of O_3 and NO_x make levels of O_3 inextricably linked: Ozone production is dependent on the state of NO_x , as NO_2 and NO increase the production and dissociation of O_3 , respectively. Consequently, an increased NO/ NO_2 ratio reduces the ozone concentration (Melkonyan and Kuttler, 2012). Analysis the results of Mannouba station reveals a different NO_x effect, when the NO_x concentrations is over 200 ppb, an increase of NO_x concentrations leads to a lower decrease of O_3 concentrations than at the other stations. An increase in O_3 concentrations is seen above 280 ppb of NO_x concentrations. This is presumably due to the location of this station, which includes small forests in the west and chemical plants in the south which promote VOCs emissions, then the increase of both O_3 and NO_x concentrations. A positive effect is detected for the

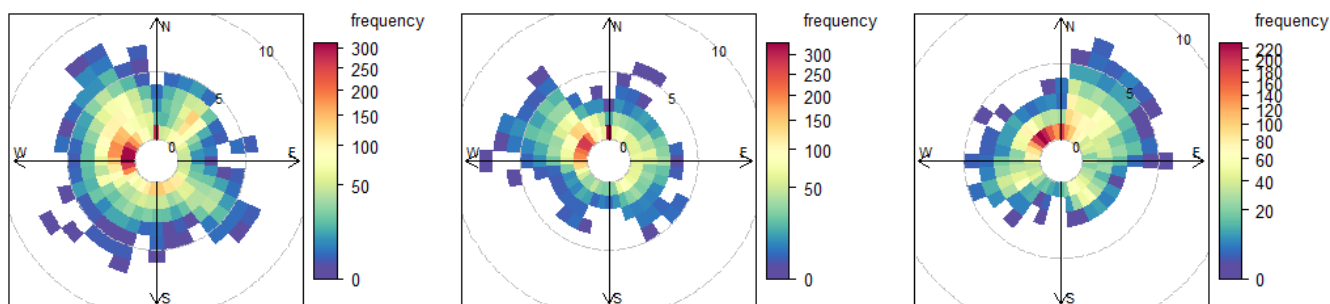


Figure 6: Wind speed-direction frequencies for three Meteorological Stations (from left to the right) Gazela, Mannouba and Bab Aliwa. Each cell gives the total number of hours the wind was from that wind speed/direction (period of 2008-2009). The number of hours is coded as a color scale shown to the right. The dashed circular grey lines show the wind speed scale.

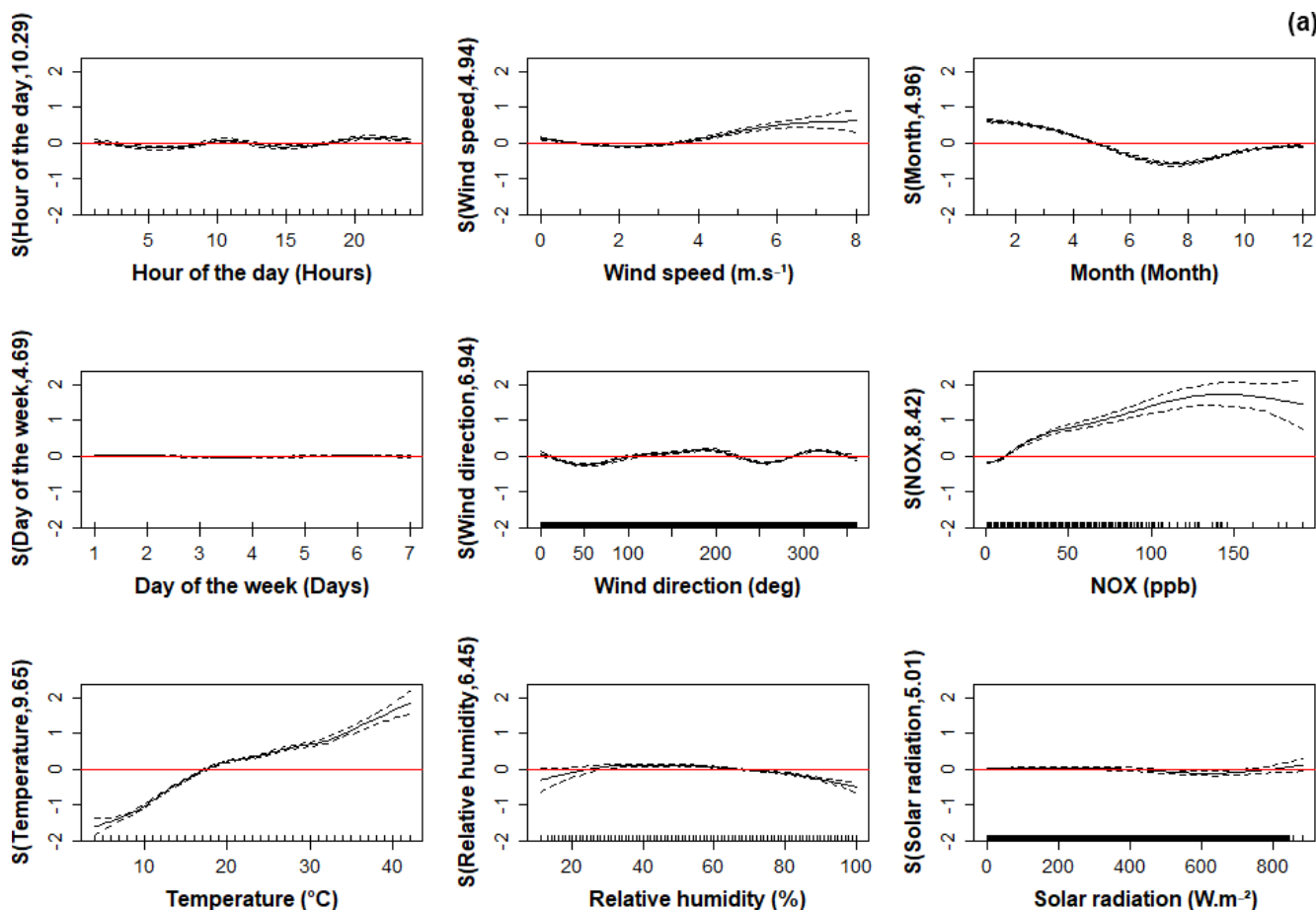


Figure 7a: GAM estimated relationships for temporal, meteorological and traffic variables on PM_{10} concentration for Gazela. The x-axis represents increasing variations. The y-axis indicates the contribution of the smoother to the fitted values. The region between the dashed lines represents the 95% confidence interval.

variable Month on O_3 concentration in warm months (spring and summer). In this period, there is an increase in temperatures and in the intensity of solar radiation. These meteorological conditions promote the mixing process of pollutants and O_3 formation. The ozone evolution is controlled not only by the influence of climate but also by the movement of pollutants. In fact, the same result was found in two regions: Spain and Italy which belong to the Mediterranean climate (Domínguez-López et al., 2014; Myriokefalitakis et al., 2016).

PM_{10}

The impact of traffic and site location on PM_{10}

Atmospheric PM_{10} are multicomponent aerosols. They originate from a variety of mobile, stationary and other natural sources, and are also formed in the atmosphere through chemical and physical processes. SO_2 (mainly issued from industrial sector) and NO_x (mainly issued from transport sector) are two precursors of secondary particulate matter (Harrison, Jones and Lawrence, 2004). Their chemical and physical compositions vary widely. Many studies showed that the PM_{10} yearly, daily and hourly average concentration exceeds the Tunisian and the European standard limits at all the sampling stations (Bouchlaghem et al., 2009). A significant proportion of PM_{10} in Tunis has many sources like sea salt, mineral dust (Calzolari et al., 2015). In the

Mediterranean Tunisian regions, the average seasonal evolution of PM_{10} is characterized by a winter maximum (November and December) (Bouchlaghem and Nsom, 2012). On the other hand, ozone concentration reaches its maximum values during summer period under the great photochemical activity and the effect of land-sea breeze. This difference has been highlighted in many studies and has been explained by the formation of PM_{10} as a complex mixture of many chemical species. Indeed, both the proximity to traffic sources and the different types of air mass scenarios make PM_{10} formation rather complex and associated with geographic, temporal and meteorological conditions. In Tunis, we found that the final model explained between 56% and 59% of the variance of log-transformed PM_{10} . The highest value of R^2 was found at Bab Aliwa station and the aggregate impact of meteorological variables accounting for 29%. The estimated effects of independent variables of the model are shown in Fig. 7 (a), (b) and (c) for three stations in Tunis. The model shows how the association of PM_{10} concentrations varies with the levels of other variables. The association between NO_x concentrations and PM_{10} concentrations was non-linear with $edf=8.41, edf=8.48$ and $edf=8.12$ at Gazela, Mannouba and Bab Aliwa respectively (Table 4) and is characterized by a general positive effect. It is reasonable and also found in Munir et al. (2013). Actually both NO_x and PM_{10} are largely issued from road traffic. The curve for Bab Aliwa is the one going farthest to the

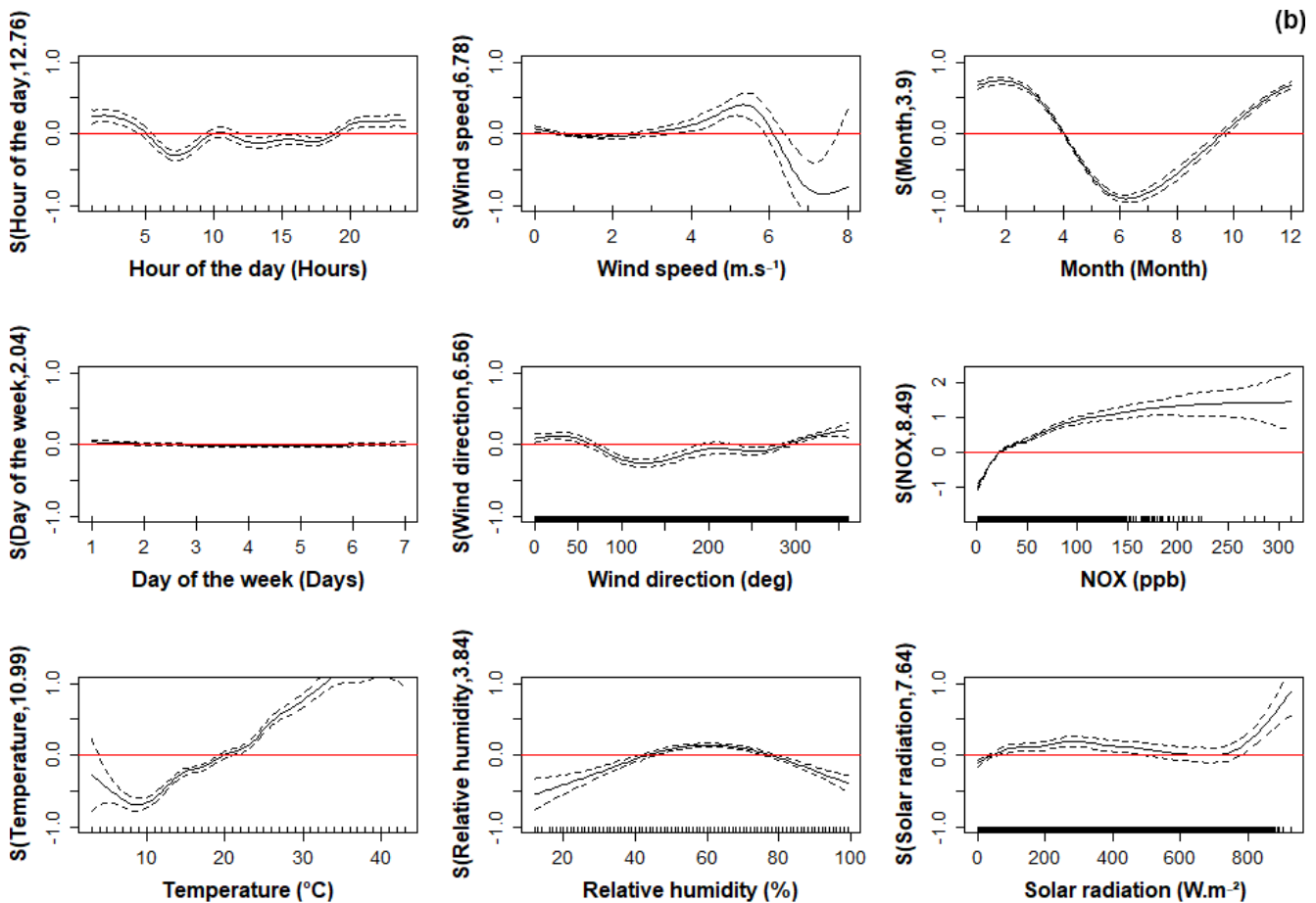


Figure 7b: GAM estimated relationships for temporal, meteorological and traffic variables on PM_{10} concentration for Mannouba. The x-axis represents increasing variations. The y-axis indicates the contribution of the smoother to the fitted values. The region between the dashed lines represents the 95% confidence interval.

right meaning that it is the location where the highest number of vehicles was observed. This might be logically explained by the fact that in this location, we found the biggest bus station and the most popular cemetery in the country. SO_2 and NO_x are the two sources of secondary particulate matter and have mostly a positive effect on PM_{10} (Harrison, Jones and Lawrence, 2004). NO_x concentration in Gazela station may be affected by Tunis airport located in the South east of the station.

The influence of local meteorology on PM_{10}

A non-linear association was observed between PM_{10} and wind speed. This variable has a positive effect on PM_{10} concentration from 4 $m.s^{-1}$ to 8 $m.s^{-1}$ at Gazela site. The curves for Mannouba and Bab Aliwa (Fig. 7 (b) and Fig. 7 (c)) reached the peak at 5 $m.s^{-1}$ then decrease. The same wind behavior was observed in three sites and was found in Belušić, Herceg-Bulić and Bencetić Klaić (2015): For large wind speeds, PM_{10} concentration decrease. This result was as expected as low wind and stable atmospheric conditions support higher concentrations of PM_{10} . We note however that the decrease in PM_{10} levels at higher winds observed in the present study is in contrast to the result found in Makkah by Munir et al., (2013) and in Maribor by Lešnik, Mongus and Jesenko (2019). Wind direction had variable association with PM_{10} : $edf=6.88$ at Bab Aliwa site (Table 4). Several curves

were observed for different sites. In the first station, Gazela, (center of Fig. 7 (a)), PM_{10} exhibit a first maximum concentration for wind direction around 170° . This can be explained by localized effect of the road. The secondary maximum is observed around 320° , clearly reflecting the effect the small factory situated north of the study area. As Bab Aliwa is based next to taxi and bus stations, this particular measuring site is subject to PM_{10} transport by southeast winds. For relative humidity, the results are very clear especially for Gazela and Mannouba sites, which find that high humidity was associated to low PM_{10} concentration. So, the curves go downward for humidity better than 80%. This agrees with previous findings of Aldrin and Haff (2005) and Belušić, Herceg-Bulić and Bencetić Klaić (2015). Particles are then removed from contaminated surface air by wet deposition in precipitation added to dry deposition (Giri, Murthy and Adhikary, 2008). The estimate curves of temperature have the same slope for the various locations. Temperature was named as the most significant meteorological variable for Bab Aliwa ($F=179.51$, p -value <0.001) and Gazela ($F=175.75$, p -value <0.001) sites. Interpretation of the curves (lower left of Fig. 7 (a), (b) and (c)) can be expressed as follows: increasing temperature corresponds with increasing PM_{10} with a notable positive effect for temperature above $20^\circ C$. It's important to note that this finding agrees the result from PM_{10} studies (Bouchlaghem

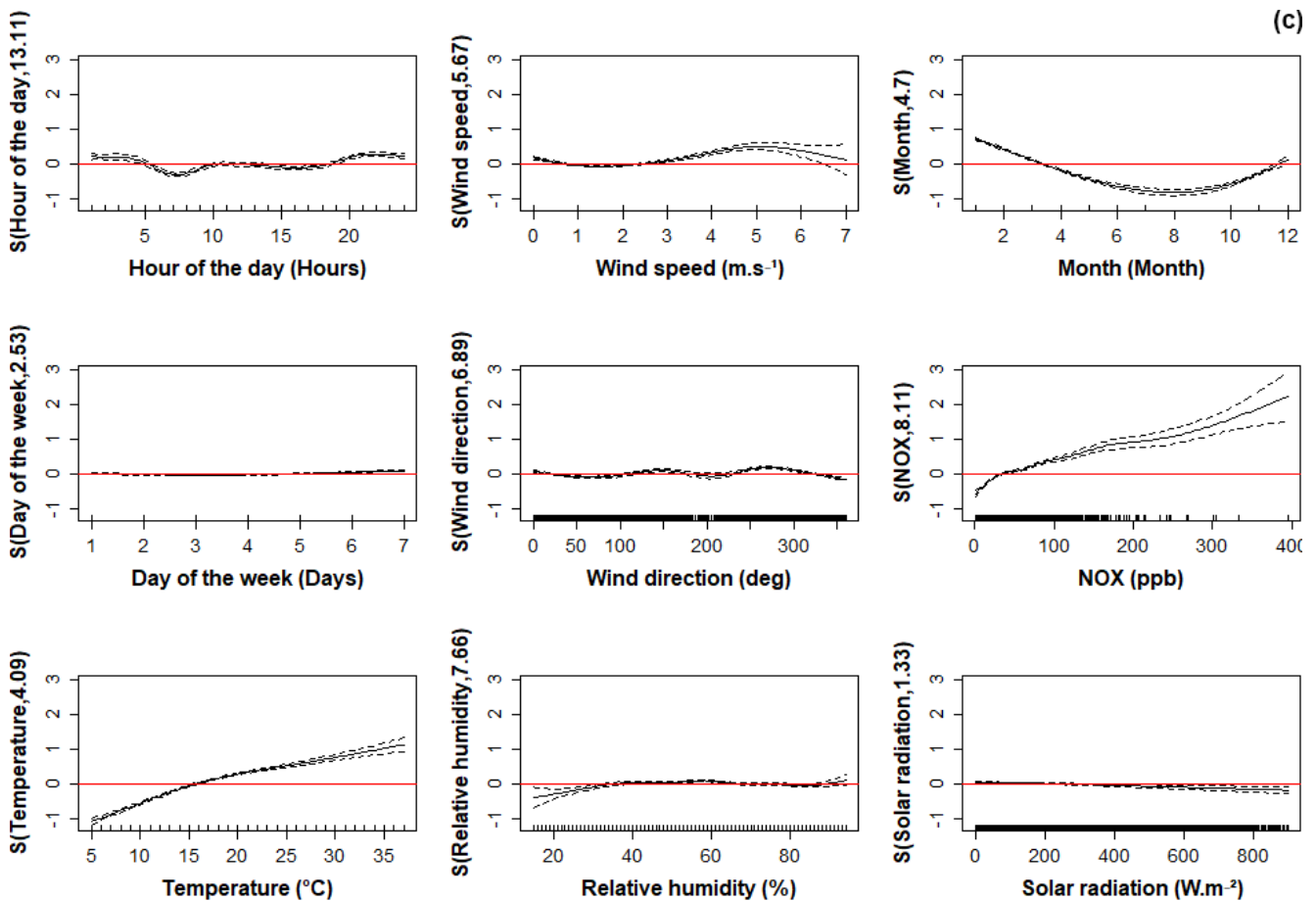


Figure 7c: GAM estimated relationships for temporal, meteorological and traffic variables on PM_{10} concentration for Bab Aliwa. The x-axis represents increasing variations. The y-axis indicates the contribution of the smoother to the fitted values. The region between the dashed lines represents the 95% confidence interval.

Table 4: Model estimates of the effects of predictors on PM_{10} (all sites). *edf*=effective degrees of freedom of the smooth function terms (*edf*>1 indicate non-linear relationships); *F* value is an approximate *F*-test, *SE*=asymptotic standard error. *** Significant at the 0.000 level

	Gazela Site		Mannouba Site		Bab Aliwa Site	
Smooth terms	<i>edf</i>	<i>F</i>	<i>edf</i>	<i>F</i>	<i>edf</i>	<i>F</i>
s(Hour of the Day)	10.29	18.56***	12.75	11.91***	13.10	25.12***
s(Day of the Week)	4.68	2.44*	2.03	2.31	2.53	14.90***
s(Temperature)	9.64	175.75***	10.98	31.93***	4.09	179.51***
s(Wind Speed)	4.93	54.24***	6.78	9.86***	5.66	44.19***
s(Wind Direction)	6.93	77.03***	6.56	22.76***	6.88	20.84***
s(Relative Humidity)	6.44	31.08***	3.84	40.03***	7.66	7.21***
s(Month)	4.95	333.96***	3.89	353.675***	4.70	326.94***
s(NO_x)	8.41	137.12***	8.48	120.59***	8.12	51.35***
s(Solar Radiation)	5.00	4.76***	7.64	9.27***	1.31	6.54**
Linear terms	Estimate	SE	Estimate	SE	Estimate	SE
Intercept	3.72	0.005	3.9	0.007	4.26	0.006
Explained Deviance	58.5%	-	54.5%	-	60.1%	-
GCV score	0.27	-	0.36	-	0.18	-

and Nsom, 2012). However, the positive relationship between temperature and PM₁₀ is probably explained by the dust layer created over three sites especially during peak hours.

The impact of time variables on PM₁₀

The time variable hour of the day (HD) has a non-linear association with PM₁₀ concentration. It was mainly used to account the effect of traffic. At the study stations, PM₁₀ concentration fall to a minimum between 7:00-8:00 and increase until 10:00, this corresponds to the morning peak traffic flow. In Bab Aliwa site, an evening peak traffic flow was noted at around 21:00. This second peak is probably due to people’s daily commuting between the capital and the suburbs. Curves of partial effect of the variable Month pointed out that in all measuring sites, PM₁₀ is characterized by a winter maximum (December-January-February). This result is consistent with the data of Bouchlaghem and Nsom (2012), who found a winter PM₁₀ peak in five different stations (traffic, industrial and residential) in Tunisia. This is presumably due to the influence of low mixing in the atmosphere and the advection of Saharan plumes. We note the absence of the second peak observed during the summer in the previous works (Bouchlaghem and Nsom, 2012). The slight effect of Saharan dust can be explained by the temporal difference between the South and the North of Tunisia and the geographical locations of the monitoring stations far from the southwest origin of the Saharan event. Since the Mannouba station is placed close to agriculture fields, plowing during the autumn season (September-October) promotes increasing PM₁₀ concentrations.

Assessment of the model performance

Table 5: Statistical evaluation of the model for all pollutants at Gazela site for the entire study period

	O ₃ (ppb)	PM ₁₀ (µg.m ⁻³)
IOA	0.91	0.77
RMSE	6.46	38.21
Modified RMSE	6.46	38.21
Measurement standard deviation	12.11	51.81
Model standard deviation	10.21	34.82
Measurement mean	67.89	56.50
Model mean	67.89	56.50

Various metrics (RMSE, modified RMSE, measurement standard deviation, model standard deviation and IOA (see Appendix A)) were used to assess the model performance. This statistical evaluation of the model on the original scale is presented in

Table 5 is for all variables at Gazela site; other pollutants and measuring site data are not shown here as the results are similar to these. The first criterion for model evaluation was checked

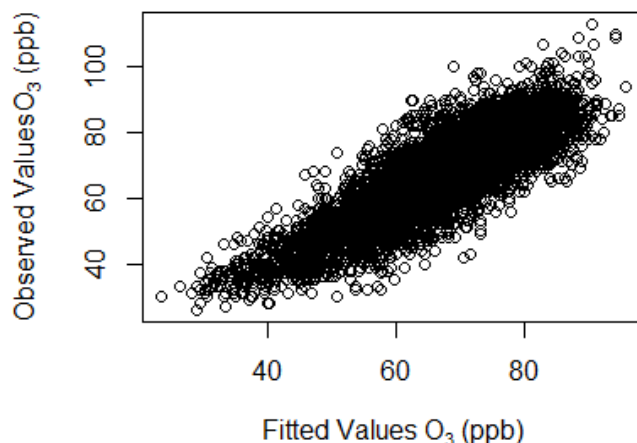


Figure 8: Plot of response against fitted values O₃ concentrations at Gazela shows a positive linear relationship with a good deal of scatter

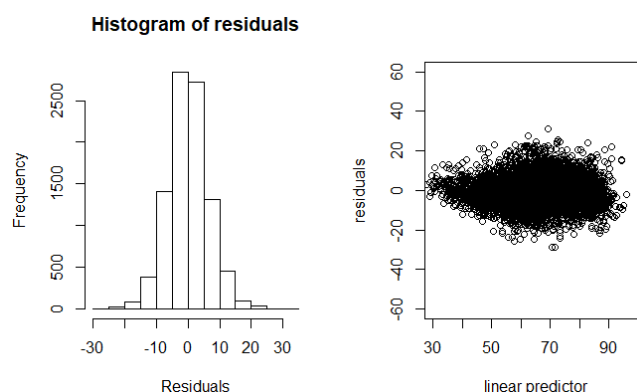


Figure 9: Residual plots for O₃ (ppb) at Gazela for the period 2008–2009. Left: histogram of residuals, exhibiting a normal distribution Right: the relationship between residuals and fitted values. The majority of residuals group around zero, as expected. The x-axis range on the left-hand plot and the y-axis range on the right-hand plot are the same.

and both RMSE and Modified RMSE are less than measurement standard deviation. In addition, the index of agreement is 0.91 and 0.77 for O₃ and PM₁₀, respectively, which corresponds to a good compromise between modeled and measured values. Fig. 8 shows the relationship between the response and fitted values of O₃ concentration at Gazela site. PM₁₀ and other measuring site data are not shown as they are similar to those presented in this figure. This figure shows a positive linear relationship with a good deal of scattering. Residual plots are also used to characterize model efficacy. Fig. 9 clearly shows that the majority of residuals group around zero, as expected. The right-hand scatter plot which describes the relationship between residuals and fitted values suggest that variance is approximately constant as the mean increases. The left-hand plot, the residual histogram, exhibits a normal distribution for O₃ at Gazela.

Conclusions

The objective of this work was to estimate the relationship between each of two pollution variables, namely concentrations of PM₁₀ and tropospheric ozone O₃ and NO_x concentrations (taking as a proxy of traffic) as well as a set of meteorological variables for the urban area of Tunis. To achieve this objective,

a statistical methodology is used based on the Generalized Additive Model (GAM). We have shown that the GAM can model the non-linear effect of the covariates. The model is additive on the log scale and the estimates were made on hourly data collected during two years at three different locations in Tunis. The model provides a reasonably good fit in terms of the explained variance. For all stations, O_3 was easier to model (i.e. with more explanatory power and higher values of R^2). The most significant important variables for O_3 are NO_x , wind direction and relative humidity. The impact of temperature and NO_x is the strongest for PM_{10} , followed by relative humidity and wind variables. The time variables (hour of the day, day of the week and month) appear to have a particular impact on air quality. In this study, the variable Month plays a significant role in the characterization of the study area as a function of time. In fact, we note the seasonal behavior of O_3 and PM_{10} pollutants, with the highest concentrations in summer and winter, respectively. These results allow a first and fast analysis of the air pollution due to O_3 and PM_{10} in 3 locations in Tunis. It emphasizes the critical role of the local conditions on the air pollution, and especially the emissions and the weather as two main drivers of urban air pollution. Our findings suggest focusing on model improvement as future work. The addition of precipitation and traffic density (number of vehicles) variables could help to improve the model assessment. So, it is necessary to take into account all the sources of emissions exhaustively. In summary, the use of GAM in combination with partial residual plots offered an effective way to outline the relationships between temporal, meteorological and traffic variables and air pollution. Although our study did not detail chemical and physical aspects of air pollution, the results produced were reasonable and comparable to other studies. Furthermore, the results may be considered as relevant because research work on air pollution is insufficient in Tunisia. To this end, after quantifying the influence of all used variables, we plan to use GAM and GAMM (Hastie and Tibshirani, 1990; Wood, 2006) models to forecast pollutant concentrations.

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Appendix A

edf: The effective degrees of freedom (edf) estimated from generalized additive models were used as a proxy for the degree of non-linearity in stressor-response relationships. An edf of 1 is equivalent to a linear relationship, an edf > 1 and ≤ 2 is a weakly non-linear relationship, and an edf > 2 indicates a highly non-linear relationship.

GCV: generalized cross validation score can be taken as an estimate of the mean square prediction error based on a leave-one-out cross validation estimation process. We estimate the model for all observations except *i*, then note the squared

residual predicting observation *i* from the model. Then we do this for all observations. GCV criteria is numerically stable and efficient, but its computation become extensive especially when several smoothing parameters have to be estimated.

F-statistic: An F statistic is a value you get when you run an ANOVA test or a regression analysis to find out if the means between two populations are significantly different. In regression case, the F value is the result of a test where the null hypothesis is that *all of the regression coefficients are equal to zero*. In other words, the model has no predictive capability. Basically, the f-test compares your model with zero predictor variables (the intercept only model), and decides whether your added coefficients improved the model.

Asymptotic Standard Error: Asymptotic standard error is an approximation to the standard error, based upon some mathematical simplification. In regression analysis, the term "standard error" refers either to the square root of the reduced chi-squared statistic, or the standard error for a particular regression coefficient (as used in, say, confidence intervals).

VIF: Variance Inflation Factor detects multicollinearity in regression analysis. For an independent variable X_i , it can be calculated by the formula below using R-squared values:

$$VIF_i = \frac{1}{1 - R_i^2}$$

IOA: Index of Agreement is a standardized measure of the degree of model prediction error which varies between 0 and 1. IOA=1 represents full agreement and IOA=0 indicates no agreement at all.

$$IOA = 1 - \frac{\sum_{i=1}^n (O_i - P_i)^2}{\sum_{i=1}^n (|P_i - \bar{O}| + |O_i - \bar{O}|)^2}$$

RMSE: The Root Mean Square Error is used to measure the difference between values predicted and values observed.

$$RMSE = \sqrt{\frac{1}{n} \sum_{i=1}^n (O_i - P_i)^2}$$

$$Modified\ RMSE = \sqrt{\frac{1}{n} \sum_{i=1}^n (O_i - \bar{O} - P_i + \bar{P})^2}$$

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Research article

Does apparent temperature modify the effects of air pollution on respiratory disease hospital admissions in an industrial area of South Africa?

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Received: 8 June 2021 - Reviewed: 16 July 2021 - Accepted: 13 September 2021

<https://doi.org/10.17159/caj/2021/31/2.11366>

Abstract

Background: Temperature and air pollution are often treated as separate risk factors and very few studies investigated effect modification by temperature on air pollution, and the impact of this interaction on human health in Africa. This study therefore investigated the modifying effects of temperature on the association between air pollution and respiratory disease (RD) hospital admissions in South Africa.

Methods: RD admission data (ICD10 J00-J99) were obtained from two hospitals located in Secunda, South Africa between 1 January 2011 to 31 October 2016. Ambient NO₂, SO₂, PM₁₀, PM_{2.5}, temperature and relative humidity data were obtained from the South African Weather Services. A case-crossover epidemiological study design was applied and lag0-1 was used. Models were adjusted for public holidays and apparent temperature (Tapp). Days were classified as warm (Tapp>75th percentile), cold (Tapp<25th percentile) and normal (Tapp 25th-75th percentile).

Results: Of the 14 568 RD admissions, approximately an equal number of females and males were admitted. The average daily NO₂, SO₂, PM_{2.5} and PM₁₀ levels were 12.4 µg/m³, 8.5 µg/m³, 32.3 µg/m³ and 68.6 µg/m³, respectively. Overall, a 10 µg/m³ increase in SO₂ on warm days was associated with an increase in RD hospital admissions: 8.5% (95% Conf. Int: 0.4%, 17.2%) and 8.4% (95% Conf. Int: 0.3%, 17.1%) after adjustment for PM_{2.5} and PM₁₀, respectively. However, increasing PM_{2.5} or PM₁₀ levels was associated with an increase in RD hospital admissions on normal days, after adjusting for SO₂. On cold days there were significant associations between the SO₂ and RD admissions among the 0-14 year age group, after adjusting for either PM_{2.5} (6.5%; 95% Conf.Int: 0.9%, 12.4%) or PM₁₀ (5.5%; 95% Conf.Int: 0.3%, 11.1%).

Conclusions: These results indicate that the risk of RD hospital admission due to ambient air pollution exposure is different on cold, normal and warm days in Secunda.

Keywords

Particulate matter, SO₂, apparent temperature, respiratory disease, hospital admissions, South Africa, case-crossover.

Introduction

Many epidemiological studies have demonstrated the independent effects of air pollution and temperature on health (Zhang et al., 2018; Chen et al., 2017; Wichmann, 2017). Rising temperature is one of the key climatic change indicators which affects human health directly and indirectly leading to deaths, illnesses and the aggravation of respiratory diseases (Wichmann, 2017). Cardiovascular diseases such as ischemic heart disease have been attributed to high temperature (Zacharias et al., 2014). Studies have shown that increase in temperature can lead to increased mortality (Li et al., 2013; Petkova et al., 2013). It has also been shown that both low temperature and high temperature can increase the risk of respiratory diseases (Michelozzi et al., 2009; Zhao et al., 2018; Su et al., 2014).

Air pollution also affects human health, especially the respiratory system which is usually the first point of contact in the human body (Dadbakhsh et al., 2015). In urban areas, anthropogenic emissions give rise to high levels of air pollution and the commonly found anthropogenic and natural air pollutants are SO₂, NO_x, O₃, volatile organic compounds and suspended particulate matter (PM) (Rahman, 2016; Norman et al., 2007).

Exposure to ambient levels of air pollution is an important determinant of emergency room visits and hospital admissions for acute and chronic respiratory symptoms (Szyzkowicz et al., 2018). There has been significant increase in ischemic heart disease deaths attributable to heat wave of 1.9-fold (685 deaths

per year) in 2021–2050, and 5.1-fold (1801 deaths per year) in 2069–2098 compared to baseline years of 2000–2010 (Zacharias et al., 2014). Increases from 1980 baseline in heat-related mortality of approximately 22.2, 49.4, and 91 % in 2020s, 2050s, and 2080s, respectively, using the A2 scenario (Li et al., 2013). Increase in heat-related mortality to six to nine times present rates in 2080s using RCP 8.5 (Petkova et al., 2013).

Respiratory morbidity in South Africa

In South Africa, evidence suggests that the prevalence of respiratory morbidity is increasing (Masekela et al., 2018). A study on the epidemiology of asthma in South Africa reports an approximate 5% increase in lifetime and 12-month wheeze amongst children and adolescents between 1995 and 2002 (Zar et al., 2007). The increase in respiratory symptoms was associated with deteriorating air quality (Naidoo et al., 2013).

In Durban, children living in industrial areas with higher levels of ambient air pollution have more asthma and hyper-reactive airways than children living further away from industrial areas (Naidoo et al., 2013).

Similarly, people living close to mine-dumps in South Africa have poorer respiratory health outcomes compared to people living further away (Nkosi et al., 2015; Nkosi et al., 2015b).

However, the modification effects of ambient temperature on the association between respiratory morbidity and air pollution in South Africa have not been explored.

An earlier study observed an association between daily ambient apparent temperature and daily all cause mortality between 2006 and 2010 with almost half a million deaths out of a population of about 12 million in Cape Town, Durban and Johannesburg (Wichmann, 2017).

Another study showed that there was a modification effect of temperature on air pollution associated with CVD hospital admissions in Cape Town (Lokotola et al., 2020).

It is therefore important to explore the modification effect of ambient temperature on the association between respiratory morbidity and air pollution in South Africa.

This is also important because the mean annual temperature in South Africa increased by at least 1 °C during the last 50 years which is 1.5 times the global average (Engelbrecht et al., 2015; Ziervogel G et al., 2014; MacKellar et al., 2014).

It has been projected that by 2100, warming will reach around 3–4°C along the South African coast, and 6–7°C inland, thus higher than the global average warming (Department of Environmental Affairs, 2010).

Many studies have reported on the interaction between ambient temperature and air pollution on respiratory morbidity (McCormack et al., 2016; Iranpour et al., 2020). However,

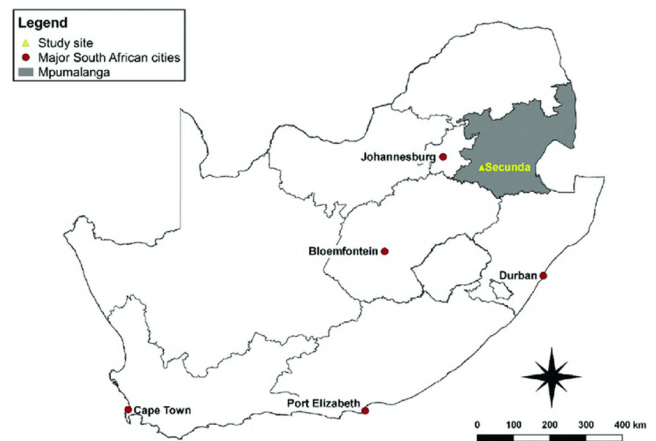


Figure 1: Map of South Africa showing the location of the study site, Secunda. Courtesy: Emslie et al. 2020

there is accumulating evidence that the warm/hot increasing temperature effects are enhanced by high pollution levels (especially PM₁₀ and ozone) and vice versa and that effects of pollutants are enhanced by the presence of high temperature (Analitis et al., 2018).

In Hefei, China, a study found synergy between PM₁₀ concentrations and temperature in their effects on mortality (Qin et al., 2017). Given the frequent simultaneous exposure to ambient temperature and air pollution, one can expect to see the synergistic effects of these factors in human physiology (Analitis et al., 2018). It has been shown that high temperature could modify the effects of air pollution on daily mortality and high air pollution might enhance the air temperature effects (Chen et al., 2018).

The knowledge of the modifying effects of temperature on the association between RD hospital admission and air pollution will help policy makers, and inform risk assessments (Kan et al., 2008). Therefore, we investigated the effects of apparent temperature on the association between air pollution and respiratory disease (RD) hospital admissions in Secunda, which is situated in the inland part of South Africa.

Secunda is located in the Highveld Priority Area (HPA), which was declared 14 years ago in 2007 as such to manage and address the poor air quality in the area (NEMA, 2004; <http://www.saaqis.org.za/documents/Highveld%20Priority%20Area%20Declaration.pdf>). Air quality in the HPA consistently exceeds national ambient air quality standards (NAAQS) due to both industrial and non-industrial sources (NEMA, 2004).

The Highveld area in South Africa is characterised by poor ambient air quality and elevated concentrations of criteria pollutants due to the concentration of industrial and non-industrial sources (Held et al., 1996).

Secunda was identified to be an air quality hotspot in the Highveld Priority Area Air Quality Management Plan due to frequent exceedances of the SO₂ standards.

The main emissions in Secunda are from the petrochemical industry and energy sector in the region. Others agriculture, domestic fuel burning, mining activities, veld fires, and power stations which are sources of $PM_{2.5}$, PM_{10} , SO_2 and NO_2 (SSI Environmental, 2013).

Secunda lies at the heart of South Africa's coal mining industry and still experiences high air pollution levels (NEMA, 2004). (Figure 1). Secunda produces the most polluting liquid fuels in the world through the Sasol's Synfuels facility (Myllyvirta, 2020). To improve air quality in the HPA, an Air Quality Management Plan (AQMP) was developed in accordance with the National Environmental Management Air Quality Act 2004.

Material and methods

Ethical approval (reference 132/2018) was obtained from the Research Ethics Committee, Faculty of Health Sciences, University of Pretoria in 2018.

Study design

In this study, a case-crossover epidemiology study design was used to explore the modifying effects of temperature on the association between major air pollutants, including sulphur dioxide (SO_2), nitrogen dioxide (NO_2) and particulate matter less than 10 microns in diameter (PM_{10}) and 2.5 microns in diameter ($PM_{2.5}$) and hospital admissions for RD in Secunda, South Africa. (Carracedo-Martínez et al., 2010).

This study design was developed as a variant of the case-control design to study the effects of transient exposures on emergency events, comparing each person's exposure in a time-period just prior to a case-defining event with the person's exposure at other times (Carracedo-Martínez et al., 2010). If the control days are chosen close to the event day, personal characteristics that vary slowly over a short time period of 24 hours are controlled for by matching (Carracedo-Martínez et al., 2010). Such characteristics may include co-morbidities (e.g. HIV status, hypertension, smoking status and so forth). Nevertheless, such characteristics may be potential effect modifiers, i.e. indicate susceptibility. However, information on such characteristics was not provided by the hospitals.

A time-stratified approach was applied to select the control days, defining the day of RD hospital admission as the case day and the same day of the week in the same month and year as control days (i.e. theoretically 3 to 4 control days per case day) (Carracedo-Martínez et al., 2010).

Hospital admission data

Individual-level RD hospital admission data (International Classification of Disease, 10th version [ICD-10] (J00–J99)) were obtained from two private hospitals in Secunda, after ethical approval. The two hospitals are from the same hospital group. Data were available electronically from 1 January 2011 to 31 October 2016.

Air pollution and weather data

Hourly $PM_{2.5}$, PM_{10} , NO_2 and SO_2 data from 2011–2016 were obtained from the South African Weather Services through the South African Air Quality Information Systems (SAAQIS) for the study period, after signing a data agreement. A network of air pollution monitors in Secunda continuously measures real-time concentrations of the criteria air pollutants using equivalent methods of the United States Environmental Protection Agency and in accordance with ISO 17025 guidelines (National Environmental Management: Air Quality Act, 2004).

Hourly temperature ($^{\circ}C$) and relative humidity (%) data were obtained from the South African Weather Service (SAWS) for the study period 1 January 2011 – 31 October 2016, after signing a data agreement. The Secunda monitoring station is 11.8km and 16.9km in relation to the two hospitals.

Apparent temperature

Models were adjusted for apparent temperature (Tapp) which reflects the physiological experience of combined exposure to humidity and temperature and thereby better captures the response on health than temperature (Steadman, 1984).

Saturation vapour pressure

$$= 6.112 \times 10^{(7.5 \times \text{temperature } ^{\circ}C / (237.7 + \text{temperature } ^{\circ}C))} \quad (1)$$

Actual vapour pressure

$$= (\text{relative humidity (\%)} \times \text{saturation vapour pressure}) / 100 \quad (2)$$

Dew point temperature $^{\circ}C$

$$= (-430.22 + 237.7 \times \ln(\text{actual vapour pressure})) / (-n(\text{actual vapour pressure}) + 19.08) \quad (3)$$

Apparent temperature $^{\circ}C$

$$= -2.653 + (0.994 \times \text{temperature } ^{\circ}C) + 0.0153 \times (\text{dew point temperature } ^{\circ}C) \quad (4)$$

Statistical analysis

Correlation between the air pollutants and Tapp were investigated using Spearman correlation analyses. Most studies on temperature as a modifier of the health effects of air pollution selected short lags, e.g. lag0 (same day of exposure as day of hospital admission), lag1 (day prior to day of hospital admission) or lag0-1 (mean of lag0 and lag1). The results in the present study will focus on lag0-1, as done in other studies (Li et al., 2017; Chen et al., 2017).

The association between the air pollutants and RD hospital admissions was investigated using conditional logistic regression models (R Development Core Team, 2019). Two pollutant models were investigated, which included $PM_{2.5}$ and SO_2 , $PM_{2.5}$ and NO_2 , PM_{10} and SO_2 and PM_{10} and NO_2 as the air pollutants were not strongly correlated with each other ($p < 0.05$). Models were adjusted for a public holiday variable (binary variable) and Tapp. The shape (i.e. linear or non-linear) of the association between the Tapp and RD hospital admissions

was investigated. First Tapp was included as a natural spline with 3 degrees of freedom (df) (non-linear term) in the models. Whether the non-linear term of Tapp improved the model was checked with log likelihood ratio tests, i.e. compared it to a model that included Tapp as a linear term. It was observed that the non-linear term of Tapp did not add value to the model and Tapp was then included as a linear term. Air pollutants were added as linear terms in the model, as done in many studies (Li et al., 2017; Chen et al., 2017).

The associations are presented as the percent excess risk in RD hospital admissions per 10µg.m⁻³ increase in an air pollutant level. This approach is commonly applied in other studies (Li et al., 2017; Chen et al., 2017). Susceptibility of age groups (<15 years, 15–64 years and ≥65 years) and sex (male/female) on warm and cold days was investigated in stratified analyses followed by models with interaction terms.

Intra-individual factors cannot be examined as effect modifiers due to the nature of the case-crossover design. However inter-individual variation using an interaction term between the effect modifier and an air pollutant in the conditional logistic regression model, can detect a p-value for interaction.

Stratified analyses were conducted to examine the interactive effects of temperature and air pollution on RD hospital admission. Temperature was divided into three levels –warm, normal and cold days. Warm and cold days were defined as days when Tapp was higher than the 75th percentile of Tapp of the study period and lower than the 25th percentile of Tapp, respectively. Normal days were those equal or higher than the 25th percentile of Tapp, but lower or equal to the 75th percentile of Tapp. Other studies have used a similar approach (Chen et al., 2017; Li et al., 2017; Chen et al., 2013).

Results

Descriptive statistics

Of the 14 568 RD hospital admissions in this study, 49.3% (n=7 179) were males and the highest number of patients admitted for RD in a day was 26 (Table 1a). The mean Tapp for the study period was 14.2°C, PM₁₀ peaked at 496.9 µg.m⁻³ and PM_{2.5} peaked at 262.4 µg.m⁻³ (Table 1b). During the study period, daily PM₁₀ and PM_{2.5} levels exceeded the daily WHO air quality guidelines on 721 (34%) and 1081 (51%) of the 2131 days, respectively. The daily WHO air quality guidelines for PM_{2.5} and PM₁₀ are 25 µg.m⁻³ and 50 µg.m⁻³, respectively. The annual PM₁₀ mean concentrations were above the annual WHO guideline (20 µg.m⁻³), except in 2015. However, the annual mean values of SO₂ and NO₂ in Secunda between 2011 and 2016 were significantly lower than their NAAQS and WHO annual means.

All air quality variables were positively correlated (Table 2). PM₁₀ and PM_{2.5} were strongly correlated (r=0.946, p < 0.05), whilst SO₂ and NO₂ were weakly correlated (r = 0.085, p < 0.05). Air quality variables were negatively correlated with temperature and relative humidity (r=-0.483 to -0.120) (Table 2).

Table 1a: Summary statistics of health outcomes in Secunda, 1 January 2011 – 31 October 2016 (2 131 days).

Variable	Mean	Min	25%	Median	75%	Max
Respiratory disease hospital admissions						
All ages and both sexes	6.8	0.0	3.0	6.0	10.0	26.0
Female patients (n=7 389)	3.5	0.0	1.0	3.0	5.0	17.0
Male patients (n=7 179)	3.4	0.0	1.0	3.0	5.0	15.0
0-14 year olds (n=6 915)	3.2	0.0	1.0	2.0	5.0	20.0
15-64 year olds (n=6 531)	3.1	0.0	1.0	2.0	4.0	16.0
≥65 year olds (n=1 122)	0.5	0.0	0.0	0.0	1.0	4.0

Table 1b: Summary statistics of air pollutants and meteorological conditions in Secunda, 1 January 2011 – 31 October 2016 (2 131 days).

Variable	Mean	Min	25%	Median	75%	Max
PM ₁₀ (µg.m ⁻³)	68.6	0.0	19.4	43.3	97.9	496.9
PM _{2.5} (µg.m ⁻³)	32.3	0.0	10.7	19.9	41.8	262.4
SO ₂ (µg.m ⁻³)	8.5	0.0	3.8	6.7	10.9	73.2
NO ₂ (µg.m ⁻³)	12.4	0.5	6.7	11.0	16.7	73.8
Tapp (0C)	14.2	-1.0	9.4	14.9	18.9	26.4
Temperature (0C)	15.6	1.7	11.7	16.5	19.3	26.5
Relative humidity (%)	58.5	16.6	47.3	60.3	70.3	94.1

Abbreviations: PM₁₀: particulate matter with an aerodynamic diameter of less than 10 µm; PM_{2.5}: particulate matter with an aerodynamic diameter of less than 2.5 µm; SO₂: sulphur dioxide; NO₂: nitrogen dioxide, Tapp: apparent temperature

Table 2: Spearman correlation coefficients between air pollution and weather variables.

Variable	PM _{2.5}	SO ₂	NO ₂	Tapp	Temp	RH
PM ₁₀	0.946	0.235	0.216	-0.446	-0.398	-0.483
PM _{2.5}		0.238	0.259	-0.507	-0.477	-0.406
SO ₂			0.085	-0.180	-0.176	-0.200
NO ₂				-0.263	-0.261	-0.120
Tapp					0.983	0.242
Temperature						0.106

Abbreviations: PM₁₀: particulate matter with an aerodynamic diameter of less than 10 µm; PM_{2.5}: particulate matter with an aerodynamic diameter of less than 2.5 µm; SO₂: sulphur dioxide; NO₂: nitrogen dioxide; Tapp: apparent temperature; RH: relative humidity. All correlations were significant (p < 0.05)

Exposure - response estimates

In the unstratified analysis (i.e. entire Tapp range), there was no association between any of the pollutants and RD hospital admission. In the stratified analysis, a $10 \mu\text{g.m}^{-3}$ increase in SO_2 was associated with a significant increase in hospital admissions for RD among the 0-14 year age-group (4.9% (0.3%, 9.7%)) on cold days. Also, a $10 \mu\text{g.m}^{-3}$ increase in NO_2 led to an increase (8.0% (1.3%, 15.1%)) in RD hospital admissions among males on normal days. However, there was no association between either $\text{PM}_{2.5}$ or PM_{10} and RD hospital admission. (Table 3).

In the second model, for all ages combined, a $10 \mu\text{g.m}^{-3}$ increase in $\text{PM}_{2.5}$ and PM_{10} was associated with a 3.5% (0.1%, 7.0%) and 1.7% (0.3%, 3.1%) increase in RD hospital admissions after adjusting for SO_2 on normal days. Also, for all ages combined, a $10 \mu\text{g.m}^{-3}$ increase in SO_2 on warm days was associated with 8.5% (0.4%, 17.2%) and 8.4% (0.3%, 17.1%) increase in RD hospital admission after controlling for $\text{PM}_{2.5}$ and PM_{10} , respectively. Conversely, on cold days, a $10 \mu\text{g.m}^{-3}$ increase in SO_2 was associated with increased hospital admissions among the 0-14 year age-group after controlling for the two types of particulate matter- $\text{PM}_{2.5}$ (6.5% (0.9%, 12.4%)) and PM_{10} (5.5% (0.3%, 11.1%)). (Table 4).

Sensitivity analysis

Median Tapp used to classify days

The only robust result was found with SO_2 . Similar to the main analysis (Tables 3 and 4), a $10 \mu\text{g.m}^{-3}$ increase in the level of SO_2 increased RD hospital admission in the 0-14 year olds on cold days in both the two level models (Tables 5 and 6). Also, a $10 \mu\text{g.m}^{-3}$ increase in SO_2 was associated with an increase in hospital admissions among the female participants on warm days after adjusting for $\text{PM}_{2.5}$ as in the two pollutant model of the main analyses. The effect estimates for SO_2 in the sensitivity analysis were lower than those of the main analysis except in the one-pollutant level (9.0% (0.3%, 18.4%)-Table 5) where it was higher than in the main analysis (4.9% (0.3%, 9.7%)) (Table 3).

Discussion

This study explored the modifying effects of temperature on the association between NO_2 , SO_2 , $\text{PM}_{2.5}$ and PM_{10} and RD hospital admission over a five-year period in Secunda, South Africa.

RD hospital admissions associated with SO_2 concentrations were affected by temperature extremes while the particulate matters ($\text{PM}_{2.5}$ and PM_{10}) had effect on RD admission during normal temperature. Overall, SO_2 was significantly associated with increased hospitalizations on warm days after adjusting for $\text{PM}_{2.5}$ or PM_{10} . The same applied to children between 0 and 14 years old. During normal temperature, $\text{PM}_{2.5}$ and PM_{10} were associated with increased hospitalizations after adjusting for SO_2 . There was an increase in the hospital admissions of the female participants when exposed to a $10 \mu\text{g.m}^{-3}$ increase in PM_{10} (adjusted for SO_2) and SO_2 (adjusted for $\text{PM}_{2.5}$) on normal and warm days respectively. With either of the particulate matters

(PM_{10} and $\text{PM}_{2.5}$), there was an increase in the RD hospital admission during normal temperature. These findings highlight the need to effectively manage air pollutants especially SO_2 in areas where temperature extremes are common.

The Highveld Priority Area, within which Secunda is located, is the home of many coal mining operations and coal fired power stations which are the main sources of SO_2 emissions. SO_2 is a gas produced by fuel combustion and one of the major sources of combustion pollution is traffic (Enkh-Undraa et al., 2019). Overall, SO_2 was associated with increased RD hospital admission during the warm periods in this study. This is probably because high temperature leads to an increase in sulphate aerosols due to faster SO_2 oxidation (Jacob and Winner, 2009; Luhana et al., 2007) and sulphate aerosols are considered to be the most irritating acid aerosol for the respiratory tract (Duarte et al., 2014). This is an important finding because in the next 100 years, the average temperature of South African inland where Secunda is located is projected to increase by 6-7°C (Department of Environmental Affairs, 2010) and this means that more RD hospital admissions should be expected in the future.

Furthermore, a $10 \mu\text{g.m}^{-3}$ increase in SO_2 could lead to an increase in RD hospital admissions among female patients on warm days, but no effect of temperature was observed among their male counterparts. Different studies have shown that female patients show higher susceptibility to SO_2 than male patients (Zhou et al., 2019; Zhang et al., 2014). The increase in RD hospital admissions among the female patients might be due to females having smaller lung tissue and trachea than males (Oiamo and Luginaah, 2013) resulting in a greater deposition of inhaled particles in their lungs. Females have fewer red blood cells than males, and thus may be more sensitive to the toxicological influences of SO_2 (Chen et al., 2005). Men and women also differ in their response to extreme temperatures. Women sweat less, have a higher working metabolic rate, and have thicker subcutaneous fat that prevents them from cooling themselves as efficiently as men. This shows that women, as a population, are less tolerant of an imposed heat stress (Duncan, 2006).

However, on cold days, a $10 \mu\text{g.m}^{-3}$ increase in SO_2 increased hospital admissions in children of 0-14 years but not in the older age-groups. SO_2 is a highly reactive gas whose concentration is very seasonal, peaking in the winter period (Morakinyo et al., 2020). It has been observed that children are more vulnerable than adults to air pollutants such as SO_2 by virtue of their increased susceptibility and the higher doses received (Mielzynska-Svach et al., 2013; Kochi et al., 2017) as they breathe higher volumes of air, their body systems are still developing and they have little control over their environment unlike adults (Salvi, 2007; Heinrich et al., 2002; Pikhart et al., 2001). Furthermore, exposure to cold temperatures reduces the functions of the nasal epithelium and reduces the capacity to protect the lower respiratory tract. This causes disorganization of the epithelium, nasal muciliary defence mechanisms and leaving the distal acinar airways more vulnerable to air pollutants (Lowen et al., 2007).

In this study, the effects of PM_{2.5}, PM₁₀ and NO₂ on respiratory disease hospital admissions were not robust enough as effects were different in the main analyses and the sensitivity analyses—the temperature effects depended on the categorization and levels of Tapp. In the main analysis, PM_{2.5} and PM₁₀ increased RD hospital admission during normal temperature on adjusting for SO₂, showing that extremes of temperature did not affect the effects of the particulate matters on RD hospital admissions in Secunda. This is contrary to the results of the 2 level Tapp and other studies that did not use similar classification of Tapp or temperature in cold, normal and warm/hot and when there was no adjustment for SO₂. In the 2 level Tapp, PM_{2.5}, PM₁₀ and SO₂ had effects during the warm temperature and this is similar to many studies (Zhang et al., 2018).

Tapp has been shown to be the most important predictor of heat-related mortality (Zhang et al; 2014). This is contrary to the findings of Barnett et al. (2010) which showed that there was no single temperature measure that is superior to others. Tapp has been applied in several studies (Wichmann et al., 2012; Wichmann et al., 2011; Lokotola et al., 2020). For example, in the warm period, an inter-quartile range increase in maximum apparent temperature (Tappmax) was associated with an increase of 7% (95% CI: 1%, 13%) in RD admissions in Greater Copenhagen, Denmark (Wichmann et al., 2011). Also, in South Africa,

This study is limited by the use of patient records from private hospitals. In South Africa, users of private hospitals are more likely to have high incomes, white-collar occupations and be gainfully employed. These factors are significant predictors of health insurance ownership (Kiriga et al., 2005). In South Africa, only the wealthiest 16% of the population can afford private health insurance to cover the costs of private-sector services (McIntyre and van den Heever, 2007). Therefore, the results cannot be extrapolated to the general South African population as the results represent the middle and upper socio-economic classes. It was postulated that including data from public hospitals would include people from the lower socio-economic class as people living in poor socio-economic conditions generally live closer to industrial areas and suffer more from the ill effects of air pollution (Naidoo et al., 2013), and could potentially show stronger associations with hospitalisation and air pollution levels. However, South African public hospitals have poor state of records management. Medical records are not being managed properly, resulting in a lack of effective systems for opening, tracking and indexing files (Marutha and Ngoepe, 2017).

It was assumed that air quality and temperature were homogenous for Secunda, which might give rise to measurement error. There might also be a potential lagged effects among participants who were not admitted immediately with the appearance of the symptoms, missing the milder cases that are not admitted to the hospital at all. Also, this study did not consider factors such as socioeconomic status, physical activities and pre-existing diseases as potential confounders

because these factors would not change within the month of case and control days.

Conclusion

SO₂ was associated with RD hospital admission in children aged 0-14 years during cold temperature but in females during warm temperatures. Both PM_{2.5} and PM₁₀ were associated with RD hospital admissions when the temperature was normal. This epidemiological evidence will help policy makers in South Africa to accept that policy interventions are needed to improve air quality as well as address the climate change-related health risks.

Author contributions

B.G.O. and J.W: Research design, methodology, statistical analyses, interpretation of results and writing the manuscript.

Financial interests declaration

None declared.

Acknowledgements

The authors would like to thank the South African Weather Services for the air pollution and weather data and the private hospital group for the hospital admission data. Authors express gratitude towards Prof Kuku Voyi and Dr Nico Claassen for preliminary feedback provided during the PhD studies of the 1st author.

To the memory of Constance Makwela (RIP) ,the research assistant for the PhD project.

Conflicts of interest

The authors declare no conflict of interest.

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Table 3: Percentage change (95% CI) in daily respiratory disease hospital admissions per 10 µg/m³ increase in an air pollutant level (lag0-1) in Secunda, South Africa on normal, warm and cold days by age groups and sex.

Air pollutant	Tapp	All	0-14 year olds	15-64 year olds	≥65 year olds	Females	Males
PM _{2.5}	Warm	5.2 (-5.3, 16.9)	11.5 (-3.3, 28.5)	-1.7 (-17.2, 16.7)	-2.1 (-33.4, 43.8)	-0.4 (-14.7, 16.5)	10.0 (-4.6, 26.9)
	Normal	2.3 (-0.2, 5.0)	2.2 (-1.4, 6.1)	2.9 (-0.9, 6.9)	-0.8 (-10.3, 9.6)	1.5 (-1.9, 5.0)	3.4 (-0.4, 7.4)
	Cold	0.1 (-1.3, 1.5)	-0.1 (-2.0, 1.9)	0.0 (-2.0, 2.1)	1.4 (-3.3, 6.3)	0.4 (-1.5, 2.3)	-0.2 (-2.1, 1.8)
PM ₁₀	Warm	0.8 (-1.7, 3.3)	3.0 (-0.3, 6.5)	-2.1 (-6.0, 1.9)	0.1 (-10.5, 11.9)	-0.5 (-4.1, 3.3)	1.8 (-1.6, 5.3)
	Normal	0.9 (-0.1, 2.0)	1.0 (-0.6, 2.6)	0.8 (-0.8, 2.3)	1.9 (-2.4, 6.5)	0.9 (-0.6, 2.4)	1.0 (-0.6, 2.6)
	Cold	-0.1 (-0.8, 0.7)	-0.1 (-1.2, 1.0)	0.0 (-1.2, 1.1)	0.1 (-2.5, 2.7)	0.0 (-1.0, 1.1)	-0.1 (-1.2, 1.0)
NO ₂	Warm	3.8 (-4.4, 12.8)	0.0 (-12.0, 13.6)	5.7 (-5.9, 18.7)	13.9 (-16.0, 54.5)	1.5 (-9.6, 14.0)	6.3 (-5.6, 19.6)
	Normal	1.2 (-3.2, 5.8)	1.3 (-4.8, 7.7)	1.0 (-5.7, 8.2)	1.9 (-16.1, 23.6)	-5.2 (-11.0, 1.0)	8.0 (1.3, 15.1)
	Cold	-0.7 (-4.4, 3.2)	-2.2 (-7.4, 3.2)	0.2 (-5.6, 6.3)	5.4 (-7.9, 20.6)	-1.6 (-6.9, 3.9)	0.2 (-5.0, 5.8)
SO ₂	Warm	6.3 (-1.3, 14.5)	9.4 (-2.4, 22.6)	4.7 (-5.9, 16.4)	0.1 (-23.1, 30.2)	10.5 (-0.5, 22.8)	2.5 (-7.7, 13.8)
	Normal	-0.4 (-3.8, 3.2)	0.9 (-4.2, 6.3)	-1.0 (-5.9, 4.1)	-4.6 (-16.9, 9.5)	-0.9 (-5.6, 4.1)	0.2 (-4.7, 5.4)
	Cold	1.5 (-1.4, 4.5)	4.9 (0.3, 9.7)	-0.9 (-5.1, 3.4)	-0.9 (-9.9, 9.0)	-0.2 (-4.3, 4.1)	3.3 (-0.9, 7.6)

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

Table 4: Percentage change (95% CI) in daily respiratory disease hospital admissions per 10 µg/m³ increase in an air pollutant level (lag0-1) in Secunda, South Africa on normal, warm and cold days by age groups and sex.

Air pollutant	Tapp	All	0-14 year olds	15-64 year olds	≥65 year olds	Females	Males
PM _{2.5} adjusted NO ₂	Warm	1.9 (-8.7, 13.8)	9.5 (-6.1, 27.6)	-5.7 (-20.9, 12.5)	-4.2 (-35.5, 42.4)	-3.6 (-18.3, 13.7)	6.6 (-8.2, 23.6)
	Normal	2.0 (-1.1, 5.3)	4.1 (-0.5, 8.8)	0.3 (-4.3, 5.1)	-1.3 (-12.6, 11.5)	2.1 (-2.1, 6.5)	2.2 (-2.4, 7.1)
	Cold	-0.2 (-1.9, 1.5)	-0.4 (-2.8, 2.1)	-0.1 (-2.6, 2.4)	0.8 (-5.1, 7.0)	0.2 (-2.1, 2.5)	-0.6 (-3.0, 1.9)
NO ₂ adjusted PM _{2.5}	Warm	4.8 (-4.1, 14.5)	-0.1 (-12.8, 14.3)	6.1 (-6.4, 20.3)	25.7 (-10.9, 77.3)	3.7 (-8.7, 17.7)	6.0 (-6.3, 20.0)
	Normal	-2.4 (-8.4, 4.0)	-2.1 (-10.2, 6.7)	-4.3 (-13.6, 6.0)	1.4 (-22.2, 32.1)	-9.5 (-17.3, -0.8)	5.2 (-4.0, 15.2)
	Cold	-1.0 (-4.8, 3.1)	-2.3 (-7.7, 3.3)	-0.4 (-6.4, 6.0)	5.1 (-8.6, 20.8)	-2.0 (-7.4, 3.8)	0.0 (-5.4, 5.7)
PM _{2.5} adjusted SO ₂	Warm	-2.2 (-15.3, 13.0)	1.5 (-17.9, 25.3)	-0.6 (-19.8, 23.3)	-27.6 (-57.9, 24.8)	-8.8 (26.3, 13.0)	3.8 (-14.7, 26.3)
	Normal	3.5 (0.1, 7.0)	2.8 (-1.9, 7.7)	5.9 (0.5, 11.6)	-4.7 (-16.1, 8.3)	2.8 (-1.8, 7.6)	4.4 (-0.7, 9.6)
	Cold	-0.3 (-2.6, 2.0)	-1.5 (-4.7, 1.9)	0.6 (-2.9, 4.23)	0.6 (-6.9, 8.8)	1.6 (-1.6, 4.9)	-2.1 (-5.3, 1.2)
SO ₂ adjusted PM _{2.5}	Warm	8.5 (0.4, 17.2)	12.1 (-0.5, 26.2)	6.8 (-4.3, 19.3)	-1.6 (-26.2, 31.2)	11.6 (0.0, 24.6)	5.5 (-5.4, 17.8)
	Normal	-2.2 (-5.9, 1.7)	-1.3 (-6.9, 4.6)	-3.0 (-8.3, 2.7)	-3.2 (-16.6, 12.4)	-1.8 (-7.0, 3.7)	-2.5 (-7.9, 3.2)
	Cold	1.6 (-1.9, 5.2)	6.5 (0.9, 12.4)	-2.2 (-7.0, 2.9)	0.3 (-10.9, 13.0)	-1.3 (-6.2, 3.7)	4.6 (-0.4, 9.9)
PM ₁₀ adjusted NO ₂	Warm	-0.3 (-3.0, 2.4)	2.9 (-0.7, 6.6)	-4.2 (-8.3, 0.1)	-2.4 (-13.3, 9.8)	-2.2 (-6.1, 1.8)	1.2 (-2.4, 5.0)
	Normal	0.7 (-0.6, 2.0)	1.6 (-0.3, 3.6)	-0.4 (-2.4, 1.5)	2.2 (-2.9, 7.5)	0.9 (-0.9, 2.7)	0.4 (-1.5, 2.4)
	Cold	-0.2 (-1.1, 0.7)	-0.2 (-1.6, 1.2)	-0.1 (-1.5, 1.2)	-0.3 (-3.5, 2.9)	0.0 (-11.2, 1.3)	-0.4 (-1.7, 1.0)
NO ₂ adjusted PM ₁₀	Warm	5.0 (-4.0, 14.9)	-1.5 (-14.2, 13.1)	7.4 (-5.2, 21.7)	26.9 (-10.4, 79.5)	4.5 (-8.0, 18.8)	5.6 (-6.9, 19.7)
	Normal	-1.8 (-7.6, 4.4)	-0.8 (-8.6, 7.6)	-3.6 (-12.9, 6.5)	-2.1 (-24.3, 26.6)	-9.0 (-16.6, -0.7)	6.2 (-2.7, 16.0)
	Cold	-0.9 (-4.8, 3.1)	-2.3 (-7.7, 3.3)	-0.4 (-6.3, 6.0)	5.9 (-7.9, 21.7)	-1.9 (-7.3, 3.9)	0.1 (-5.3, 5.8)
PM ₁₀ adjusted SO ₂	Warm	-0.7 (-3.6, 2.3)	1.4 (-2.8, 5.8)	-2.4 (-6.7, 2.0)	-3.5 (-16.6, 11.7)	-2.0 (-6.2, 2.4)	0.4 (-3.6, 4.6)
	Normal	1.7 (0.3, 3.1)	1.5 (-0.5, 3.6)	1.6 (-0.4, 3.7)	3.5 (-2.2, 9.5)	2.2 (0.2, 4.2)	1.2 (-0.8, 3.2)
	Cold	-0.2 (-1.3, 1.0)	-0.3 (-2.0, 1.5)	0.1 (-1.7, 1.9)	-1.2 (-5.0, 2.7)	0.3 (-1.3, 2.0)	-0.6 (-2.3, 1.1)
SO ₂ adjusted PM ₁₀	Warm	8.4 (0.3, 17.1)	12.3 (-0.3, 26.4)	6.7 (-4.5, 19.1)	-1.4 (-26.1, 31.6)	11.5 (-0.1, 24.5)	5.7 (-5.3, 17.9)
	Normal	-2.5 (-6.3, 1.4)	-1.7 (-7.4, 4.3)	-2.4 (-7.8, 3.2)	-7.8 (-21.1, 7.8)	-2.9 (-8.1, 2.6)	-2.1 (-7.5, 3.6)
	Cold	1.6 (-1.8, 5.0)	5.5 (0.3, 11.1)	-1.8 (-6.4, 3.0)	2.5 (-8.1, 14.4)	-0.5 (-5.1, 4.4)	3.6 (-1.1, 8.6)

Warm: Apparent temperature > 75th percentile; Cold: Apparent temperature < 25th percentile; Normal: Apparent temperature >= 25th and <= 75th percentile. Bold text: Significant (p < 0.05)

Table 5: Percentage change (95% CI) in daily respiratory disease hospital admissions per 10 µg/m³ increase in an air pollutant level (lag0-1) in Secunda, South Africa on warm and cold days by age groups and sex.

Air pollutant	Tapp	All	0-14 year olds	15-64 year olds	≥65 year olds	Females	Males
PM _{2.5}	Warm	8.2 (1.3, 15.6)	12.0 (2.0, 23.0)	4.0 (-6.2, 15.2)	10.7 (-13.8, 42.1)	9.4 (-0.4, 20.3)	7.0 (-2.5, 17.5)
	Cold	0.7 (-0.4, 1.8)	0.2 (-1.4, 1.8)	1.0 (-0.7, 2.7)	2.2 (-1.7, 6.4)	1.1 (-0.4, 2.6)	0.2 (-1.4, 1.8)
PM ₁₀	Warm	1.6 (0.0, 3.2)	3.0 (0.8, 5.3)	-0.2 (-2.7, 2.3)	2.7 (-3.6, 9.4)	2.7 (0.4, 5.0)	0.6 (-1.6, 2.9)
	Cold	0.4 (-0.2, 1.0)	0.2 (-0.6, 1.1)	0.5 (-0.3, 1.4)	1.0 (-1.1, 3.1)	0.6 (-0.2, 1.4)	0.2 (-0.6, 1.1)
NO ₂	Warm	-0.2 (-11.0, 11.8)	-6.2 (-20.7, 11.0)	2.6 (-13.1, 21.2)	23.7 (-20.8, 93.2)	-6.3 (-20.4, 10.2)	6.2 (-9.6, 24.6)
	Cold	2.6 (-2.7, 8.3)	2.1 (-5.2, 9.9)	1.6 (-6.6, 10.5)	14.2 (-6.7, 39.9)	-0.4 (-7.7, 7.4)	5.8 (-1.9, 14.2)
SO ₂	Warm	13.5 (0.4, 28.3)	25.3 (4.8, 49.7)	8.4 (-9.5, 29.7)	-24.4 (-54.2, 24.7)	19.9 (0.8, 42.7)	7.7 (-9.5, 28.0)
	Cold	4.5 (-1.2, 10.5)	9.0 (0.3, 18.4)	1.8 (-6.3, 10.5)	-3.0 (-19.6, 17.0)	2.2 (-5.5, 10.6)	6.8 (-1.3, 15.6)

Warm: Apparent temperature ≥ 50th percentile; Cold: Apparent temperature < 50th percentile. Bold: p < 0.05

Table 6: Percentage change (95% CI) in daily respiratory disease hospital admissions per 10 µg/m³ increase in an air pollutant level (lag0-1) in Secunda, South Africa on normal, warm and cold days by age groups and sex.

Air pollutant	Tapp	All	0-14 year olds	15-64 year olds	≥65 year olds	Females	Males
PM _{2.5} adjusted NO ₂	Warm	3.2 (-4.2, 11.2)	11.5 (0.4, 23.9)	-6.1 (-16.4, 5.4)	10.0 (-16.6, 45.0)	2.3 (-8.2, 14.0)	-0.1 (-2.0, 2.0)
	Cold	0.4 (-1.0, 1.7)	-0.4 (-2.3, 1.6)	1.0 (-1.1, 3.0)	1.8 (-3.1, 7.0)	0.7 (-1.1, 2.6)	4.0 (-6.1, 15.3)
NO ₂ adjusted PM _{2.5}	Warm	0.6 (-6.0, 7.7)	-4.5 (-13.8, 5.8)	3.2 (-6.4, 13.8)	22.6 (-6.8, 61.1)	-3.6 (-12.6, 6.4)	4.7 (-4.8, 15.1)
	Cold	0.6 (-2.8, 4.1)	1.2 (-3.4, 6.1)	-1.2 (-6.5, 4.3)	5.8 (-6.6, 19.8)	-0.4 (-5.1, 4.6)	1.5 (-3.3, 6.6)
PM _{2.5} adjusted SO ₂	Warm	4.6 (-3.3, 13.3)	7.9 (-3.8, 21.1)	1.4 (-9.9, 14.1)	6.9 (-20.8, 44.1)	4.9 (-6.2, 17.4)	4.3 (-6.8, 16.7)
	Cold	-0.3 (-2.1, 1.6)	-1.5 (-4.0, 1.2)	0.9 (-1.9, 3.7)	0.3 (-5.7, 6.8)	0.9 (-1.6, 3.5)	-1.6 (-4.2, 1.0)
SO ₂ adjusted PM _{2.5}	Warm	4.3 (-1.0, 9.9)	5.0 (-2.8, 13.5)	5.5 (-2.1, 13.7)	-10.1 (-27.5, 11.4)	7.9 (0.2, 16.1)	1.0 (-6.2, 8.7)
	Cold	2.0 (-0.6, 4.6)	4.7 (0.8, 8.7)	-0.2 (-3.8, 3.6)	-0.4 (-8.7, 8.6)	0.6 (-2.9, 4.2)	3.4 (-0.2, 7.2)
PM ₁₀ adjusted NO ₂	Warm	0.9 (-0.8, 2.7)	3.4 (1.0, 5.9)	-2.4 (-5.1, 0.4)	2.6 (-4.3, 10.0)	1.8 (-0.7, 4.4)	0.0 (-2.4, 2.5)
	Cold	0.3 (-0.4, 1.0)	0.1 (-0.9, 1.2)	0.4 (-0.6, 1.5)	0.8 (-1.8, 3.4)	0.4 (-0.5, 1.4)	0.1 (-0.9, 1.2)
NO ₂ adjusted PM ₁₀	Warm	0.3 (-6.4, 7.4)	-6.2 (-15.5, 4.0)	4.1 (-5.6, 14.8)	21.9 (-7.5, 60.7)	-4.5 (-13.5, 5.5)	5.1 (-4.6, 15.7)
	Cold	0.5 (-2.9, 3.9)	0.8 (-3.8, 5.5)	-1.0 (-6.3, 4.5)	6.2 (-6.1, 20.0)	-0.4 (-5.0, 4.5)	1.3 (-3.4, 6.3)
PM ₁₀ adjusted SO ₂	Warm	0.9 (-0.8, 2.7)	2.4 (-0.1, 4.9)	-0.9 (-3.5, 1.8)	2.5 (-4.6, 10.0)	1.6 (-0.8, 4.2)	0.2 (-2.2, 2.6)
	Cold	0.1 (-0.8, 1.1)	-0.1 (-1.5, 1.2)	0.4 (-1.0, 1.8)	0.1 (-3.0, 3.3)	0.6 (-0.7, 1.8)	-0.3 (-1.6, 1.0)
SO ₂ adjusted PM ₁₀	Warm	4.5 (-0.7, 10.0)	5.1 (-2.6, 13.4)	6.0 (-1.6, 14.1)	-10.4 (27.7, 11.0)	7.7 (0.1, 15.9)	1.4 (-5.7, 9.1)
	Cold	1.6 (-0.8, 4.1)	3.7 (0.1, 7.5)	0.0 (-3.5, 3.6)	-0.3 (-8.2, 8.2)	0.6 (-2.7, 4.1)	2.6 (-0.8, 6.2)



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Research article

Quantifying potential particulate matter intake dose in a low-income community in South Africa

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Received: 27 January 2021 - Reviewed: 8 June 2021 - Accepted: 29 August 2021

<https://doi.org/10.17159/caj/2021/31/2.9426>

Abstract

Understanding how exposure to particulate matter impacts human health is complex. Personal exposure is a function of the pollution concentrations measured at any given place and time. The health impacts of this exposure are, in part, determined by how high pollutant concentrations are and how much pollution can potentially enter the body. This study considered data gathered in the winter of 2013 in a low-income community on the Mpumalanga Highveld, South Africa, which is a geographical area known for its high air pollution levels. Data collected by GPS monitors worn by individuals in the community were used to understand in which microenvironments people spend most of their time. Participants spent time in five main micro-environments: (highest rank first) inside a house, directly outside a house, on a dirt road, on a tar road, and on an open field. Eight days' worth of ambient, indoor and personal particulate matter measurements were paired with individual GPS positioning data for one study participant. We identified pollutant concentrations where the person spent time and how much particulate matter the person potentially inhaled. Highest concentrations were measured inside the dwelling and directly outside the dwelling of the individual. When comparing directly (ranging from 0.02 – 0.76 mg) - and indirectly (0.02 – 0.34 mg) derived time-weighted potential intake doses, directly derived intake doses were higher and more likely to represent how much particulate matter was potentially inhaled by the participant. This study suggests that people living in communities on the Mpumalanga Highveld are exposed to unacceptably high air pollution levels in places in which they spend most of their time. Direct exposure and intake dose assessments are an important element of environmental health studies to supplement data collected by stationary monitors in order to better understand exactly what people are breathing.

Keywords

air pollution exposure, household air pollution, micro-environments

Introduction

The effect of air pollution on human health is dependent on many factors (Wilson et al., 2000). Initial steps towards better understanding how air pollution influences health include 1) identifying how much air pollution an individual comes into contact with at the breathing zone and for how long, 2) determining how much of this air pollution is inhaled, and finally 3) understanding how much pollution is taken up into the body, where the chemical characteristics of the pollutant determine how it can affect organs and bodily systems (Figure 1).

Air pollution exposure

Stationary ambient air quality monitors are often used as a proxy for personal air pollution exposure. Ambient concentrations, however, are not necessarily indicative of the

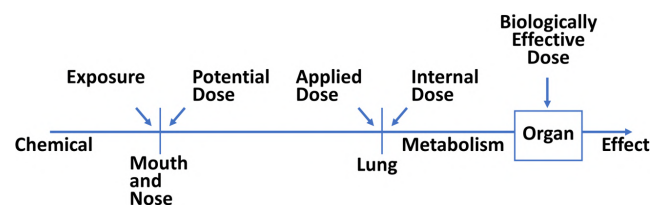


Figure 1: Illustration of Inhalation Route: Exposure and Dose (U.S. EPA 1992)

pollutant concentrations that people are exposed to especially within low-income residential communities where air pollution exposure is highly variable and where people spend their time in different micro-environments (Bruce et al., 2002; Ferro et al., 2004; Diapouli, 2011; Lim et al., 2012). Exposure to air pollution is typically a function of time spent in proximity to pollution

sources in multiple locations (Vette et al., 2001). Even though significant emissions stem from outdoor sources, people spend most of their time in an indoor environment where significant sources also exist (Vette et al., 2001). These sources contribute to the total dose of air pollution inhaled by an individual on a typical day. Consequently, to understand air pollution exposure and how much a person can potentially breathe in, air pollution needs to be measured in every micro-environment in which an individual spends time throughout the day. For this, micro-environment and time-activity pattern identification are essential (Park et al., 2020).

Potential air pollution intake dose

Determining how much air pollution is *potentially* inhaled by an individual to then be taken up by the body requires knowledge of the concentration of the pollutant which the individual is exposed to at the breathing zone, the breathing rate of the individual, as well as the time spent inhaling the air pollution at that concentration.

Breathing rates differ by age and gender, and determine how much and how deeply a specific pollutant is inhaled into the respiratory system (Smith, 1993; Wilson et al., 2000). An individual's physiology, body size and activity level also influence a person's breathing rates (Wilson et al., 2000). Normal breathing rates for adults (men and women combined) range from 12.2 m³/day (for ages 81 years and older) to 16.0 m³/day (for ages 31 to 51 years) (U.S. EPA, 2011).

It is possible to derive direct and indirect exposure to air pollution, as well as direct and indirect potential intake dose estimates. Direct methods consider inhalation-based exposure assessments using personal monitors located in the breathing zone (Abbey et al., 1999; Cong et al., 2021). Indirect methods include questionnaires/ time-activity diaries and measurements taken at centrally located, stationary monitoring sites. This estimation considers time spent in specific micro-environments and matches this information with the average particulate matter (PM) concentrations measured in those environments (Jones et al., 2000; Mdluli, 2007). Holistic, total exposure estimates would consider a combination of both direct and indirect measurements with detailed personal activity patterns.

Though ambient and indoor particulate matter data has been collected in low-income communities in South Africa, few studies have considered ambient, indoor, and personal air pollution concentrations simultaneously. To the best of our knowledge, no studies have considered personal exposure to PM concentrations in various micro-environments and presented corresponding potential intake doses. This study aims to fill this research gap.

Methods

Sampling site

The low-income community chosen for this study is situated in



Figure 2: GPS coordinates plotted on an image of the community to identify common micro-environments. Multiple individuals' GPS data are illustrated on this figure to avoid disclosing personal information about the participant in this case study. The different colours represent coordinates of different individuals on different days and have no other meaning.

the Gert Sibande District Municipality in Mpumalanga, South Africa. It is located within the Highveld Priority Area known for poor air quality caused mainly by cumulative emissions stemming from sources such as domestic burning and industrial activities, but also road dust and waste burning (Nkosi et al., 2017).

PM data collection and processing

Ambient PM_{2.5} and PM₁₀ concentrations were measured at a centrally located, stationary site using Horiba BAM and MetOne E-Bam instruments, respectively.

Personal PM₄ concentrations of one adult who gave informed consent were monitored on different days using a TSI SidePak AM510 photometric monitor for a range of one to eight hours per day. This adult also carried a mobile GPS which tracked personal movements. Indoor PM₄ concentrations were measured in the dwelling of the individual using a TSI DustTrak photometric monitor (Models 8520 and 8530). The indoor and personal PM₄ measurements were corrected according to a specific photometric calibration factor obtained for the DustTrak and SidePak instruments (Language et al. 2016).

Measurements were taken between 21 – 28 August 2013. The GPS coordinate readings represented daytime movement patterns ranging from 09:00 to 18:00. No position readings were taken during the night, but it can be assumed that the individual spent less time outside of the dwelling during the evening and spent most of the time indoors at night.

Identification of micro-environments

To track everyday movement patterns, Global Positioning System (GPS) monitors were worn by a sub-sample of 20 consenting community members who were participants in a larger air quality study, and who also wore personal air quality monitors and spent most of their time within the community. Micro-environments were identified by plotting dots of all

recorded GPS readings over the period of the study on a SPOT 6/7 (2015) satellite image using QGIS version 2.14.1 (Figure 2). Areas on the map where the dot-density was highest were identified as places frequently visited whilst wearing the personal monitoring device.

Sampling was conducted as part of a larger sampling campaign in the winter of 2013. This study was conducted under the oversight of the North-West University Health Research Ethics Committee (NWU-00066-13-S3).

Corresponding personal exposure and micro-environments

The personal PM₄ concentrations were paired with GPS tracking data at ten-minute average intervals. Figure 3 illustrates the personal PM₄ concentrations measured in the places in which the individual spent time on 25 August 2013.

Indoor PM₄ measurements for the dwelling in which the individual spent the most time were also considered together with ambient PM_{2.5} concentrations. No detailed behavioural information was considered, and this is recognised as a limitation of the study since exposure is closely associated with behavioural patterns (Terblanche et al. 1992).

Direct and indirect intake doses

Total (integrated) exposure, which considers the period an individual comes into contact with an exposure concentration, is derived by using Equation (1):

$$E = \int_{t_1}^{t_2} C(t) dt \tag{1}$$

Where E is the magnitude of exposure, C(t) is the exposure concentration as a function of time and t is time, t₂ – t₁, representing the exposure duration (ED) (U.S. EPA 1992). Once the magnitude of exposure has been determined, it is possible to calculate the *potential* dose, or the amount of a pollutant that *could* be inhaled (through the mouth or nose), all of which is not absorbed by the body.

Equation 2 shows the *potential* dose for inhalation as the integration of the pollutant intake rate (concentration of the pollutant in the air times the intake rate of the air, C times IR) over time (U.S. EPA 1992):

$$D_{pot} = \int_{t_1}^{t_2} C(t)IR(t)dt \tag{2}$$

D_{pot} is *potential* dose and IR(t) is the inhalation rate of an individual. The quantity t₂ – t₁ represents the period over which exposure is examined (U.S. EPA 1992). The above methods describe how the potential intake of exposure concentrations was assessed.

Direct *potential* intake doses were derived using equation 2 by using the direct personal measurements read by the personal monitor. A short-term inhalation rate for an average middle-aged woman at light-intensity activity was used for the purposes

of this exercise (0.012 m³/min) (U.S. EPA, 2011). GPS / Personal PM₄ data did not represent a full day’s worth of data, hence a short-term breathing rate was deemed appropriate.

To indirectly derive *potential* intake doses over the same period, instead of personal PM measurements, stationary indoor PM₄, ambient PM_{2.5} measurements and movement patterns determined by the GPS monitor were used. PM₄ concentrations were directly compared to PM_{2.5} concentrations as these have been found to be comparable in another study, which identified the relationship between PM of different size fractions measured at the same time and place. The median ratio of PM_{2.5}:PM₄ was found to be close to one (0.92) (Language 2019) supporting the comparison of the two PM size fractions in this study.

Results

Identified micro-environments

Considering the plotted GPS coordinates in figure 2, a total of five different micro-environments were identified based on GPS monitor data for numerous study participants. Most time was spent (highest rank first) inside a house, directly outside a house, on a dirt road, on a tar road, and in an open field. Most of the personal movements (on average 87%) were concentrated inside- and directly outside of dwellings.

Personal exposure in different micro-environments

The participant wearing the personal PM₄ monitor was exposed to a wide range of PM₄ concentrations in the locations in which the individual spent time on each day between 21 and 28 August. Figure 3 illustrates an example of personal exposure on 25 August 2013 where the individual was exposed to PM₄ concentrations ranging from 36 µg/m³ between 13:00 and 13:10 directly outside a house and 2 781 µg/m³ between 18:00 and 18:10 inside a house.

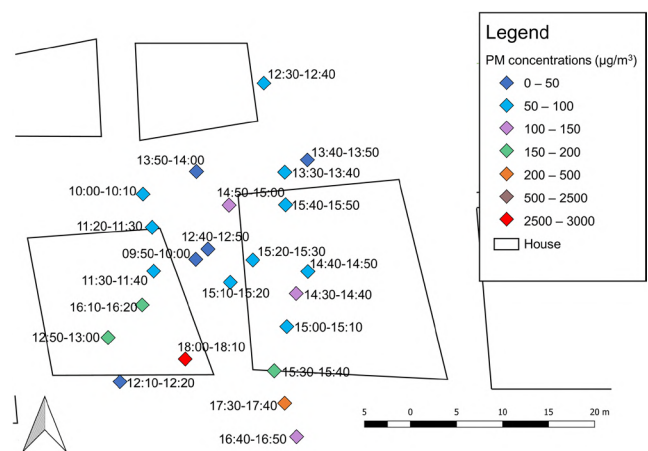


Figure 3: GPS tracks (dots) showing ten-minute average personal PM₄ concentration exposure on 25 August 2013. Micro-environments displayed are “Inside a house” (delineated by an outlined rectangle) and “Directly outside a house” (the space outside the rectangles).

When plotting similar figures for every other day between 21 and 28 August 2013 (the dates on which this specific individual wore the monitors), it was found that personal exposure concentrations ranged from low concentrations to unacceptably high concentrations, and often reached levels into the mg/m³ range. When comparing daily average PM concentrations, personal, indoor and ambient concentrations were highly variable (Table 1).

Table 1: Comparing daily average personal PM measurements with daily average ambient and indoor PM measurements (µg/m³) ± standard deviation

Date	Personal PM ₄	Indoor PM ₄	Ambient PM _{2.5}
21-Aug-13	32 (1.42)	189 (387.55)	35 (6.60)
22-Aug-13	13 (7.58)	259 (1111.50)	37 (19.90)
23-Aug-13	100 (33.82)	207 (381.71)	39 (16.45)
24-Aug-13	100 (30.88)	147 (302.35)	40 (14.05)
25-Aug-13	192 (845.96)	237 (672.13)	42 (13.55)
26-Aug-13	58 (21.27)	87 (51.07)	33 (9.71)
27-Aug-13	71 (36.88)	125(108.43)	37 (8.61)
28-Aug-13	176 (228.57)	272 (350.77)	61 (29.26)

Table 2: Directly and indirectly derived potential intake doses (mg) between 21 – 28 August 2013 for the number of hours for which personal PM and GPS measurements were available

Direct potential dose for which personal measurements were available		
Date	Dose (mg)	Time (Hours)
21-Aug-13	0.02	1.00
22-Aug-13	0.05	4.83
23-Aug-13	0.52	7.17
24-Aug-13	0.21	3.50
25-Aug-13	0.76	5.83
26-Aug-13	0.19	4.33
27-Aug-13	0.17	2.67
28-Aug-13	0.34	3.50
Indirect time-weighted potential dose for the hours in which ambient, indoor and personal measurements were simultaneously available		
Date	Dose (mg)	Time (Hours)
21-Aug-13	0.02	1.00
22-Aug-13	0.08	4.83
23-Aug-13	0.03	7.17
24-Aug-13	0.14	3.50
25-Aug-13	0.35	5.83
26-Aug-13	0.25	4.33
27-Aug-13	0.14	2.67
28-Aug-13	0.23	3.50

Potential intake doses

Potential PM₄ intake doses ascertained using direct and indirect methods, varied (Table 2). Potential intake dose estimates using the direct potential dose derivation method ranged between 0.02 - 0.76 mg for the time during which GPS measurements were available (i.e. between 09:00 and 17:00). Indirect potential intake doses for the same time ranged between 0.02 - 0.34 mg. Potential intake doses calculated in an indirect manner, using stationary monitoring data, were generally lower compared to potential intake doses calculated from direct personal exposure measurements. On the 23rd of August 2013, for example, the direct potential dose calculated was 0.52 mg over 7 hours, whereas the indirectly derived potential dose for the same period was 0.03 mg (0.49 mg less). In Figure 3, on 25 August 2013, the individual wearing the monitor potentially inhaled a directly derived PM dose of 0.76 mg. When considering the indirectly derived dose for that same day, the individual potentially inhaled a dose of 0.35 mg (0.41 mg of PM less). Highest potential doses mainly corresponded to the days on which readings were available for the longest period of time (Table 2 - final column).

Discussion

Particulate matter exposure and potential intake doses

Air pollution exposure of an individual who resides in a low-income town on the Mpumalanga Highveld, where air pollution is an identified health concern, was investigated (Balmer, 2007; Mdluli, 2007; Garland et al., 2017). Five main micro-environments were identified as places in which time was spent. These were 1) inside a house, 2) directly outside a house, 3) on a dirt road, 4) on a tar road and 5) on an open field. On average, 90% of the time was spent in and around a house. Less time was spent on roads and open fields.

Ambient, indoor and personal PM measurements were found to exceed ambient standards in numerous micro-environments, where PM levels even went into the milligram range.

GPS tracking data and personal PM₄ measurements of a participant were paired at ten-minute average intervals over an eight-day period (between 21 and 28 August 2013). By plotting the matched datasets onto an aerial photograph, location patterns helped understand how much PM₄ the individual was exposed to in each micro-environment where time was spent.

Exposure to highest concentrations could not be pinpointed to the indoor environment only, because high personal PM measurements were also recorded when the individual was in the ambient environment. Nevertheless, the most extreme exposure concentrations were recorded in the indoor environment and directly outside of houses.

Direct and indirect potential intake doses for PM₄ were calculated and compared. A trend of lower potential doses for indirectly derived methods was evident on five of the eight days

studied. On the other three days, the indirectly derived doses were either higher than or equal to the potential dose derived by direct methods. These differences are likely attributable to the fact that stationary monitors used for the indirect methods did not measure the actual concentrations the individual was exposed to at the breathing zone. These monitors measure concentrations in the identified micro-environment in which they were positioned, thereby representing only a proxy for what the individual was potentially exposed to.

The resulting figures are representative of doses of PM potentially inhaled during winter as a result of time spent exposed in the ambient and the indoor environment. Though indirectly-derived potential doses fell below what has been demonstrated to be inhaled in other low-income communities (e.g. when compared to potential inhalation doses in Bangladesh where doses ranged between 4.4 mg and 5.8 mg of PM_{2.5} per day (Chowdhury et al., 2012)), it should be kept in mind that, for smaller PM size fractions there is no threshold below which there is no discernible health effect (Zhao et al., 2020).

The average potential intake dose of PM_{2.5} has been estimated to range between 7.0 – 17.5 mg per cigarette actively smoked (Pope et al., 2009). The results in this study show that the amount of PM potentially inhaled by an individual in their living environment during a few hours are not far from the amount of PM inhaled while actively smoking when extrapolated to an entire day. This exposure could lead to cardiovascular and respiratory health effects without the individual even actively being aware of it (Pope et al., 2009).

It is likely that the directly derived total daily potential intake dose of the individual would be much higher than what has been demonstrated here. This is because peak exposure concentrations during domestic burning times for heating or cooking in the early morning may not have been included, and because the directly derived potential doses have been estimated with an incomplete dataset (only for a few hours a day, for which data were available). As time-activity journals were not kept by the participant of the study, it was not possible to identify the source of the PM to which the participant was exposed at different times. These are clear limitations of the study.

Directly- versus indirectly derived exposure and potential intake dose assessments

An age-old scientific debate between scientists exists, which compares the value of directly- and indirectly derived air quality exposure assessments for air pollution environmental-epidemiologic studies (National Research Council 1997; Wilson 2006; Diapouli et al., 2011). Both methods have advantages and disadvantages depending on the research question. Direct measurements, in this study represented by personal PM₄ measurements and GPS readings, are generally the most accurate estimate of concentrations which people are exposed to at the breathing zone. Indirect measures (stationary air quality

monitors) are less expensive, less time- and resource-intensive, and cover a larger temporal and spatial scale (National Research Council 1997).

In the case of this study, ambient- and indoor PM measurements help better understand indicative personal total exposure rates, if used to derive indirect potential intake doses along with micro-environment data. These measurements help answer the question of whether air quality in the community complies with ambient standards. The more individual-focused elements of this study, like the identification of micro-environments, for instance, help prioritise where specifically to focus action to reduce air pollution. Similarly, by pairing location with personal PM exposure, priority exposures can be identified for risk mitigation and air pollution management strategies.

Gathering more information on the biology of the individual (e.g., weight and breathing rate) as well as understanding the chemical characteristics of the pollutant in question would assist in better understanding how much of the pollutant is eventually taken up into the body and how it impacts on it.

Conclusions

Residents of low-income communities on the Mpumalanga Highveld are chronically exposed to air that is not safe to breathe (Wernecke et al., 2015; Language et al., 2016). This study exemplifies that a typical individual living in one of these communities is exposed to high PM₄ concentrations in the spaces in which the most time is spent during a typical day (in winter for this study) and that directly derived potential intake doses, are more likely to represent what an individual breathes in.

This is the first South African study to link PM exposure to micro-environments, to directly- and indirectly-determine this exposure and to then compare potential PM₄ intake doses in these environments, which contributes to our understanding of PM exposure in South African low-income communities.

Direct- and indirect exposure and potential dose intake assessments have been conducted in this study. Micro-environments have also been determined. The next step towards filling South Africa's exposure- and dose-response function gap could be to build upon this study design and to then determine health impacts using daily health diaries and / or medical surveys, coupled with personal measurements and time-activity patterns to support the information collected by more general means, such as ambient air quality monitoring coupled with community surveys. This is a necessary next step in understanding how air pollution affects the people living in communities on the Highveld where air pollution is a serious problem.

Acknowledgements

Data collection was conducted by North-West University and The Nova Institute.

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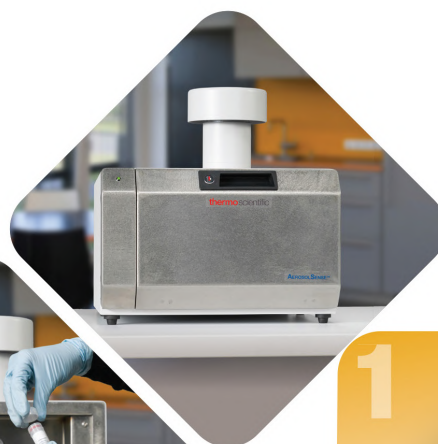


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Research article

The quality of the first and second Vaal Triangle Airshed Priority Area Air Quality Management Plans

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Received: 15 September 2021 - Reviewed: 22 October 2021 - Accepted: 11 November 2021

<https://doi.org/10.17159/caj/2020/31/2.12178>

Abstract

In response to deteriorating air quality, South Africa implemented national programmes that aim to manage and regulate ambient air quality and air pollution. Air Quality Management Plans (AQMPs) are clear outlines of measures and resources needed to achieve air quality objectives in a given geographical area and require support from government, business, industry, non-governmental organisations (NGOs) and the public. The success of the AQMPs depends primarily on the support of all stakeholders and the quality of the management plan. The Vaal Triangle Airshed Priority Area (VTAPA) was declared in 2006 as an area where ambient air quality standards are exceeded or may cause adverse air quality impacts. This research study focused on the VTAPA to evaluate the quality of the first and second-generation AQMPs for the VTAPA. Quality evaluation includes an analysis of procedures, processes, methods and documents. Effectiveness refers to the results of individual activities; therefore, the extent to which the AQMP met the expected outcomes of the review package defined the quality of the AQMP report. Both the first and draft second-generation AQMPs were considered to be of good quality. The first-generation AQMP was found to be of better quality than that of the draft second-generation AQMP. Funding mechanisms need to be investigated to assist in implementing intervention strategies in the AQMP as both the first and draft second-generation AQMPs were found to lack the potential to secure funds. Though the draft second-generation AQMP was found to be of lesser quality, the source apportionment study for identification of all sources as well as a better-outlined air quality management system was found to be good improvements to the AQMP.

Keywords

air quality management, quality, priority areas, airshed, Vaal Triangle

Introduction

Research studies worldwide have reliably recorded the devastating effect of ambient air pollution on human health. It has been estimated that annually at least seven million deaths worldwide are due to the impact of air pollution (Mannucci & Franchini, 2017). According to Altieri and Keen (2016); McCarthy (2020); Robinson (2019), the economic burden associated with air pollution equated to 3.3% of the world's gross domestic product (GDP) in 2018, 3.8% of China's GDP in 2007, 5% of GDP in the U.S in 2014 and 6.0% of South Africa's GDP in 2012, accounting for 7.4% of deaths associated with exposure to PM_{2.5}.

Effects of exposure to poor ambient air on human health varies from mild upper respiratory irritation to severe chronic respiratory and cardiac diseases (Katoto et al., 2019). According to Stats S.A. (2018), cardiovascular disease, respiratory disease, and HIV/AIDS constitute three of the five leading causes of death in South Africa. An estimated 14,356 premature deaths

in 2012 were caused by Acute Lower Respiratory Infection (ALRI), Chronic Obstructive Pulmonary Disease (COPD), lung cancer, Ischemic Heart Disease (IHD), and stroke from all causes (Langerman & Pauw, 2018).

Many countries, including South Africa, have implemented national programmes to manage and regulate ambient air quality and air pollution (Garland et al., 2017). Air Quality Management Plans (AQMPs) are clear outlines of measures and resources needed to execute a strategy or strategies for achieving a particular objective on air quality (DEA, 2018a). The AQMPs set out a course of action to achieve air quality objectives in a given geographical area. To reduce the effects of poor air quality in South Africa, the National Environmental Management Air Quality Act 39 of 2004 (NEM: AQA) requires that all spheres of government and emitters develop AQMPs and emissions reduction management plans (Tshehla & Wright, 2019). The NEM:AQA permitted the establishment of priority areas for

interventions in air quality management to ensure compliance with national air quality management standards and to monitor possible adverse impacts on human health (Wright et al., 2011).

Three national priority areas have been declared to date, with Vaal Triangle Airshed declared in 2006, Highveld declared in 2007 and Waterberg-Bojanala in 2015. Efforts have been put in place to enhance and sustain good air quality in those areas (DEA, 2018a). The Vaal Triangle Airshed Priority Area (VTAPA) faces complex and persistent air pollution challenges due to its extensive commercial, agricultural, residential, industrial, and mining activities close to each other (Scorgie et al., 2003). After years of implementation of the VTAPA AQMP since its publication in March 2009, a second-generation AQMP was developed in June 2020 to define the baseline and assess any improvements to air quality since the initial air quality management plan was initiated (DEFF, 2020). The second-generation AQMP aimed to develop new approaches and action plans, focused on a deeper understanding of the relationships between cause and effect to ensure further progress and eventual compliance (DEA, 2018c).

This study is aimed to evaluate the quality of the AQMP for the VTAPA. The evaluation focused on both the first- and second-generation draft AQMPs. Evaluation of the quality of the second-generation AQMP for the VTAPA will enable the stakeholders of the VTAPA to identify and eliminate any shortcomings of the AQMP should there be any. Lee and Colley (1992) indicate that the success of an Environmental Impact Assessment (EIA) process depends on the quality of environmental statements; therefore, it is fair to assume that the success of any management plans execution depends on the quality of the plan itself. The assessment of the quality of the document used to improve ambient air quality could contribute to the uncovering of the deficiencies not only of the first generation AQMP but also of the second-generation plan (DEA, 2019b). It is only through systematic assessment that we can recognise their particular strengths and weaknesses and determine if their overall output is good enough to provide a framework for ensuring that they meet the desired standard or outcome (Berke & Godschalk, 2009). Efforts to assess their quality are necessary in order to make gradual changes in future versions through the AQMP review process (Hossu et al., 2020).

Sadler (1998) identified four aspects of EIA effectiveness as: 1) The quality of the reports, 2) The effect on decision making, 3) The effectiveness of prediction and management of the impacts, 4) monitoring and post-auditing. Based on Sadler 1996's analysis, it is clear that effectiveness evaluations encompasses more aspects than quality and therefore this study is only limited to quality of AQMP in VTAPA and not effectiveness thereof. The Lee and Colley package has been adopted and adapted to define quality evaluation criteria for the VTAPA AQMP. This criteria was informed by Manual for Air Quality Management Planning (the manual) (DEAT, 2008), secondly, The National Framework for Air Quality Management in South Africa (the framework); and lastly the National Environmental Management: Air Quality Act 39 of 2004 requirements with respect to aspects of an AQMP.

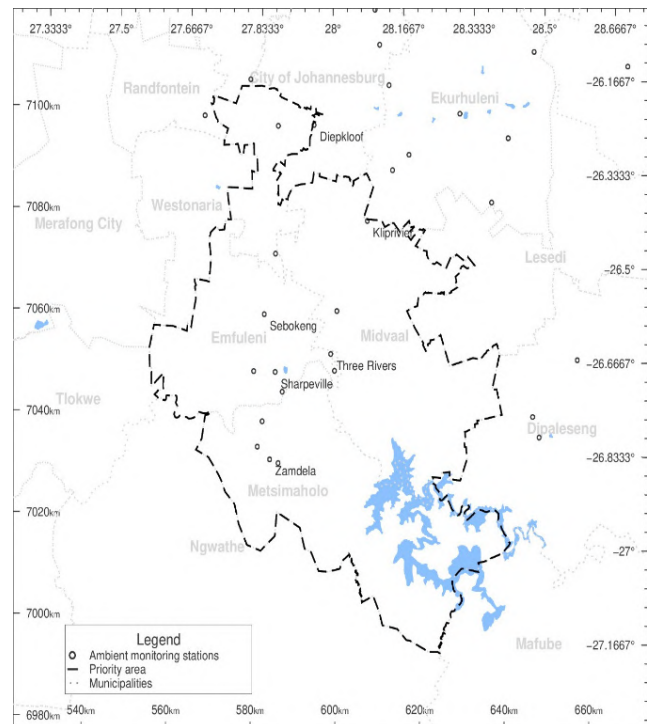


Figure 1: The VTAPA and ambient monitoring sites

Air quality management

According to Gulia et al. (2015) and Sivertsen and Bartonova (2012), developing countries cannot effectively implement AQMPs due to the lack of stakeholder commitment, weak policies, standards and regulations and the absence of air quality data and emission inventories. Further research into understanding all sources of emissions and the identification of unknowns, including eliminating uncertainties, is needed to address all sources of pollution (Kim & Lee, 2018).

The National Environmental Management: Air Quality Act requires the development of a framework for air quality management by the minister. The National Framework for Air Quality Management was first developed in 2007, with a review done in 2012 and 2017 (DEA, 2018a). The framework's successes include establishing the National Ambient Air Quality Standards (NAAQS), three air quality priority areas, and the South African Air Quality Information System (SAAQIS). Priority area management relies on the collaborative effort from all government spheres, industry, and the broader community, and SAAQIS significantly improves the availability of information to establish AQMPs going forward (Scorgie, 2012).

Vaal Triangle Airshed Priority Area

The VTAPA was declared in 2006 and comprised a portion of the City of Johannesburg Municipality, Emfuleni, Midvaal, and Metsimaholo Local Municipalities (Figure 1) (DEA & NWU, 2018; DEA, 2018c; DEAT, 2006). The VTAPA has high emissions from various industrial sources, including a coal-fired power plant, collieries, and quarries. This area is heavily populated with large high-density informal residential settlements, where coal

and wood-burning are typical and have exceeded the health and NAAQS (Anegarn & Scorgie, 1997; DEA & NWU, 2018; DEAT, 2009; Feig et al., 2014; Lindeque, 2018; Mathee & von Schirnding, 2003; Mundackal et al., 2014).

Ambient air quality trend analysis

The South African Air Quality Information System (SAAQIS) holds a live database of ambient air quality across the monitoring stations in the country. It is available for all stakeholders to view (DEA, 2018a). Six ambient monitoring stations (Figure 1) have been set up in the Vaal Triangle locations of Diepkloof, Kliprivier, Sebokeng, Sharpeville, Three Rivers and Zamdela (Sasolburg) (DEA, 2018c; Feig et al., 2014).

Figure 2 is a representation of the particulate matter (PM₁₀ and PM_{2.5}) annual average data from 2007 to 2018 as presented in the "State of Air Report and National Air Quality Indicator" of 2018 (DEA, 2019a). These graphs illustrate the state of compliance of the different areas within the VTAPA with the NAAQS over the past ten (10) years which indicates that almost all are in noncompliance. Fugitive dust, fires, mining, transportation, electricity generation, industrial activities, domestic fuel burning, and traffic are PM sources (Altieri & Keen, 2019; de Lange et al., 2019; Kim et al., 2000). Conclusions made by the State of Air report indicated that PM is the most significant concern due to the numerous pollution sources, even though climatic conditions are an essential factor (DEA, 2019a). Therefore, increased action from national, provincial, and local levels of government would be required to decrease particulate matter concentrations to meet the standards.

It should be noted that a reduction in pollutants cannot solely be a factor of good intervention implementation because,

from the air quality perspective, climatic conditions also play a significant role (Lewis et al., 2020). Furthermore, major policy shifts in the energy, mining and transport sectors would be critical to achieving clean air goals, in addition to the continuous and successful implementation of emission reduction strategies (DEA, 2019a).

Methodology

The Lee and Colley Review Package was adopted and modified in this study to review the quality of the VTAPA AQMP (Lee & Colley, 1992). There has not been a study that evaluates or assesses air quality management plans in South Africa. Research available internationally focuses on the improvement of air quality through different models to compare the ambient air quality before and after management interventions have been put in place (Berhane et al., 2016; Cheng et al., 2019; D'Elia et al., 2009; Ghodousi et al., 2017; Kim & Lee, 2018; Mardones & Cornejo, 2020; Pisoni et al., 2019; Thunis et al., 2017; Wang et al., 2016; Wang & Hao, 2012). This method has been successfully and widely used in the review of the quality of environmental impact assessments and modified for strategic environmental assessment reports, environmental management programmes as well as health impact assessments similar to this study (Anifowose et al., 2016; Bonde & Cherp, 2000; Chang et al., 2013; Cilliers et al., 2015; Fischer, 2010; Fredsgaard et al., 2009; Hallatt et al., 2015; Sandham & Pretorius, 2008; Sandham et al., 2013b; Swanepoel et al., 2019). The LCRP, though seemingly complex, is relatively simple, easy to learn, and easily adaptable with minor changes to suit the application (Lee et al., 1999). The LCRP is robust in that it can be amended to satisfy the legislative requirements of different countries while staying mainly in its original form (Lee & Colley, 1992). In addition, the

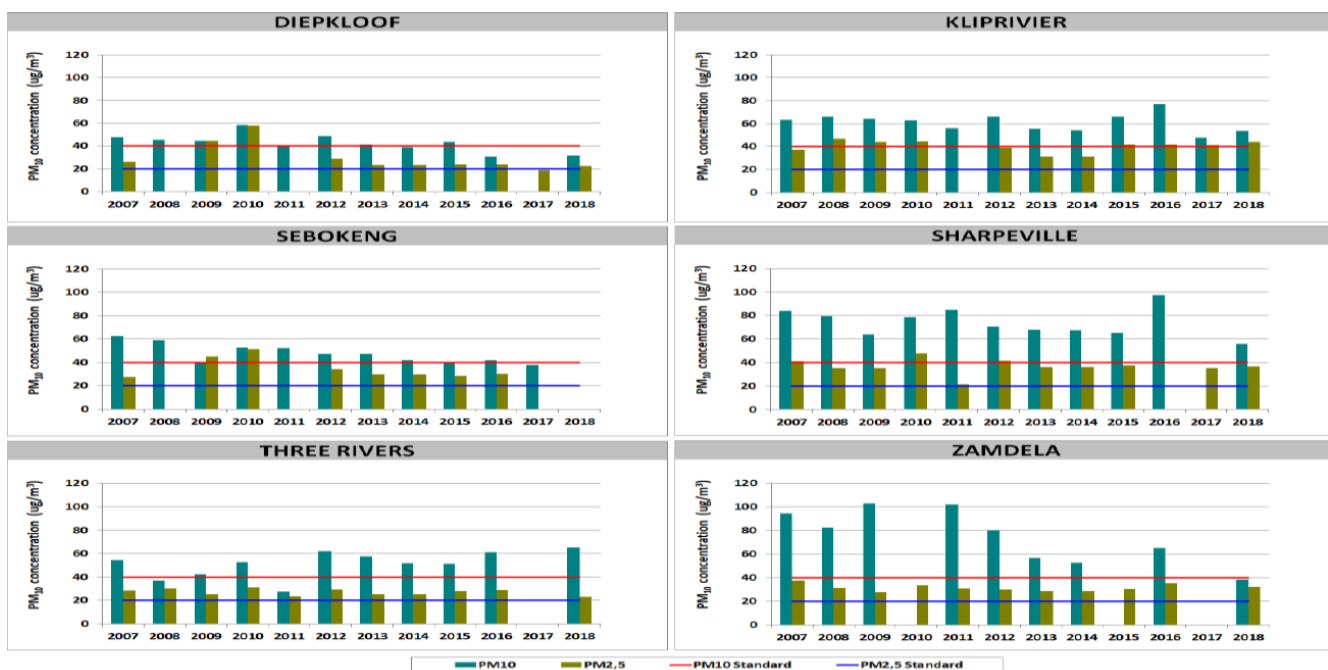


Figure 2: PM₁₀ and PM_{2.5} yearly average data from 2007 to 2018 comparing the Vaal Triangle's compliance with the NAAQS (DEA, 2019a)

LCRP provides the framework and methodology for quality evaluation for any subject. Three documents were used as guidelines in the development of the review package: firstly, the Manual for Air Quality Management Planning (the manual) (DEAT, 2008); secondly, the National Framework for Air Quality Management in South Africa (the framework); and lastly the National Environmental Management: Air Quality Act 39 of 2004.

In this study, a qualitative research method was adopted using a single case study, specifically reviewing the quality of the first-generation and the second-generation draft AQMP for the VTAPA. The VTAPA AQMP was chosen as a case study because among the priority area AQMPs, it was the first AQMP to be developed and the only one with a second-generation plan. Therefore it was essential to assess the quality of the second-generation version of the plan since the air quality in the area could not be sufficiently improved since the development of the first generation AQMP (Tshehla & Wright, 2019).

The LCRP has been successfully and widely used in the review of the quality of environmental impact assessments and has also been widely or adapted or modified for strategic environmental assessment reports, environmental management programmes as well as health impact assessments (Anifowose et al., 2016; Bonde & Cherp, 2000; Chang et al., 2013; Cilliers et al., 2015; Fischer, 2010; Fredsgaard et al., 2009; Hallatt et al., 2015; Sandham & Pretorius, 2008; Sandham et al., 2013b; Swanepoel et al., 2019).

The LCRP requires at least two reviewers to independently do the assessment (Lee et al., 1999). The reviewers later discuss or compare the differences of the assessment outcomes and re-evaluate to resolve the differences in those assessments to decide the terms of the scoring (Pöder & Lukki, 2011). However, similar to used only one reviewer due to similarities in the reviews and very little variance in the two reviewers' assessments, a single reviewer was used for this research.

The methodological principles of the LCRP, as the hierarchical approach and the use of letters for rating, were used in the design of the AQMP review package (Cilliers, 2016; Pöder & Lukki, 2011; Retief, 2007).

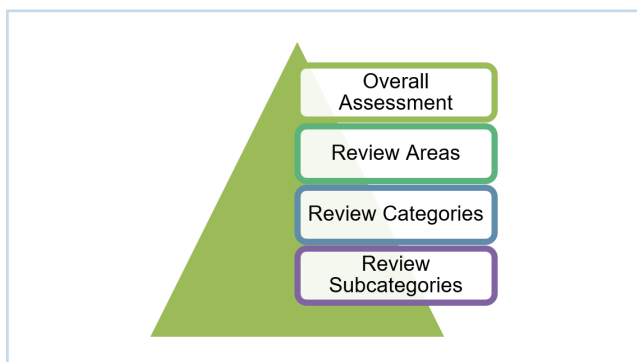


Figure 4: Pyramid structure of Lee and Colley review method (Sandham & Pretorius, 2008)

A review starts at the base of the pyramid with subcategories which are given a score of A-F based on the success of a task, and then the score is combined to provide an overall rating for each category as shown in figure 3 (Sandham et al., 2013a). Subcategories refer to actions/activities that must be done to ensure that the activities listed in the category are performed well (Lee et al., 1999).

Review categories are the activities that need to be undertaken within review areas. Review areas are the main activities of the AQMP (i.e. baseline air quality assessment) (Lee et al., 1999). The A-F sliding score indicates the level of quality, with scores of A-C reflecting the good quality and rating of D-F relatively poor quality (Bonde & Cherp, 2000). As part of the modification LCRP in this study, the categories and subcategories of the review package were developed using the three documents, i.e. the manual, the National Framework, and the Air Quality Act. This study has developed nine (9) review areas for the AQMP review package.

Table 1: A-F assessment score table with explanations (Lee & Colley, 1992)

Symbol	Explanation
A	Relevant tasks well performed; no important tasks left incomplete.
B	Generally satisfactory and complete; only minor omissions and inadequacies.
C	It can be considered just satisfactory despite omissions and/or inadequacies.
D	Parts are well attempted but must, as a whole, be considered just unsatisfactory because of omissions or inadequacies.
E	Not satisfactory, significant omissions or inadequacies.
F	Very unsatisfactory, important tasks(s) poorly done or not attempted.
N/A	Not applicable. The Review topic is not relevant, or it is irrelevant in the context of this statement.

Methodological limitations

The second-generation AQMP for the VTAPA was assessed in its draft format as the plan's development was ongoing at the time of this study. Although there is an inherent risk in analysing a document out for public review, the quality checks done by the Project Steering Committee and the expert panel includes members of academia, regulators, industry experts and NGO's, as well as the National air quality officer, means that the draft document is essentially scientifically correct and has gone through a vigorous vetting process (DEFF, 2020).

Data analysis, results and discussion

Review packages and checklists for quality assessment in environmental impact reports have been the main tools used internationally and locally and consist of set criteria of rating

evaluation tasks against the Environmental Impact Report (Lee et al., 1999; Sandham et al., 2013a). Quality reviews are undertaken to systematically evaluate the strength and weaknesses of plans to judge the overall quality and that they are of a good standard (Berke & Godschalk, 2009). Refer to appendix A for a comprehensive comparison of the review scores per review area. The second-generation draft AQMP was not analysed in Review Areas 8 and 9, these were not analysed because, at the time of the analysis, the second-generation AQMP was still in its draft format and was undergoing its public comment phase of the AQMP development.

Overview of the AQMP – Review Area 1

This study shows that AQMPs for both the first and second generation did well to identify overall and specific goals in line with air quality problem areas of the region. Section 1.4 of the National Framework requires that goals and objectives be "SMART" (specific, measurable, achievable, realistic, and timeous) and informed by section 2 principles of the NEMA. However, the overall goals set out in Section 5.3 of the first-generation AQMP do not have timeframes, thereby not fulfilling the SMART principle. Compliance with the minimum emissions standards and fugitive dust, veld fires, and awareness in the second-generation AQMP were also not SMART because these goals did not outline the timeframe with which the goal would be met although the different objectives may have included timeframes.

Both AQMPs fail to adequately describe the different land uses, topography, landscape, and natural resources, including the socio-economic status of the region. Section 2 of the manual requires that the socio-economic impacts be addressed in all interventions. Therefore, it was pertinent that the socio-economic status should be known. According to the summary of the health study provided for in the second-generation draft AQMP, the socio-economic status of an area and its fuel use make people vulnerable to air pollution (DEFF, 2020). This category scored a C for both AQMPs, indicating that it can be considered satisfactory despite omissions and/or inadequacies.

Implementation of Chapter 3 NEM: AQA – Review Area 2

Despite a health study being conducted, people living in areas where fossil fuels are used for household purposes remains unknown in the VTAPA. A health impact study in the study area determined poor health related to energy use in the household coupled with poor hygiene practices, overcrowding, and lifestyle choices (DEFF, 2020).

Industrial air pollution sources were only identified in the hotspot area in the first-generation AQMP. Dust-generating sources identified in the VTAPA include mining operations, Eskom's ash dump, the petrochemical sector, and iron and steel sectors, which were identified in Section 2.4.2. Section 2.4.2.1 of the first generation AQMP provides no distinction between Section 21 (listed air quality activities) and Section 23 (Controlled Emitters) industries as per Section 16 of the NEM: AQA, which requires a

quantification of Section 21 and Section 23 facilities however it is to be noted that Section 21 and 23 requirements were not published at the time of publication of the first generation AQMP. No obligations with respect to international agreements were referenced. However, best practice guidelines and strategies for local and international examples were used, including the progress and shortfalls of best practices. Furthermore, the roles and responsibilities for all the spheres of government were well laid out.

Section 3.3 of the second-generation draft AQMP report mentions the total number of point sources within the region including the number of Section 21 (listed air quality activities) and Section 23 (controlled emitters). Other pollution sources are well-represented in terms of source profiling. International agreements, best practice principles and plans for improved air quality as required by the Act have not been identified. Although the second-generation draft AQMP mentions that best practice guidelines will be used throughout the document, it does not stipulate which and where this learning is coming. In some instances, further study is needed in terms of best practices. Such an omission does not inspire confidence that adequate research was done on the best practice of air quality. The use of best practice guidelines assists in using methods, techniques, and technologies that are already tried and tested and have proven success or failure (DEAT, 2009).

The lack of capacity within the government may have led to the poor implementation of the identified interventions (DEFF, 2020). This led to the conclusion that the local government option, as a lead for implementation, is inadequate to effectively tackle and comply with air pollution challenges (Gollata & Newig, 2017).

Therefore, both the first and second-generation draft AQMPs were given a score of C.

Air quality goal setting – Review Area 3

Overall, the primary pollutant impacting health identified in the plans is PM_{10} , as outlined in Section 5.2.1 in the first generation and Section 2.2 in the second-generation draft AQMP.

The first-generation AQMP has assessed the regional impacts and greenhouse gases were considered in Section 2.3, 3.1.1, and 4.9 of the report. No evidence could be found that the second-generation draft AQMP had documented environmental and climate change-related impacts as required by subcategories 3.1.2 and 3.1.3 of the review package. Furthermore, regional and transboundary issues are also poorly expressed. Similarly, greenhouse gases and indoor exposure risks are also poorly represented in the second-generation draft AQMP.

The provision of training, institutional building, and information management has been well laid out and maintained from the first-generation to the draft second-generation of the AQMPs. The referencing of legislation throughout both texts is excellent, with obligations to create specific laws achieved.

The first-generation AQMP fared better than the second-generation draft AQMP. The first generation scored overall a B which is satisfactory and complete with only minor omissions and inadequacies. On the other hand, the second-generation draft AQMP was considered just satisfactory, despite omissions and/or inadequacies, scoring a C.

Baseline air quality assessment – Review Area 4

Both AQMPs described the boundaries of the AQMPs well. They included the regions that fall within the VTAPA as well as the affected municipalities. However, the AQMPs did not provide information on the urban populated extensions and the boundaries of the most populated areas.

The climate and climatic conditions such as wind, temperature and precipitation data are well-presented in both AQMPs. The climate and meteorology data are crucial as it indicates the dispersion conditions within the area. The dispersion potential is attributed mainly to climatic conditions, particularly the wind field (DEA, 2018c). Thus, having this information for the baseline assists greatly in modelling the pollution potential of emissions within the area and predicting their movements when analysing wind data. An explanation of the air stability and temperature inversions and impacts thereof were well explained within the first-generation AQMP in Section 4.1 of the baseline report.

Nevertheless, this was not investigated thoroughly in the second-generation draft AQMP. An inversion layer in the atmosphere traps pollutants within that layer. This results in the pollutants being easily transported by the wind. There is a high dispersion potential in the case of strong winds, and the inverse is true for slight breezes (Thomas, 2008). Therefore, this is an essential omission that the second-generation draft AQMP suffers as it describes the behaviour of the pollutants as a direct result of the climatic conditions. Both AQMPs score a B in this review area.

Air quality management system – Review Area 5

A well-placed monitoring network, working optimally under the recommended standards and guidelines, is critical to monitor the pollution trends of the area (DEAT, 2009; DEFF, DEFF Department of Environment Forestry and Fisheries, 2020). Modelling plays a part both in the planning stage of initiatives and in providing insight into the efficacy of intervention after it has been implemented (Lewis et al., 2020). Having confidence in the air quality management systems gives assurance in any analysis done in line with the data provided (DEAT, 2008). A large data set improves the statistical power of research, particularly over a more extended period, accounting for meteorological factors (Lewis et al., 2020).

The air quality monitoring systems reflected for the second-generation draft AQMP report had more than enough information available to ensure that the monitoring network

was well-established and covered the entire study area. Going forward, additional monitoring points have also been identified for the VTAPA to expand the monitoring network (DEFF, 2020).

Regarding the ambient monitoring network for the first-generation AQMP, there were no set standards to measure against as the National Ambient Air Quality Standards had not yet been promulgated when the plan was published. The DEA monitoring stations were only operational for a short time before the development of the AQMP and did not fulfil the three years required for data availability (DEAT, 2009).

The identification of sources was grouped in sectors (industry, commercial, mines and ash dumps, vehicles and domestic fuel burning) and not in terms of point, line and area as required by the manual. The first-generation AQMP scored a B in this review area, while the second-generation AQMP scored an A overall.

Gap analysis and challenges – Review Area 6

Section 3.6 of the manual requires that a gap analysis is conducted. In the first-generation AQMP, the gap analysis was carried out in a problem analysis and objectives setting, whereby a fault tree analysis was conducted in Section 4 of the AQMP for each of the problem complexes identified. In this study, eleven (11) problem complexes were identified and were further divided into emission and non-emission problem complexes. The manual requires that gaps be identified in many ways, including, but not limited to, the inadequacies of monitoring data, emissions inventory, stakeholder consultation, complaints, capacity, and funding constraints, to name a few (DEAT, 2008). Complaints data (complaints lodged) was overlooked entirely in terms of adequately trending the complaints received in the VTAPA, and these could have provided invaluable information for problem areas (DEAT, 2008).

The draft second-generation AQMP complaints data was not considered when evaluating weaknesses and challenges for the plans. However, stakeholder participation was evident in the second-generation draft AQMP, where the voice or opinions of stakeholders were noted as concerns in the implementation of the AQMP for the VTAPA. The first- and second-generation AQMPs score a C and B, respectively, indicating good performance.

Intervention strategies – Review Area 7

The implementation of intervention strategies is proposed to be directly linked to improved ambient air quality (DEAT, 2009). Improved ambient air quality was used as a marker for effective intervention implementation; Section 5 of the second-generation draft AQMP models the emissions performance after successfully implementing all intervention strategies (DEFF, 2020).

Section 5 of the first-generation AQMP highlighted all the intervention strategies implemented in terms of policy and legislative changes, as required by Section 3.9 of the manual.

Several standards, manuals, and publications were planned for in the first generation AQMP as intervention strategies relating to governance. These standards, manuals and publications included the National Ambient Air Quality Standards, National Framework for air quality management, Listed Activities and National Emission Standards, and declaration of small industries as controlled emitters, to name a few significant governance interventions that have since been established.

The first-generation AQMP identified interventions for each air pollution source and specific facilities, including Sasol, Eskom, and Samancor. Furthermore, all the intervention strategies identified indicated the time frames (short-, medium- and long-term) for implementation. Yet, they did not tackle the technical or socio-economic impacts of the intervention as required by the manual in Section 3.8.2.1. Dispersion modelling for potential reduction in pollution after implementing the reduction strategy was not included as required by the manual in Section 3.28.2. The estimated cost for the projects proposed was included for each of the strategies in most cases. However, the benefits associated therewith were not included as required by Section 3.8.2.1 of the manual. Measures to reduce emissions from different sources were well-documented, along with responsibilities to implement the reduction strategies for each problem complex.

The second-generation draft AQMP does not supply the intervention strategies that each facility would implement for emissions reduction however the requirement to comply with the MES provides guidance in terms of the expectation from government in terms of compliance. Each emission source needs to provide information on existing emission reduction strategies (DEAT, 2008). General strategies have been identified, and time frames for execution have been specified from short to long-term. The budget implication for each strategy was also included as required in Section 3.8.2.1 of the manual. The dispersion model predictions showed an observed reduction in emissions (PM_{10} , $PM_{2.5}$, NO_2 , O_3 and SO_2) if all intervention strategies were implemented within the committed timeframes (DEFF, 2020). Measures to reduce emissions from point, line and area sources were well documented, along with responsibilities to implement the reduction strategies. However, in some cases, the responsibility might need to be clarified when the roles are split with different entities (government, NGOs, CBOs) as indicated in the Implementation plans for domestic waste burning and biomass emissions.

The collaboration of Non-Governmental Organisations (NGOs) and/or Community-Based Organisations (CBO) in assisting the municipalities with conducting awareness and educational campaigns are prevalent in both AQMPs. Despite this, it is unclear if there were Public-Private Partnerships (PPPs) to facilitate these agreements. Section 5.9.2.3 of the framework identifies PPPs as one of the strategies that can be used to improve capacity through awareness. The first-generation AQMP indicated collaboration between Eskom, Sasol and the government in implementing the Basa Njengo Magogo top-

down fire making method. However, the second-generation draft AQMP does not have such promises or commitments.

Where the interventions are to be conducted by government organisations, the municipal fund has been identified in the first-generation AQMP. In the second-generation AQMP, there has been no mention of where the funds will be coming from. Section 4.2.5 of the national framework identifies the national and provincial governments as the principal responsibility for these funds. For some projects, it has been recognised as an enabling factor for intervention funds raised from offset projects and social responsibility initiatives projects. These statements do not assure that funds are available, and there is no legal requirement that forces industries to provide funding for projects identified by the department. Industries had already identified viable projects that they will implement as part of the postponement applications when Atmospheric Emissions Licenses (AEL) were issued (SRK consulting, 2019). This concern over the lack of adequate budgetary allocations is identified as a risk that would result in an inability to achieve the goals in the second-generation draft AQMP.

The overall goals, targets and objectives for the VTAPA have been well-summarised for each of the problem complexes in the first- and second-generation draft AQMPs. In terms of goal one (1), specifically of the second-generation draft AQMP, it stipulates that industries need to comply with the Minimum Emissions Standards (MES) by 2025 as an objective. Some industries, however, have received postponements on their MES limits, allowing for postponements of the 2020 MES limits. (DEA, 2018c). Therefore, the industry's expectation will comply with MES by 2025 does not conform to the current situation. Perhaps the timeframe for compliance with this requirement should have been extended up to 2030. Overall, for this Review Area, the first-generation AQMP scored a B, which is generally satisfactory, and the second-generation draft AQMP scored a C which is just satisfactory.

Communication and stakeholder participation – Review Area 8

According to the framework, successful development and implementation of the AQMP relies on the participation of multiple stakeholders (DEA, 2018a). According to the manual, stakeholder participation should take place early at goal setting phase through consultation. It was clear that stakeholder participation has taken place as per the requirements of the manual. This is further supported by the provision of concerns highlighted by stakeholders within the AQMP, which resulted from various stakeholder interventions allowing them to voice their concerns (DEAT, 2009). The AQMP has indicated awareness campaigns and communication channels that ought to be used for communication and stakeholder participation. The MSRSG and workshops were the platforms used for the AQMP development (DEAT, 2009). The first-generation AQMP scored a B overall in this review area, which is generally satisfactory and complete.

Reporting, monitoring and review – Review Area 9

According to Section 3.11.2 of the manual, the annual performance of the AQMP should be provided. The content should include: firstly, the extent to which the AQMP was implemented; secondly, air quality management initiatives; thirdly, compliance of the AQMP to the applicable standards; fourthly, how the priority area performed in achieving the targets; and lastly, any amendments to the plan (DEAT, 2008). The AQMP at the time stipulated that this function was a multi-stakeholder function and, therefore, the MSRSG would be the platform through which this annual performance would be undertaken. A framework was to be developed as part of the AQMP process.

The provision of funding has been identified as a requirement in Section 3.1.2 of the manual, including explaining any budget constraints in Section 3.6. The second-generation draft AQMP refers to various potential funding mechanisms, although it is unclear whether these mechanisms have been finalised. For both plans, there is a barrier to implementing the AQMP because, without funds, most of the mechanisms identified, especially from the regulator's point of view, cannot be implemented.

According to the planning arrangements in the first-generation AQMP, the plan was to be revised after five (5) years unless stipulated otherwise. Eleven years after it was published, a second-generation draft report was published. Among other things, it was delayed by the need to adequately identify the sources of pollution in the area to create interventions that cater to the primary pollution sources (DEFF, 2020). Therefore, the review process of the AQMP failed to meet its commitment of a 5-yearly review. A score of B was determined for the first-generation AQMP.

Conclusion

The main aim of this study was to determine the quality of the VTAPA AQMPs. The quality of the AQMP was determined using the requirements as stated in the manual, the framework and the NEM: AQA, effectiveness evaluation was not undertaken as a part of this paper. Evaluating the quality of the first-generation AQMP could assist in unearthing shortcomings of the first-generation AQMP and those of the second-generation draft AQMP and indicating why the initial plans were not effective.

In general, the first-generation AQMP was of better quality than that of the second-generation draft AQMP. The first-generation AQMP scored a B overall for its quality assessment which is generally satisfactory and complete with only minor omissions and inadequacies. In contrast, the second-generation draft AQMP scored a C, which is just satisfactory despite minor omissions. These documents included goals, fact bases, policies, public participation, and plan provisions for implementation and monitoring as required for plans to be considered of good quality (Lyles & Stevens, 2014).

Several shortfalls were identified which affected both plans, these shortfalls were identified in the following areas: (1) the description of the socio-economic status; (2) the identification of international agreements; (3) the assessment of regional and greenhouse impacts; (4) description of urban population extension and urban agglomeration boundary; (5) complaints data; (6) and lastly, funding. The failure of planners to predict demographic and economic change inevitably restricted the reach of planning at the outset (Talen, 1997). Funding for intervention plans as a primary driver for development was a shortfall identified in this review for both AQMPs. According to (Talen, 1997) (1) political complexity and lack of consensus in society, (2) vagueness and lack of data, and (3) the lack of funding and level of community support are some of the factors that may lead to poor implementation of plans.

Implementing task teams and further awareness and training in different platforms have also been well-outlined. Some intervention strategies were implemented by government departments, which do not form part of the VTAPA Multi Stakeholder Reference Group (MSRSG). To curb waste and tyre burning, the public needs to be made aware of its impacts. Therefore, awareness campaigns need to have a far wider reach than what is allotted in the draft plan.

The intervention strategies identified for the second-generation draft AQMP are more projects-orientated instead of investigation, research and policy-driven, as with the first-generation AQMP. Lewis et al. (2020) note that a reduction in pollutants cannot solely be a factor of good intervention implementation because the atmospheric conditions such as temperature, wind and precipitation also play a significant role from an air quality perspective in terms of pollution dispersion potential.

The lack of capacity within the government may have led to the poor implementation of the identified interventions (DEFF, 2020). This led to the conclusion that the local government option for implementation may be inadequate in effectively tackling and complying with air pollution challenges at a regional scale (Gollata & Newig, 2017). Local authorities have stronger administrative powers; however, they lack sufficient compliance capabilities such as legal and financial backing and are reluctant to enact higher levels of regulation (Gollata & Newig, 2017).

The first-generation AQMP quality assessment reveals that it is of good quality because it met the review package's requirements better than the second-generation draft AQMP. However, the first-generation AQMP still failed to meet the overall objectives set, which includes improving the ambient air quality of the region.

Consequently, the conclusion is that the second-generation draft AQMP requires more input to perform better since it has performed poorly compared to the first-generation AQMP in general. Some of the information that may improve the AQMP quality other than funding includes, but are not limited to: (1)

the description of socio-economic impacts; (2) the identification of areas that use fossil fuels; (3) a reference to international agreements; (4) best practice examples both nationally and internationally; and (5) lastly, using complaints data to outline emission excursion trends. However, there were many improvements that the second-generation draft AQMP provided: the health and the source apportionment studies as well as a better-outlined air quality management system.

Despite being of good quality, the first-generation's implementation did not result in the desired outcomes due to external factors beyond the plan. Therefore, the quality of a plan does not necessarily mean it will be implemented well so long as the external factors impacting upon the plan are not addressed, and therefore high implement ability does not translate to a good plan (Talen, 1997; Tian & Shen, 2011). As discussed by (Talen, 1997), these include but are not limited to political complexities, lack of information, lack of funding and an inability to link cause and effect. The energy, mining and transportation sectors need major policy shifts if the country is to successfully move towards a path of pollution reduction (DEA, 2019a). Interventions created for any management plan need to be effective and yield sustainability in their implementation (Wright & Oosthuizen, 2009).

Recommendations and areas of future research

To improve the quality of the AQMP, funding mechanisms need to be investigated to assist in implementing intervention strategies. The industries that cause any air pollution could be used to generate revenue in the form of environmental pollution taxes/levies for the implementation of AQMP intervention strategies, this can be achieved by having regulations in line with the NEMA polluter-pays principle. This can be achieved by having a regulation in line with the NEMA polluter-pays principle like the implementation of the Carbon Tax Act.

Future research in this field should include evaluating and comparing the quality and effectiveness of AQMPs in the different air quality Priority Areas; the AQMP review package can be used in further studies evaluating the quality of other AQMPs, and a should be developed to assess effectiveness.

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Appendix A Summary tables of review areas 1-9

RA, Category, subcategory	First-generation	Draft Second generation
1. Overview of the AQMP	C	C
1.1 The AQMP clearly sets the goals and objectives of the VTAPA AQMP	A	A
1.1.1 The overall goals of the AQMP have been identified	A	A
1.1.2 The specific goals of the AQMP have been identified	A	A
1.1.3 The goals and objectives are linked to the specific air quality problems of the area	A	A
1.1.4 Objectives are SMART (Specific, Measurable, Realistic and Timeous)	C	B
1.2 Geographical area within which the AQMP will be implemented are described	C	C
1.2.1 The geographical area is mapped out	A	A
1.2.2 Description of the area, land use, topography, landscape and natural resources are described	D	D
1.3 The description of socio-economic status is provided for	F	F
1.3.1 The description of demographics is addressed	F	F
1.3.2 Socio-economic status is outlined	F	F

	First-generation AQMP	Draft second-generation AQMP
2. Implementation of Chapter 3 of Air Quality Act – Section 16 and 17	C	C
2.1 Identification of the main air pollution sources	A	A
2.2 Identification of areas where fossil fuel are used for domestic use	D	F
2.2.1 List of areas that use fossil fuel for domestic use	C	C
2.2.2 Health status of persons living in areas of high fossil fuel use for domestic use	F	D
2.3 Identification of all industrial air pollution sources	D	D
2.3.1 Inventory of Section 21 industries	D	C
2.3.2 Inventory of Section 23 industries	F	C
2.3.3 Inventory of industries generating dust	D	B
2.3.4 Inventory of industries that may cause air pollution but are not listed	C	F
2.4 International Agreements	F	F
2.5 Best practice guidelines identified for air quality management and listed	A	F
2.6 Roles and responsibilities	B	B

	First-generation AQMP	Draft second-generation AQMP
3. Air Quality goal setting	B	C
3.1 Identification of primary and secondary pollutants of concern	B	C
3.1.1 Pollutants impacting health	B	B
3.1.2 Environmental impact-related	B	F
3.1.3 Climate change-related	D	F
3.2 Assessment of impact of industrial activities	A	A
3.3 Assessment of regional issues	D	D
3.4 Assessment of greenhouse gases	C	D
3.5 Assessment of indoor exposure	D	D
3.6 Provision made for training, institutional building and information management	A	A
3.7 Reference made to compliance with legislative requirements	A	A

	First-generation	Draft Second generation
4. Baseline air quality assessment	B	B
4.1 Area description and definition	C	C
4.1.1 Description of administrative boundaries	A	A
4.1.2 Description of region or municipality	A	A
4.1.3 Description of Priority Area definition	B	A
4.1.4 Description of urban populated extension	F	F
4.1.5 Description of urban agglomeration boundary	F	F
4.2 Meteorology and climate description	A	B
4.2.1 Description of the climate of the area	A	A
4.2.2 Presentation of wind, temperature and precipitation data	A	A
4.2.3 Description of air stability and temperature inversions	A	C
4.3 Information about the population distribution and population density presented	B	B
4.4 Evaluation of baseline air quality data	B	B
4.4.1 Description of air quality monitoring programmes	B	B
4.4.2 Description of quality assurance and quality control (QA/QC) programme	B	C
4.4.3 Description of current air quality	B	B
4.4.4 Identification of sources and emissions	A	A
4.4.5 National and Provincial requirements	F	B
4.4.6 Adequacy of AQM structures	C	B
4.4.7 Inventory of current procedures and methods	A	A

	First-generation	Draft Second generation
5. Air Quality Management system	B	A
5.1 Air quality monitoring	B	A
5.1.1 Ambient air quality monitoring network	C	A
5.1.2 Location of monitoring stations	B	B
5.1.3 Source monitoring	A	A
5.1.4 Continuous meteorological monitoring	C	A
5.2 Emissions inventory	B	A
5.2.1 Identification of types of sources – point, line, area	C	A
5.3 Atmospheric dispersion modelling conducted for the area	B	A

	First-generation	Draft Second generation
6. Gap Analysis and challenges	C	B
6.1 A gap analysis conducted	C	B
6.2 Description of pressures and challenges	C	B
6.3 Problems associated with enforcement and compliance	C	A
6.4 Stakeholder consultation	B	B
6.5 Complaints data	F	F
6.6 Description of problems associated with enforcement and compliance	B	B

	First-generation	Draft Second generation
7. Intervention strategies	B	C
7.1 General intervention strategies available	A	C
7.1.1 Intervention strategies on policy implementation and legislative changes	A	B
7.1.2 Intervention strategies on the use of international best practice	A	D
7.2 Intervention strategies relating to air quality	C	B
7.2.1 Air pollution source has identified existing emission reduction initiatives and their effectiveness	B	C
7.2.2 Potential reduction strategies – short-, medium- and long-term	A	A
7.2.3 A description of each strategy	C	C
7.2.4 Dispersion modelling	F	A
7.2.5 Estimation of expected costs and benefits	B	B
7.2.6 Roles to implement the reduction strategy	A	C
7.2.7 Measures to reduce emissions from mobile sources identified	B	B
7.2.8 Measures to reduce emissions from area sources	A	A
7.3 Implementation of intervention strategies	B	C
7.3.1 Implementation task teams	A	A
7.3.2 Short-, medium- and long-term actions	A	A
7.3.3 Conducting awareness and educational campaigns	A	A
7.3.4 Public-Private Partnership	C	F
7.3.5 By-Laws developed	D	F
7.3.6 Funding	D	F
7.3.7 A summary of the entire AQMP is provided	C	C

	First-generation	Second-generation draft
8. COMMUNICATION AND STAKEHOLDER PARTICIPATION	B	
8.1 Stakeholder participation	A	
8.2 Awareness campaign and communication	C	
8.2.1 Engaging with the stakeholders	C	
8.2.2 Stakeholders to participate in the AQM planning process	B	
8.2.3 Workshops with interested and affected parties	A	
8.2.4 Awareness raising and building capacity	D	
8.3 Examples of successful implementation	B	
8.4 Benchmarking	B	

	First-generation AQMP	Second-generation draft AQMP
9. REPORTING, MONITORING AND REVIEW	B	
9.1 Reporting	A	
9.2 Monitoring	B	
9.2.1 Strategic issues	B	
9.2.2 Communication and public participation	B	
9.2.3 Financial plan	F	
9.2.4 Air quality management implementation programme	D	
9.2.5 Review of AQMPs	C	



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- Air Quality Management Plans (AQMP)
- Air Quality Impact Assessments (AQIA)
- Emissions Inventories (EI)
- Dispersion Modelling (DM)
- Atmospheric Emission License (AEL) Applications

Air Quality Monitoring

- Continuous Ambient Air Quality And Meteorological Monitoring
- Stack Monitoring
- Dustfall Monitoring (SANAS Accredited)

Climate Change

- Framework Strategies
- GHG Emission Inventories

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Research article

Source apportionment of ambient fine and coarse aerosols in Embalenhle and Kinross, South Africa

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Received: 23 August 2021 - Reviewed: 16 October 2021 - Accepted: 22 November 2021

<https://doi.org/10.17159/caj/2020/31/2.11980>

Abstract

The South African Highveld is recognised as a region having significant negative ambient air quality impacts with its declaration as an Air Quality Priority Area in 2007. Such areas require the implementation of specific air quality intervention strategies to address the air quality situation. A greater understanding of the composition of the atmospheric aerosol loading and the contributing air pollution sources will assist with the formulation and implementation of these strategies. This study aims to assess the composition and sources of the aerosol loading in Embalenhle and Kinross located on the Highveld. Fine ($PM_{2.5}$) and coarse ($PM_{2.5-10}$) aerosol samples were collected during summer and winter, which were quantified by gravimetry. Wavelength-Dispersive X-Ray Fluorescence (WD-XRF) spectrometry and Ion Chromatography (IC) analysis were applied to determine the chemical compositions of aerosol samples on filters. Mean $PM_{2.5}$ concentrations in Embalenhle and Kinross ranged from 16.3 to 34.1 $\mu\text{g}/\text{m}^3$ during winter and 7.4 to 19.0 $\mu\text{g}/\text{m}^3$ during summer. Mean $PM_{2.5-10}$ concentrations ranged from 10.3 to 114 $\mu\text{g}/\text{m}^3$ during winter and 5.9 to 11.2 $\mu\text{g}/\text{m}^3$ during summer. Si, Al, S, SO_4^{2-} and NH_4^+ were the most abundant species in $PM_{2.5}$ during both seasons. The elements Na and Ca were also abundant at both sites during winter and summer, respectively. In $PM_{2.5-10}$, Si, Al, SO_4^{2-} and F were the most abundant species during both seasons. The element Na was also abundant at both sites during winter with S and Ca also having high abundances at Embalenhle and Kinross, respectively, during summer. Source apportionment was performed by Positive Matrix Factorisation (PMF), which resolved five sources. Dust, secondary aerosols, residential combustion, wood and biomass burning, and industry were determined to be the contributing sources. Any measures to mitigate particulate air pollution on the Highveld should consider these key sources.

Keywords

Particulate matter, source apportionment, positive matrix factorisation, backward trajectories.

Introduction

South Africa, and especially the industrialised Highveld, is a significant anthropogenic and natural aerosol region of the southern Hemisphere (Held, 1996). South Africa has one of the three largest industrialised economies on the African continent and boasts significant mining and metallurgical industries (Venter et al., 2012). It is the largest regional energy producer on the continent, generating most of the electricity in coal-fired power plants (Tiitta et al., 2014; Josipovic et al., 2019). The Highveld region has been recognised as a region having significant negative ambient air quality impacts and as such, was declared an Air Quality Priority Area on 23 November 2007. Power generation activities are located on the Highveld close to extensive coalfields. Other significant emission sources include household fuel burning, motor vehicles, heavy industries and mining activities (DEA, 2012).

Exposure to elevated concentrations of fine particulate matter is of significant health concern in several regions of South Africa (Altieri and Keen, 2019). The adverse human health effects associated on exposure to elevated levels of particulate matter have been well documented by the World Health Organisation (WHO). They include a range of acute and chronic respiratory and cardiovascular effects, irreversible changes in physiological function (e.g., lung function) and premature mortality (WHO, 2006). There is a need to develop strategies to improve the air quality and diminish the health risks associated with airborne particulate matter. A greater understanding of the composition of the atmospheric aerosol loading and the contributing air pollution sources is an essential step to achieving this.

Source apportionment is used to identify air pollution sources and to quantify their contribution to ambient pollution levels

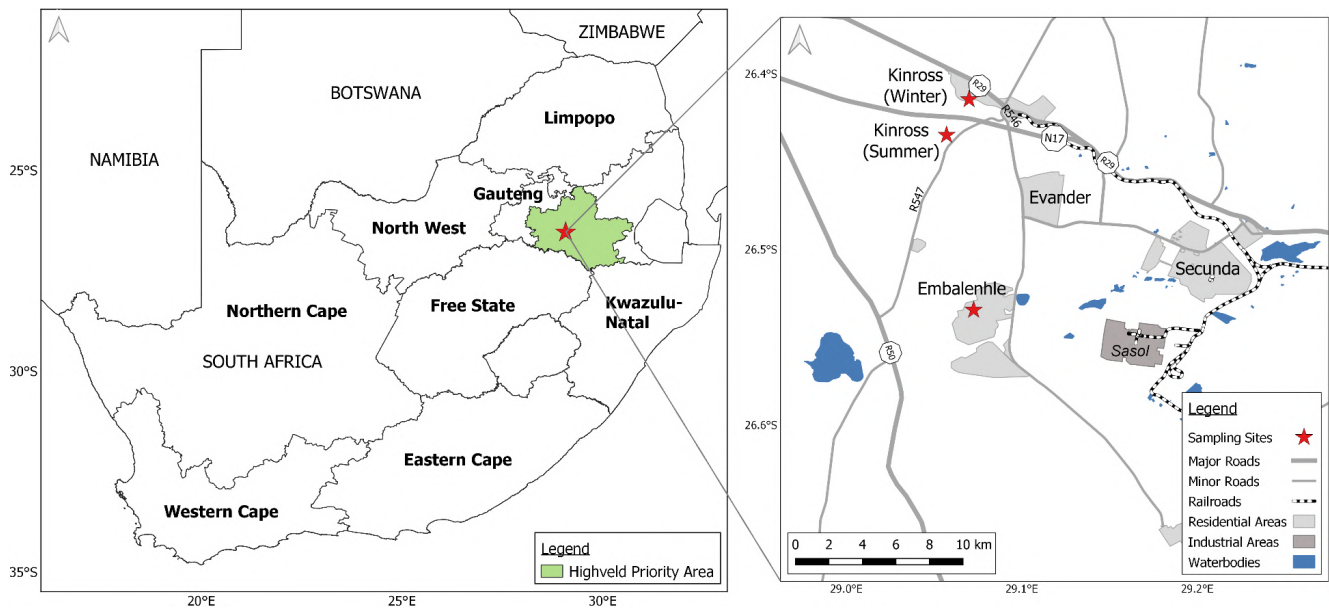


Figure 1: Location of the HPA in South Africa and the aerosol sampling sites in Embalenhle and Kinross

in an airshed (Belis et al., 2014). Different source assessment methods include emission inventories, dispersion models and receptor models (Viana et al., 2008; Belis et al., 2014). Receptor models commonly utilised include, amongst others, Positive Matrix Factorisation (PMF) and Chemical Mass Balance (CMB). A key determinant in selecting an appropriate model for a particular airshed is the degree of knowledge required about the contributing sources, prior to the application of a receptor model (Viana et al., 2008). Multivariate models such as PMF require less knowledge of the contributing sources, while CMB requires detailed understanding of the sources and their compositions.

Within South Africa, several aerosol source apportionment studies have been undertaken to understand aerosol characteristics and their attributions. Annegarn et al. (1998) used the CMB model to apportion particulate matter to sources in Soweto township in the Gauteng Province. The CMB model was also applied to studies in the Vaal Triangle in the Gauteng Province (Engelbrecht et al., 1996a, 1998), Qalabotjha in the Free State Province (Engelbrecht et al., 2002). Olifantsfontein on the Highveld (Engelbrecht et al., 1996b) and Nelspruit in the Mpumalanga Province (Engelbrecht et al., 1996c). However, only a few studies have applied the PMF model. Tiitta et al. (2014) used PMF to characterise the organic aerosol properties of aerosol mass spectra from a grassland site in the North West Province (Welgegund). More recently, Tshehla and Djolov (2018) used PMF to apportion sources of particulate matter from an industrialised rural area (Steelpoort and surrounding areas) in the Limpopo Province. The primary sources identified from source apportionment studies in South Africa include mineral dust, industry, residential fuel burning, sea salt and biomass burning (Maenhaut et al., 1996; Annegarn et al., 1998; Piketh et al., 1999; Engelbrecht et al., 2002). However, for the most part, such studies are limited in South Africa, with additional research required to understand other aerosol sources and their contributions (Mathuthu et al., 2019).

This study aims to identify the air pollution sources contributing to the aerosol loading at two residential areas located on the Highveld, i.e., Embalenhle and Kinross, as well as to apportion their contribution to the aerosol loading by applying the PMF model.

Materials and methods

Site description

The Highveld Priority Area (HPA) covers an area of 31 106 km² and forms part of South Africa's elevated inland plateau. The terrain of the HPA is relatively flat, ranging from approximately 1400 m above sea level in the north-west to 1900 m above sea level in the southeast. The HPA experiences a temperate climate with warm, wet summers and cool, dry winters.

Aerosol sampling was undertaken at two sites in the HPA, namely, Embalenhle and Kinross in the Mpumalanga Province (Figure 1). Sampling was undertaken at Embalenhle school (-26.535 °S, 29.073 °E) and Kinross (-26.435 °S, 29.057 °E in summer and -26.415 °S, 29.070 °E in winter). Embalenhle is a low-income residential area, approximately 8 km to the west of the town of Secunda. Kinross is a low-density residential area located about 12 km north of Embalenhle. Kinross was selected as an upwind sampling site to determine any other additional significant contributing sources in the area (other than emissions from Embalenhle).

Both residential areas are located close to a major petrochemical refinery and associated open-cast mining activities. Matla and Kriel coal-fired power stations are located approximately 27 km and 30 km, respectively, to the north of these two sites. Based on the Census 2011, domestic fuels such as coal, wood and paraffin continue to be used in Embalenhle even though most households are electrified.

Aerosol sampling

Aerosol sampling was undertaken during winter from 15 to 31 July 2016 and 5 to 17 August 2016 and during summer from 14 to 28 February 2017 at Embalenhle and Kinross.

Sampling was undertaken using the 'Gent' stacked filter unit (SFU) sampler (Maenhaut et al., 1994; Hopke et al., 1997). The sampler collects particles with an aerodynamic diameter (AD) $\leq 10 \mu\text{m}$ in separate coarse and fine fractions. The coarse fraction corresponds to aerosols collected with an AD between 10 and $2.5 \mu\text{m}$, while the fine fraction corresponds to aerosols collected with an AD $\leq 2.5 \mu\text{m}$. Two 47-mm Nuclepore® polycarbonate filters with pore diameters of $8.0 \mu\text{m}$ and $0.4 \mu\text{m}$ are sequentially used during sampling. The flow rate was set to 16 L/min. The filters were weighed prior to and after sampling using an XP26 DeltaRange Microbalance (Mettler-Toledo AG, Greifensee, and CH), to determine the deposited gravimetric mass after 12 hours of exposure. Samples were collected twice a day, from approximately 10:00 – 22:00 and 22:00 – 10:00 to capture the bimodal concentration peak. After sampling, the Nuclepore® filters were stabilised for 24 hours in a temperature and humidity-controlled environment, prior to weighing and then placed in a stabilised environment prior to chemical analysis. Blanks were also collected during both sampling campaigns, comprising 10% of the total collected samples during each campaign.

Chemical analyses

Elemental Analysis

Wavelength-Dispersive X-Ray Fluorescence (WD-XRF) spectrometry analysis was performed using a Panalytical Axios^{max} spectrometer. This instrument implemented sequential sampling of the selected element and has a rhodium anode x-ray tube with a 4 kW generator. The system uses helium gas as a medium within the analysis chamber. An area with a 20 mm diameter was analysed on each sampled filter. MICROMATTER™ calibration standards were used for calibration purposes, which are National Institute of Standards and Technology (NIST) traceable reference materials. These standards have a Nuclepore® polycarbonate aerosol membrane backing mounted in a 25 mm ring mount. Each element had two calibration points, i.e., a very light standard ranging between 3 - $8 \mu\text{g}\cdot\text{cm}^{-2}$ and a heavier standard ranging between 40 - $60 \mu\text{g}\cdot\text{cm}^{-2}$.

Elements determined using SuperQ software included: sodium (Na), magnesium (Mg), aluminium (Al), silicon (Si), phosphorus (P), sulphur (S), chlorine (Cl), potassium (K), calcium (Ca), titanium (Ti), vanadium (V), chromium (Cr), manganese (Mn), iron (Fe), nickel (Ni), zinc (Zn) and lead (Pb). All measurements were corrected by the average of the blank values from each campaign.

Ionic Analysis

Ion chromatography (IC) analysis of the water-soluble aerosol fraction was undertaken with a Dionex ICS-3000 system consisting of two flow lines (Conradie et al., 2016). One flow

line was used to detect anion species and the other flow line for cation species. Before chemical analysis was commenced, the filters were leached in 10 mL de-ionised water in an ultrasonic bath for 30 min. A five-point calibration from 20 ppb to 500 ppb, was conducted using certified stock solutions obtained from Industrial Analytical (Muyemeki et al., 2021). Ionic species determined included: fluoride (F⁻), chloride (Cl⁻), nitrate (NO₃⁻), sulphate (SO₄²⁻), sodium (Na⁺), ammonium (NH₄⁺), potassium (K⁺), calcium (Ca²⁺), magnesium (Mg²⁺) and the organic acids, formic, acetic and oxalic acid. All measurements were corrected by the average of the blanks from each campaign.

Meteorological data

Temperature, relative humidity and rainfall data were obtained from the South African Weather Services' weather station in Secunda (-26.497 °S; 29.186 °E), while wind speed and wind direction were retrieved from the ambient air quality monitoring station operated by Sasol in Embalenhle (-26.552 °S; 29.112 °E).

Positive matrix factorisation

PMF is a multivariate factor analysis technique that deconstructs a matrix of speciated sample data into two matrices: factor contributions and factor profiles (Norris et al., 2014). The PMF model equation can be expressed as follows:

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

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

In this paper, source apportionment was undertaken using the EPA PMF 5.0 software package (Norris et al., 2014). Model input included receptor (ambient) concentrations and realistic uncertainties. For the ambient files, the water-soluble ionic form instead of the elemental form was retained for Cl⁻, Na⁺ and K⁺. K⁺ was selected over K as a better biomass burning tracer species. Species that were below the detection limit (BDL) in all samples collected were excluded from further analysis. Negative values in the dataset were recognised as BDL values and were replaced by half of the method detection limit (MDL). The corresponding uncertainties were calculated as $(5/6) \times \text{MDL}$ (Polissar et al., 1988). The MDL for each element was defined as three times the standard deviation of the average values of the blank filters (Barbaro et al., 2019) (supplementary Table 1). For concentrations greater than the MDL, uncertainties were calculated using the formula (Norris et al., 2014):

$$\text{Unc} = \sqrt{(\text{Error Fraction} \times \text{Concentration})^2 + (0.5 \times \text{MDL})^2} \quad (2)$$

Three to six PMF factors were evaluated in the model. Each of these models was run 100 times using randomised seeds. The final solution was selected on the basis of the interpretability of the factors and the results of the model error estimation

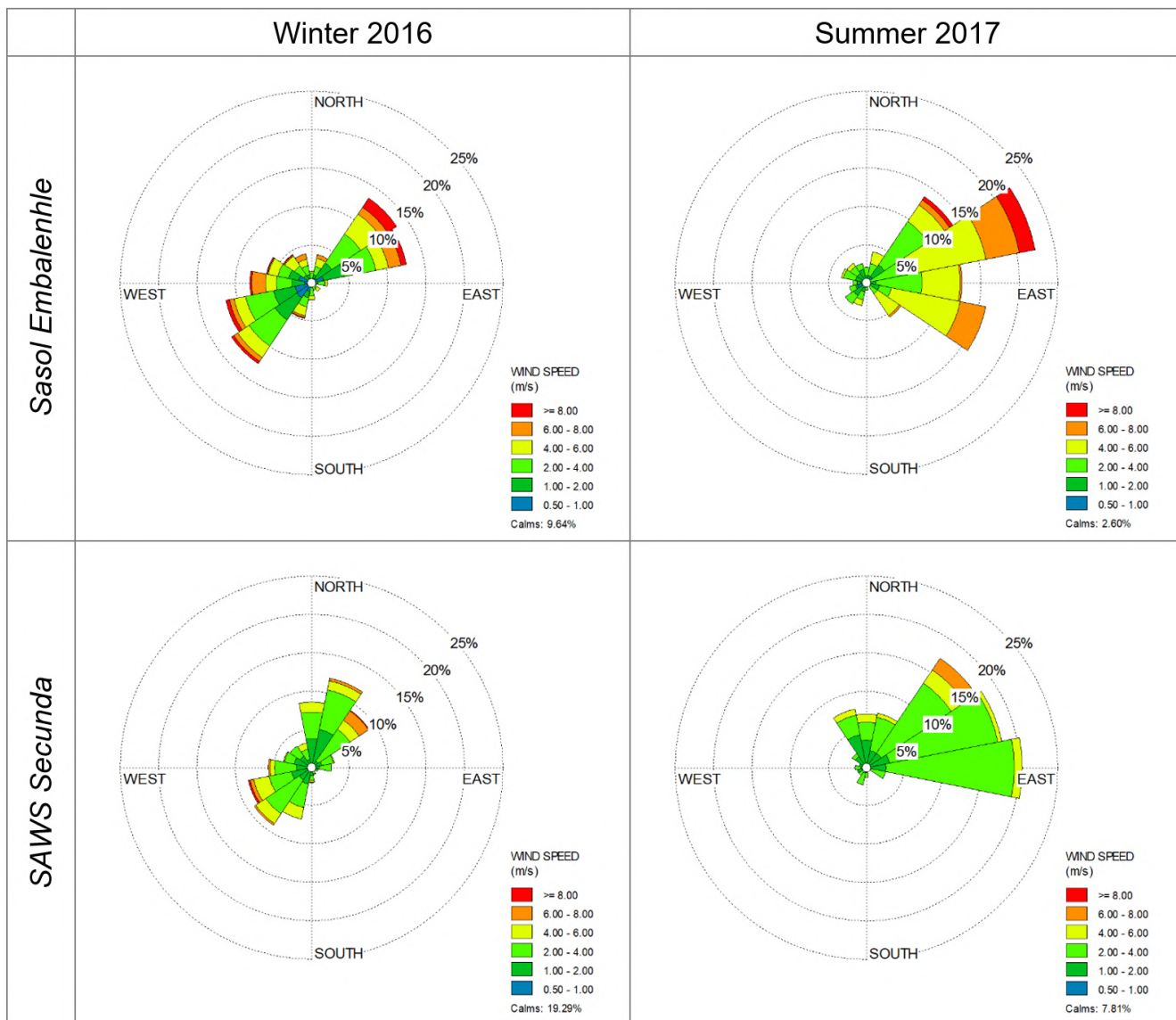


Figure 2: Wind field of Embalenhle and Secunda during the sampling campaigns

methods. Source types were assigned to each PMF factor based on known indicator species and by comparison to known source profiles from the USEPA’s SPECIATE database (USEPA, 1999). It is recognised that source profiles vary considerably across different regions of the world and where available, comparison was made with local source profiles (Engelbrecht et al., 2000, 2001, 2002).

Particulate mass was included in the PMF model as a ‘total variable’. In this study, the reconstructed particulate mass was used in the PMF model instead of the measured gravimetric mass as this produced a more stable solution. Reconstruction of the gravimetric mass included the following aerosol components: (1) inorganic ions of SO_4^{2-} , NO_3^- and NH_4^+ ; (2) geological minerals = $2.20 Al + 2.49 Si + 1.63 Ca + 1.94 Ti + 2.42 Fe$ known as the IMPROVE ‘soil’ formula; (3) sea salt, estimated as $Cl + 1.4486 Na$, where 1.4486 is the ratio of the concentration of all elements except Cl in sea water to the Na concentration in

sea water (Maenhaut et al., 2002); (4) trace elements (excluding S and geological minerals) summed in their elemental form; (5) remaining mass or ‘others’, which was assumed to be non-crustal K based on the formula $K - 0.6 Fe$ (Maenhaut et al., 2002), the sum of organic acids (Putaud et al., 2000) and other species unaccounted for. The detailed methodology is outlined in Chow et al. (2015).

Air mass trajectory analysis

For each sampling campaign, the air mass history was analysed using back trajectories calculated with the HYbrid Single-Particle Lagrangian Integrated Trajectory (HYSPPLIT) version 4.8 modelling software. This software was developed by the National Oceanic and Atmospheric Administration (NOAA) Air Resources Laboratory (ARL) (Stein et al., 2015; Rolph et al., 2017). The model was run with the GDAS meteorological archive (spatial resolution of $1^\circ \times 1^\circ$) produced by the US National Weather Service’s National Centre for Environmental Prediction

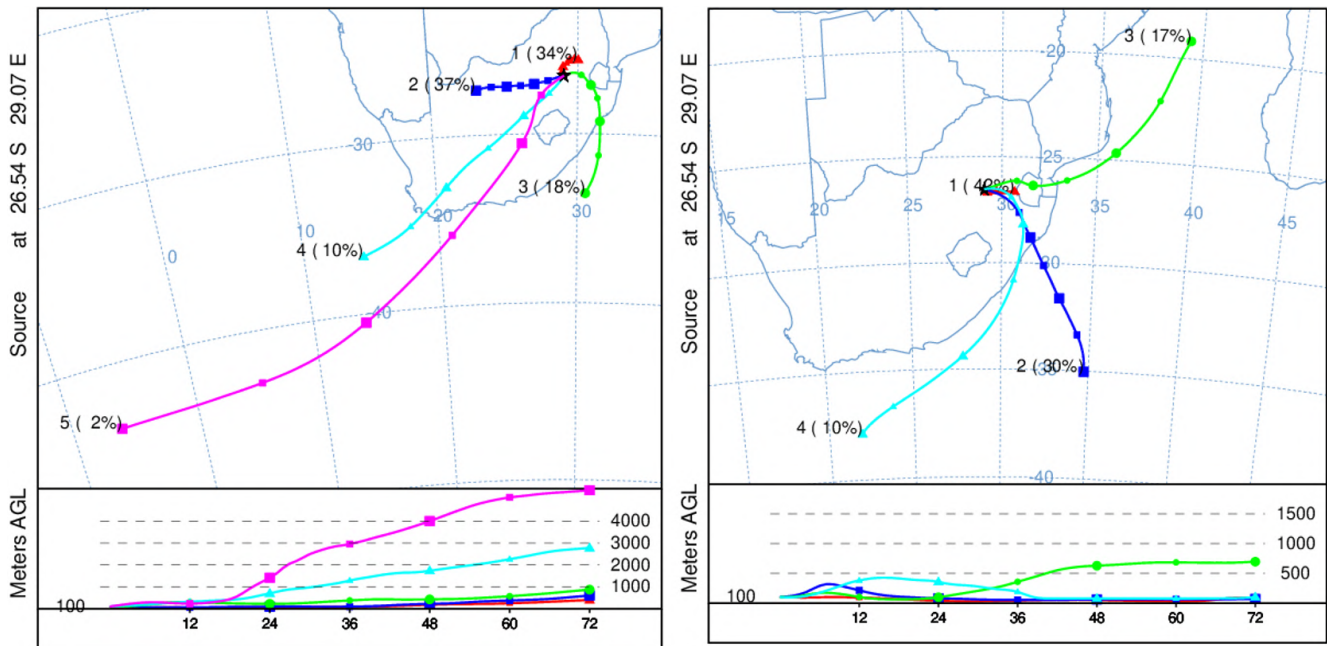


Figure 3: 72-hour backward trajectory cluster means for air masses arriving at Embalenhle during the winter 2016 (left) and summer 2017 (right) sampling campaigns

and archived by ARL (Air Resources Laboratory, 2012). Individual 72-hour back trajectories for air masses at an arrival height of 100 m above ground level were calculated. The geomorphology in HYSPLIT is not well defined therefore an arrival height of 100 m above ground level was selected as lower arrival heights could result in increased error margins on single trajectory calculations (Tiitta et al., 2014). Errors for individual trajectories range from 15 to 30% of the back trajectory distance travelled (Stohl, 1998; Riddle et al., 2006). The individual trajectories were then grouped into clusters of similar air mass origins using the trajectory cluster analysis tool in the HYSPLIT model. The number of clusters retained was determined by the percent change in the total spatial variance (TSV) whereby a significant increase in the change of TSV indicates that ‘different’ rather than ‘similar’ clusters are being paired (Draxler et al., 2020).

Results and discussion

Meteorological conditions

During the winter sampling campaign in 2016, a high-pressure system dominated most of the sampling period, resulting in sunny and cool to warm conditions. Daily average temperatures ranged from -1.0 °C to 26.2 °C. From 24 to 27 July 2016, an upper-air cut-off low prevailed, resulting in rainfall and a subsequent reduction in aerosol concentrations. Winds were moderate to fast and originated predominantly from the northeasterly and southwesterly sectors (Figure 2).

A low-pressure surface trough prevailed for most of the summer sampling campaign in 2017. For the first few days of the sampling campaign, a tropical storm to the east of the country resulted in rain in Embalenhle on 14 February 2017. Rainfall was

also experienced from 19 – 25 February 2017. Rainfall is one of the main mechanisms for scavenging aerosol particles from the atmosphere (Zheng et al., 2019). Daily average temperatures were warm to hot and ranged from 11.9 °C to 30.2 °C. Surface winds remained moderate to high, with winds originating mainly from the northeast, east-northeast and east (Figure 2).

Long-range transport clusters

Two dominant low-level air masses influenced the area during winter (Figure 3). Air mass 1 (accounting for 37% of the trajectories) originated from the west and passed over the mining areas of the western Bushveld Igneous Complex in the North-West Province. A significant portion of the global ferrochromium and platinum group metals is produced in this region (Venter et al., 2016). Air mass 2 (accounting for 34% of the trajectories) originated from the northeast via a short pathway. This air mass passed over the Mpumalanga Highveld before arriving at the sites.

The remaining three high-level trajectories (together accounting for 30% of the trajectories) originate from the Atlantic and Indian Oceans.

During summer, four air masses were identified (Figure 3). Air mass 1, originating via a short pathway from the east, was the dominant air mass and accounts for 43% of the trajectories. The remaining three trajectories (accounting for 30%, 17% and 10% of the trajectories) originated from the Atlantic and Indian Oceans.

Gravimetric concentrations

The ambient mass concentrations measured in the two size fractions at Embalenhle and Kinross during the sampling

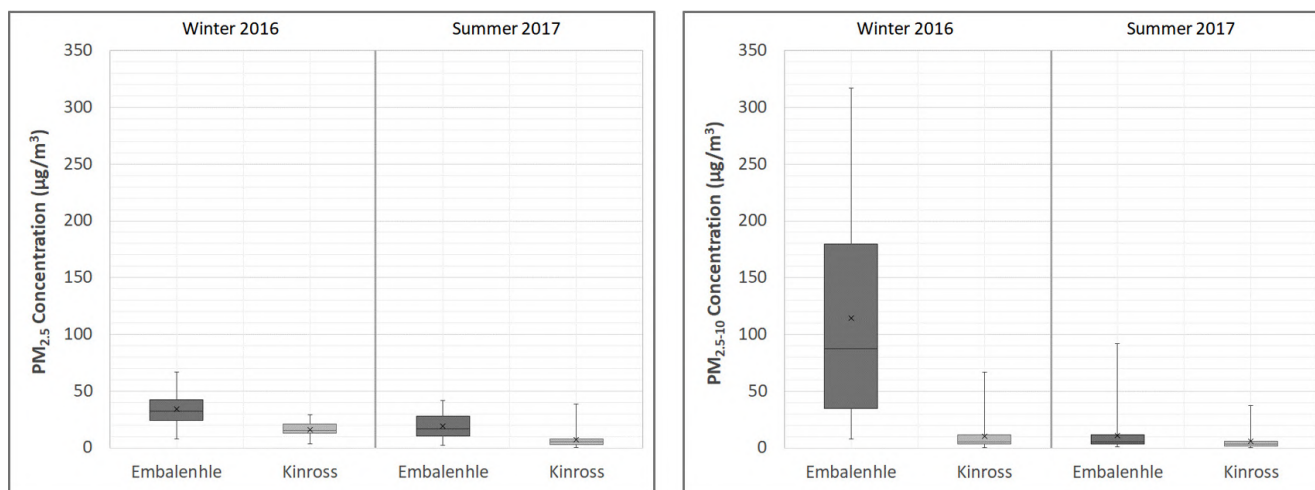


Figure 4: Ambient $PM_{2.5}$ (left) and $PM_{2.5-10}$ (right) mass concentrations at Embalenhle and Kinross during the sampling campaigns

campaigns are presented in Figure 4. It is evident that the median $PM_{2.5}$ and $PM_{2.5-10}$ concentrations are higher at Embalenhle than the upwind site of Kinross during both seasons. This indicates the contribution of localised air pollution sources in the township, especially during the winter months when residential fuel burning is most widespread.

During winter, median $PM_{2.5}$ concentrations were $32.5 \mu\text{g}/\text{m}^3$ and $15.6 \mu\text{g}/\text{m}^3$ at Embalenhle and Kinross, respectively, decreasing to $16.9 \mu\text{g}/\text{m}^3$ and $5.6 \mu\text{g}/\text{m}^3$ at Embalenhle and Kinross, respectively, during summer. Median $PM_{2.5-10}$ concentrations were substantially higher at Embalenhle during winter ($87.6 \mu\text{g}/\text{m}^3$), decreasing to $5.4 \mu\text{g}/\text{m}^3$ during summer. At Kinross, median $PM_{2.5-10}$ concentrations of $5.3 \mu\text{g}/\text{m}^3$ and $3.5 \mu\text{g}/\text{m}^3$ were recorded during winter and summer, respectively.

The seasonal difference in aerosol concentrations is reflective of the changes in source emission strength and the prevailing meteorological conditions during each season.

Chemical composition

The elemental and ionic composition of $PM_{2.5}$ and $PM_{2.5-10}$ at each site during winter and summer are graphically represented in Figure 5. The mean concentrations and standard deviations are numerically presented in supplementary Tables 2 and 3. Spearman correlation coefficients are numerically given in supplementary Tables 4 – 11.

Elemental

The elements Si, Al and S were the dominant species at Embalenhle and Kinross during both seasons in $PM_{2.5}$. Na was also abundant at both sites during winter and Ca during summer. Strong correlations are seen between Si, Ca, Fe, Na, Mg, Al and Ti suggesting either a crustal origin or coal combustion origin of these species (Maenhaut et al., 1996). The abundance of Ca and Na has also been reported by Venter et al. (2017) in trace metal concentrations at Welgegund in the North-West Province. The abundance of S, as well as the strong correlations seen

with the secondary species, SO_4^{2-} and NH_4^+ , indicates that coal combustion is an important source at these sites. Coal is used as a residential fuel in Embalenhle and a petrochemical plant is located to the immediate right of Embalenhle. Several coal-fired power stations are also present in the region, with Kriel Power Station and Matla Power Station the closest to Kinross. Back trajectory analysis shows that the sites are impacted by air masses from the Highveld region as well as from air pollution sources in the western Bushveld Igneous Complex during winter (Figure 3).

In $PM_{2.5-10}$, Si, Na and Al were the most abundant elements at Embalenhle and Kinross during winter. Contributions from Si and Al remained high during summer with S and Ca also abundant at Embalenhle and Kinross, respectively. Strong correlations between Al, Si, Ca, Fe and Mg indicate a dominant crustal origin of these species.

Heavy metals such as V, Ni, Cr and Zn are also present in low abundances in the datasets. V and Ni can be associated with the petrochemical industry (Bosco et al., 2005; de la Campa et al., 2011) and oil combustion sources (Moreno et al., 2010; Viana et al., 2008). Zn and Cr can also originate from waste burning and motor vehicles (de Bruin et al., 2006; Pant and Harrison, 2012).

Ionic

SO_4^{2-} and NH_4^+ were the most abundant species in $PM_{2.5}$. This has also been seen in a number of studies undertaken at various sites in South Africa (Tiitta et al., 2014; Conradie et al., 2016; Venter et al., 2018; Muyemeki et al., 2021). These species are strongly correlated at both sites, again indicating a common secondary origin, such as coal combustion, of these species.

The species K^+ , an excellent tracer for wood and biomass burning (Zhang et al., 2013; Yu et al., 2016), is present in higher abundances during winter, as also observed by Venter et al. (2018) at Welgegund. This is consistent with the fire burning season, which reaches maximum intensity in South Africa in

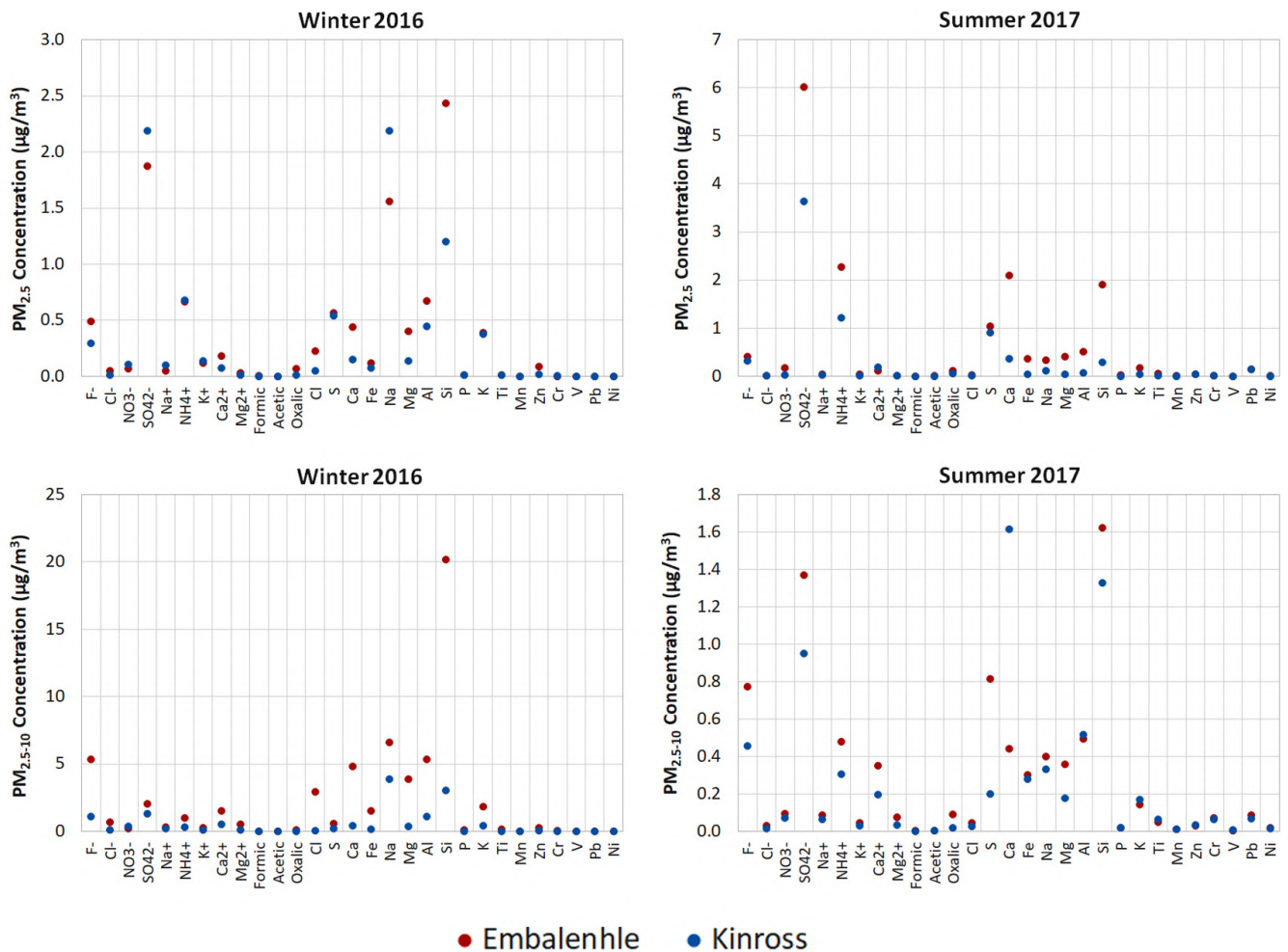


Figure 5: Mean elemental and ionic compositions of $PM_{2.5}$ (top) and $PM_{2.5-10}$ (bottom) at Embalenhle and Kinross during the sampling campaigns

September (Scholes et al., 1996a and b). Aurela et al. (2016) found that the chemical composition of fresh biomass burning plumes was comprised mainly of SO_4^{2-} , OC, EC, NH_4^+ and K^+ in the fine fraction, while Na^+ , Cl^- , NO_3^- and oxalate were divided between the fine and coarse fractions. K^+ was in the form of potassium chloride (KCl) in fresh smoke. K^+ is correlated with Na^+ in both size fractions at Embalenhle and Kinross, as well as Cl^- , NO_3^- , SO_4^{2-} and NH_4^+ in some of the datasets, supporting a contribution from wood and biomass burning.

The presence of Cl^- in the datasets can also originate from coal combustion, waste burning (Liu et al., 2017; Yang et al., 2018) and sea salt (Vasconcellos et al., 2007). While air masses originating from the Indian and Atlantic Oceans are evident in the back trajectory analysis (Figure 3), Cl^- and Na^+ are poorly correlated in most of the datasets which does not support a significant marine source of these species.

In $PM_{2.5-10}$, SO_4^{2-} and F^- are the most abundant species at both sites during both seasons. High abundances of Ca^{2+} and NH_4^+ are also seen during winter and summer, respectively. During both seasons, SO_4^{2-} is strongly correlated with NH_4^+ and moderately correlated with NO_3^- at Embalenhle and Kinross. F^- shows good correlations with various species such as SO_4^{2-} , K^+ , Mg , Al and

Si which can be associated with coal combustion, wood and biomass burning, soil dust and industry.

Ratios of NO_3^-/SO_4^{2-} can be used to evaluate the sources of these two ionic species (Gao et al., 1996). NO_3^-/SO_4^{2-} ratios > 1 indicate a greater contribution of NO_3^- from vehicle emissions while NO_3^-/SO_4^{2-} ratios < 1 indicate a significant contribution of SO_4^{2-} from industrial activities. Mean NO_3^-/SO_4^{2-} ratios for both sites indicate that industrial activities are the dominant source of these species during both seasons (Table 1).

Table 1: Mean NO_3^-/SO_4^{2-} ratios of $PM_{2.5}$ and $PM_{2.5-10}$ at Embalenhle and Kinross during the sampling campaigns

Sampling Site	Winter 2016		Summer 2017	
	$PM_{2.5}$	$PM_{2.5-10}$	$PM_{2.5}$	$PM_{2.5-10}$
Embalenhle	0.06 ± 0.04	0.14 ± 0.20	0.03 ± 0.04	0.14 ± 0.17
Kinross	0.06 ± 0.04	0.44 ± 0.31	0.01 ± 0.01	0.10 ± 0.09

SO_4^{2-} and NH_4^+ abundances are significantly higher during summer compared to winter. During summer, the increase in relative humidity favours SO_4^{2-} production (Aurela et al., 2016; Shikwambana and Sivakumar, 2019). The prevailing wind field, combined with the high contribution of these species in the

Table 2: Mean elemental and ionic concentrations ($\mu\text{g}/\text{m}^3$) at Embalenhle and Zamdela during winter and summer. Concentrations below the detection limit are shown as BDL.

Species	Embalenhle				Zamdela			
	Winter 2016		Summer 2017		Winter 2018		Summer 2018	
	PM _{2.5}	PM _{2.5-10}	PM _{2.5}	PM _{2.5-10}	PM _{2.5}	PM _{2.5-10}	PM _{2.5}	PM _{2.5-10}
Cl	0.22	2.92	0.03	0.05	0.14	2.37	0.08	0.10
S	0.57	0.58	1.05	0.81	1.90	2.30	1.74	1.90
Ca	0.44	4.81	2.10	0.44	0.15	2.80	0.21	1.13
Fe	0.12	1.52	0.37	0.30	17.60	2.21	0.36	1.14
Na	1.56	6.63	0.34	0.40	0.44	4.00	0.05	1.88
Mg	0.40	3.87	0.42	0.36	0.00	2.29	0.01	0.78
Al	0.67	5.34	0.51	0.50	0.05	4.51	0.71	1.54
Si	2.43	20.19	1.90	1.62	0.27	13.59	1.33	4.87
P	0.01	0.08	0.03	0.02	0.02	0.11	0.01	0.04
K	0.39	1.81	0.18	0.14	0.99	1.84	0.17	0.41
Ti	0.02	0.14	0.06	0.05	0.02	0.21	0.05	0.07
Mn	0.00	0.01	0.01	0.01	0.06	0.07	0.03	0.03
Zn	0.09	0.28	0.04	0.03	0.18	0.21	0.28	0.10
Cr	0.00	0.06	0.02	BDL	6.32	0.59	0.08	0.17
V	0.00	0.00	0.00	0.00	0.05	0.01	0.04	0.00
Pb	0.00	0.00	BDL	0.09	0.08	0.02	0.07	0.02
Ni	0.00	0.02	0.01	0.02	1.90	0.18	0.02	0.05
F ⁻	0.49	5.32	0.42	0.77	0.51	2.73	2.25	1.41
Cl ⁻	0.05	0.66	0.02	0.03	0.03	0.30	0.03	0.01
NO ₃ ⁻	0.07	0.22	0.18	0.09	0.36	2.92	1.31	0.68
SO ₄ ²⁻	1.87	2.03	6.02	1.37	4.21	6.59	6.75	4.16
Na ⁺	0.05	0.29	0.05	0.09	0.05	0.38	0.38	0.13
NH ₄ ⁺	0.66	0.97	2.27	0.48	1.91	2.79	2.50	1.35
K ⁺	0.12	0.26	0.04	0.04	0.34	0.67	0.08	0.07
Ca ₂ ⁺	0.18	1.54	0.11	0.35	0.04	1.04	0.28	0.46
Mg ₂ ⁺	0.03	0.54	0.02	0.08	0.00	0.16	0.06	0.06
Formic	0.01	0.02	0.00	0.00	0.01	0.01	0.01	0.00
Acetic	0.00	0.01	0.01	0.00	0.00	0.00	0.01	0.01
Oxalic	0.07	0.13	0.12	0.09	0.13	0.22	0.15	0.11

coarse fraction, suggests emissions from neighbouring coal-fired power stations are impacting Embalenhle and Kinross.

Comparison of measured concentrations

For contextualisation of the elemental and ionic concentrations measured at Embalenhle (and Kinross), the findings have been compared to concentrations measured in Zamdela in the Free State Province (Muyemeki et al., 2020) (Table 2). Similar to Embalenhle, Zamdela is a low-income township and is also located within a Priority Area, the Vaal Triangle Airshed Priority

Area. The township is also located close to petrochemical activities.

In PM_{2.5}, Fe was the most abundant species at Zamdela during the winter which is likely due to ferromanganese (FeMn) activities in the area (Venter et al., 2017). This is also seen in the higher Mn recorded at Zamdela compared to the minor contribution at the two Highveld sites. Si, which was the most abundant PM_{2.5} species at Embalenhle during the same season, was significantly lower at Zamdela. Na concentrations at the two

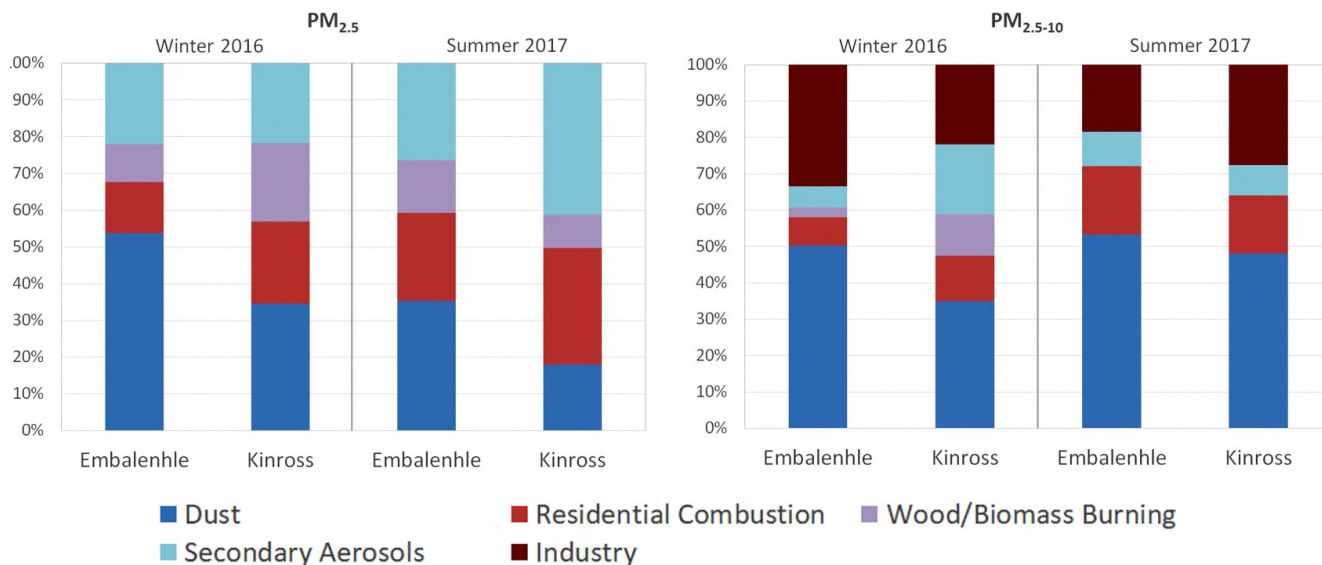


Figure 6: Source apportionment of $PM_{2.5}$ (left) and $PM_{2.5-10}$ (right) at Embalenhle and Kinross during the winter 2016 and summer 2017 sampling campaigns

Highveld sites were substantially higher than those recorded at Zamdela during winter. Na concentrations remained high at Embalenhle during summer while Kinross and Zamdela showed similar concentrations.

K^+ concentrations were higher at Zamdela compared to the two Highveld sites suggesting a stronger influence of wood and biomass burning at this site.

The heavy metals, Cr and Ni, were considerably higher at Zamdela with Zn and V also higher than Embalenhle and Kinross. This is likely due to the stronger industrial influence in the Vaal Triangle.

The secondary species, SO_4^{2-} and NH_4^+ , were seen to be abundant at all three sites during both seasons. However, SO_4^{2-} and NH_4^+ concentrations were over double those at Embalenhle and Kinross during winter, although summer concentrations were similar at Embalenhle. S concentrations were also higher than those reported for the Highveld sites during both seasons.

In $PM_{2.5-10}$, Si was the most abundant species at both the Highveld and Zamdela sites (with the exception of Na and Ca at Kinross in winter and summer, respectively). Al concentrations were similar at Embalenhle and Zamdela during winter but lower at Embalenhle during summer.

SO_4^{2-} and NH_4^+ concentrations were higher at Zamdela compared to the Highveld sites, during both seasons. However, F concentrations at Zamdela were lower during both seasons. F concentrations at Embalenhle were almost double those measured at Zamdela during winter.

Source apportionment

The PMF model identified dust, residential combustion, secondary aerosols, wood and biomass burning, and industry as the main sources contributing to the aerosol loading in Embalenhle and Kinross. The source apportionment results are

shown in Figure 6. Four and five factor profiles were identified to be the most representative in the fine and coarse fraction, respectively.

Fine fraction

A dust profile was identified by the presence of traditional soil elements such as Ca, Mg, Si, Al, Fe, Ti and Mn. This profile accounted for 55% and 42% of the reconstructed $PM_{2.5}$ mass concentrations at Embalenhle and Kinross, respectively, during winter and 35% and 18% during summer. The dust contribution at Embalenhle during winter is overestimated and likely represents a mixed dust and coal combustion profile, given the similarity of the species in each source. This has been confirmed through CMB modelling for the site (not given in this paper), which shows both a dust (~22%) and coal combustion (~38%) contribution.

Residential combustion is distinguished by the presence of Cl, Zn, Na^+ , NH_4^+ and K^+ in the profiles. Zn can be emitted from the burning of waste while the presence of K^+ can be attributed to the burning of unsorted domestic and garden waste (Muyemeki et al., 2020). Residential combustion accounts for 12 to 13% (winter) and 9 to 24% (summer) of the reconstructed $PM_{2.5}$ mass concentrations at both sites.

A secondary inorganic aerosol profile is detected by the presence of secondary species such as SO_4^{2-} , NH_4^+ , and NO_3^- (Crilley et al., 2017). Secondary aerosols are found predominantly in the fine fraction and contribute to 22% and 25% of the reconstructed $PM_{2.5}$ mass concentrations at Embalenhle and Kinross, respectively, during winter. During summer, the contribution increases to 26% and 41%, respectively, due to the increase in relative humidity during this season. Coal combustion is the dominant source of secondary aerosols as the burning of coal releases significant quantities of the precursor gases, SO_2 , NO_x and NH_3 , which then oxidise into secondary particles (Watson et al., 1994).

A wood and biomass burning profile, identified by the abundance of K^+ as well as Cl^- , SO_4^{2-} , NO_3^- , NH_4^+ , Na^+ and organic acids. Wood is also used as a residential fuel in Embalenhle although to a much lesser degree than coal. This source accounted for 11% and 20% of the reconstructed $PM_{2.5}$ mass concentrations at Embalenhle and Kinross, respectively, during winter. During summer, this source contribution is 14% and 32%.

Coarse fraction

In the coarse fraction, dust accounted for 35 to 51% of the reconstructed $PM_{2.5-10}$ mass concentrations during winter and 46 to 50% during summer, at both sites. This source represents both a natural (wind-blown) and anthropogenic component of dust. Elements such as Zn, V, Pb, Ni and Cr are also seen in the profiles which can be associated with resuspended dust from paved and unpaved roads. Zn, Pb, Ni and Cr are associated with dust from tyre wear and braking (Adachia and Tainoshob, 2004; Hjortenkrans et al., 2007) while V could also originate from neighbouring industrial activities.

Residential combustion contributes to 9 to 13% (winter) and 14 to 23% (summer) of the reconstructed $PM_{2.5-10}$ mass concentrations at both sites.

Secondary aerosols account for 4% and 19% at Embalenhle and Kinross, respectively, during winter and 10% and 9%, respectively, during summer.

A wood and biomass burning profile is only seen at Embalenhle (2%) and Kinross (11%) during winter.

Industrial activities are also identified in the coarse fraction with the presence of SO_4^{2-} , NH_4^+ , S, Al, Si and the heavy metals, Cr, V, Ni and Pb. Industry contributes to 33% and 22% of the reconstructed $PM_{2.5-10}$ mass concentrations at Embalenhle and Kinross, respectively, during winter and 18 to 32% during summer. The presence of V and Ni indicates a contribution from the neighbouring petrochemical plant with coal-fired power stations also contributing, especially during summer.

Conclusions

Aerosol sampling undertaken at Embalenhle and Kinross during winter and summer shows Si, Al, S, SO_4^{2-} and NH_4^+ to be the most abundant $PM_{2.5}$ species during both seasons. The elements Na and Ca were also abundant at both sites during winter and summer, respectively. In $PM_{2.5-10}$, Si, Al, SO_4^{2-} and F were the most abundant species during both seasons. The element Na was also abundant at both sites during winter with S and Ca also having high abundances at Embalenhle and Kinross, respectively, during summer.

PMF modelling provided an assessment of dust (natural and anthropogenic), residential combustion, secondary aerosols, wood and biomass burning, and industry as the primary sources contributing to the aerosol loading in Embalenhle and Kinross. Of these, dust and secondary aerosols are the most significant

contributors. Emissions from neighbouring industrial activities are also seen to influence the air quality in these two areas. Emission reduction measures to be implemented in the region should consider these sources.

It is noted that the findings of this study are based on two short-term sampling campaigns. As such, the findings of this paper are not necessarily representative of the entire winter and summer seasons. Aerosol concentrations are influenced by various factors such as meteorological conditions and emission source strengths which show considerable spatial and temporal variability.

Acknowledgements

The authors wish to acknowledge the South African Weather Services for providing the meteorological data. NOAA ARL is acknowledged for the provision of the HYSPLIT transport and dispersion model and/or READY website.

Collection of aerosol data from the sampling sites was made possible through the loan of the 'Gent' SFU samplers from Professor Willy Maenhaut at Ghent University.

Jan-Stefan Swartz and Belinda Venter are thanked for performing the IC and XRF analyses, respectively, at the North-West University. Andrew Venter is acknowledged for his assistance in the chemical preparation procedures.

Author contributions

Nicola Walton analysed the data for Embalenhle and Kinross, applied the PMF model to the datasets and drafted this paper. This manuscript was reviewed by each of the authors listed above.

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Eskom drives just energy transition for a greener future in energy

Eskom has embarked on a Just Energy Transition drive to reduce its carbon footprint, while at the same time promoting local industrialisation and ensuring that no jobs are lost in the process.

Given that some of its coal power stations are approaching the end of their life, this is the ideal opportunity to increase capacity with clean technologies by repowering and repurposing our stations. This in turn will contribute to continued economic stimulus, particularly in the communities close to the stations.

Eskom will be seeking to secure significant financing to support its accelerated transition away from coal, increase clean capacity and to protect workers, communities and businesses whose livelihoods are threatened by the shutdown programme.

André de Ruyter, Eskom Group Chief Executive, has indicated that several “repowering and repurposing” opportunities have already been identified.

Pilot project

The Camden, Komati, Grootvlei and Hendrina power stations are scheduled for shutdown in the near-term. Komati has been generating electricity since 1961, and its last operational unit is scheduled to be shut down in 2022, signalling the start of a coal shutdown programme.

Among other opportunities, Eskom is planning to convert the workshops at the Komati power station in Mpumalanga to a factory that will manufacture and assemble a containerised microgrid solution. Eskom has already piloted a microgrid system near Ficksburg in the Free State. This consists of solar photovoltaic (PV) generation, battery storage and intelligent energy management. These technologies have been integrated

into a standard, low-voltage reticulation network, where electricity is delivered to consumers through conductor wires in a local distribution network.

The containerised microgrid solutions could be deployed in far-flung regions nationally where grid-tied electricity is proving to be hugely expensive to supply. There is also great potential to market it to the rest of Africa where, in many cases, the percentage of electrified homes is much lower than in South Africa.

Besides the microgrid manufacturing, Eskom is also working on repowering options through the deployment of solar PV and battery storage.

A major concern related to the shutdown of coal stations is job losses at the stations and linked mines as well as communities near the power stations. Eskom has conducted extensive socio-economic impact studies at Komati, Grootvlei and Hendrina to understand these impacts, and ensure that a robust plan is developed in collaboration with social partners, to manage these negative impacts. We will be conducting these studies for each of our coal stations as they approach shutdown.

As Eskom transitions away from coal, it estimates that nearly 300 000 net jobs could be created in the construction, operation and maintenance of new Eskom and non-Eskom wind and solar PV plants. This figure is based on the assumption that South Africa will grow the local manufacturing ability and attract the investment needed to produce wind and solar components.

Eskom has a comprehensive JET Strategy that details additional opportunities such as battery storage.



Eskom implemented this microgrid in the Free State as a pilot project



Komati Power Station in Mpumalanga

Just: Doing better for people and the planet, and growing localisation

Energy: Cleaner, sustainable electricity provision

Transition: Transformational change of business models, attracting green financing

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