Clean Air Journal

ISSN 1017 - 1703
Vol 29 No 2
November / December 2019

Official publication of the
National Association for Clean Air

CLEAN AIR JOURNAL

ISSN 1017-1703
November / December 2019
Volume 29, No. 2

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

Website and archive copies
www.cleanairjournal.org.za

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Editorial
The use of long-term datasets for informing and evaluating air quality policy initiatives

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https://doi.org/10.17159/caj/2019/29/2.7723

Political institutions generally desire to use the best available scientific evidence when formulating policy. In South Africa, the declaration of the Priority Areas where 'ambient air quality standards are being, or may be, exceeded' (according to the National Environmental Management: Air Quality Act, 2004) is an example of a policy initiative that was prompted by quantitative evidence – measurements of ambient air pollution levels. National Priority Areas have been declared in the Vaal Triangle in 2006, in the Highveld in 2007 and in the Waterberg-Bojanala region in 2012.

The Department of Environment, Forestry and Fisheries (DEFF, formerly the Department of Environmental Affairs) established a network of 6 ambient air quality monitoring stations in the Vaal Triangle and 5 monitoring stations in the Highveld Priority Area to inform the emission reduction strategies and track progress in reducing ambient pollution levels. The summarised data from these monitoring stations is presented annually by the National Air Quality Officer in the State of the Air Report. In this issue of the Clean Air Journal, there are two papers (Feig et al., 2019 and Govender and Sivakumar, 2019) that have subjected the 10+ years of data from these monitoring stations to more rigorous trend analysis, which allows reflection on progress made to date and future regulatory priorities.

Govender and Sivakumar (2019) analyse long-term changes in $PM_{2.5}$ and O_3 concentrations in the Vaal Triangle Airshed Priority Area, while Feig et al. (2019) examine trends in PM_{10}, $PM_{2.5}$ and SO_2 in the Vaal Triangle Airshed and Highveld Priority Areas. For both studies, the ambient air quality data was extracted from the South Africa Air Quality Information System (SAAQIS) database and Theil-Sen trend analysis was performed using the Open Air Package in R (in addition to other analyses).

The results of these studies show that there is still widespread non-compliance with ambient particulate matter levels in the Vaal Triangle and Highveld Priority Areas (there is non-compliance with the annual ambient PM_{10} standard at seven of the eleven monitoring stations and non-compliance with the annual average $PM_{2.5}$ standard at eight of the eleven monitoring stations (Feig et al., 2019)). There are significant downward trends at most of the monitoring stations, however (PM_{10} and $PM_{2.5}$ concentrations are declining at eight and nine of the eleven sites, respectively (Feig et al., 2019)). The annual rate of decline is generally small, ranging between 1 and 5 µg/m^3/year.

SO_2 is not a variable of concern in the Priority Areas (except for at eMalahleni) (Feig et al., 2019), but O_3 is of concern in the Vaal (Govender and Sivakumar, 2019). Perhaps surprisingly, Feig et al. (2019) demonstrate that ambient SO_2 levels in the Priority Areas are lower than SO_2 levels in the large South African metropolitan centres in the 1960s.There is a statistically significant increasing trend in O_3 concentrations at two of the six monitoring sites, and no significant trends in monthly O_3 concentrations at the other four sites in the Vaal (Govender and Sivakumar, 2019). Since O_3 is a secondary pollutant, there is a need to understand the emission and transport of precursors in order to design effective initiatives to reduce ambient O_3 levels.

These studies demonstrate the value of long-term datasets, noting that there needs to be high data availability (ensured through regular calibration and maintenance), quality control of the data (in this case by both the network operators and the authors of the papers) and data archiving in a way that is easily accessible to all who have an interest in the data.

References

Feig, G., Garland, R.M., Naidoo, S., Maluleke, A. and van der Merwe, M. (2019) 'Assessment of changes in concentrations of selected criteria pollutants in the Vaal and Highveld priority areas', Clean Air Journal, 29(2). https://doi.org/10.17159/caj/2019/29/2.7464.

Govender, K. and Sivakumar, V. (2019) 'A decadal analysis of particulate matter ($PM_{2.5}$) and surface ozone (O_3) over Vaal Priority Area, South Africa', Clean Air Journal, 29(2). https://doi.org/10.17159/caj/2019/29/2.7578.

Commentary
IPCC Land report

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https://doi.org/10.17159/caj/2019/29/2.7701

The IPCC produced its seminal report "People, land and climate in a warming world" earlier this year. It is the first such report developed by all three of the IPCC working groups working together (WG1 on climate change science, WG2 on impacts and adaptation, and WG3 on mitigation), and included direct collaboration with the task force on National Greenhouse Gas Inventories (TFI). It was also the first IPCC report to have a majority of developing country scientists in its authorship list. Clearly then, this report marks an effort by the IPCC to represent a broad range of scientific and applied perspectives to the issue of how land, and its management, could play a role in the climate system, in reducing vulnerabilities of human society to climate change, and in helping to mitigate climate change through understanding and managing emissions from land use and land management.

The seven chapters of the report comprise, after a chapter on framing and context, Land-climate interactions, Desertification, Land degradation, Food security as four basic building blocks, and then two chapters looking at interlinkages and between these building blocks, and finally risk management and decision making. The Summary for Policy Makers (SPM) cuts across these topics cleverly, covering four main areas in developing overall take-home messages from what is an extremely comprehensive report. These four areas are People, land and climate in a warming world, Adaptation and mitigation response options, Enabling response options, and Action in the near-term.

The SPM presents the case that human society already consumes somewhere between a quarter to a third of the potential net primary production from the land surface for food, fibre, timber and energy. This estimate has been in the literature for some time now, but the report constrains the estimate well. Furthermore, the report presents how rapidly human demand for resources from the land surface are ramping up, both due to population growth and increasing wealth and buying power, while almost an eight of the world's population remains undernourished. The result has been degradation of about a quarter of the earth's land surface, with soil erosion rate exceeding soil formation rate by 20 to 100 times. Climate change is exacerbating this trend, and people living in degraded and desertified areas are particularly badly affected. While the land surface has helped to sequester around 6 GtCO2 per annum of anthropogenic emissions, projections suggest that climate change could weaken this sink. The report highlights a number of implications for food supply and food security, with risks escalating unequally in different parts of the world.

There is good news, as well, and this relates especially to how efforts to combat desertification, degradation and the decline of food security can also serve to increase our ability to adapt to and mitigate climate change. Several response options are considered in the report, and include sustainable food production, improved and sustainable forest management, soil organic carbon management, ecosystem conservation and land restoration, reduced deforestation and forest degradation, and reduced food loss and waste. However, these responses are effective over very different timelines, leading to a need to prioritise in their implementation. Conservation of high carbon ecosystems would be immediately effective, but less immediate responses include reforestation and afforestation, and ecosystem restoration, including the reclamation of systems with degraded soils.

While these responses all play a role in helping society to adapt to and mitigate climate change, their effectiveness has a definite ceiling, as they sinks for carbon fill up in most ecosystems, save a few such as peatlands that may sequester carbon for centuries. Afforestation of so-called "degraded" ecosystems is a bone of contention in southern Africa, and indeed in many subtropical regions, where species rich grasslands can be characterised as anthropogenic, and thus degraded. Policy makers in the global south need to be aware of this issue before signing onto afforestation schemes that could be ruinous for local people and their livelihoods. Another issue is the species chosen for afforestation programs, with a long history of inadequately assessed introductions of invasive species a clear warning to those who would restore forests with exotic tree species. The report finds that if deployed at appreciable scales for carbon sequestration, afforestation, biochar production and biomass based fuel production, these efforts could greatly increase demand for new land conversion, and should thus usefully be limited in scale.

Some apparently low hanging fruit for carbon management are to be found in improved management options, which require neither any further land use change nor more demand for land conversion and land cover change. However, an increasing awareness of dietary choices which are prompted by non-sustainable meat production may affect markets for meat even if it is produced sustainably, a potentially adverse outcome that policy makers in the global south should be vigilant about.

As we currently understand it, options that limit global warming to 1.5C include more land-based mitigation than do higher warming pathways. In other words, land is crucial for achieving this low warming outcome, and the results projected include that direct climate change impacts on land systems would be less severe. The report finds that delaying action on climate change mitigation and adaptation responses across sectors would cause increasingly adverse impacts on terrestrial ecosystems and food production. Ironically, delayed action also increases the demand for widespread land-based mitigation responses, which could limit their future effectiveness. This is the policy equivalent of human society painting itself into a corner. Indeed, the outcomes of this would include irreversible impacts on some ecosystems, and even the acceleration of emissions from some high-carbon ecosystems such as high latitude peatlands, which would in turn exacerbate anthropogenic warming. Such a fate is clearly something we would want to avoid.

News
A Global Statement for Air Pollution and Health

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https://doi.org/10.17159/caj/2019/29/2.7636

Statement
The Statement is available at:
https://air-pollution.health

Keywords
Air pollution, human health, wellbeing, climate change, environmental health

Air pollution poses a major threat to human health. It is estimated that 91% of the world's people live in places that did not meet the air quality guidelines of the World Health Organization (WHO, 2019a). Exposure to air pollution is well-established to be associated with adverse health effects, including respiratory conditions, either exacerbating existing diseases or increasing susceptibility to respiratory and other infections (WHO, 2019b). Currently many areas in South Africa exceed National Ambient Air Quality Standards. Priority Areas of air pollution hotspots are being managed to try to improve air quality (SA DEA, 2019).

In July 2019, at the United Nations headquarters in New York, the Academy of Sciences of South Africa (ASSAf) joined the science academies of Germany, Brazil, and the United States, as well as the US National Academy of Medicine, in issuing an urgent call to citizens, governments, and businesses to reduce global air pollution. The delegation presented a science-policy statement to senior United Nations representatives and high-level diplomats. ASSAf was represented at the event by Executive Officer, Professor Himla Soodyall, and was joined by Senior Specialist Scientist in the Environment and Health Research Unit of the South African Medical Research Council (SAMRC) Dr Caradee Wright. Dr Jacqueline McGlade, former Chief Scientist to the United Nations, led the scientific delegation (Figure 1).

National Academies are a forum in which scientists from all disciplines come together. This creates a unique position to address pressing issues; in this case, air pollution and health, to lobby that air pollution is moved up the policy agenda. The statement appeals for emissions controls in all countries as well as proper monitoring of key pollutants, especially $PM_{2.5}$, and stresses that funding is needed to invest in air pollution reduction measures to mirror the scale of the problem. Decisive action by stakeholders in all sectors would culminate in cost-effective management of air pollution.

The success and urgency with which this initiative will be met relies on an increase in international stakeholders therefore, including the public, policy makers, and researchers. September 2019 saw national action being taken. Approximately 200 delegates from the Environmental Health fraternity attended a two-day conference celebrating World Environmental Health Day 2019. A WHO-facilitated dialogue on air pollution and health was held and several actions were proposed which the National Department of Health committed to pursue in the coming months.

Prof Soodyall, Dr McGlade and Dr Wright in NYC, June 2019.

References
WHO, 2019a. Ambient air pollution [Internet]. World Health Organization. 2019 [cited 31 July 2019]. Available from: https://www.who.int/airpollution/ambient/en/

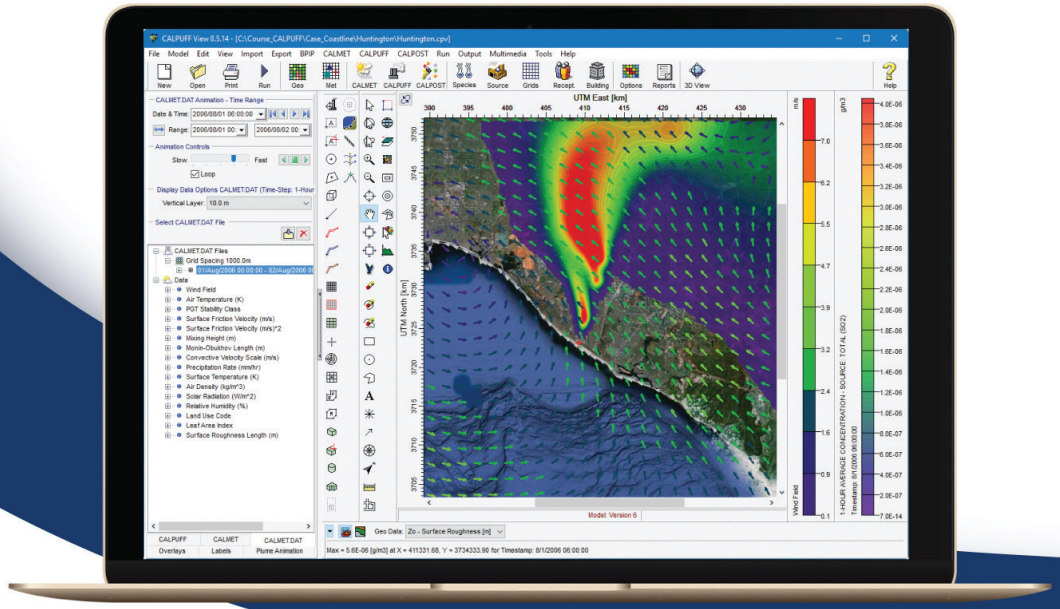
WHO, 2019b. Ambient (outdoor) air quality and health [Internet]. Who.int. 2019 [cited 31 July 2019]. Available from: https://www.who.int/en/news-room/fact-sheets/detail/ambient-(outdoor)-air-quality-and-health

South Africa: Department of Environmental Affairs, 2019. Air quality [Internet]. [cited 31 July 2019]. Available from: https://www.environment.gov.za/sites/default/files/reports/environmentoutlook_chapter10.pdf

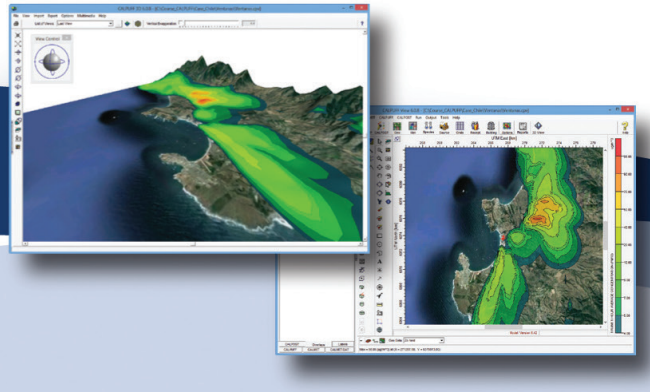
CALPUFF View

Long Range Puff Air Dispersion Model

Lakes Software



- Support for Latest US EPA Models
- CALMET Parallel
- Plume Animation
- Percentile & Rolling Averages
- Auto-Download of Maps
- Auto-Download of Geophysical Data



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News

A new atmospheric pollen monitoring network for South Africa

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https://doi.org/10.17159/caj/2019/29/2.7645

Introduction

It is estimated that 4 out of 5 children in the world are breathing unsafe air (https://www.who.int/airpollution/en/). The majority of focus around air quality examines air pollutants like SO_2, CO, and particulate matter with attendant health problems. However, there are other bioaerosols such as pollen and fungal spores that also affect health. Modelling data from Europe indicate that climate change will dramatically increase pollen levels by as much as 4-5 fold (Katelaris and Beggs 2018). Greater attention to this aspect of air quality is thus urgently needed in South Africa.

Pollen allergy and climate change

Airborne pollen and fungal spores are known as aeroallergens. These aeroallergens may trigger asthma (airway), allergic rhinitis (nose) and conjunctivitis (eye) in an estimated 20-30% of South Africans (Ait-Khaled et al 2007). Pollen, fungal spores and house dust mites and the three commonest aeroallergens that allergy sufferers react too. When pollen is inhaled and comes into contact with the mucous membrane of the respiratory tract, it rapidly extends a pollen tube, often within 90 seconds (Solomon 2005). The tube releases recognition substances in the form of enzymes, which are predominantly proteins and glycoproteins. It is these proteins and glycoproteins that cause an immune response to be mounted in allergic individuals as briefly outlined in Figure 1.

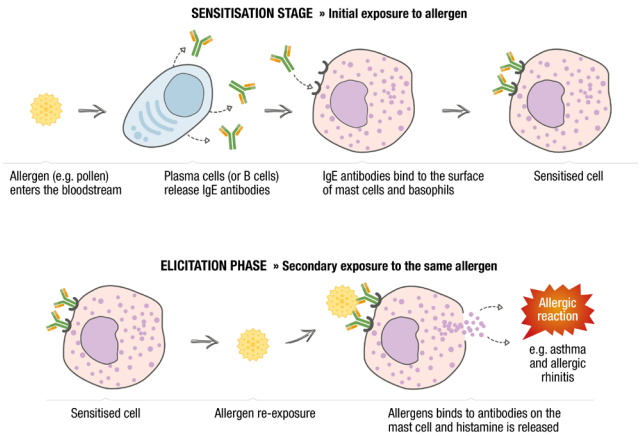

Figure 1: *Schematic representation of the basic immune mechanisms of an immediate type-1 allergic reaction, resulting in the rapid release of histamine and other substances inducing the typical clinical allergic pattern of symptoms (From Jeurink 2005)*

The pollen of plants that have brightly coloured flowers, pollinated by insects, birds or small mammals like mice, rarely trigger allergy. Pollen that is allergenic is usually produced in large quantities, is windborne and it is strongly associated with weather parameters, especially temperature, humidity, rainfall and wind speed and direction. Grass is a good example of an allergenic pollen and is the major pollen allergen in many countries. Climate Change is altering weather patterns and thereby changing the length of the pollen season, as some plants increase their pollen production under conditions of increased CO_2 or higher temperatures (Albertine et al 2014). The impact of these changes is that exposure of the allergic individual to pollen allergens is likely to increase. This is especially relevant to ragweed, an invasive weed that is known to have been introduced to South Africa but has not been detected at pollen monitoring sites so far. Ragweed is the major weed allergen in many Northern Hemisphere countries. The pollen season for grasses and trees is likely to lengthen and become less predictable as a consequence of Global Warming. For these reasons, it is becoming increasingly important for countries to routinely measure pollen in the air, especially focused on the most densely populated cities.

Pollen and pollutants

Air pollution is a growing global concern; and climate change looks set to make pollen an increasing problem. What is worse, is that there appears to be a synergist effect of air pollutants on allergic responses to pollen proteins. Epidemiological data indicates that allergic subjects living close to major roads, with exposure to diesel exhaust particles, have more severe allergic disease (Motta et al 2006). One of the drivers of this surge in symptoms appears to be the interaction between pollen and diesel exhaust particles. The mechanism that brings about this effect is damage to the exine, or outer cell wall of the pollen grain, rendering the pollen more fragile, so that it readily fractures and releases allergenic cytoplasmic particles (Motta et al 2006). Aggravation of asthma and allergic rhinoconjunctivitis has also been linked to exposure to particulate matter (PM); and in several SA provinces and cities, due to veld fires and burning biomass, PM levels exceed WHO recommendations (Baldacci et al 2015). The size of the particles (in μm) aerosolised is important in the consequent airway/allergic effects, as the size of the particles impact their ability to penetrate into the respiratory tract. PM_{10}, and $PM_{2.5}$ only penetrate the upper respiratory tract, while ultrafine particles $PM_{0.1}$ are able to penetrate the alveoli.

These smallest particle, $PM_{0.1}$ can lead to molecular changes and thought to be key to changing the way the immune system interacts with allergen at the airway interface (Baldacci et al 2015).

Pollen monitoring in seven South African cities

Pollen monitoring has always been sporadic in South Africa. Several areas have never been monitored e.g. Eastern Cape or Mpumalanga; while others Gauteng were last consistently monitored in the 1990s (Potter and Cadman). This limited local data and increasing realisation of the changing environment and likely worsening impact on health has led us to launch a national campaign to monitor pollen across SA. The project was initiated and is being led by the Allergy and Immunology Unit at the University of Cape Town Lung Institute; and includes clinicians, aerobiologist, palynologists and students at six universities. Funding has been raised through corporate sponsorship. In August 2019 the National Pollen Campaign established pollen monitoring sites where pollen spore traps were set up in: Johannesburg, Pretoria, Durban, Bloemfontein, Kimberley and Port Elizabeth and together with the existing Cape Town site a pollen monitoring network has been created. Teams of clinicians and scientists in each city are using 7 day recording volumetric spore traps, mounted on rooftops to trap the pollen and fungal spores contained in a measured air volume onto sticky cellulose strips. The harvested strips are examined using light microscopes to identify and count the air spora and to produce the daily pollen count, which is the number of pollen grains contained in one cubic meter of air per day. The weekly pollen counts, together with information on how and why pollen monitoring is being done, key facts for allergy sufferers and about allergenic plants are available through our website – www.pollencount.co.za. This makes the information readily available to individual patients; clinicians who treat patients with allergic disease and scientists. This is already leading to increased diagnosis and triaging of patients to correct treatment.

Vegetation biomes and SA cities

South Africa has a high range of biodiversity relative to its size and the seven cities chosen as the initial pollen sampling sites are both large urban provincial capital centres, but also reflect many of the SA different biomes. Fynbos is a feature of the Western Cape, Durban is in a subtropical climate zone, Pretoria and Johannesburg are in Grassland/ Savanna regions. Kimberley and Bloemfontein are close to maize crops and Port Elizabeth is a melting pot of vegetation biomes. Tree pollen from the sites is predominantly from Northern Hemisphere aliens that have been introduced, but white stinkwood: Celtis africana and kareeboom or Rhus, now named Searsia, have been identified from pollen monitoring in South Africa.

Where to from here? Pollen Samplers for Mpumalanga, Limpopo and North West Province

A major strategic aim of the project is to have consistent pollen monitoring in at least one site for each of SA nine provinces. Thus, we are looking to expand to the additional three provinces that are not currently part of the National Pollen Campaign. Further funding and logistical planning is being undertaken, and we would welcome contributes from interested scientists. A further aim is to continue the pollen monitoring programme for five years. Undoubtedly the longer the project continues and the wider its reach, the more impact it is likely to have. We believe that initiatives like this, striving to achieve long-term national pollen data will be of major benefit to individual patients, climate change scientists and healthcare and environmental policy makers assessing the impact of climate change on allergenic pollen in SA and consequently its potential future impact on human health.

References

Katelaris CH, Beggs PJ. Climate change: allergens and allergic diseases. *Intern Med J*. 2018;48(2):129-34.

Ait-Khaled N, Odhiambo J, Pearce N, Adjoh KS, Maesano IA, Benhabyles B, et al. Prevalence of symptoms of asthma, rhinitis and eczema in 13- to 14-year-old children in Africa: the International Study of Asthma and Allergies in Childhood Phase III. *Allergy*. 2007;62(3):247-58.

Solomon WR. Airborne pollen: a brief life. J Allergy Clin Immunol. 2002;109(6):895-900.

Albertine JM, Manning WJ, DaCosta M, Stinson KA, Muilenberg ML, Rogers CA. Projected carbon dioxide to increase grass pollen and allergen exposure despite higher ozone levels. *PLoS One*. 2014;9(11):e111712.

Motta AC, Marliere M, Peltre G, Sterenberg PA, Lacroix G. Traffic-related air pollutants induce the release of allergen-containing cytoplasmic granules from grass pollen. I*nt Arch Allergy Immunol*. 2006;139(4):294-8.

Baldacci S, Maio S, Cerrai S, Sarno G, Baiz N, Simoni M, et al. Allergy and asthma: Effects of the exposure to particulate matter and biological allergens. *Respir Med*. 2015;109(9):1089-104.

Potter PC, Cadman A. Pollen allergy in South Africa. *Clin Exp Allergy*. 1996;26(12):1347-54.

Jeurink, Prescilla V., and Huub FJ Savelkoul. “Induction and regulation of allergen-specific IgE.” *Allergy Matters: New approaches to allergy prevention and management* 10 (2005) p18.

News
School on Climate and Environmental Modelling in the West African Region

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https://doi.org/10.17159/caj/2019/29/2.7594

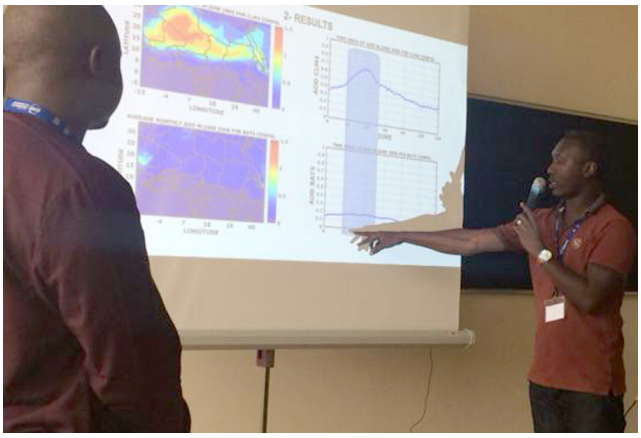
The West African climate is characterized by complex atmospheric dynamics and growing anthropogenic pressures which when its combined with climate change and variability, lead to many environmental impacts. Regional climate models allowing coupling with oceans, surface hydrology, atmospheric chemistry and biogeochemical cycles are appropriate for such complex processes investigation. Therefore, regional climate models offer interesting frameworks for climate impact assessments over West Africa. However, these models complexity require high computing power and appropriate knowledge.

An international workshop on climate and environmental modelling was held from March 11th - 15th at the Pole Scientifique et d'Innovation (PSI) of the University Félix Houphouet-Boigny (Campus of Bingerville, Abidjan, Côte d'Ivoire). This international workshop led by the Numerical Simulation Pole on "Climate and Sustainable Development" was co-organised by the International Centre for Theoretical Physic (ICTP), The Ministry of High Education and Scientific Research of Cote d'Ivoire, in collaboration with Institute of Research and development (IRD), the Centre National de Recherche Scientifique(CNRS) of France, ATOS, WASCAL and the African Centre of Excellence on "Climate Change, Biodiversity and Sustainable Agriculture" (CEA-CBAD). The workshop convened about 50 participants from West Africa, Cameroon and South Africa.

The main objective of the workshop was the reinforcement of the regional climate modelling capacities for a better understanding of local and regional climate process associated with climate change and variability as well as different related impacts.

Specific objectives covered by the workshop were:

- introducing basis of regional climate and earth system modelling;
- Strengthening the expertise of national teams working on regional climate models and their environmental impacts (air quality, agriculture, water resources, health…).
- Reinforcing local and regional interdisciplinary platforms to foster integrated regional climate modelling studies and their applications;



Participants presenting their "small project" results on 15 March 2019

Some participants with their training certificates

- Proposing extensive hands-on regional climate and Earth system modelling sessions using HPC facilities;
- Showing examples of climate and earth system studies over sub-Saharan Africa
- Building a network of African scientists contributing to CORDEX-Africa initiative and fully involved in the future CORDEX Flagship Pilot Studies envisionned for West and Central Africa.

This workshop was divided into theoretical and practical sessions. Models fundamental basics such as atmospheric dynamics and numerical representation, parametrisation, hydrostatic assumption, resolution (temporal, spatial and vertical), aerosol interactions with African climate as well as simulated PM2.5 surface concentrations and their health impacts were discussed. These models fundamental basements were discussed by prominent scientists:

- Dr Hubert Gallée, Université Grenoble Alpes, IGE, Grenoble (France)
- Dr Fabien Solmon, LA, Toulouse (France) and ICTP
- Model developer Mr Graziano Guiliani, ICTP, Trieste (Italy)
- Dr Bamba M. Sylla, WASCAL Competence Centre, Ougadougou (Burkina Faso)
- Dr N'Datchoh E. Toure, UFHB, Abidjan (Cote d'Ivoire)

Then, practical sessions allowed participants to run their own "small projects" using regional modelling tools such as MAR and RegCM4 on the Ivorian supercomputer named WOMBELE. With its peak performance of 322 teraflops and storage capacity of 5PB, it is the second supercomputer in Africa. Côte d'Ivoire is the first country in West Africa to set up a national supercomputing centre with the purpose of sharing computing systems between universities and industrials. Atos' Center for Excellence in Parallel Programming supports the Côte d'Ivoire Ministry in their ambitious project with two concurrent actions:

- By delivering organisational recommendations for setting up an efficient operational structure and a balanced decisional system for sharing computing resources between national research communities.
- Setting up application knowledge transfer to the national research ecosystem,

so as to push national excellence to a level recognized by the HPC community. Many actions are scheduled such as regular trainings, workshops, and creating HPC academic training.

On the Friday 15th March, participants presented their results from numerical experiments for examples reforestation of Sahara into Savana and assessment of the associated impact on West African Monsoon system, high resolution simulation of a convective system over the West African region, simulation of dust outbreaks and climatic impacts on the West African region, model intercomparison between MAR and RegCM4 regional climate models.

The final discussion rose the issues about how to optimally invest in research infrastructure and scientist education in Africa for tackling numerical studies on climate and impacts.

Dedication

This paper is dedicated to the memory of late Prof Abdourahamane Konare who coordinated the project of the Ivorian HPC with the ambition to offer a powerful numerical platform and research environment to all African scientists who need more computational resources to make their research work.

Acknowledgement

Special thanks to ICTP, DGRI, UFHB, IRD, LA, ATOS, IGE, WASCAL, CEA-CCBAD for the financial support which allowed the organisation of the workshop.

Research brief

Reducing exposure to $PM_{2.5}$ in South Africa leads to significant avoided premature mortalities and has large associated economic benefits

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https://doi.org/10.17159/caj/2019/29/2.7698

Exposure to high levels of fine particulate matter ($PM_{2.5}$) is a health and environmental concern in many areas across South Africa. Chronic exposure to $PM_{2.5}$ is associated with increased mortality risks, and there is a link between high concentrations of air pollutants and large health costs. The Global Burden of Disease (GBD) estimates that chronic exposure to $PM_{2.5}$ in South Africa leads to 14,356 total deaths annually. A World Bank study estimated 19,802 deaths annually due to air pollution in South Africa, with a total welfare loss equivalent to 3.12% of South Africa's GDP. However, these international studies rely on a combination of global models, satellite retrievals, and national level statistics to conduct a national-scale analysis of what is a highly geospatially explicit problem.

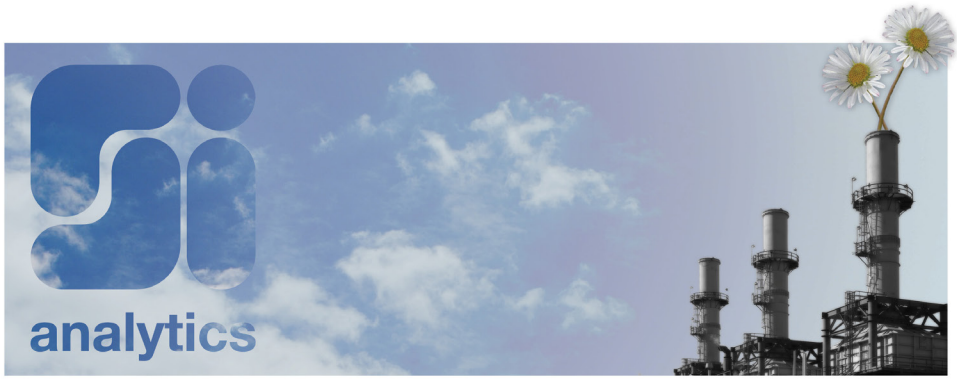
This study addresses the same question as the GBD and World Bank study – how many premature mortalities are due to chronic exposure to $PM_{2.5}$ annually in South Africa, and what is the associated cost to the economy? The difference here is that we apply a spatially explicit approach using a tool developed by the US EPA, the environmental Benefits Mapping and Analysis Program (BenMAP; https://www.epa.gov/benmap). BenMAP was initialized with South Africa-specific shape files, census and mortality data from StatsSA, air pollution data from SAAQIS, and a variety of health impact functions from the literature. Instead of one national-level calculation, a calculation was conducted for each municipality across South Africa, using municipality-specific input data for the year 2012.

We estimate 28,000 premature mortalities associated with chronic exposure to $PM_{2.5}$ across South Africa, with a total welfare loss equivalent to 4.5% of South Africa's GDP. This is significantly higher than the estimates produced by the international community and highlights the importance of accounting for regional heterogeneity within a country.

In a more policy-relevant analysis, we also estimated the total premature mortalities avoided if every monitoring station across South Africa met the annual average NAAQS for $PM_{2.5}$. We find that 14,000 premature mortalities could be avoided if existing standards were met across the country, with an associated economic cost equivalent to 2.2% of South Africa's GDP. Meeting the NAAQS in 2012 would have saved 155.8 billion Rands (2019 ZAR). This study provides additional motivation for the importance of meeting existing standards. There is an economic cost of reducing air pollution, but it is important to remember that there is also an economic cost of not reducing air pollution.

Reference

Altieri, KE, Keen, S. Public health benefits of reducing exposure to ambient fine particulate matter in South Africa. *Sci Total Environ* 2019, 684: 610-620



Company profile

Originally formed in 1976, SI Analytics provides air monitoring solutions to industry, government and research organisations. Our analytical instrumentation offers continuous measurement of both surrounding air pollution and chimney emissions.

We design, manufacture, supply, install, commission, train and provide after sales service for either individual monitors, or integrated systems in southern Africa.

Being a founding member of Europa Environmental gives us the expertise to supply and support advanced environmental monitoring technology. To meet your environmental monitoring needs, our aim is to bring you the world's best instrumentation, spare parts, service, technical support and training at the most affordable prices.

Services

Due to government regulatory requirements, our clients are under increasing pressure to collect emissions data. This is not part of their core business - their expertise lies in manufacturing the products they sell. Our expertise lies in assisting them to manage their data and compliance issues.

We offer a wide range of services such as ambient air quality and continuous emissions monitoring which include:

- environmental monitoring,
- certified USEPA, European EN and UK MCerts equipment sales,
- data management and reporting,
- service and maintenance contracts,
- equipment rentals,
- Indoor air quality assessments and,
- short courses (For example: operator monitoring assessment training).

Products

Stand alone products, integrated systems, spare parts, local factory repair, and on-site maintenance are all part of what SI Analytics has to offer, thus providing tailor-made solutions to client applications.

Instrumentation from the most popular products includes:

Ecotech:	manufactures a range of ambient air quality instruments.
Grimm:	mobile and stationary ambient dust monitors.
Ecochem:	extractive high sensitivity gas analyser systems.
Procal Analytics:	in-situ continuous stack gas analysers.
Chromatotec:	chlorine, hydrocarbon and sulphur speciation analysers.

Clients

SI Analytics has successfully installed and maintained various analysers and systems for a variety of clients such as:

We also provide data management and monthly reporting services for clients throughout South Africa, Botswana, Mozambique, Zambia and Zimbabwe, amongst others.

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Research article

Measurement of atmospheric black carbon in some South Mediterranean cities: Seasonal variations and source apportionment

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Received: 12 April 2019 - **Reviewed:** 4 June 2019 - **Accepted:** 10 September 2019
https://doi.org/10.17159/caj/2019/29/2.7500

Abstract

This study aims to investigate, for the first time in Algeria, the atmospheric black carbon (BC) concentrations over one year measured at the Scientific Observatory of Algiers and to compare their concentration levels with other Mediterranean cities (i.e., Athens and Crete). The diurnal cycles as well as seasonal variations of BC concentrations were evaluated and attributed to their emission sources (fossil fuel: BC_{ff} and wood burning: BC_{wb}). The annual mean concentrations of BC, BC_{ff} and BC_{wb} were 1.113±2.030, 1.064±2.002 and 0.049±0.262 μgm^{-3}, respectively. The highest seasonal mean concentrations were recorded in summer and autumn with 1.283±1.346 and 1.209±1.149 μgm^{-3} for BC and 1.217±1.431 and 1.177±1.151 μgm^{-3} for BC_{ff}, respectively. However, the lowest mean concentrations were recorded in winter and spring with 1.023±1.189 and 0.966±0.964 μgm^{-3} for BC and 0.933±1.177 and 0.956±0.874 μgm^{-3} for BC_{ff}, respectively. For BC_{wb}, the highest mean concentrations were reached in winter and summer with 0.090±0.055 and 0.066±0.050 μgm^{-3}, respectively, very likely due to the forest fires and long-range transport of air pollution from Europe. The lowest mean concentrations of BC_{wb} were recorded in autumn and spring with 0.032±0.033 and 0.010±0.021 μgm^{-3}, respectively. Seggregating BC levels into eight wind sectors, showed that the prevailing BC pollution with concentrations reaching up to 5.000 μgm^{-3} originated from the North-West wind sector. A source apportionment of BC for the wet and dry period was also perfomed followed by a back trajectory cluster analysis for long-range transport.

Keywords

Aerosol, black carbon, atmospheric pollution, source apportionment, seasonal variation, year modulation.

Introduction

Anthropogenic emissions of aerosols in the atmosphere have increased historically affecting air quality, human health and regional radiative forcing (Seinfeld and Pandis, 2006). Soot or black carbon (BC) is considered a reliable indicator of air pollution at a regional scale (Streets et al., 2001). Because of its submicron diameter, BC is capable of provoking numerous respiratory diseases and impacting the cardiovascular system, penetrating deep into the lungs and being deposited on the pulmonary alveoli (Cheng et al., 2014). Black carbon plays a great role in the climate system and is responsible for direct and semi-direct effects on regional and global climate (Bond et al.,

2013; Wang et al., 2015) as it absorbs solar radiation and affects the thermal stability of the atmosphere and precipitation (Jose et al., 2016). BC includes elemental carbon (EC, present as graphite), the dominant light absorbing material, that is generally co-emitted and coated by polycyclic aromatic hydrocarbons and humic-like substances or brown carbons (Andreae and Gelencser., 2006).

In addition, once emitted into the atmosphere, BC is subject to atmospheric aging that leads to particles of aged BC that are internally mixed with soluble components such as sulfates or nitrates and also water soluble organics, which increase aerosol solubility (Jennings et al., 1996). Such particles (aged BC) can contribute to the number concentrations of cloud condensation nuclei (CCN) (Bahadur et al., 2012).

Furthermore, during rainfall the fresh BC, which is not hygroscopic, can be removed from the atmosphere by wet scavenging (below-cloud scavenging), while aged hygroscopic BC particles can be removed both by in-cloud and below-cloud scavenging due to condensation of secondary inorganic aerosols on BC (Gadhavi and Jayaraman, 2010).

The physical and chemical properties of aerosols change significantly when BC is mixed with dust aerosols during dust events (Pu et al., 2015). The binding of organic pollutant particles with BC influences their transport and limits their bioavailability (Ali et al., 2014). It was proved that BC adsorbs strongly to pesticides, polyaromatic hydrocarbons (PAHs), biphenyls (PCBs) and polychlorinated dibenzo-p-dioxins (PCDDs), which can exacerbate water and soil pollution and human disease (Lohmann et al., 2005). BC can travel hundreds to thousands of kilometers in the atmosphere, but in sediments, it can live up to several millions of years (Masiello and Druffel, 1998). During monsoon season, low BC concentrations are associated with higher wind speeds and rainfall (Begam et al., 2016). The diurnal evolution of the atmospheric boundary layer (ABL) is a determinant factor for the variation of BC concentrations during the nighttime (Nair et al., 2007). The surface heating during the daytime increases the ABL height and results in a dilution of atmospheric aerosols in the ABL, thus decreasing BC concentration (Jose et al., 2016). In Algeria, similarly to other southern Mediterranean countries, no detailled studies of BC levels have been reported to date, to the best of our knowledge. The present study aims to document, for the first time, BC air pollution in Algiers through a full year of BC measurement performed at the observatory of Algiers from 1 June 2014 to 31 May 2015.

Experimental

Description of the measurement site

The sampling station is located at the Scientific Observatory of Bouzaréah, Algiers at the Centre de Développement des Energies Renouvelables (CDER). The site is situated at 36.8°N, 3.0°E, at 345m above the sea level in the highest Algiers plateau and at about 1 km from the Mediterranean Sea coast. It is also located about 6 km to the north-west of the Algiers downtown and has no significant industrial and agricultural combustion activities surrounding the sampling site at a distance of several kilometers; however, there is a forest about 3 km to the west (see Figure 1).

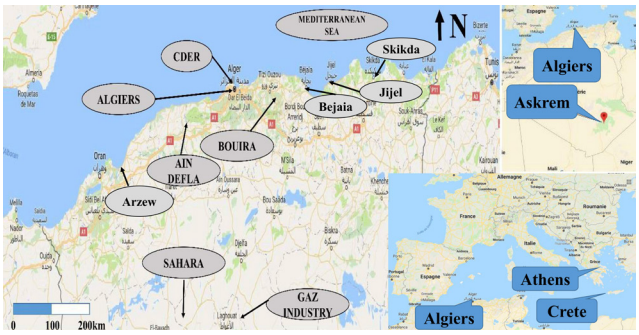

Figure 1: *Location of investigated sampling site.*

Instruments

Aethalometer

From 1 June 2014 to 31 May 2015, continuous BC observations have been performed at a height of 3m above ground level using an aethalometer (model AE-33 of Magee Scientific, USA).

The aethalometer AE33 instrument measures the light beam attenuation in seven wavelengths (370, 470, 520, 590, 660, 880 and 950 nm), operating with a flow rate of 5 L/min, a 1-minute measurement interval and an automatic zero calibration with an accuracy of 0.001 μgm^{-3}. According to the manufacturer, the instrument sensitivity is 0.03 μgm^{-3} at 1 minute and its detection limit is 0.005 μgm^{-3} for a 1-hour mean. The aethalometer uses the patented Dual Spot method to compensate for the 'spot loading effect' and provides a real-time output of the 'loading compensation' parameter, which may provide additional information about the physical and chemical properties of the aerosol (aethalometer Model AE33, user manual). The aethalometer AE33 used in the present study is equipped with a sampling head, which has an inlet with a diameter allowing the entry of $PM_{2.5}$. The BC measured in the present study was obtained by the following equation.

$$BC_{reported} = BC_{zeroloading} * (1 - k * ATN) \quad (1)$$

where $BC_{reported}$ is the BC measured by the aethalometer, $BC_{zeroloading}$ is the BC measured by the instrument without loading effect, k is the loading compensation parameter, and ATN is the attenuation of light beam in the wavelength of measurement. The equations below are taken from the aethalometer model developed by Sciare et al., (2011). Aerosol absorption coefficients (b_{abs}) were obtained by equations 2 and 3.

$$b_{abs,470nm} = BC_{470nm} * 14.54 / 1000 \quad (2)$$

$$b_{abs,950nm} = BC_{950nm} * 7.19/1000 \quad (3)$$

where 14.54 and 7.19 are the Mass Absorption Efficiency (MAE) in the two wavelengths, 470nm and 950nm, respectively, and are provided by the manufacturer. We used the measurements at 470nm in order to avoid the absorption by the dust at 370nm. The equations 4 to 7 enabled the calculation of the BC_{ff} (fossil fuel) and BC_{wb} (wood burning).

$$b_{abs,\lambda} = b_{absff,\lambda} + b_{abswb,\lambda} \quad (4)$$

$$b_{abs,ff,470nm} / b_{abs,ff,950nm} = (470/950)^{-\alpha ff} \quad (5)$$

$$b_{abs,wb,470nm} / b_{abs,wb,950nm} = (470/950)^{-\alpha wb} \quad (6)$$

$$BC_{ff} = BC * b_{abs,ff,950nm} / b_{abs,950nm} \quad (7)$$

where $b_{abs,\lambda}$ is the light absorption coefficient at the wavelength λ (we used 950nm wavelength), $b_{abs,ff,\lambda}$ is the light absorption coefficient for the BC_{ff} and $b_{abs,wb,\lambda}$ is the light absorption coefficient of BC_{wb}, α_{ff} and α_{wb} are the Angstrom exponents for fossil fuel and wood burning in Algiers, respectively. The α_{ff} and α_{wb} calculated by Sciare et al. (2011) were equal to 1 and 2 respectively. The combination of these equations using dedicated software developed by Sciare et al. (2011) was applied for the calculation of BC_{ff} and BC_{wb}. In addition to BC, biomass-burning aerosols contain a substantial fraction of organic substances, which absorb in the N-UV and blue part of the spectrum in contrast to the N-IR wavelength range, resulting in α_{wb} larger than α_{ff} (Zotter et al., 2017). It is also important to mention that BC exists in mixed nature (from fossil fuel and wood burning at the same time).

A calculation of the mass absorption cross-section (MAC) has been performed in this study by using the following equation:

$$\sigma_{air} = \frac{S * (\Delta ATN/100)}{BC * Fin\Delta t * C} \quad (8)$$

where, S=spot size; t=time, C=multiple scattering parameter (Weingartner et al., 2003), σair=mass absorption crossection.

$$Fin = Fout * (1-\zeta) \quad (9)$$

where, Fout=measured flow, ζ= leakage factor.

$$ATN = -100 * \ln(\frac{I}{I_0}) \quad (10)$$

Where, I0=reference signal and I=spot signal.

Meteorological data

Local meteorological parameters such as air temperature, relative humidity, wind speed and direction, atmospheric pressure and rain intensity were monitored by the CHEMS network of the CDER (composed of a weather and radiometric station) at 5-minute intervals using instruments situated near the aethalometer.

Traffic informations

Two national roads exist near the site about 1 km to the north and about 2km to the east, and roads with light traffic are about 200m to the south-east and 1.5 km to the west of the monitoring site. These roads can be sources of BC emissions. Algiers's car fleet includes more than 1,400,000 vehicles of all categories, with 31.32% using diesel and 68.68% using petrol as of the end of 2014, and about 1 million cars coming daily from the other regions. The car fleet in Algeria is old (51.11% of vehicles' age is greater than 20 years) (http://www.ons.dz/-Au-31-12-2014-.html) and has more than 5,000,000 cars of all categories, with 34.29% powered by diesel versus 65.71% by petrol. Almost all goods are transported by road. Algiers has also a train station, a harbor, an airport, and an industrial area situated at about 4, 5, 25 and 20 km from the measurement site, respectively.

Results and discussion

Monitoring of black carbon

One-year observations of BC, BC_{ff} and BC_{wb} with hourly, diurnal, and seasonal evolutions allow better understanding of high pollution events due to the rush hour traffic emissions, wild fires, oil industry to the south, celebration events (coinciding with high BC emissions due to abusive use of pyrotechnic products) and long-range air pollution transport from Europe or neighboring countries.

Black carbon variability

Figure 2 presents BC concentrations recorded at the scientific observatory of Bouzaréah (from 1 June 2014 to 31 May 2015), along with BC_{ff} and BC_{wb} calculated for the site. The BC concentrations were recorded at 950 nm wavelength, and the missing data are indicated with blanks. The equal distance between the high peaks is explained by the moving of the tape roll when the attenuation (ATN) at 370 nm reaches 100 (Sciare et al., 2011), which leaves gaps of two to three minutes. BC and BC_{ff} concentration levels were high with several peaks reaching 50.000 μgm^{-3}, however, BC_{wb} values were low and usually close to 0 μgm^{-3}. The BC, BC_{ff} and BC_{wb} mean concentrations were 1.113±2.030, 1.064±2.002 and 0.049±0.262 μgm^{-3}, respectively. It is worth noting that the BC_{ff} represents 95.60% of BC total concentration mass, suggesting that the main BC pollution originated from fossil fuel (traffic and oil industry). It is important to mention that the main source of heating in Algiers and in Algeria in general is natural gas, which is also the principal source of electricity production (96%). Therefore, the predominant BC_{ff} emissions are diesel, gasoline and kerosene used in the transport sector.

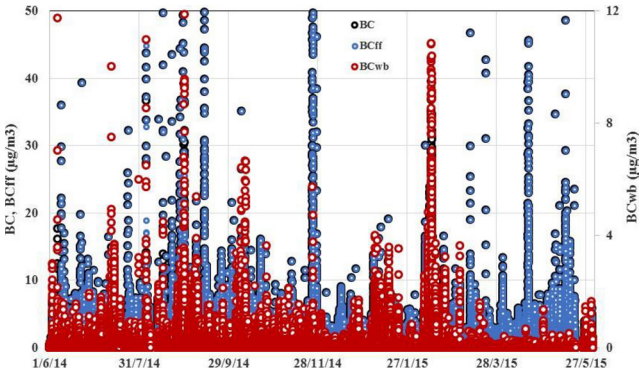

Figure 2: *One-year of BC, BC_{ff} and BC_{wb} measurements.*

The monthly averages of BC varied between 0.759±0.669 and 1.556±2.366 µgm^{-3} as presented in the Figure 3. The results are close to the previous ones presented in the Figure 2 with large standard deviation values due to the huge quantity of results treated in the present study.

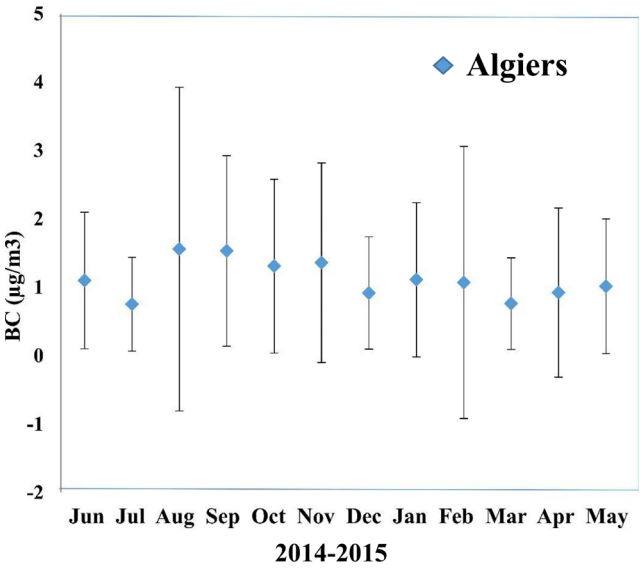

Figure 3: *Variation of monthly BC averages and standard deviations during the year of measurement.*

The daily peaks of BC exceeding 5.000 µgm^{-3} could be related to forest fires or a regional source (heating) which can be confirmed by the BC_{wb} concentrations, e.g., on 29 August 2014, the mean concentration of BC_{wb} from 02:00 to 09:00 was 0.450 µgm^{-3}. On 4 August 2014, BC_{wb} mean concentration from 04:00 to midnight (00:00) was 0.180 µgm^{-3} and on 7 September 2014, BC_{wb} mean concentration from midnight (00:00) to 15:00 was 0.055 µgm^{-3}. The yearly percentage of BC_{ff} was very high (95.60%), compared to BC_{wb} (4.40%). This result revealed that the main sources of BC in Algeria are local activities and road traffic rather than forest fires and cooking.

These results could be of great importance for air quality management policy. The measured BC concentrations can be compared with literature data from other locations as presented in Table 1. The annual average of BC recorded at the Algiers Observatory was much lower than that recorded in Anantapur (India), Prague (Czech), Athens (Greece) and Rome (Italy). In contrast, it was higher than measured values in Santa Cruz de Tenerife (Spain), Crete (Greece) and Finland. Chiloane et al., (2017) recorded BC mean concentrations ranging between 0.7 and 1.4 µgm^{-3} in background sites and sites influenced by industrial activities and/or nearby settlements in South Africa.

A thorough investigation of BC variation in the present study is needed for a comparison and characterization in order to explain these findings.

The highest seasonal mean concentrations of BC, BC_{ff} and BC_{wb} were recorded in summer with 1.283±1.346, 1.217±1.431 and 0.066±0.050 µgm^{-3}, respectively, which could be explained by the scarcity of rains and winds on the one hand and the increase of visitors to the north of Algeria during summer holidays on the other hand. The high BC_{wb} levels in summer are attributed to the forest fires recorded during that season.

The second most polluted season was the autumn, where mean concentrations reaching 1.209±1.149 µgm^{-3} for BC and 1.177±1.151 µgm^{-3} for BC_{ff} were recorded, explained by the intensive socioeconomic activity started after the summer holidays which is reflected by the traffic road increase, however, the BC_{wb} mean concentration was lower at 0.032±0.033 µgm^{-3} due to the decrease of the magnitude of forest fires during this season, and the low use of wood burning in Europe. During winter, the mean concentrations of BC and BC_{ff} were lower than in autumn (1.023±1.189 and 0.933±1.177 µgm^{-3}, respectively); however, the BC_{wb} mean concentration was the highest at 0.090±0.055 µgm^{-3}, which can be related to air masses originating from Europe and having a high BC_{wb} load (due to domestic heating emissions).

During spring, the mean BC, BC_{ff} and BC_{wb} concentrations were the lowest with 0.966±0.964, 0.956±0.874 and 0.010±0.021 µgm^{-3}, respectively, attributed to air masses coming mostly from the north-west, with low BC_{wb} from Europe (low use of wood and coal for heating in spring), south, and local sources with a very high rate of BC_{ff} (98.96%) due to petroleum industry and traffic.

For the sake of comparison, Table 2 reports the seasonal variations of BC measured in this study and those reported in the literature.

The mean concentrations of BC recorded in the present study during summer was higher than those measured in Prague (Czech) and Mahabaleshwar (India). In contrast, the BC concentrations measured in this study were lower than mean concentrations documented for six large Brazilian cities. The results recorded in winter are higher than those measured in Paris and Toulouse, but lower than the mean concentrations obtained in Cairo (Egypt), Prague (Czech Republic) and six large Brazilian cities. For the spring season, the concentration levels were higher than those reported in Stockholm (Sweden), but, lower than the mean concentrations recorded in Prague and

Table 1: *Comparison of [BC] (in µg m^{-3}) measured in the present study and other concentrations reported in the literature.*

Station	Type of site	BC Concentration	Period	Reference
Algiers (Algeria)	Suburban	1.113±2.030	June 2014–May 2015	Present study
Hyderabad (India)	Urban	15.91	March (2010–2012)	(Jose et al. 2016)
Hyderabad (India)	Urban	9.84	June (2010–2012)	(Jose et al. 2016)
Anantapur (India)	Semi-arid, Suburban	3.03		(Reddy et al., 2012)
Indian Himalayas	Background	0.90±0.60	2005-2014	(Hooda et al., 2018)
Prague (Czech)	Suburban	1.71	Sep 2009–Aug 2010	(Vodička et al. 2013)
Barcelona (Spain), Lugano (Switzerland), London, and North Kensington (England)	Urban background	1.7–1.9	2009	(Reche et al. 2011)
London, Marylebone (England)	Urban traffic	7.8	2009	(Reche et al., 2011)
Bern (Switzerland)	Urban traffic	3.5	2009	(Reche et al., 2011)
New York(USA)	Urban	1.38	Jan–Feb 2004	(Venkatachari et al., 2006)
Mexico City(Mexico)	Urban	3.4	Apr 2003	(Salcedo et al., 2006)
Hong Kong (China)	Coastal rural	2.4	Jun 2004–May 2005	(Cheng et al., 2006)
Huelva (Spain)	Urban background	0.7	2009	(Reche et al., 2011)
Santa Cruz de Tenerife (Spain)	Urban background	0.8	2009	(Reche et al., 2011)
Finland	Urban	1.71	Sep 2009–Aug 2010	(Saarikoski et al., 2008)
Baltic Sea	Coastal rural	0.6	2008–2009	(Byčenkienė et al., 2011)
South of China	Oceanic site	0.28–2.14	Daily BC averages	(Wu et al., 2013)

Table 2: *Comparison of seasonal [BC] (in µgm^{-3}) measured in the present study and other values reported in the literature.*

Station	Type of site	BC Concentration	Season	Reference
Algiers (Algeria)	Suburban	1,283±1.346	Summer	Present study
		1,209±1.149	Autumn	
		1,023±1.181	Winter	
		0,966±0.964	Spring	
Prague (Czech)	Suburban	1.26	Spring	(Vodička et al. 2013)
Prague (Czech)	Suburban	0.87	Summer	(Vodička et al. 2013)
Prague (Czech)	Suburban	2.06	Autumn	(Vodička et al. 2013)
Prague (Czech)	Suburban	2.66	Winter	(Vodička et al. 2013)
Six large Brazilian cities		2.30–7.10	Summer	(De Miranda et al. 2012)
Six large Brazilian cities		4.0–13.1	Winter	(De Miranda et al. 2012)
Stockholm (Sweden)	Rural	0.36	Spring	(Krecl et al. 2011)
Stockholm (Sweden)	Street canyon site	5.39	Spring	(Krecl et al. 2011)
Cairo (Egypt)		9.9	2004 Autumn	(Mahmoud et al. 2008)
Cairo (Egypt)		6.9	2005 Spring	(Mahmoud et al. 2008)
Paris (France)	Suburban site	0.9	Winter	(Laborde et al. 2013)
Toulouse, France	Coastal urban	0.95	2005 Winter	(Saha and Despiau. 2009)
Mahabaleshwar (India)	Rural	0.303	Summer	(Singla et al. 2019)

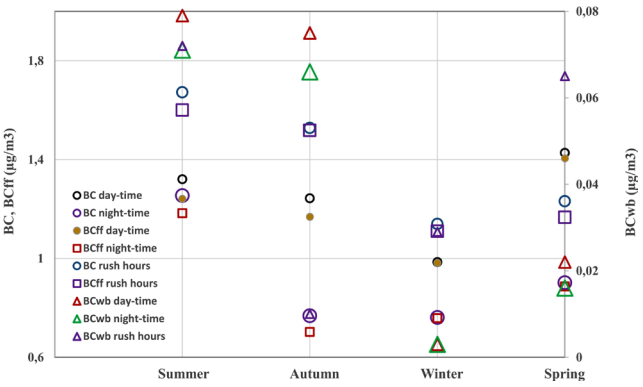

Figure 4: *Daytimes, nighttimes and rush hours mean concentrations of BC, BC_{ff} and BC_{wb} quarterly averages.*

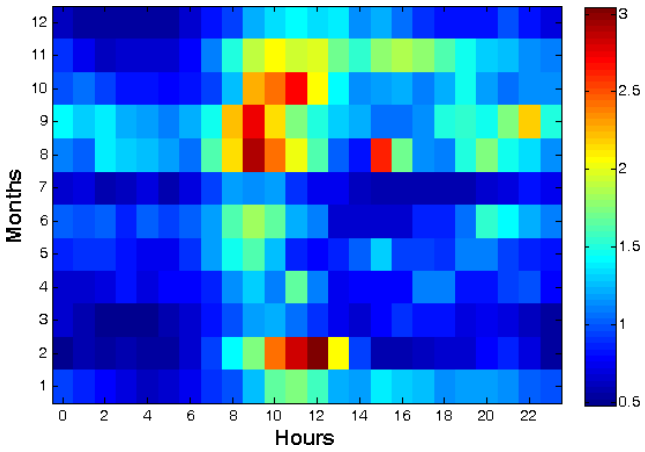

Figure 5: *Month-hour plot of BC (µgm^{-3}) during the year of measurement.*

Cairo. Finally, during autumn, the mean concentrations recorded in Algiers were lower than those measured in Prague (Czech). The difference between the seasonal mean concentrations of BC recorded in Algiers and other cities could depend not only on the national emissions related to wild fires, industry, and transport, but also on the long-range transport pathways which will be studied in the rest of the study.

Figure 4 displays the seasonal variation of the daytime, nighttime and rush hour mean concentrations of BC, BC_{ff} and BC_{wb}, during the studied one-year period.

It shows that during the daytime, spring was the most polluted season with BC and BC_{ff} mean concentrations reaching up to 1.427±1.045 and 1.405±1.043 µgm^{-3}, respectively, followed by the summer season when concentrations of 1.320±0.958 µgm^{-3} for BC and 1.241±1.504 µgm^{-3} for BC_{ff} were recorded.

These high mean concentrations of BC with high rates of BC_{ff} reaching 98.46% in spring season can be explained by the decrease of rains, the increase of airflow from the south charged with BC_{ff} emitted by oil industry and the air masses coming from Europe.

In summer, the rate of BC_{ff} concentration was less at 94.00% due to the wild fires emitting BC_{wb}. The lowest mean BC and BC_{ff} concentrations were recorded in the winter season at 0.985±1.409 and 0.982±1.401 µgm^{-3}, respectively, which can be explained by the wet scavenging by the rain and the dispersion by winds.

During nighttime, summer is the most polluted season by BC and BC_{ff} with concentrations of 1.254±1.306 and 1.183±1.306 µgm^{-3}, respectively, followed by the autumn season with 0.902±0.779 and 0.886±0.781 µgm^{-3}, because of the increased use of cars during the night contrary to autumn and winter seasons. It is worth noting that the majority of the industries in Algeria work 24 hours a day, leading to increased emissions of BC_{ff} by cars and machines during the night.

As to BC_{wb}, the highest mean concentrations were recorded during the summer followed by the autumn with 0.071±0.571 and 0.066±0.028 µgm^{-3} respectively, which is due to the forest fires in summer and wood burning coming from Europe in autumn with a scarcity of rains in Algeria. The lowest BC_{wb} was observed in the spring with 0.003±0.194 µgm^{-3}, which can be explained by the wet scavenging by rains and the dispersion by winds.

As the cooking and heating in Algeria is from natural gas, the BC_{wb} measured in our study could be due to cooking and heating from Europe and from wild fires during the summer (both in Europe and in Algeria). The BC_{wb} in the daytime was higher than BC_{wb} at night during summer, autumn and spring, which is due to the fires recorded in Algeria in summer with a higher frequency and magnitude of spread during the day than at night and the low use of heating in Europe during spring and autumn. However, in winter, we recorded in Algiers a similar average of BC_{wb} concentration in the daytime and in the nighttime, which could be explained by the long-range transport from Europe with a high use of wood and coal for heating and cooking during the day and the night.

For the BC and BC_{ff} average concentrations in rush hours (07:00 and 10:00), summer and autumn were the most polluted with 1.672±1.297 and 1.528±1.167 µgm^{-3} for BC and 1.600±1.327 and 1.518±1.171 µgm^{-3} for BC_{ff}, respectively. These concentrations, which were higher than the yearly average, can be explained by the citizens' behavior during peak hours in Algeria: leaving work and school at the same time, and lunch between midday and 13:00 for most citizens. The seasonal maximum levels of BC_{wb} during rush hours (between 11:00 and 14:00) were recorded in summer and spring with 0.381±0.232 and 0.183±0.151 µgm^{-3}, respectively, most likely associated with cooking activities in restaurants and wild fires.

In Figure 5, we present the month-hour variation plot of BC during the period of measurement in Algiers as plotted by Hooda et al. (2018). The figure presents better the variation of BC depending on hours and months and completes the previous interpretations. It is clear that the BC peaks were observed in the summer during the rush hours. The BC concentrations during daytimes were very high during spring months, which is

in line with the previous interpretations. The figure summarizes what was presented previously and gives a more precise view of the hourly averages during the months studied.

Figure 6 shows the mean diurnal cycle of BC, BC_{ff} and BC_{wb} concentrations during the studied period of one year. The diurnal cycle reflects the combined effect of variations in anthropogenic emissions, surface meteorology and ABL dynamics (Ramachandran and Rajesh, 2007). Targino and Krecl, (2017) reported that local traffic was by far the most important source of BC in street canyon in a mid-sized city in southern Brazil, with hourly mean concentration peaking during the rush hours at 5.840 μgm^{-3} in the morning at 06:00 and at 4.550 μgm^{-3} in the afternoon at 18:00.

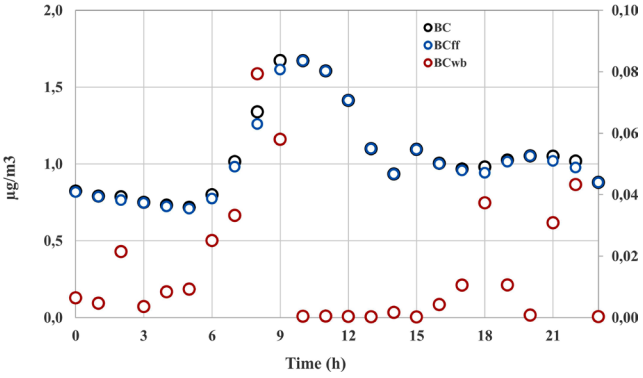

Figure 6: *Mean diurnal cycle of mean BC, BC_{ff} and BC_{wb} concentrations during the year of measurement.*

In the present study, the hourly mean concentrations of BC and BC_{ff} were higher than 1.300 μgm^{-3} between 08:00 and midday (12:00) with maximum concentration of 1.673 μgm^{-3} at 09:00, due to the high traffic. In contrast, the concentrations were as low as 0.718 μgm^{-3} for BC and 0.709 μgm^{-3} for BC_{ff} at 05:00. For BC_{wb}, low hourly mean concentrations were recorded with two peaks at 08:00 and 09:00 reaching 0.079 and 0.058 μgm^{-3}, respectively, most likely related to the indoor and outdoor cooking and heating.

In Figure 7, a comparison of BC daily variability with the means of wind speed (ws) and recorded rainfall has been presented. An inversely proportional relationship between the wind speed and rainfall on the one hand and BC concentrations on the other hand was observed. A statistical study of daily data revealed a correlation between ws and rainfall increase and BC decrease, which is very clear in Figure 7. When ws was less than 5 $m.s^{-1}$, the BC decreased with a good correlation (correlation coefficient K=0.16); however, BC concentrations increased with the ws higher than 5 $m.s^{-1}$. This result could be explained by the dispersion of BC pollution when winds are moderate. In contrast, the increase of BC when ws is higher than 5 m s^{-1} could be due to the atmosphere disruption. Nevertheless, a relationship between rainfall and BC decrease was recorded, with a better correlation when rainfall is less than 3mm. BC dispersion by winds and the wet scavenging by rains can explain the decrease of BC. High wind speed may increase the vertical mixing thereby diluting particles among them black carbon in

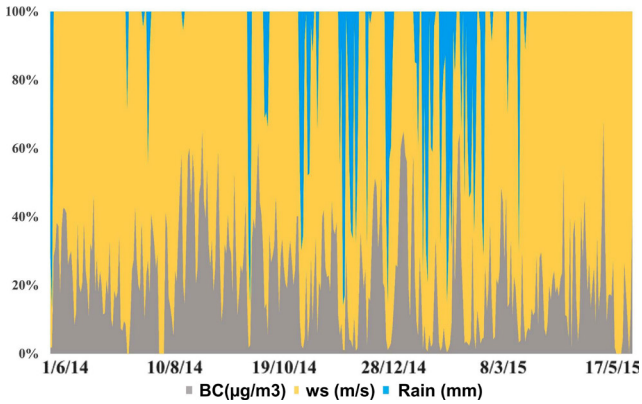

Figure 7: *BC daily variability with the wind speed (ws) and rainfall during the year of measurement.*

addition to the dispersion of BC by winds, which decreases the BC concentrations. Low atmospheric boundary layer height and low wind speeds during winter can be also attributed to high accumulation of BC aerosols (Begam et al., 2016).

To study the relationship between the atmospheric boundary layer (ABL) and BC concentrations, Table 3 presents the averages of BC vs ABL height. Cheng et al. (2014) revealed that the BC mass concentrations decreased during the afternoon hours, due to the boundary layer dynamics and a lower car fleet volume. Begam et al., 2016, confirmed the inverse proportional relationship between the ABL level and BC concentrations. In the present study, it was observed that BC levels were high when the boundary layer was lower than 1000m, and decreases with the ABC height, which is explained by the pressure applied by the ABL on the air pollutants, decreasing the volume and increasing the concentrations.

Table 3: *Variation of BC averages and Atmospheric Boundary Layer heights.*

ABL (m)	BC average (µg/m³)
1000	1.16
2000	1.10
3000	1.06
4000	1.05
5000	1.05

Sources of black carbon

Figures 8a,b,c,d show some pollution events (pollution from the Sahara, sport and religious celebrations, forest fires, long-range transport from Europe as well as a day without cars). The hourly averages of BC, BC_{ff} and BC_{wb} concentrations during 7 September 2014 are displayed in Figure 8a. High levels of BC and BC_{ff} reach 9.043 and 8.996 μgm^{-3}, respectively, between 09:00 and 10:00. A high peak of BC_{wb} reaching 0.133 μgm^{-3} between 10:00 and 11:00 was also measured. These high levels of BC_{wb} were attributed to the forest fires in Ain Defla in the south-west

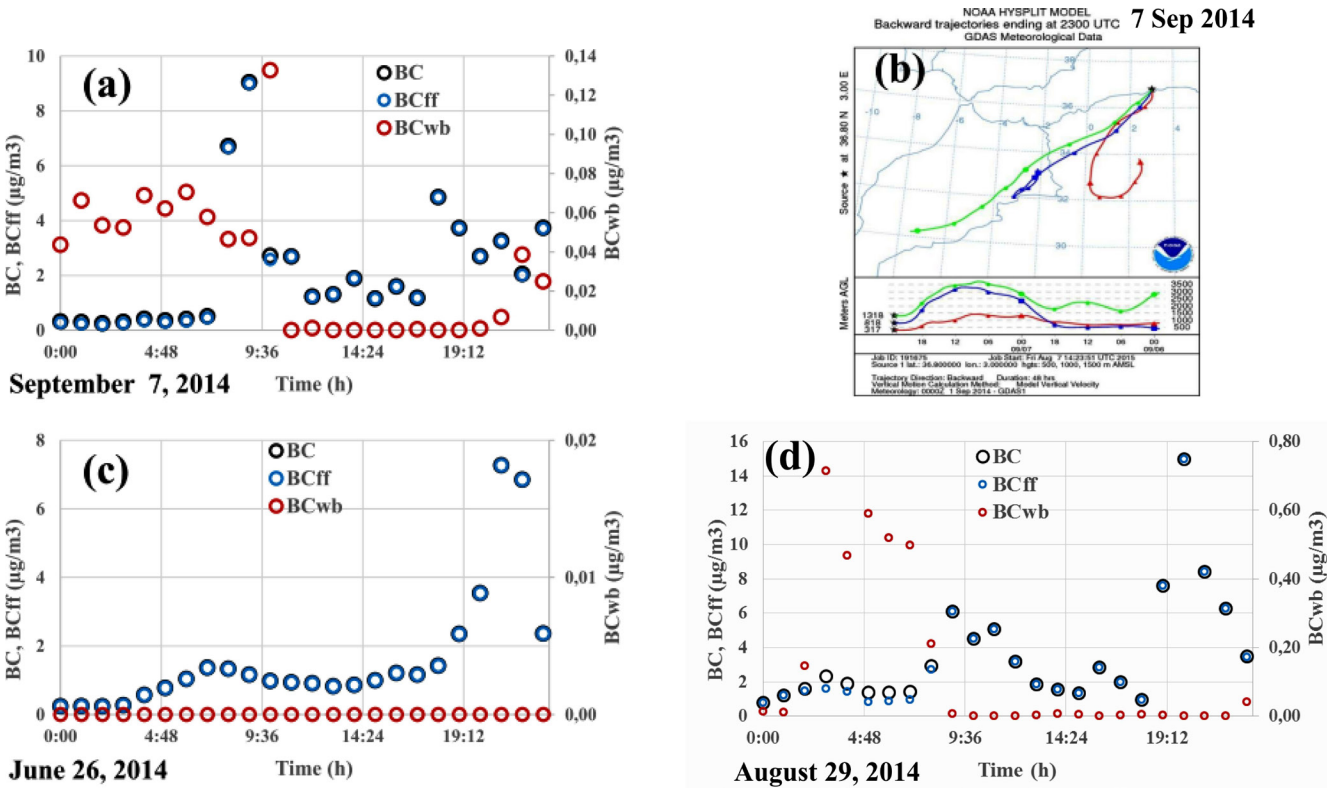

Figure 8: *Variation of BC, BC_{ff} and BC_{wb} concentrations on polluted days (7 September, 26 June and 29 August 2014).*

of Algiers that occured during the same period, as cited by the Algerian Directorate General of Forestry, and affecting the region as confirmed by the air mass trajectories shown in Figure 8b. The high levels of BC and BC_{ff} can be associated with other air masses coming from the petroleum industry in the Sahara (Figure 8b).

Figure 8c displays the hourly averages of BC, BC_{ff} and BC_{wb} on 26 June 2014, when the national soccer team of Algeria came for the first time to the second round of the 2014 World Cup. High levels of BC and BC_{ff} were recorded during the night of 26 June exceeding 7.000 μgm^{-3} between 21:00 and 23:00 with 98.88% of the BC being BC_{ff}. This pollution has been produced by traffic emissions and excessive use of pyrotechnic products exacerbated during this particular celebration event. The hourly averages of BC, BC_{ff} and BC_{wb} on 29 August 2014 are shown in Figure 8d. High peaks of BC and BC_{ff} reached up to 14.000 μgm^{-3} between 20:00 and 21:00 and high levels of BC_{wb} reached 0.715 μgm^{-3} between 03:00 and 04:00 and 0.589 μgm^{-3} between 05:00 and 06:00 with a daily mean concentration of 0.199 μgm^{-3}.

The Hysplit back trajectories (Figure 9a) corroborated the occurrence of huge forest fires in the east of Algiers (300 km away) in Bejaia and Jijel, as announced by the Director General of Forestry. The fires started on 27 August 2014 and the plume arrived at Algiers on 29 August 2014. The mean concentrations of BC and BC_{ff} between 20:00 and 21:00 were similar and very high (i.e., 14.953 μgm^{-3}), suggesting the dominance of fossil fuel sources of BC after the extinction of the forest fires. Interestingly, Figure 9b compares the variation of BC in two successive celebrations years of the Birthday of Muslim Prophet (Mawlid Enabawi) coincided with the nights of 2 January 2015 and 23 December 2015. High levels of BC concentrations were recorded with peaks exceeding 9.000 μgm^{-3} on 23 December and 5.000 μgm^{-3} on 2 January, and daily mean concentrations of 3.477 μgm^{-3} on 23 December, and 0.559 μgm^{-3} on 2 January, occuring during tremendous use of pyrotechnic products. Seidel and Birnbaum (2015) reported an increase of the US-average mean hourly $PM_{2.5}$ values reaching 21.000 μgm^{-3} 21:00 to 22:00 during the celebration of the national independence day in the US (4 July 1999-2013) and a decrease to zero by noon the day after (5 July 1999-2013). The authorities implemented a day without traffic road in the center of Algiers. This special case helps to assess to what extent the impact of traffic on BC emissions is important for BC levels in the Algiers city. Figure 9c presents a comparison of BC variations between an ordinary day (7 August 2015) and a day without cars in Algiers (31 July 2015).

A substantial drop in BC concentrations of between 47.3% and 92.4% was observed during the day without cars relative to the days with cars, considering that the air mass sources are local during the ordinary (day with cars) day and from the south-west from the region having no oil industry, for the day without cars as shown in Figure 10. The BC average concentrations during the day and the night of this day without cars were 0.564 and 0.543 μgm^{-3}, respectively, representing 50% on the annual average, which demonstrates the role of car fleet in BC levels in Algiers.

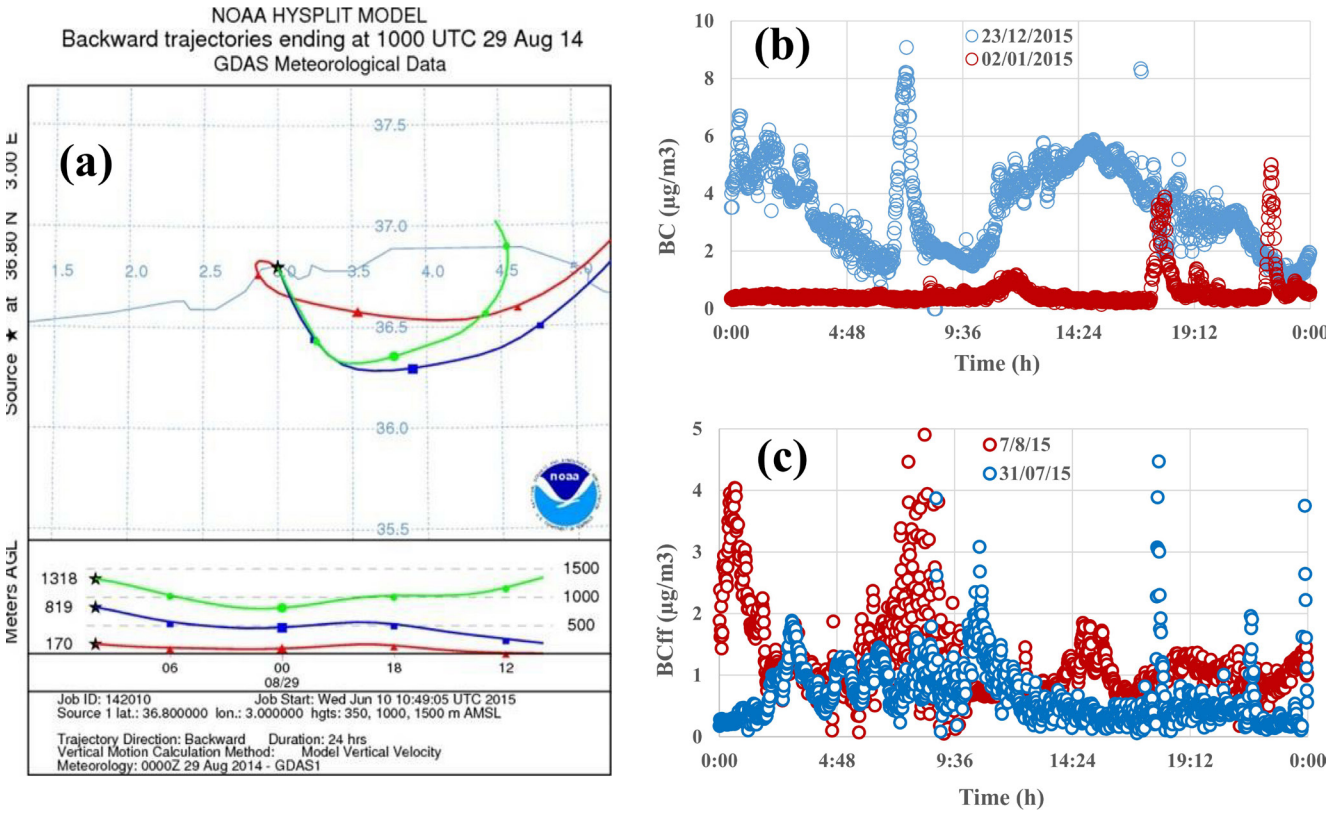

Figure 9: *Variation of BC during pollution events (29 August 2014, 2 January, 23 December, 31 July and 7 August 2015).*

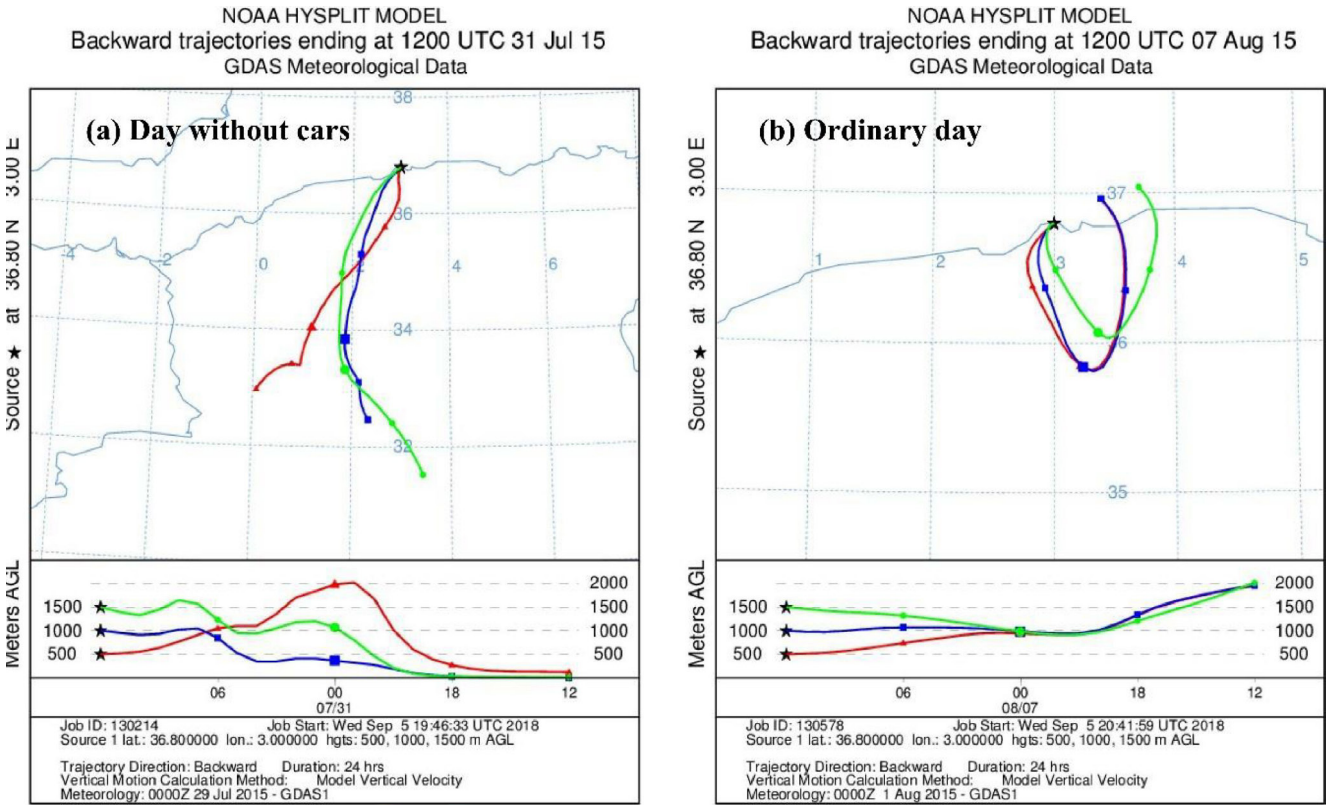

Figure 10: *Back trajectories confirming pollution events (31 July and 7 August 2015).*

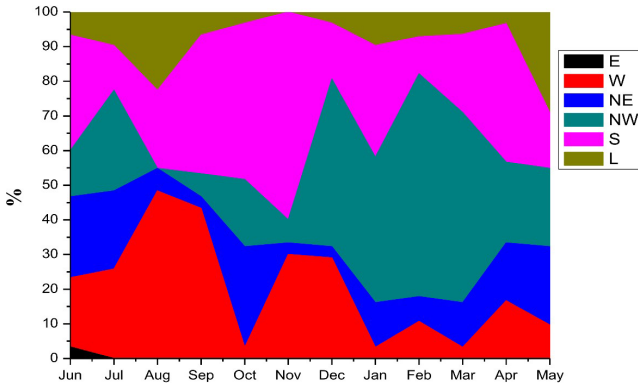

Figure 11: *Wind sources map of the measurement site during one year of BC measurement.*

Annual source apportionment of black carbon

Figure 11 displays the variation of wind origin at 500m above ground level at the measurement site based on 365 air mass back trajectories performed using the Hysplit model for the year of the measurements. These maps are important for the interpretation of the high peaks of BC air pollution and their emission sources. From July to September 2014, the westerly sector was the main sector of wind origin, with average frequency of occurrence of 25.80% in July, 48.38% in August and 43.33% in September. In October and November 2014, the winds came mostly from the south with frequency of occurrence of 45.16% and 60.00%, respectively. For December 2014, January, February and March 2015, the principal source of winds was from north-west with frequencies of 48.38%, 41.93%, 64.28% and 54.84%, respectively. April was characterised by southerly winds coming from the Sahara, with frequency of occurrence of 40.00%. In May, the winds were local (29.03%), from the north-east (22.58%) and the north-west (22.58%).

The relationship between BC and the source apportionment of winds (measured by CHEMS network of CDER) for the four seasons of the measurement period is depicted in Figure 12. A filtering of the BC coming from the eight wind sectors has been performed. In summer, BC predominantly came from the west (23.94%), the south (24.89%) and the east (23.08%). The polluted air masses came from the west and north-west with mean concentrations reaching 5 μgm^{-3} for southerly airflow and 3 μgm^{-3} for westerly airflow; however, the air masses coming from the east were cleaner (concentrations below 1 μgm^{-3}). During the autumn, the predominant BC mass fraction directions were from the south (34.80%), the west (32.39%) and the east (12.96%), with BC mean concentrations reaching up to 4 μgm^{-3} for the westerly and 3 μgm^{-3} for the southerly and the easterly sectors. In the winter, the prevailing air masses came from the west (34.40%), the south (19.97%) and the north-west (13.10%). The BC mean concentrations coming from the west, the south and north-west reached up to 3, 3 and 2 μgm^{-3}, respectively. In spring, BC mass fractions came mostly from the east (26.88%), the south (26.58%) and the west (22.88%). The BC mean concentrations reached up to 3 μgm^{-3} for the southerly and 2 μgm^{-3} for the easterly and the westerly sectors. Air masses rich in BC came from the west of Algiers.

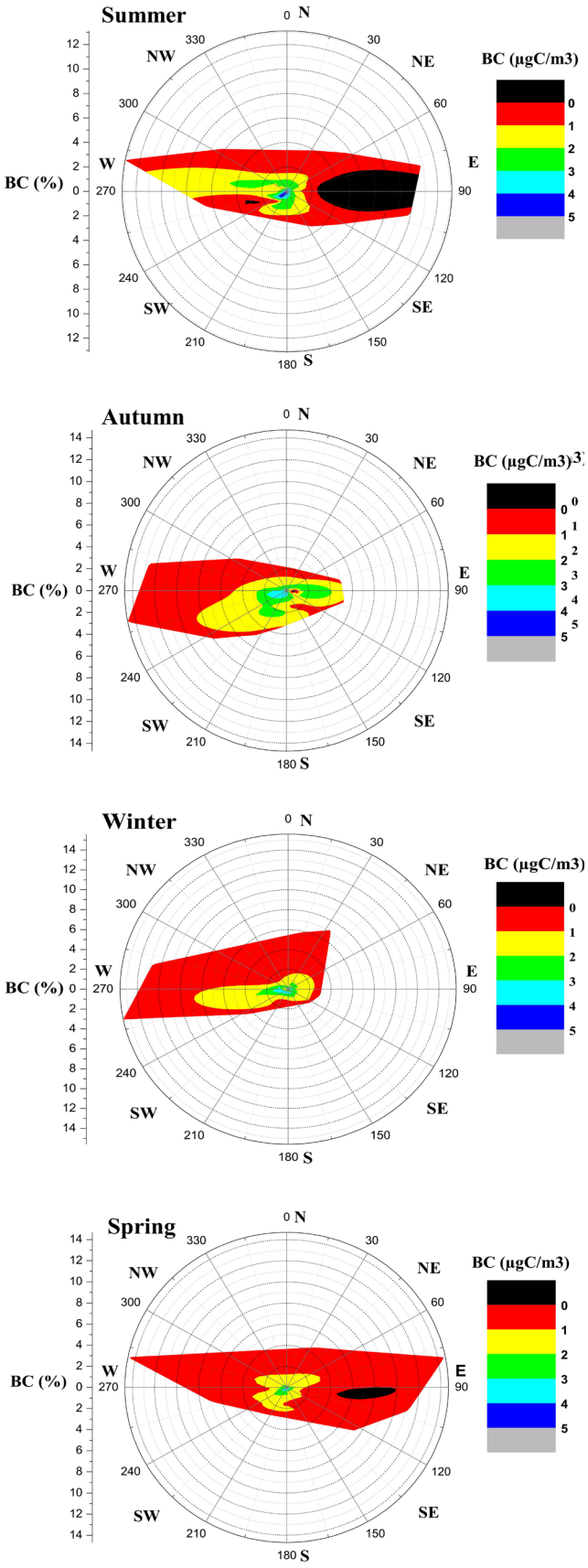

Figure 12: *Variation of BC versus the source apportionment of winds.*

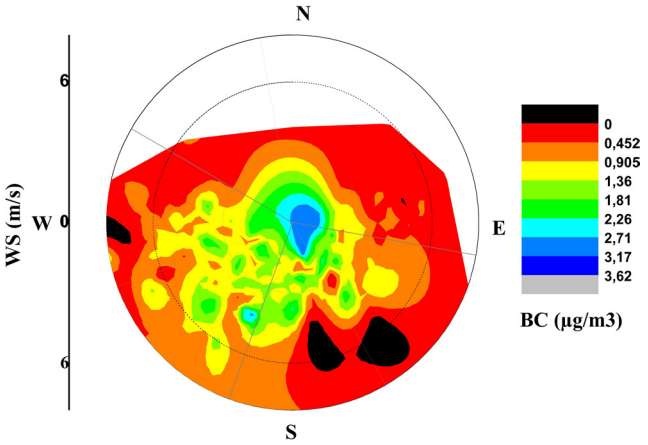

Figure 13: *. Variation of BC versus wind speed and wind direction for the entire sampling period.*

During the wet period, high BC mean concentrations of 1.333 µgm^{-3} were recorded in association with southerly winds (petroleum industry), followed by local winds with 1.238 µgm^{-3} (car fleet) despite the wet scavenging of BC. The highest mean concentrations of BC recorded during the dry period were from the easterly (1.380 µgm^{-3}) and westerly (1.377 µgm^{-3}) directions, coinciding with the emissions from the fires, harbour and airport to the east and petroleum refinery to the west.

Figure 13 displays a non-parametric wind regression analysis involving wind speed and source apportionment of winds (measured by CHEMS network of CDER) and BC concentrations, which could reveal more information on the source apportionment of BC. The mean concentration of BC was 0.725 µgm^{-3} when ws was higher then 4 m s^{-1}; however, it reached 1.806 µgm^{-3} for ws lower than 2 m s^{-1}, reflecting the dispersion of BC by winds. Figure 13 revealed also that the prevailing sources of BC were from the south, north-east and north-west with concentrations reaching up to 3.170 µgm^{-3} recorded when the wind speed was lower than 2 m s^{-1}. However, despite southerly winds reaching 4 m s^{-1}, high levels of BC of up to 3.170 µgm^{-3} have been recorded from the southerly direction, suggesting the emissions from the oil industry.

For a better understanding of the source apportionment of BC and its long-range transport pathways, a back trajectory cluster analysis has been performed. Hysplit 120-hour back trajectories arriving at the site every 6 hours for 365 days (from 1 June 2014 to 31 May 2015) were carried out. The 6-hour time intervals were centered around midnight (00:00), 06:00, 12:00 and 18:00. The trajectories arriving at the site at a height of 500m A.S.L, were performed by the model Hysplit developed by NOAA (Figure 14a) for the year of measurement. We defined six clusters (north-east, north-west, south, east, west and local as performed by Kouvarakis et al. (2000). A percentage of wind sources for each one of the four intervals (00:00, 06:00, 12:00 and 18:00) was calculated, followed by the calculation of the average of BC, BC_{ff} and BC_{wb} for each cluster during the four intervals. The north-westerly direction was predominant with percentages reaching up to 69, 66, 65 and 62% at 18:00, 12:00, 00:00 and 06:00,

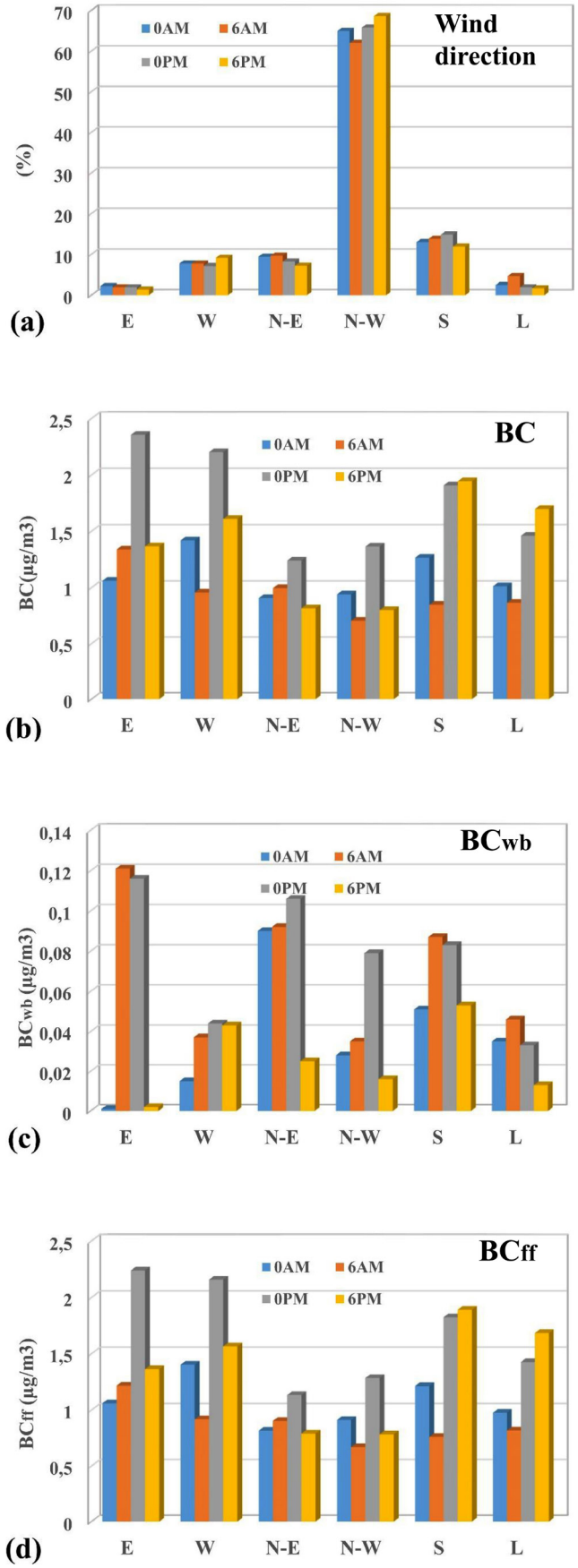

Figure 14: *Variation of BC, BC_{ff} and BC_{wb} concentrations with back trajectory cluster analysis.*

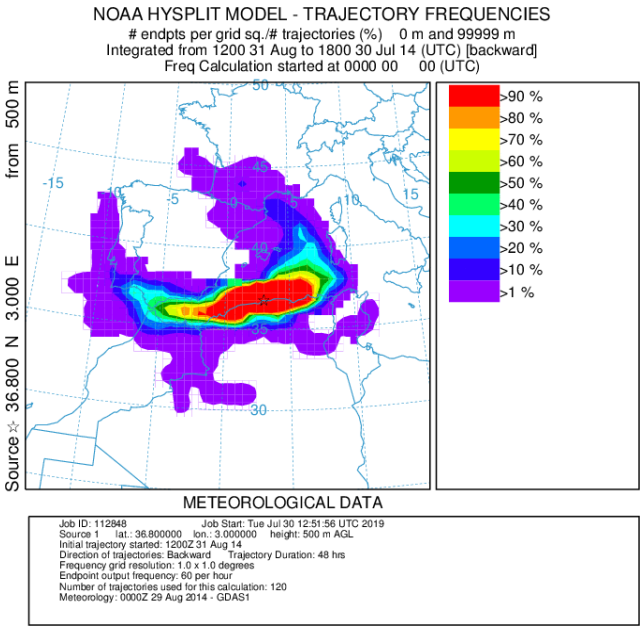

Figure 15: *. Backward trajectory frequency in summer (30 July-31 August) 2014 to Algiers site.*

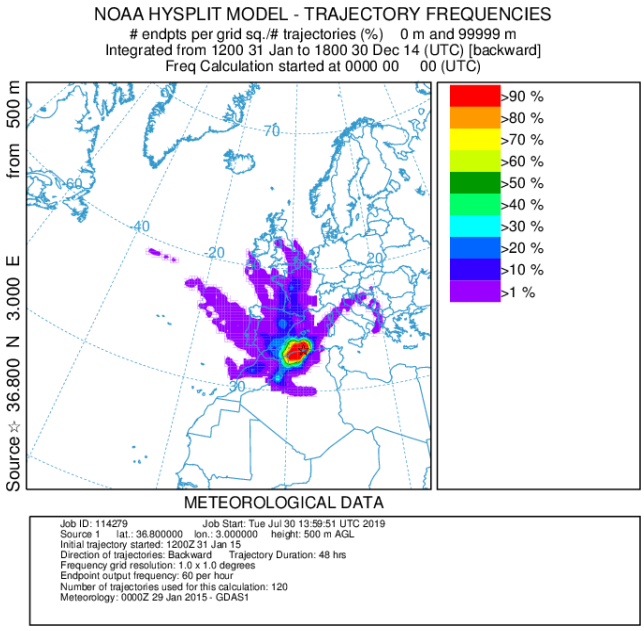

Figure 17: *. Backward trajectory frequency in winter (30 December 2014-31 January 2015) to Algiers site.*

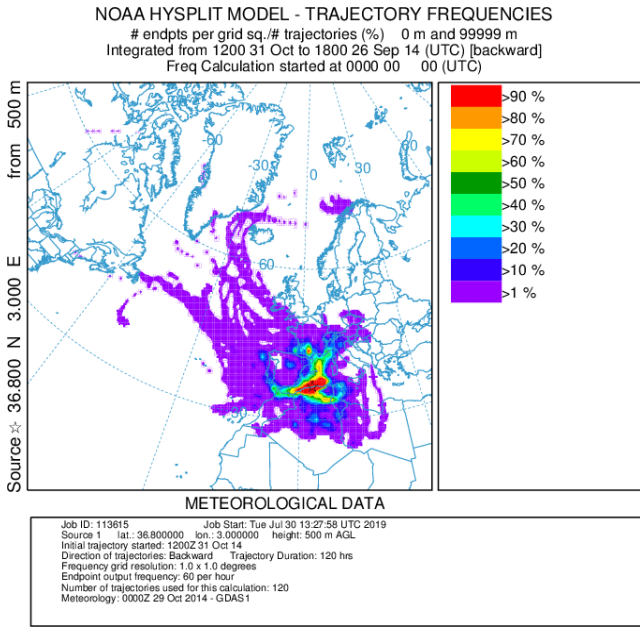

Figure 16: *Backward trajectory frequency in autumn (September-October) 2014.*

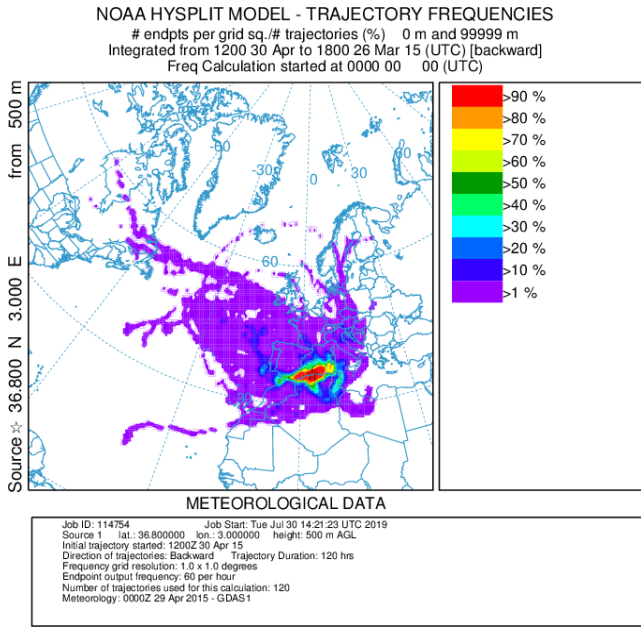

Figure 18: *Backward trajectory frequency in spring (26 March-30 April) 2015 to Algiers site.*

respectively, followed by the southerly direction with 15, 14, 13 and 12% at 12:00, 06:00, 00:00 and 18:00, respectively. These results confirm that the prevailing winds during the year of study were north-westerly followed by the southerly. Figures 14 b, c and d, display respectively the percentages of BC, BC_{ff} and BC_{wb} for the wind directions of the same four intervals during the year of measurements. For the midnight (00:00) cluster, the highest BC and BC_{ff} mean concentrations were recorded from the west followed by the south with 1.417 and 1.402 μgm^{-3} for BC and 1.263 and 1.212 μgm^{-3} for BC_{ff}, respectively, which can be explained by the presence of the oil refinery of Arzew in the west and the oil industry (pumping, processing, storage and transport) in the south. The highest mean concentrations of BC and BC_{ff} observed for the 06:00 cluster came from the east and the north-east for BC with 1.336 and 0.994 μgm^{-3}, respectively, and from the east and the west for BC_{ff} with 1.215 and 0.917 μgm^{-3}, respectively. The occurrence of such BC concentrations can be related to the harbor activities, the refineries to the east and west, the car fleet, and wood burning from Eastern Europe. With regards to the midnight (00:00) cluster, BC and BC_{ff} sources were mainly from the east followed by the west with concentrations of 2.354 and 2.200 μgm^{-3} for BC and 2.238 and 2.156 μgm^{-3} for BC_{ff}, respectively, which can be due to the harbor activities and petroleum refineries of Bejaia and Skikda in the east and of Arzew in the west.

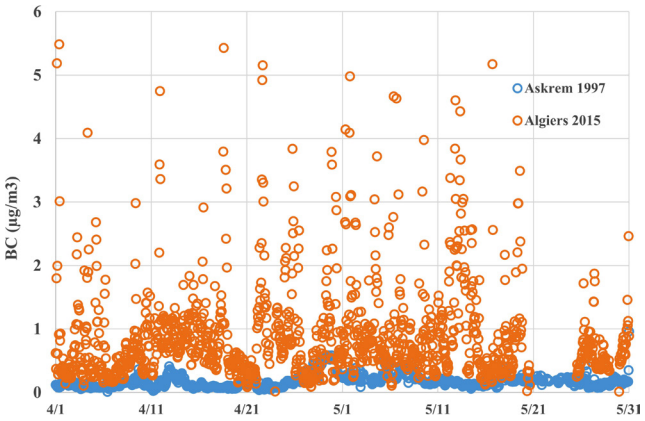

Figure 19: *Comparison of BC concentrations at Algiers and Askrem stations.*

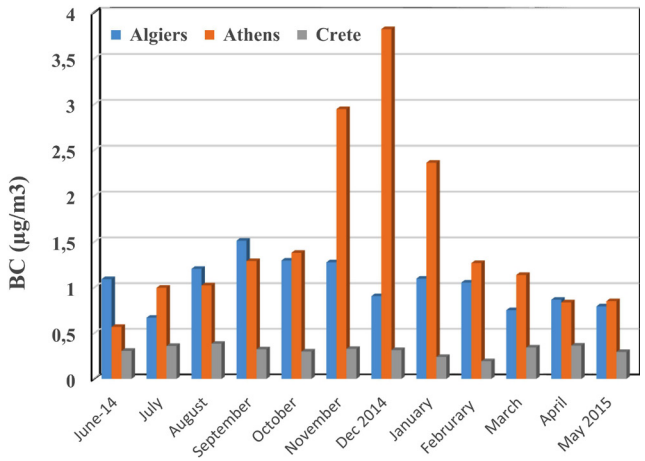

Figure 20: *Comparison of monthly average BC concentrations at Algiers, Athens and Crete sites.*

For the 18:00 cluster, the predominant BC and BC_{ff} mean concentrations were recorded from the south and in association with local airflow with 1.942 and 1.696 µgm^{-3} for BC and 1.889 and 1.683 µgm^{-3} for BC_{ff}, respectively, which can be due to the oil industry in the south and the local car fleet. For the case of BC_{wb} at midnight (00:00), the highest mean concentrations were predominantly from the north-east at 0,090 µgm^{-3}, which is double the previous average, due to wood burning in Europe, followed by from the south at 0,051 µgm^{-3}, close to the yearly mean, due to wild fires in the south of Algiers.

As to the 06:00 and midday clusters, the prevailing sources of BC_{wb} were to the east and the north-east with high means reaching up to 0.121 and 0.092 µgm^{-3}, and 0.116 and 0.106 µgm^{-3}, respectively, explained by wood burning in Eastern Europe and Tunisia. Finally, for the 18:00 cluster, the BC_{wb} predominant mean concentrations were close to the annual average and came from the south with 0.053 µgm^{-3} due to cooking in the southern Algeria cities and from the west with 0,043 µgm^{-3}, very likely derived from cooking in the western Algeria cities and from wood burning in Morocco.

For a better interpretation of the previous results, trajectory frequency maps were performed by the Hysplit model. Figure 15 displays the trajectory frequency in summer (July-August), where winds came mainly from the north-east and north-west, which is in line with the previous results bringing BC_{ff} and BC_{wb} emitted by fires and human activities.

Figure 16 presents the trajectory frequency map during autumn months (September-October) 2014. The prevailing winds were from the west, the south and the north-east, reinforcing the results presented in the meteorological map.

Figure 17 displays the trajectory frequency map in winter (December 2014-January 2015). The predominant winds were from the west, the south and the north-east, reinforcing the results presented in the meteorological map (Figure 11).

The trajectory frequency map during spring months (March-April) 2015 is shown in Figure 18. The prevailing winds during spring were from the north-east, the north-west and the south, which is in line with the previous results.

Anthropogenic sources of BC

We further focus on the anthropogenic part of BC emissions. For this purpose, a comparison of the levels of BC recorded in Algiers, Athens, Crete and Tamanrasset (the GAW/WMO reference station of Askrem) was performed.

Figure 19 displays the mean concentrations of BC recorded in Askrem, a background station situated at 23.27°N, 5.63°E and an altitude of 2730 m a.s.l, from April and May 1997. In contrast to the mean BC concentration measured in Algiers during April-May 2015, i.e., 0.936 µgm^{-3}, the BC mean concentration recorded at the Askrem GAW station was 0.168 µgm^{-3}. The BC concentrations in Algiers, which is heavily impacted by anthropogenic emissions, were 79.78% higher than at the Askrem background station.

Figure 20 shows a comparison of BC recorded in Algiers, Athens and Finokalia, Crete (Greece) during the year of measurement. The measurement site of Thissio (Athens), is situated in a city near the Akropolis, which is a much-visited museum. The Finokalia station is a background site used as a reference for Greek and European stations, 80 km east of Heraklion (Crete). The monthly average of BC in the urban agglomeration of Athens were similar to those recorded in Algiers, except in November, December and January, when the average concentrations were for Athens and Algiers respectively 2.949 and 1.278, 3.819 and 0.907 and 2.364 and 1.099 µgm^{-3}. The high BC concentrations in Athens during winter could be due to wood burning for heating. The monthly mean BC levels observed at the background station of Finokalia, Crete varied mostly between 0.2 and 0.5 µgm^{-3} with an annual average of 0.314 µgm^{-3}, which represents 20.36% and 28.16% of the annual averages recorded in Athens and Algiers, respectively.

BC modelling

We further compare the observed BC with modelled results. For this, we used the TM4-ECPL global model developed by

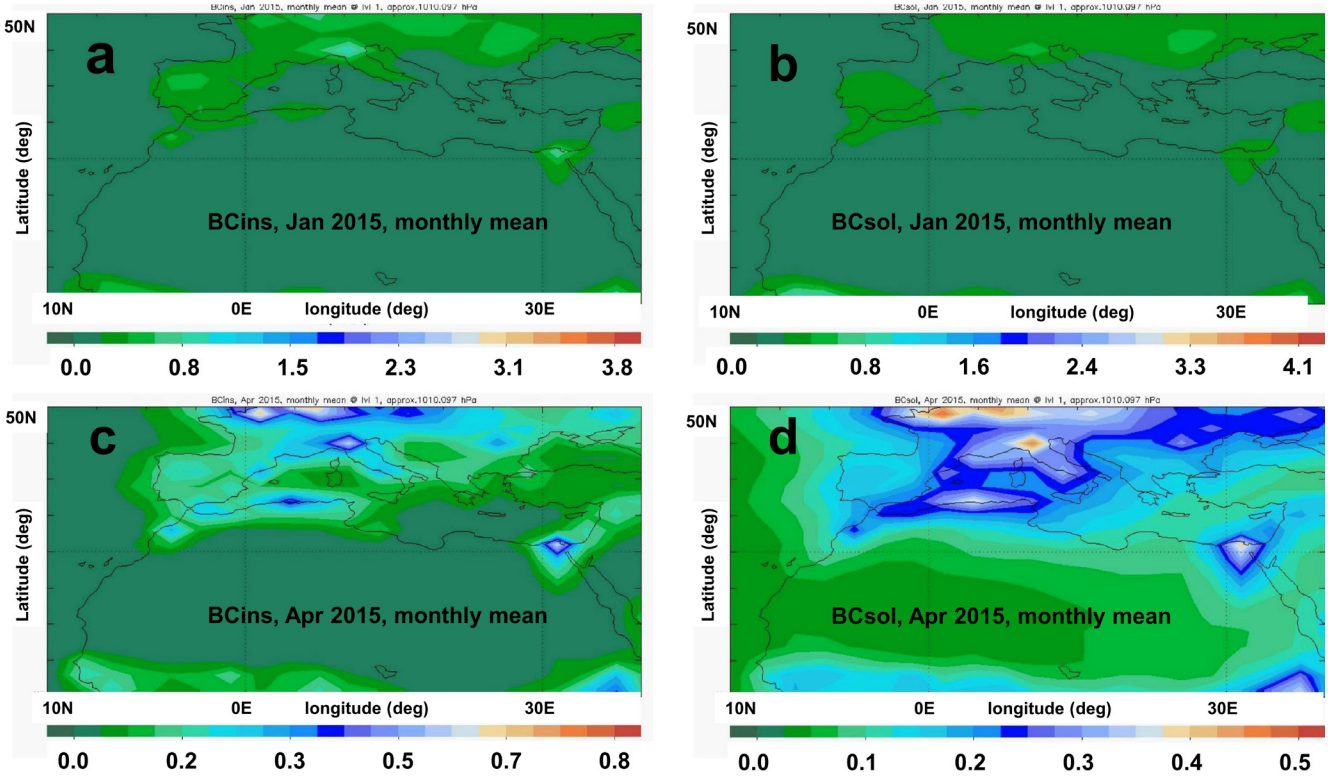

Figure 21: *Variation of BC_{ins} and BC_{sol} in January and April 2015 using TM4-ECPL global model.*

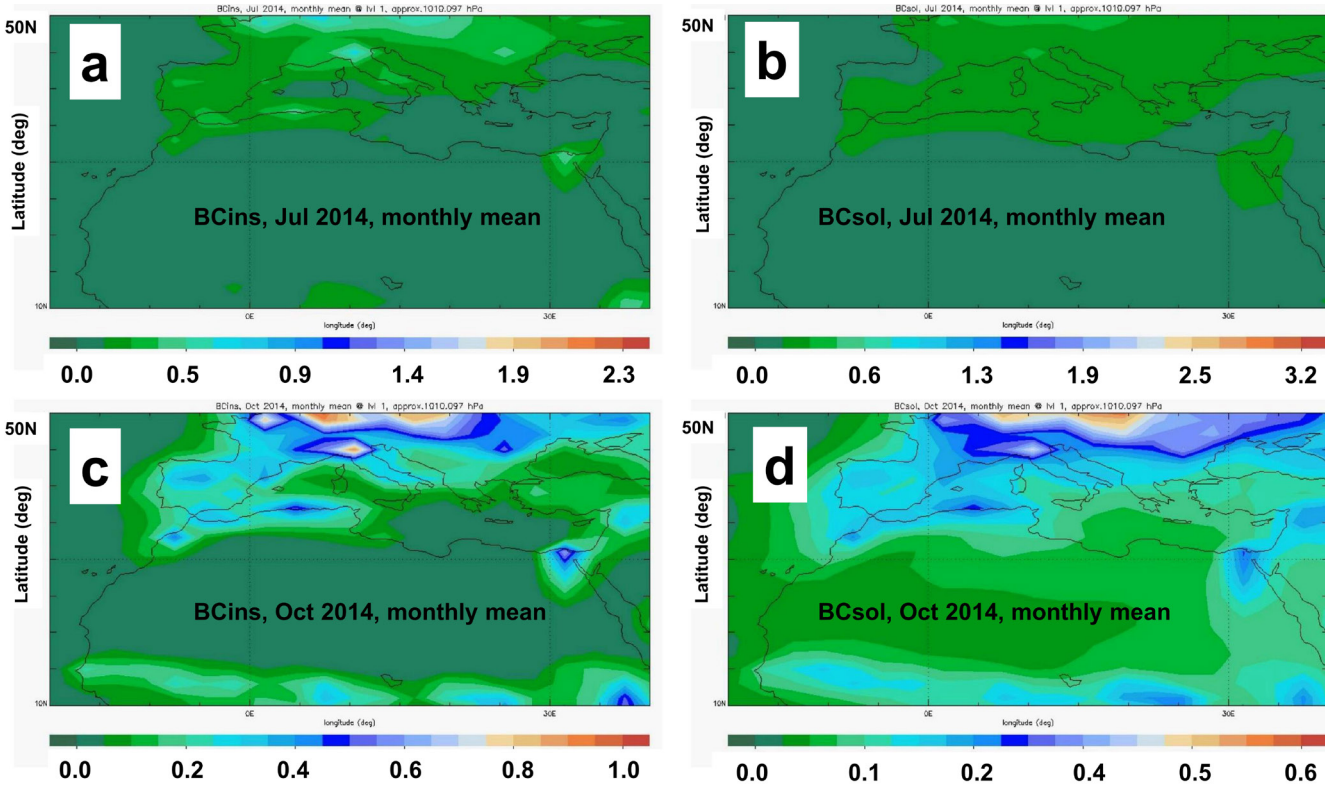

Figure 22: *Monthly average concentrations of BC_{ins} and BC_{sol} in July and October 2014 modelled with TM4-ECPL.*

the University of Crete (Greece). The model, which has been described in detail by Daskalakis et al. (2015), accounts for multiphase chemistry as well as all major aerosol types, including carbonaceous aerosols, both insoluble (BC_{ins}) and soluble (BC_{sol}) black carbon. The modelling of BC_{sol} and BC_{ins} is important to follow the atmospheric concentrations of fresh insoluble BC (BCins) and the aged soluble BC (BCsol) that can be transported to the soil and ground water by wet scavenging. The results of the modeling are depicted in Figures 21 and 22 for July and October 2014 and January and April 2015, respectively. The months were chosen for a representative comparison of measured and modeled results for each month in the middle of each season. Figures 21a, b, c and d present the modeled BC_{ins} in January 2015, BC_{sol} in January 2015, BC_{ins} in April 2015 and BC_{sol} in April 2015, respectively. Figures 22a, b, c and d depict the modeled BC_{ins} in July 2014, BC_{sol} in July 2014, BC_{ins} in October 2014 and BC_{sol} in October 2014, respectively. BC_{ins} and BC_{sol} were higher in the center of Africa and the north of Europe, due to forest fires and wood burning. The modelled BC_{ins} and BC_{sol} in Algiers were respectively 0.50 and 0.30 μgm^{-3} in July 2014, 0.50 and 0.40 μgm^{-3} in October 2014, 0.40 and 0.40 μgm^{-3} in January 2015 and 0.45 and 0.40 μgm^{-3} in April 2015. Therefore, the total modelled BC for the four months was 0.80, 0.90, 0.80 and 0.85 μgm^{-3}, respectively, which were very close to the mean measured BC concentration levels (0.745, 1.308, 1.108 and 0.917 μgm^{-3}).

In order to follow the variation of BC_{ff} and BC_{wb} by using the model proposed by Zotter et al. (2016), a statistical study has been performed. This statistical study was based on replacing the Angstrom exponents α_{ff}=1 and α_{wb}=2, used in the aethalometer model by α_{ff}=0.90 and α_{wb}=1.682 proposed by Zotter et al. (2016), followed by a calculation of the new seasonal BC, BC_{ff} and BC_{wb} average concentrations. A decrease of seasonal BC_{ff} of 9.15 and 7.08% in winter and autumn, respectively, was calculated using the Zotter et al. (2016) model, compared to an increase of BC_{wb} of 96.97 and 90.43% for spring and autumn, respectively. This result indicates that BC_{wb} was undervalued by the model applied in our study, in contrary to the Zotter et al. (2016) model especially in spring and autumn.

Angstrom exponent

Another important factor for the characterization of the source of measured BC during the year of study is the Angstrom exponent (α). Favez et al., (2009) revealed a low spectral dependence of black carbon light absorption (α ~ 1), in contrast, it is much higher for other aerosol components, i.e. hematite and brown carbon. Sandradewi et al. (2008) recorded light absorption exponents of 1.1 for traffic and 1.8–1.9 for wood burning, calculated from the light absorption at 470 and 950 nm. Soni et al. (2011) observed minimum values of the Angstrom exponent during May and maximum values during the winter period (December and January), related to a gradual decrease in the coarse particle concentration from summer to winter months. The Angstrom exponent (calculated at 470 and 950nm) shown in Figure 23 varied mostly between 0.5 and 1.5 with an annual

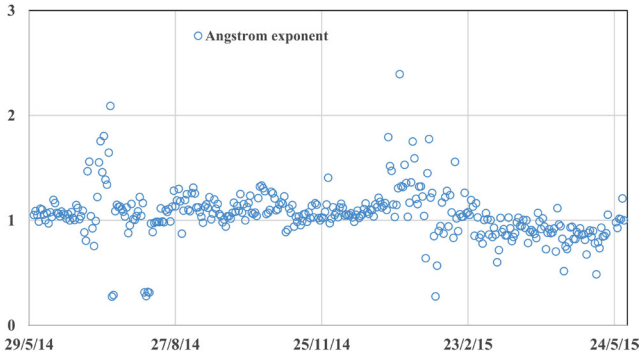

Figure 23: *Daily variation of Angstrom exponent during the year of measurement at Algiers site.*

average of 1.07, indicating that the main source of BC during the year of measurement was from fossil fuel burning, in line with the findings presented in this investigation.

Mass absorption cross-section

The mass absorption cross-section (MAC) is also a good indicator of the source and the aging of BC particles. The MAC values are related to the aerosol mixing state, size, and morphology (Bond and Bergstrom, 2006), and increase with coating thickness or water at high relative humidity (Schnaiter, 2005). Laborde et al. (2013) revealed that the MAC average was ~7.3 m^2g^{-1} for traffic emissions due to refractory BC (rBC) cores wich are mainly uncoated and small. However, for the case of wood burning, the MAC average was ~7.8 m^2g^{-1} explained by thicker coating and bigger rBC core size. Higher average MAC (~8.8 m^2g^{-1}) were recorded with aged aerosols because of the thicker coating compared to aerosols from traffic and wood burning (Schnaiter, 2005). Cao et al. (2015) reported median MAC values of 20.0, 33.7, 29.1 and 27.6 m^2g^{-1} during the spring, summer, autumn and winter, respectively, in China. However, the seasonal means of MAC values recorded in Switzerland were much smaller with 8.9, 9.5, 10.9, and 9.9 m^2g^{-1} in spring, summer, autumn and winter, respectively (Lavanchy et al., 1999). The high MAC values recorded in China could be due to biomass burning, relatively large increase in the symmetrical particles and cluster-like structures emitted by motor vehicles, and the secondary and aged aerosols under high relative humidity (60–80%) and strong solar radiation.

In the present study, the seasonal means of MAC during summer, autumn, winter and spring were 10.97, 15.56, 30.27 and 17.12 m^2g^{-1}, respectively. These high MAC values could be due to aged aerosols coming from Europe and secondary aerosols under high humidity and strong solar radiation, considering that the seasonal relative humidity was 58.38, 61.78, 64.12 and 62.03% in summer, autumn, winter and spring, respectively. The highest monthly averages for MAC were recorded in February, December and March at 36.18, 30.54 and 26.92 m^2g^{-1}, respectively, when, relative humidity was high, reaching 63.94, 65.44 and 65.44%, respectively. The lowest MACs were recorded in July, September and October at 9.61, 1.75 and 11.88 m^2g^{-1}, respectively, with

lower relative humidity of 58.00, 58.77 and 55.53%, respectively. This difference between the monthly variations in MAC could also be due to the clear skies in February and March especially, in contrast to the other months, and the wind in winter months bringing aged aerosols. July and September were also characterized by fresh aerosols due to wild fires as presented in the previous sections.

Conclusions

Observations of BC, BC_{ff} and BC_{wb} over one year of measurement were performed for the first time in Algeria in order to better understand the levels, the occurrence, the sources and the seasonal modulations of BC air pollution. Hourly, diurnal, and seasonal variations in BC levels have been investigated. BC sources were found to be associated with traffic, wild fires, and the oil industry in the south of Algeria. Celebration events and long-range transport of pollution from Europe and neighboring countries have also been investigated.

The main source of BC pollution in Algiers city was found to be fossil fuel combustion accounting for 95.60% of the total annual mean BC levels, whereas BC_{wb} contributed only 4.40%.

The highest seasonal mean concentrations were recorded in summer and autumn at 1.283 and 1.209 μgm^{-3} for BC and 1.217 and 1.177 μgm^{-3} for BC_{ff}, respectively. The lowest mean concentrations were recorded in winter and spring at 1.023 and 0.966 μgm^{-3} for BC and 0.933 and 0.956 μgm^{-3} for BC_{ff}, respectively.

For BC_{wb}, the highest mean concentrations were reached in winter and summer at 0.090 and 0.066 μgm^{-3}, respectively, due to the forest fires and long-range transport of air pollution from Europe. The lowest mean concentrations of BC_{wb} were recorded in autumn and spring at 0.032 and 0.010 μgm^{-3}, respectively.

The BC pollution predominantly came from the west of Algiers, probably associated with the petroleum refinery in Arzew (Oran) situated 400 km from Algiers.

A source apportionment study of BC has been carried out for the wet and dry period of the studied year, followed by back trajectory cluster analysis for a better understanding of the long-range transport pathways. The highest BC mean concentrations of 1.333 μgm^{-3} in the wet period were recorded in association with southerly winds, followed by local pollution contributing 1.238 μgm^{-3} of BC. During the dry period, the highest mean concentrations of BC were observed in association with airflow from the East and West directions at concentrations of 1.380 and 1.377 μgm^{-3} respectively.

The mass absorption cross-section (MAC) and Angstrom exponent were used to investigate the source and the ageing of BC measured in the present study.

The present study allowed a comparison of BC mean concentrations recorded in Algiers, Crete and Athens (Greece), and Tamanrasset (GAW/WMO reference station of Askrem), revealing that the anthropogenic emissions were 79.78, 90.26 and 46.50% higher than at the Askrem background station for Algiers, Athens and Crete, respectively.

The annual average BC concentration recorded at the Algiers Observatory (suburban site) was much lower than that recorded in Anantapur (India), Prague (Czech), Athens (Greece) and Rome (Italy), but was higher than values measured in Santa Cruz de Tenerife (Spain), Crete (Greece) and Finland.

Acknowledgements

We gratefully acknowledge the NOAA Air Resources Laboratory (ARL) for the provision of the HYSPLIT transport and dispersion model and READY website (http://www.ready.noaa.gov) used in this publication.

The authors are thankful to the National Meteorological Office, Algeria especially to Mr Mimouni Mohamed, for providing the Askrem data.

We thank also Dr Pavlos Zarmpas, Dr Georgos Kouvarakis, Georgos Fanourgakis and Dimitrios Amanatidis at the University of Crete (Greece) for their assistances with the modelling.

References

Ali, U., Syed, J.H., Junwen, L., Sánchez-García, L., Malik, R.N., Chaudhry, M.J.I., Arshad, M., Li, J., Zhang, G., Jones, K.C. 2014, 'Assessing the relationship and influence of black carbon on distribution status of organochlorines in the coastal sediments from Pakistan', *Environmental Pollution* 190: 82–90. https://doi.org/10.1016/j.envpol.2014.03.024.

Andreae, M.O., Gelencser, A. 2006, 'Black carbon or brown carbon? The nature of light-absorbing carbonaceous aerosols', *Atmospheric Chemistry and Physics* 6:3131-3148. https://doi.org/10.5194/acpd-6-3419-2006.

Bahadur, R., Russell, L.M., Jacobson, M.Z., Prather, K., Nenes, A., Adams, P. and Seinfeld, J.H. 2012, 'Importance of composition and hygroscopicity of BC particles to the effect of BC mitigation on cloud properties: Application to California conditions: Effect of CV particles on clouds', *Journal of Geophysical Research: Atmospheres* 117(D9). https://doi.org/10.1029/2011JD017265.

Begam, G.R., Vachaspati, C.V., Ahammed, Y.N., Kumar, K.R., Babu, S.S. and Reddy, R.R. 2016, 'Measurement and analysis of black carbon aerosols over a tropical semi-arid station in Kadapa, India', *Atmospheric Research*, 171:77–91. https://doi.org/10.1016/j.atmosres.2015.12.014.

Bond, T.C. and Bergstrom, R.W. 2006, 'Light Absorption by Carbonaceous Particles: An Investigative Review', *Aerosol Science and Technology* 40(1):27–67. https://doi.org/10.1080/02786820500421521.

Bond, T. C., Doherty, S. J., Fahey, D. W., Forster, P. M., Berntsen, T., DeAngelo, B. J., Flanner, M. G., Ghan, S., Kärcher, B., Koch, D., Kinne, S., Kondo, Y., Quinn, P. K., Sarofim, M. C., Schultz, M. G., Schulz, M., Venkataraman, C., Zhang, H., Zhang, S., Bellouin, N., Guttikunda, S. K., Hopke, P. K., Jacobson, M. Z., Kaiser, J. W., Klimont, Z., Lohmann, U., Schwarz, J. P., Shindell, D., Storelvmo, T., Warren, S. G. and Zender, C. S. 2013, 'Bounding the role of black carbon in the climate system: A scientific assessment', *Journal of Geophysical Research* 118(11): 5380–5552. https://doi.org/10.1002/jgrd.50171.

Byčenkienė, S., Ulevicius, V. and Kecorius, S. 2011, 'Characteristics of black carbon aerosol mass concentration over the East Baltic region from two-year measurements', *Journal of Environmental Monitoring* 13(4):1027. https://doi.org/10.1039/c0em00480d.

Cao, J., Zhu, C., Ho, K., Han, Y., Shen, Z., Zhan, C. and Zhang, J. 2015, 'Light attenuation cross-section of black carbon in an urban atmosphere in northern China', *Particuology* 18:89–95. https://doi.org/10.1016/j.partic.2014.04.011

Cheng, Y., Lee, S.C., Ho, K.F., Wang, Y.Q., Cao, J.J., Chow, J.C. and Watson, J.G. 2006, 'Black carbon measurement in a coastal area of south China', *Journal of Geophysical Research* 111(D12). https://doi.org/10.1029/2005JD006663.

Cheng, Y.-H., Liao, C.-W., Liu, Z.-S., Tsai, C.-J. and Hsi, H.C. 2014, 'A size-segregation method for monitoring the diurnal characteristics of atmospheric black carbon size distribution at urban traffic sites', *Atmospheric Environment* 90:78–86. https://doi.org/10.1016/j.atmosenv.2014.03.023.

Chiloane, K. E., Beukes, J. P., van Zyl, P. G., Maritz, P., Vakkari, V., Josipovic, M., Venter, A. D., Jaars, K., Tiitta, P., Kulmala, M., Wiedensohler, A., Liousse, C., Mkhatshwa, G. V., Ramandh, A. and Laakso, L. 2017, Spatial, temporal and source contribution assessments of black carbon over the northern interior of South Africa, *Atmos. Chem. Phys*. 17(10):6177–6196. https://doi.org/10.5194/acp-17-6177-2017.

Daskalakis, N., Myriokefalitakis, S. and Kanakidou, M. 2015, 'Sensitivity of tropospheric loads and lifetimes of short lived pollutants to fire emissions', *Atmospheric Chemistry and Physics* 15(6):3543–3563. https://doi.org/10.5194/acp-15-3543-2015.

De Miranda, R.M., de Fatima Andrade, M., Fornaro, A., Astolfo, R., de Andre, P.A. and Saldiva, P. 2012, 'Urban air pollution: a representative survey of $PM_{2.5}$ mass concentrations in six Brazilian cities', *Air Quality, Atmosphere & Health* 5(1):63–77. https://doi.org/10.1007/s11869-010-0124-1.

Favez, O., Cachier, H., Sciare, J., Sarda-Estève, R., Martinon, L. 2009, 'Evidence for a significant contribution of wood burning aerosols to $PM_{2.5}$ during the winter season in Paris, France', *Atmospheric Environment* 43:3640–3644. https://doi.org/10.1016/j.atmosenv.2009.04.035.

Gadhavi, H. and Jayaraman, A. 2010, 'Absorbing aerosols: contribution of biomass burning and implications for radiative forcing', *Annales Geophysicae* 28(1):103–111. https://doi.org/10.5194/angeo-28-103-2010.

Hooda, R. K., Kivekäs, N., O'Connor, E. J., Collaud Coen, M., Pietikäinen, J.-P., Vakkari, V., Backman, J., Henriksson, S. V., Asmi, E., Komppula, M., Korhonen, H., Hyvärinen, A.-P. and Lihavainen, H. 2018, 'Driving factors of aerosol properties over the foothills of Central Himalayas based on 8.5 years continuous measurements, *J. Geophys. Res. Atmos*. 123(23):13421–13442. https://doi.org/10.1029/2018JD029744.

Jennings, S.G., Spain, T.G., Doddridge, B.G., Maring, H., Kelly, B.P. and Hansen, A.D.A. 1996, 'Concurrent measurements of black carbon aerosol and carbon monoxide at Mace Head', *Journal of Geophysical Research: Atmospheres* 101(D14):19447–19454. https://doi.org/10.1029/96JD00614.

Jose, S., Niranjan, K., Gharai, B., Rao, P.V.N. and Nair, V.S. 2016, 'Characterisation of absorbing aerosols using ground and satellite data at an urban location, Hyderabad', *Aerosol and Air Quality Research* 16(6): 1427–1440. https://doi.org/10.4209/aaqr.2014.09.0220.

Krecl, P., Targino, A.C. and Johansson, C. 2011, 'Spatiotemporal distribution of light-absorbing carbon and its relationship to other atmospheric pollutants in Stockholm', *Atmospheric Chemistry and Physics* 11(22):11553–11567. https://doi.org/10.5194/acp-11-11553-2011.

Laborde, M., Crippa, M., Tritscher, T., Jurányi, Z., Decarlo, P.F., Temime-Roussel, B., Marchand, N., Eckhardt, S., Stohl, A., Baltensperger, U., Prévôt, A.S.H., Weingartner, E., Gysel, M. 2013, 'Black carbon physical properties and mixing state in the European megacity Paris', *Atmospheric Chemistry and Physics* 13(11):5831–5856. https://doi.org/10.5194/acp-13-5831-2013.

Lavanchy, V.M.H., Gäggeler, H.W., Nyeki, S. and Baltensperger, U. 1999, 'Elemental carbon (EC) and black carbon (BC) measurements with a thermal method and an aethalometer at the high-alpine research station Jungfraujoch', *Atmospheric Environment* 33(17):2759–2769. https://doi.org/10.1016/S1352-2310(98)00328-8.

Lohmann, R., MacFarlane, J.K. and Gschwend, P.M. 2005, 'Importance of black carbon to sorption of native PAHs, PCBs, and PCDDs in Boston and New York harbor sediments', *Environmental Science & Technology* 39(1):141–148. https://doi.org/10.1021/es049424+.

Mahmoud, K.F., Alfaro, S.C., Favez, O., Abdel Wahab, M.M. and Sciare, J. 2008, 'Origin of black carbon concentration peaks in Cairo (Egypt)', *Atmospheric Research* 89(1–2):161–169. https://doi.org/10.1016/j.atmosres.2008.01.004.

Masiello, C.A. and Druffel, E.R.M. 1998, 'Black carbon in deep-sea sediments', *Science*, New Series 280(5371):1911–1913. https://doi.org/10.1126/science.280.5371.1911.

Nair, V.S., Moorthy, K.K., Alappattu, D.P., Kunhikrishnan, P.K., George, S., Nair, P.R., Babu, S.S., et al. 2007, 'Wintertime aerosol characteristics over the Indo-Gangetic Plain (IGP): Impacts of local boundary layer processes and long-range transport: Winter aerosols over Indo-Gangetic Plain', *Journal of Geophysical Research: Atmospheres* 112(D13). https://doi.org/10.1029/2006JD008099.

Pu, W., Wang, X., Zhang, X., Ren, Y., Shi, J.-S., Bi, J.-R., Zhang, B.-D. 2015, ' Size distribution and optical properties of particulate matter (PM_{10}) and black carbon (BC) during dust storms and local air pollution events across a loess plateau site', *Aerosol and Air Quality Research*, 15(6): 2212-2224. https://doi.org/10.4209/aaqr.2015.02.0109.

Ramachandran, S. and Rajesh, T.A. 2007, 'Black carbon aerosol mass concentrations over Ahmedabad, an urban location in western India: Comparison with urban sites in Asia, Europe, Canada, and the United States', *Journal of Geophysical Research* 112(D6). https://doi.org/10.1029/2006JD007488.

Reche, C., Querol, X., Alastuey, A., Viana, M., Pey, J., Moreno, T., Rodríguez, S., et al. 2011, 'New considerations for PM, Black Carbon and particle number concentration for air quality monitoring across different European cities', *Atmospheric Chemistry and Physics* 11(13):6207–6227. https://doi.org/10.5194/acp-11-6207-2011.

Reddy, B.S.K., Kumar, K.R., Balakrishnaiah, G., Gopal, K.R., Reddy, R.R., Reddy, L.S.S., Ahammed, Y.N., Narasimhulu, K., Moorthy, K.K., Babu, S.S. 2012, 'Potential source regions contributing to seasonal variations of black carbon aerosols over Anantapur in Southeast India', *Aerosol and Air Quality Research* 12(3):344–358. https://doi.org/10.4209/aaqr.2011.10.0159.

Saarikoski, S., Timonen, H., Saarnio, K., Aurela, M., Järvi, L., Keronen, P., Kerminen, V.-M., Hillamo, R. 2008, 'Sources of organic carbon in fine particulate matter in northern European urban air', *Atmospheric Chemistry and Physics* 8(20):6281–6295. https://doi.org/10.5194/acp-8-6281-2008.

Saha, A. and Despiau, S. 2009, 'Seasonal and diurnal variations of black carbon aerosols over a Mediterranean coastal zone', *Atmospheric Research* 92(1):27–41. https://doi.org/10.1016/j.atmosres.2008.07.007.

Salcedo, D., Onasch, T.B., Dzepina, K., Canagaratna, M.R., Zhang, Q., Huffman, J.A., DeCarlo, P.F., Jayne, J.T., Mortimer, P., Worsnop, D.R., Kolb, C.E., Johnson, K.S., Zuberi, B., Marr, L.C., Volkamer, R., Molina, L.T., Molina, M.J., Cardenas, B., Bernabé, R.M., Márquez, C., Gaffney, J.S., Marley, N.A., Laskin, A., Shutthanandan, V., Xie, Y., Brune, W., Lesher, R., Shirley, T., Jimenez, J.L. 2006, "Characterization of ambient aerosols in Mexico City during the MCMA-2003 campaign with Aerosol Mass Spectrometry: results from the CENICA Supersite", *Atmospheric Chemistry and Physics* 6(4):925–946. https://doi.org/10.5194/acp-6-925-2006.

Sandradewi, J., Pre´vo^t, A.S.H., Szidat, S., Perron, N., Alfarra, M.R., Lanz, V.A., Weingartner, E., Baltensperger, U., 2008b. Using aerosol light absorption measurements for the quantitative determination of wood burning and traffic emission contributions to particulate matter. *Environmental Science and Technology* 42:3316–3323. https://doi.org/10.1021/es702253m.

Schnaiter, M. 2005, 'Absorption amplification of black carbon internally mixed with secondary organic aerosol', *Journal of Geophysical Research*, 110(D19). https://doi.org/10.1029/2005JD006046.

Sciare, J., d'Argouges, O., Sarda-Estève, R., Gaimoz, C., Dolgorouky, C., Bonnaire, N., Favez, O., Bonsang, B., Gros, V. 2011, 'Large contribution of water-insoluble secondary organic aerosols in the region of Paris (France) during wintertime: Wintertime water-insoluble SOA', *Journal of Geophysical Research: Atmospheres* 116(D22). https://doi.org/10.1029/2011JD015756.

Seidel, D.J. and Birnbaum, A.N. 2015, 'Effects of Independence Day fireworks on atmospheric concentrations of fine particulate matter in the United States', *Atmospheric Environment*, 115:192–198. https://doi.org/10.1016/j.atmosenv.2015.05.065.

Seinfeld, J.H. and Pandis, S.N. 2006, 'Atmospheric Chemistry and Physics: From Air Pollution to Climate Change, 2. ed.', Hoboken, NJ.

Singla, V., Mukherjee, S., Kashikar, A.S., Safai, P.D. and Pandithurai, G. 2019, 'Black carbon: source apportionment and its implications on CCN activity over a rural region in Western Ghats, India', *Environmental Science and Pollution Research*, 26(7):7071-7081. https://doi.org/10.1007/s11356-019-04162-w

Streets, D.G., Gupta, S., Waldhoff, S.T., Wang, M.Q., Bond, T.C. and Yiyun, B. 2001, 'Black carbon emissions in China', *Atmospheric Environment* 35(25): 4281-4296. https://doi.org/10.1016/S1352-2310(01)00179-0.

Targino, A.C. and Krecl, P. 2017, 'Local and Regional Contributions to Black Carbon Aerosols in a Mid-Sized City in Southern Brazil', *Aerosol and Air Quality Research* 16(1):125–137. https://doi.org/10.4209/aaqr.2015.06.0388.

Venkatachari, P., Zhou, L., Hopke, P.K., Felton, D., Rattigan, O.V., Schwab, J.J. and Demerjian, K.L. 2006, 'Spatial and temporal variability of black carbon in New York City: SPATIAL AND TEMPORAL VARIATIONS OF BC', *Journal of Geophysical Research: Atmospheres* 111(D10). https://doi.org/10.1029/2005JD006314.

Vodička, P., Schwarz, J. and Ždímal, V. 2013, 'Analysis of one year's OC/EC data at a Prague suburban site with 2-h time resolution', *Atmospheric Environment* 77:865–872. https://doi.org/10.1016/j.atmosenv.2013.06.013.

Wang, H., Nie, L., Liu, D., Gao, M., Wang, M. and Hao, Z. 2015, 'Physico-chemical characterization and source tracking of black carbon at a suburban site in Beijing', *Journal of Environmental Sciences* 33:188–194. https://doi.org/10.1016/j.jes.2015.05.001.

Weingartner, E., et al. (2003), Absorption of Light by Soot Particles: Determination of the Absorption Coefficient by Means of Aethalometers *Journal of Aerosol Science* 34:1445-1463. https://doi.org/10.1016/S0021-8502(03)00359-8.

Wu, D., Wu, C., Liao, B., Chen, H., Wu, M., Li, F., Tan, H., Deng, T., Li, H., Jiang, D., Yu, J.Z. 2013, 'Black carbon over the South China Sea and in various continental locations in South China', *Atmospheric Chemistry and Physics* 13(24):12257–12270. https://doi.org/10.5194/acp-13-12257-2013.

Zotter, P., Herich, H., Gysel, M., El-Haddad, I., Zhang, Y., Močnik, G., Hüglin, C., Baltensperger, U., Szidat, S., Prévôt, A.S.H. 2017, 'Evaluation of the absorption Ångström exponents for traffic and wood burning in the Aethalometer based source apportionment using radiocarbon measurements of ambient aerosol', *Atmospheric Chemistry and Physics* 17:4229-4249. https://doi.org/10.5194/acp-2016-621.

Research article

Aerosol characterisation including oxidative potential as a proxy of health impact: a case of a residential site in a highly industrialised area

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Received: 4 February 2019 - **Reviewed:** 20 March 2019 - **Accepted:** 25 August 2019
https://doi.org/10.17159/caj/2019/29/2.7517

Abstract

This study aimed to characterise aerosols sampled in the vicinity of a major industrialised area, i.e. the Vaal Triangle. It included the determination of oxidative potential as a predictive indicator of particle toxicity. Aerosol samples were collated through the cascade filtering during an eight-month period (12 h over three days in one week). Three size fractions were analysed for organic carbon (OC), black carbon (BC) and oxidative potential (OP), while ionic content was presented as monthly and seasonal concentrations. The continuous measurement of black carbon by an optical attenuation instrument was collated concurrently with cascade filtering. The carbonaceous content was low compared to the ionic one. Within the carbonaceous concentrations, the organic carbon was higher than concentrations of black carbon in both seasons in the ultra-fine fraction; the opposite was the case for the fine fraction, while the coarse fraction concentrations of organic carbon in the dry season had higher concentrations than black carbon in the wet season and organic carbon in the wet season. The OP tended to increase as the size was decreasing for wet season aerosols, whereas, for the dry season, the highest OP was exerted by the fine fraction. The ultrafine fraction was the one showing the most contrasting OP between the two seasons. Continuous monitoring indicated that the higher BC concentrations were recorded in the dry/winter part of the year, with the daily pattern of concentrations being typically bimodal, having both the morning and evening peaks in both seasons. Within the ionic content, the dominance of sulphate, nitrate and ammonium was evident. Multiple linear correlations were performed between all determined compounds. Strong correlations of carboxylic acids with other organic compounds were revealed. These acids point to emissions of VOC, both anthropogenic and biogenic. Since they were equally present in both seasons, a mixture of sources was responsible, both present in the wider area and throughout the year.

Keywords

carbon, ions, acellular assays, cascade impactors, aethalometer

Introduction

Context and problem

Pollution is now recognised to be a huge environmental threat, with air pollution being a leading cause of premature human death (Landrigan et al., 2018). The different air pollutants, their doses and time of exposure and in particular the pollutant mixtures to which humans are usually exposed lead to adverse impacts on human (and similarly to animal) health that range from acute to chronic effects. Epidemiological studies and animal model data state that primarily affected systems are the cardiovascular and the respiratory system (Kampa and Castanas, 2007). In terms of the health effects of the particulate matter (PM), there has been substantial progress in the evaluation of

PM health effects at different time-scales of exposure and in the exploration of the concentration-response function (Pope and Dockery, 2006).

Inhalation toxicology research has demonstrated that the generation of reactive oxygen species (ROS) and the associated induction of oxidative stress in target cells may represent a key pathway by which ambient fine and ultrafine (sub-micrometre) size particles cause adverse health effects. Particle surface reactivity, metals and redox cycling organic compounds are properties shown to be involved in ROS generation.

Air quality (AQ) regulations in most countries rely on PM volumetric mass without taking into consideration PM composition. Since PM is a complex mixture including metallic, inorganic and organic compounds, with differing proportions depending on the sources of emissions and ambient atmospheric conditions, alternative metrics such as the oxidative potential (OP) can and have been proposed to predict PM toxicity (Boogaard et al., 2012).

Oxidative properties of aerosol are considered to be important attributes to explain many of the aerosol biological/health effects (Borm et al., 2007). As such, aerosol OP constitutes a unifying factor explaining its pathological activity and has been considered as an additional metric to determine aerosol health impact (Donaldson et al., 1996; Borm et al., 2007). Li et al. (2003) indicated that the ultra-fine mode of ambient aerosols collected from Los Angeles (USA) had a higher intrinsic oxidative capacity than the fine and coarse aerosol modes.

To date, not many health impact studies have been undertaken in Africa. The POLCA project ("POLlution des Capitales Africaines") in western Africa was a pioneering study that addressed the physicochemical characterisation of aerosol pollution in two west African capitals (Bamako, Mali and Dakar, Senegal). These cities experience high population growth. They are impacted by a variety of emission sources, dominated by traffic and domestic burning, and strongly subjected to contrasted meteorological conditions (Doumbia et al., 2012).

The sampling campaigns in these West African capitals were performed during the dry season, and the influence of the Saharan dust events and biomass burning was recognised in addition to prevailing anthropological pollution sources, mainly traffic and domestic burning (Assamoi and Liousse, 2010). Val et al. (2013) demonstrated that the finest size fractions of these aerosols induced a pro-inflammatory response in human bronchial cells, indicating that aerosol pollution in West African cities may have a strong impact on its population health. These cities have high ambient aerosol concentrations originating from anthropogenic sources known to contribute to health effects (Doumbia et al., 2012; Liousse and Galy-Lacaux, 2010; Cassee et al., 2013).

South Africa has one of the largest economies in Africa and remains the largest industrialised regional energy producer. South Africa is on a continuing trend of increasing fossil fuel consumption and demand for electricity. Most of the electricity in South Africa is produced by coal-fired power stations. A substantial fraction of liquid fuel is also distilled from coal as well as from natural gas. The main axes of South Africa's economy remain the large mining and associated metallurgical industries (e.g. Beukes et al., 2010). Domestic biomass and fossil fuel combustion for space heating and cooking is also widely practised, especially in informal settlements that are found around most towns and cities (e.g. Kimemia et al., 2010; Vakkari et al., 2013; Butt et al., 2016; Makonese et al., 2016).

All of the afore-mentioned has led to increased environmental concerns with atmospheric pollution being a major worry. New industrial installations in South Africa are being equipped with cleaner technologies. However, emissions of sulphur dioxide (SO_2), oxides of nitrogen (NO_x), black carbon (BC) and carbon dioxide (CO_2) are predicted to increase.

These emissions, combined with a potential change in biomass burning due to global warming, and the associated dryer climate in certain parts of southern Africa, can significantly influence the regional and global climate (Boko et al., 2007). Climate change may also enhance migration to already densely populated urban areas, and potentially increase environmental concerns.

Overall, the atmospheric PM in this region originates from a mixture of natural and anthropogenic emission sources (such as industry, domestic burning, biomass burning, transport), and is spatially and temporally variable (Tiitta et al., 2014). On the southern African sub-continent, number concentrations and optical properties of sub-micrometre aerosol particles have been investigated intensively during the SAFARI 1992 and 2000 measurement campaigns (Swap et al., 2003; Ross et al., 2003; Eck et al., 2003). Recently, more studies have been published on the aerosol measurements regionally giving more insight into the sub-micrometre number-size distribution, formation of secondary particles, and trends and loadings of non-refractive species (Hirsikko et al., 2012; Vakkari et al., 2013; Tiitta et al., 2014; Sundström et al., 2015). The aerosol organic compounds were studied (Booyens et al., 2015), BC modelled results were compared with the regional BC (continuous) measurements (Kuik et al., 2015), and impacts of aerosols from residential activities were also modelled (Butt et al., 2016).

This study focused on characterising aerosol pollution impacting a residential suburb amid a major South African industrialised area, i.e. the Vaal Triangle. In addition to the carbonaceous and ionic content, the oxidative potential of the aerosols was determined to provide a predictive indicator of their toxicity.

The particles from 0.25 to over 2.5 µm in aerodynamic diameters were collected through three cascade impactor devices running in parallel at the Vaal Park Primary School, in Sasolburg. They were sampled for 12 hours in each of three sampling days during the last week of every month from March 2012 to October 2012 to encompass the three local weather seasons: entire autumn, entire winter and early to mid-spring. The thermal analytical method was used to determine the black carbon (BC) and organic

carbon (OC) content (Cachier et al., 1989), whereas (water soluble) ionic compounds were determined by ion chromatography and inductively coupled-plasma-mass spectroscopy. Three different acellular assays were applied to characterise the oxidative potential. A determination of trace metals content was out of the scope for this study.

Site selection

The study area is colloquially called the Vaal Triangle, after the three towns forming each of its three spatial angles (Sasolburg, Vanderbijlpark and Vereeniging). The Vaal Triangle area is where a large part of the South African petrochemical and other chemical industries is located. In addition, other large point sources, including a coal-fired power station (without SO_x and NO_x scrubbing) and several large (iron) metallurgical smelters, are also within this area. This area, together with the southern section of Gauteng was proclaimed a national air pollution area for intervention in terms of the South African National Environmental Management: Air Quality Act (Government Gazette Republic of South Africa, 2007) termed the "Vaal Triangle Priority Area" (constituting effectively more intensely monitored air-shed air quality (AQ) management area). The Vaal Triangle is also home to a number of formal (towns and townships) and informal settlements (slums), which mainly use coal, paraffin and wood as fuel sources. This, in turn, impacts directly on the health and well-being of the people residing in these communities. Other sources of concern contributing to the pollution emission mixtures within the area include transport-related emissions, biomass burning (domestic/anthropogenic and wild/natural), water treatment works and landfill areas, agricultural activities and various other fugitive sources. This is a strong indication that air quality in this area often exceeds or is close to exceeding National Ambient Air Quality Standards for criteria pollutants.

The study sampling site was set at the grounds of a primary school in the target area (Vaalpark Primary School). The sampling site's latitude is 26˚46' 2" south, and the longitude is 27˚51' 12" east (Figure 1). The sampling site was chosen for its proximity to the Sasolburg industrial complex. This was done to determine the level of aerosol pollution possibly received by the population working, visiting and residing in close proximity to the major petrochemical complex (Sasol Industries) and metallurgical industries (in Vanderbijlpark and Vereeniging municipalities) (Figure 1).

Regional weather and climate

Geo-morphologically, the sampling site is located on the South African Highveld (an elevated plateau of 1400 m a.s.l. to 1700 m a.s.l.). Laakso et al. (2012) recently gave a concise description of the weather conditions over the South African Highveld. The description in this paper is, therefore, limited.

Over the South African Highveld, literally all precipitation falls during the wet season (from mid-October to end of April), with almost no precipitation during the dry season (May to mid-October). The exact duration of the dry and wet seasons can differ between years, especially if the onset of rains is late. This distinct

Figure 1: *Location of the Vaalpark Primary School sampling site within the Vaal Triangle area (Source: Google Earth with own markings).*

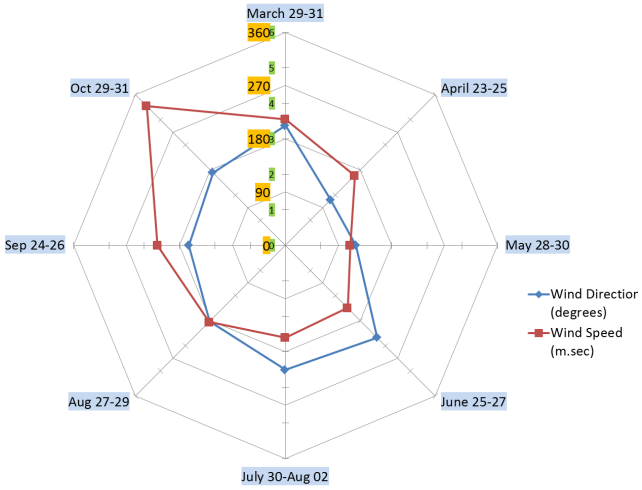

Figure 2: *The wind speeds and directions near the site over the sampling periods (source: SASOL wind data).*

precipitation cycle strongly affects pollutant concentrations, i.e. enhanced wet scavenging (wet deposition/wash-out) of pollutants during the wet season, while pollution levels are increased during the dry season through the occurrence of large-scale biomass burning and wind-blown dust emissions.

An important attribute of the regional climate is protracted periods of cold mainly night temperatures with often sub-zero temperatures, which contribute to lower relative humidity in the ambient air as well as drying moisture content from vegetation. The lower temperatures also coincide with increased use of coal, paraffin and wood as fuels in low-income residential areas (formal and informal). This contributes to the overall ambient pollution levels and subsequently to the increase in aerosol concentrations. During the winter and early spring months, multiple inversion layers form regularly at various altitudes. Such layered atmospheric structure significantly reduces vertical mixing, impacting significantly on the residence time of atmospheric pollutant concentrations. The region is also dominated by anti-cyclonic circulation, especially during the

winter months (June-Aug). Such re-circulation can trap pollutants for a number of weeks (Garstang et al., 1996; Tyson and Preston-Whyte, 2000; Freiman and Piketh, 2002, Mafusire et al., 2016).

The more frequent and highest wind speeds are observed during the end of the dry season (August, September and part of October), towards the beginning of the wet season (mid- to end-October) (Figure 2).

During the wet season, besides higher precipitation, higher temperatures and relative humidity with frontal and convective air mass movements enable horizontal and vertical mixing of the atmosphere over the area, and across the entire region, allowing better dispersion and dilution of pollutants.

Methods

Aerosol filter (impactor) sampling

The filter collection of aerosol was performed by means of three cascade impactors (5-stage Sioutas-type impactor) at the flowrate of 9 l.min^{-1}, running in parallel for 12 h a day over three days (8am-8pm), resulting in 36 h of sampling per month. Each cascade impactor (Sioutas) was mounted with four 25 mm diameter filters, and one 37 mm filter (one Sioutas with polycarbonate nuclepore filters with 1 µm porosity). A second (Sioutas) impactor, equipped with quartz filters (QMA, Whatman), was dedicated to carbonaceous aerosol measurements (for black carbon – BC and organic carbon – OC determination). A third cascade impactor ran with Teflon filters (Zefluor, Pall Corporation®) for water-soluble ions and trace element analysis. Aerodynamic particle diameters collated by the cascade impactors were in the following cut-off size ranges (marked as M5 - M1 in Table 1): <0.25 µm, 0.25-0.50 µm, 0.50-1 µm, 1-2.5 µm, and ≥2.5 µm. For the data analysis, the results were presented in three particle size fractions, which were combined out of five sizes: ultrafine particle, UF [<0.25 µm], fine, F [>0.25<1 µm], and coarse C [>1µm and >2.5 µm]. For oxidative potential measurements, particles from the same season were pooled.

Aethalometer (BC) measurement

Black carbon (BC) equivalent concentrations were also measured by using an optical attenuation method applied in a seven wave-length aethalometer (Magee Scientific®, model AE-42) ranging from 370 to 950 nm wavelengths. Just the periods towards the end of June 2012 and at the beginning of July 2012 were excluded due to temporary instrument stoppage, which left some periods with data gaps.

Meteorological data

An essential task in this study was to correctly allocate collated data into two main weather seasons with similar prevailing climatic conditions, i.e. wet and dry season, as to better categorise the entire dataset. This, we based on the following:

i. prevailing climatic patterns (Tyson and Preston-Whyte, 2000),
ii. the NOAA's ARL data for the sampling week and the South African Weather Service (SAWS) precipitation information because the measurements at the Vaalpark site did not include any rain gauge
iii. a nearby Sasol company AQ monitoring station (Vaal Eco-Park) acquisition of the wind speed and wind directions data.

From this, a decision was taken to keep the April filter samples within the wet season dataset, while the May data/filters were allocated to the dry season. September's filters were included in the dry season set, while October's filters were included in the wet season. There were several days with rain at the end of October (when our aerosol samplings were taken). Although October could fit in either season, the data were kept in the "wet season" data grouping.

Analytical methods

Carbonaceous content analysis by a thermal method

Black carbon (BC) and total carbon (TC) contents were determined from quartz filters with a thermal method developed by Cachier et al. (1989). Two similar aliquots of the same filter were separately analysed. One portion was directly examined for total carbon content (TC). The other portion was first submitted to a pre-combustion step (2 h at 340°C under pure oxygen) to eliminate OC and then analysed for BC content. Organic carbon (OC) concentrations were calculated as the difference between TC and BC.

Major ions (ion chromatographer) analysis

This method was used to determine water-soluble compounds. Teflon filters are weighted on a METTLER MC21S electronic microbalance with a sensitivity of 1 µg and measure the precision of ± 5 µg, to determine the aerosol mass loadings before analysis. In order to determine the water-soluble components (WSOC), half of the samples are extracted in 15 ml of Milli-Q water (resistivity ≈ 18.2 MΩ) by ultrasonic stirring for 15 minutes. Three ion chromatographs, DX-100, ICS 1000 and DX 500 of Dionex were equipped respectively with an anion exchange column IonPac AS40, a cation exchange column IonPac AS50 and a carbonate exchange column IonPac AS50, which are used for separating the ions. A combination of 1.8 mM Na_2CO_3 and 1.7 mM $NaHCO_3$ is used as the anion eluent, while a 20 mM solution of methane sulphonic acid (CH_3SO_2OH) is used as the cation eluent, and 100 % water de-ionised is used for eluting carbonate ion. WSOCs obtained were Cl^-, SO_4^{2-}, NO_3^-, CO_3^{2-}, K^+, Na^+, NH_4^+, Mg^{2+} and Ca^{2+} expressed in ppb. The limits of detection were less than 10 ppb for anions and cations.

Aerosol oxidative potential (OP) analysis

The three size fractions presented above were reconstituted from sampled nuclepore membrane filters. Recovery of the particles was achieved as previously described by Ramgolam et al. (2009). Briefly, polycarbonate membranes were sonicated (Ultrasonic Processor, Bioblock Scientific), 3 × 5 sec at 60 Watt in 15mM HEPES (2-[4-(2-hydroxyethyl)piperazin-1-yl]ethane-sulfonic acid) (Life Technologies, Gibco®). Blank filters were prepared in the same way and were used as a control in the experiments. Particle

suspensions were stored at -20°C (to prevent degradation and bacterial development) until use, and were again sonicated (3 × 10 sec) just before dilution in the culture media for acellular assays.

Three different acellular assays were performed to characterise the OP:

- the scission of plasmid DNA;
- the consumption of dithiothreitol (DTT), a reducing agent, and
- the depletion of antioxidants (AO): ascorbic acid (AA), uric acid (UA) and glutathione (GSH) in synthetic lung surfactant, according to the methods specified in Crobeddu et al. (2017).

Plasmid assay

It served as a determination of the ability of PM to induce DNA breaks. As such, it is a good indicator of PM ability to generate free radicals reactive enough to break DNA.

The DNA used was a plasmid (φX174) of 5386 bp. It is super-helicoidal except where there is a simple strand DNA break leading to a relaxed form. These forms can be separated by gel electrophoresis and quantified to evaluate their respective proportion.

Plasmid (290 ng) was incubated with 100 µg/ml particles in the presence of hydrogen peroxide (0.4 mM) in 15 mM Hepes buffer for 4 hours at 37°C under gentle agitation (Rotolab 3D). After incubation, samples were electrophoresed on a 1 % agarose gel at 50 V for 5 hours in a 1X Tris-Borate-EDTA buffer (Euromedex). At the end of the migration, DNA bands were visualised using ethidium bromide (2 µg/ml) by soaking for 10 min. The fluorescence is visualised on UV trans-illuminator and the intensity of the fluorescence of the super-helicoidal and relaxed forms was quantified using "Image J" software in order to express the results as a percentage (%) of relaxed form.

Anti-oxidant depletion assay

It served as a determination of the ability of PM to deplete anti-oxidants in a synthetic lung lining fluid. Three specific anti-oxidants used were: ascorbic acid (AA), uric acid (UA) and glutathione (GSH).

PM of each size fraction at 25, 50 and 100 µg/ml were incubated for 4 h at 37°C with AA, UA and GSH, under agitation. At the end of every incubation, each sample was centrifuged in 0.22 µm spins (Costar®, Fisher Scientific) (30 sec at 12 000 g) to eliminate the nanoparticle. AA, UA, GSH and the oxidised GSH form (GSSG) were analysed by means of reversed-phase HPLC (Shimadzu HPLC system interfaced with the "LabSolution" software). Samples were injected onto a C18 column (length 250 mm; internal diameter 4.6 mm; particle size 5 µm) at 40°C. The mobile phase composition for the gradient system was 25 mM sodium phosphate monobasic; 0.5 mM octane sulphonic acid (pH 2.7) for mobile phase A; and 100 % acetonitrile for mobile phase B. The gradient programme was 0-5min 0 % B; 5-10 min 0-12 % B; the initial conditions (100 % A) were then maintained for 10 min.

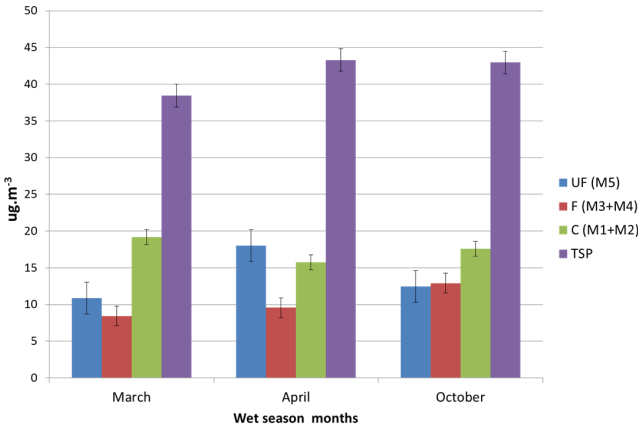

Figure 3: *Vaalpark aerosol concentrations (µg.m^{-3}) over the wet season sampling period for all main sizes (UF, F, C) in comparison with total suspended particulates (TSP), mean ± standard error of measurement (SEM).*

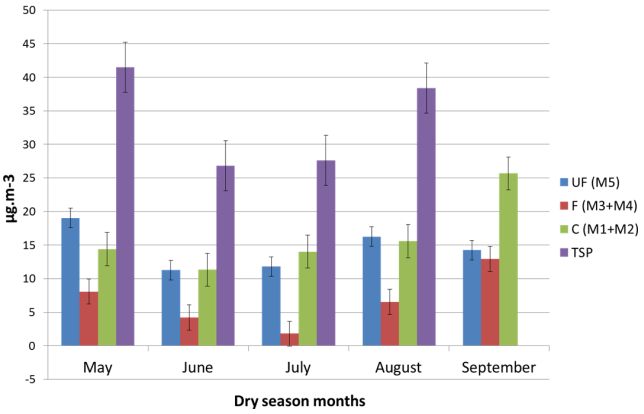

Figure 4: *Vaalpark aerosol concentrations (µg.m^{-3}) over the dry season sampling period for all main sizes (UF, F, C) in comparison with total suspended particulates (TSP), (mean ± SEM).*

The products were monitored spectrophotometrically at 243 nm (AA); 280 nm (UA) and 210 nm (GSH; GSSG) and quantified by integration of the peak absorbance area.

DTT (dithiothreitol) consumption assay

The redox-active compounds catalyse the reduction of oxygen to superoxide by DTT, which is oxidised to disulphide (Kumagai et al., 2002).

PM of each size-fraction at 25, 50 and 100 µg/ml was incubated with 200 µM of DTT for 1 h at 37°C. After incubation, samples were centrifuged for 15 min at 3500 g at 4°C to eliminate PM. 5 mM of DTNB was added to the supernatant. The absorbance was analysed at 405 nm (micro-plate reader Multiskan-EX, Thermo Scientific).

For all the valid samples, the final results' blank sample values were taken into account at the time of calculation.

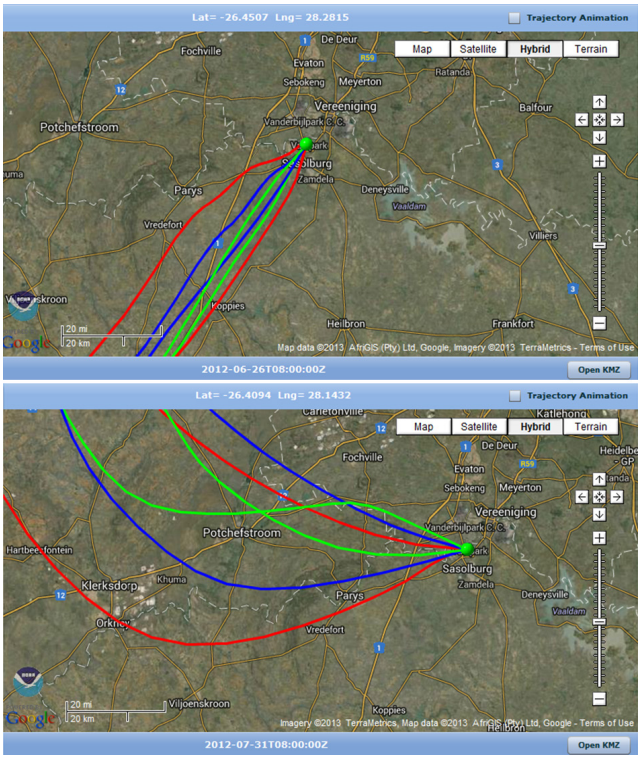

Figure 5: *Back trajectories (HYSPLIT 6-hourly, arriving heights at 200 m) for the June (upper figure) and July (lower figure) sampling periods (after ARL, GDAC, NOAA).*

Results

Aerosol filter concentrations, its carbonaceous ratios and seasonal trends

Considering the distribution of the aerosol filter concentrations over each main season, it was evident that the concentrations were comparable between the months of the wet season (Figure 3).

During two dry season months (June and July), concentrations decreased 'unseasonably', which constituted the anomaly for this region (dry and cold temperature, low ambient relative humidity, increased residential burning as well as the start of occasional wildfires/biomass burning) (Figure 4).

For these two anomalous months (June and July), air mass movement and origin were investigated. The wind speeds during these sampling periods increased and the back trajectory analysis for the sampling period indicated that the air masses were overpassing from the S-SW (June) and NW-W (July) directions (Figure 5) (ARL, GDAC, NOAA, 2013). The prevailing air mass originated from cleaner and essentially background regions. This influenced the aerosol concentrations at the sampling site. Therefore, these questionable concentrations could be attributed to the intrusions of cleaner air masses that changed the prevailing ambient air concentrations (Figure 5).

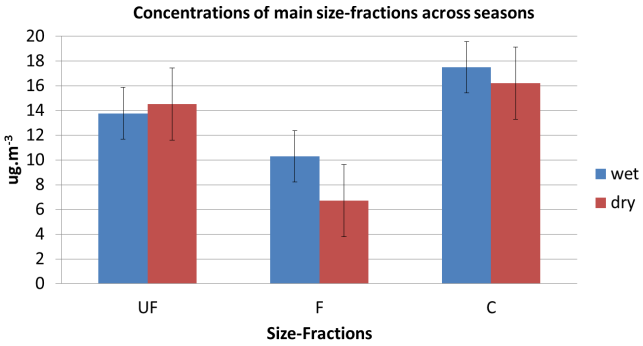

Figure 6: *Concentrations over three main sizes over the entire sampling period (mean ± SEM).*

Considering the concentrations averaged over the main sizes across the two main seasons, the following trend is evident: overall, the coarse fraction was recorded highest and Ultra-Fine 2nd highest concentrations over both seasons (Figure 6). The fine fractions were lower in both seasons.

The concentrations of BC, OC and TC for the three size-fractions and across the two main seasons were determined. Whatever the season, the highest carbonaceous concentrations were observed in the UF fraction, followed in C fraction and then in F fraction (Table 1). Both the organic carbon (OC) and black carbon (BC) had higher concentrations in the dry season compared to the wet season for all fractions.

Table 1: *Carbonaceous content mean concentrations; BC, OC and TC in the wet season*

Wet season ($\mu g.m^{-3}$)	M5	M3+ M4	M1+M2	**Total carbonaceous**
	UF	**F**	**C**	
BC	0.210	0.105	0.230	0.545
OC	0.352	0.056	0.183	0.591
TC	0.562	0.161	0.413	
Dry season ($\mu g.m^{-3}$)	M5	M3+ M4	M1+M2	**Total carbonaceous**
	UF	**F**	**C**	
BC	0.350	0.158	0.236	0.745
OC	0.546	0.111	0.433	1.09
TC	0.897	0.269	0.668	

In general, the concentrations of OC were higher than those of BC, whatever the season, except for the fine fraction.

The following ratios, i.e. OC/BC, BC/TPM (total particulate matter) and TC/TPM concentration ratios were considered for each season and were higher in the dry season (except for the UF OC/BC ratio). The highest ratios of OC/BC, BC/TPM and TC/TPM were found for the ultra-fine fraction (UF), followed by the coarse (C) and fine (F) fraction during the wet season (Table 2). For the dry season, the OC/BC ratio was the highest. The lowest ratios were found for each BC/TPM and TC/TPM in the wet season (Table 2).

Table 2: *Main carbonaceous ratios over three major mean size fractions for the wet and dry season*

Wet season ratios	UF	F	C
OC/BC	1.678	0.526	0.795
BC/TPM	0.008	0.004	0.008
TC/TPM	0.021	0.006	0.015
Dry season ratios	**UF**	**F**	**C**
OC/BC ratios	1.559	0.701	1.836
BC/TPM	0.013	0.006	0.009
TC/TPM	0.034	0.01	0.025

Aethalometer measured BC trends

Since this instrument ran continuously, the diurnal trend for the equivalent BC was determined using the mean BC results obtained from the absorptions near infrared (880 nm) wavelength (Hansen et al., 1984). The results are presented as two main South African Highveld seasons (Figure 7).

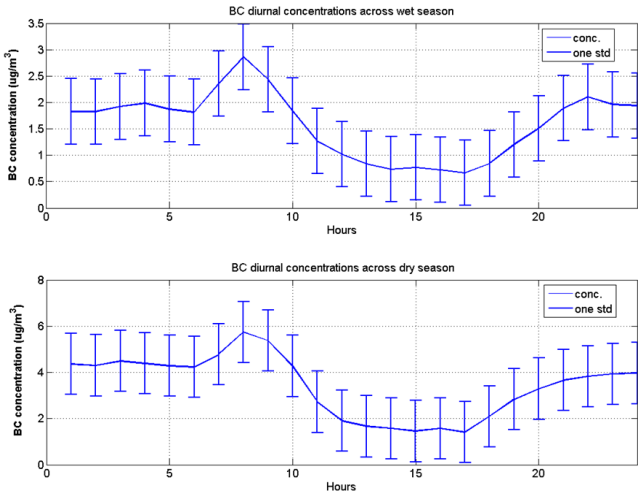

Figure 7: *Diurnal aethalometer BC concentration trends. Wet season sampling (upper graph) and dry season sampling period (lower graph) (mean ± one STD).*

The observed bimodal diurnal distribution is typical of the BC pollution reported (and $PM_{2.5}$ and PM_{10} trends) for South Africa's low-cost residential settlements over the SA Highveld region (Hersey et al., 2015; Venter et al., 2012). However, the morning peaks of the Vaalpark aethalometer BC data appear higher than the evening peaks for both seasons. As expected, the dry period had higher concentrations overall. However, the evening peak was lower than the morning peak. The mid-day to mid-afternoon diurnal concentration levels were low. Should there be much industrial pollution reaching/impacting the sampling site, these concentrations would be higher after the break of nocturnal and development of convective boundary layer, ~3-4h after sunrise (Korhonen et al., 2014; Gierens et al., 2018)

Inter-comparison of aethalometer BC with filter determined BC content

The comparison of BC results from filter analysis and aethalometer data obtained for the same days is shown in Figure 8. Both sets of data followed a similar trend during the entire sampling period, gradually increasing in concentrations from the late spring towards winter and higher levels over winter as expected. Although there were substantial differences between their absolute values, it needs to be taken into account that the filter sampling ran 12 h a day while the aethalometer measured 24 h. As elaborated earlier, the June and July 'dip' in the trend level is explained by the cleaner air mass overpasses (HYSPLIT modelled back trajectories, ARL, NCAR, NOAA, 2013). This is also evident when both datasets for the same period were plotted alongside one another (Figure 8).

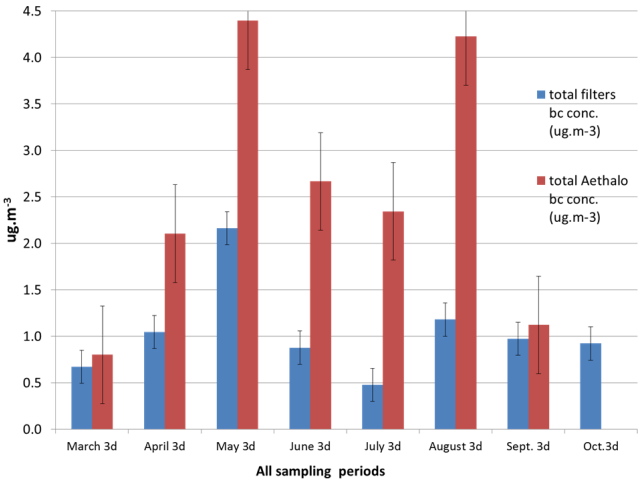

Figure 8: *Comparison of filter sampling BC with aethalometer BC trends over the same three days in each sampled month. Note that the aethalometer values were adjusted 30% lower to account for scattering and other reasons as per Hansen et al. (1984). Impactor concentrations were unchanged (mean ± SEM).*

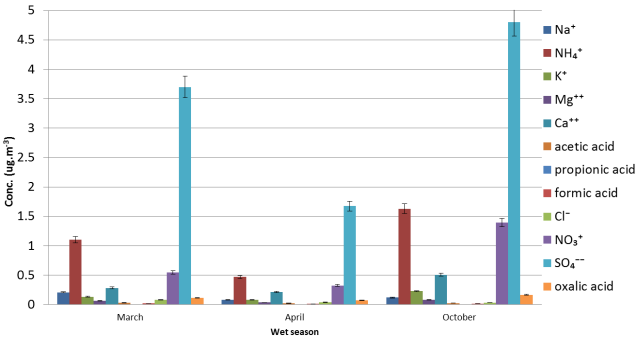

Figure 9: *Ionic concentrations over the wet season sampling periods (mean ± SEM).*

Ionic aerosol content

The content of important non-carbonaceous compounds was also determined. To a certain extent, the ionic concentrations reflect similar trends to the carbonaceous measurements.

We considered the ionic concentrations over the two main sampling season periods and also per species (Figure 9).

Considering the dry season months, the concentrations in the May, August and September sampling periods followed the well-known trends of increased concentrations of S and N compounds. However, two mid-dry season sampling months (June and July) were at much lower levels (Figure 10).

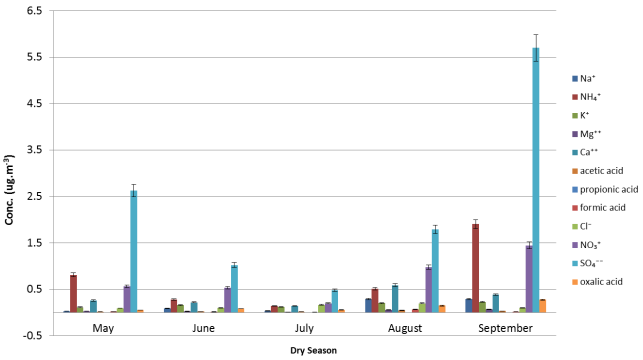

Figure 10: *Ionic composition concentrations over the dry season sampling periods (mean ± SEM).*

The same reason for the lower concentration levels was applicable, as in case of carbonaceous content concentrations: the sampling site weather played a crucial role – having been influenced by the cleaner air masses arriving from the regional background. This background has neither much industrial and residential burning, nor wildfires. Similarly, the emissions from traffic in the regional background are also limited.

Relative contributions of aerosol components

Considering the wet season, it is evident that the sulphate, ammonium and nitrate were dominant species (Figure 11). Black carbon and organic carbon appear almost equally (~3 %). However, the majority of the aerosol content was sulphate, and this fact points to the prevailing anthropogenic origin of the aerosol sampled at the sampling site. Together with nitrate and ammonium, these compounds represent more than two-thirds of the filter-sampled aerosol content (Figure 11).

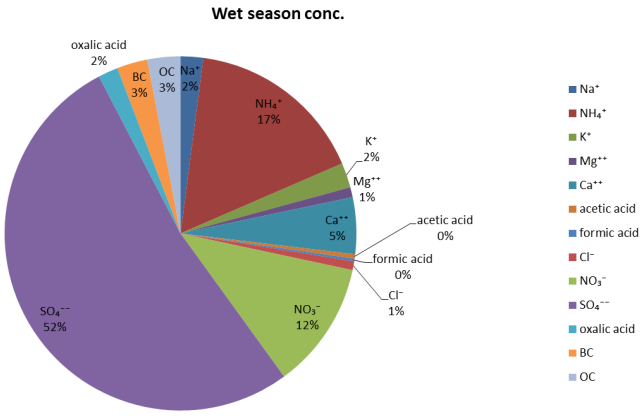

Figure 11: *Aerosol composition concentrations' relative fraction in the wet season (%).*

The dry season had just slightly different aerosol composition concentrations. The fractions of sulphate (45 %) and ammonium (14 %) were lower, while the relative fractions of BC (5 %), OC

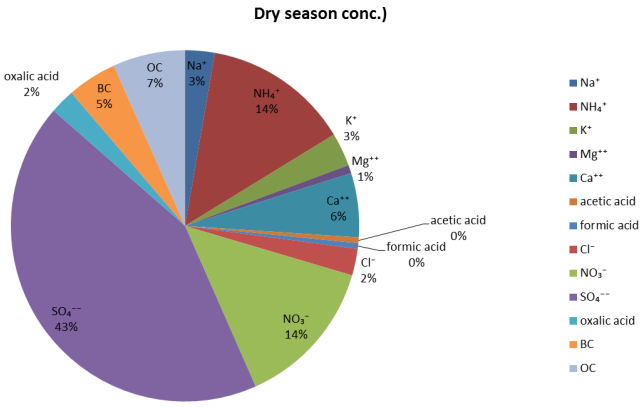

Figure 12: *Aerosol composition concentrations' relative fraction in the dry season (%).*

(7 %) and nitrate (14 %) increased. This points to an increased influence of the biomass and residential fossil fuel burning and not necessarily a decrease in industrial emissions (Figure 12).

It is also essential to look at the ratios between the major ions and the BC and OC with regard to the total particulate matter (TPM).

The higher sulphate ratios within TPM and higher sulphate over BC ratios indicate the prevalence of industrial combustion emissions, while increased BC/TPM ratios indicate the prevalence of domestic and biomass burning combustion emissions. The lack of higher sulphate ratios in the dry season can be explained by the fact that most of the production (petrochemical and metallurgical industries) stacks are designed to be high, over ~200 m (Beukes et al., 2012, Held et al., 1996), so the emissions are released aloft the inversion capping layer in the colder and drier part of the year. The reduced moisture content also impedes the formation of sulphate (and nitrate), leaving the residential and traffic pollution content higher, likely emanating from within, as well as farther from the concerned area (Figure 13 a-d).

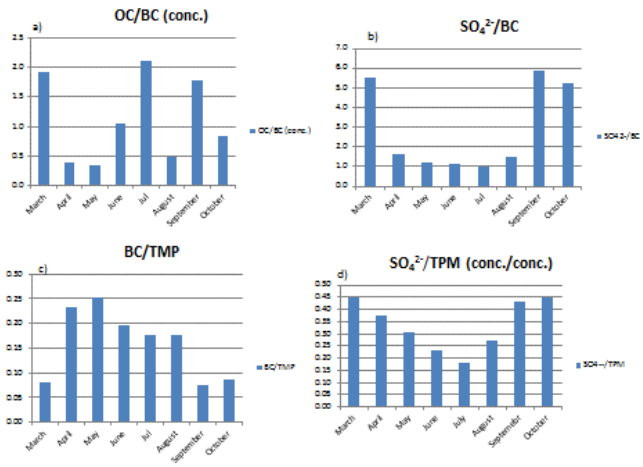

Figure 13: *Ratios between aerosol major compounds over the main sampling periods: a) OC/BC, b) sulphate/BC, c) BC/TPM d) sulphate/TPM.*

The linear regression as the coefficient of correlation (Pearson), between each of ions, was calculated, and the output (R) has been indicated as a matrix. Certain high correlations (in dark red)

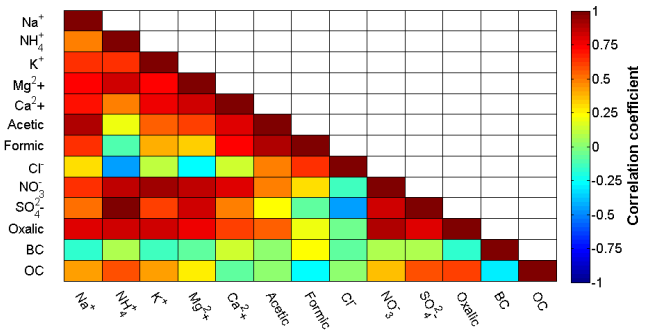

Figure 14: *Linear regression correlation (R values) matrix among all ionic species and carbonaceous fractions (OC and BC).*

and some low to anti-correlations (in yellow, green and blue) have been revealed (Figure 14).

The correlation calculations did not indicate any unexpected association. To gain more insight into the species correlations, a multiple-linear regression (MLR) with root mean square error (RMSE) output was modelled using Matlab® for all the main ions, taking BC as the dependent variable, while others were left as an independent.

The model output (MLR) also indicated that the Mg^{2+}, Cl^- and oxalic were negatively associated and NH_4^+ and formic positively with BC. The model showed that to accurately predict BC (i.e. the change in RMSE reaches a minimum), NH_4^+, Mg^{2+}, Formic, Cl^- and oxalic were the most important and the OC was less significant (Figure 15).

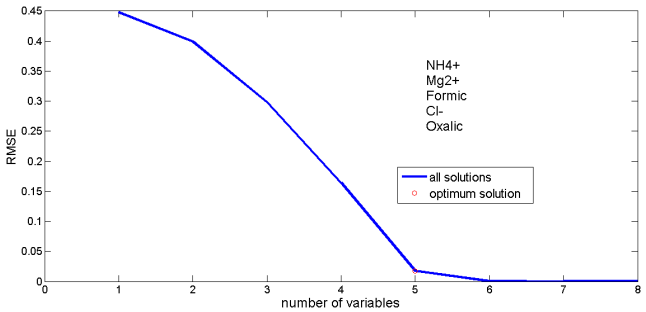

Figure 15: *Ionic MLR modelled, where BC was dependant variable and all other ions independent.*

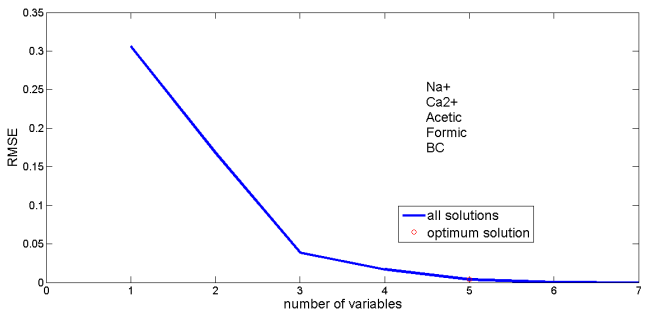

Figure 16: *Ionic MLR modelled in the instance where OC was dependent variable and all other ions independent.*

Another multiple-linear regression with RMSE was modelled for all the main ions taking OC as the dependent variable while others were left as an independent. This MLR output indicated that the acetic ions and BC are negatively associated, while Na^+, Ca^{2+} and formic ions are positively associated with OC. Modelled showed that to accurately predict OC (i.e. the change in RMSE reaches a minimum), Na^+, Ca^{2+}, Acetic, Formic and BC were the most important (Figure 16).

Aerosol oxidative potential

Plasmid assay results

To induce DNA breaks, the production of free radicals, such as hydroxyl radical (OH•), is needed. Adding H_2O_2 (reactive oxygen species) to the incubation mixture allows the Fenton reaction to take place, if there is the presence of transition metals in the aerosol. All samples increased the production of DNA damages as measured by the relaxation of the plasmid DNA by comparison with the control (T) and H_2O_2 (reactive oxygen species) alone, suggesting the presence of transition metals. The effects of aerosol from the wet season were increasing as the PM size was decreasing. For the dry season, the F fraction is the most reactive, whereas C and UF exhibited a similar effect (Figure 17).

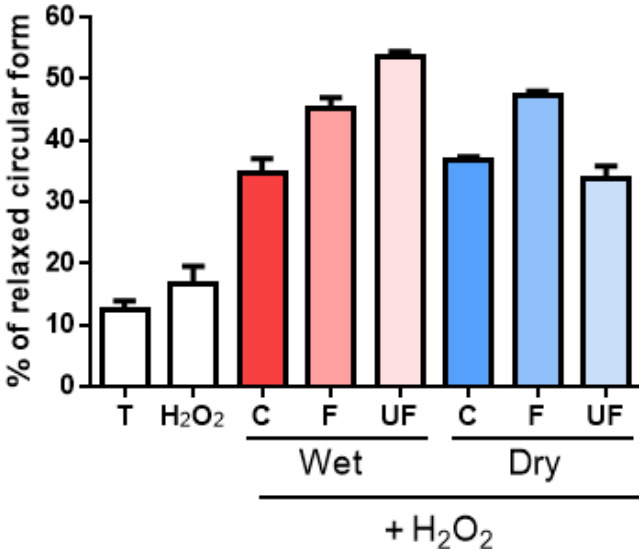

Figure 17: *Intrinsic oxidative capacity characterised by plasmid scission assay (%) of relaxed plasmid circular form relative to super-helicoidal form, for the coarse (C), fine (F) and ultra-fine (UF) size fractions of wet (W) and dry (D) season used at 100 µg/mL: n=2, T=control (mean ± SEM)..*

Anti-oxidant depletion assay results

All samples induced an ascorbic acid (AA) and glutathione (GSH) depletion (Figure 18 top and middle) with concomitant production of oxidised glutathione (GSSG) (Figure 18 bottom). There was no depletion of uric acid (UA) for either season or different dilutions (therefore not shown). For wet season samples, there is a trend of more AA and GSH depletion as the aerosol fraction size is decreasing. For the dry season aerosols, we observed a stronger effect of the fine fraction compared to the other size fractions. The coarse fraction depleted AA and GSH only at the highest concentrations tested.

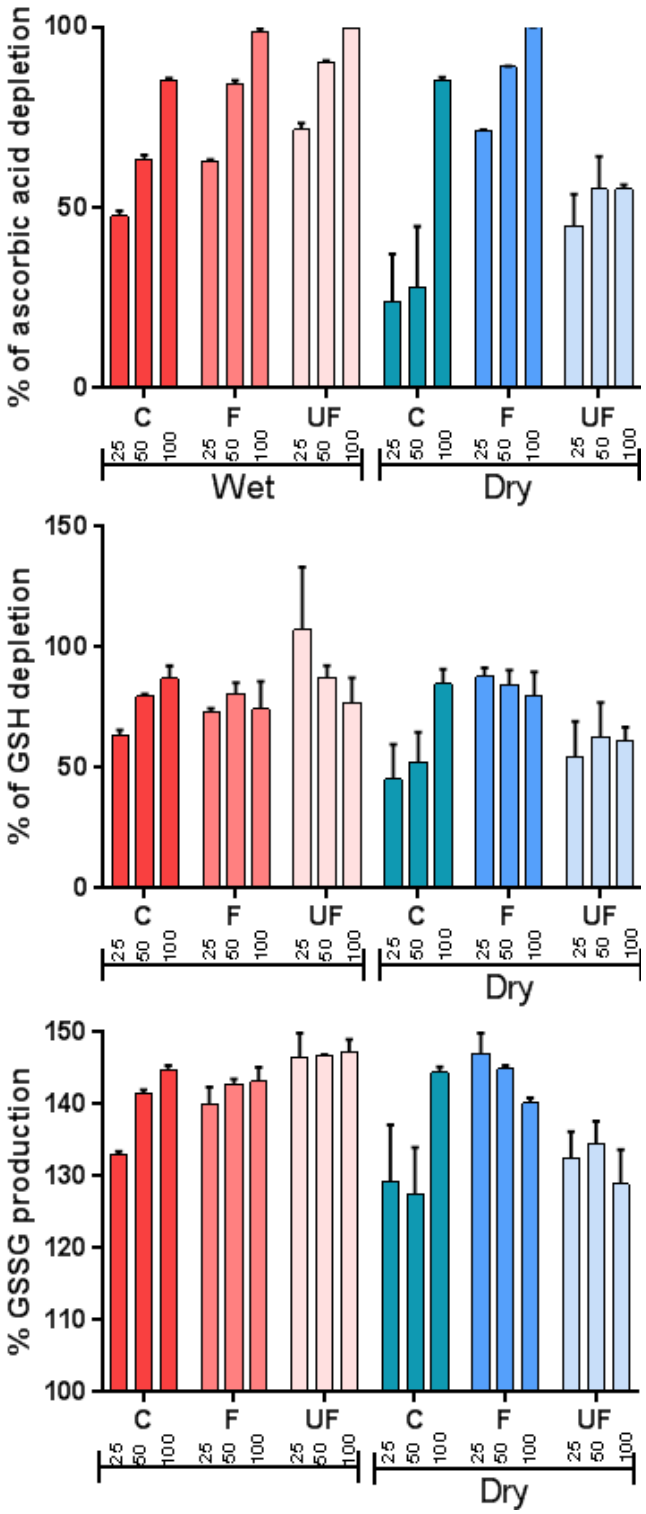

Figure 18: *Intrinsic oxidative capacity characterised by the anti-oxidant assay for the coarse (C), fine (F) and ultra-fine (UF) size fractions of wet and dry season at different concentrations. Ascorbic acid depletion (top graph); reduced glutathione (GSH) depletion (middle graph); and oxidised glutathione (GSSG) production in a synthetic lung lining fluid (bottom graph), n=2 independent experiments with three replicates, values are mean ± SEM.*

DTT consumption assay results

All samples induced a significant dose-dependent DTT depletion (Figure 19). For wet season samples, as seen for plasmid assay and anti-oxidant depletion assays, we observed that as the size is decreasing, the DTT depletion was more pronounced and significantly different for the ultrafine fraction compared to the fine and coarse fractions. The DTT depletion was generally more pronounced with PM from the wet season considering the same size fraction. For the dry season, the higher effect was observed for the fine fraction and is significant at the highest concentration, whereas the other size fractions exhibited similar oxidative potential (OP) (Figure 19).

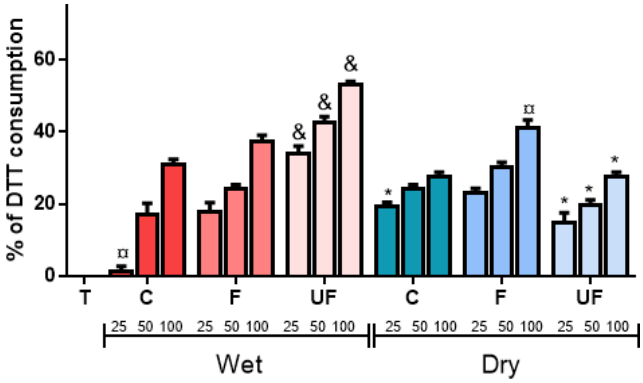

Figure 19: *Intrinsic oxidative capacity characterized by DTT depletion for the coarse (C), fine (F) and ultra-fine (UF) size fractions of wet and dry season at different concentrations (µg/ml). n=3 independent experiments with three replicates, different from wet samples of the same size and dose, different from other sizes whatever the season, different from other fractions of the same season (values are mean ± SEM).*

The three OP assays consistently showed that for aerosols sampled in the wet season, the OP tends to increase as the size is decreasing. By contrast, for the dry season, the size-fraction exhibiting the highest OP is the fine mode. The UF fraction showed the most contrasting OP between the two seasons.

Discussion

To a certain extent, the gravimetric mass of the filter samples was low, and we attribute this to the type of the environment where the sampling was taken, i.e. an affluent, fully electrified residential neighbourhood, as well as to the prevailing weather patterns effectively upwind from the nearby pollution sources. However, the aerosols still contained representative mixtures of chemical compounds, such as sulphate, nitrate and ammonium, strongly indicating the prevalence of the anthropogenic sources. The carbonaceous content of both organic and black carbon was not high, and they are mostly similar in content in both seasons, although an evident higher organic content in the dry season indicates the typical influence of the biomass burning season, which is prevalent in the 2nd part of the dry season (from mid-winter to mid-spring).

The diurnal pattern seen from the continuous data indicates bimodal peaks, with the morning peak being higher than the evening peak. The plausible explanation is that the traffic into and around the area and some residential fossil fuel burning increase concentrations in the morning. While the evening peak is not as pronounced, it is indicative of reduced emissions in comparison

to the morning peaks (reduced traffic and reduced residential burning). This is understandable for the sampling site, which is in a residential electrified area and although a number of electrified households in SA use fossil fuels for heating, this activity is less pronounced than in low-income settlements (Hersey et al., 2015; Venter et al., 2012).

A question was posed why the concentrations of major ionic compounds (being characteristic of industrial combustion emissions) in March and October were similar compared with the months in the dry season (May, September). We attribute this to the regular existence of a more turbulent boundary layer (with higher solar radiation and fewer inversions and horizontal stratification) allowing entrainment/fumigation of the pollution from the industrial sources nearby. The months of March and October fit this description, although April concentrations were reduced, and the April period meteorology had played a determining role.

It is important to point to the presence of some carboxylic acids (oxalic, formic, acetic) found among particulate organic matter. However, similar relative content of these carboxylic acids in both seasons indicates that there are a number of sources/precursors of these species during both seasons. Since the main precursors to carboxylic acids are VOC (from both anthropogenic and biogenic sources), their presence throughout the year is expected. The degradation of anthropogenic hydrocarbons photo-chemically, as well as oxidation of isoprene and monoterpene emissions, leads to the formation of such acids (Herrmann et al., 2005; Lim et al., 2005). Among the identified carboxylic acids, oxalic acid is the most abundant compound in both seasons. Oxalic acid is frequently observed as one of the most abundant single organic compounds in tropospheric particles. Sources of oxalic acid in the atmosphere are numerous. Biomass burning (Yamasoe et al., 2000) and vehicular exhaust (Kawamura and Kaplan, 1987) are two primary emission sources for oxalic acid.

Atmospheric oxidation processes provide secondary mechanisms for oxalic acid formation from both anthropogenic and biogenic volatile organic compound emissions (e.g. Kerminen et al., 2000; Warneck, 2003).

In the studied environment, there are both types of sources present throughout the year as well as seasonally. While the anthropogenic emissions from industries are more or less constant, plus emission transport from out of the area, the domestic biomass burning is not specific to the case study area, although there are a number of low-income dwellings in its vicinity that rely on biomass burning for cooking and heating. In addition, in the winter months, intentional and spontaneous biomass burning in the form of veld (grassland and savannah) fires is a substantial source of pollutant emissions.

A significant correlation (coefficient of determination) was found between potassium (K) as a well-known fire tracer (Gao et al., 2003; Andreae et al., 1998) with oxalic acid. Correlations between potassium (K) and oxalic acid per seasons resulted in high correlations (R^2=0.87 in dry and R^2=0.99 in wet season). This indicates that the oxalic acid would be strongly correlated to fire origin, both domestic uses as well as biomass burning seasonal fires.

The US Department of Health and Human Services (1992) indicated that oxalic acid aids a range of detrimental pulmonary impacts. Acetic acid is a strong eye, skin and mucous membrane irritant. Any prolonged skin contact with glacial acetic acid may result in tissue destruction.

Although formic acid shares most of the chemical properties of other carboxylic acids, it has low toxicity. However, the concentrated formic acid is corrosive to the skin (Reutemann and Kieczka, 2002). Such characteristics hint that the carboxylic acids do play a role in OP of collated aerosol.

Interestingly, to the best of our knowledge, this study was the first to compare the OP of PM according to the size fraction in South Africa. It revealed that the fine fraction exhibited a constant OP whatever the season. By contrast, the ultrafine fraction exhibited the highest OP in the wet season compared to the other size fraction, whereas it was equal to the coarse fraction in the dry season. These data suggest the role of the particle composition on the ultrafine OP that would warrant deeper chemical characterisation. We can speculate that metals are involved in OP, since the DNA scission can only occur through hydroxyl radical production that is known to be mediated by transition metals (Crobeddu et al. 2017).

The acidity of the particles due to the significant content in oxalic and formic acid could favour the bioavailability of metals. Size-wise, UF and F fractions of aerosol in both seasons are capable of evident OP when exposed to in-vitro acellular human cell proxies.

Conclusions

Aerosol OP was characterised on particle size fractions collected in a heavily industrialised area at a residential suburb through a set of three different assays. By detecting ROS generated by different particle characteristics, OP can be seen as an integrative indicator of aerosol exposure enabling the prediction of the aerosol effects on human/animal health within the South African context. In this study, in the wet season, the fine (F) and ultra-fine (UF) fractions were proven to exhibit stronger OP compared to the coarse (C) fraction. In addition, the UF fraction had higher OP in the wet season compared to the dry season when elevated pollution levels are usually recorded on the SA Highveld due to its climate and land topography. The investigated air mass movement point out that cleaner (background) air mass influx dominated large periods of the sampling campaign, and that the sampling site is upwind of major pollution sources in the area, such as industrial, and low-cost and slum dwellings. However, should more particulates have been sampled in the dry season, this could have exerted different effects. Nevertheless, the chemical analysis confirmed the predominance of anthropogenic compounds present in collated aerosol – sulphate, nitrate and ammonium.

The role of combustion from transport and residential burning is also evident through the continuous BC sampling having typical bimodal peaks and a much stronger peak in inversion capped the cold and dry season. Furthermore, inevitably, we could pick up evidence of biomass burning, first in BC/OC content and ratios, as well as in the presence of the organic acids, which are generally secondary pollutants resulting from oxidation of VOC. The organic acids emanate from both anthropogenic and biogenic sources such as biomass burning, transport emission as well as industrial production and effluents such as carboxylic acids.

Overall, the sampled aerosols represent the majority of the pollution sources being present well in the atmosphere of the wider area and are not mainly and only generated by the industrial emission despite being so spatially close to major chemical and metallurgical industries. Residential use of fossil and biomass burning with seasonal veld fires contributes to emissions of solid particulate matter in the area. In terms of overall health effect, by considering the OP assays, we showed significant oxidative properties for the finer sizes and in the more turbulent atmosphere, although the main chemical contributors to OP could not be determined. Combined aerosol characterisations, including OP with experimental biological studies, with relevant target cells *in vitro* (and *ex vivo*), are necessary to decipher the link between chemical composition, OP and cellular oxidative stress within the context of South African aerosols. Such *in vitro* studies would allow establishing the predictivity of OP as an exposure indicator to be used in epidemiological studies.

Acknowledgements

We acknowledge the French GDRI for making possible the collaboration between the French and SA institutions as well as the NRF, its SA counterpart, in this collaboration. We also kindly thank the SAWS for the provision of rainfall data and SASOL Ltd for the provision of wind data.

References

Air Resources Laboratory, Gridded Meteorological Data Archives, [online] available at http://www.arl.noaa.gov/archives.php ([accessed November 2013].

Andreae, M. O., Andreae, T. W., Annegarn, H., Beer, J., Cachier, H., le Canut, P., Elbert, W., Maenhaut, W., Salma, I., Wienhold, F. G., Zenker, T. 1998 Airborne studies of aerosol emissions from savanna fires in southern Africa, 2, Aerosol chemical composition, J. Geophys. Res., 103, 32,119-32,128. https://doi.org/10.1029/98JD02280

Assamoi E-M., Liousse C. 2010, A new inventory for two-wheel vehicle emissions in West Africa for 2002. Atmos. Environ, 44:3869-3996. https://doi.org/10.1016/j.atmosenv.2010.06.048

Beukes, P., Dawson, N., van Zyl, P. G. 2010. Theoretical and practical aspect of Cr (VI) in the South African FeCr industry, S. African Inst. Min. M., 110, 743-750.

Boko, M. I., Niang, A., Nyong, C., Vogel, A., Githeko, M., Medany, B., Osman-Elasha, R., Tabo, Yanda, P. Africa, in. Climate Change 2007: impacts, adaptation and vulnerability. The contribution of working group II to the fourth assessment report of the intergovernmental panel on climate change, edited by Parry, M. L., Canziani, O. F., Palutikof, J. P., van der Linden, P. J., and Hanson, C. E. Cambridge University Press, Cambridge UK, 433-467.

Boogaard, H., Janssen, N.A.H., Fischer, P.H., Kos, G.P.A., Weijers, E.P., Cassee, F.R., van der Zee, S.C., de Hartog, J.J., Brunekreef, B., Hoek, G., 2012. Contrasts in oxidative potential and other particulate matter characteristics collected near major streets and background locations. Environ Health Perspect. 120, 185-191. https://doi.org/10.1289/ehp.1103667

Booyens, W., Van Zyl, P.G., Beukes, J.P., Ruiz-Jimenez, J., Kopperi, M., Riekkola, M., Josipovic, M., Venter, A.D., Jaars, K., Laakso, L. 2015. Size-resolved characterisation of organic compounds in atmospheric aerosols collected at Welgegund, South Africa. J Atmos. Chem. 72, 43-64. https://doi.org/10.1007/s10874-015-9304-6

Borm, P.J.A, Kelly, F., Künzli, N., Schins, R.P.F., Donaldson, K., 2007. Oxidant generation by particulate matter: from biologically effective dose to a promising, novel metric. Occup. Environ Med; 64, 73-74. https://doi.org/10.1136/oem.2006.029090

Butt E. W., Rap, A., Schmidt, A., Scott, C. E., Pringle, K. J., Reddington, C. L., Richards, N. A. D., Woodhouse, M. T., Ramirez-Villegas, J., Yang, H. 2016. The impact of residential combustion emissions on atmospheric aerosol, human health, and climate. Atmos. Chem. Phys., 16, 873-905. https://doi.org/10.5194/acp-16-873-2016

Cachier H, Brémond MP, Buat-Ménard P: 1989. Determination of atmospheric soot carbon with a simple thermal method. Tellus B Chem. Phys. Meteorol., 41(B):379-390. https://doi.org/10.3402/tellusb.v41i3.15095

Cassee F.R., Héroux M.E., Gerlofs-Nijland M.E., Kelly F.J. 2013. Particulate matter beyond mass: recent health evidence on the role of fractions, chemical constituents and sources of emission. Inhal. Toxicol. (14):802-12. https://doi.org/10.3109/08958378.2013.850127

Celo, V., Dabek-Zlotorzynska, E., Mathieu, D., Okonskaia, I. 2010; Validation of a Simple Microwave-Assisted Acid Digestion Method Using Microvessels for Analysis of Trace Elements in Atmospheric PM2.5 in Monitoring and Fingerprinting Studies. The Open Chemical and Biomedical Methods Journal 3, 143-152. https://doi.org/10.2174/1875038901003010143

Crobeddu B., Aragao-Santiago L., Bui L.C., Boland S., Baeza Squiban A. 2017. Oxidative potential of particulate matter 2.5 as predictive indicator of cellular stress. Environ Pollut. 230,125-133. https://doi.org/10.1016/j.envpol.2017.06.051

Donaldson, K., Beswick, P.H., Gilmour, P.S. 1996. Free radical activity associated with the surface of particles: A unifying factor in determining biological activity? Toxicol Lett., 88, 293-298. https://doi.org/10.1016/0378-4274(96)03752-6

Doumbia, E.H.T., Liousse, C., Galy-Lacaux C., Ndiaye, S.A., Diop, B., Ouafo, M Assamoi, E.M., Gardrat, E., Castera, P., Rosset, R., Akpo, A., Sigha, L. 2012. Real-time black carbon measurements in West and Central Africa urban sites. Atmos. Environ. 54, 529-537. https://doi.org/10.1016/j.atmosenv.2012.02.005

Eck T. F., Holben B. N., Ward D. E., Mukelabai M., M. Dubovik O., Smirnov A., Schafer J. S., Hsu N. C., Piketh, S. J., Queface, A., Le Roux J., Swap R. J., Slutsker, I. 2003. Variability of biomass burning aerosol optical characteristics in southern Africa during the SAFARI 2000 dry season campaign and a comparison of single scattering albedo estimates from radiometric measurements, J. of Geophys Res.-Atmos, Volume 108, issue D13. https://doi.org/10.1029/2002JD002321

Favez, O, Haddad, I. E, Piot, C., Bor'eave, A., Abidi, E., Marchand, N., Jaffrezo, J.-L., Besombes, J.-L., Personnaz, M.-B., Sciare, J. 2010. Inter-comparison of source apportionment models for the estimation of wood burning aerosols during wintertime in an Alpine city (Grenoble, France). Atmos. Chem. Phys., 10, 5295-5314. https://doi.org/10.5194/acp-10-5295-2010

Freiman, T. and Piketh, S. J. 2002. Air Transport into and out of the Industrial Highveld Region of South Africa, Journal of Applied Meteorology, 42, 994-1002. https://doi.org/10.1175/1520-0450(2003)042<0994:ATIAOO>2.0.CO;2

Gao, S., D. A. Hegg, P. V. Hobbs, T. W. Kirchstetter, B. I. Magi, and M. Sadilek. 2003. Water-soluble organic components in aerosols associated with savanna fires in southern Africa: Identification, evolution, and distribution, J. Geophys. Res., 108(D13), 8491. https://doi.org/10.1029/2002JD002324

Garstang, M., Tyson, P.D., Swap, R., Edwards, M., Kallberg, P., Lindesay, J.A. 1996. Horizontal and vertical transport of air over southern Africa. Journal of Geophysical Research-Atmospheres, 101, 23721-23736. https://doi.org/10.1029/95JD00844

Gierens, R. T., Henriksson, S., Josipovic, M., Vakkari, V., van Zyl, P. G., Beukes, J. P., Wood, C. R. and O'Connor, E. J. 2018. Observing continental boundary-layer structure and evolution over the South African savannah using a ceilometer, Theor. Appl. Climatol., 1-14. https://doi.org/10.1007/s00704-018-2484-7

Hansen, A.D.A., Rosen, H., Novakov, T. 1984; The aethalometer - An instrument for real-time measurement of optical absorption by aerosol particles. The Science of The Total Environment, 36, 191-196. https://doi.org/10.1016/0048-9697(84)90265-1

Herrmann, S.M., Anyambab, A., Tucker, C.J. 2005. Recent trends in vegetation dynamics in the African Sahel and their relationship to climate. Global Environmental Change 15, 394-404. https://doi.org/10.1016/j.gloenvcha.2005.08.004

Hersey, S. P., Garland, R., M. Crosbie, E, Shingler, T., Sorooshian, A., Piketh, S., Burger, R. 2015. An overview of regional and local characteristics of aerosols in South Africa using satellite, ground, and modelling data, Atmos. Chem. Phys., 15, 4259-4278, www.atmos-chem-phys.net/15/4259/2015/. https://doi.org/10.5194/acp-15-4259-2015

Hirsikko, A., Vakkari, V., Tiitta, P., Manninen, H. E., Gagne, S., Laakso, H., Kulmala, M., Mirme, A., Mirme, S., Mabaso, D., Beukes, J. P., Laakso, L. 2012. Characterisation of sub-micron particle number concentrations and formation events in the western Bushveld Igneous Complex, South Africa. Atmos. Chem. Phys., 12, 3951-3967. https://doi.org/10.5194/acp-12-3951-2012

Kampa, M. and Castanas, E. 2008. Human health effects of air pollution, Environmental Pollution, 151, 362-367. https://doi.org/10.1016/j.envpol.2007.06.012

Kawamura, K. and Kaplan, I. R. 1987. Motor Exhaust Emissions as a Primary Source for Dicarboxylic Acids in Los Angeles Ambient Air. Environ. Sci. Technol., 21, 105-110. https://doi.org/10.1021/es00155a014

Kerminen V-M., Ojanen, C., Pakkanen, T., Hillamo, R., Aurela, M. and Kinen, J.M. 2000. Low-Molecular-Weight Dicarboxylic Acids In An Urban And Rural Atmosphere, J. Aerosol Sci. Vol. 31, No. 3, pp. 349-362. https://doi.org/10.1016/S0021-8502(99)00063-4

Kimemia, D., Annegarn, H., Makonese, T., Molapo, V. and Robinson, J. 2010. Characterisation of domestic biomass combustion technologies used in Setswetla, Alexandra township, Gauteng, 18 DUE, Cape Peninsula University of Technology, Cape Town, 29-31 March 2010, 167-173, 2010

Koike, E. and Kobayashi, T. 2006. Chemical and biological oxidative effects of carbon black nanoparticles, Chemosphere, 65, 946-951. https://doi.org/10.1016/j.chemosphere.2006.03.078

Korhonen K, Giannakaki E., Mielonen T., Pfüller, A., Laakso L., Vakkari V., Baars H., Engelmann R., Beukes J.P., Van Zyl P.G., Ramandh A, Ntsangwane L, Josipovic M, Tiitta P., Fourie G., Ngwana I., Chiloane K., Komppula M. 2014. Atmospheric boundary layer top height in South Africa: measurements with lidar and radiosonde compared to three atmospheric models. Atmos Chem Phys 14(8):4263-4278. https://doi.org/10.5194/acp-14-4263-2014

Kuik, F., Lauer, A., Beukes, J. P., Van Zyl, P. G., Josipovic, M., Vakkari, V., Laakso, L., Feig, G. T. 2015. The anthropogenic contribution to atmospheric black carbon concentrations in southern Africa: a WRF-Chem modelling study. Atmos. Chem. Phys., 15, 8809-8830. https://doi.org/10.5194/acp-15-8809-2015

Kumagai, Y., Koide, S., Taguchi, K., Endo, A., Nakai, Y., Yoshikawa, T., Shimojo, N. 2002. Oxidation of Proximal Protein Sulfhydryls by Phenanthraquinone, a Component of Diesel Exhaust Particles. Chem. Res. Toxicol, Volume 15, Number 4, pp. 483-489. https://doi.org/10.1021/tx0100993

Laakso, L., Vakkari, V., Virkkula, A., Laakso, H., Backman, J., Kulmala, M., Beukes, J. P., van Zyl, P. G., Tiitta, P., Josipovic, M., Pienaar, J. J., Chiloane, K., Gilardoni, S., Vignati, E., Wiedensohler, A., Tuch, T., Birmili, W., Piketh, S., Collett, K., Fourie, G. D., Komppula, M., Lihavainen, H., de Leeuw, G., and Kerminen, V.-M. 2012. South African EUCAARI measurements: seasonal variation of trace gases and aerosol optical properties, Atmos. Chem. Phys., 12, 1847-1864. https://doi.org/10.5194/acp-12-1847-2012

Landrigan, P.J., Fuller, R., Acosta, N. et al. 2018.The Lancet Commission on pollution and health. The Lancet Commissions, 391, 462-512

Li N., Sioutas, C., Cho, A., Schmitz, D., Misra, M., Sempf, J., Wang, M., Oberley, T., Froines, J. Nel, A. 2003. Ultrafine Particulate Pollutants Induce Oxidative Stress and Mitochondrial Damage, Environ. Health Perspect., 2003 Apr; 111(4): 455-460. https://doi.org/10.1289/ehp.6000

Life Technologies, Gibco, [online] available at: https://www.thermofisher.com/za/en/home/brands/life-technologies.html [accessed Sept. 2018].

Lim, L. P., Lau, N.C, Garrett-Engele, P. Grimson, A., Schelter, J. M., Castle, J., Bartel, D. P., Linsley P. S.; Johnson J. M. 2005. Microarray analysis shows that some microRNAs downregulate large numbers of target mRNAs, Nature, volume 433, pages 769-773. https://doi.org/10.1038/nature03315

Liousse C. and Galy-Lacaux C. 2010. Urban pollution in West Africa. Meteorology, 71, 45-49. https://doi.org/10.4267/2042/37377

Liousse, C., Cachier, H., Jennings, S.G. 1993. Optical and thermal measurements of black carbon aerosol content in different environments: Variation of the specific attenuation cross-section, sigma (σ). Atmos. Environ. Part A. General Topics, 27, 1203-1211. https://doi.org/10.1016/0960-1686(93)90246-U

Mafusire, G., Annegarn, H.J., Vakkari, V. Beukes, J. P. Josipovic, M., van Zyl, P. G. and Laakso, L. 2016. Sub-micrometer aerosols and excess CO as tracers for biomass burning air mass transport over southern Africa, J. Geophys. Res. Atmos., 121. https://doi.org/10.1002/2015JD023965

Magee Scientific®, model AE-42, USA [online] available at: https://mageesci.com/our-products/ae42/ [accessed Sept. 2018].

Makonese, T., Masekameni, M.D., Annegarn, H.J. Energy use scenarios in an informal urban settlement in Johannesburg, South Africa. In: Proceedings of the domestic use of energy conference, 31 March-2 April 2016. Cape Town: Cape Peninsula University of Technology. 1-6. 10.1109/DUE.2016.7466703. https://doi.org/10.1109/DUE.2016.7466703

Micro-plate reader Multiskan-EX, Thermo Scientific [online] available at: https://www.selectscience.net/products/thermo-scientific-multiskan-ex/?prodID=10022 [accessed Sept. 2018].

Occupational Safety and Health Guideline for Acetic Acid, Centers for Disease Control and Prevention, US Department of Health and Human Services, 1992, USA.

Petzhold, A., Kopp, C., Niessner, R. 1997. the dependence of the specific attenuation cross-section on black carbon mass fraction and particle size. Atmos. Environ., 31, 661-672. https://doi.org/10.1016/S1352-2310(96)00245-2

Pope and Dockery. 2006. Health Effects of Fine Particulate Air Pollution: Lines that Connect. J Air Waste Manag Assoc , 56, 709742. https://doi.org/10.1080/10473289.2006.10464485

QMA, Whatman, [online] available at: https://www.sigmaaldrich.com/catalog/product/aldrich/wha1851047?lang=en&region=ZA&gclid=Cj0KCQjwguDeBRDCARIsAGxuU8YlRrKbuCUdYmukqj9H8cjLs3A70-Y6Rg9j4ciLSbjfDPc091Xmeg8aAnpuEALw_wcB [accessed Sept. 2018].

Ramgolam K., Favez O., Cachier H., Gaudichet A., Marano F., Martinon L., Baeza-Squiban A. 2009. Size-partitioning of an urban aerosol to identify particle determinants involved in the proinflammatory response induced in airway epithelial cells, Part Fibre Toxicol, 6:10. https://doi.org/10.1186/1743-8977-6-10

Reutemann W. and Kieczka H. 2002. Formic Acid in Ullmann's Encyclopedia of Industrial Chemistry, Wiley-VCH, Weinheim.

Ross, K.E., Piketh, S. J., Bruintjes, R. T., Burger, R. P., Swap, R. J., Annegarn, H. J. 2003. Spatial and seasonal variations in CCN distribution and the aerosol-CCN relationship over southern Africa. J. of Geophys Res.-Atmos 108, 8481-8498. https://doi.org/10.1029/2002JD002384

Sauvain, J., Deslarzes, S., Riediker, M. 2008. Nanoparticle reactivity toward dithiothreitol. Nanotoxicology, 2, 121-129. https://doi.org/10.1080/17435390802245716

Seinfeld, J. H. and Pandis, S. N. 2006. Atmospheric Chemistry and Physics: from Air Pollution to Climate Change, 2nd Edition, J. Wiley & Sons, New York.

Sundström, A.-M., Nikandrova, A., Atlaskina, K., Nieminen, T., Vakkari, V., Laakso, L., Beukes, J.P., Arola, A., Van Zyl, P.G., Josipovic, M., Venter, A.D., Jaars, K., Pienaar, J.J., Piketh, S., Wiedensohler, A., Chiloane, E.K., De Leeuw, G. and Kulmala, M. 2015. Characterization of satellite-based proxies for estimating nucleation mode particles over South Africa, Atmos. Chem. Phys., 15, 4983-4996. https://doi.org/10.5194/acp-15-4983-2015

Swap R.J., Annegarn, H.J., Suttles, J.T., King, M.D., Platnick, S., Privette, J.L., Scholes, R.J. 2003. Africa burning: A thematic analysis of the Southern African Regional Science Initiative (SAFARI 2000). J Geophys Res Atmos 108, 8465-8479. https://doi.org/10.1029/2003JD003747

The Government Gazette Republic of South Africa, 4 May 2007 No. 29864, [online] available at http://www.info.gov.za/view/DownloadFileAction?id=73046 [accessed October 2013].

Tiitta, P., Vakkari, V., Croteau, P., Beukes, J. P., van Zyl, P. G., Josipovic, M., Venter, A. D., Jaars, K., Pienaar, J. J., Ng, N. L., Canagaratna, M. R., Jayne, J. T., Kerminen, V. M., Kokkola, H., Kulmala, M., Laaksonen, A., Worsnop, D. R., and Laakso, L. 2014. Chemical composition, main sources and temporal variability of PM1 aerosols in southern African grassland, Atmos. Chem. Phys., 14, 1909-1927. https://doi.org/10.5194/acp-14-1909-2014

Tyson, P. D. and Preston-Whyte, R. A. 2000. The Weather and Climate of Southern Africa, Oxford University Press, Oxford, UK.

Ultrasonic Processor, Bioblock Scientific [online] available at: https://www.fishersci.com/us/en/products/ITN31F6R/equipment-instruments.html [accessed Sept. 2018].

Vakkari, V., Beukes, J. P., Laakso, H., Mabaso, D., Pienaar, J. J., Kulmala, M., Laakso, L. 2013. Long-term observations of aerosol size distributions in semi-clean and polluted savannah in South Africa. Atmos. Chem. Phys., 13, 1751-1770. https://doi.org/10.5194/acp-13-1751-2013

Val S., Martinon L., Cachier H., Yahyaoui A., Marfaing H., Baeza-Squiban A. 2011. Role of size and composition of traffic and agricultural aerosols in the molecular responses triggered in airway epithelial cells. Inhal. Toxicol. 23 (11):627-640. https://doi.org/10.3109/08958378.2011.599445

Val, S., Liousse, C., Doumbia, E.H.T., Galy-Lacaux, C., Cachier, H., Marchand, N., Badel, A., Gardrat, E., Sylvestre, A., Baeza-Squiban, A. 2013. Physico-chemical characterization of African urban aerosols (Bamako in Mali and Dakar in Senegal) and their toxic effects in human bronchial epithelial cells: description of a worrying situation. Particle and Fibre Toxicol. 10:10. https://doi.org/10.1186/1743-8977-10-10

Venter, A.D., Vakkari, V., Beukes, J.P., van Zyl P.G., Laakso, H., Mabaso, D., Tiitta, P., Josipovic, M., Kulmala, M., Pienaar, J.J., Laakso, L. 2012. An air quality assessment in the industrialised western Bushveld Igneous Complex, South Africa, S. Afr. J. Sci. 108, 1059. https://doi.org/10.4102/sajs.v108i9/10.1059

Warneck, P. 2003. In-cloud chemistry opens a pathway to the formation of oxalic acid in the marine atmosphere. Atmos. Environ., 37, 2423-2427. https://doi.org/10.1016/S1352-2310(03)00136-5

Yamasoe, M.A., Artaxo, P., Miguel, A.H., Allen, A.G. 2000. Chemical composition of aerosol particles from direct emissions of vegetation fires in the Amazon Basin: water-soluble species and trace elements. Atmos. Environ., 34, 1641-1653. https://doi.org/10.1016/S1352-2310(99)00329-5

Zefluor, Pall Corporation [online] available at: https://www.opticsplanet.com/pall-zefluor-ptfe-membrane-filters-pall-life-sciences-p5pj047.html [accessed Sept. 2018].

Research article

Aerosol optical properties and direct radiative effect over Gobabeb, Namibia

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Received: 15 February 2019 - **Reviewed:** 17 July 2019 - **Accepted:** 17 Sept 2019
https://doi.org/10.17159/caj/2019/29/2.7518

Abstract

Atmospheric aerosols contribute significantly to the uncertainty in radiative forcing effects that influence the climate as well as posing a significant health risk to humans. The climatic implications of aerosols are dependent on many variables, including aerosol size, shape, chemical composition, and position in the atmospheric column. The radiative impact of aerosols transported over the west coast of southern Africa has been found, in particular, to be complicated by the aforementioned aerosol properties. This study investigated the columnar optical properties of aerosols over Gobabeb, Namibia (23.5621° S, 15.0409° E, 405 m asl) using sunphotometer data between December 2014 and November 2015. Aerosol mean optical depth AOD_{500} had its maximum and minimum values in 2015 August (0.37±0.30) and June (0.06±0.02), respectively. The Angström parameter was mostly above unity during the study period and indicated the prevalence of fine particles for the most part of the year with maximum and minimum values observed in August 2015 (1.44±0.19) and December 2014 (0.57±0.19), respectively. The columnar water vapor was highest in January (2.62±0.79) and lowest in June (0.76±0.27). The volume size distribution showed the fine particles having a mean radius of about 0.16 µm and the coarse mode had variation in sizes with a radius ranging between 3 µm and 7 µm. The single scattering albedo at visible wavelengths ranged between 0.83 and 0.91. The phase function was high at small angles but minimum at about 140° in all seasons. The radiative forcing showed a heating effect in all seasons with maximum and minimum in winter (9.41 Wm^{-2}) and autumn (3.64 Wm^{-2}), respectively. Intercomparison of the sunphotometer data with the Moderate Resolution Imaging Spectroradiometer (MODIS) showed that the satellite sensor overestimates the aerosol loading compared to the ground-based sunphotometer measurements. Both sets of observations were better correlated during the spring and winter seasons than for the summer and autumn.

Keywords

Aerosol Optical Depth, Volume Size Distribution, Radiative Forcing, Single Scattering Albedo, MODIS

Introduction

Atmospheric aerosol studies over the past years have yielded progress in unraveling their impacts on health and the climate system. These advances have been driven by extensive ground-based measurement networks, in situ-measurements during field campaigns, satellite-based instrument observations around the globe and finally modeling efforts that integrate and synthesize current scientific understanding. Their optical, radiative and microphysical properties are as a result, not totally as evasive as they used to be in previous years. The uncertainties of these properties are mostly due to the spatial and temporal variability of aerosol emissions, loading, transport and the diversity of the mechanisms of transformation and formation in the atmosphere (Stocker et al., 2013).

There have been more than four decades of observation

and measurements of aerosols in southern Africa and their seasonality has been established (Chester et al., 1972; Diner et al., 2001; Laakso et al., 2012; Losno et al., 1992). Studies showed that much of the aerosols produced in southern Africa are being transported over the Atlantic through the south-west coast (Swap et al., 1996). While the aerosol concentration transported is very low during the transition from summer to autumn, the concentration escalates dramatically as the season goes from winter to spring (Losno et al., 1992). The sudden rise in aerosol concentration has been attributed to biomass burning, industrial emission, and aeolian erosion (Piketh et al., 1999; Swap et al., 1996). The period between July and September has the highest recorded occurrence of open biomass burning (Eck et al., 2013; Vakkari et al., 2018).

The complex interactions of the coastal stratocumulus cloud deck off Namibia and Angola are thought to have a potentially important radiative, biogeochemical and hydrological impact on the local, regional and global climate (Hegg et al., 2012; Platnick et al., 2001; Zhou et al., 2017). Several large-scale regional and land-atmosphere experiments conducted in the southern African region over the past decades, including the Transport and Atmospheric Chemistry near the Equator-Atlantic (TRACE-A); the Southern African Fire-Atmosphere Research Initiative (SAFARI-92) and the Southern African Atmospheric Research Initiative SAFARI-2000 have shown that: a) Southern Africa is an important region of the world in terms of global emissions to the atmosphere and a good natural laboratory to evaluate the earth, land and ocean-atmosphere interactions, and b) critical gaps remain in our understanding of the fate and impacts of the emissions on the functioning of the regional land-atmosphere-ocean systems. To partially fill these gaps, the international Sea-Earth-Air Linkages in southern Africa (SEALS-sA) program has recently been initiated in order to promote coordinated field experiments and long-term monitoring along the west coast of southern Africa. In this framework, the Gobabeb Research and Training Centre hosts an ongoing research program consisting of the long-term measurements of meteorological parameters, aerosols, and fog. In this paper, we discuss the preliminary results of observations of the columnar aerosol optical properties provided by the Aerosol Robotic Network (AERONET) sunphotometer located at the station and compared with satellite remote sensing observations from MODIS.

Site and instrumentation

Site

Gobabeb Research and Training Centre (Gobabeb; 23.5621° S, 15.0409° E, 405 m above sea level; https://deims.org/182ac129-f0b8-4808-9842-0c7c841802cc (accessed Aug 14, 2019)) is located in the Namib Desert on the southwestern African coast. This desert stretches nearly 2000 km from South Africa to Namibia and extends to Angola. Gobabeb is located along the ephemeral Kuiseb River, with gravel plains to the north and sand dunes to the south. Precipitation is mainly in the summer and autumn months (December to May) with an average annual precipitation of 10 mm and a temperature of 21° C (Eckardt et al., 2013). Although the site is 70 km from the coast it is still influenced by cold coastal air from over the Benguela current. Consequently, clouds and fog are a fairly common occurrence at Gobabeb (average of 115 fog days/annum; SASSCAL, 2019). Precipitation over this region is triggered directly and indirectly by large circulation systems including easterly waves associated with tropical temperate troughs, the west coast low-pressure system, westerly temperate cold fronts and associated coastal low-pressure system and finally cut-off lows that develop over the subcontinent (Eckardt et al., 2013; Tyson and Preston-Whyte, 2000).

Instrumentation

Sunphotometer

The sun/sky photometer (CE-318 CIMEL Inc, Paris, France) is a radiometer capable of taking both direct sun and diffuse sky measurements (Holben et al., 1998). The instrument keeps track of the sun with the sensor head being within approximately 1° of the sun. The instrument takes direct sun measurements at eight spectral channels of 340, 380, 440, 500, 675, 870, 940 and 1020 nm with the 940 nm band used to measure columnar water vapor approximately every 15 minutes throughout the day. The diffuse radiance measurements in the solar almucantar or principal plane are taken at four spectral bands 440, 675, 870 and 1020 nm in the mornings and afternoon mostly at low solar elevation. From these measurements, the single scattering albedo (SSA), refractive index, volume size distribution, etc. are retrieved using an inversion algorithm. More details regarding the error estimation, uncertainties, and calibration protocols have been presented elsewhere (Dubovik et al., 2002; Dubovik and King, 2000).

Measurements are transmitted through the METEOSAT geostationary satellite being received and monitored from NASA Goddard Space Flight Center in the USA. AERONET observations are available at three levels; level 1.0 corresponding to raw data, level 1.5 corresponding to cloud-screened observations and level 2.0 corresponding to quality-assured data to which recent calibration has been applied. The uncertainty for AOD retrieval under cloud screened condition for wavelengths greater than 440 nm is < ± 0.01 and for shorter wavelengths < ± 0.02 or less than ± 5% uncertainty in the retrieval of the sky radiance measurements. Errors of retrieving particles in the size range ($0.1 \leq r \leq 7\mu m$) do not exceed 10% except for very small sizes less than 0.1 µm and higher than 7 µm. SSA has an uncertainty of about 0.03 – 0.05 depending on aerosol loading and aerosol types. Real and imaginary parts of the refractive index have uncertainties of about 0.3 – 0.5 and ± 0.04, respectively (Alam et al., 2012; Dubovik et al., 2002). In this paper, we present a preliminary analysis using level 2.0 for the direct sun measurement and level 1.5 for the inversion products for December 2014 to November 2015 due to scanty data for the inversion product. The problem of scanty data for inversion products in places of lower aerosol optical depth was studied in detail by Li et al. (2014) from 44

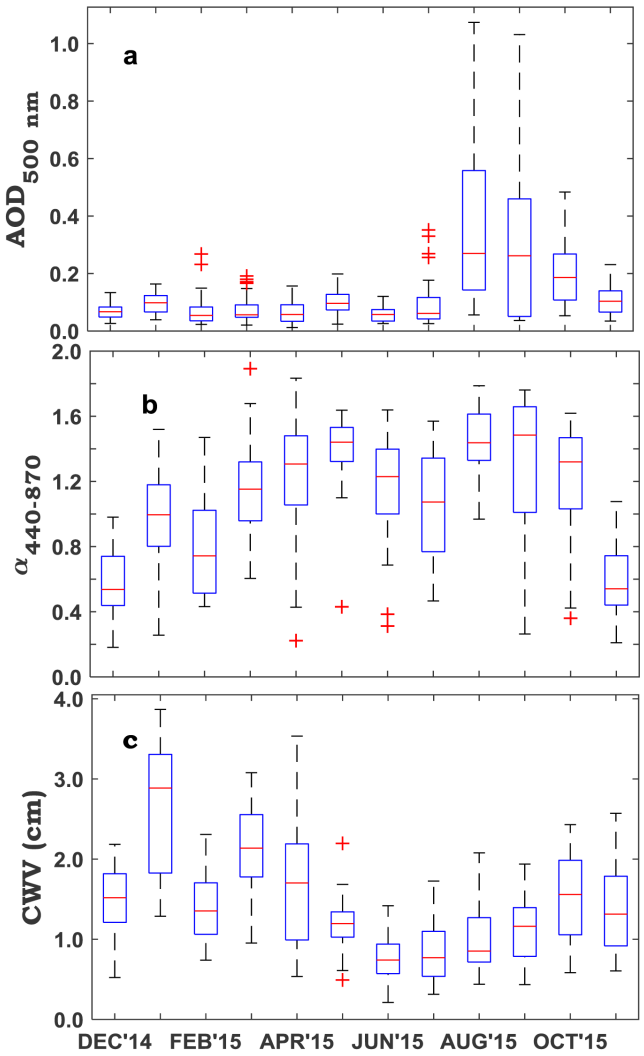

Figure 1: *(a) Aerosol optical depth at 500 nm, (b) Angström parameter at 440-870 wavelength and (c) Columnar water vapor content. The red line shows the median value of each month, the red crosses are the outliers, the vertical hinges represent data points from the lower to the upper quartile (i.e., 25th and 75th percentiles) and the whiskers represent data points from the 5th to 95th percentiles for the period December 2014 to November 2015 over Gobabeb.*

AERONET centers who then proposed some further screening to the level 1.5 that was adopted in this paper.

MODIS

MODIS has spatial resolutions of 250, 500 and 1000m with 36 spectral channels spanning 415 to 14235 nm capable of measuring radiances at the top of the atmosphere (TOA). It possesses two separate sensors employed in earth observation (King et al., 2003; Kumar et al., 2015). The two sensors Terra (1999) and Aqua (2002) orbit the earth crossing the equator at 10:30 Local Solar Time providing information about aerosols over land and ocean (Kaufman et al., 1997). The special algorithm involved in the retrieval of aerosol data is different for both land and ocean with accuracy over land lower than that of over the ocean (Levy et al., 2007; Remer et al., 2005; Sreekanth and Kulkarni, 2013; Tanré et al., 1997). The instrument with its cloud mask performs above twenty tests that incorporate cirrus detection tests to denote clear or cloudy pixels at a 1 x 1 km resolution. To enhance accuracy, pixels of surface reflectance greater than 0.15 are not considered (Chung et al., 2002; Kumar et al., 2015). For the present study, MODIS Terra (MOD08_D3) level 3 daily data were downloaded from https://giovanni.gsfc.nasa.gov/ (accessed October 5, 2017) and the data compared with that of AERONET. This dataset incorporates the aerosol models and polarization information for calculating the radiative transfer introduced by Remer et al. (2008). Ichoku et al. (2003) had earlier shown that MODIS can effectively monitor different types of aerosols. This ability to monitor was not just that of global but even regional. They carried out the validation of MODIS using AERONET during the SAFARI 2000 since southern Africa was dominated by smoke aerosols which stem from biomass burning particularly during August-September months. Here we did not limit our data to just these two months but we have extended this to a whole year.

Result and discussion

Major causes of variability in aerosol properties and air pollutant concentrations in southern Africa were extensively discussed by Laakso et al. (2012). The major causes were natural, anthropogenic and meteorological conditions existing in the region. During the winter and spring (June-November). there are common occurrences of wildfires in these months, this leads to a significant increase in the aerosol loading. In addition to these, there is lower boundary layer causing decrease in the mixing height and thereby aiding re-circulation of aerosols. Also there are higher industrial activities (electricity consumption becomes higher) and widespread domestic space heating. Whereas during the summer and autumn (December-May) months, mixing heights are higher, weather favoring advection of pollutants and precipitation during these seasons also causes wet deposition thereby leading to lower pollutant concentrations.

Monthly variability of AOD_{500}, $\alpha_{440-870}$, CWV

Monthly variability of aerosol optical depth at 500 nm (AOD_{500}), Angström exponent ($\alpha_{440-870}$) and columnar water vapor (CWV) at Gobabeb are shown in (Figure 1). The boxplot for each month revealed the median, the first and the third quartiles and cases of outliers. This helped to have a good representation of the measure of the central tendency of each month. AOD_{500} measured at Gobabeb had the highest loadings recorded between August and October 2015. This is consistent with previous measurements undertaken over the subcontinent in the past decades (Eck et al., 2003b; 2003a; Queface et al., 2011). The subcontinent experiences open biomass burning emissions during this period. The burning season injects a significant pulse of aerosols into the atmosphere that is transported across the entire region (Adesina et al., 2015; Eck et al., 2003a; Hersey et al., 2015; Kumar et al., 2013). The boxplot also indicated that the daily variability of aerosol optical depth within the month was moderate from December 2014 to July 2015 and afterward the daily variation became high from August to October 2015 due to the influence of open biomass burning that can be natural

Table 1: *The monthly summary of the mean, mode and standard deviation (SD) of the AOD_{500}, $\alpha_{440-870}$, and CWV at Gobabeb between December 2014 and November 2015.*

Season	Month	AOD_{500}			$\alpha_{440-870}$			CWV (cm)		
		Median	Mean	SD	Median	Mean	SD	Median	Mean	SD
Summer	Dec '14	0.07	0.07	0.02	0.54	0.57	0.19	1.52	1.46	0.42
	Jan '15	0.10	0.10	0.04	0.99	0.95	0.33	2.89	2.62	0.79
	Feb '15	0.05	0.07	0.06	0.74	0.80	0.31	1.35	1.43	0.44
Autumn	Mar '15	0.06	0.08	0.05	1.15	1.20	0.31	2.14	2.09	0.58
	Apr '15	0.06	0.07	0.04	1.31	1.22	0.41	1.70	1.73	0.90
	May '15	0.10	0.10	0.04	1.44	1.39	0.22	1.20	1.18	0.32
Winter	Jun '15	0.06	0.06	0.02	1.23	1.16	0.32	0.74	0.76	0.27
	Jul '15	0.06	0.10	0.09	1.07	1.07	0.32	0.77	0.84	0.38
	Aug '15	0.27	0.37	0.30	1.44	1.44	0.19	0.85	1.02	0.45
Spring	Sep '15	0.26	0.31	0.30	1.48	1.31	0.38	1.16	1.09	0.40
	Oct '15	0.19	0.20	0.10	1.32	1.20	0.34	1.56	1.54	0.52
	Nov '15	0.10	0.11	0.05	0.54	0.59	0.23	1.31	1.35	0.50

or anthropogenic (Vakkari et al., 2018). There were few outliers during some of the months of low AOD_{500} that may be as a result of a meteorological effect, as discussed by Eckardt et al. (2013).

The $\alpha_{440-870}$ is a qualitative factor determining aerosol size. It can be used to determine the relative abundance of the accumulation to the coarse mode. Higher values represent the dominance of the accumulation mode and vice versa (Alam et al., 2012). Unlike the AOD_{500} it has greater daily variability almost every month of the year. Apart from a few months, i.e. January, February, November and December, Gobabeb experienced high values of $\alpha_{440-870}$. Pawar et al. (2015) associated high $\alpha_{440-870}$ with corresponding low AOD_{500} with the continental average type of aerosols. This is what seems to be prevalent between March and July. The CWV also showed high variability especially during the first quarter of the year. The sea surface temperature affects the moisture content of the atmosphere in this region. Factors such as El Niño Southern Oscillation, Benguela upwelling and the influence of the western Indian Ocean all contribute to this variability (Eckardt et al., 2013).

Table 1 gives the summary statistics of the aerosol properties. Where there are high standard deviations, the mean and the mode show a noticeable difference. The mean AOD_{500} (with the standard deviation) was highest in 2015 August with a value of 0.37±0.30 and lowest in June with a value of 0.06±0.02. September and October also had high values of 0.31±0.30 and 0.20±0.10, respectively. $\alpha_{440-870}$ value had a maximum of 1.44±0.19 in August 2015 and a minimum of 0.57±0.19 in December 2014. It had values greater than 1.0 from March to October 2015. The high values show the prevalence of accumulation mode during this period. The Namib Desert, in general, receives little rainfall throughout the year, with an annual average of 25 mm (Eckardt et al., 2013). There was a relatively high amount of water vapor present in the atmosphere at the beginning of the year with the maximum in January 2015 having a value of 2.62±0.79 (cm) and a minimum of 0.76±0.27 (cm) in June. Winter months receive less rainfall than other months of the year at Gobabeb (Eckardt et al., 2013). The seasonal correlation coefficients between AOD_{500} and CWV (not shown) were 0.58, 0.50, 0.55 and 0.62 for summer, autumn, winter, and spring, respectively and the p-value in each case was < 0.0001. Though the correlation coefficient was not very high, it is statistically significant at a 95% confidence level.

Seasonal variability of the aerosol volume size distribution

The volume size distribution (VSD) presented in Figure 2 accounted for the mixture of different types of aerosols present and advected into a location that is conditioned by scavenging processes and meteorological influences (Ali et al., 2014). In the AERONET retrievals, the volume size distribution is represented in a lognormal form as

$$\frac{dV(r)}{d\ln r} = \sum_{i=1}^{n} \frac{C_i}{\sqrt{2\pi}\sigma_i} exp\left[\frac{-(\ln r - \ln R_i)^2}{2\sigma_i^2}\right] \quad (1)$$

Where C_i is the particle volume concentration, R_i is the median or geometric mean radius, σ_i is the variance or width of each mode, r is the particle radius, and n is the number of lognormal aerosol modes. In Figure 2 the VSD shows a bimodal lognormal distribution that varies according to seasons, i.e. summer (DJF), autumn (MAM), winter (JJA) and spring (SON). This distribution shows the ratio of the volume of the coarse and the accumulation particles present in each season. The spectral dependence and amplitude of the optical depth are normally responsible for the variations in the two modes (Bi et al., 2011). There were no significant changes in the size of the accumulation mode throughout the seasons as the radius centered around 0.16 µm. The volume in this mode was higher

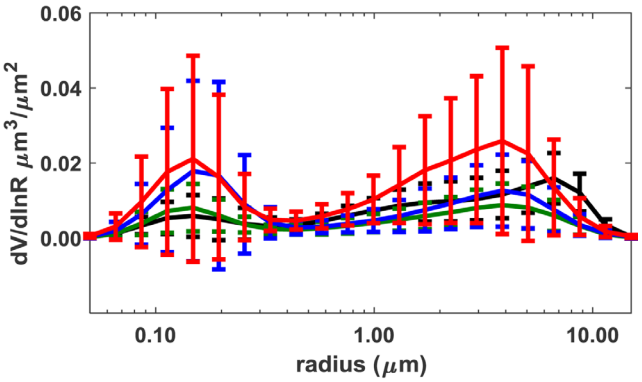

Figure 2: *AERONET retrieved mean volume size distribution in different seasons along with the standard deviations*

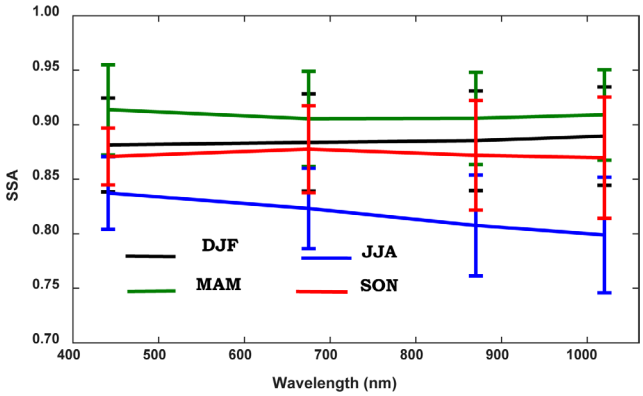

Figure 3: *Seasonal AERONET mean retrieved single scattering albedo at four different wavelengths along with the standard deviations*

for winter and spring and lower during summer and autumn. Spring had the highest volume concentration in both the accumulation and coarse modes. The size of the coarse particles had a noticeable variation with a radius ranging from 4 to 7 µm. The coarse particle distribution is lower in winter but slightly higher in summer when compared to accumulation mode. There seemed to be no difference in the distribution of both modes during autumn.

Seasonal variability of single scattering albedo

The single scattering albedo (SSA) is an important property in understanding aerosol radiative forcing and relates the ratio of scattering to the extinction coefficient. It depends on the aerosol refractive index (that is on the composition) as well as the volume size distribution of the aerosol (Dubovik and King, 2000). SSA presents a distinct spectral behavior depending on the type of aerosol; increase with wavelength will indicate that aerosols absorb more in the UV part of the spectrum. Such aerosols are large in size representing UV-absorbing compounds such as iron oxides from desert dust while a decrease with wavelength is mostly characteristic of fine aerosols that absorb at longer wavelengths. Such small size particles are associated with black carbon from biomass or urban/industrial aerosol (Dubovik et al., 2002). The seasonal plot of the SSA is shown in Figure 3. In summer, there is a slight increase from the shorter to the longer wavelengths (values of 0.881, 0.883, 0.885 and 0.890 at (440, 675, 870 and 1020) nm, respectively). This indicates that a particle of larger sizes predominates during this season. These particles can be of the desert origin or formed from the hygroscopic growth of aerosols in the atmosphere.

The columnar water vapor content is high during this season especially as seen in January (Figure 1) and so can favor this growth. In autumn, the SSA decreased from 0.914 to 0.905 and 0.906 and later increased to 0.910 at the aforementioned wavelengths. This implies that the season is not particularly dominated by either fine or coarse particles as aerosols of different sizes make a comparable contribution to the columnar aerosol loading. In winter, the spectral line has a constant decrease with wavelength, it means a higher absorption at a longer wavelength. This relates more to the predominance of

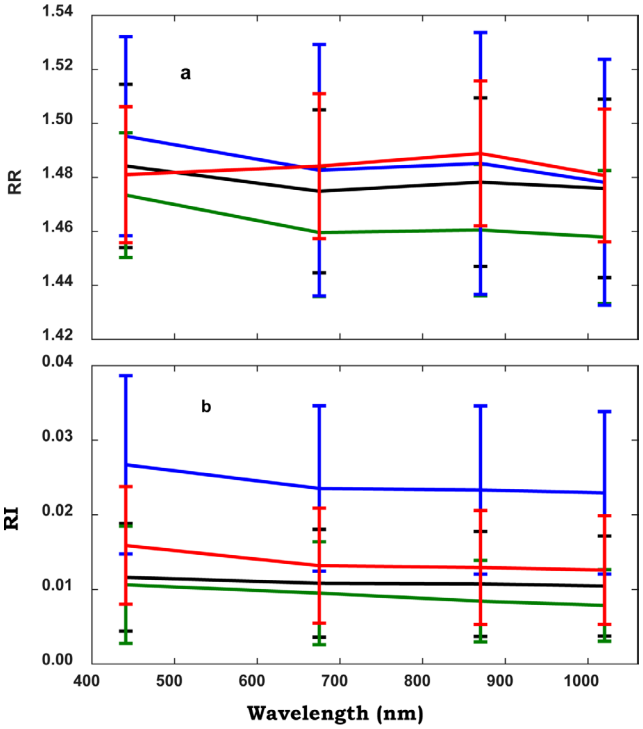

Figure 4: *Seasonal AERONET mean retrieved refractive index (a) real (b) imaginary at four different wavelengths with the standard deviations*

biomass or urban/industrial aerosols with values of 0.837, 0.823, 0.808 and 0.799, respectively. Just like in the winter the spectral line in spring first increased in the lower but later decreased in the longer wavelength having values of 0.871, 0.878, 0.872 and 0.870, respectively.

Previous research found that the effect of the biomass season in southern Africa is more pronounced during the spring season than in the winter (Chiloane et al., 2017) and for Gobabeb this seems to be the same. The combined seasonal effect on SSA, is more pronounced in winter than spring. The effect started from mid-winter in July, culminating in a maximum in August and September, with October and November having a negligible effect. The combined seasonal effect, therefore, is more pronounced in winter than spring.

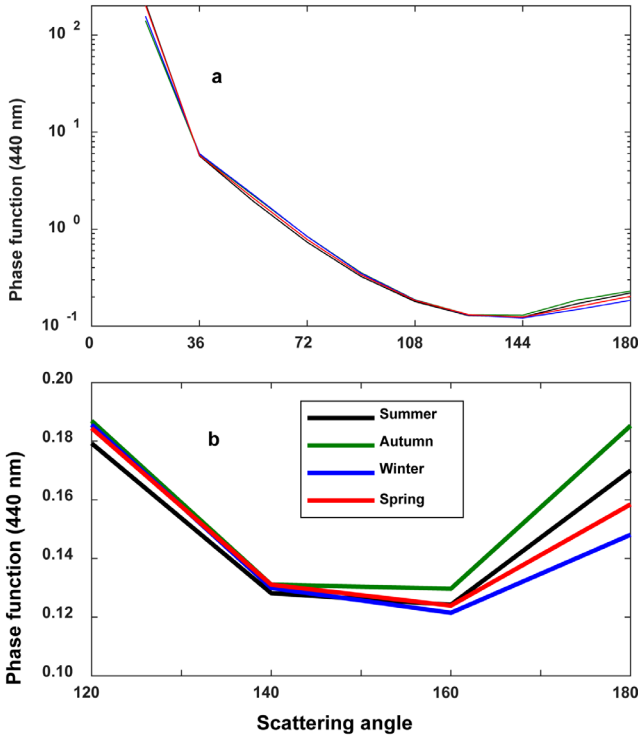

Figure 5: *Seasonal averages of phase function from scattering angle (a) 0–180° and (b) 120–180° at 440 nm wavelength*

Ichoku et al. (2003) recorded 0.88 and 0.84 at visible wavelengths (440 – 675 nm) as the range of SSA in southern Africa, while Queface et al. (2011) showed that at Skukuza it is between 0.91 and 0.89 and for Mongu 0.87 and 0.83, respectively. For Gobabeb, it ranged between 0.88 and 0.87.

Seasonal variability of refractive Index

The refractive index is made of the real part (RR) and the imaginary part (RI). It reflects the ability of aerosols to scatter and absorb incoming radiation. The complex refractive indices for aerosol particles depend on the chemical composition. High values of the RR indicate scattering while high values of the RI indicate absorption. In Figure 4, the seasonal RR and RI for Gobabeb is shown. Comparing the RR and the RI, it can be observed that the RI exhibits larger dependence than the RR. The RR ranges between 1.47 and 1.49 in the shortest wavelength for all seasons but the RI show higher absorption for winter (0.027) followed by the spring (0.016). The high absorption during these seasons may be attributed to the influence of veld fires and open biomass burning.

Seasonal variability of the phase function

Phase function relates to the intensity and angular distribution of the scattering of the incident stream of light by the particles. It is influenced by the internal structure, size distribution, shape of particles and the refractive index (Bi et al., 2011; Bibi et al., 2016). The scattering angle depends on the non-sphericity of the particles. The phase function (Figure 5) is maximum at small angles for all seasons. The minimum value of the phase function occurs at 140° (see Figure 5b), while it is maximum at 0°. More backscatter occurs during the autumn and the summer that shows the predominance of coarse particles during those seasons. Climate forcing estimation and resolution of atmospheric correction problems are linked with phase functions at angles greater than 90° (Bibi et al., 2016; Kokhanovsky, 1998).

Aerosol radiative forcing

Aerosol radiative forcing is defined as the difference between the downward TOA and the upward (surface) fluxes of the short wave radiation with and without aerosols. This quantity is made available through the AERONET inversion code (calculated in the solar spectrum (0.2 – 4.0 μm)) and the assumptions used in calculating it has been extensively discussed in earlier works (Dubovik and King, 2000; Dubovik et al., 2006; García et al., 2011). Earlier works showed that there is a good correlation between SBDART (Ricchiazzi et al., 1998), a software package usually employed in calculating aerosol radiative forcing and AERONET (Adesina et al., 2014; Alam et al., 2012; Ali et al., 2014). García et al. (2011) further suggested that the value recorded for the surface forcing by AERONET is overestimated and needed to be corrected by (1-SA) where SA is the surface albedo. In this paper, we have employed this method to make a correction to the surface forcing. Table 2 shows the seasonal radiative forcing over Gobabeb. The radiative forcing at the surface ranges from –6.46 Wm^{-2} in autumn to –4.00 Wm^{-2} in winter. The TOA ranges from –0.87 Wm^{-2} in spring to 2.95 Wm^{-2} winter. The resultant effect on the atmosphere was minimum in autumn with a value of 3.64 Wm^{-2} and 9.41 Wm^{-2} in winter being maximum. The resultant is a warming effect throughout all the seasons.

Table 2: *Seasonal mean of aerosol radiative forcing over Gobabeb for December 2014 to November 2015*

Season	Surface (Wm^{-2})	TOA (Wm^{-2})	Atmosphere (Wm^{-2})
Summer	-4.89	0.16	5.05
Autumn	-4.00	-0.36	3.64
Winter	-6.46	2.95	9.41
Spring	-4.91	-0.87	4.04

Intercomparison of MODIS AOD and AERONET AOD

Apart from establishing a long term database for climatological studies, intercomparison of satellite sensors with ground-based instruments can aid the improvement of coverage and accuracy deficiency involved in the use of a single sensor (Adesina et al., 2016; Alam et al., 2012; Kang et al., 2016). The datasets from MODIS (Dark target product) that matched the AERONET data for the period under consideration was used. Since both sensors retrieve at different wavelengths, i.e. 550 nm for MODIS and 500 nm for AERONET, both AODs were interpolated to a common wavelength (550 nm) using the power law

$$AOD_{\lambda 2} = AOD_{\lambda 1} \left(\frac{\lambda 2}{\lambda 1}\right)^{-\alpha} \quad (2)$$

Where α is the $\alpha_{440\text{-}870}$ (Alam et al., 2014; Kang et al., 2016; Kumar et al., 2015).

The AOD from both sensors (Figure 6) showed that the MODIS was able to reproduce similar features as that of AERONET despite the fact that it overestimates the ground measurement. Both sensors recorded the lowest values in June 0.05 and 0.11 for AERONET and MODIS, respectively while for both, maximum occurred in September with 0.29 and 0.36, respectively. There was a slight increase in AOD in May for both sensors and then a decrease before it started rising to the maximum in September.

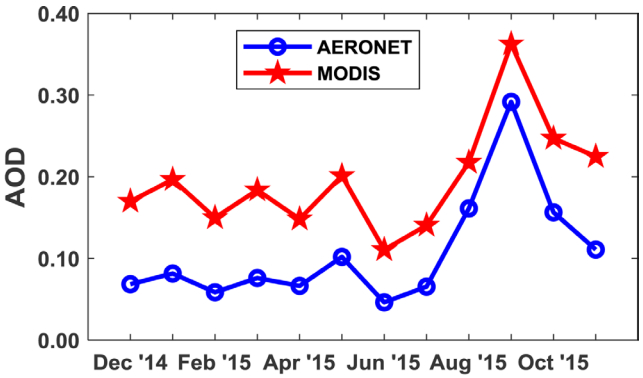

Figure 6: *MODIS and AERONET AOD_{550} monthly variability during the study period*

The intercomparison between the two sensors (Figure 7) was carried out using linear regression of the scatter plots for each season. The slope and intercept of the regression lines are of high importance (Adesina et al., 2016; Alam et al., 2014; Levy et al., 2010; More et al., 2013). The slope shows the degree of the satellite retrieval bias as a result of aerosol model assumptions, pixel choice, instrument calibration or uncertainties associated with large AOD when it departs from unity (Adesina et al., 2016; More et al., 2013; Zhao et al., 2002). The deviation of the intercept from zero is attributed to either incorrect assumptions of the reflectance of the surface when AOD is small or as a result of the difference in terms of the spatial coverage of both sensors. The slopes of both the summer and the autumn indicate that MODIS overestimates AERONET for these seasons. The coefficient of determination for these two seasons was equally very low, i.e. 0.39 and 0.36, respectively. Clouds and fog that are prevalent during these seasons might have contributed to the wide disparity between the two sensors. The slope in winter and spring was similar although the coefficient of determination was much better for spring than in winter. In all seasons, the intercept was negligible showing a good retrieval of the AOD over the location. Ichoku et al. (2003) found that MODIS underestimates AERONET, particularly over Zambian locations, while Kumar et al. (2015) found a good correlation coefficient of 0.78 over Skukuza in South Africa. MODIS and AERONET were also found to have R^2 = 0.72 over Lahore (Gupta et al., 2013), R^2 = 0.76 over Jaipur (Tripathi et al., 2005), and R^2 ranging from 0.61 to 0.76 over four locations on the Indo-Gangetic plains in India (Bibi et al., 2015). The correlation coefficient between MODIS and AERONET is also found to differ in terms of season. Alam et al. (2014) found R^2 = (0.66 and 0.68) for pre-monsoon and post-

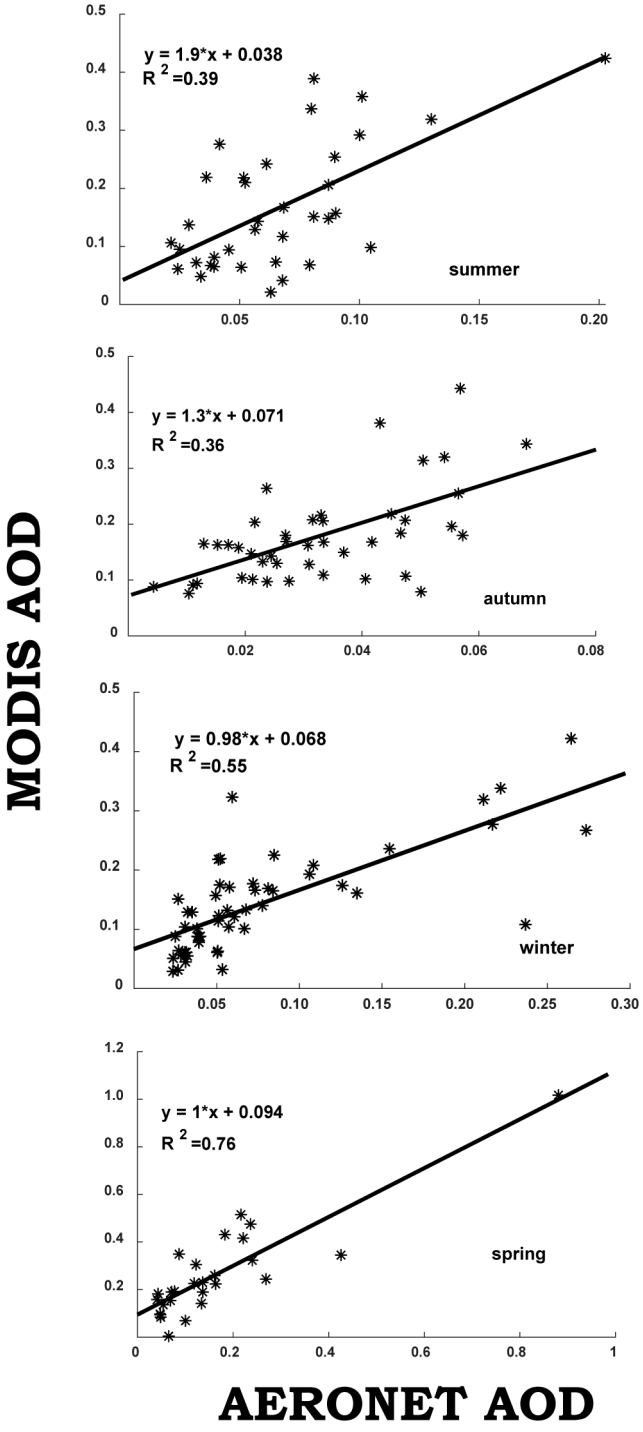

Figure 7: *MODIS and AERONET AOD_{550} monthly variability during the study period*

monsoon seasons over Lahore while Prasad et al. (2007) found R^2 = (0.47 and 0.29) for the winter and summer respectively. A correlation coefficient of 0.76-0.80 was equally found over the Mediterranean basin (Floutsi et al., 2016).

Conclusion

Gobabeb experienced low AOD_{500} from the beginning of the year to July followed by an increase from August to October. It subsequently decreased again until the end of the year. The increase in August to October can be attributed to long-range transport of biomass burning aerosols as a result of atmospheric circulation processes. The high AOD_{500} with corresponding high $\alpha_{440\text{-}870}$ during this period corroborates this. The VSD of aerosols over the location showed various particle sizes of the coarse mode in the different seasons with the largest sizes during the summer. This may be due to the hygroscopic growth of aerosols because of the presence of high water vapor content. The spring season also had an almost equal presence of coarse and fine particles. The SSA showed a strong wavelength dependence (characteristic of biomass burning) during the winter season, while other seasons did not - perhaps due to a mixture of various types of aerosol particles. The imaginary refractive index showed a strong absorption property of aerosols in winter, followed by spring. The phase function showed that particles capable of backscattering are more present in autumn followed by summer, then spring and lowest in winter. This can also point to the prevalence in coarse particles present in these seasons. Aerosol radiative forcing had a heating effect in all seasons with the highest during the winter. An intercomparison between the sunphotometer and the MODIS sensor showed that though MODIS reproduced a similar trend as that of sunphotometer, it overvalues it in all the months. The two instruments correlate better during the spring than all other seasons.

Acknowledgments

Funding for this research is contributed by the Groupement de Recherche International “Atmospheric Research in Southern Africa and the Indian Ocean” (ARSAIO, CNRS/NRF) and the PHC PROTEA (contract n. 863243K) of the NRF and the French Ministries of National Education, of Research and of Foreign Affairs and International Development. The sunphotometer data are from AERONET and the MODIS data used in this paper were produced with the Giovanni online data system, developed and maintained by the NASA GES DISC.

References

Adesina, A. J., Kumar, K. R. and Sivakumar, V. (2015). Variability in aerosol optical properties and radiative forcing over Gorongosa (18.97 oS, 34.35 oE) in Mozambique. Meteorology and Atmospheric Physics, 127(2), 217-228. https://doi.org/10.1007/s00703-014-0352-2

Adesina, A. J., Kumar, K. R., Sivakumar, V. and Griffith, D. (2014). Direct radiative forcing of urban aerosols over Pretoria (25.75 S, 28.28 E) using AERONET Sunphotometer data: First scientific results and environmental impact. Journal of Environmental Sciences, 26(12), 2459-2474. https://doi.org/10.1016/j.jes.2014.04.006

Adesina, A. J., Kumar, K. R., Sivakumar, V. and Piketh, S. J. (2016). Intercomparison and assessment of long-term (2004-2013) multiple satellite aerosol products over two contrasting sites in South Africa. Journal of Atmospheric and Solar-Terrestrial Physics, 148, 82-95. https://doi.org/10.1016/j.jastp.2016.09.001

Alam, K., Sahar, N. and Iqbal, Y. (2014). Aerosol characteristics and radiative forcing during pre-monsoon and post-monsoon seasons in an urban environment. Aerosol Air Qual. Res, 14(1), 99-107. https://doi.org/10.4209/aaqr.2013.05.0154

Alam, K., Trautmann, T., Blaschke, T. and Majid, H. (2012). Aerosol optical and radiative properties during summer and winter seasons over Lahore and Karachi. Atmospheric Environment, 50, 234e245. https://doi.org/10.1016/j.atmosenv.2011.12.027

Ali, M., Tariq, S., Mahmood, K., Daud, A. and Batool, A. (2014). A study of aerosol properties over Lahore (Pakistan) by using AERONET data. Asia-Pacific Journal of Atmospheric Sciences, 50(2), 153-162. https://doi.org/10.1007/s13143-014-0004-y

Bi, J., Huang, J., Fu, Q., Wang, X., Shi, J., Zhang, W., Huang, Z. and Zhang, B. (2011). Toward characterization of the aerosol optical properties over Loess Plateau of Northwestern China. Journal of Quantitative Spectroscopy and Radiative Transfer, 112(2), 346-360. https://doi.org/10.1016/j.jqsrt.2010.09.006

Bibi, H., Alam, K., Blaschke, T., Bibi, S. and Iqbal, M. J. (2016). Long-term (2007-2013) analysis of aerosol optical properties over four locations in the Indo-Gangetic plains. Applied optics, 55(23), 6199-6211. https://doi.org/10.1364/AO.55.006199

Bibi, H., Alam, K., Chishtie, F., Bibi, S., Shahid, I. and Blaschke, T. (2015). Intercomparison of MODIS, MISR, OMI, and CALIPSO aerosol optical depth retrievals for four locations on the Indo-Gangetic plains and validation against AERONET data. Atmospheric Environment, 111, 113-126. https://doi.org/10.1016/j.atmosenv.2015.04.013

Chester, R., Elderfield, H., Griffin, J., Johnson, L. and Padgham, R. (1972). Eolian dust along the eastern margins of the Atlantic Ocean. Marine Geology, 13(2), 91-105. https://doi.org/10.1016/0025-3227(72)90048-5

Chiloane, K. E., Beukes, J. P., Van Zyl, P. G., Maritz, P., Vakkari, V., Josipovic, M., Venter, A. D., Jaars, K., Tiitta, P. and Kulmala, M. (2017). Spatial, temporal and source contribution assessments of black carbon over the northern interior of South Africa. Atmospheric Chemistry and Physics. https://doi.org/10.5194/acp-17-6177-2017

Chung, C. E., Ramanathan, V. and Kiehl, J. T. (2002). Effects of the South Asian absorbing haze on the northeast monsoon and surface-air heat exchange. Journal of Climate, 15(17), 2462-2476. https://doi.org/10.1175/1520-0442(2002)015<2462:EOTSAA>2.0.CO;2

Diner, D., Abdou, W., Bruegge, C., Conel, J., Crean, K., Gaitley, B., Helmlinger, M., Kahn, R., Martonchik, J. and Pilorz, S. (2001). MISR aerosol optical depth retrievals over southern Africa during the SAFARI-2000 dry season campaign. Geophysical Research Letters, 28(16), 3127-3130. https://doi.org/10.1029/2001GL013188

Dubovik, O., Holben, B., Eck, T., Smirnov, A., Kaufman, Y., King, M., Tanre, D. and Slutsker, I. (2002). Climatology of aerosol absorption and optical properties in key worldwide locations. J. Atmos. Sci, 59, 590-608. https://doi.org/10.1175/1520-0469(2002)059<0590:VOAAOP>2.0.CO;2

Dubovik, O. and King, M. D. (2000). A flexible inversion algorithm for retrieval of aerosol optical properties from Sun and sky radiance measurements. Journal of Geophysical Research, 105(D16), 20673-20696. https://doi.org/10.1029/2000JD900282

Dubovik, O., Sinyuk, A., Lapyonok, T., Holben, B. N., Mishchenko, M., Yang, P., Eck, T. F., Volten, H., Muñoz, O. and Veihelmann, B. (2006). Application of spheroid models to account for aerosol particle nonsphericity in remote sensing of desert dust. Journal of Geophysical Research: Atmospheres, 111(D11). https://doi.org/10.1029/2005JD006619

Eck, T., Holben, B., Reid, J., Mukelabai, M., Piketh, S., Torres, O., Jethva, H., Hyer, E., Ward, D. and Dubovik, O. (2013). A seasonal trend of single scattering albedo in southern African biomass-burning particles: Implications for satellite products and estimates of emissions for the world's largest biomass-burning source. Journal of Geophysical Research: Atmospheres, 118(12), 6414-6432. https://doi.org/10.1002/jgrd.50500

Eck, T., Holben, B., Reid, J., O'neill, N., Schafer, J., Dubovik, O., Smirnov, A., Yamasoe, M. and Artaxo, P. (2003b). High aerosol optical depth biomass burning events: A comparison of optical properties for different source regions. Geophysical Research Letters, 30(20). https://doi.org/10.1029/2003GL017861

Eck, T., Holben, B., Ward, D., Mukelabai, M., Dubovik, O., Smirnov, A., Schafer, J., Hsu, N., Piketh, S. and Queface, A. (2003a). Variability of biomass burning aerosol optical characteristics in southern Africa during the SAFARI 2000 dry season campaign and a comparison of single scattering albedo estimates from radiometric measurements. Journal of Geophysical Research: Atmospheres, 108(D13). https://doi.org/10.1029/2002JD002321

Eckardt, F., Soderberg, K., Coop, L., Muller, A., Vickery, K., Grandin, R., Jack, C., Kapalanga, T. and Henschel, J. (2013). The nature of moisture at Gobabeb, in the central Namib Desert. Journal of arid environments, 93, 7-19. https://doi.org/10.1016/j.jaridenv.2012.01.011

Floutsi, A., Korras-Carraca, M., Matsoukas, C., Hatzianastassiou, N. and Biskos, G. (2016). Climatology and trends of aerosol optical depth over the Mediterranean basin during the last 12 years (2002-2014) based on Collection 006 MODIS-Aqua data. Science of the Total Environment, 551, 292-303. https://doi.org/10.1016/j.scitotenv.2016.01.192

García, O., Díaz, J., Expósito, F., Díaz, A., Dubovik, O., Derimian, Y., Dubuisson, P. and Roger, J.-C. (2011). Shortwave radiative forcing and efficiency of key aerosol types using AERONET data. Atmospheric Chemistry and Physics Discussions, 11(12), 32647-32684. https://doi.org/10.5194/acpd-11-32647-2011

Gupta, P., Khan, M. N., da Silva, A. and Patadia, F. (2013). MODIS aerosol optical depth observations over urban areas in Pakistan: quantity and quality of the data for air quality monitoring. Atmospheric pollution research, 4(1), 43-52. https://doi.org/10.5094/APR.2013.005

Hegg, D., Covert, D., Jonsson, H. and Woods, R. (2012). A simple relationship between cloud drop number concentration and precursor aerosol concentration for the regions of Earth's large marine stratocumulus decks. Atmospheric Chemistry and Physics, 12(3), 1229-1238. https://doi.org/10.5194/acp-12-1229-2012

Hersey, S., Garland, R. M., Crosbie, E., Shingler, T., Sorooshian, A., Piketh, S. and Burger, R. (2015). An overview of regional and local characteristics of aerosols in South Africa using satellite, ground, and modeling data. Atmospheric chemistry and physics (Print), 15, 4259. https://doi.org/10.5194/acp-15-4259-2015

Holben, B., Eck, T., Slutsker, I., Tanre, D., Buis, J., Setzer, A., Vermote, E., Reagan, J., Kaufman, Y. and Nakajima, T. (1998). AERONET-A federated instrument network and data archive for aerosol characterization. Remote sensing of environment, 66(1), 1-16. https://doi.org/10.1016/S0034-4257(98)00031-5

Ichoku, C., Remer, L. A., Kaufman, Y. J., Levy, R., Chu, D. A., Tanré, D. and Holben, B. N. (2003). MODIS observation of aerosols and estimation of aerosol radiative forcing over southern Africa during SAFARI 2000. Journal of Geophysical Research: Atmospheres, 108(D13). https://doi.org/10.1029/2002JD002366

Kang, N., Kumar, K. R., Hu, K., Yu, X. and Yin, Y. (2016). Long-term (2002-2014) evolution and trend in Collection 5.1 Level-2 aerosol products derived from the MODIS and MISR sensors over the Chinese Yangtze River Delta. Atmospheric Research, 181, 29-43. https://doi.org/10.1016/j.atmosres.2016.06.008

Kaufman, Y., Tanré, D., Remer, L. A., Vermote, E., Chu, A. and Holben, B. (1997). Operational remote sensing of tropospheric aerosol over land from EOS moderate resolution imaging spectroradiometer. Journal of Geophysical Research: Atmospheres, 102(D14), 17051-17067. https://doi.org/10.1029/96JD03988

King, M. D., Menzel, W. P., Kaufman, Y. J., Tanré, D., Gao, B.-C., Platnick, S., Ackerman, S. A., Remer, L. A., Pincus, R. and Hubanks, P. A. (2003). Cloud and aerosol properties, precipitable water, and profiles of temperature and water vapor from MODIS.

IEEE Transactions on Geoscience and Remote Sensing, 41(2), 442-458. https://doi.org/10.1109/TGRS.2002.808226

Kokhanovsky, A. (1998). Variability of the phase function of atmospheric aerosols at large scattering angles. Journal of the Atmospheric Sciences, 55(3), 314-320. https://doi.org/10.1175/1520-0469(1998)055<0314:VOTPFO>2.0.CO;2

Kumar, K. R., Sivakumar, V., Reddy, R., Gopal, K. R. and Adesina, A. J. (2013). Inferring wavelength dependence of AOD and Ångström exponent over a sub-tropical station in South Africa using AERONET data: Influence of meteorology, long-range transport and curvature effect. Science of the Total Environment, 461, 397-408. https://doi.org/10.1016/j.scitotenv.2013.04.095

Kumar, K. R., Yin, Y., Sivakumar, V., Kang, N., Yu, X., Diao, Y., Adesina, A. J. and Reddy, R. (2015). Aerosol climatology and discrimination of aerosol types retrieved from MODIS, MISR and OMI over Durban (29.88 S, 31.02 E), South Africa. Atmospheric Environment, 117, 9-18. https://doi.org/10.1016/j.atmosenv.2015.06.058

Laakso, L., Vakkari, V., Virkkula, A., Laakso, H., Backman, J., Kulmala, M., Beukes, J., Van Zyl, P., Tiitta, P. and Josipovic, M. (2012). South African EUCAARI measurements: seasonal variation of trace gases and aerosol optical properties. Atmospheric Chemistry and Physics, 12(4), 1847-1864. https://doi.org/10.5194/acp-12-1847-2012

Levy, R., Remer, L., Kleidman, R., Mattoo, S., Ichoku, C., Kahn, R. and Eck, T. (2010). Global evaluation of the Collection 5 MODIS dark-target aerosol products over land. Atmospheric Chemistry and Physics, 10(21), 10399-10420. https://doi.org/10.5194/acp-10-10399-2010

Levy, R. C., Remer, L., Mattoo, S., Vermote, E. and Kaufman, Y. (2007). Second-generation algorithm for retrieving aerosol properties over land from MODIS spectral reflectance. J. Geophys. Res, 112(D13), D13. https://doi.org/10.1029/2006JD007815

Li, J., Carlson, B. E., Dubovik, O. and Lacis, A. A. (2014). Recent trends in aerosol optical properties derived from AERONET measurements. Atmospheric Chemistry and Physics, 14(22), 12271-12289. https://doi.org/10.5194/acp-14-12271-2014

Losno, R., Bergametti, G. and Carlier, P. (1992). Origins of atmospheric particulate matter over the North Sea and the Atlantic Ocean. Journal of atmospheric chemistry, 15(3-4), 333-352. https://doi.org/10.1007/BF00115403

More, S., Pradeep Kumar, P., Gupta, P., Devara, P. and Aher, G. (2013). Comparison of aerosol products retrieved from AERONET, MICROTOPS and MODIS over a tropical urban city, Pune, India. Aerosol and air quality research, 13(1), 107-121. https://doi.org/10.4209/aaqr.2012.04.0102

Pawar, G., Devara, P. and Aher, G. (2015). Identification of aerosol types over an urban site based on air-mass trajectory classification. Atmospheric Research, 164, 142-155. https://doi.org/10.1016/j.atmosres.2015.04.022

Piketh, S., Annegarn, H. and Tyson, P. (1999). Lower tropospheric aerosol loadings over South Africa: The relative contribution of aeolian dust, industrial emissions, and biomass burning. Journal of Geophysical Research: Atmospheres, 104(D1), 1597-1607. https://doi.org/10.1029/1998JD100014

Platnick, S., King, M., Hobbs, P., Osborne, S., Piketh, S. and Ruinjes, R. (2001). Cloud and Radiation Studies during SAFARI 2000. Paper presented at the AGU Fall Meeting Abstracts.

Prasad, A. K., Singh, S., Chauhan, S., Srivastava, M. K., Singh, R. P. and Singh, R. (2007). Aerosol radiative forcing over the Indo-Gangetic plains during major dust storms. Atmospheric Environment, 41(29), 6289-6301. https://doi.org/10.1016/j.atmosenv.2007.03.060

Queface, A. J., Piketh, S. J., Eck, T. F., Tsay, S.-C. and Mavume, A. F. (2011). Climatology of aerosol optical properties in Southern Africa. Atmospheric Environment, 45(17), 2910-2921. https://doi.org/10.1016/j.atmosenv.2011.01.056

Remer, L. A., Kaufman, Y., Tanré, D., Mattoo, S., Chu, D., Martins, J. V., Li, R.-R., Ichoku, C., Levy, R. and Kleidman, R. (2005). The MODIS aerosol algorithm, products, and validation. Journal of the Atmospheric Sciences, 62(4), 947-973. https://doi.org/10.1175/JAS3385.1

Remer, L. A., Kleidman, R. G., Levy, R. C., Kaufman, Y. J., Tanré, D., Mattoo, S., Martins, J. V., Ichoku, C., Koren, I. and Yu, H. (2008). Global aerosol climatology from the MODIS satellite sensors. Journal of Geophysical Research: Atmospheres, 113(D14). https://doi.org/10.1029/2007JD009661

Ricchiazzi, P., Yang, S., Gautier, C. and Sowle, D. (1998). SBDART: A research and teaching software tool for plane-parallel radiative transfer in the Earth's atmosphere. Bulletin of the American Meteorological Society, 79(10), 2101-2114. https://doi.org/10.1175/1520-0477(1998)079<2101:SARATS>2.0.CO;2

Sreekanth, V. and Kulkarni, P. (2013). Spatio-temporal variations in columnar aerosol optical properties over Bay of Bengal: Signatures of elevated dust. Atmospheric Environment, 69, 249-257. https://doi.org/10.1016/j.atmosenv.2012.12.031

Stocker, T., Qin, D., Plattner, G., Tignor, M., Allen, S., Boschung, J., Nauels, A., Xia, Y., Bex, B. and Midgley, B. (2013). IPCC, 2013: climate change 2013: the physical science basis. Contribution of working group I to the fifth assessment report of the intergovernmental panel on climate change.

Swap, R., Garstang, M., Macko, S., Tyson, P., Maenhaut, W., Artaxo, P., Kållberg, P. and Talbot, R. (1996). The long-range transport of

southern African aerosols to the tropical South Atlantic. Journal of Geophysical Research: Atmospheres, 101(D19), 23777-23791. https://doi.org/10.1029/95JD01049

Tanré, D., Kaufman, Y., Herman, M. and Mattoo, S. (1997). Remote sensing of aerosol properties over oceans using the MODIS/EOS spectral radiances. Journal of Geophysical Research: Atmospheres, 102(D14), 16971-16988. https://doi.org/10.1029/96JD03437

Tripathi, S., Dey, S., Chandel, A., Srivastava, S., Singh, R. P. and Holben, B. (2005). Comparison of MODIS and AERONET derived aerosol optical depth over the Ganga Basin, India. Paper presented at the Annales Geophysicae. https://doi.org/10.5194/angeo-23-1093-2005

Tyson, P. D. and Preston-Whyte, R. A. (2000). weather and climate of southern Africa: Oxford University Press.

Vakkari, V., Beukes, J. P., Dal Maso, M., Aurela, M., Josipovic, M. and van Zyl, P. G. (2018). Major secondary aerosol formation in southern African open biomass burning plumes. Nature Geoscience, 11(8), 580. https://doi.org/10.1038/s41561-018-0170-0

Zhao, T. X., Stowe, L. L., Smirnov, A., Crosby, D., Sapper, J. and McClain, C. R. (2002). Development of a global validation package for satellite oceanic aerosol optical thickness retrieval based on AERONET observations and its application to NOAA/NESDIS operational aerosol retrievals. Journal of the Atmospheric Sciences, 59(3), 294-312. https://doi.org/10.1175/1520-0469(2002)059<0294:DOAGVP>2.0.CO;2

Zhou, X., Ackerman, A. S., Fridlind, A. M., Wood, R. and Kollias, P. (2017). Impacts of solar-absorbing aerosol layers on the transition of stratocumulus to trade cumulus clouds. Atmospheric Chemistry and Physics, 17(20), 12725-12742. https://doi.org/10.5194/acp-17-12725-2017

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Research article

A decadal analysis of particulate matter ($PM_{2.5}$) and surface ozone (O_3) over Vaal Priority Area, South Africa

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Received: 17 July 2019 - **Reviewed:** 10 September 2019 - **Accepted:** 4 November 2019

https://doi.org/10.17159/caj/2019/29/2.7578

Abstract

Atmospheric pollutants that affect human health most significantly are particulate matter ($PM_{2.5}$) and surface ozone (O_3). This paper analysed the long-term temporal trends for $PM_{2.5}$ and ground level O_3 for six air quality monitoring stations in the Vaal Triangle Area of South Africa from 2007 to 2017. Research has been conducted on the short-term temporal trends for $PM_{2.5}$ concentration and surface O_3 concentrations. There are no studies that have focussed on the long-term temporal trends for $PM_{2.5}$ and O_3 in the Vaal Triangle Area of South Africa, because these air quality monitoring stations have only existed for a period of approximately 11 years. The data used in this study is derived from ground-based instruments from the South African Weather Service. Temporal patterns for time of day, days of the week, and seasons were observed for all air quality stations. $PM_{2.5}$ concentration increased during early mornings and late afternoons, with higher concentration during weekdays than weekends and an increase from late winter through to spring and summer. Surface O_3 concentrations peaked during the spring and summer months and during midday when there was maximum sunlight acting as a catalyst for photochemical reactions. The long term trends illustrated that there has been no significant decrease in annual average concentration for $PM_{2.5}$ in four of the six stations and surface O_3 for the six stations in the past 10 years in the Vaal Triangle Area of South Africa.

Keywords

Particulate matter; Tropospheric ozone; Long term temporal trends; Human health; Mortality; Morbidity

Introduction

A changing climate, anthropogenic and natural emissions, and meteorological variables are key drivers which influence the concentration as well as dispersion of air pollutants on a spatial and temporal scale (Silva et al., 2016). Climate change has an impact on air pollution by changing the amount, intensity and the timing of extreme heat events, air mass movements, rainfall and other meteorological occurrences that influence pollutant concentration (Fiore et al., 2015). Meteorological factors influence the concentration of air pollution at the source through dispersion, dilution, chemical transportation over large areas as well as dry and wet deposition (Kinney, 2018). Climate change has an impact on air quality and contrariwise air quality affects climate change and this may have a negative impact on human health (Orru et al., 2017, Fiore et al., 2015). Being exposed to ambient, surface level particulate matter with an aerodynamic diameter of less than 2.5 μgm^{-3} ($PM_{2.5}$) and ozone (O_3) has also been shown to have an impact on early mortality rates and morbidity (Tshehla et al., 2019; Kinney,2018).

$PM_{2.5}$ has a short lifespan of a few days in the atmosphere and is formed from a variety of sources resulting in global temporal and spatial heterogeneity (Ramanthan et al., 2001). $PM_{2.5}$ originates from primary sources of emissions or secondary sources when gases react in the atmosphere to form $PM_{2.5}$ (Kinney, 2008). The majority of sources of $PM_{2.5}$ are fuel combustion by motor vehicles, furnaces and power plants, mining, windblown dust

and household combustion and biomass burning (Fuzzi et al., 2015; Karagulian et al., 2015; Kinney, 2018). $PM_{2.5}$ can exacerbate pre-existing cardiopulmonary disease and has been associated with cases of lung cancer mortality (Villa et al., 2016; Melamed et al., 2016; Lehtomäki et al., 2018). Chronic exposure to $PM_{2.5}$ is responsible for untimely deaths (Silva et al., 2016, Apte et al., 2018).

Tropospheric ozone is a greenhouse gas that has a negative effect on human health and the environment. The formation of surface ozone occurs in the presence of high ambient temperatures and sunlight (Kinney, 2018). Ozone is a secondary pollutant that is formed as a result of precursors (which are volatile organic compounds (VOCs) and nitrogen oxides (NO_x)) reacting with sunlight. The concentration of ozone is influenced by the amount of sunlight for photochemical reactions, dry deposition, and precursors' concentrations, which in turn is affected by industrial and vehicle emissions. The regional transport of ozone is influenced by meteorological conditions (Gao et al., 2017, Monks et al., 2015). This has led to O_3 being monitored in the troposphere because ozone has a harmful effect on people and ecosystems (Monks et al., 2015; Derwent et al., 2018,). It is therefore imperative to study how ozone is being formed in a particular region over a long period of time. Through the analysis of O_3 concentrations, we are able to detect if the people living in close proximity to the Vaal Triangle Air-Shed Priority Area (VTAPA) are being chronically exposed to high levels of ozone that impact negatively on human health.

The aim of this study was to investigate the temporal trends in concentrations of $PM_{2.5}$ and ground-level O_3 for a period of 10 years from 2007 to 2017 for six air quality monitoring stations in the VTAPA and compare these results with the limit for acceptable concentrations set by the National Ambient Air Quality Standard (NAAQS) for South Africa. The VTAPA was declared a National Priority Area in 2006. Studies that informed the declaration of the VTAPA were short-term studies that used passive samplers to collect data over short periods of time for outdoor or indoor air pollutants and the concentrations were averaged over months, which does not take into consideration trends for day and night (Terblanche et al., 1993; van Horen et al., 1993; Scorgie et al., 2003, Zunckel, 2004 and Shezi et al., 2018). A new project to compile an emission inventory has been commissioned by the Department of Environment Forestry and Fisheries (DEFF) using 2011 census data, 2016 community survey data for the VTAPA and air quality data from 2007-2017(South African Department of Environment, Forestry and Fisheries, 2019).

The driving forces that contribute to elevated $PM_{2.5}$ and surface O_3 concentrations need to be understood and therefore long-

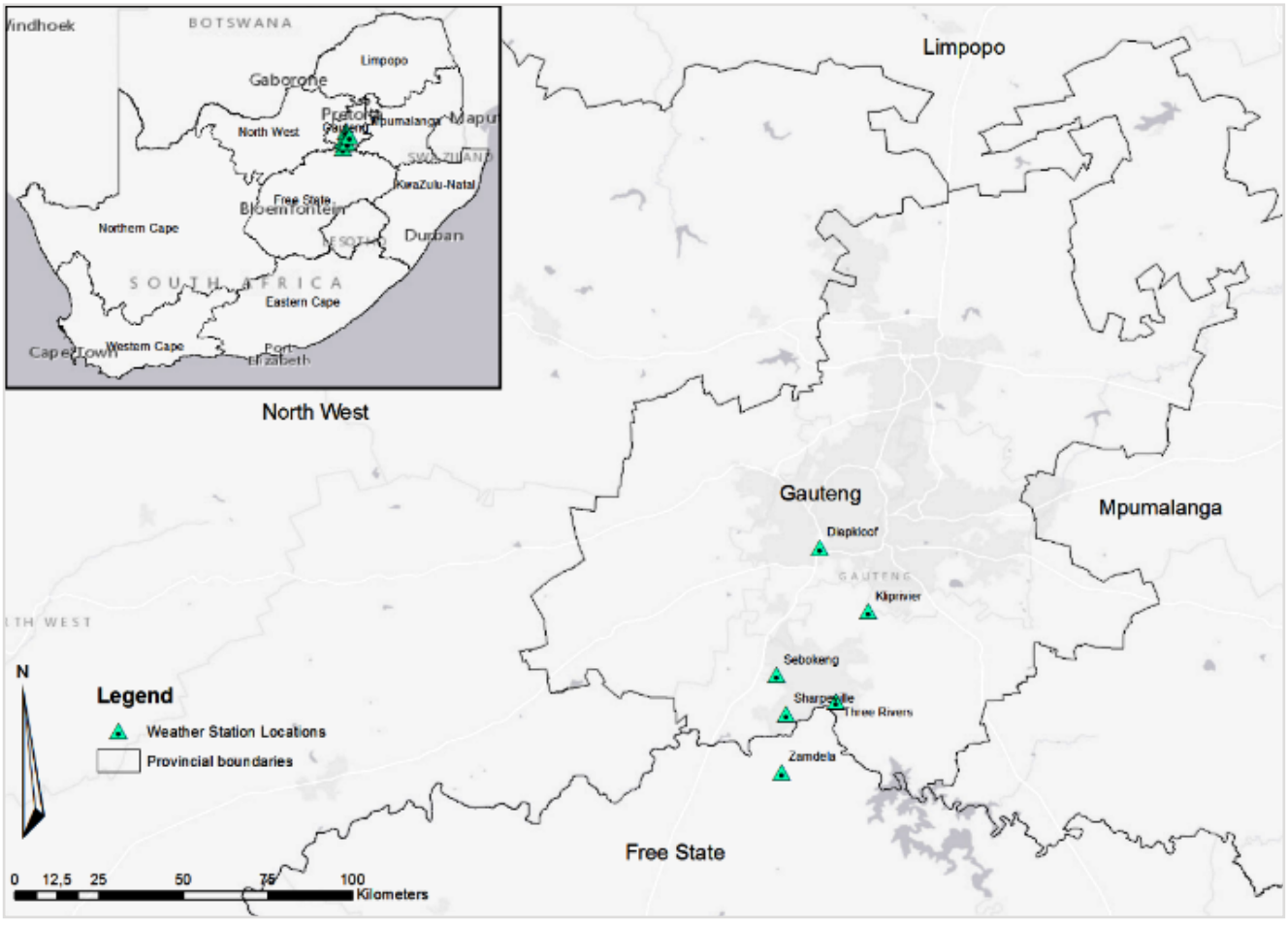

Figure 1: *Geographical map of the selected six air quality monitoring stations in the Vaal Triangle Airshed Priority Area of South Africa.*

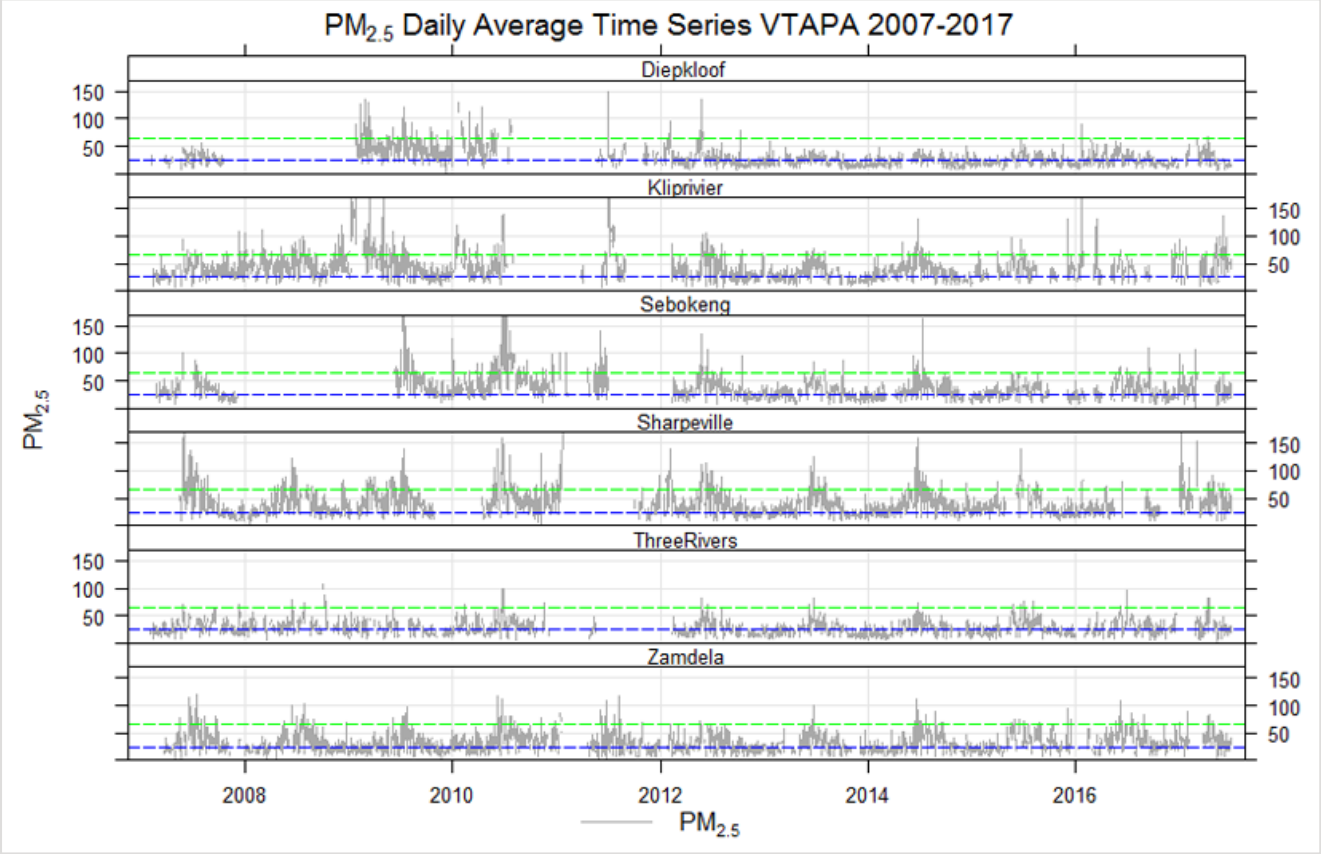

Figure 2: *Daily Average of $PM_{2.5}$ concentration of hourly data for February 2007 to June 2017*

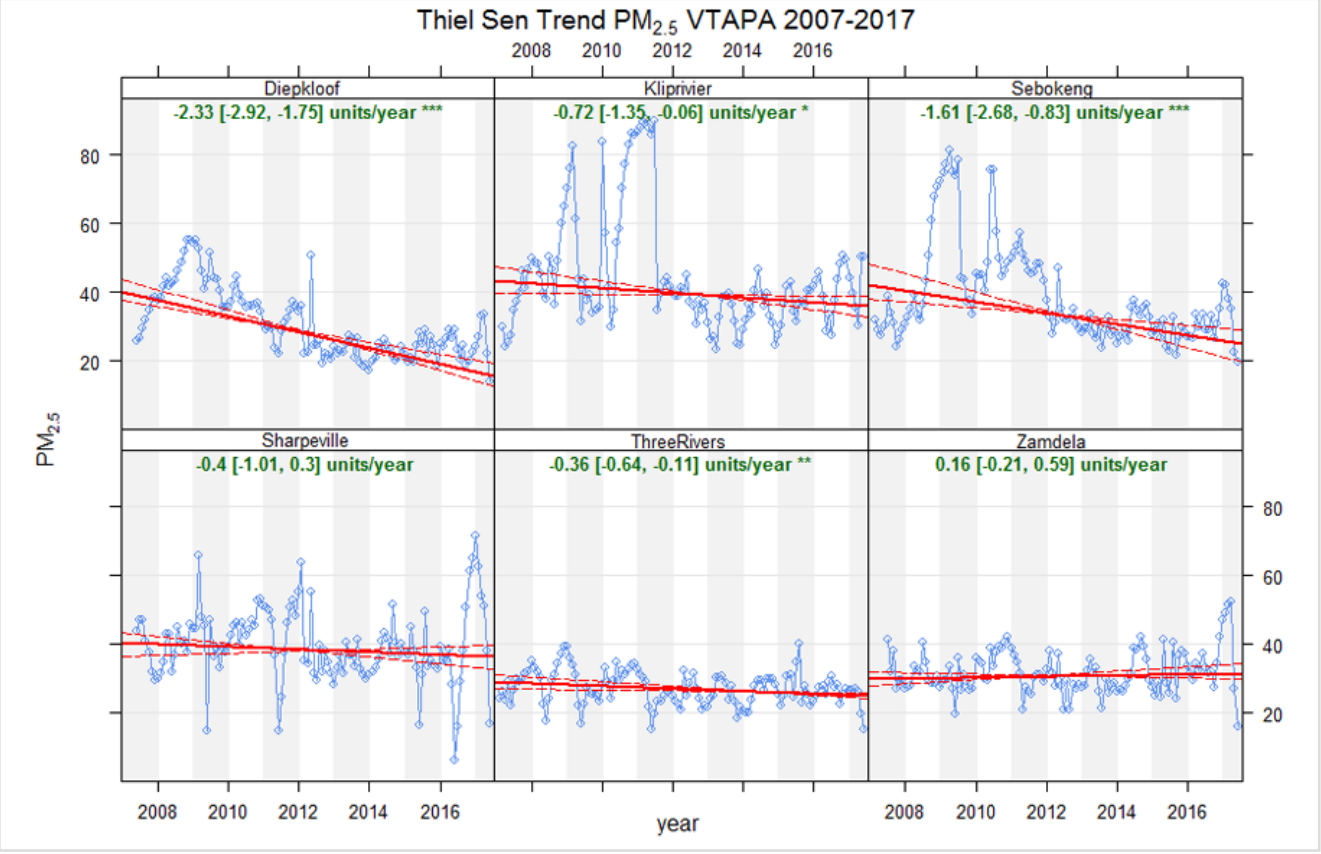

Figure 3: *Mann Kendall and Theil-Sen trend analysis for $PM_{2.5}$ concentrations for 2007-2017*

term observation studies are crucial to determine the long term trends of these pollutants. The VTAPA is located in the Gauteng and Free State Provinces of South Africa. The data used in this paper is secondary data acquired from the South African Weather Service (SAWS). The data is collected by the DEFF monitoring network.

A similar study for long term observations of spatial and temporal patterns was conducted in Richards Bay for PM_{10} and SO_2 (Okello et al., 2018), but our current study is focussed on an inland region i.e. the VTAPA, and considers $PM_{2.5}$ and O_3.

Study site

The air quality monitoring stations for the VTAPA are Diepkloof, Kliprivier, Sebokeng, Sharpeville, Three Rivers and Zamdela. The VTAPA is an area that has mining operations, metal and steel plants, collieries, a coal-fired power station and petrochemical industries that contribute to dust, gas and particulate emission (Scorgie et al., 2003; Feig et al., 2015). Domestic fuel burning is an important source for space heating and cooking as this is an affordable source of energy in the VTAPA (Tshehla et al., 2019). In South Africa, summer months are from December to February, March to May is autumn, June to August is winter, and September to November is spring. Figure 1 below illustrates the geographical positions of the air quality stations in the VTAPA in South Africa. The exact geolocation is tabulated in the Table 1.

Table 1: *Air quality monitoring stations' location*

Name of Station	Latitude	Longitude
Diepkloof	-26.250733	27.9564167
Sebokeng	-26.587805	27.84022
Sharpeville	-26.689833	27.86775
Zamdela	-26.844889	27.8551111
Three Rivers	-26.658306	27.99822
Kliprivier	-26.42033	28.084889

Data and Methodology

Ground based observations for hourly concentrations of $PM_{2.5}$, and hourly as well as 8-hour rolling concentrations for tropospheric O_3 were compared and analysed for decadal data from 1 February 2007 to 31 June 2017 for six stations that are part of the VTAPA.

The maintenance of each monitoring station is crucial to ensure that good quality data is produced and it meets the requirements for air quality stations by the South African National Accreditation System (SANAS). Data needs to be supplied for 90% of the monitoring period prior to validation checks being applied to the data (SANAS, 2013). The calibration frequency for each monitoring station occurs on a quarterly basis with a minimum of two weekly checks using a precise known concentration for individual gas analysers. One of the four quarterly calibrations should be undertaken by an accredited calibration laboratory (SANAS, 2013).

Statistical analyses were conducted using Open Air Package in R version 3.4.4 (Carslaw et al., 2012). The data was cleaned to remove negative values and repeated values for hourly data. Data availability at each monitoring station was greater than 60% for each year after data was recovered and quality checked.

The hourly data was used to plot the daily average time series for each station in figure 2 to visually illustrate the $PM_{2.5}$ daily averages in comparison to daily NAAQS and World Health Organisation (WHO) limit of 65 μgm^{-3} and 25 μgm^{-3}, respectively (World Health Organisation, 2006; NAAQS, 2009) The calendar plot in R was plotted for each station to determine the frequency of exceedance for each day and the results are tabulated in table 2. The annual average concentration for $PM_{2.5}$ was calculated for all six stations for each of the 10 years during 2007-2017 using R and is presented in table 3.

The time variation plots, averaged by hours of the day, days of the week and months of the year for each pollutant, were created for each of the six stations. The purpose of this exercise was to determine if the levels of pollutants differ during working days (Monday to Friday) and weekends (Saturday and Sunday). It could also identify the consistency of pollutant levels during working days if there are emission from industries and factories surrounded by the selection of stations. An advantage of considering time variation plots is that the source of the pollutant is more likely to be inferred (Faridi et al., 2018). Emissions of certain sources are more pronounced during a particular time of day or during a particular season because of varying reasons such as people use more fuels during winter for space heating and this leads to an increase in emissions of particulate matter or meteorological conditions (Scorgie et al., 2003).

The long term trends in $PM_{2.5}$ and O_3 were analysed using the Mann Kendall and Theil-Sen calculations for 2007-2017 for each of the six stations. The Mann Kendall and Theil-Sen calculations (Carslaw et al., 2012) were used to determine if there is a significant downward or upward trend over the 10 years for the concentrations of $PM_{2.5}$ and O_3. The symbols in the graph in figure 3 and figure 7 indicate the statistical significance with $p < 0.001$ = ***, $p < 0.01$ = **, $p < 0.05$ = * and $p < 0.1$ = +.

Results and discussion

Particulate Matter ($PM_{2.5}$)

The frequency of allowable exceedances of the limit value for $PM_{2.5}$ per year is 4 according to the NAAQS. Results in figure 2 and table 2 below illustrate that most of the stations have more exceedances per year than the NAAQS limit of 4, with Kliprivier having the most amount of exceedances over the last 10 years. For the past five years the Diepkloof monitoring station has been compliant with the NAAQS limit as the sources of emissions at Diepkloof are from household combustion and very limited industrial activities (Feig et al., 2015).

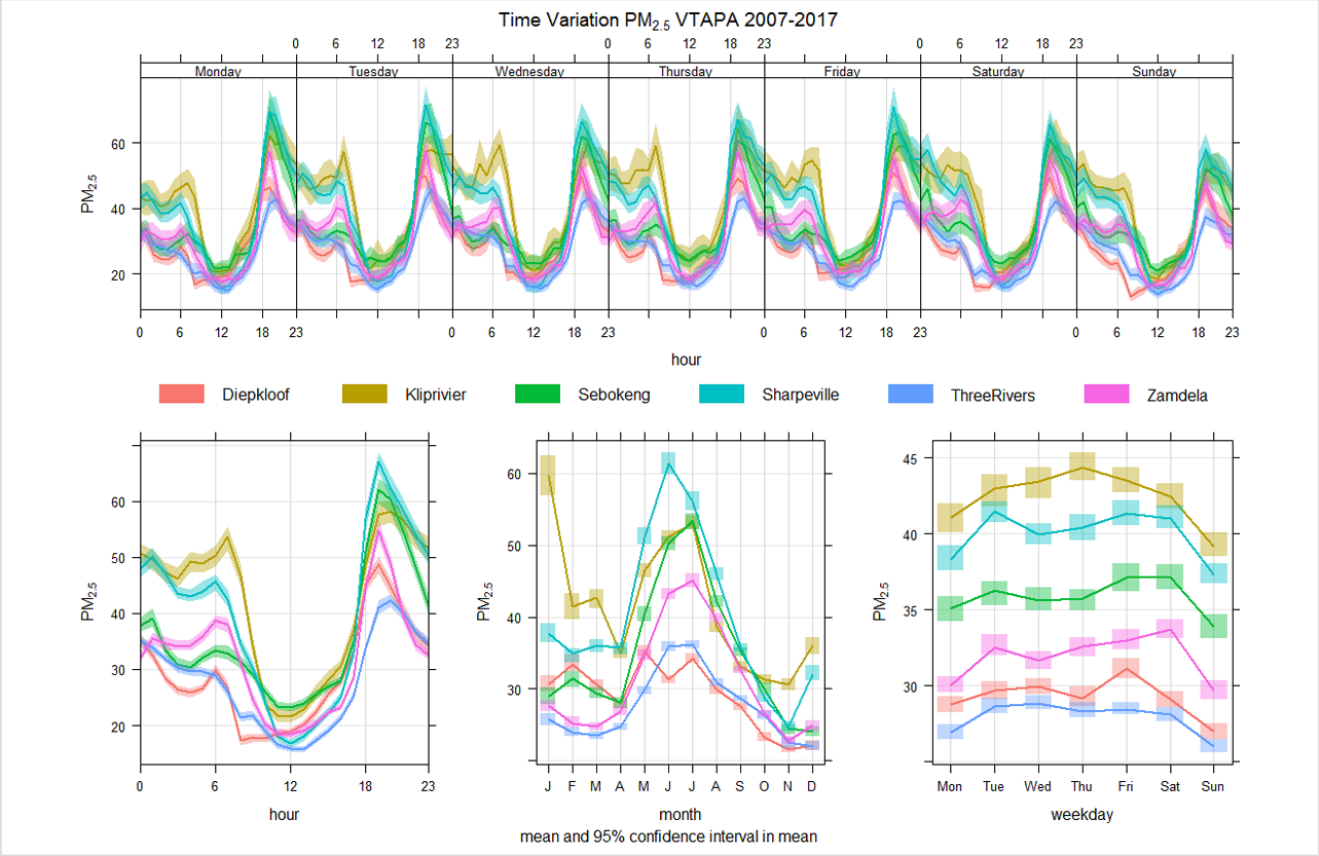

Figure 4: *Time variation plot for daily average of $PM_{2.5}$ concentration of hourly data for February 2007 to June 2017*

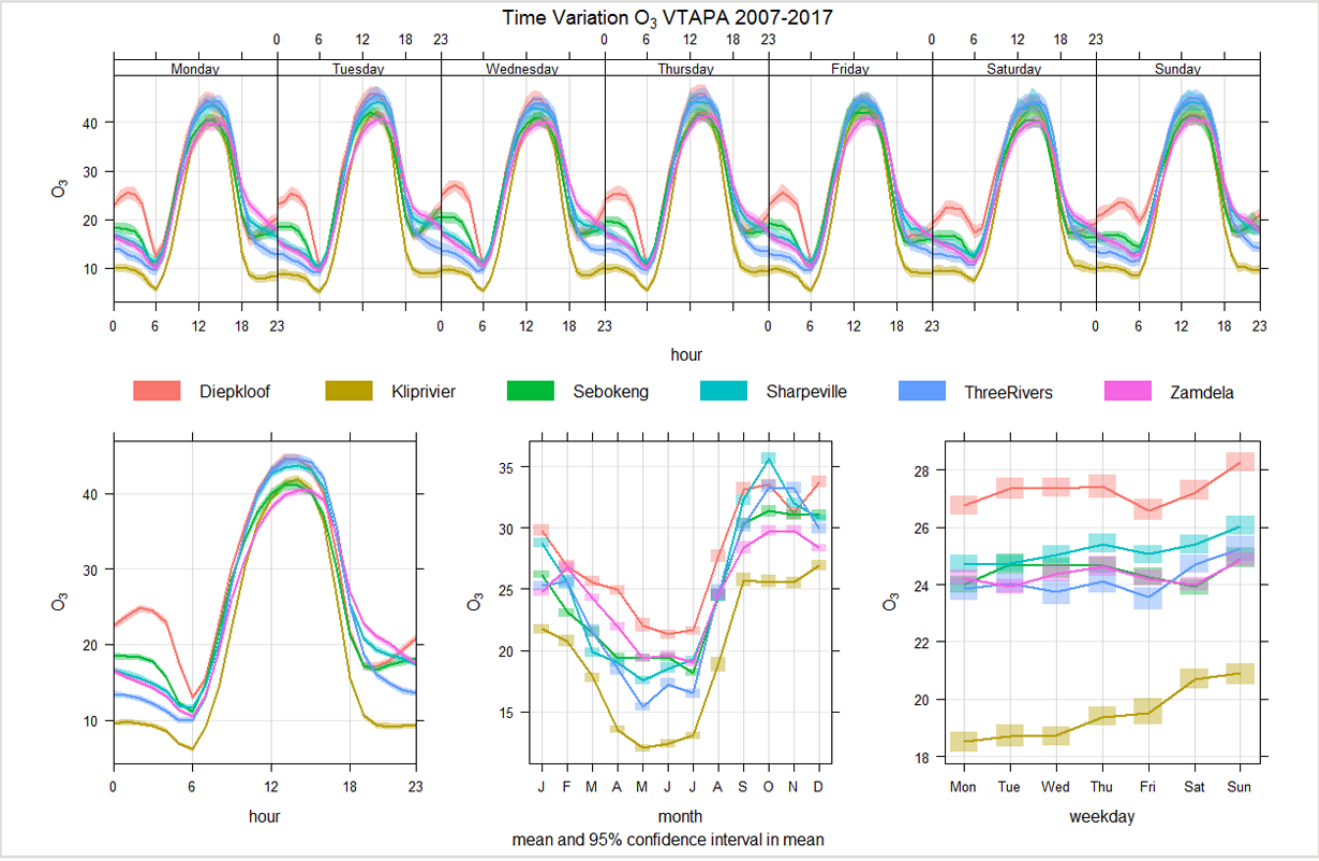

Figure 5: *O_3 time variation plot for daily average of concentration of hourly data for February 2007 to June 2017*

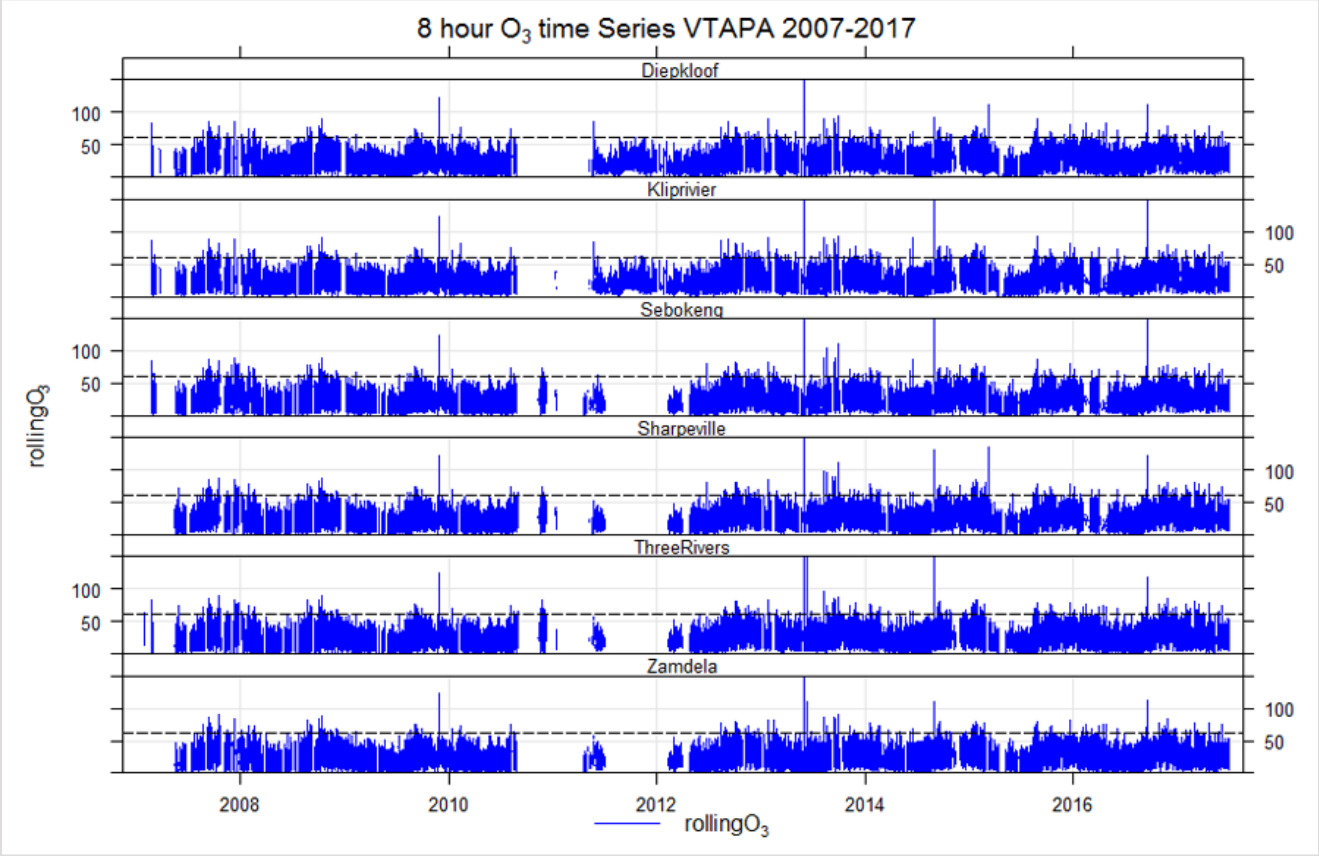

Figure 6: *Time Series Plot illustrating the 8-hourly O_3 concentration for the period February 2007 to June 2017*

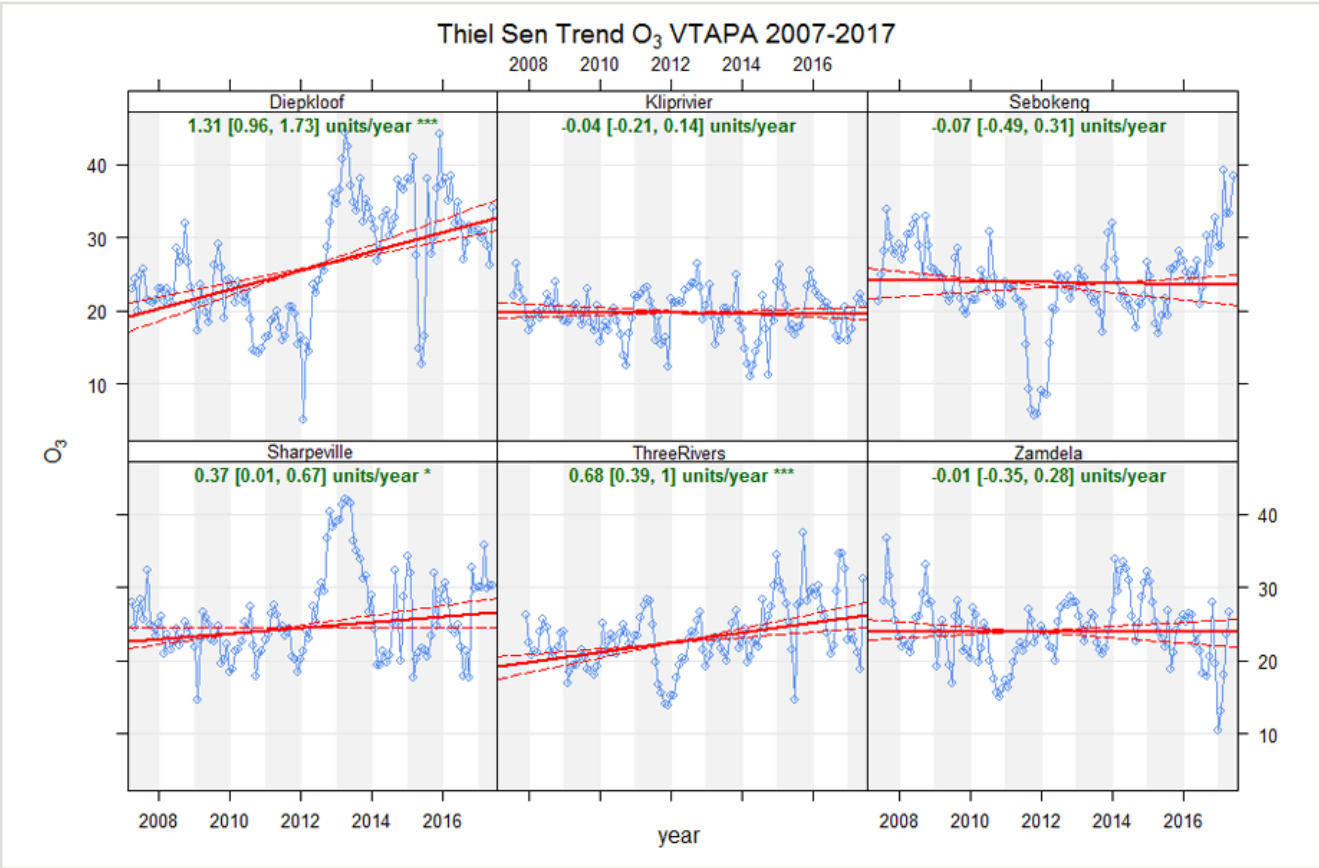

Figure 7: *Mann Kendall and Thielsen for O_3 concentrations for 2007-2017*

Table 2: *Frequency of Daily $PM_{2.5}$ Exceedances for the VTAPA*

Year	Klipriver	Sharpeville	Sebokeng	Three Rivers	Zamdela	Diepkloof
2007	49	10	10	3	25	0
2008	No Data	No Data	No Data	11	17	No Data
2009	84	33	32	5	7	41
2010	49	78	18	13	20	50
2011	30	24	24	1	18	1
2012	28	18	18	3	8	11
2013	11	7	7	2	5	0
2014	21	10	10	1	16	0
2015	19	1	1	5	11	0
2016	39	16	15	2	20	2
2017	38	5	5	2	10	2
Totals	368	202	140	48	157	107

Table 3: *Annual Average Concentration of $PM_{2.5}$ (μgm^{-3}) for the VTAPA*

Year	Klipriver	Sharpeville	Sebokeng	Three Rivers	Zamdela	Diepkloof
2007	36.9	45.4	32.3	28.7	36.5	27.5
2008	47.3	37.4	NaN	32.0	31.2	NaN
2009	54.0	44.0	46.7	27.2	28.3	45.0
2010	52.2	48.9	53.0	32.5	36.4	58.5
2011	57.9	42.6	55.6	24.4	31.6	30.2
2012	37.6	39.5	33.6	26.0	29.1	27.0
2013	30.8	34.8	29.0	24.4	29.6	23.0
2014	35.5	38.5	30.8	25.6	30.0	22.6
2015	38.6	35.8	28.5	27.6	30.7	23.3
2016	44.9	31.2	31.2	28.6	35.2	24.5
2017	49.8	49.4	34.3	27.9	37.1	26.1

The NAAQS annual mean concentration limit for $PM_{2.5}$ is $25\mu gm^{-3}$. The annual average concentration limit for $PM_{2.5}$ has been exceeded for every year from 2007-2017 for Kliprivier, Sharpeville, Sebokeng and Zamdela stations. Diepkloof had 6 exceedances years during 2007-2017 for $PM_{2.5}$ annual average concentration in comparison to NAAQS limit for annual average concentration. At Diepkloof there are not many heavy industries and no major mines in close proximity (Feig et al., 2015).

The Mann Kendall and Theil-Sen calculations computed in R indicate the monthly mean de-seasonalised trend analysis for the six stations for a period of 10 years (figure 3). There is a downward trend in the yearly concentration of $PM_{2.5}$ for Diepkloof and Sebokeng which is highly significant ($p < 0.001$) and a slightly significant declining trend at the level of 0,01 at Three Rivers. At Kliprivier the decreasing trend was statistically significant at the level of 0,05. At Sharpeville the declining trend remained the same for each of the 10 years. The concentration of $PM_{2.5}$ at Zamdela was increasing slightly for each year but it is not statistically significant.

The diurnal variation graph (Figure 4) illustrates the temporal variation of $PM_{2.5}$ concentrations to infer the sources of the $PM_{2.5}$. The analysis was done for weekdays and week-ends to determine whether emissions are greater over weekdays or week-ends and what is contributing to the differing levels of emissions during these times. The diurnal variation for $PM_{2.5}$ concentration for weekdays and week-ends is consistent across the six air quality stations for the VTAPA. The temporal trend of $PM_{2.5}$ concentrations across all six stations peaks in the early morning and in the late afternoon. The peaks in $PM_{2.5}$ concentration correspond to the peaks in domestic use of fuel for heating and cooking during 6 am to 7 am and 6 pm to 7 pm and also corresponds to high volumes of traffic that contributes to fine dust from the roads becoming airborne (South African Department of Environment, Forestry and Fisheries; 2019). Dispersion is inhibited due to adverse conditions in meteorology and this is another factor that contributes to the morning and evening peaks in $PM_{2.5}$ concentrations (de Lange et al., 2019). The decrease in $PM_{2.5}$ concentrations during daylight is a result of greater mixing in the atmosphere because the planetary boundary layer is broken up (de Lange et al., 2019).

The monthly average $PM_{2.5}$ concentrations are highest during the winter months for all the stations and this is likely because of biomass burning that normally takes place in winter and early spring in Southern Africa (Laban et al.,2015) and burning biofuels to keep warm during winter (Tshehla et al., 2019). Another factor that affects the winter $PM_{2.5}$ concentrations is the meteorological conditions (de Lange et al.,2019). The atmospheric concentration of $PM_{2.5}$ is affected by dispersion and wet deposition (Veechi.,2004 and de Lange et al.,2019). A study conducted by the DEFF at Sharpeville for 5 days during winter found that 68% of active fires were as a result of domestic burning (South African Department of Environment, Forestry and Fisheries; 2019). During the weekdays the concentration of $PM_{2.5}$ has a slightly higher peak than week-ends. The decrease in $PM_{2.5}$ concentrations over weekends can be attributed to less traffic resulting in less airborne dust that is similar to trends found in other studies (Tan et al., 2013 and Faridi et al., 2018).

Ozone (O_3)

We now examine the O_3 concentrations for the six stations in VTAPA for February 2007 to June 2017. In figure 5 the time variation plot for O_3 was plotted to graphically illustrate the times that the O_3 concentrations peak. The concentration of

ozone fluctuates with the time of day, with peak concentrations closer to midday due to increased photochemical reactions as a result of solar radiation being intense (Faridi et al., 2018)

The O_3 concentration shows a dwindling trend from the evening 6 pm until 12pm due to no photochemical reactions, a decrease in O_3 because of dry deposition and O_3 being depleted through the titration of NO_x which is consistent with studies done by Faridi et al., (2018) and Laban et al., (2015).

The O_3 concentration starts to increase in late winter with a maximum during spring to summer seasons. The O_3 concentration is a minimum during autumn (Von Schneidemesser et al., 2015). The O_3 concentration was higher during weekends than weekdays and this could be as a result of decreasing NO_x concentrations due to fewer vehicle emissions on a weekend. This occurrence is referred to as the "holiday effect" (Tan et al., 2013).

The South African NAAQS rolling 8-hour mean concentration limit value is 61 ppb for tropospheric O_3. The limit value is represented by a dashed black horizontal line in Figure 6 and values over 61 ppb are counted as limit value exceedances. Figure 6 illustrates that an exceedance at one station often coincides with exceedances at the other stations and this can be attributed to chemical transport between the air quality stations (Jacob et al., 2009; Von Schneidemesser et al., 2015). A study conducted by Masuku et al. (2014) in the VTAPA found that unusually high concentrations of ozone was attributed to biomass burning. The O_3 formed in the Vaal Triangle could be as a result of NO_x and VOC species from power station or other industrial emissions that are a distance away and not from species that are produced in the vicinity of the air quality monitoring station (Masuku et al., 2014).

The Mann Kendall and Thielsen calculations shown in figure 7 indicate that the monthly mean de-seasonalised trend analysis for O_3 concentration shows a significant increasing trend at Diepkloof and Three Rivers at the 0.001 level. Annual average O_3 concentrations for Diepkloof have increased by 1.31 ppb/per year, at Sharpeville by 0.37 ppb/per year and at Three Rivers by 0.68 ppb/per year. The decreasing O_3 concentration trends at Kliprivier, Sebokeng and Zamdela are not statistically significant.

Discussion

This research study analysed $PM_{2.5}$ and O_3 concentrations as collected at six ambient monitoring stations within the VTAPA. It was found that the pollutant concentrations are characterised by strong temporal and seasonal trends where $PM_{2.5}$ shows increased concentrations during the winter months and during the early hours of the day and the early hours of the evening with these trends being attributable to household combustion activities, windblown dust and domestic fuel burning, as well as the planetary boundary layer that breaks up around midmorning (de Lange et al., 2019). The long term trends in annual $PM_{2.5}$ concentrations for 2007-2017 indicate that there has been a highly significant decrease in $PM_{2.5}$ concentrations at Diepkloof and Sebokeng ($p<0,001$). A similar study was conducted by Cairncross, (2016) for $PM_{2.5}$ in different regions of South Africa, with the VTAPA being one of the regions, from 2012-2015. The results of this study show that there were many exceedances of the NAAQS daily limit of 40µgm^{-3} of $PM_{2.5}$ in the VTAPA stations, which resonates with this study. The annual average $PM_{2.5}$ NAAQS limit of 25 µgm^{-3} was exceeded at all four stations from 2012-2015, except at Diepkloof station from 2013-2015, which is consistent with this study.

The O_3 concentration varies according to season and time of the day. The peak values in ozone concentration are from 10 am to 6 pm (local time) and during late winter and spring. A study conducted in the VTAPA in 2014 by Laban et al. (2015) found that the O_3 concentration exceeded the NAAQS limit of 61ppb for an 8 hour running average which is consistent with this study. The long term trend indicates an upward trend in the annual average O_3 concentration for three of the six stations. There is a downward trend in O_3 concentration that is static and is not statistically significant for the other three stations in the VTAPA. The increase in O_3 from 2007-2017 for three of the sites can be likely attributed to an increase in photochemical reactions as well as an increase in O_3 precursor emissions (Jang et al., 2017). The concentration of O_3 is affected by O_3 precursors such as nitrogen oxides (NO_x) and volatile organic compounds (VOCS). The VTAPA has high volumes of traffic as there are industries that use trucks as well as domestic vehicles for people's day to day use and this provides a local source of NO_x and VOC.

There has been a significant decrease in $PM_{2.5}$ concentrations at only two of the six stations and no significant decrease in O_3 concentrations at any of the six stations in the VTAPA. This is of concern because long term exposure to $PM_{2.5}$ and O_3 increases the morbidity and mortality (Kinney, 2018) of people living in the VTAPA. There needs to be more enforcement to ensure that industries are complying with their emission limits and not exceeding limits set out by NAAQS. Projects that enable community members to get access to the use of clean fuels for cooking and space heating will also play an important role in improving the quality of air in VTAPA and reducing mortality and morbidity.

References

Apte, J.S., Brauer, M., Cohen, A.J., Ezzati, M. and Pope III, C.A. 2018, 'Ambient $PM_{2.5}$ reduces global and regional life expectancy', *Environmental Science & Technology Letters*, 5(9):546-551. https://doi.org/10.1021/acs.estlett.8b00360

Caiazzo, F., Ashok, A., Waitz, I.A., Yim, S.H. and Barrett, S.R. 2013, 'Air pollution and early deaths in the United States. Part I: Quantifying the impact of major sectors in 2005', *Atmospheric Environment*, 79:198-208. https://doi.org/10.1016/j.atmosenv.2013.05.081

Cairncross, E., 2016. The State of South Africa's air quality monitoring network and its air quality. *Paper presented at the National Association for Clean Air*.

Carslaw, D.C. and Ropkins, K., 2012, 'Openair — an R package for air quality data analysis', *Environmental Modelling & Software*, 27:52-61. https://doi.org/10.1016/j.envsoft.2011.09.008

de Lange, A., Garland, R.M. and Dyson, L.L., 2019, 'Estimating particulate matter (PM) concentrations from a meteorological index for data-scarce regions: A pilot study', *Atmospheric Pollution Research*. https://doi.org/10.1016/j.apr.2019.05.004

Derwent, R.G., Manning, A.J., Simmonds, P.G., Spain, T.G. and O'Doherty, S., 2018, ' Long-term trends in ozone in baseline and European regionally-polluted air at Mace Head, Ireland over a 30-year period', *Atmospheric Environment*, 179:279-287. https://doi.org/10.1016/j.atmosenv.2018.02.024

Faridi, S., Shamsipour, M., Krzyzanowski, M., Künzli, N., Amini, H., Azimi, F., Malkawi, M., Momeniha, F., Gholampour, A., Hassanvand, M.S. and Naddafi, K., 2018,' Long-term trends and health impact of $PM_{2.5}$ and O_3 in Tehran, Iran, 2006–2015', *Environment international*, 114:37-49. https://doi.org/10.1016/j.envint.2018.02.026

Feig, G.T., Vertue, B., Mabaso, D., Naidoo, S. and Ncgukana, N., 2015, 'Measurement of atmospheric black carbon in the Vaal Triangle and Highveld Priority Areas', *Clean Air Journal= Tydskrif vir Skoon Lug*, 25(1): 46-50. https://doi.org/10.17159/2410-972X/2015/v25n1a4

Fiore, A.M., Naik, V. and Leibensperger, E.M., 2015, 'Air quality and climate connections', *Journal of the Air & Waste Management Association*, 65(6):645-685. https://doi.org/10.1080/10962247.2015.1040526

Fuzzi, S., Baltensperger, U., Carslaw, K., Decesari, S., Denier van der Gon, H., Facchini, M. C., Fowler, D., Koren, I., Langford, B., Lohmann, U., Nemitz, E., Pandis, S., Riipinen, I., Rudich, Y., Schaap, M., Slowik, J. G., Spracklen, D. V., Vignati, E., Wild, M., Williams, M., and Gilardoni, S., 2015,' Particulate matter, air quality and climate: lessons learned and future needs', *Atmospheric Chemistry & Physics*, 15:8217-8299. https://doi.org/10.5194/acp-15-8217-2015

Gao, W., Tie, X., Xu, J., Huang, R., Mao, X., Zhou, G. and Chang, L., 2017,' Long-term trend of O_3 in a mega City (Shanghai), China: Characteristics, causes, and interactions with precursors', *Science of The Total Environment*, 603:425-433. https://doi.org/10.1016/j.scitotenv.2017.06.099

Jacob, D.J. and Winner D.A. 2009,' Effect of climate change on air quality', *Atmospheric environment*, 43(1): 51-63. https://doi.org/10.1016/j.atmosenv.2008.09.051

Karagulian, F., Belis, C.A., Dora, C.F.C., Prüss-Ustün, A.M., Bonjour, S., Adair-Rohani, H. and Amann, M. 2015,'Contributions to cities' ambient particulate matter (PM): A systematic review of local source contributions at global level', *Atmospheric environment*, 120:475-483. https://doi.org/10.1016/j.atmosenv.2015.08.087

Kinney, P.L., 2008, 'Climate change, air quality, and human health. *American journal of preventive medicine*, 35(5), 459-467. https://doi.org/10.1016/j.amepre.2008.08.025

Kinney, P.L. 2018,'Interactions of Climate Change, Air Pollution, and Human Health. *Current environmental health reports*, 5(1):179-186. https://doi.org/10.1007/s40572-018-0188-x

Laban, T.L., Beukes, J.P. and Van Zyl, P.G. 2015, 'Measurement of surface ozone in South Africa with reference to impacts on human health', *Clean Air Journal=Tydskrif vir Skoon Lug*, 25(1):9-12.

Lehtomäki, H., Korhonen, A., Asikainen, A., Karvosenoja, N., Kupiainen, K., Paunu, V.V., Savolahti, M., Sofiev, M., Palamarchuk, Y., Karppinen, A. and Kukkonen, J. 2018,'Health Impacts of Ambient Air Pollution in Finland', *International journal of environmental research and public health*, 15(4):736. https://doi.org/10.3390/ijerph15040736

Masuku, N., Feig, G., Vertue, B., Ncipha, X., Mabaso, D., Naidoo, S., Tshehla, C. and Ngcukana, N. 2014,' Analysis of a period of elevated ozone concentration reported over the Vaal Triangle on 2 June 2013', *Clean Air Journal=Tydskrif vir Skoon Lug*, 24(1):10-16.

Melamed, M.L., Schmale, J. and von Schneidemesser, E. 2016,'Sustainable policy—key considerations for air quality and climate change', *Current opinion in environmental sustainability*, 23:85-91. https://doi.org/10.1016/j.cosust.2016.12.003

Morgan, W.T., Paasonen, P., Righi, M. and Sindelarova, K. 2015,'Chemistry and the linkages between air quality and climate change', *Chemical Reviews*, 115(10):3856-3897. https://doi.org/10.1021/acs.chemrev.5b00089

Monks, P.S., Archibald, A.T., Colette, A., Cooper, O., Coyle, M., Derwent, R., Fowler, D., Granier, C., Law, K.S., Mills, G.E. and Stevenson, D.S. 2015, 'Tropospheric ozone and its precursors from the urban to the global scale from air quality to short-lived climate forcer', *Atmospheric Chemistry and Physics*, 15(15):8889-8973. https://doi.org/10.5194/acp-15-8889-2015

NAAQS (National Ambient Air Quality Standards) (2009). Air quality standard. Notice No. 1210, 24 December.

Okello, N.O., Camminga, S., Okello, T.W. and Zunckel, M. 2018,'Spatial and Temporal Trends of PM_{10} and SO_2 in the Richards Bay Area', *Clean Air Journal=Tydskrif vir Skoon Lug*, 28(2):80-89. https://doi.org/10.17159/2410-972x/2018/v28n2a20

Orru, H., Ebi, K.L. and Forsberg, B. 2017,'The interplay of climate change and air pollution on health', *Current environmental health reports*, 4(4):504-513. https://doi.org/10.1007/s40572-017-0168-6

Ramanathan, P. J., Crutzen, J. T., Kiehl, D., and Rosenfeld D. 2001, 'Aerosols, climate, and the hydrological cycle science', 294(5549):2119-2124. https://doi.org/10.1126/science.1064034

Roser, M. and Ritchie, H. 2013,'Indoor Air Pollution, *Our World in Data*. Available at: https://ourworldindata.org/indoor-air-pollution

SANAS. 2013, TR 07-03. Supplementary Requirements for the accreditation of continuous ambient air quality monitoring stations.

South African Department of Environmental Affairs (DEA). The Second Generation Vaal Triangle Airshed Priority Area Air Quality Management Plan Draft Baseline Assessment Report [document on the internet]c2019[cited 2019 October 1].

Shezi, B. and Wright, C.Y. 2018,'Household air pollution exposure and respiratory health outcomes: a narrative review update of the South African epidemiological evidence, '*Clean Air Journal*', 28(1):43-56. https://doi.org/10.17159/2410-972X/2018/v28n1a11

Silva, R.A., West, J.J., Lamarque, J.F., Shindell, D.T., Collins, W.J., Dalsoren, S., Faluvegi, G., Folberth, G., Horowitz, L.W., Nagashima, T. and Naik, V. 2016, 'The effect of future ambient air pollution on human premature mortality to 2100 using output from the ACCMIP model ensemble', *Atmospheric chemistry and physics*, 16(15):9847-9862. https://doi.org/10.5194/acp-16-9847-2016

Scorgie, Y., Kneen, M.A., Annegarn, H.J. and Burger, L.W. 2003, 'Air pollution in the Vaal Triangle-quantifying source contributions and identifying cost-effective solutions', *Clean Air Journal= Tydskrif vir Skoon Lug*, 13(2):5-18.

Tan, P.H., Chou, C. and Chou, C.C.K. 2013, 'Impact of urbanization on the air pollution "holiday effect" in Taiwan', *Atmospheric Environment*, 70:361-375. https://doi.org/10.1016/j.atmosenv.2013.01.008

Terblanche, A.P., Opperman, L., Nel, C.M., Reinach, S.G., Tosen, G. and Cadman, A. 1992, 'Preliminary results of exposure measurements and health effects of the Vaal Triangle Air Pollution Health Study', *South African medical journal=Suid-Afrikaanse tydskrif vir geneeskunde*, 81(11):550-556.

Terblanche, A.P., Nel, C.M., Opperman, L. and Nyikos, H.1992, 'Exposure to air pollution from transitional household fuels in a South African population', *Journal of exposure analysis and environmental epidemiology*, 3:15-22.

Tshehla C, Wright CY. 2019, '15 Years after the National Environmental Management Air Quality Act: Is legislation failing to reduce air pollution in South Africa?',*South African Journal of Science* 115(9/10):27-29. https://doi.org/10.17159/sajs.2019/6100

Van Horen, C., Eberhard, A., Trollip, H. and Thorne, S.1993, 'Energy, environment and urban poverty in South Africa', *Energy Policy*, 21(5):623-639. https://doi.org/10.1016/0301-4215(93)90045-H

Vecchi, R., Marcazzan, G., Valli, G., Ceriani, M. and Antoniazzi, C.,2004,'The role of atmospheric dispersion in the seasonal variation of PM_1 and $PM_{2.5}$ concentration and composition in the urban area of Milan (Italy)',*Atmospheric Environment*, 38(27):4437-4446. https://doi.org/10.1016/j.atmosenv.2004.05.029

Villa, T.F., Gonzalez, F., Miljievic, B., Ristovski, Z.D. and Morawska, L. 2016, 'An overview of small unmanned aerial vehicles for air quality measurements: Present applications and future prospective', Sensors, 16(7):1072. https://doi.org/10.3390/s16071072

Von Schneidemesser, E., Monks, P.S., Allan, J.D., Bruhwiler, L., Forster, P., Fowler, D., Lauer, A., Morgan, W.T., Paasonen, P., Righi, M. and Sindelarova, K. 2015, 'Chemistry and the linkages between air quality and climate change', *Chemical Reviews*, 115(10):3856-3897. https://doi.org/10.1021/acs.chemrev.5b00089

World Health Organization, 2006, '*WHO Air quality guidelines for particulate matter, ozone, nitrogen dioxide and sulfur dioxide: global update 2005: summary of risk assessment*,' (No. WHO/SDE/PHE/OEH/06.02). Geneva: World Health Organization.

Zunckel, M., Venjonoka, K., Pienaar, J.J., Brunke, E.G., Pretorius, O., Koosialee, A., Raghunandan, A. and Van Tienhoven, A.M. 2004, 'Surface ozone over southern Africa: synthesis of monitoring results during the Crossborder Air Pollution Impact Assessment project', *Atmospheric Environment*, 38(36):6139-6147. https://doi.org/10.1016/j.atmosenv.2004.07.029

Research article

Assessment of changes in concentrations of selected criteria pollutants in the Vaal and Highveld priority areas

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Received: 9 October 2019 - **Reviewed:** 31 October 2019 - **Accepted:** 20 November 2019
https://doi.org/10.17159/caj/2019/29/2.7464

Abstract

Ambient air pollution has important impacts on a variety of environmental issues, particularly on human health and ecosystem processes. A key tool for understanding the impacts of atmospheric pollution is through the long-term measurement of the ambient concentrations of criteria atmospheric pollutants. Monitoring of ambient pollution concentrations has been conducted in two of the National Air Quality Priority Areas since 2007. During this time period, significant changes in the management of air pollution have occurred, including the adoption of the ambient air quality standards, and the implementation of Section 21 emission standards. This paper examines the long-term evolution of ambient concentrations for PM and SO_2 in the Vaal Triangle Airshed Priority Area and Highveld Priority Area. These trends will be evaluated against the implementation of management interventions and the variation in the measured concentrations and emerging areas of concern are highlighted.

Keywords

Particulate matter, SO_2, Priority areas, air quality trends, Vaal Triangle Priority Area, Highveld Priority Area, Theil-Sen.

Introduction

In the time since the promulgation of the National Environmental Management Air Quality Act (NEM:AQA) (DEA, 2004) considerable changes have occurred in the air quality management landscape in South Africa (Tshehla and Wright, 2019), these include:

- The introduction of National Ambient Air Quality Standards (NAAQS) (DEA, 2009, 2012b),
- The identification of activities resulting in atmospheric emissions from industry (Section 21 of the Act) (DEA, 2010) and the subsequent setting of atmospheric emission limits (DEA, 2013a, 2015a), including certain printing industry activities (DEA, 2016).
- The promulgation of dust control regulations (DEA, 2013b),
- The identification and development of emission standards for controlled emitters as described under section 23 of the NEMAQA including; small scale char and charcoal plants (DEA, 2015b), temporary asphalt plants (DEA, 2014b), small scale boilers (DEA, 2013c),
- The declaration of greenhouse gases as priority air pollutants (DEA, 2014a)

While this legislation has been enacted, certain areas have been identified as being of particular concern and have been declared as air quality priority areas, these include: the Vaal Triangle Airshed Priority Area (VTAPA; DEAT, 2006), the Highveld Priority Area (HPA;DEAT, 2007) and the Waterberg-Bojanala Priority Area (DEA, 2012a). Within these areas, particular concern has been placed in the development and review of management plans, including the VTAPA in 2009, the HPA in 2012 and the Waterberg in 2015 (DEA, 2015c).

Beyond the scope of changing legislation, there have been changes in the emissions profiles within the priority areas that have had significant impacts on emission profiles in the priority areas (Pretorius et al., 2015). These include:

- The increase in the number of vehicles in the country has contributed to changes in the emissions profiles, including an increase in vehicle emissions. Between January 2009 and June 2018, South Africa has seen an increase in motor vehicles, rising from ~9 to ~12 million. Notable increases have been seen in the Gauteng and Mpumalanga provinces (where VTAPA and HPA are located), with increases from ~3.5 to ~4.7 million in Gauteng, and ~600 000 to ~900 000 motor vehicles in Mpumalanga have been recorded (ENATIS, 2018).
- The closure of Highveld Steel in February 2015 (Goldswain, 2016), which resulted in a decrease in industrial emissions in the HPA.
- The large scale domestic electrification programmes in the priority areas which resulted in the rapid electrification, across South Africa more than 5 million households received access to electricity between 1990 and 2007 in South Africa (Bekker et al., 2008) which resulted in a reduction in household emissions but an increase in electricity consumption and associated emissions at the power generation plants.

Although changes in power generation including the recommissioning of Camden, Grootvlei and Komati power stations to counter the electricity shortages experienced from 2007 (ESKOM, 2011), and the introduction of the Kusile and Medupi power stations into the national grid have improved the output capacity, this only sustains the country's reliance on coal, further delaying emission reductions, especially with non-compliant ageing power plants (Pretorius et al., 2015).

Concurrent with the development of legislation and the rollout of air quality management plans, there has been the installation of air quality monitoring infrastructure. Starting in 2007, the Vaal Triangle Airshed Priority Area Ambient Air Quality Monitoring Network was established with monitoring sites located at identified air pollution hotspots, followed by the Highveld Priority Area ambient air quality monitoring network in 2008. These networks have approximately 10 years of data that are available through the South African Air Quality Information System (SAAQIS). These data can be used to identify trends in ambient concentrations in order to identify if the interventions made have been effective, and if there are changing priorities in which pollutants are of greatest concern.

Methods and Materials

Data for the 11 monitoring stations in the VTAPA and HPA priority area ambient air quality monitoring networks were requested from SAAQIS at an hourly temporal resolution (Table 1). The data were provided in a quality controlled form, but a subsequent quality control of the data was conducted to remove a limited number of negative concentration values and the removal of a significant number of values below the detection limit of the instruments (typically zero values) and other anomalous measurements. The data were analysed using the R language for statistical computing (R Core Team, 2013), specifically using the Open Air Package (Carslaw and Ropkins, 2012). The trends in the concentrations of PM_{10}, $PM_{2.5}$, and SO_2 were calculated for each station for the period of available data using the Theil-Sen trend analysis following a deseasonilisation step as recommended, which uses the Loess method. The Theil-Sen Estimator approach uses monthly averages, for which an 80% data availability threshold was set. A comparison of the trends in the continuously monitored SO_2 concentrations can be made with a dataset of measurements conducted by the CSIR extending from 1959-1968 (Kemeny and Halliday, 1972; Kemeny, 1980; Kemeny and Vleggaar, 1983; Walker, Ellerbeck and Kemeny, 1986; Walker, Galpin and Pienaar, 1987). The historical CSIR measurements were made using the Hydrogen Peroxide Method, which consisted of passing the air volume through a dilute solution of hydrogen peroxide and measuring the change in the pH of the solution through titration with a sodium borate solution (Kemeny, 1980). Comparison with these historical results serves as a valuable benchmark as to how the ambient concentrations of SO_2 have changed in the last 60 years.

Results

PM_{10}

Using a 75% data capture threshold, it is clear that all the monitoring stations in the VTAPA and HPA have been out of compliance with the historical (red line) and/or current (orange line) annual NAAQS (Figure 1). This corresponds to the results presented by the National Air Quality Officer in the annual State of the Air Report (individual values presented may differ slightly based on the data completeness requirements or the data cleaning protocol that have been used). As of 2016 (the last year with full data used in this assessment), it was only the Hendrina and Middelburg sites that complied with the annual standard, and both of these sites are located in middle income communities. The Zamdela, Witbank, Three Rivers and Sharpeville sites were non-compliant with the historical standard. From this it is clear that significant problems related to the concentrations of PM_{10} occur in the majority of the monitoring sites in the VTAPA and HPA.

The monthly PM_{10} concentrations have shown a general decrease at all the sites in the VTAPA and HPA (Figure 2), except for Sharpeville (which shows a large increase in 2016), Kliprivier and Three Rivers. This decrease has been significant at the $p < 0.001$ confidence interval at Diepkloof (-1.56 $\mu g/m^3$/year), Sebokeng (-1.37 $\mu g/m^3$/year), Zamdela (-4.68 $\mu g/m^3$/year), Hendrina (-1.97 $\mu g/m^3$/year), Middelburg (-3.54 $\mu g/m^3$/year) and Secunda (-4.35 $\mu g/m^3$/year). At the $p < 0.01$ confidence level Ermelo shows a trend of -1.22 $\mu g/m^3$/year and at the $p<0.05$ level, Sharpeville shows a decreasing trend of -1.16 $\mu g/m^3$/year (the strength of the trend is negatively influenced by the 2016 value). The Witbank site shows a decrease but this is not significant, while the Kliprivier site shows a non-significant increase. The Three Rivers site shows an increase of 0.99 $\mu g/m^3$/year at a $p<0.05$ confidence interval.

Table 1: *Monitoring site Location and dominant emission sources*

Site	Priority Area	Location	Dominant Emissions Sources
Diepkloof	VTAPA	-26.2507S 27.9564E	Vehicles, Mine Tailings and Domestic Combustion
Ermelo	HPA	-26.4934S 29.9690E	Domestic Combustion
Hendrina	HPA	-26.1319S 29.7343E	Electricity Generation
Kliprivier	VTAPA	-26.4203S 28.0848E	Domestic Combustion, Vehicles
Middelburg	HPA	-25.7960S 29.4636E	Regional Industry
Sebokeng	VTAPA	-26.5879S 27.8409E	Domestic Combustion, Industry
Secunda	HPA	-26.5485S 29.0800E	Industry, Domestic Combustion, Mine Tailings
Sharpeville	VTAPA	-26.6898S 27.8677E	Domestic Combustion, Industry
Three Rivers	VTAPA	-26.6569S 27.9993E	Electricity Generation
Witbank	HPA	-25.8778S 29.1886E	Domestic Combustion, Industry
Zamdela	VTAPA	-26.8448S 27.8551E	Domestic Combustion, Industry

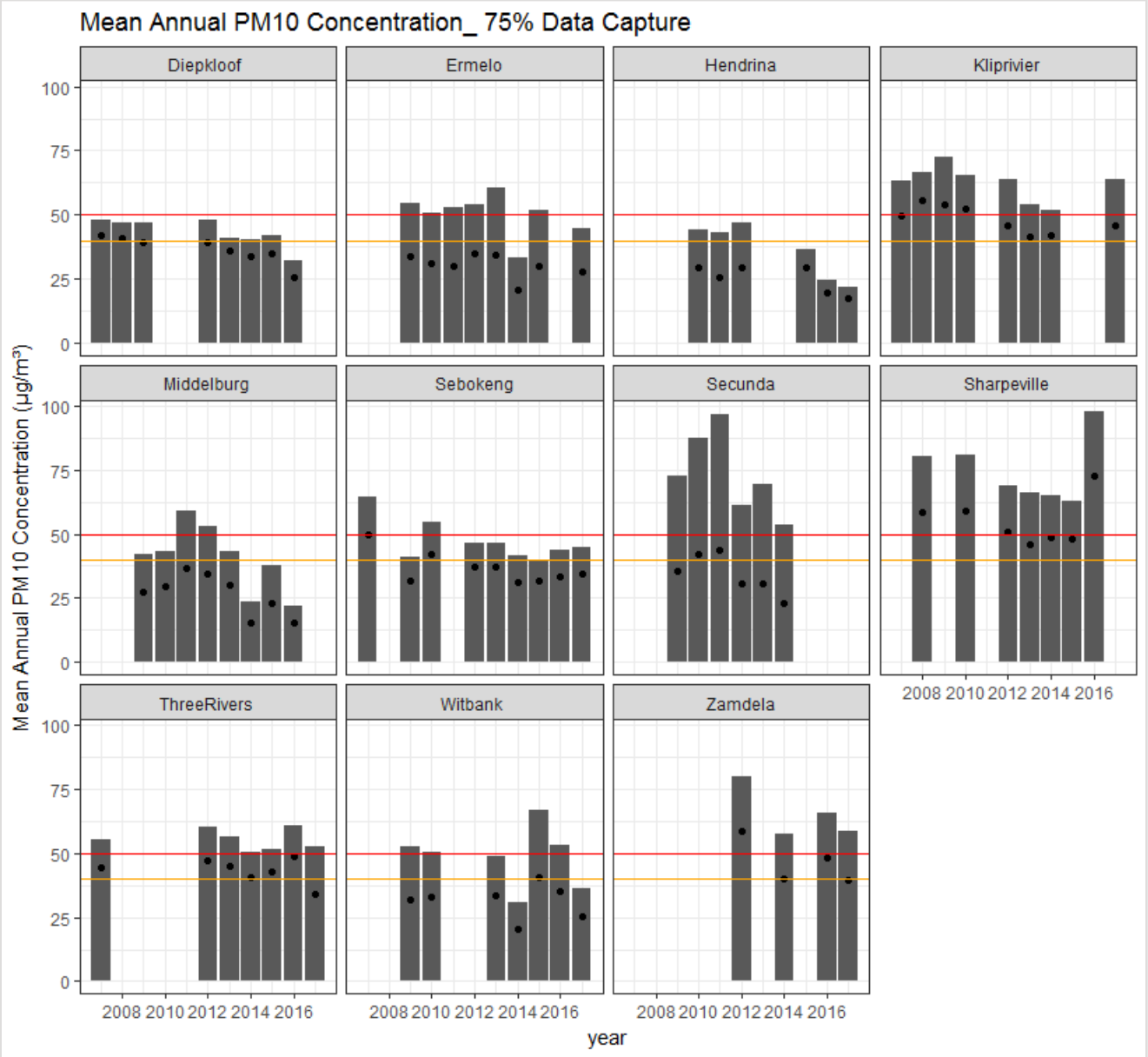

Figure 1: *Annual average PM_{10} concentrations for the VTAPA and HPA (the black dot represents the median, the red horizontal line represents the historical (2009-2014) annual standard of 50µg/m^3, the orange line represents the current (2015-present) annual standard of 40µg/m^3)*

Using the trend in the change in PM_{10} concentrations over the measurement period, a simplistic assessment was made of how long it would take for each of the monitoring stations to reach compliance with the national PM_{10} standard based on the annual average concentration for the last full year with data (Table 2), it is acknowledged that this is an overly simplistic approach as it is not expected that the ambient concentrations of PM_{10} will follow a continuous linear trend. However, this approach is illustrative as to how long it may take to get to compliance following the long term current trend. Currently three stations are in compliance with the PM_{10} annual standard for the remainder it is (simplistically) expected that compliance will be reached between 2018 for Secunda and 2065 for Sharpeville. In the Sharpeville case, the annual concentration for 2016 (57.2 $\mu g/m^3$) was considerably higher than for 2015 (22.78 $\mu g/m^3$) in this case if the 2015 annual value is used with a continuation of the historical trend, compliance is expected by 2035.

$PM_{2.5}$

Using the 75% data capture threshold, it is clear that most of the monitoring sites are in exceedance of the current annual standard for $PM_{2.5}$, it is only Hendrina and Middelburg that are below the current annual NAAQS (Figure 3). From this it is clear that there are still significant problems relating to the concentrations of $PM_{2.5}$ over the VTAPA and HPA.

Monthly $PM_{2.5}$ concentrations show a decreasing trend for all the sites, except for Zamdela (a non-significant increase in $PM_{2.5}$) (Figure 4). A decreasing concentration at a confidence interval $p \leq 0.001$ was found for: Diepkloof (-2.33 $\mu g/m^3$/year), Middelburg

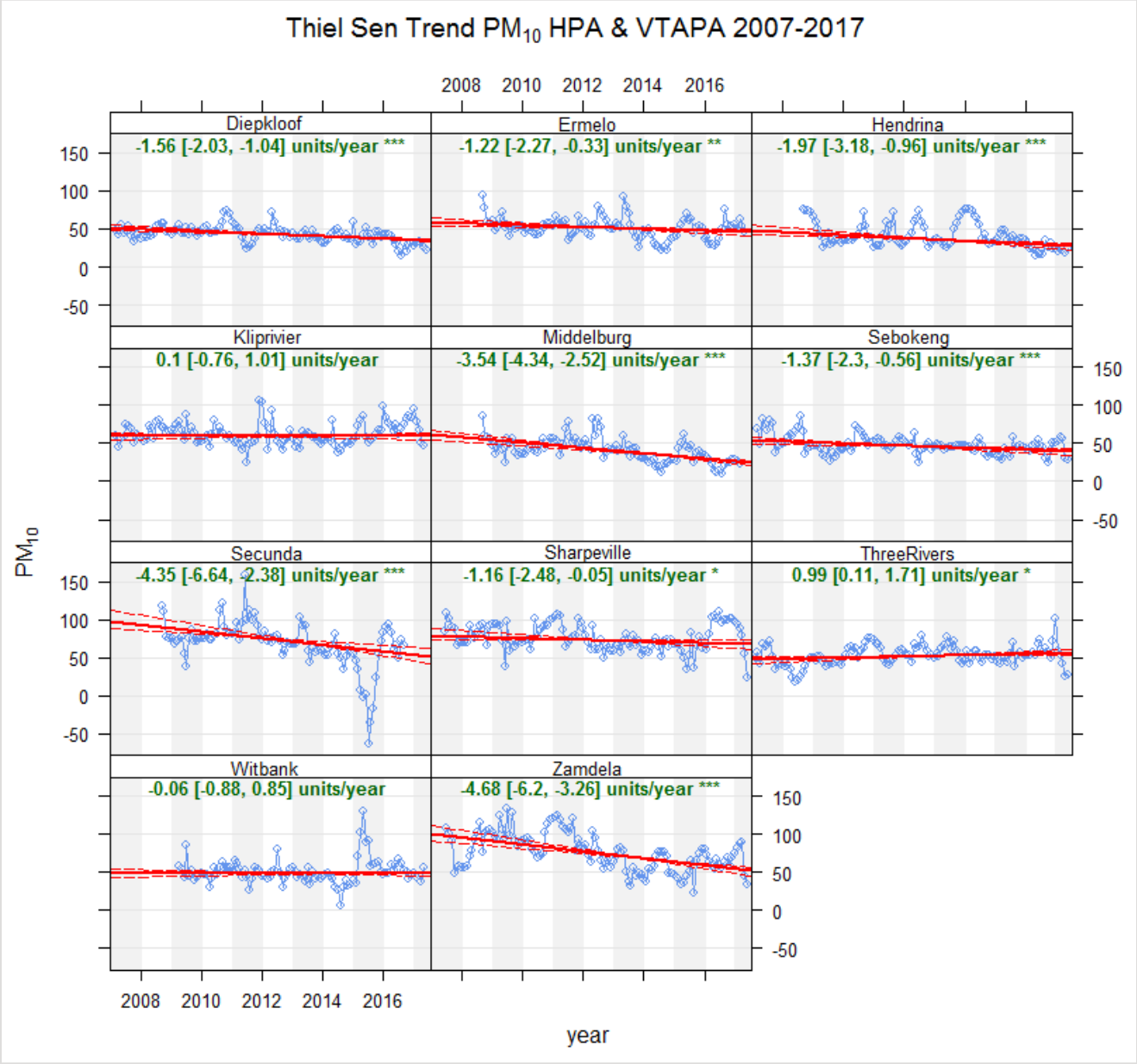

Figure 2: *Thiel-Sen Trend analysis for monthly PM_{10} at the VTAPA and HPA air quality monitoring sites, *** indicates significance at the p<0.001 confidence level, ** indicates significance at the p<0.01 confidence level and * indicates significance at the p<0.05 confidence level*

Table 2: *Summary of trends in PM_{10} concentrations in the VTAPA and HPA (NS = not significant)*

Site	Priority Area	Average annual PM_{10} Concentration (µg/m³), most recent full year	Year of last measurement	Difference from NAAQS	Rate µg/m³/year	Significance	Years to compliance with current standard	Year of compliance with current standard
Diepkloof	VTAPA	31.99	2016	-8.01	-1.56	0.999	0	In compliance
Ermelo	HPA	51.73	2015	11.73	-1.22	0.99	9.6	2026
Hendrina	HPA	24.43	2016	-15.57	-1.97	0.999	0	In compliance
Kliprivier	VTAPA	51.88	2014	11.88	0.1	NS	No Trend	
Middelburg	HPA	21.90	2016	-18.1	-3.54	0.999	0	In compliance
Sebokeng	VTPA	43.97	2016	3.97	-1.37	0.999	2.9	2019
Secunda	HPA	53.82	2014	13.82	-4.35	0.999	3.2	2018
Sharpeville	VTAPA	97.2	2016	57.2	-1.16	0.95	49	2065
	VTAPA	62.78	2015	22.78	-1.16	0.95	19.6	2035
Three Rivers	VTAPA	61.15	2016	21.15	+0.99	0.95	Worsening PM_{10} conditions	
Witbank	HPA	53.35	2016	13.35	-0.06	NS	No Trend	
Zamdela	VTAPA	65.72	2016	25.72	-4.68	0.999	5.5	2022

Table 3: *Summary of trends in $PM_{2.5}$ concentrations in the VTAPA and HPA (NS = not significant)*

Site	Average annual $PM_{2.5}$ Concentration (µg/m³)	Year of last measurement	Difference from NAAQS	Rate µg/m³/year	Significance	Years to compliance	Year of compliance with current standard	Year of compliance with 2030 standard
Diepkloof	24.46	2016	4.46	-2.33	0.999	1.9	2018	2021
Ermelo	21.91	2015	1.91	-1.75	0.999	1.1	2016	2019
Hendrina	13.21	2016	-6.79	-1	0.999	In compliance		
Kliprivier	35.47	2014	15.47	-0.72	0.95	21.5	2035	2042
Middelburg	10.56	2016	-9.44	-1.88	0.999	In compliance		
Sebokeng	31.15	2016	11.15	-1.61	0.999	6.9	2023	2026
Secunda	25.41	2014	5.41	-1.84	0.999	3	2017	2020
Sharpeville	35.75	2015	15.75	-0.4	NS	No Trend		
Three Rivers	28.56	2016	8.56	-0.36	0.99	23.7	2039	2053
Witbank	22.95	2016	2.95	-1.12	0.999	2.6	2016	2023
Zamdela	30.71	2015	10.71	0.16	NS	No Trend		

Table 4: *Summary of trends in SO_2 concentration in the VTAPA and HPA (NS = not significant)*

Site	Average annual SO_2 Concentration (ppb)	Year of last measurement	Difference from NAAQS	Rate ppb/year	Significance	State of compliance with current standard
Diepkloof	4.18	2016	-14.82	-0.3	0.999	In compliance
Ermelo	10.76	2016	-8.42	-0.52	0.999	In compliance
Hendrina	9.5	2016	-8.5	-0.73	0.999	In compliance
Kliprivier	5.17	2014	-13.83	0.01	NS	In compliance, no trend
Middelburg	5.77	2015	-13.23	-0.52	0.999	In compliance
Sebokeng	4.96	2015	-14.04	0.08	NS	In compliance, no trend
Secunda	6.83	2016	-12.17	-0.23	NS	In compliance, no trend
Sharpeville	5.79	2016	-13.21	-0.1	NS	In compliance, no trend
Three Rivers	5.93	2016	-13.07	+0.08	0.95	In compliance
Witbank	19.29	2014	+0.29	-0.21	NS	Out of compliance, no trend
Zamdela	8.47	2016	-10.53	+0.03	NS	In compliance, no trend

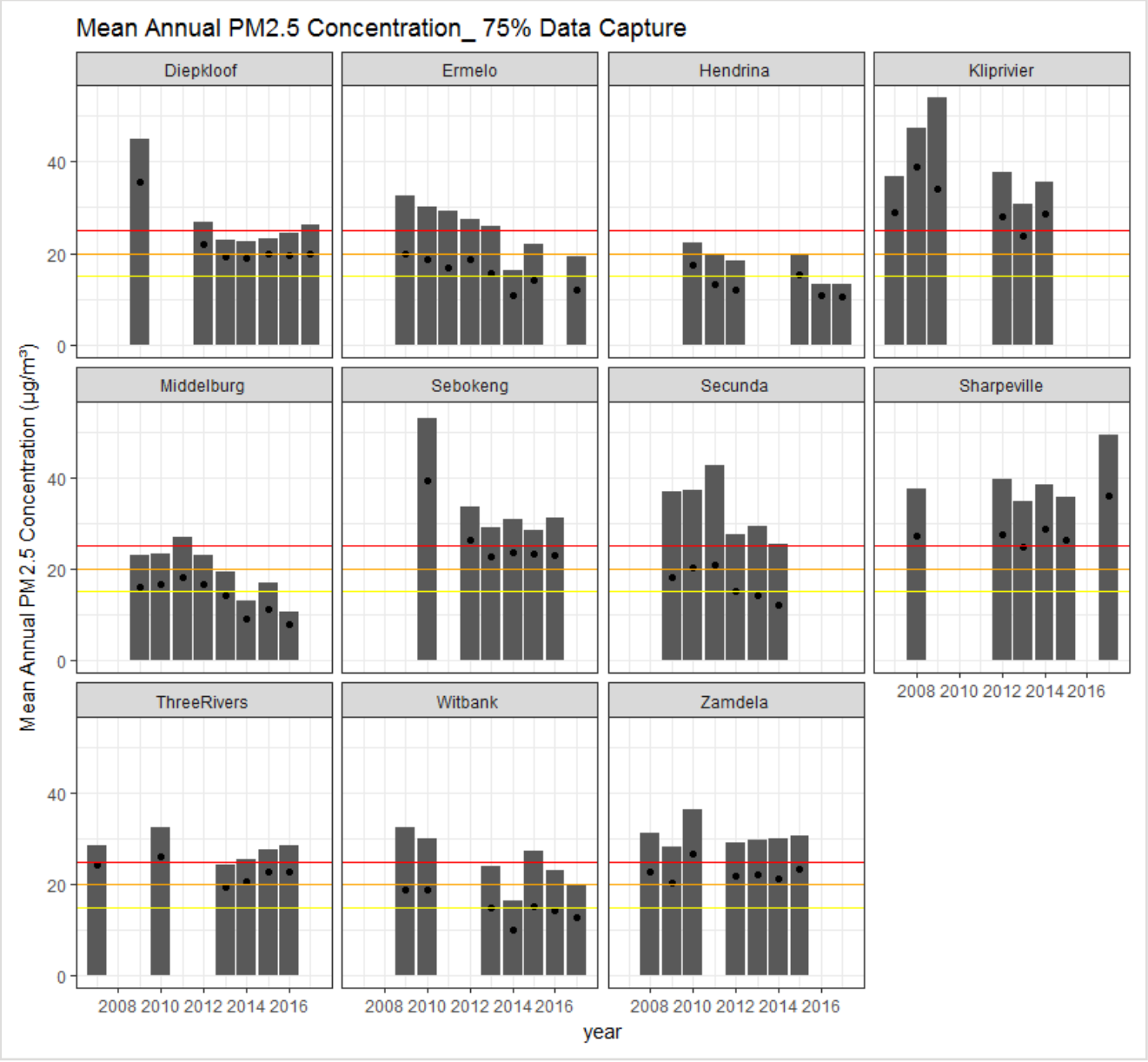

Figure 3: *Annual average $PM_{2.5}$ concentrations (µg/m³) for the VTAPA and HPA (black dot represents the median), the red horizontal line represents the historical (2012-2015) annual standard of 25µg/m³, the orange line represents the current (2016-2029) annual standard of 20µg/m³, and the yellow line represents the future (2030) annual standard of 15 µg/m³).*

(-1.88 µg/m³/year), Secunda (-1.84 µg/m³/year), Ermelo (-1.75 µg/m³/year), Sebokeng (-1.61 µg/m³/year), Witbank (-1.12 µg/m³/year), and Hendrina (-1 µg/m³/year). A decreasing trend with a confidence interval of p <0.01 was found for Three Rivers (-0.36 µg/m³/year), while Kliprivier showed a decreasing trend (-0.72 µg/m³/year) at the p<0.05 level. Sharpeville and Zamdela did not show a statistically significant trend in the $PM_{2.5}$ concentrations.

Using the trend in the change in $PM_{2.5}$ concentration over the measurement period, a simplistic assessment was made of how long it would take for each of the monitoring stations to reach compliance with the current national $PM_{2.5}$ standard, based on the long term trend and extending the trend line the from the latest available data point, i.e. concentrations for 2016. Sharpeville is the exception due to the unusually high concentration value in 2016. The long term trendline was extended from the value in 2015 into the future to determine the year in which the annual concentration for the last full year with data (Table 3) and the year in which compliance with the 2030 standard is expected to be met. Currently two of the sites comply with both the current and future $PM_{2.5}$ standards, these sites are Hendrina and Middelburg. For the sites that are non-compliant with the NAAQS, compliance was expected to be reached between 2016 for Ermelo to 2039 for Three Rivers. For the future standard, compliance is expected between 2019 and 2053 for Ermelo and Three Rivers respectively. Since no statistically significant trend could be established for Zamdela and Sharpeville it is difficult to estimate when compliance could be reached at current rates.

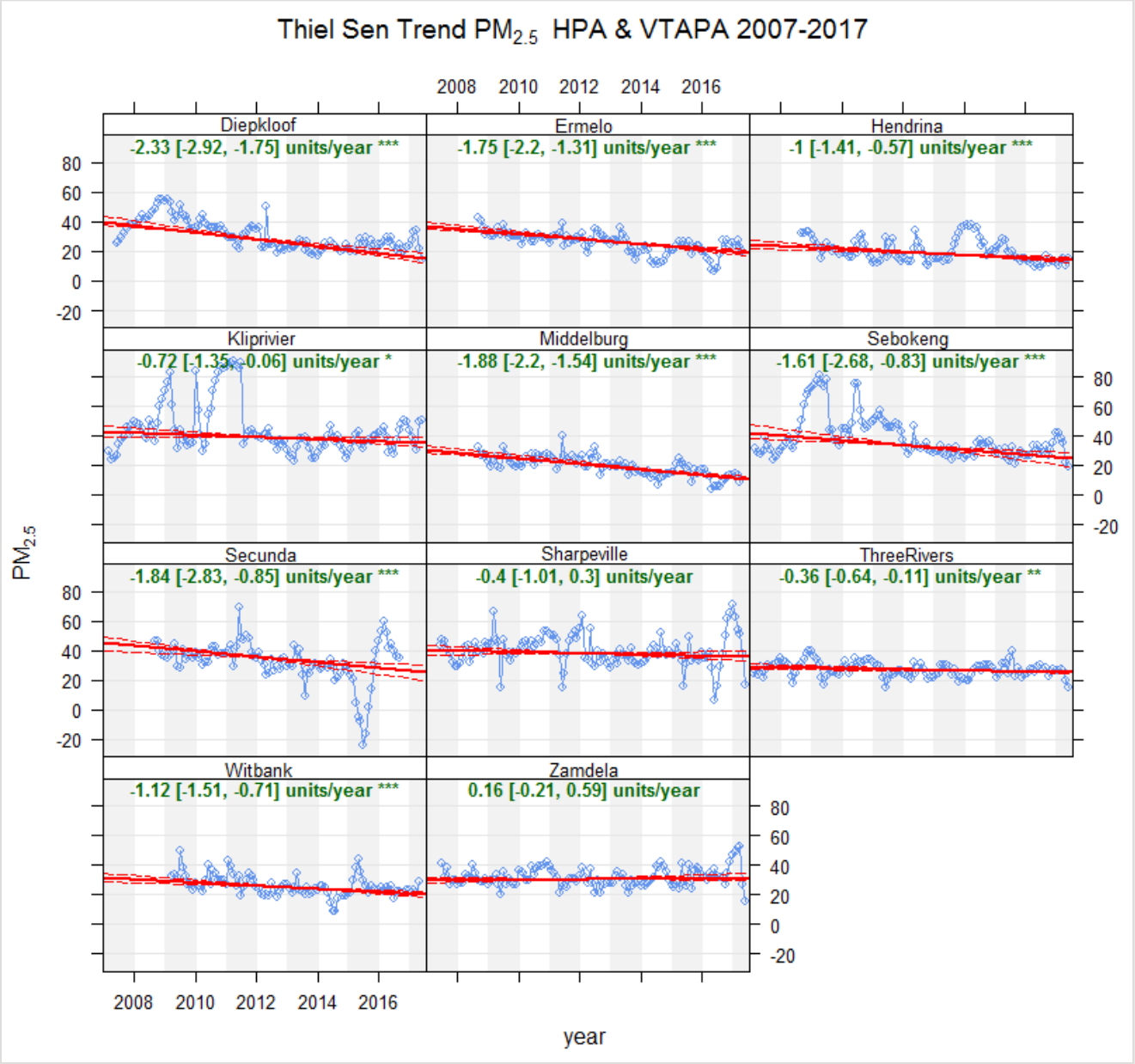

Figure 4: *Thiel-Sen trend in monthly $PM_{2.5}$ concentration ($\mu g/m^3$)over the VTAPA and HPA *** indicates significance at the p<0.001 confidence level, ** indicates significance at the p<0.01 confidence level and * indicates significance at the p<0.05 confidence level*

SO_2

Using a 75% data capture requirement, it is only the Witbank site that exceeds the annual SO_2 standard within the priority areas (Figure 5). SO_2 has not proven to be out of compliance with the NAAQS.

In comparison to the PM measurement where there was a significant decreasing trend in almost all the monitoring sites, the trends in SO_2 concentrations were only significant at five of the 11 sites (Figure 6). A significant decrease in SO_2 concentrations at the p<0.001 level was found at Hendrina (-0.73 ppb/year), Ermelo (-0.52 ppb/year), Middelburg (-0.52 ppb/year), and Diepkloof (-0.3 ppb/year). At the Three Rivers site an increasing trend of 0.08 ppb/year in the SO_2 concentration was observed at the p<0.05 confidence level. No significant trends were observed at the other sites (Table 4).

Historical SO_2 concentrations

Long term historical data are not available for many of the sites under consideration in this study, however the measured SO_2 concentrations reported for the major metros in South Africa during the 1960s show that historically the ambient concentrations of SO_2 were considerably higher than is currently recorded. Within Johannesburg, Durban and East London annual average SO_2 concentrations above 20ppb were common (Figure 7).

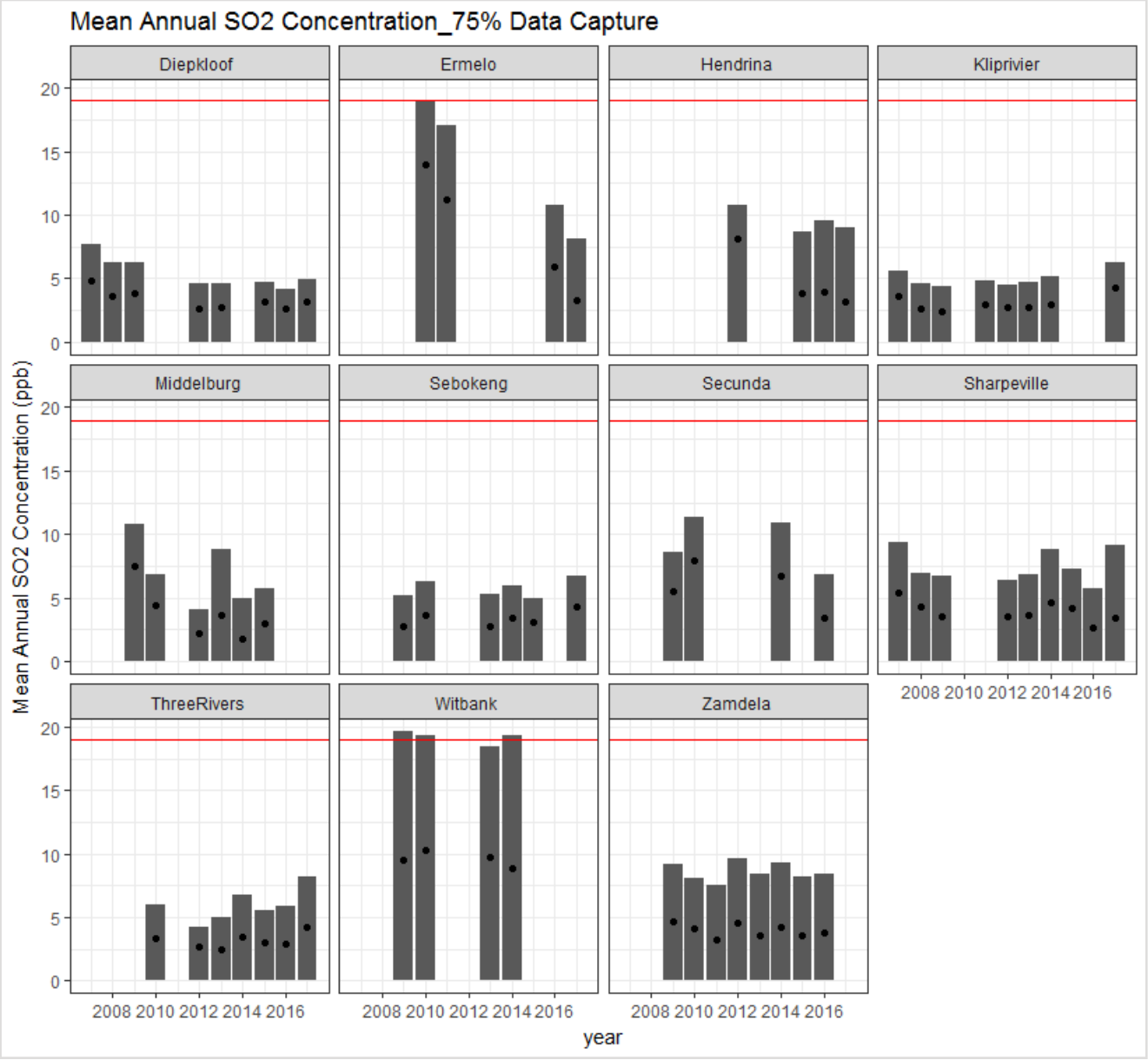

Figure 5: *Annual average SO_2 concentrations (ppb) for the VTAPA and HPA (black dot represents the median), the red horizontal bar represents the annual standard of 19 ppb).*

Discussion

Considerable efforts have been expended over the last decade and a half in the development of the legislative framework to govern air quality management in South Africa, however it is frequently stated that the strategic objectives have not been met (Tshehla and Wright, 2019). It was reported that with 70 % of the planned interventions from the Vaal Triangle Priority Area Air Quality Management Plan having been implemented, there was no proportional improvement in air quality (Senene, 2018). However, these reports are often based on a fairly cursory analysis of whether compliance criteria have been met. The long term dataset of reasonably comprehensive and good quality data that is growing in the VTAPA and HPA provides a good opportunity to assess the long term trends in pollution in these areas.

As stated by the National Air Quality Officer, in previous State of Air Reports, the compliance with the PM_{10} and $PM_{2.5}$ standards is a significant problem over the Vaal and Highveld regions with almost all of the monitoring sites being non-compliant with the current NAAQS this is well known and has been previously extensively documented (Venter et al., 2012; Hersey et al., 2015; Wernecke et al., 2015; Feig, et al, 2016; Garland et al., 2017). However, annual SO_2 is generally (with the exception of Witbank) considerably lower than the NAAQS. These results are similar to those reported by Feig et al, (2016) for the Waterberg priority area and Venter et al. (2012), for the Western Bushveld Igneous complex, which identified the major sources of SO_2 as high stack emissions and PM_{10} from domestic combustion emissions. Across the Highveld annual average concentrations of SO_2 were reported as being below the NAAQS (Lourens et al.,

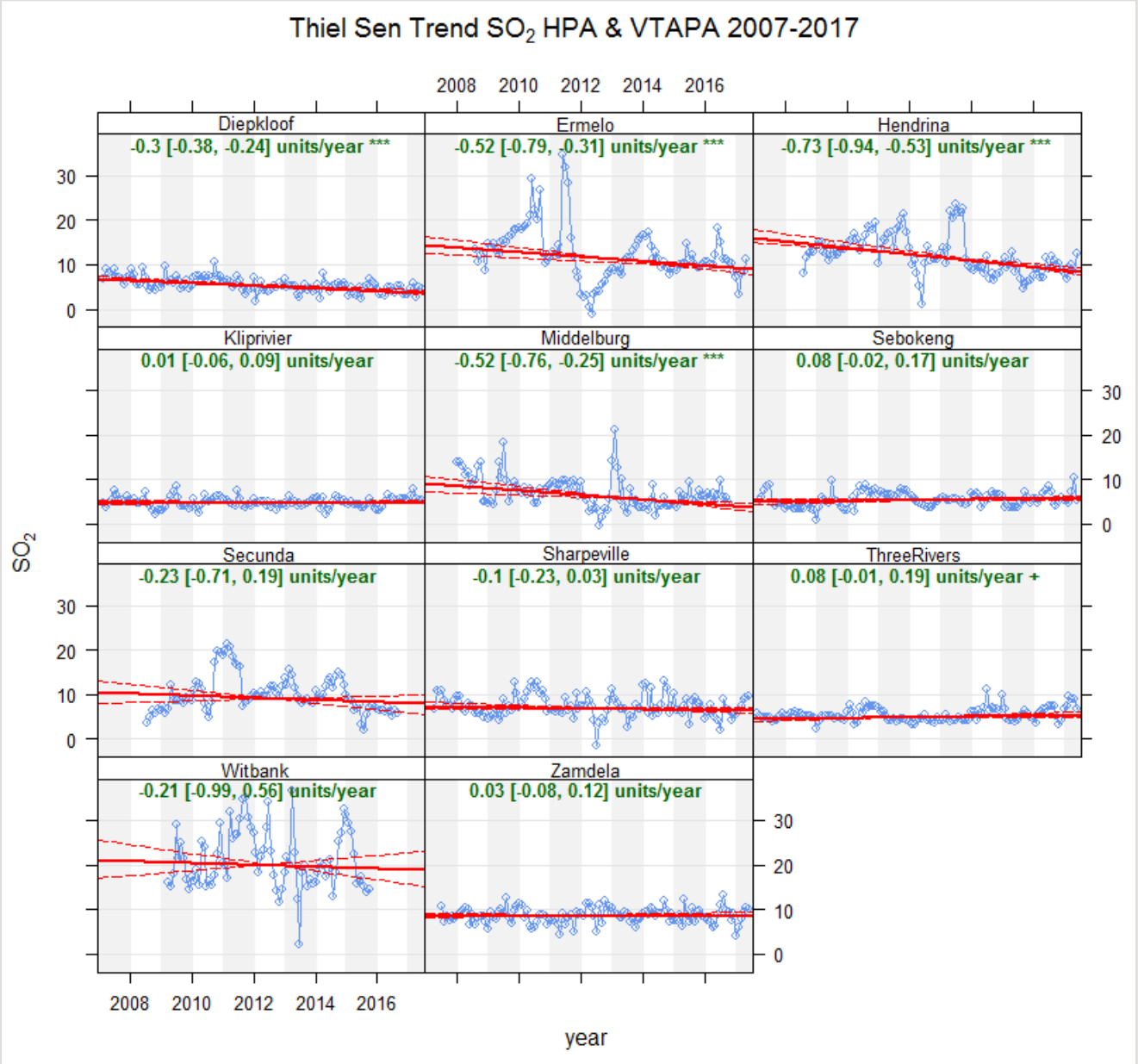

Figure 6: *Thiel-Sen trend in SO_2 concentration (ppb) over the VTAPA and HPA *** indicates significance at the p<0.001 confidence level, ** indicates significance at the p<0.01 confidence level and * indicates significance at the p<0.05 confidence level*

2011). At the KwaDela site (located in the Highveld between Ermelo and Secunda), health risks associated with both indoor and outdoor exposure to particulate matter was identified as a risk (Wernecke et al., 2015).

The trend in the concentrations of particulate matter is largely negative, with the exception of Three Rivers for PM_{10} and Zamdela for $PM_{2.5}$ (which did not show any statistical significance).

Previous studies into the long term temporal and spatial analysed of pollutants in the priority areas has been done previously including the work done by Sangeetha and Sivakumar, (2019), where monitoring stations were grouped and averaged according to broad spatial location. In this study the seasonal variability of measured SO_2 was discussed, but the long term trend was not analysed. Further comparison between ground based measurements of SO_2 and satellite based estimates were performed for the Sharpeville site in the VTAPA (Sangeetha et al., 2017). In a remote sensing based study that looked at SO_2 concentrations over the HPA and increasing trend in SO_2 emissions was reported (Shikwambana and Tsoeleng, 2019) it is however unclear how an emissions value was obtained from the ambient data that was used in the study. In addition to these recent studies the long term trends in SO_2 obtained from the historical CSIR studies in these it showed a reduction in the SO_2 concentrations between the 1960s and 1970s and a plateau in concentration in the 1980s (Kemeny, 1980; Kemeny and Vleggaar, 1983).

The general decrease in PM concentrations observed across the

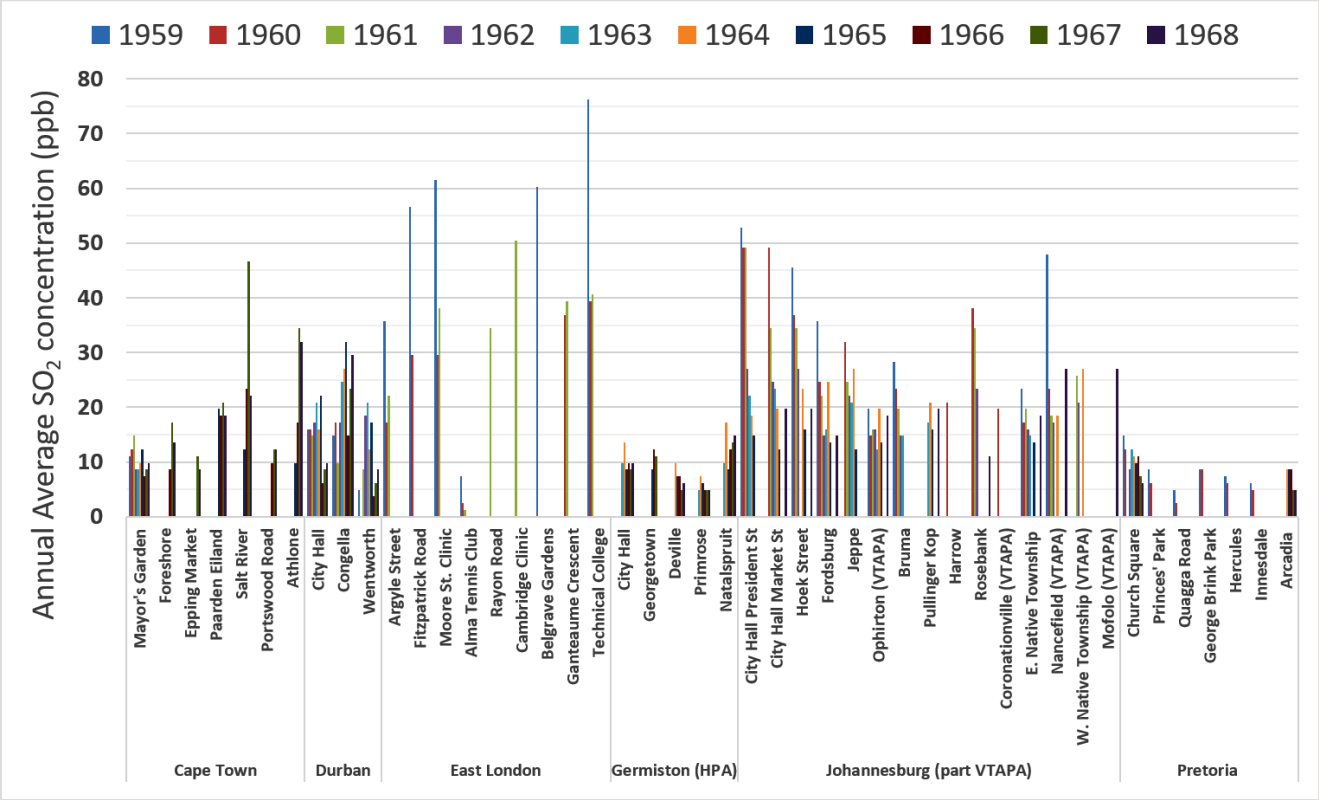

Figure 7: *Historical data of annual average SO_2 concentrations (ppb) in major South African cities 1959-1968. Data used in this analysis were previously published in Kemeny and Halliday (1972); Kemeny et al. (1980); Kemeny and Vleggaar (1983); Walker, Ellerbeck and Kemeny (1986).*

VTAPA and HPA indicates that improvements in air quality are occurring; however, there is still considerable variability in the rate of improvements between sites. For PM_{10}, the greatest rate of improvement in ambient concentration is seen for the Secunda and Zamdela sites at -4.35 and -4.68 µg/m^3/year respectively, while at Kliprivier and Witbank, no significant changes were observed; and a significant increasing trend was observed at Three Rivers. Based on the initial ambient concentrations and the rate of decrease in the PM_{10} concentration, the expected time until compliance with the annual NAAQS ranges from less than three years for Sebokeng to 49 years for Sharpeville, this is dependent on the assumption that the current linear trend is to continue. It is acknowledged that this assumption is a simplification, and thus is used for illustrative purposes in this context. Similarly, for $PM_{2.5}$, the rate of decrease in ambient concentrations ranges from -2.3 for Diepkloof to -0.3 µg/m^3/ year for Three Rivers. Compliance with the annual NAAQS is expected to take between 1 year and 39 years for Ermelo and Sharpeville, respectively. By the time the 2030 NAAQS for $PM_{2.5}$ comes into effect, it is expected that only Kliprivier, Sharpeville and Three Rivers will still be out of compliance, based on the current trends.

For both $PM_{2.5}$ and PM_{10}, the concentrations at Sharpeville showed a strong increase in 2016. It is not known whether this is the result of a short term temporary localised event or if it is indicative of a more significant local change in the emissions profile at the site.

As recently demonstrated through an assessment and cost performed for the entire country using the BENMAP model, significant impacts on health and associated economic costs for South Africa from not meeting the NAAQS for $PM_{2.5}$ (Altieri and Keen, 2019). Changes in the ambient concentration of PM_{10} and $PM_{2.5}$ are expected to have an impact on the ambient concentrations of co-emitted pollutants such as black carbon (Feig et al., 2015; Kuik et al., 2015).

In contrast to the decreasing trend in particulate matter, there is little to no trend in the concentrations of SO_2 over the VTAPA and HPA monitoring stations, where a significant increasing trend was found in five of the 11 sites. Four of the sites showed a negative trend in SO_2 (Diepkloof, Ermelo, Hendrina and Middelburg) which ranged between -7.3 ppb/year for Hendrina to -0.3ppb/year for Diepkloof. Supporting the previous findings, the ambient concentrations of SO_2 at the annual averaging period are in compliance with the NAAQS (Lourens et al., 2011; Venter et al., 2012; Wernecke et al., 2015).

Long term historical measurements at some of the major South African cities in 1960s to 1980s show a decreasing trend in SO_2 concentration during the 1960s and 1970s however it levelled out during the 1980s at the end of the measurement time series (Kemeny and Halliday, 1972; Kemeny, 1980; Kemeny and Vleggaar, 1983). During the time period between the observations reported in the Kemeny papers and the beginning of the observations in the VTAPA and HPA reported here there has been a considerable decrease in the ambient

concentrations, once again highlighting the importance of continuous observation record.

With the availability of a long term data set of ambient air quality concentrations, it is valuable to assess the state of air quality not just in terms of the compliance in terms of the annual standards, but also to examine the trend in concentration to determine how quickly sites are moving towards compliance and to focus attention on the locations where both the ambient concentrations are out of compliance and where little progress is being made towards meeting the NAAQS.

Conclusion

Despite the existence of the current air quality management regime for a period of 15 years (since the promulgation of the NEM:AQA in 2004) the air quality in the VTAPA and HPA is still considered to be poor and these areas are out of compliance with the PM_{10} and $PM_{2.5}$ NAAQS. However, in most instances over the monitoring time period, the ambient concentrations of particulate matter are improving and in some cases are improving fairly rapidly. These trends are not as evident for SO_2 concentrations, however, in contrast to PM, there is only one station where ambient SO_2 concentrations exceeded annual NAAQS.

This study is intended to provide a simple approach to identify where and at what rate the ambient air quality is improving, or to identify locations where improvements are not being observed. This serves as a guidance for air quality managers to consider and focus their management interventions. This analysis can be updated annually to continue to quantify trends, as long as data quality and data capture rates are high.

Author contributions

Gregor Feig conceptualised the study and wrote most of the text. Rebecca Garland contributed to the conceptualisation of the study and the review and drafting of the text. Seneca Naidoo contributed to the data analysis. Amukelani Maluleke contributed to the capture and analysis of the historical data. Marna Van der Merwe provided guidance on the Thiel-Sen analysis and provided valuable input in the review and editing of the paper.

Acknowledgements

We would like to thank DEA, SAAQIS and SAWS for the provision of the data and the hard work and dedication involved in collecting and processing the data.

This research was funded by the CSIR Parliamentary Grant. Mr Amukelani Maluleke's stay at the CSIR was made possible by the NRF internship program.

References

Altieri, K. E. and Keen, S. L. (2019) 'Public health benefits of reducing exposure to ambient fine particulate matter in South Africa', *Science of the Total Environment*. Elsevier B.V., 684, pp. 610–620. doi: 10.1016/j.scitotenv.2019.05.355.

Carslaw, D. C. and Ropkins, K. (2012) 'Openair - An R package for air quality data analysis', *Environmental Modelling and Software*. Elsevier Ltd, 27–28, pp. 52–61. doi: 10.1016/j.envsoft.2011.09.008.

DEA (2004) *National Environmental managment: Air Quality Act No. 39 of 2004*. South Africa.

DEA (2009) *National Environmental Managment Act (39/2004): National Ambient Air Quality Standards.*

DEA (2010) *National Environmental Management: Air Quality Act (39/2004): List of activities which result in atmospheric emissions which have or may have a significant detrimental effect on the environment, including health, social conditions, economic conditions, ec.*

DEA (2012a) *Declaration of the Waterberg National Priority Ares. South Africa.*

DEA (2012b) *National Ambient standard for particulate matter with aerodynamic diameter less than 2.5 micron meters ($PM_{2.5}$). South Africa.*

DEA (2013a) *National Environment Management: Air Quality Act, 2004(39/2004): List of activities which result in atmospheric emissions which have or may have a significant detrimental effect on the environment, including health, social conditions, economic conditions,.*

DEA (2013b) *National Environmental Management: Air Quality Act 2004 (Act No 39 of 2004) National Dust Control Regulations.*

DEA (2013c) *National Environmental Management Act: Declaration of a small boiler as a controlled emitter and estblishment of emission standards.*

DEA (2014a) *Declaration of Greenhouse Gases as Priority Air Pollutants.*

DEA (2014b) *Declaration of Temporary Asphalt Plants as a Controlled Emitter and establishment of emission standards.*

DEA (2015a) *Amendments to National List of Activities which result in atmospheric emission which have or may have a significant detrimental effect on the environment, including health, social conditions, economic conditions ecological conditions or cultural heritage.*

DEA (2015b) *Declaration of small-scale char and small-scale charcoal plants as controlled emitters and establishment of emission standards.*

DEA (2015c) *The Waterberg-Bojanala Priority Area Air Quality Management Plan and Threat Assessment.*

DEA (2016) '*Declaration of certain printing industry activities as controlled emitters and establishment of emission standards*'.

DEAT (2006) *Declaration of the Vaal Triangle Air-Shed priority area in terms of section18(1) of the National Environmental Management: Air Quality Act 2004, (Act No 39 of 2004).*

DEAT (2007) *Declaration of the Highveld as a priority area in terms of section 18 of the National Environmental Management: Air Quality Act (39/2004).*

ENATIS (2018) *eNATIS*. Available at: http://www.enatis.com/index.php/statistics/13-live-vehicle-population.

ESKOM (2011) *ESKOM Integrated Report 2011*. Johannesburg. Available at: http://financialresults.co.za/2011/eskom_ar2011/downloads/eskom-ar2011.pdf.

Feig, G. T. et al. (2015) 'Measurement of atmospheric black carbon in the Vaal Triangle and highveld priority areas', *Clean Air Journal = Tydskrif vir Skoon Lug*, 25(1), pp. 46–50. Available at: http://reference.sabinet.co.za/webx/access/electronic_journals/cleanair/clainair_v25_n1_a14.pdf%5Cnhttp://dx.doi.org/10.17159/2410-972X/2015/v25n1a4.

Feig, G. T., Naidoo, S. and Ncgukana, N. (2016) 'Assessment of ambient air pollution in the Waterberg Priority Area 2012-2015', *Clean Air Journal*, 26(1), pp. 21–28. doi: http://dx.doi.org/10.17159/2410-972X/2016/v26n1a9.

Garland, R. M. et al. (2017) 'Air quality indicators from the Environmental Performance Index : potential use and limitations in South Africa', *Clean Air Journal*, 27(1), pp. 33–41. doi: 10.17159/2410-972X/v27n1a8.

Goldswain, Z. (2016) 'Highveld Closed', *Witbank News*, 17 February. Available at: https://witbanknews.co.za/61365/highveld-closed/.

Hersey, S. P. et al. (2015) 'An overview of regional and local characteristics of aerosols in South Africa using satellite, ground, and modeling data', *Atmospheric Chemistry and Physics*, 15(8), pp. 4259–4278. doi: 10.5194/acp-15-4259-2015.

Kemeny, E. (1980) 'Long-Term Trends in smoke and Sulphur Dioxide Pollution in the Republic of South Africa', *Clean Air Journal*, 5(4), pp. 11–20.

Kemeny, E. and Halliday, E. C. (1972) 'The influence of yearly weather variations on smoke and sulphur dioxide pollution in Pretoria', *Archiv fur Meteorologie, Geophysik und Bioklimatologie, Serie B*, 20(1), pp. 49–78. https://doi.org/10.1007/BF02243314

Kemeny, E. and Vleggaar, C. M. (1983) 'Long-Term Trends in Smoke and Sulphur Dioxide Pollution in South Africa', *Clean Air Journal*, 6(4), pp. 21–22.

Kuik, F. et al. (2015) 'The anthropogenic contribution to atmospheric black carbon concentrations in southern Africa: A WRF-Chem modeling study', *Atmospheric Chemistry and Physics*, 15(15), pp. 8809–8830. doi: 10.5194/acp-15-8809-2015.

Lourens, A. S. M. et al. (2011) 'Spatial and temporal assessment of gaseous pollutants in the Highveld of South Africa', *South African Journal of Science*, 107(1/2). https://doi.org/10.4102/sajs.v107i1/2.269

Pretorius, I. et al. (2015) 'A perspective on South African coal fired power station emissions', *Journal of Energy in Southern Africa*, 26(3), pp. 27–40. Available at: http://www.scielo.org.za/scielo.php?script=sci_arttext&pid=S1021-447X2015000300004&nrm=iso. https://doi.org/10.17159/2413-3051/2015/v26i3a2127

R Core Team (2013) 'R: A language and environment for statistical computing'. Vienna: R Foundation for Statistical Computing.

Sangeetha, S. K. et al. (2017) 'SO_2 seasonal variation and assessment of Ozone Monitoring Instrument (OMI) measurements at Sharpeville (27.86°E; 26.68°S) a South African ground-based station', *International Journal of Remote Sensing*. Taylor & Francis, 38(23), pp. 6680–6696. doi: 10.1080/01431161.2017.1363433.

Sangeetha, S. K. and Sivakumar, V. (2019) 'Long-term temporal and spatial analysis of SO_2 over Gauteng and Mpumalanga monitoring sites of South Africa', *Journal of Atmospheric and Solar-Terrestrial Physics*. Elsevier Ltd, 191(March), p. 105044. doi: 10.1016/j.jastp.2019.05.008.

Senene, V. (2018) 'Vaal triangle Air-shed Priority Area Souce apportionment Study: Preliminary results'. Kimberley: DEA. Available at: http://www.airqualitylekgotla.co.za/assets/2018_5.4-vtapa-source-apportionment-study.pdf.

Shikwambana, L. and Tsoeleng, L. T. (2019) 'Impacts of population growth and land use on air quality . A case study of Tshwane , Rustenburg and Emalahleni , South Africa', *South African Geographical Journal*. Routledge, pp. 1–14. doi: 10.1080/03736245.2019.1670234.

Tshehla, C. and Wright, C. Y. (2019) '15 Years after the National Environmental Management Air Quality Act : Is legislation failing to reduce air pollution in South Africa ?', *South African Journal of Science*, 115(9), pp. 2–5. https://doi.org/10.17159/sajs.2019/6100

Venter, A. D. et al. (2012) 'An air quality assessment in the industrialised western Bushveld Igneous Complex, South Africa', *S Afr J Sci*, 108, pp. 1–10. https://doi.org/10.4102/sajs.v108i9/10.1059

Walker, N., Galpin, J. S. and Pienaar, A. E. (1987) 'Predicting Urban Smoke concentrations', *Clean Air Journal*, 7(4), pp. 18–19.

Walker, N. P., Ellerbeck, R. H. and Kemeny, E. (1986) 'National Survey of Smoke and Sulphur Dioxide: Quality of smoke measurements', *Clean Air Journal*, 7(1), pp. 27–29.

Wernecke, B. et al. (2015) 'Indoor and ambient particulate matter exposure on the Mpumalanga Highveld – a case study', *Clean Air Journal*, 25(15), pp. 12–16. https://doi.org/10.17159/2410-972X/2015/v25n2a1

Research article

An Environmental Justice Perspective on Air Quality Offsets

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Received: 18 October 2019 - **Reviewed:** 6 December 2019 - **Accepted:** 12 December 2019
https://doi.org/10.17159/caj/2019/29/2.7499

Abstract

Air quality offsets in South Africa are intended to counterbalance the harm caused by atmospheric emissions and deliver a net ambient air quality benefit in the affected airshed. In practice, they are implemented as a condition of leniency from compliance with the Minimum Emission Standards, and they focus on converting solid fuel burning households in low-income communities to cleaner forms of energy. Air quality offsets are not supported by all stakeholders, with non-governmental organisations in particular voicing vociferous objections. To date, there have only been very limited analyses of the ethical dimensions of air quality offsets. In this paper, air quality offsets and the Minimum Emission Standards are examined and compared from the perspective of three notions of environmental justice: distributive justice, which focuses on the distribution of environmental burdens and benefits; procedural justice, which considers inclusion and exclusion in decision- and policy-making processes; and justice as recognition, which focuses on the cultural and institutional processes that determine recognition, misrecognition and non-recognition of various groups. It is found that air quality offsets should guide action that promotes distributive justice because they are focussed on reducing polluting emissions in vulnerable, low-income communities that are exposed to the highest levels of ambient pollution. From a procedural justice perspective, South Africa's legislative processes provide for involving most stakeholders in decision-making processes. Air quality offset initiatives should be evaluated once they have been implemented at scale to determine whether they have indeed aided in redressing injustices. Assessment criteria could include whether the air quality-related health risk of vulnerable communities has been reduced, whether community members have participated in the design and implementation of interventions, and whether marginalised members of the community have benefitted from the interventions.

Keywords

air quality offsets, minimum emission standards, environmental justice.

Introduction

Air quality offsets are one of several measures introduced by the South African government to improve ambient air quality. In practice, they are implemented by polluting facilities as a condition of a postponement of compliance with the Minimum Emission Standards. They take the form of reducing emissions from domestic burning, waste burning and veld fires in low-income residential areas in close proximity to the polluting facilities.

Air quality offsets, as contemplated in the Air Quality Offsets Guideline, 2016 are unprecedented globally, in that the basis of equivalence is ambient air quality, and improvements in ambient concentrations of one pollutant, for example particulate matter (PM), may be traded for emissions of another pollutant, for example sulphur dioxide (SO_2). Emission trading schemes in other parts of the world take the form of market-based schemes that allow facilities to trade in pollution reduction credits. These are usually for the same pollutant, and are intended to achieve a mandated reduction in emissions at least cost (Krupnick, Oates and van de Verg, 1983; Drury et al., 1999).

Despite the focus of air quality offset projects on improving air quality in dense low-income communities that are exposed to the poorest ambient air quality in South Africa, offsets have received severe criticism from non-governmental organisations

(NGOs). The Centre for Environmental Rights (2015), for example, has categorically stated that they and their clients 'do not agree, in principle, with the use of offsets as a management tool to avoid compliance with legislation.' Another NGO, groundWork, has reported that Eskom's air quality offsets project 'has created more problems than solutions' (Molefe, 2018).

In light of the novelty and contentious nature of South Africa's air quality offsets programme, this paper assesses air quality offsets from an environmental justice perspective. Environmental justice here is taken to mean the ways in which the environment and social difference are intertwined, and the justice of this interrelationship (after Walker, 2012). An analysis of this nature is perhaps premature, given that air quality offsets have not been implemented at scale yet, and so the promotion (or not) of justice through the offsets programme cannot be evaluated. However, ethical theory can be used both to guide action and to evaluate actions (Driver, 2007). Criticism of the air quality offsets programme in South Africa has largely been directed towards the policy, and not so much the implementation of interventions. As such, the purpose of this analysis is to offer assertions as to whether air quality offset policy is adequately designed to guide actions that promote environmental justice. Thereafter, suggestions are made for evaluating the offset implementations from an environmental justice perspective.

Materials and methods

To provide the necessary background for the evaluation of air quality offsets from an environmental justice framework, legislation pertaining to air quality offsets in South Africa is reviewed, and offset implementation programmes are discussed based on Eskom and Sasol's plans, since between them they account for the majority of offsets initiatives. Environmental justice is introduced, drawing in particular on the work of Walker (2012), who identifies three concepts of justice – distributive justice, procedural justice and justice as recognition – which are discussed further in section 4.2. These three aspects of justice form the framework for the analysis in this paper. Use is made of Walker's (2012) proposal that claims about environmental justice should comprise of three elements (Figure 1):

i. an analysis of the evidence to determine the state of equality or inequality;
ii. an explanation identifying why the inequalities and injustices exist. This reasoning on why the inequality fails to satisfy a justice principle is linked to:
iii. a normative claim about justice/injustice.

Walker (2012) argues that the academic literature on environmental justice has tended to focus either on analysing justice concepts and theories, drawing on various philosophical and political traditions, or on the demonstration of patterns of inequality. An attempt is made in this study to explore both elements and the linkages between them in other to make a normative claim about what is just or fair.

The status of air quality on the South African Highveld is first assessed in terms of an environmental justice framework in order to provide context for the evaluation of air quality offsets. The term 'Highveld' is used here in the geographical sense, referring to the high-altitude plateau in the South African interior.

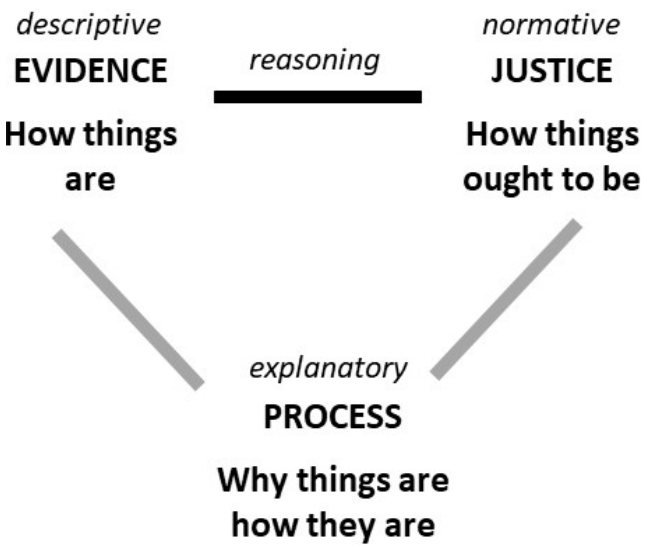

Figure 1: *The three elements of environmental justice claims (after Walker, 2012)*

Air quality offsets are evaluated through an environmental justice lens, relative to the Minimum Emission Standards for Listed Activities Identified in terms of Section 21 of the National Environmental Management: Air Quality Act, 2004 (henceforth referred to as the Minimum Emission Standards (MES)), since offsets are usually required as a condition of a delay in full or immediate compliance with the MES. This analysis focuses on South Africa's air quality offsets policy and regulatory framework, as led by the national Department of Environment, Forestry and Fisheries (DEFF, previously the Department of Environmental Affairs), and the way in which air quality offsets have been conceived of in Eskom and Sasol's implementation plans. Criteria for assessing the effectiveness of the implementation of offset projects from an environmental justice perspective are proposed.

This analysis takes place within the context of South Africa as 'a society based on democratic values, social justice and fundamental human rights' (according to the Preamble of the Constitution of the Republic of South Africa, 1996), and assumes political and societal views aligned with this.

Status of air quality offsets in South Africa

Air quality offsets are but one type of environmental offsetting in South Africa (Government of South Africa, 2015). Offsets are also employed for biodiversity, wetlands, water resources and carbon management. Environmental offsets are a response to the National Development Plan's statement that 'South Africa faces urgent developmental challenges in terms of poverty,

unemployment and inequality, and will need to find ways to "decouple" the economy from the environment, to break the links between economic activity, environmental degradation and carbon intensive energy consumption. In the past, resources were exploited in a way that was deeply unjust and left many communities excluded from economic opportunities and benefits while the natural environment was degraded. The country must now find a way to use its environmental resources to support an economy that enables it to remain competitive, while also meeting the needs of society' (Government of South Africa, 2015).

Legal status of air quality offsets

The Air Quality Offsets Guideline was published on 18 March 2016 (Notice 333, Government Gazette No. 39833) in terms of the National Environmental Management Act, 1998 (Act No. 107 of 1998) section 24J (a). The Guideline provides 'guidance on situations under which offsets can be applied during the implementation of the atmospheric emission licensing system…' The Guideline establishes principles for air quality offsets including that they are outcome based (on ambient air quality improvements); an offset does not need to be 'like for like'; there is transparency by the implementers and the authorities; receiving parties need to consent to participate; offsets projects should be sustainable in the long-term; and projects should be measurable and scientifically robust. It also stipulates that an offset programme should be subject to 'detailed and transparent' public participation.

The legal requirement to implement air quality offsets is stipulated in the atmospheric emission licences (AELs) of specific facilities, but there is almost no detail on what this entails. For example, the AEL for Sasol Synfuels issued on 31 March 2015 contains the condition that 'The facility must implement an offset programme to reduce PM and SO_2 pollution in the ambient air / receiving environment and the implementation plan is to be presented to the NAQO [National Air Quality Officer] and the licencing authority by 30 June 2015 after agreement, followed by an appropriate public participation process.' Duvha Power Station's AEL issued on 28 June 2017 stipulates that 'Eskom Duvha Power Station is required to provide and implement, a specific and time bound Atmospheric Emission Off-Set Plan to reduce PM in the ambient/receiving environment that must be approved by the Atmospheric Emission Licensing Authority annually.' Importantly, these conditions requiring air quality offsets in facilities' AELs were included as a requirement of an approved postponement of compliance with the MES.

Implementation of air quality offsets

Eskom and Sasol have published Air Quality Offset Implementation Plans for their facilities that are required to implement offsets. According to Sasol's offset plans for Secunda (Sasol, 2017) and for the Sasolburg operations and Natref (Sasol, 2016), Sasol's offset projects are implemented in Zamdela and eMbalenhle, the two large low-income residential areas in close proximity to their operations, and in Lebohang, which is a little further away, at the request of the Licencing Authority. Sasol intends to insulate up to 7600 solid fuel-burning formal homes in eMbalenhle (5200 homes) and Lebohang (2400 homes). Sasol expresses intent to insulate and swop stoves for 1400 to 1800 serviced informal homes by June 2020 (provided a successful insulation solution for informal dwellings is found). Sasol is assisting with veld fire management around Secunda and has looked into measures to suppress dust on untarred roads (although since their contribution to ambient air quality is minimal, they will not be pursuing this further). In Zamdela and surrounds, Sasol is focussing on reducing emissions from veld fires, waste burning and vehicles. Education and awareness activities are undertaken in all three communities, with special focus on the schools.

As of June 2018, Sasol had insulated 500 formal RDP houses in eMbalenhle. They had also completed the insulation of 24 serviced informal dwellings with spray polyurethane foam and swopped their coal-burning stoves for a low emission coal stove or liquid petroleum gas (LPG) stove and heater in Lebohang. In Sasolburg, 20 staff members from the Fire and Traffic Departments were trained in vehicle emission testing to promote routine vehicle testing by the local authorities. 100 waste skips were placed in Zamdela in 2018, and approximately 10 000 tons of waste removed from the community (Sasol, 2018). Eskom proposes to reduce emissions from domestic burning in 40 000 households on the Mpumalanga Highveld by 2025 by insulating the dwellings and swopping the coal stoves for a cleaner alternative (probably electricity or LPG heaters and stoves) (Eskom, 2017c, 2017a). Communities are selected for offsets based on the following criteria:

i. Communities exposed to highest ambient air quality impact from the facility are prioritised
ii. Only communities where there is non-compliance with ambient air quality standards may be selected
iii. Only communities where there is opportunity for offsets may be selected

In southern Gauteng near Lethabo Power Station, the focus is on reducing emissions from the burning of waste (Eskom, 2017b).

Eskom has completed a pilot study in KwaZamokuhle (adjacent to Hendrina town, Mpumalanga). Initially 120 households were insulated (60 with ceilings and 60 with ceilings and insulation on three walls) and each household received either an electricity subsidy (of R200 per month for the winter months) or their coal stove was swapped for a low emission coal stove, or their coal stove was swapped for an LPG heater and LPG stove (Langerman et al., 2018). Eskom subsequently insulated another 30 households and swapped their coal stoves for electric heaters and stoves. Eskom is currently busy with the contracting process to implement offsets on a larger scale.

Arguments for and against air quality offsets

Air quality offsets have been embraced by implementing industries and government. The Air Quality Offsets Guideline states that 'offsets are required to assist in sustaining required standards of environmental quality while achieving sustainable

rates of economic growth'. Eskom emphasizes the need to reduce exposure to air pollution at least cost, and propounds that 'household or community offsets are a more effective way of reducing human exposure to harmful levels of air pollution, than is retrofitting power stations with emission abatement technology at exorbitant costs'(Eskom, 2019).

Air quality offsets are vehemently opposed by NGOs, however. Their arguments against offsets are summarised in the submissions made by the Centre for Environmental Rights and their clients on the draft Air Quality Offsets Policy published in 2014, and on the draft Air Quality Offsets Guideline published in 2015 (Centre for Environmental Rights, 2014, 2015), and expanded on in Life After Coal's submission on Eskom's 2019 application for leniency from the MES (Centre for Environmental Rights, 2019). The NGOs 'do not agree, in principle, with the use of offsets as a management tool to avoid compliance with legislation', in particular the MES. Offsets themselves are perceived to have 'no overarching legislative or policy framework' (Centre for Environmental Rights, 2014). Furthermore, the design of offsets is considered to be flawed because a relaxation of SO_2 emissions from power stations, for example, may be traded for a reduction in ambient PM concentrations. They feel that 'offsets must result in a balancing of losses and gains in the same attribute or variable of concern,' (Centre for Environmental Rights, 2019), and that 'determining a reliable and defensible basis for determining equivalency between the impacts of regulatory relaxation and the offset effort is fundamental to any system of offsets' (Centre for Environmental Rights, 2014).

The NGOs also contend that the benefits of offsets are distributed unfairly. Air quality offsets are not a valid substitute for compliance with the MES, because the measures operate at different scales ('offsets could endorse higher levels of regional pollution while reducing pollutants at a localised level' (Centre for Environmental Rights, 2019)). They refer to 'implications for equity and justice' arising because 'offset activities may unfairly favour some communities at the expense of others (e.g. where electricity or gas subsidies are provided, or improvements/ retrofits to houses are made)' (Centre for Environmental Rights, 2014). In addition, 'the implications of offsets for public health will be affected by the timeframes permitted for implementing offsets and attaining required air quality standards' (Centre for Environmental Rights, 2014). For example, Eskom has not progressed beyond the pilot stage of their programme. Lastly, some polluting facilities may be unfairly advantaged over others by offsets because 'offsets represent a subsidy to pollution-generating activities' (Centre for Environmental Rights, 2014).

The NGOs purport that the responsibility for the implementation of offset-type projects should lie with the state, rather than with polluting industries. The CER perceives of air quality offsets as 'outsourcing government's responsibility toward human settlements in need of alternative forms of clean energy' (Centre for Environmental Rights, 2019) and proposes rather that 'the responsibility for tackling the problems of domestic air pollution would best be placed at the local authority and/ or community health level, supported by national policy' (Centre for Environmental Rights, 2014).

The views of households who have received offset interventions are not well known. The implementation is reported on favourably from research conducted by the offset project teams (most of the internal reports assessing the effectiveness of the pilot projects have not been published, but Eskom's 2017 progress report indicates that 80% of households who were approached to trade their coal stove for either a low emission coal stove or an LPG heater and stove agreed to participate, and of those more than 90% elected to keep their new stoves rather than swopping back to their old coal stoves after one winter (Matimolane, 2017)). However, a report by groundWork alleges that they 'discovered a number of challenges that the community is facing. From the way the project was presented and communicated, to the financial implication of replacing coal stoves with electric ones, to shoddy workmanship on ceilings and walls, to commitments and promises that were never fulfilled.' The two households that were interviewed in KwaZamokuhle reported a leaking roof, being unable to afford electricity for the electric stove that replaced their coal stove, insulation peeling off the walls, and being unable to open windows due to the wall insulation (Molefe, 2018).

The notion of justice

Environmental justice has been conceived within the much larger field of social justice.

Social justice

There are two schools of thought on justice that developed during the European Enlightenment. One approach focuses on identifying ideal institutions and associated rules of behaviour for a perfectly just society. This approach has been called "transcendental institutionalism" and has been led by the work of people such as Thomas Hobbes, Jean-Jacques Rousseau, Immanuel Kant and John Rawls.

Rawlsian justice is underpinned by the conception of 'justice as fairness.' Fairness can be seen as a demand for impartiality, where evaluations avoid bias, take note of the interests and concerns of others, and avoid being influenced by vested interests or prejudices. Rawlsian justice is concerned with setting up 'just institutions' that constitute the basic structure of society, and requires that people's behaviour complies entirely with the proper functioning of these institutions (Rawls, 1971). Rawls (1993) proposes two principles of justice:

i. Each person has an equal right to a full scheme of basic liberties which is compatible with a similar scheme of liberties for all.
ii. Social and economic inequalities are to be arranged so that they are attached to offices and positions open to all under conditions of fair equality of opportunity, and so that they are to the greatest benefit of the least advantaged members of society.

The second approach to justice is comparative rather than concentrating on ideals, and focuses on social realizations resulting from actual institutions and actual behaviour. This 'realization-focused comparison' has been elucidated by Adam Smith, Mary Wollstonecraft, Karl Marx and Amartya Sen, among others. Sen (2009) argues that an assessment of justice requires a focus on the lives that people are able to lead, not on just the institutions and behaviour compliant to these institutions.

Environmental justice draws from both schools of thought. The principle of 'justice as fairness' is frequently invoked. Environmental government departments are designed to be 'just institutions' and they pass legislation to ensure that society's behaviour complies with the requirements of their departments. In conceiving of environmental legislation, however, government is also very cognisant of the realities of vulnerable and marginalised people, and the need to find a balance between preventing environmental harm and supporting economic development.

Environmental justice

Environmental justice has been defined by the United States Environmental Protection Agency as 'the fair treatment and meaningful involvement of all people regardless of race, color, national origin, or income, with respect to the development, implementation, and enforcement of environmental laws, regulations, and policies.' Environmental justice takes place at different scales: for people, for communities and for non-human species and ecosystems. Inherent in environmental justice is the idea that disadvantaged people should not be subjected to disproportionate environmental impacts.

Three concepts of environmental justice have been elucidated by Walker (2012):

i. Distributive justice focuses on the distribution of environmental resources (positive) and harms (negative).
ii. Procedural justice is concerned with the way decisions are made, who is involved and has influence, and who has access to the formal justice system. It looks at inclusion and exclusion in decision-making processes around environmental and social issues.
iii. Justice as recognition emphasises who is given respect, and who is or is not valued, which is related to prejudice and discrimination.

The analyses that follow consider each of these three notions of environmental justice.

Environmental justice applied to ambient air quality on the South African Highveld

The environmental justice framework is first applied to ambient air quality on the South African Highveld (where offset programmes are currently targeted), in order to provide a context for the assessment of air quality offsets that follows. A distributive justice framework is adopted, in line with Walker's (2012) suggestion that it is most relevant when considering air quality.

Many of South Africa's mines, coal-fired power stations, smelters, other industries and a coal-to-liquids plant are clustered on the Highveld. A large portion of South Africa's population resides in the large metropolitan centres located there (26% of South Africa's total population resides in Gauteng alone (StatsSA, 2019)). The air pollution produced by the mining, industrial and urban activities has resulted in the declaration of two inter-provincial Priority Areas (the Highveld Priority Area in eastern Gauteng and Mpumalanga, and the Vaal Triangle Airshed in the northern Free State and southern Gauteng), where special management measures have been put in place to address the poor air quality.

The following three questions posed by Bell (2004) are examined when considering a distributive justice claim for air quality on the Highveld:

i. Who are the recipients of environmental benefits and burdens?
ii. What is to be distributed?
iii. What is the principle of distribution?

Firstly, the "community of justice" that matters when considering the environmental burdens and benefits of air quality on the South African Highveld are all people that breathe the air, which is all people living or working on the Highveld (and indeed, beyond).

Secondly, the environmental burden of poor air quality is often measured in terms of ambient air quality concentrations and compliance with ambient standards, but can perhaps be better quantified in terms of personal exposure. These measures are surrogates for an increase in the risk of contracting or dying from a number of diseases due to exposure to air pollution. It is crudely estimated that there are over 7 000 premature deaths in South Africa each year due to an increased incidence of stroke, ischemic heart disease, lung cancer, acute lower respiratory disease and chronic obstructive pulmonary disease (in order of decreasing significance) (WHO, 2016). Increased incidence of negative health outcomes (for example premature deaths or hospital admissions) provide a useful indication of the costs of poor air quality, because they also account for the vulnerability of those being exposed. These metrics point to the type of evidence needed to make judgements about justice in air quality matters.

Lastly, Bell (2004) has identified three principles that are generally applied in environmental justice scholarship to determine the right principle of distribution. These are first, a 'principle of equality', which in the case of air quality would mean that everyone should breathe air of uniform quality; second, a 'principle of equality plus a guaranteed standard', where there is no inequality and also a minimum standard ensured for all; and third, 'a guaranteed minimum with variation above that minimum according to personal income and spending choices', in which, beyond an ensured minimum, people can reasonable express their preferences. The ambient air quality management approach followed in South Africa, and indeed

in many countries, adopts the third principle of distribution, by legislating and then attempting to enforce ambient air quality standards (published in 2009 and 2012 by the Department of Environmental Affairs in South Africa).

Walker (2012) proposes that the focus of his environmental justice claim-making framework (Figure 1) used for a distributive justice analysis for air quality should be on patterns of exposure, vulnerability and responsibility. Claims regarding exposure to air pollution on the South African Highveld are made in Table 1, assuming that justice requires a guaranteed minimum standard of air quality (the National Ambient Air Quality Standards), with variation above that minimum according to preference.

There is widespread non-compliance with ambient PM standards across the Highveld. In 2018, there was non-compliance with the annual PM_{10} standard of 40 µg/m^3 at 14 of the 20 operational ambient air quality monitoring stations on the Highveld, and non-compliance with the ambient $PM_{2.5}$ annual standard at 8 of the 9 operational monitoring stations in the Vaal Triangle Airshed and the Highveld Priority Areas (Khumalo, 2019). $PM_{2.5}$ is monitored at very few urban sites in South Africa, so more monitoring is needed for conclusions to be drawn about air quality levels in cities.

Highest levels of PM are measured in low-income residential areas where domestic burning occurs (Hersey et al., 2015) (Figure 2). Ambient PM_{10} concentrations in the major urban areas are on average slightly higher than PM_{10} levels in industrial areas, although concentrations in industrial areas are usually higher than in urban areas in the summer. The actual levels of particulate pollution to which people are exposed are significantly higher than the ambient levels in communities that practise domestic burning, as demonstrated by Wernecke (2018) for KwaZamokuhle and Kwadela, coal-using communities on the Mpumalanga Highveld (Figure 2; Table 3).

Highest exposure levels among lower socio-economic groups is also widely found in North America and Europe, where it has been consistently shown that lower-income communities and minority groups are more exposed to air pollution from both point and mobile sources (Morello-Frosch and Jesdale, 2006; Tian, Xue and Barzyk, 2013; Taylor, 2014; Bullock, Ard and Saalman, 2018; Barnes, Chatterton and Longhurst, 2019; European Environment Agency, 2019). Similarly, in the Korba region in India, where coal mines and coal-fired power stations are clustered, marginalised social groups (the officially-designated Scheduled Caste (lowest caste) and Scheduled Tribe (indigenous people)) are disproportionately exposed to the environmental risks associated with coal extraction (Oskarsson and Bedi, 2018).

The high levels of exposure on the Highveld can be accounted for by the emissions of pollutants from a multitude of sources, the proximity of people to these emission sources, poor dwelling quality, and meteorology. Tall stack sources disperse pollutants effectively, which means that plumes are considerably diluted before they come to ground level, but they affect a large area. Emissions from surface sources such as domestic burning, waste burning, veld fires and vehicles are much lower in quantity, but a much higher fraction of the emissions is inhaled (called inhalation intake fraction) (Humbert et al., 2011). The poor quality of housing in low-income areas, which usually takes the form of uninsulated (i.e. no ceilings) formal RDP homes or poorly constructed informal dwellings, often built out of highly conductive materials, increases the energy demand for heating. Clean energy is often inaccessible or unaffordable for these communities. For example, around 22% of households

Table 1: *Environmental justice claims about air quality on the South African Highveld, based on the framework of Walker (2012)*

	Exposure	Vulnerability	Responsibility
Exposure	Widespread non-compliance with ambient PM standards. Highest exposure for low-income, solid/ liquid fuel-burning communities.	People living in low-income communities, children, the elderly, and people with pre-existing conditions are particularly vulnerable to air pollution.	Almost everyone is responsible for air pollution in some way. Greatest responsibility rests with large industries and power stations that emit highest quantities of pollutants, and solid/liquid fuel-using households who are responsible for emissions causing highest exposure.
Process	Exposure levels are high on the Highveld because of the multitude of sources, the high intake fraction of surface emissions and unfavourable dispersion conditions, especially in winter.	Low-income communities are highly vulnerable because of their poorer baseline health status, lower quality dwellings, inability to afford cleaner energy and an inferior standard of health care provided. Low-income communities are partly a legacy of South Africa's past.	Large industries and power stations are in many cases unable/unwilling to comply with emission standards. Energy poverty and no access to cleaner energy results in domestic use of 'dirty fuels'.
Justice	Exposure levels (including to indoor air) should not exceed National Ambient Air Quality Standards.	There should be monitoring of air to which the most vulnerable are exposed. There should be an adequate minimum standard of health care accessible to all.	Emission reduction should be financed by those most responsible for causing air pollution.

in Gauteng and 8% of households in Mpumalanga are not connected to the national electricity grid (StatsSA, 2019). Pollution is exacerbated by the sub-tropical climate on the Highveld. Low wind speeds result in stagnation of pollutants, frequent nocturnal temperature inversions trap air pollutants near the surface, recirculating air flow brings pollution back into the area after it has exited, and the lack of rain in the winter means the atmosphere is not effectively cleansed in these months (Tyson and Preston-Whyte, 2000).

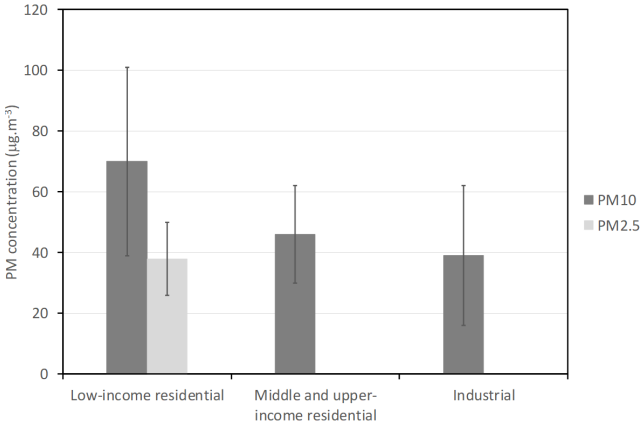

Figure 2: *Annual average PM_{10} and $PM_{2.5}$ concentrations (µg/m³) measured at the surface monitoring stations at different site types in South Africa's five major urban areas, the VTAPA and the HPA (after Hersey et al., 2015)*

Table 2: *Average daily particulate matter concentrations (µg/m³) for indoor air, outdoor air and personal exposure levels in two coal-using communities on the Highveld (Wernecke, 2018).*

Kwadela	**Winter 2013 & 2014**	**Summer 2014 & 2015**
Indoor PM_4	166	99
Personal PM_4	70	38
Ambient $PM_{2.5}$	36	18
KwaZamokuhle	**Winter 2016**	**Summer 2016**
Indoor PM_4	193	65
Personal PM_4	161	53
Ambient $PM_{2.5}$	70	42

The widespread non-compliance with ambient air quality standards on the Highveld is considered undesirable by all who reside in these areas. There is widespread agreement between almost all interested parties, from government to NGOs to industries and communities, on the need to achieve ambient air quality standards.

With regards to the vulnerability of people exposed to air pollution on the Highveld, health studies show that children, the elderly, and people with existing medical conditions are most vulnerable to the effects of poor air quality (Royal College of Physicians of London, 2016). In particular, gestation, infancy and early childhood are vulnerable times because organs are developing. The heart, brain, hormone systems and immunity can all be harmed by air pollution.

In addition to being exposed to higher levels of pollution, communities of lower economic status generally have compromised health status (CSDH, 2008) (because of limited access to sufficient healthy food, among many other factors). Furthermore, there appears to be an amplified "triple jeopardy" health impact, beyond the additive effect of higher exposure levels and compromised health status (Buzzelli, 2018). In South Africa, lower income communities are reliant on public health services, which are often inferior to the private health services available to higher income individuals. This further compounds the vulnerability of lower socioeconomic communities to health risks from air pollution.

A just society requires that specific measures are put in place to protect those who are more vulnerable to the effects of poor air quality. For example, an adequate minimum standard of health care should be available to all, with additional health care available to children and the elderly. Ambient air quality monitoring networks should ensure there is adequate coverage in low income communities so that the authorities are aware of the severity of the air quality in these areas and can take appropriate steps to manage the situation.

Lastly, there are different opinions as to how the responsibility for poor air quality should be allocated. Responsibility is typically assigned on the basis of quantities of emissions, but an argument could also be made for assigning responsibility based on contribution to ambient air quality or exposure (which depends not just on source strength but also on the proximity of people breathing the air to the source). Moreover, responsibility is usually assigned to the owner of a facility producing emissions, but responsibility could also be allocated to the consumer of the goods/services produced (except perhaps to consumers of electricity in South Africa, since there is a statutory monopoly on the generation, transmission and sale of electricity). In the case of the South African Highveld, almost everyone is responsible for the poor air quality in some way, but the greatest responsibility rests with large industries, power stations and related activities such as mining (DEA, 2011), high income individuals who own their own vehicles and consume a lot of electricity and other commodities, and solid/liquid fuel-burning households (who in some cases contribute the greatest amount of pollution to ambient levels in areas that experience the worst air quality in the country (e.g. Chidhindi et al., 2019)).

Emissions are usually linked to economic factors. Large industries and power stations are often unable or unwilling to comply with the MES because of the large costs associated with retrofitting abatement technology (Eskom, 2019). Low-income households are unable to afford cleaner sources of energy and so resort to using dirty fuels like coal, wood and paraffin (Pauw et al., 2008).

From a normative perspective, it is generally agreed that the

'polluter pays' principle should apply and action should be financed by those most responsible for causing air pollution.

An environmental justice analysis of air quality offsets

Air quality offsets are now considered from distributive justice, procedural justice and justice as recognition perspectives. Since air quality offsets are often implemented as a substitute for full compliance with the MES, the two pieces of legislation are contrasted.

Distributive justice

For an analysis of how air quality offsets fare in terms of distributive justice, we again answer the questions posed by Bell (2004) (Table 3), and then use Walker's (2012) claim-making framework, focusing on patterns of exposure, vulnerability, responsibility and access to resources.

Table 3: *The benefits, costs and principle of distribution for air quality offsets versus the Minimum Emission Standards*

	Air quality offsets	**Minimum Emission Standards compliance**
What is distributed?	Benefits: Reduced health risk, improved dwellings, greater cooking convenience?, greater indoor thermal comfort? Costs: Smaller increase in cost of production	Benefits: Reduced health risk Costs: Greater increase in cost of production, higher electricity tariff
Recipients of benefits/ burdens	Benefits: Usually low income communities	Benefits: Everyone in the (large) airshed Costs: Electricity users, consumers and shareholders
Principle of distribution	Communities selected for offsets based on ambient air quality impact of implementing facility, non-compliance with ambient standards, and opportunities for offsets	MES apply stricter standards to newer facilities than to older facilities. MES postponement applications adjudicated by the NAQO and Licencing Authorities.

Firstly, with reference to distributed benefits, both air quality offsets and the MES are designed to improve ambient air quality and reduce the health risk of exposed communities. Air quality offsets implemented at household level have other benefits that can favourably influence the quality of life of the household members, for example improved dwelling quality, more comfortable indoor temperature, and more convenient cooking facilities. The installation of emission abatement retrofits creates business and employment opportunities. The costs of the emission reductions are borne by the polluting facilities in both cases, and these in turn reduce the profits of the companies and/or are passed through to the consumers. Eskom estimates that the full cost of compliance with the MES is capital costs of R182 billion (in 2018 real terms) and annual operating costs of at least R5.9 billion per annum. This translates to the electricity tariff being 7-10% higher than it would be in the absence of emission abatement retrofits. Eskom has instead opted for a reduced emission reduction plan estimated to cost R67 billion (in 2018 real terms) over the next 10 years, with annual operating costs of R0.9 billion, and the implementation of air quality offsets (Eskom, 2019). In this case, air quality offsets as a substitute for full compliance with the MES avoid a 4-8% increase in the electricity tariff.

Secondly, the health benefits of emission reductions achieved due to reduction of industrial emissions are received by all who live in the airshed (often at a regional scale), while the benefits of offset projects are confined to the communities where the offsets are implemented.

There are several possible principles of distribution when selecting communities for air quality offset interventions. Eskom has prioritised communities primarily on the basis of impact of the polluting facility on ambient air quality, and then on need and feasibility (i.e. where opportunities for offsets exist). It would perhaps be more equitable to prioritise communities based on need (i.e. those experiencing the worst ambient air quality levels), irrespective of their location relative to the implementing facility (but this would be less palatable to communities who are impacted by the emissions from facilities). With regards to distributing the responsibility for reducing emissions between facilities, the MES applies stricter limits for facilities that were constructed after the standards were published, than for those that were constructed before the standards were published. Applications for postponement of compliance with the MES are adjudicated by the NAQO together with the Licensing Authorities.

I offer the following distributive justice claims about air quality offsets and the MES, with respect to exposure, vulnerability, responsibility and access to resources:

Apropos exposure to air pollution as a result of the implementation of air quality offsets, the evidence suggests that there will be large reductions in exposure to PM, and potentially also SO_2 and NO_x, but the reductions will be limited to recipient communities. Conversely, all people residing on the South African Highveld will experience small reductions in $PM_{2.5}$ exposure due to MES compliance. Communities close to large facilities that reduce their emissions will experience larger reductions in SO_2 exposure. Atmospheric dispersion of pollutants and the resulting differences in inhalation intake fraction are the processes that account for the differences in magnitude and geographical location of the reductions in

exposure. Sources addressed by offsets emit lower quantities of pollution almost directly into the air that people breathe, and pollutants may be trapped near the surface. Emissions from these ground-level sources usually have a high inhalation intake fraction (Humbert et al., 2011). Facilities targeted by the MES emit much higher quantities of pollutants from tall stacks, and these pollutants are transported to a large area. Because the pollutants are greatly dispersed before they reach the surface, the emissions from the facilities have an intake fraction several orders lower in magnitude than indoor emissions. From a normative perspective, as the NGOs point out, a basis for equivalence between emission reductions achieved through air quality offsets and the MES is required (Centre for Environmental Rights, 2014). This author is of the opinion that a 'like for like' equivalence will not be the most effective way of achieving exposure reductions through offsets, since polluting facilities are often unable to comply with SO_2 emission standards, for example, while the main pollutant of concern in low-income communities is PM. Instead, it is proposed that an equivalent reduction in health risks should be achieved by the offset project to that that would have been achieved through MES compliance, taking into account the entire domain influenced by emission reductions brought about by each of the two measures.

With regards to vulnerability, offsets specifically target emission reduction in low-income communities that are both exposed to the worst air quality (Hersey et al., 2015) and are more vulnerable to the effects of the air pollution because of their generally poorer health status and inferior health services provided. In particular, children and the elderly, who are particularly vulnerable to air pollution (Royal College of Physicians of London, 2016), often spend more time indoors than adults in formal employment, and will particular benefit from a reduction in domestic burning emissions. There is unfortunately a real danger of neglecting some of the most vulnerable households in the communities when implementing offsets, such as households residing in informal dwellings. Informal dwellings are non-uniform and often poorly constructed, and so cannot easily be insulated with the any of the methods used by Eskom and Sasol for formal houses. The difficulty in insulating these households may result in them being neglected entirely. Immigrants are also particularly vulnerable because they are less able to access state services like housing and health care.

In terms of the implications for justice, then, air quality offsets promote actions to redress injustices to vulnerable groups. Some of the most vulnerable households, like immigrants and those that reside in informal dwellings, should also receive offset interventions.

The responsibility for reducing emissions at facilities in order to comply with the MES clearly lies with the facilities themselves. The responsibility for implementing offsets currently also rests with the facilities, since the offsets are a condition of the facilities' AELs, and also with the recipients of the offset interventions (who are required to alter their cooking habits or maintain their new ceilings, for example). However, responsibility for some of the interventions being undertaken, including facilitating recycling and refuse removal, insulating dwellings and switching households to cleaner sources of energy usually lies with local authorities, government departments responsible for housing and those responsible for energy, respectively, and with households themselves. Indeed, section 26 (2) of the Constitution of the Republic of South Africa assigns the responsibility for the realisation of the right for everyone to have adequate housing to the state. Using existing departments to implement offsets would theoretically benefit from expertise and economies of scale that are already in place. However, then facilities could not be held responsible for offsets that do not deliver the expected improvement in ambient air quality. There is probably value in exploring different models of implementing offsets through government departments or another central agency.

Eskom and Sasol have tried to very carefully navigate the issue of disputed responsibility for service provision by not taking over any of the state functions of providing housing, removing refuse or testing vehicles, but have rather supported the state functions by insulating the dwellings, providing waste skips and training officers in vehicle emissions testing. The provision of cleaner energy is a function of both the state (through providing connections to the national electricity grid) and the market (through distributing fuels like LPG, coal, paraffin and wood). Eskom and Sasol have more aggressively intervened in this space in their attempts to switch fuels used by households.

From a normative perspective, then, according to the 'polluter pays' principle, facilities (and their customers) should be required to pay for their own emission reductions (which essentially is internalising the full costs of production). Since air quality offsets are a substitute for emission reduction at the facilities themselves, they should also be funded by the facilities. Polluting facilities should not perhaps take over responsibility for providing state services, but they can definitely support the state.

Large facilities obviously have access to more resources than low-income communities. Indeed, it could be argued that poverty is the main reason for poor air quality in low-income communities due to the lack of affordability of cleaner fuels, the poor quality of housing that necessitates a lot of energy for heating in winter, and even payment of low rates means basic services like refuse removal are inadequately funded. However, many polluting facilities also claim that they are unable to finance full compliance with the MES. The economic burden of emission reduction is minimised if reductions in exposure to poor air quality are achieved at least cost.

Procedural justice

Procedural justice is a second conception of environmental justice that complements distributive justice, and focuses on the procedure of justice rather than just the outcome of justice. Procedural justice is concerned with inclusion and exclusion in decision-making processes (Walker, 2012). The processes

Table 4: *Information publicly available on Air Quality Offsets and the Minimum Emission Standards*

Information provider	Information	Location
South African Air Quality Information System (SAAQIS)	All air quality acts, regulations and notices, strategies, policies, guidelines and municipal by-laws	https://saaqis.environment.gov.za
	Ambient air quality	
Parliamentary monitoring group	Summary of decisions on MES postponement applications by 31 March 2015	http://pmg-assets.s3-website-eu-west-1.amazonaws.com/171107Postponements_Report.docx
Sasol	2019 MES Postponement applications	https://www.srk.co.za/en/za-sasol-2019-mes-postponement-applications
	Previous applications for postponement of the MES, decisions issued and air quality offset implementation plans	https://www.srk.co.za/en/za-sasol-postponements
	Integrated Report/Sustainability Report provides annual summary of the status of offsets and postponement applications	e.g. http://www.integratedreport.sasol.com/sustainability/driving-sustainable-air-quality.php
Eskom	2019 MES postponement applications	http://www.naledzi.co.za/public-documents-naledzi.php
	Annual emission reports for 2017/18	http://www.naledzi.co.za/public-documents-naledzi.php
	Air quality offset implementation plans	http://www.eskom.co.za/AirQuality/Pages/PlansReports.aspx
Centre for Environmental Rights	CER's submissions on the offsets guidelines to the DEA	https://cer.org.za/programmes/pollution-climate-change/submissions-on-draft-regulations-guidelines-and-declarations
	Challenges to MES postponement applications by Eskom and Sasol	https://cer.org.za/programmes/pollution-climate-change/litigation
	Appeal of power stations' AELs	https://cer.org.za/programmes/pollution-climate-change/litigation
	Eskom's 2017 and 2018 MES applications and CER's objections.	

of scrutiny here are the formulation of the air quality offsets guideline and the Minimum Emission Standards, the decision on who is required to implement offsets, the decision on what the AEL requirements regarding offsets are, and the formulation of the facilities' air quality offset implementation plans.

The following aspects can be the subject of procedural justice claims (Stephens, Bullock and Scott, 2001; Schlosberg, 2007):

i. The availability of environmental information that is required for effective participation.
ii. Inclusion in environmental decision-making and policy-making processes in terms of who is able to participate and the respect given to participants.
iii. Access to legal processes for challenging decision-making and protecting environmental rights.
iv. Inclusion in community-based participatory research in which scientists collaborate with community members.

Again, Walker's (2012) framework for environmental justice claim-making (Figure 1) is followed, considering the evidence for inequality, an explanation as to why the inequality occurs and a normative claim about justice, with reference to these four points.

Most information pertaining to air quality offsets and the MES is available online (see Table 4 for a list). The internet and South Africa's laws, including the requirement for public participation in environmental processes and the Promotion of Access to Information Act, 2000, greatly facilitate the provision of information. There are, however, several key pieces of information that are not currently available but should be, including the scientific and economic studies underpinning the formulation of the MES, the documents from the MES standards setting process, and the reports on progress and effectiveness of offset implementation. The Eskom air quality offsets website has not been updated since 2017.

Environmental decision-making and policy development processes are generally inclusive. Industries and NGOs were well represented at the SABS standard-setting process for the MES. Unfortunately other affected parties like Labour were less well represented. The SABS process was eventually abandoned because consensus could not be reached by all parties. A period of public comment was given for the draft Air Quality Offsets Guideline and its preceding draft Air Quality Offsets Policy. Public participation is a legal requirement for industries when applying for postponement of compliance to the MES and developing air quality offset implementation plans. Public meetings on MES postponement applications and offset implementation plans are usually held by facilities in affected low-income communities to facilitate participation of more vulnerable people. The NGOs have made lengthy submissions on the major postponement applications and have challenged the wording of offset conditions in facilities' AELs (for example, the appeals lodged by the CER on behalf of their clients against the AELs of four power stations in 2016, available at https://cer.org.za/programmes/pollution-climate-change/litigation). Illiterate people, who comprise around 21% of South African adults between the ages of 35 and 64 (Statistics South Africa, 2017), still struggle to participate.

Industries frequently access legal processes, for example when requesting postponement of compliance to the MES, but access to legal processes by members of the public is generally hampered by the high cost of legal fees. Here NGOs, like the CER, groundWork and many others, play a vital role in giving a voice to marginal communities. There is a concern, however, that the interests of communities will not be represented if they conflict with the interests of the NGOs. In a just society, all would have equal access to legal processes, regardless of income or education level.

Detailed community-based research has been conducted in support of the air quality offsets programmes, including the conducting of quality of life assessments prior to and after implementation of the intervention, and a comprehensive assessment of the effectiveness of the interventions. Not only have households been the subject of rigorous research, but members of the communities have participated in the research through conducting household surveys and operating ambient air quality monitoring instrumentation sited in the community, for example. (This information has been directly communicated to the author by members of the implementation team in KwaZamokuhle). Unfortunately, most of this research has only been published in confidential industry reports, and needs to be published in the public domain.

Justice as recognition

Justice as recognition is concerned with who is respected or valued and who is discriminated against (Walker, 2012). Schlosberg (2004) argues that misrecognition is fundamental to the production of distributional inequalities. Misrecognition may occur by institutions of the state or more subtly through social norms and the way in which people interact with each other.

While South African environmental legislation has come a long way in recognising all individuals, everyone is certainly not recognised equally. Studies show that, in many instances, racism and gender discrimination apply just as much to air quality issues as to general societal issues (Drury et al., 1999; Siddiqui et al., 2005 for example). There are a few other groups of people who should be mentioned. The illiterate are often invisible. In addition, immigrants are often maligned or disparaged. They also do not have access to all the services that South Africans do, such as health care (even if legally entitled to it) (Lepodise, 2018) and housing. Undocumented migrants are particularly marginalised because of their insecure legal status. An estimated four million migrants are hosted in South Africa currently (United Nations, 2017).

Evaluating air quality offset implementation

The environmental justice implications of air quality offsets can, of course, only properly be evaluated once offset interventions have been executed on a large scale. Some principles for evaluating the roll-out of offset projects from an environmental justice perspective are proposed here. Exact metrics still need to be developed, preferably in consultation with a larger group of affected parties. These metrics should ideally be incorporated into an air quality offsets standard which regulates offset planning, implementation and reporting requirements.

The promotion of distributive justice will depend on whether the air-quality related health risk of vulnerable communities has been reduced through the implementation of an offset initiative. Health risk depends on susceptibility (which depends on things like nourishment, age and pre-existing health conditions of community members), exposure to air pollution, and access to health care. Air quality offsets specifically target reducing exposure through reducing emissions (although the awareness programmes may also reduce exposure through encouraging people to avoid particularly smoky environments, for example), but they may also indirectly reduce susceptibility of people to air pollution-related illnesses through means such as improving the thermal comfort of dwellings.

The simplest metric to measure the effectiveness of air quality offsets is avoided emissions (relative to the baseline). Distributive justice is served if inequalities in exposure between lower income communities and higher income communities is reduced. Changes in quality of life of community members affects vulnerability and susceptibility to air quality-related health risks, so metrics also need to take into account indirect costs and benefits of an intervention, such as changes to housing quality and thermal comfort, training provided and jobs created. Certainly, an offset intervention should not result in a net decline in quality of life.

The question then arises as to what a sufficient offset for a facility is. If air quality offsets are viewed as a strict substitute

for compliance with the MES, then the equivalence could be determined in terms of ambient air quality levels, exposure levels, or health risk. Impact on health risk is the most complete metric, but the most difficult to measure.

The principles of the Air Quality Offsets Guideline encourage procedural justice in air quality offset programmes. Specific requirements for a procedurally just community intervention could include that community members participate meaningfully in the design and implementation of an intervention; that the community has access to information about their exposure and their participation in the project (such as ambient air quality measurements and results of community-based research); and that their participation is voluntary. Consultation should be at household level, and not just with community leaders.

To advance justice as recognition, interventions should be designed to be as inclusive as possible of groups who are particularly vulnerable or who tend to be marginalised, such as immigrants, the illiterate, people who live in informal dwellings, the unemployed and even women. The inclusion of under-recognised people could be promoted through the development of criteria upfront regarding who is to receive offset interventions. These criteria should favour the most marginalised communities and community members, as far as technical considerations allow.

Conclusions

Air quality offsets have been assessed from an environmental justice perspective, considering the context of ambient air quality on the South African Highveld where low-income communities are exposed to the highest air pollution levels. The potential of the Air Quality Offsets Guideline to guide environmentally just action has been compared with that of the MES, since offsets are usually implemented as a condition of postponement of compliance with the MES.

In summary, air quality offsets, as conceived in the Air Quality Offsets Guideline (2016) and the industries' offset implementation plans, have the potential to be used to promote environmental justice. From a distributive justice perspective, air quality offsets focus air quality improvements on vulnerable communities that are exposed to highest concentrations of pollution. In terms of procedural justice, decision-making and policy formulation processes are generally inclusive, but lower socio-economic groups are still somewhat disadvantaged in their access to legal processes and information like evaluations of offset effectiveness. Offsets can play a role in achieving the Sustainable Development Goals, particularly goals 3 (good health and well-being), 7 (affordable and clean energy), 10 (reduced inequalities) and 11 (sustainable cities and communities).

The realisation of the potential of offsets to promote environmental justice can only be determined once offsets have been implemented at scale. Such an assessment of the results of offset interventions should holistically consider the many factors that influence the effects of poor air quality on illness (including the underlying vulnerability and quality of life of members of a community), as well as the air quality in the community relative to the baseline air quality in the absence of the interventions, and relative to nearby higher income communities. It should also consider the effective participation of the community in the offsets project design and implementation, and be as inclusive as possible in terms of which households are selected to participate.

Note

An earlier version of this paper was presented at the National Association for Clean Air (NACA) Conference in October 2019 and was published in its Proceedings.

References

Barnes, J. H., Chatterton, T. J. and Longhurst, J. W. S. (2019) 'Emissions vs exposure: Increasing injustice from road traffic-related air pollution in the United Kingdom', *Transportation Research Part D: Transport and Environment*, 73, pp. 56–66. doi: 10.1016/j.trd.2019.05.012. https://doi.org/10.1016/j.trd.2019.05.012.

Bell, D. (2004) 'Environmental justice and Rawls' difference principle', *Environmental Ethics*, 26(3), pp. 287–306. https://doi.org/10.5840/enviroethics200426317.

Bullock, C., Ard, K. and Saalman, G. (2018) 'Measuring the Relationship between State Environmental Justice Action and Air Pollution Inequality, 1990–2009', *Review of Policy Research*, 35(3), pp. 466–490. https://doi.org/10.1111/ropr.12292

Buzzelli, M. (2018) 'Air pollution and respiratory health: Does better evidence lead to policy paralysis?', in Holifield, R., Chakraborty, J., and Walker, G. (eds) *The Routledge Handbook of Environmental Justice*. Oxon: Routledge, pp. 327–337. https://doi.org/10.4324/9781315678986-26

Centre for Environmental Rights (2014) 'Comments on the Department of Environmental Affairs' Draft Air Quality Offset Policy'. Available at: https://cer.org.za/wp-content/uploads/2016/08/CER-submissions-on-Draft-AQ-Offset-Policy_21-Feb-2014_final.pdf (Accessed: 14 July 2019).

Centre for Environmental Rights (2015) 'Submissions on Department of Environmental Affairs' Draft Air Quality Offsets Guideline dated January 2015'. Available at: https://cer.org.za/wp-content/uploads/2016/08/Offsets-AQ-Gdlne-CER-27-July-2015_final.pdf (Accessed: 13 July 2019).

Centre for Environmental Rights (2019) 'Submissions on Eskom's application for suspension, alternative limits and/or postponement of compliance with the Minimum Emission Standards of 10 of its coal-fired power stations'.

Available at: http://www.naledzi.co.za/assets/documents/e11350a1ea6af4657cf960c82e3ec60b.pdf (Accessed: 14 July 2019).

Chidhindi, P. et al. (2019) 'Assessing the impact of Eskom power plant emissions on ambient air quality over KwaZamokuhle', *Clean Air Journal*, 29(1). https://doi.org/10.17159/2410-972x/2019/v29n1a2.

CSDH (2008) *Closing the gap in a generation: Health equity through action on the social determinants of health*. Final Report of the Commission on Social Determiinants of Health. Geneva: World Health Organisation.

DEA (2011) *Highveld Priority Area Air Quality Management Plan*. Pretoria: Department of Environmental Affairs, p. 233. Available at: http://www.saaqis.org.za/documents/HIGHVELD%20PRIORITY%20AREA%20AQMP.pdf.

Driver, J. (2007) *Ethics: The Fundamentals*. Malden, MA: Blackwell (Fundamentals of Philosophy).

Drury, R. T. et al. (1999) 'Polluting trading and environmental justice: Los Angeles' failed experiment in air quality policy', *Duke Environmental Law and Policy Forum*, 9, pp. 231–289.

Eskom (2017a) *Air Quality Offsets Implementation Plan for Gert Sibande District Municipality: Tutuka, Majuba, Camden and Grootvlei Power Stations: March 2017 update*. ENV17_R074. Sunninghill: Eskom Holdings SOC Limited, p. 59. Available at: http://www.eskom.co.za/AirQuality/Documents/AQoffsetPlanGertSibandeMar17.pdf.

Eskom (2017b) *Air Quality Offsets Implementation Plan for Lethabo Power Station: March 2017 update*. ENV17-R075. Sunninghill: Eskom Holdings SOC Limited, p. 61. Available at: http://www.eskom.co.za/AirQuality/Documents/AQoffsetPlanLethaboMar17.pdf.

Eskom (2017c) *Air Quality Offsets Implementation Plan for Nkangala District Municipality: Hendrina, Arnot, Komati, Kriel, Matla, Kendal and Duvha Power Stations: March 2017 update*. ENV17_R073. Eskom Holdings SOC Limited, p. 66. Available at: http://www.eskom.co.za/AirQuality/Documents/AQoffsetPlanNkangalaMar17.pdf.

Eskom (2019) *Applications for suspension, alternative limits and/or postponement of the Minimum Emission Standards (MES) compliance timeframes for Eskom's coal and liquid fuel fired power stations: Summary document.* Eskom ENV18-R245 rev 2.1. Available at: http://naledzi.co.za/assets/documents/21fc8413fc875fff57cdbe6c68916241.pdf.

European Environment Agency (2019) *Air quality in Europe - 2019 report*. 10/2019. Copenhagen: European Environment Agency. doi: 10.2800/822355. https://doi.org/10.1016/S0306-3747(19)30103-4

Government of South Africa (2015) 'Environmental Offsets Discussion Document'. Department of Environmental Affairs.

Hersey, S. P. et al. (2015) 'An overview of regional and local characteristics of aerosols in South Africa using satellite, ground, and modelling data', *Atmospheric Chemistry and Physics*, 15, pp. 4259–4278. https://doi.org/10.5194/acp-15-4259-2015.

Humbert, S. et al. (2011) 'Intake Fraction for Particulate Matter: Recommendations for Life Cycle Impact Assessment', *Environmental Science & Technology*, 45(11), pp. 4808–4816. https://doi.org/10.1021/es103563z.

Khumalo, T. (2019) '2018 State of the Air Report and National Air Quality Indicator', in *14th Air Qulity Governance Lekgotla*. Stellenbosch: Department of Environmental Affairs.

Krupnick, A. J., Oates, W. E. and van de Verg, E. (1983) 'On marketable air-pollution permits: the case for a system of pollution offsets', *Journal of Environmental Economics and Management*, 10, pp. 233–247. https://doi.org/10.1016/0095-0696(83)90031-1

Langerman, K. E. et al. (2018) 'Moving Households to Cleaner Energy through Air Quality Offsets', in *Proceedings of the Domestic Use of Energy Conference 2018. Domestic Use of Energy Conference 2018*, Cape Town, pp. 2–9. Available at: https://drive.google.com/drive/folders/1BwgLT6OLXN-Wh2KE4vjt_RaaQc180h2R.

Lepodise, O. (2018) 'Medical Xenophobia: Public hospitals deny migrants health care services – SAHRC', *Daily Maverick*, 29 March.

Matimolane, M. (2017) *Eskom Air Quality Offset Programme: Progress Report March 2017*. ENV17-R070. Eskom Holdings SOC Limited, p. 18. Available at: http://www.eskom.co.za/AirQuality/Documents/ProgressReportMar2017.pdf (Accessed: 14 September 2019).

Molefe, T. (2018) 'Government: Wheels come off the Eskom offset'. Available at: https://www.groundwork.org.za/archives/2018/news20180814-Wheels_come_off_the_Eskom_offset.php (Accessed: 13 July 2019).

Morello-Frosch, R. and Jesdale, B. M. (2006) 'Separate and unequal: Residential segregation and estimated cancer risks associated with ambient air toxics in U.S. metropolitan areas', *Environmental Health Perspectives*, 114(3), pp. 386–393. https://doi.org/10.1289/ehp.8500

Oskarsson, P. and Bedi, H. P. (2018) 'Extracting environmental justice: Countering technical renditions of pollution in India's coal industry', *The Extractive Industries and Society*, 5(3), pp. 340–347. doi: https://doi.org/10.1016/j.exis.2018.05.003.

Pauw, C. J. et al. (2008) *Air pollution in dense, low-income settlements in South Africa: Issue 1*. Report to The Royal Danish

Embassy and the Dept. of Environmental Affairs and Tourism.

Rawls, J. (1971) *A Theory of Justice*. Cambridge, MA: Harvard University Press.

Rawls, J. (1993) *Political liberalism*. New York: Columbia University Press.

Royal College of Physicians of London (ed.) (2016) *Every breath we take: the lifelong impact of air pollution: report of a working party*. London: Royal College of Physicians of London.

Sasol (2016) *Joint Offset Implementation Plan: Sasol Sasolburg operations and Natref*. Sasol Limited. Available at: https://www.srk.co.za/sites/default/files/File/South-Africa/publicDocuments/SASOL_Postponements/SASOLBURG_NATREF_OFFSET_IMPLEMENTATION_PLAN.pdf.

Sasol (2017) *Offset implementation plan: Sasol Secunda*. Sasol Limited, p. 64. Available at: https://www.srk.co.za/sites/default/files/File/South-Africa/publicDocuments/SASOL_Postponements/SECUNDA_OFFSET_IMPLEMENTATION_PLAN.pdf.

Sasol (2018) *Sasol Limited Sustainability Report*. Sasol Limited. Available at: http://www.integratedreport.sasol.com/sustainability/ebook/files/assets/common/downloads/publication.pdf?uni=fb96ccdb3507d367ba26d0f42a4565ad.

Schlosberg, D. (2004) 'Reconceiving environmental justice: global movements and political theories', *Environmental Politics*, 13(3), pp. 517–540. https://doi.org/10.1080/09644010420002290 25.

Schlosberg, D. (2007) *Defining environmental justice: Theories, movements and nature*. Oxford: Oxford University Press. https://doi.org/10.1093/acprof:oso/9780199286294.001.0001.

Sen, A. (2009) *The Idea of Justice*. Cambridge, MA: Harvard University Press. https://doi.org/10.2307/j.ctvjnrv7n

Siddiqui, V. et al. (2005) 'Eye and respiratory symptoms among women exposed to wood smoke emitted from indoor cooking: a study from southern Pakistan', *Energy for Sustainable Development*, 9(3), pp. 58–66. https://doi.org/10.1016/S0973-0826(08)60524-4.

Statistics South Africa (2017) *Education Series Volume III Educational Enrolment and Achievement*, 2016. 92-01–03. Pretoria, p. 120. Available at: http://www.statssa.gov.za/publications/Report%2092-01-03/Report%2092-01-032016.pdf. StatsSA (2019) 'General Household Survey 2018'. Statistics South Africa.

Stephens, C., Bullock, S. and Scott, A. (2001) *Environmental Justice: Rights and means to a healthy environment*. Swindon: ESRC Global Environmental Change Programme.

Taylor, D. (2014) *Toxic Communities: Environmental Racism, Industrial Pollution*, and Residential Mobility. NYU Press.

Tian, N., Xue, J. and Barzyk, T. (2013) 'Evaluating socioeconomic and racial differences in traffic-related metrics in the United States using a GIS approach', *Journal of Exposure Science and Environmental Epidemiology*, 23(2), pp. 215–222. https://doi.org/10.1038/jes.2012.83.

Tyson, P. D. and Preston-Whyte, R. A. (2000) *The Weather and Climate of Southern Africa*. Oxford University Press.

United Nations (2017) I*nternational Migration Report 2017*. New York.

Walker, G. (2012) *Environmental justice: Concepts, Evidence and Politics*. Oxon: Routledge. https://doi.org/10.4324/9780203610671.

Wernecke, B. (2018) *Ambient and Indoor Particulate Matter Concentrations on the Mpumalanga Highveld*. MSc dissertation. North-West University.

WHO (2016) *Ambient air pollution: a global assessment of exposure and burden of disease*. Geneva: World Health Organisation.