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"Pathways to effective air quality management in South Africa"

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The 2022 Conference of the National Association for Clean Air will again be held back-to-back with the Department of Forestry, Fisheries and the Environment's Air Quality Governance Lekgotla.

The NACA Council has decided that the 2022 Annual NACA Conference will take place in Gauteng and will be held onsite (face-to-face) only. The theme of this year's conference is ***"Pathways to effective air quality management in South Africa"***. We will advise as soon as a conference venue has been confirmed in conjunction with our colleagues from the Department of Forestry, Fisheries and the Environment (DFFE)

The conference will start with the joint DFFE and NACA Multi-Stakeholder Workshop on Wednesday morning followed by lunch.

After lunch, NACA will host a **technical session**. The topic of this session will be confirmed. The **GCRF Mine Dust & Health Network** have advised that they will also be hosting a workshop again as an integral part of this year's conference line-up.

The conference will be opened by NACA's president. We look forward to receiving feedback from the 16th Air Quality Governance Lekgotla and to seeing our colleagues from the public sector again.

We are planning a full and interesting programme with a host of keynote and invited speakers to address all attendees on current air quality topics. We expect to host two very full and interesting conference days with between 25 and 30 research papers.

We furthermore wish to call on our generous sponsors who have continued to support us during the pandemic and hope that we can offer you an even better return on your support during the upcoming onsite event.

We will revert back to you soon to confirm the conference venue and with a sponsorship and exhibition prospectus.

We truly look forward to seeing all of you again as there is nothing quite like a face-to-face event to network, and bring together researchers and practitioners from academia and industry to report on the latest scientific and technical advances.

The conference will also feature poster sessions and possibly three-minute talks.

Day two of the conference will get off to an early start and is expected to end at five o'clock on Friday afternoon.

The Organising Committee of the 2022 NACA Conference invites submissions of papers for the annual conference. Presenters are requested to register and submit their abstracts on the electronic submission and evaluation system. Guidelines for papers and posters will be made available on the conference page of the NACA website.

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31 August 2022

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7 September 2022

Authors resubmit papers if required indicating how reviewers' comments were addressed
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Editorial
Ambient air pollution on the Highveld: An airshed at a watershed moment?

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Background

On 18 March 2022, the Pretoria High Court found that chronic air pollution is a violation of Section 24 of the South African Constitution, and that South Africans have a right to an environment that is not harmful to their health and well-being (CER, 2022). This "has important implications for communities forced to live with the debilitating effects of air pollution on the Mpumalanga Highveld, and more broadly for constitutional jurisprudence and government accountability" (CER, 2022).

Since 2019, two environmental justice groups, groundWork and the Vukani Environmental Justice Movement, represented by the Centre for Environmental Rights (CER), sought recourse from the High Court on the high air pollution levels in the Highveld Priority Area (HPA). The basis of this "Deadly Air" Case was a declaration that "the poor air quality in the Highveld Priority Area is a breach of the residents' right to an environment that is not harmful to their health and wellbeing" (CER, 2022).

The HPA is known for its poor air quality. Numerous studies have reported widespread non-compliance with the PM and O_3, as well as NO_x and SO_2 National Ambient Air Quality Standards (NAAQS) (Steyn and Kornelius 2018; Feig et al., 2019; Chindhindi et al., 2019; Morosele and Langerman 2020). There are a range of air pollution sources which contribute to the poor air quality in the HPA, including industry, roads, vehicles, mining, power generation, biomass burning, wind-blown dust, domestic fuel use practices and waste burning, to name a few (Ross et al., 2007; Nkosi et al., 2018; Walton et al., 2021). The negative impacts of the air pollution are felt by many who reside on the Highveld and even beyond. Much pressure has been placed on government by civil society and legal experts to improve the air quality in the region, leading to many difficult discussions. Consequently, over the past years, the air quality in the airshed has become the focus of many air pollution-related research studies, legal debates and media releases.

Legislation governing air quality on the Highveld and the Minimum Emission Standards

A comprehensive set of laws, standards and guidelines exists in South Africa to govern air quality. This stems from the Constitution, which ensures that clean air is a fundamental human right (RSA, 1996). Under the National Environmental Management: Air Quality Act, 2004 (RSA, 2005), standards exist to govern the levels of pollutants that are emitted into the air from major industrial activities (e.g., Minimum Emission Standards (MES) (RSA, 2019) and Dust Control Regulations (RSA, 2013)). Similarly, standards have been set to measure the ambient air quality at ground-level, where people breathe (RSA, 2009b; RSA 2012a). A National Framework outlines the governance, measurement and reporting tools (e.g., South African Atmospheric Emission Licencing and Inventory Portal (SAAELIP)) systems and procedures for monitoring air quality and implementing air quality management strategies (DEA, 2018). This includes the declaration of Air Quality Priority Areas as well as the drafting of Air Quality Management Plans (RSA, 2007; RSA, 2009a; RSA, 2012b; DEA, 2012).

Larger emitters require an Atmospheric Emission Licence to operate and are mandated to comply with the MES for specific criteria pollutants at source. When created, the MES phased in more lenient emission limits in 2015 and stricter limits in 2020. The MES are the regulations that have the authority needed to achieve significant emission reductions from large sources.

Despite the existence of a world class legislative framework in South Africa to govern air quality, its implementation and compliance to its regulations has often been difficult to achieve. Many industries report that they are unable to comply fully with the MES, citing the unaffordable cost of emission abatement retrofits as the primary reason. Aging technology, water and space constraints, and production considerations are other reasons given for the inability to reduce emissions by the stipulated dates. Larger facilities in particular, like power stations and oil refineries, have struggled to comply. Some of these facilities have applied to the Department of Forestry, Fisheries and the Environment (DFFE) for postponement of the MES compliance timeframes, essentially asking government to allow them to continue operating as usual until the facilities are able to comply, or until they reach the end of their life.

This has been a highly contentious and ongoing legal wrangle. In the most recent developments, and more than a decade after the MES were introduced, government has rejected many postponement applications which will potentially result in many facilities being forced to cease operations.

What lies ahead?

The High Court decision ordering the DFFE to clean up the air has been widely welcomed, but how this will be achieved is still unclear. Going forward, protracted legal battles delaying action to reduce pollution, or the possible premature closure of facilities without appropriate replacements which will hamper economic growth, lead to job losses and an increase in poverty, should ideally be avoided. Unemployed people are often unable to afford clean energy carriers and are exposed to the highest pollution levels in the country (Hersey et al., 2015). Achieving a balance between emission reduction and economic growth is essential.

Minister Creecy has indicated that the DFFE will be drafting new air quality regulations, and that they will continue to engage with communities, experts, NGOs, energy producers and other stakeholders in the area. It is essential that other government departments and agencies – those dealing with housing, health, energy and water – also are active in the dialogue and way forward. The new regulations can only be effective if different interest groups agree, that cleaning the air we breathe is a priority, in order to save lives. Are consumers of some products (like electricity) willing to pay the higher prices needed to fund the emission abatement retrofits? Can water be repurposed from other uses for pollution abatement? Furthermore, resources need to be available to enable compliance with the new regulations.

Beyond the need to reduce emissions from significant pollutant sources, the approach focus for emission reductions needs to be holistic. Studies show that many sources contribute to pollution levels. In addition to large point sources like power stations, smelters, refineries and industries, smaller source like vehicle emissions, unpaved roads, residential wood/coal burning, waste burning, veld fires and mines can also be extremely important at a local level. Prioritising sources based on their contribution to exposure levels is a sensible approach which can have large impacts on ambient pollution levels while not absolving large emitters from meeting their regulatory requirements. Limited resources can be assigned to emission reduction from sources that have the greatest impact on health, informed by a robust evidence base (e.g., outcomes from cost-benefit analysis).

Conclusion

Whatever the approach adopted, it is clear that the HPA will remain an air quality hotspot for the foreseeable future, posing continued health risks to people living in the region. Given the increased pressure on industry and government to reduce emissions, to transition to cleaner energy and ultimately to improve ambient air quality, it seems as though the HPA may have reached a watershed moment – one which we hope will see the airshed move towards cleaner air.

References

Centre for Environmental Rights (CER). (2022). Major court victory for communities fighting air pollution in Mpumalanga Highveld. Available at: https://cer.org.za/news/major-court-victory-for-communities-fighting-air-pollution-in-mpumalanga-highveld.

Chidhindi, P., Belelie, M. D., Burger, R. P., Mkhatshwa, G., & Piketh, S. J. (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.

Department of Environmental Affairs (DEA). (2018). The National Framework for Air Quality Management in the Republic of South Africa. Available at: https://www.gov.za/sites/default/files/gcis_document/201805/41650gon518_0.pdf.

Department of Environmental Affairs (DEA). (2012). Manual for Air Quality Management Planning in South Africa. Available at: https://saaqis.environment.gov.za/documents/AQPlanning/MANUAL%20FOR%20AQMP%20-%20APRIL%202012%20-%20with%20cover.PDF.

Feig, G., Garland, R. M., Naidoo, S., Maluleke, A., & 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.

Hersey, S. P., 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*, 15, 4259–4278. https:// doi:10.5194/acp-15-4259-2015.

Morosele, I. P., & Langerman, K. E. (2020). The impacts of commissioning coal-fired power stations on air quality in South Africa: insights from ambient monitoring stations. *Clean Air Journal*, 30(2). https://doi.org/10.17159/caj/2020/30/2.8833.

Nkosi, N. C., Piketh, S. J., & Burger, R. P. (2018). Fine PM emission factors from residential burning of solid fuels using traditional cast-iron coal stoves. *Clean Air Journal*, 28(1). https://doi.org/10.17159/2410-972X/2018/v28n1a10.

Ross, K., Broccardo, S., Heue, K., Ferguson, K., & Piketh, S. (2007). Nitrogen oxides on the South African Highveld. *Clean Air Journal*, 16(2). https://doi.org/10.17159/caj/2007/16/2.7166.

Republic of South Africa. (1996). Constitution of the Republic of South Africa, 1996. Government Printer, Pretoria. Available at: https://www.justice.gov.za/legislation/constitution/saconstitution-web-eng.pdf.

Republic of South Africa. (2005). National Environmental Management: Air Quality Act, 2004. Government Printer, Pretoria. Available at: https://www.gov.za/sites/default/files/gcis_document/201409/a39-04.pdf.

Republic of South Africa. (2007). Notice of Intention to Declare the Highveld Priority Area in Terms of Section 18(1) of the National Environmental Management: Air Quality Act, 2004 (Act 39 of 2004). Government Printer, Pretoria. Available at: https://www.gov.za/sites/default/files/gcis_document/201409/30518.pdf.

Republic of South Africa. (2009a). Notice of Intention to Establish the Vaal Triangle Air-shed Priority Area in Terms of Section 19(1) of the National Environmental Management: Air Quality Act, 2004 (Act 39 of 2004). Available at: https://www.dffe.gov.za/

sites/default/files/gazetted_notices/nemaqa_airqualityact_g31615gon1241_0.pdf.

Republic of South Africa (2009b). National Ambient Air Quality Standards. Government Printer, Pretoria. Available at: https://www.dffe.gov.za/sites/default/files/legislations/nemaqa_airquality_g32816gon1210_0.pdf.

Republic of South Africa. (2012a). National Ambient Air Quality Standards. Government Printer, Pretoria. Available at: https://www.gov.za/sites/default/files/gcis_document/201409/35463gon486.pdf.

Republic of South Africa. (2012b). Declaration of the Waterberg National Priority Area. Available at: https://www.dffe.gov.za/sites/default/files/gazetted_notices/nemaqa_waterberg_declaration_g35435gen495_0.pdf.

Republic of South Africa. (2013). National Dust Control Regulations. Government Printer, Pretoria. Available at: https://www.gov.za/sites/default/files/gcis_document/201409/36974rg10045gon827.pdf.

Republic of South Africa. (2019). Listed Activities and Associated Minimum Emission Standards Identified in terms of Section 21 of the National Environmental Management: Air Quality Act, 2004 (Act 39 of 2004) as amended. Available at: https://cer.org.za/wp-content/uploads/2019/10/AQA-Listed-Activities-with-2019-amendment.pdf.

Steyn, M., & Kornelius, G. (2018). An economic assessment of SO2 reduction from industrial sources on the highveld of South Africa. *Clean Air Journal*, 28(1). https://doi.org/10.17159/2410-972X/2018/v28n1a9.

Walton, N. M., Piketh, S. J., van Zyl, P., Maenhaut, W., Burger, R., Language, B., & Formenti, P. (2021). Source apportionment of ambient fine and coarse aerosols in Embalenhle and Kinross, South Africa. *Clean Air Journal*, 31(2). https://doi.org/10.17159/caj/2021/31/2.11980.

Commentary

Are nature-based solutions a missing link in air quality management in South African cities?

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Foreword

Air quality in urban centres is notoriously poor: almost 50% of cities which are home to 100 000 residents or more, and more than 97% of cities in low- and middle-income countries of that size, do not meet the recently updated WHO Global Air Quality Guidelines (WHO, 2021; Jennings et al., 2021). Estimates are that 61 out of every 100,000 deaths in urban areas worldwide – totalling 1.8 million excess deaths – were due to particulate matter exposure in 2019 alone (Southerland et al., 2022). In addition to traditional air pollution reduction and mitigation methods, nature-based solutions (NBS) are increasingly being trialled to reduce air pollution exposure in cities globally. Here we discuss the potential value and necessary considerations of NBS in improving the air we breathe, and we consider how this could be applicable to a uniquely South African context. Are NBS a missing link in air quality management in South Africa?

Introduction

Because urban centres are characterised by large and dense human populations, tackling air pollution and reducing exposure to health-damaging air pollutants is a public health priority (Jennings et al., 2021). However, due to its many sources (e.g., vehicle emissions, industry and manufacturing, waste burning and outdoor cooking, to name a few), as well as complex socio-economic factors and the presence of confounding urban micro-climates, air pollution in cities remains difficult to manage, especially in developing countries (Menon et al., 2021).

In addition, cities are prone to the urban heat island effect, which presents another health threat for residents and can exacerbate the health burden placed on humans from exposure to air pollution (Menon et al., 2021).

With projections that 80% of the South African population will reside in urban areas by 2050 (Mlambo, 2018), and with ambient temperatures set to rise over the coming decades due to global climate change, it is critical that a combination of cost-effective and sustainable interventions are identified and implemented in South African cities to protect human health and biodiversity, and enhance the resilience of urban ecosystems (Liu et al., 2021).

Nature-based solutions for urban resilience

Solutions to protect, sustainably manage, and restore natural or modified ecosystems, which effectively and adaptively address societal challenges and simultaneously provide human well-being and biodiversity benefits are known as “nature-based solutions” (Cohan-Shacham et al., 2016). They include urban forests, green roofs/ walls, green spaces and parks, stormwater and detention ponds, rain gardens and bioswales, and even restored ecosystems (Liu et al., 2021; World Bank, 2021).

Nature-based solutions can be used to enhance urban resilience in the face of a wide range of urban challenges (Liu et al., 2021). When used as “green infrastructure” to complement traditional built infrastructure in urban areas, NBS have been found to contribute to improved air quality, reduced ambient air temperatures, reduced flooding, and enhanced carbon sequestration (Anderson and Gough, 2020). Over and above this, NBS have been shown to improve human mental and physical health and well-being (Ascenso et al., 2021), and when designed and implemented in a participatory manner, can favourably benefit women, youth and low-income communities (Pagano et al., 2019; Bechauf, 2021). Nature-based solutions can also be used as effective biomonitoring tools to determine the presence, quantities, temporal or spatial changes and effects of pollutants on the environment (Calfapietra, 2020; Fusaro et al., 2021; Shagjjav et al., 2022). For example, a study by Molnár et al., (2020) which assessed the usefulness of an Air Pollution Tolerance Index for environmental health, particularly considering air quality, showed that quantifying the amount of deposited dust on the surface of urban tree leaves can be an effective method for monitoring urban air quality. Similarly, some plant species can act as useful bioindicators, by developing leaf injuries or changes in vegetative periods if exposed to high concentrations of specific pollutants (Fusaro et al., 2021).

A nuanced approach to NBS for air quality management

If chosen strategically, NBS can be highly effective in taking up or removing air pollutants from the ambient air, by reducing ozone, particulate matter, nitrogen oxides and sulphur dioxide concentrations (Abhijit et al., 2017; Letter and Jaeger, 2019). Air quality improvements can occur through different ways, for example through enhanced deposition of particles on plant surfaces or the absorption of gases through plant stomata (Xing and Brimblecombe 2019). Despite their promising potential, careful planning should inform the design of NBS aimed at addressing air pollution, as shortcomings include oversimplification of design and underestimation of costs and maintenance requirements, the consequences of which could inadvertently lead to the deterioration of air quality (Schroeter at al., 2022).

A study from multiple cities in South Africa illustrated the ability of the lichen species *Parmelia caperata* to accumulate Mercury (Hg) from the ambient air, which suggests the potential to use this lichen to monitor atmospheric Hg deposition across the landscape (Panichev et al., 2019). Similarly, the lichen thallus of *Parmelia sulcate* has been used to assess the concentrations and possible sources of trace elements in the city of Tshwane, South Africa (Olowoyo et al., 2010). Vegetation barriers, e.g., shrubs or dense tree canopies can directly remove and reduce air pollution levels, but they can also present a physical barrier between humans and pollution sources (e.g., shrubs planted between roads and walk-ways in cities). This has been tested in a study in Khayelitsha, Cape Town, South Africa, where planting windbreak trees proved effective in reducing residents' exposure to ambient PM10 (Muchapondwa, 2010).

Despite evidence that promotes the use of NBS to improve air quality, research also shows that NBS are site- and context-specific, require a nuanced approach, and must be designed with specific benefits in mind (Cohan-Shacham et al., 2016; Jennings et al., 2021; Seddon et al., 2020). Care must be taken when designing NBS to ensure that unforeseen negative consequences on air quality or other important factors, such as biodiversity, do not arise. For example, trees could worsen pollution levels by preventing circulation of airflow or by producing air pollution themselves (e.g., pollen or biogenic volatile organic compounds) (Liu et al., 2022), and afforestation with non-native monocultures can negatively impact biodiversity and result in maladaptation (Seddon et al., 2020). Even though maintenance of NBS can be lower than that of traditional infrastructure, for interventions to be successful in the long-term, funding for adequate maintenance must be set aside during the design phase (Le Coent et al., 2021).

Other trade-offs need to be considered when using trees to improve air quality, including roots which can cause damage to infrastructure like roads or water pipes or the seasonal consideration of deciduous species that lose their foliage and pollution reduction potential in winter (Abhijit et al., 2017). Care must also be taken when choosing plant species for NBS, as the impacts of air pollutants can negatively impact the growth and survival of vegetation as well as the resilience of urban ecosystems.

Despite growing awareness of the potential of NBS, additional empirical evidence on their intended benefits, cost effectiveness, resilience to climate change and reliability is needed. More research and collaboration between atmospheric, natural and social scientists, NBS practitioners, and policy makers is required to ensure that NBS can be used as effective air pollution reduction interventions at a city level.

In developing contexts, where overlapping exposure to environmental health risks is a reality, it must be understood that urban greening and NBS alone cannot compensate for the systemic inequalities that lead to disproportionate burdens from environmental health risks like air pollution. Reducing this burden requires a combination of technical and socio-economic interventions (Jenings et al., 2021). This "Green Apartheid" (Venter et al., 2020) illustrates the clear need to understand the complex links between green infrastructure and human health and well-being, especially in a South African context.

Conclusions

Air pollution is one of the largest environmental health threats, causing millions of deaths annually (Landrigan et al., 2018; Jennings et al., 2021). The far-reaching benefits of well-designed NBS should be considered in cities in South Africa to improve air quality, enhance ecosystem resilience and holistically improve human health and well-being.

If weighed properly, the co-benefits and trade-offs of NBS could solve a wide range of environmental, social and economic challenges (Liu et al., 2021). If the scale of the intervention, the context and conditions of the site and the target air pollutant type are understood, the selection of plants that exhibit certain biophysical traits can enhance air pollution mitigation (Barwise and Kumar 2020).

Though NBS cannot replace the efforts which are underway to reduce or eliminate air pollution sources, they should not be discounted as cost-effective, complementary methods for holistic air quality management in South African cities, where emphasis must be placed on equitable access to green infrastructure, clean air and a healthy environment for all.

References

Abhijit, K.V. et al. (2017). Air pollution abatement performances of green infrastructure in open road and built-up street canyon environments – A review. Atmospheric Environment 162: 71-86.

Anderson, V. and Gough, W.A (2020). Evaluating the potential of nature-based solutions to reduce ozone, nitrogen dioxide, and carbon dioxide through a multi-type green infrastructure study in Ontario, Canada. City and Environment Interactions. 6:1000043. https://doi.org/10.1016/j.cacint.2020.100043.

Ascenso, A., Augusto, B., Silveira, C., Rafael, S., Coelho, S., & Monteiro, A. (2021). Impacts of nature-based solutions on the urban atmospheric environment: A case study for Eindhoven. The Netherlands. Urban Forestry & Urban Greening, 57, Article 126870. https://doi.org/10.1016/j.ufug.2020.126870.

Barwise, Y., Kumar, P. (2020). Designing vegetation barriers for urban air pollution abatement: a practical review for appropriate plant species selection. npj Clim Atmos Sci 3, 12. https://doi.org/10.1038/s41612-020-0115-3.

Bechauf, R. (2021). Nature-Based Infrastructure: A powerful tool for women's empowerment in climate adaptation. IISD Blog. Available at: https://www.iisd.org/articles/nature-based-infrastructure-powerful-tool-womens-empowerment-climate-adaptation [Accessed 22 February 2022].

Calfapetra, C. (2020). Nature-based solutions for microclimate regulation and air quality. Analysis of EU-funded projects. European Commission. Available at: https://op.europa.eu/en/publication-detail/-/publication/001a9517-d530-11ea-adf7-01aa75ed71a1/language-en [Accessed on 22 February 2022].

Cohen-Shacham E, Walters G, Janzen C, Maginnis S, editors. (2016). Nature-based solutions to address global societal challenges. Gland, Switzerland: IUCN. ISBN 978-2-8317-1812-5. https://doi.org/10.2305/IUCN.CH.2016.13.en [xiii + 97pp].

Fusaro, L., Salvatori, E., Winkler, A., Frezzini, M. A., De Santis, E., Sagnotti, L., Canepari, S. & Manes, F. (2021). Urban trees for biomonitoring atmospheric particulate matter: An integrated approach combining plant functional traits, magnetic and chemical properties. Ecological Indicators, 126, 107707. https://doi.org/10.1016/j.ecolind.2021.107707.

Jennings, V., Reid, C.E. & Fuller, C.H. (2021). Green infrastructure can limit but not solve air pollution injustice. Nat Commun 12, 4681. https://doi.org/10.1038/s41467-021-24892-1.

Letter, C., Jäger, G. Simulating the potential of trees to reduce particulate matter pollution in urban areas throughout the year. Environ Dev Sustain 22, 4311–4321 (2020). https://doi.org/10.1007/s10668-019-00385-6.

Landrigan, P. J., Fuller, R., Acosta, N. J. R., Adeyi, O., Arnold, R., Basu, N., Baldé, A. B., Bertollini, R., Bose-O'reilly, S., Boufford, J. I., Breysse, P. N., Chiles, T., Mahidol, C., Coll-Seck, A. M., Cropper, M. L., Fobil, J., Fuster, V., Greenstone, M., Haines, A., Hanrahan, D., Hunter, D., Khare, M., Krupnick, A., Lanphear, B., Lohani, B., Martin, K., Mathiasen, K. V., Mcteer, M. A., Murray, C. J. L., Ndahimananjara, J. D., Perera, F., Potočnik, J., Preker, A. S., Ramesh, J., Rockström, J., Salinas, C., Samson, L. D., Sandilya, K., Sly, P. D., Smith, K. R., Steiner, A., Stewart, R. B., Suk, W. A., Van Schayck, O. C. P., Yadama, G. N., Yumkella, K. & Zhong, M. (2018). The Lancet Commission on pollution and health. The Lancet, 391, 462-512. https://doi.org/10.1016/S0140-6736(17)32345-0.

Le Coent, P., Graveline, N., Altamirano, M. A., Arfaoui, N., Benitez-Avila, C., Biffin, T., Calatrava, J., Dartee, K., Douai, A., Gnonlonfin, A., Hérivaux, C., Marchal, R., Moncoulon, D. & Piton, G. (2021). Is-it worth investing in NBS aiming at reducing water risks? Insights from the economic assessment of three European case studies. Nature-Based Solutions (1).

Liu, H.-Y.; Jay, M.; Chen, X. (2021). The Role of Nature-Based Solutions for Improving Environmental Quality, Health and Well-Being. Sustainability 2021, 13, 10950. https://doi.org/10.3390/su131910950.

Liu, l., Seyler, B. C., Liu, H., Zhou, l., Chen, D., Liu, S., Yan, C., Yang, F., Song, D., Tan, Q., Jia, F., Feng, C., Wang, Q. & Li, Y. (2022). Biogenic volatile organic compound emission patterns and secondary pollutant formation potentials of dominant greening trees in Chengdu, southwest China. Journal of Environmental Sciences. Article in press https://doi.org/10.1016/j.jes.2021.08.033.

Menon JS and Sharma R (2021). Nature-Based Solutions for Co-mitigation of Air Pollution and Urban Heat in Indian Cities. Front. Sustain. Cities 3:705185. https://doi.org/10.3389/frsc.2021.705185.

Mlambo, V. (2018). An overview of rural-urban migration in South Africa: its causes and implications. Archives of Business Research. Vol 6(4). https://doi.org/10.14738/abr.64.4407.

Molnár, Vanda É., Dávid Tőzsér, Szilárd Szabó, Béla Tóthmérész, and Edina Simon. 2020. "Use of Leaves as Bioindicator to Assess Air Pollution Based on Composite Proxy Measure (APTI), Dust Amount and Elemental Concentration of Metals" Plants 9, no. 12: 1743. https://doi.org/10.3390/plants9121743

Muchaponwa, E. (2010). A cost-effectiveness analysis of options for reducing pollution in Khayelitsha township, South Africa. The Journal for Transdisciplinary Research in Southern Africa. Vol6(2). https://hdl.handle.net/10520/EJC111914.

Olowoyo, J.O., van Heerden, E. And Fischer,J.L. (2010). Trace element concentrations from lichen transplants in Pretoria, South Africa. Environ Scie Pollut Res. 18:663-668. https://doi.org/10.1007/s11356-010-0410-3.

Pagano, A., Pluchinotta, I., Pengal, P., Cokan, B., & Giordano, R. (2019). Engaging stakeholders in the assessment of NBS effectiveness in flood risk reduction: A participatory System Dynamics Model for benefits and co-benefits evaluation. Science of The Total Environment. 690: 543-555. https://doi.org/10.1016/j.scitotenv.2019.07.059.

Panichev, N., Mokgalaka, N. & Panicheva, S. (2019). Assessment of air pollution by mercury in South African provinces using lichens Parmelia caperata as bioindicators. Environ Geochem Health 41, 2239–2250. https://doi.org/10.1007/s10653-019-00283-w.

Schröter, B., Hack, J., Hüesker, F. et al. Beyond Demonstrators—tackling fundamental problems in amplifying nature-based solutions for the post-COVID-19 world. npj Urban Sustain 2, 4 (2022). https://doi.org/10.1038/s42949-022-00047-z.

Shagjjav, O., Bayarmaa, J., & Otgonbayar, K. (2022). Air pollution tolerance indices of selected plants around Ulaanbaatar city, Mongolia. Mongolian Journal of Biological Sciences, 20(1), 41–48. https://doi.org/10.22353/mjbs.2022.20.04.

Seddon., N., Chausson, A., Berry, P., Girardin, C, A. J., Smith, A., & Turner, B. (2020). Understanding the value and limits of nature-based solutions to climate change and other global challenges. Philosophical Transactions of the Royal Society B (Biological Sciences). 375: 1794. https://doi.org/10.1098/rstb.2019.0120.

Southerland, V. A., Brauer, M., Mohegh, A., Hammer, M. S., van Donkelaar, A., Martin, R. V., Apte, J. S., Anenberg, S. C. (2022). Global urban temporal trends in fine particulate matter (PM2·5) and attributable health burdens: estimates from global datasets. The Lancet Planetary Health, Volume 6, Issue 2, ISSN 2542-5196, https://doi.org/10.1016/S2542-5196(21)00350-8.

Venter, Z.S., Shackleton, C.M., Van Staden, F., Selomane, O. And Masterson, V.A. (2020). Green Apartheid: Urban green infrastructure remains unequally distributed across income and race geographies in South Africa. Landscape and Urban Planning. 203:103889. https://doi.org/10.1016/j.landurbplan.2020.103889.

World Health Organization. (2021). WHO global air quality guidelines: particulate matter (PM2.5 and PM10), ozone, nitrogen dioxide, sulfur dioxide and carbon monoxide. World Health Organization. https://apps.who.int/iris/handle/10665/345329. License: CC BY-NC-SA 3.0 IGO.

World Bank. 2021. A Catalogue of Nature-Based Solutions for Urban Resilience. World Bank, Washington, DC. © World Bank. Available at: https://openknowledge.worldbank.org/handle/10986/36507 License: CC BY 3.0 IGO. [Accessed 22 February 2022].

Xing, Y. and Brimblecombe, P. (2019). Role of Vegetation in deposition and dispersion of air pollution in urban parks. Atmospheric Environment. 210:73-83. https://doi.org/10.1016/j.atmosenv.2018.12.027.

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Commentary

The Minimum Emission Standards (MES) and the sabotage of public health

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Introduction

Air Emission standards for pollutants were introduced into policy through the Consultative National Environmental Policy Process in 1996. The National Environmental Management Act of 1998 (NEMA) followed in short order so as to give effect to the environmental right in section 24 of the Constitution. However, it took another seven years for the subsidiary legislation, the National Environmental Management: Air Quality Act of 2004 (AQA) to specifically mandate the development of minimum emission standards (MES). And it took another five years of stakeholder consultations and negotiations before the MES were promulgated in 2010.

Like emission reduction regimes elsewhere in the world, the purpose of emission standards is to protect human health. They also provide the means of holding polluting industries to account, which is why the major industries first resisted their introduction and have since lobbied to weaken them. Nevertheless, big industry and Eskom in particular were well aware of the health impacts of pollution. The very weak Air Pollution Prevention Act (APPA) was introduced in 1965 and was first administered by the Department of Health. In the 1970s, when Eskom was planning a new round of power station construction, the Chief Air Pollution Officer cautioned against putting them all on the Highveld (Ballim 2017). In 2006, when the AQA finally replaced the moribund APPA regime, Eskom itself commissioned studies which confirmed the direct health risks of its emissions (Scorgie and Thomas 2006a; Scorgie and Thomas 2006b). The legal implication was that, as an organ of state with constitutional obligations, Eskom was bound to act to limit its pollution well before the MES were published in 2010.

This commentary looks first at the health impacts of Eskom's coal-fired fleet and hence the 'co-benefits' of addressing climate change, and second at the legal context and contests.

The MES, air pollution and climate change: two sides of the same coin

Air pollution and climate change are inter-linked. Air pollution is a "silent public health emergency" causing 7 million premature deaths each year, and accounting for about a quarter of all heart attack deaths, and about a third of all deaths from stroke, lung cancer, and chronic obstructive pulmonary disease. Health impacts are largest among women, children, older people, and the poor (Perera 2017; WHO 2021a).

Climate change is the other side of the coin of environmental impacts on global public health. The rapidly changing climate has far-reaching and catastrophic health impacts, with the largest burden falling on the poor, who have contributed least to greenhouse gas emissions (GHGs). The 2022 floods in KwaZulu-Natal provide a brutal illustration of the point.

Therefore, urgent global action over the next decade to cut air pollution and GHGs can protect health in the short and longer terms. For example, minimising industrial and energy sector emissions can reduce the health burden of ambient air pollution, while clean and affordable household heating and cooking technologies can minimise household air pollution; and these actions have the additional co-benefits of mitigating further climate change.

Since the WHO Air Quality Guidelines of 2005, many large global population-based studies have supported the Guideline's conclusions of a significant relationship between air pollution and adverse health outcomes. Research into global mortality associated with long-term exposure to ambient $PM_{2.5}$ particulate matter in 2018 revealed it to be a more important health risk factor than previously thought (Burnett et al., 2018). Additional health outcomes associated with air pollution, and with $PM_{2.5}$ in integrated studies (EPA 2009), are cardiovascular (Malig and Ostro 2009; Atkinson et al., 2010; Chen et al., 2011; Mallone at al., 2011), respiratory (Chen et al., 2011), and total mortality (Tobias et al., 2011; Meister at al., 2012). For particulate matter there is no evidence of a safe level of exposure without adverse health effects.

Since 2015 in South Africa, various industry applications for suspension, alternative limits and/or postponement of compliance with the minimum emission standards are notable, as they are located in priority air-sheds that are generally non-compliant with national ambient air quality standards. This legal regime is outlined further below. Various modelled studies have shown severe health impacts from granting MES postponements.

A 2014 health assessment was undertaken by Lauri Myllyvirta and the Greenpeace Global Air Pollution Unit in response to Eskom's "Health impact focused cost benefit analyses" (Myllyvirta 2014). It projected that with Medupi and Kusile in full operation, emissions from Eskom's coal-fired power plants (CFPs) would be responsible for 2,400 premature deaths per year, and that excess emissions if Eskom's various applications were fully granted would result in approximately 23,000 premature deaths. Yet requiring full compliance with the MES would result in a 40% reduction in the cumulative health impact of air pollution from Eskom's CFPs.

Using data from Myllyvirta's study, air quality and health expert Dr Mike Holland assessed the health impacts and associated economic costs of emissions from Eskom's CFPs in 2016 (Holland 2017). His assessment, which focused on the role of $PM_{2.5}$ in the atmosphere following release of pollutants, such as SO_2 and NOx, estimated that the following impacts are attributable to Eskom's emissions:

Table 1: *Annual health impacts linked to coal fired generation in South Africa (Myllyvirta 2014)*

	Cases, etc	Value, $int, millions
Equivalent attributable deaths		
Lung cancer	*157*	
Ischaemic heart disease	*1,110*	
Chronic obstructive pulmonary disease	*73*	
Stroke	*719*	
Lower respiratory infection	*180*	
Total equivalent attributable deaths	**2,239**	**2,121.94**
Chronic Bronchitis (adults, cases)	2,781	64.64
Bronchitis in children aged 6 to 12	9,533	2.19
Equivalent hospital admissions	2,379	2.79
Restricted Activity Days (all ages)	3,972,902	132.72
Asthma symptom days (children 5-19yr)	94,680	1.44
Lost working days	996,628	47.05
Total costs		**2,372.78**

Finally, modelled scenarios of the health co-benefits of implementing national climate commitments consistent with the 2015 Paris Agreement temperature targets by nine representative countries, including South Africa, found that, compared with business as usual, sustainable pathways resulted in an annual reduction of 1.18 million air pollution-related deaths by 2040 (Hamilton et al., 2021).

Minimum emission standards (MES) – a legitimate government purpose to protect public health, social conditions, and the environment in air-shed priority areas

The object of the AQA, read with NEMA, is to provide measures to prevent air pollution and enhance air quality and so give effect to several constitutional rights. In its preamble, the AQA recognises that: "*the quality of ambient air in many areas … is not conducive to a healthy environment …*"; "*the burden of health impacts associated with polluted ambient air falls most heavily on the poor*"; "*air pollution carries a high social, economic and environmental cost that is seldom borne by the polluter*"; and "*minimisation of pollution through vigorous control, cleaner technologies and cleaner production practices is key to ensuring that air quality is improved*".

Three of the key regulatory instruments mandated by the AQA are the national ambient air quality standards (NAAQS), the declaration of priority air-shed areas, and the MES.

National ambient air quality standards

NAAQS have been set for eight pollutants, including nitrogen dioxide (NO_2), sulphur dioxide (SO_2), PM_{10} and $PM_{2.5}$ (DEA 2009; DEA 2012). The NAAQS are intended to be health-based, and "*broadly accepted as a proxy for air that it not harmful to health and well-being*" (DEA 2017). Nevertheless, South Africa's NAAQS are much weaker than those set out in the WHO's 2005 Air Quality Guidelines, and very much weaker than the revised WHO Guidelines published in September 2021 (WHO 2021b).

Declaration and management of air-shed priority areas

The environment minister may declare a priority area where ambient air quality standards are exceeded. The objective is to reduce air pollution, comply with NAAQS and so protect public health. South Africa has declared three priority areas: the Vaal Triangle Airshed Priority Area ("VTAPA") was declared in 2006, the Highveld Priority Area (HPA) in 2007, and the Waterberg-Bojanala National Priority Area ("WBPA") in 2012. The AQA requires that an air quality management plan is developed and implemented for each priority area and provides for regulations to enforce the plans.

List of point-source emissions activities

The minister must also publish a 'list of activities' which result in atmospheric emissions that are harmful to the environment and to people's health and which prescribe MES for each. The first list of activities was published in 2010 and allowed five years for existing plants to comply with very lenient standards by 2015, and a further five years to comply with stricter standards by April 2020. It also allowed for compliance with the MES to be postponed – for a maximum of five years – if certain criteria were satisfied, notably that the ambient air quality in the area is in compliance with the NAAQS.

Eskom's compliance with the MES – an obligation deferred

The 2010 MES were published following lengthy consultations, engaging all affected stakeholders, to set the MES. This ended with standards that are notably weaker than those in other developing countries, including India and China.

In late 2013, just ahead of the compliance deadline, Eskom applied for exemption from compliance with the 2015 MES and, when it was pointed out that this was not legally possible, for wide-ranging postponements. Sasol and other big polluters followed suit. The majority of these applications were granted despite the explicit legal criteria that the ambient air quality in the area of the operation must be in compliance with the NAAQS. The HPA, where 12 of Eskom's coal-fired power stations and Sasol's coal-to-liquid plant are situated, was not and is not in compliance.

The MES were subsequently amended in 2018. The revisions included: confirmation that no further compliance postponements of the 2015 MES are permitted; an application for a once-off postponement of compliance with the 2020 MES is permitted to 31 March 2025; facilities to be decommissioned by 31 March 2030 may apply for a once-off suspension of compliance with the 2020 MES. The amendments also introduced the application for an alternative emission limit or emission load subject to explicit criteria, including the overriding precondition that there is compliance with NAAQS in the area in which the emitting facility is based.

Nevertheless, between 2018 and 2020, Eskom applied for a combination of 5-year postponements of compliance, suspensions of compliance, and alternative (weaker) limits in relation to the MES compliance timeframes to cover 14 of its 15 coal-fired power stations. It also submitted a formal application for exemption from compliance with the MES which was dismissed.

On 30 October 2021, the National Air Quality Officer (NAQO) issued decisions on Eskom's pending applications. In short, suspension of compliance was granted to 6 stations, along with a 5-year postponement of compliance for particular pollutants for 3 stations. Eskom's applications for alternative limits were all declined, in part because NAAQS are not in compliance. The NAQO also noted that "*Eskom has made minimal effort to fully comply with the standards", and "[t]he NAQO does not have the prerogative to issue decisions that are outside the current legal provisions or are in non-compliance with the law*".

Unsurprisingly, these decisions have been appealed by Eskom, other industrial emitters, and a range of nongovernmental organisations. What was unexpected, however, was the minister's unprecedented response, proposing a public consultation process that will hear inputs from all interested & affected parties on air quality and compliance with the MES. The department's media statement explains that "*[d]ue to the complex and conflicting nature of the issues raised in the appeals received, the Minister is of the view that a consultative process will assist in ensuring that all issues arising from the appeals can be addressed in a meaningful and resolute manner", and, ". . . the consultative process would not in any way condone non-compliance with the Minimum Emissions Standards and will not impact on any present or future criminal action against non-compliance. The current appeal process will be held in abeyance pending the outcome of the consultative process*" (DFFE 2022).

The integrity of the MES regime at a crossroads

The minister, as the competent authority, has clearly arrived at an impasse that has been a decade and more in the making. On the one hand, the rule of law must be upheld and polluters, especially Eskom as the largest polluter in the country, must be compelled to comply with the MES in the interest of public health. On the other, Eskom says that 16000 MW of nominal capacity must be decommissioned if the NAQO decisions are enforced and that it cannot afford the necessary abatement technology, while the earlier neglect of maintenance coupled with misguided government action, and inaction, has left its aging fleet in tatters (BizNews 2021). The dismal management of the minerals and energy portfolio is glaringly apparent with 'emergency' procurement of new capacity stalled and all other procurement running late while it rides shotgun for fossil fuels. It will be further exposed as the scope of this public consultation process will inevitably extend beyond air quality to climate change on the other side of the coin.

The process, however, seems beset by uncertainty. In March, the minister attempted to hand responsibility for it to the Presidential Climate Commission. The commission declined, leaving it with the minister who has now initiated a process to set up an expert panel to advise her on the appeals to the NAQO's decisions. Meanwhile, the implementation of those decisions is suspended and a resolution of the matter hangs in the polluted air.

After a century of unconstrained environmental vandalism and two decades obstructing accountability, Eskom is effectively looking for exemption from MES, so to restore a right to impunity. The Terms of Reference for appointing the panel, however, says that non-compliance with the MES will not be condoned. The panel must consult widely and "provide the minister with practical options" taking account of the "constitutional right of the people to an environment that is not harmful to their health and well-being, the energy crisis and the local economic climate". They are thus asked to find a way for the minister to square the circle.

They will have six months to do so. The minister must then act with urgency. But she is right that the decision needs wider support – starting with her cabinet colleagues. It cannot finally be separated from the crisis at Eskom along with the multiple contradictions in government's management of electricity and the provision of services to all people. For a government with an aversion to responsibility, this may be a tough call. But it cannot be deferred forever except at the cost of the rule of law and of the people, the environment and, finally, the earth.

References

Atkinson, R.W., Fuller, G.W., Anderson, H.R., Harrison, R.M. and Armstrong, B. (2010). Urban ambient particle metrics and health: a time-series analysis. *Epidemiology*, 21(4):501–511. https://doi.org/10.1097/EDE.0b013e3181debc88.

Ballim, F. (2017). The Evolution of Large Technical Systems in the Waterberg Coalfield of South Africa: From Apartheid to Democracy. PhD Thesis. University of the Witwatersrand. Available at: https://wiredspace.wits.ac.za/bitstream/handle/10539/23949/Ballim_final_PhD.pdf?sequence=2&isAllowed=y [Accessed 23 May 2022].

BizNews. (2021) Resetting SA's troubled SOE, Eskom – André de Ruyter sets out how. Available at: https://www.biznews.com/sarenewal/2021/09/27/eskom-resetting [Accessed 23 May 2022].

Burnett, R., Chen, H., Szyszkowicz, M. and Spadaro, J. (2018). Global estimates of mortality associated with long-term exposure to outdoor fine particulate matter. PNAS. 115(38):9592-9597. https://doi.org/10.1073/pnas.1803222115.

Chen, R., Li, Y., Ma, Y., Pan, G., Zeng., G., Xu, X., Chen, B. and Kan, H. (2011). Coarse particles and mortality in three Chinese cities: the China Air Pollution and Health Effects Study (CAPES). *Science of the Total Environment*, 409(23):4934–4938. http://doi.org/10.1016/j.scitotenv.2011.08.058.

Department of Environmental Affairs. (2009). National Ambient Air Quality Standards 2009. Government Notice 1210 in GG 32816 of 24 December 2009. Available at: https://www.dffe.gov.za/sites/default/files/legislations/nemaqa_airquality_g32816gon1210_0.pdf [Accessed 23 May 2022].

Department of Environmental Affairs. (2012). National Ambient Air Quality Standards 2012. Government Notice 486 in GG 35463 of 29 June 2012. Available at: https://www.gov.za/sites/default/files/gcis_document/201409/35463gon486.pdf [Accessed 23 May 2022].

Department of Environmental Affairs. (2017). The National Framework for Air Quality Management in the Republic of South Africa 2017. Section 5.2.3.4. Available at: https://saaqis.environment.gov.za/Pagesfiles/2017_National_Framework.pdf [Accessed 23 May 2022].

Department of Forestry, Fishery, and Environment. (2022). Public consultation proposed on issues related to compliance with minimum emission standards. Available at: https://www.dffe.gov.za/mediarelease/compliance_minimumemissionstandards [Accessed 23 May 2022].

United States Environmental Protection Agency (EPA). (2009). Integrated science assessment for particulate matter (final report). Washington, DC, United States Environmental Protection Agency. Available at: (http://cfpub.epa.gov/ncea/cfm/recordisplay.cfm?deid=216546#Download [Accessed 05 March 2022].

Hamilton, I., Kennard, H., McGushin, A., Hoeglund-Isaksson, L., Kiesewetter, G., Lott, M., Milner, J., Purohit, P., Rafaj, P., Sharma, R., Springmann, M., Woodcock, J. and Watts, N. (2021). The public health implications of the Paris Agreement: a modelling study. *The Lancet Planetary Health*. 5(2): E74-E83. https://doi.org/10.1016/S2542-5196(20)30249-7.

Holland, M. (2017). Health impacts of coal fired power plants in South Africa. Available at: https://cer.org.za/wp-content/uploads/2017/04/Annexure-Health-impacts-of-coal-fired-generation-in-South-Africa-310317.pdf [Accessed 23 May 2022].

Malig, B.J. and Ostro, B.D. (2009). Coarse particles and mortality: evidence from a multi-city study in California. *Occupational and Environmental Medicine*. 66(12):832–839. http://dx.doi.org/10.1136/oem.2008.045393.

Mallone, S., Stafoggia, M., Faustini, A., Gobbi, G.P., Marconi, A. and Forastiere, F. (2011). Saharan dust and associations between particulate matter and daily mortality in Rome, Italy. *Environmental Health Perspectives*. 119(10):1409–1414. http://doi.org/10.1289/ehp.1003026.

Meister, K., Johansson, C. and Forsberg, B. (2012). Estimated short-term effects of coarse particles on daily mortality in Stockholm, Sweden. *Environmental Health Perspectives*. 120(3):431–436. http://doi.org/10.1289/ehp.1103995.

Myllyvirta, L. (2014). Health impacts and social costs of Eskom's proposed non-compliance with South Africa's air emission standards. Greenpeace International. Available at: https://cer.org.za/wp-content/uploads/2014/02/Annexure-5_Health-impacts-of-Eskom-applications-2014-_final.pdf [Accessed 23 May 2022].

Perera, F.P. (2017). Multiple Threats to Child Health from Fossil Fuel Combustion: Impacts of Air Pollution and Climate Change. *Environmental Health Perspectives*. 125(2):141-148. http://doi.org/10.1289/EHP299.

Scorgie, Y. and Thomas, R. (2006a). Air Pollution Health Risk Analysis of Operations of Current and Proposed Eskom Power Stations Located in the Limpopo Province. Airshed Planning Professionals (Pty) Ltd. Available at: https://cer.org.za/wp-content/uploads/2014/07/Matimba-Health-Risk-Assessment_1.pdf [Accessed 23 May 2022].

Scorgie, Y. and Thomas, R. (2006b). Eskom Mpumalanga Highveld Cumulative Scenario Planning Study: Air Pollution Compliance Assessment and Health Risk Analysis of Cumulative Operations of Current, RTS and Proposed Eskom Power Station Located within the Mpumalanga and Gauteng Provinces. Airshed Planning Professionals (Pty) Ltd. Available at: https://lifeaftercoal.org.za/virtual-library/key-information/eskom-health-studies [Accessed 23 May 2022].

Tobías, A., Perez, L., Diaz, J., Linares, C., Pey, J., Alastruey, A. and Querol, X. (2011). Short-term effects of particulate matter on total mortality during Saharan dust outbreaks: a case-crossover analysis in Madrid (Spain). *Science of the Total Environment*. 412-413:386–389. https://doi.org/10.1016/j.scitotenv.2011.10.027.

World Health Organisation (WHO). (2021a). Ambient (outdoor) air pollution. Fact Sheet. Available at: http://www.who.int/news-room/fact-sheets/detail/ambient-(outdoor)-air-quality-and-health [Accessed on 23 May 2022].

World Health Organisation (WHO). (2021b). New WHO Global Air Quality Guidelines aim to save millions of lives from air pollution. Available at: https://www.who.int/news/item/22-09-2021-new-who-global-air-quality-guidelines-aim-to-save-millions-of-lives-from-air-pollution [Accessed on 23 May 2022].

Research brief

Household air pollution and respiratory symptoms a month before and during the stringent COVID-19 lockdown levels 5 and 4 in South Africa

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In March 2020, the South African government declared a National State of Emergency as the severe acute respiratory syndrome coronavirus 2 (SARS-CoV-2) also known as coronavirus disease (COVID-19) pandemic threatened the lives of South Africans. A National Lockdown comprising five Levels was developed and implemented starting with Level 5 for 'high COVID-19 spread with low health system readiness'. Level 5 meant employees working in non-essential services and schoolchildren stayed in their dwellings with limited movement (essential supplies only).

With people spending majority of their time in their dwellings, their exposure to household air pollution (HAP) became reason for concern; especially among people who rely on so-called 'dirty fuels' as their main source of energy for cooking and / or heating. A recently published study by Wright et al. (2022) conducted a retrospective online / telephonic survey to investigate fuel use behaviours / patterns of use affecting HAP exposure and associated HAP-related respiratory health outcomes a month before and during Lockdown Levels 5 and 4, i.e., the two most stringent levels. Participants were drawn from an existing market research company panel (since field campaigns were not possible) from Gauteng, Western Cape, KwaZulu-Natal and Eastern Cape where COVID-19 cases were highest at the time of study planning.

Among 2 505 participants (72% Black African, 12% Coloured, 4% Indian/Asian and 12% White) electricity was the main energy source for heating and cooking before and during Lockdown Levels 5 and 4. Some households used less electricity and a few switched to 'dirty fuels' during Lockdown Levels 5 and 4. Unfortunately, due to the reliance on online survey questionnaires, majority of participants were from middle-to-high income groups. Fewer participants (n=250) from lower socio-economic groups were contacted by telephone to complete the questionnaire due to cost.

The prevalence of HAP-related respiratory health outcomes like wheeze, wet cough, hay fever, and shortness of breath was similar and relatively low (< 10%) before and during Lockdown Levels 5 and 4, except for dry cough (16% before; 12% after). Recall bias may have influenced these results. Most participants reported that they were cleaning more, cooking more and spending more time indoors during Lockdown Levels 5 and 4. Our most concerning finding was that one-third of participants reported presence of environmental tobacco smoke (ETS, including smoke/vape) in the dwelling (Figure 1). ETS is a form of HAP and is associated with adverse health effects, especially among children under 5 years of age. It can contribute to middle

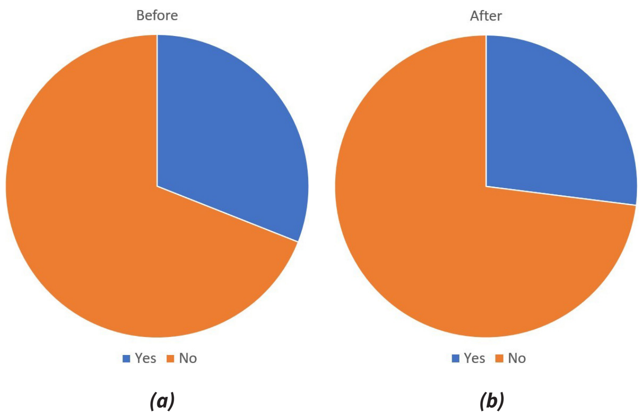

Figure 1: *Prescence of ETS (smoke/vape) in dwelling a) a month before and b) during Lockdown Levels 5 and 4. There was a 4% decline in presence of ETS in the home during Lockdown Levels 5 and 4 compared to one month before Lockdown began.*

ear disease, asthma, bronchiolitis and impaired pulmonary function among others (Hwang et al., 2012).

These are important findings for public health should South Africa return to Lockdown Levels that restrict movement and keep people and children at home indoors the majority of the time. Recommendations are needed to raise awareness about HAP, especially ETS, including how to avoid or reduce HAP to prevent associated human health impacts.

References

Hwang SH, Hwang JH, Moon JS, Lee DH. Environmental tobacco smoke and children's health. *Korean J Pediatr*. 2012;55(2):35-41. https://doi.org/10.3345/kjp.2012.55.2.35

Wright CY, Kapwata T, Abdelatif N, Batini C, Wernecke B, Kunene Z, Millar DA, Mathee A, Street R, Panchal R, Hansell A, Cordell R, Vande Hey J. Household Air Pollution and Respiratory Symptoms a Month Before and During the Stringent COVID-19 Lockdown Levels 5 and 4 in South Africa. Annals of Global Health. 2022; 88(1): 3, 1–12. https://doi.org/10.5334/aogh.3465

Research brief
Source apportionment of fine atmospheric particles using positive matrix factorization in Pretoria, South Africa

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Outdoor and indoor air pollution have been regarded as a serious issue in South Africa, with the emissions of various air pollutants and their resulting concentrations in the atmosphere being a major source of concern. For example, nearly 80% of the global population was subjected to air pollution levels that surpassed the World Health Organization (WHO) air quality guidelines in 2011. Pollution from a variety of sources has had a significant effect on air quality, posing a direct threat to the critical roles the environment plays in preserving and sustaining life by absorbing harmful ultraviolet radiation, warming the surface, and controlling the earth's temperature. Particulate matter (PM) suspended in the air for hours or days can travel a long distance, making it a long-range transported pollutant that is influenced by particle size, chemical composition, and other physical and biological characteristics. $PM_{2.5}$ (particles smaller than 2.5 µm) has received a lot of attention recently because of the negative impact it has on human health, i.e., its potential to penetrate human lungs. Furthermore, epidemiological studies have revealed a connection between PM and a variety of health problems. Source apportionment is an important air quality management tool for providing information about source contributions required for pollution abatement strategies. However, not many studies have applied air mass backward trajectory modelling with source apportionment model analysis to investigate the sources of PM.

Daily 24-hour $PM_{2.5}$ samples were collected every third day on the roof top of HW (6th floor) Snyman South Building at the School of Health Systems and Public Health (SHSPH), Prinshof Campus, University of Pretoria, from April 2017 to April 2018. At the Air Quality Laboratory of SHSPH, gravimetric analyses of $PM_{2.5}$ filters were carried out using a 1µg sensitivity microbalance (Mettler Toledo, XP6) under climate-controlled conditions (temperature and relative humidity were maintained at 21± 0.5 °C and 50 ± 5%, respectively) before and after sampling. Black carbon (BC), UVPM (a proxy for organic carbonaceous particulate matter absorbing UV light at 370 nm), the elemental composition of aerosol particles on all filters were determined using an XEPOS 5 energy-dispersive X-ray fluorescence (EDXRF) spectrometer (Spectro Analytical Instruments GmbH, Germany) at the Department of Chemistry and Molecular Biology, Atmospheric Science Division, University of Gothenburg. The Environmental Protection Agency's program EPAPMF5.0 was used to conduct the source apportionment study. The geographical origin of air masses passing through Pretoria, South Africa, was used as a proxy for long-range transport of air pollutants from distant sources and their composition. The Hybrid Single Particle Lagrangian Integrated Trajectory (HYSPLIT) program was used to generate backward trajectories for the 1-year sampling campaign.

A total of 147 $PM_{2.5}$ filter samples were obtained (with 25 duplicate samples), over a 122-day period for the 1-year sampling campaign. The average daily $PM_{2.5}$ concentration was 21.1 µg/m³. The daily mean concentration in this study was higher than the South African National Ambient Air Quality Standards (20 µg/m³) and the WHO yearly air quality guideline (10 µg/m³). The WHO daily air quality guideline for $PM_{2.5}$ (25 µg/m³) was found to be exceeded on 27% (33 days) of the sampling campaign days, with the most exceedance occurring during the winter season. The $PM_{2.5}$ mean concentrations for weekdays (88days; 20.7 µg/m³) and weekends (34days; 22.1 µg/m³) did not differ significantly from each other ($p > 0.05$). This may be attributed to other sources contributing to the pollutant levels apart from traffic, as it is expected that less traffic will be less over the weekend compared to weekdays.

The mean $PM_{2.5}$ level for this study is significantly higher than the $PM_{2.5}$ levels from the other two cities of this bigger project, Cape Town (13.3 µg/m³) and Thohoyandou (10.9 µg/m³). In comparison with other African countries, the mean $PM_{2.5}$ level were similar to the studies in Nairobi (21 µg/m³) and Accra (22.7 µg/m³) but lower to what were reported in Quagadougou (86 µg/m³) and Cairo (80 µg/m³) and Egypt (70 µg/m³.) The annual mean concentration for trace elements showed S had the highest average concentration of all the elements determined, followed by Si, Fe, K, and Ca, in that sequence. The weekly variation showed that the highest concentrations of the elements were recorded on Thursdays and lowest on Mondays for K, Pb, and Cl; Tuesday for S; and weekends for Fe, Ca. The concentrations of Pb and Cl display variation during weekdays, indicating variability in their sources, while the concentration of K, Ca, Fe, and S do not display significant variation within the weekdays, thus indicating some consistency for their possible sources. Most of the species concentration followed the seasonal pattern (winter > autumn > spring > summer) observed for $PM_{2.5}$.

The total daily $PM_{2.5}$ elemental composition dataset from the sampling campaign were used in the PMF model. Five to seven factors were investigated, but the seven factors output was presented. The mean $PM_{2.5}$ concentrations of the seven sources (i.e., are local and anthropogenic in origin) were named; fossil fuel combustion, soil dust, secondary sulphur, vehicle exhaust, road traffic, base metal/pyrometallurgical, and coal burning. Fossil fuel was found to be the major contributor with 22% of elemental mass in $PM_{2.5}$ concentrations on a yearly average. Coal combustion came in second, accounting for 18% of the total $PM_{2.5}$ mass. The combined contribution of soil dust and road traffic amounts to 4.0 $\mu g/m^3$ (22% of the total $PM_{2.5}$). Seasonal behaviour has been discovered in the known sources. When compared to other seasons, the contribution of secondary sulphur, vehicle exhaust, base metal/pyrometallurgical, and coal burning to $PM_{2.5}$ concentration levels was found to be significantly higher during winter. In comparison to other seasons, levels of fossil fuel, soil dust, and road traffic were higher in autumn ($p < 0.001$ for all tests).

Apart from local sources, air pollutants transport and regional sources have a significant impact on Pretoria's air quality. Five transport cluster pathways were identified in this study. Long-rang transport (LRT) and local sources characterized these pathways. Cluster 1 North Limpopo (NLP) is a local source from the northern province of Limpopo, which is associated with a high concentration of $PM_{2.5}$ due to anthropogenic activities happening in the area, which include coal power stations, mining, domestic fuel burning, agriculture, and veld fires. Cluster 2 Eastern Inland (EI) has its origin from the Indian Ocean while clusters 3, 4, and 5 (Short Indian Ocean SIO, Long Indian Ocean LIO, and South Westerly-Atlantic Ocean, respectively) depicts LRT sources. About 78% of the days in which the WHO and SA standard were exceeded were of local source origin (cluster NLP and EI), while only 22% was attributed to LRT sources.

To effectively develop $PM_{2.5}$ reduction strategies, the sources of PM and their contributions, as well as the contribution from each source, must be understood using a combination of transport cluster analysis. The later provides additional information on the pathway through which the sources originated.

Reference

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Research article

A street-level assessment of greenhouse gas emissions associated with traffic congestion in the city of Nairobi, Kenya

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Abstract

Traffic congestion significantly contributes to climate change due to the emissions of greenhouse gases such as carbon dioxide (CO_2), nitrous oxide (N_2O) and ozone (O_3). Rapid urbanization and poor planning coupled with increased motorization and fragmented public transport systems in cities such as Nairobi have led to increased vehicle emissions especially during heavy traffic along the various roads and within the central business district. To reduce greenhouse gas emissions in the urban transport sector, institutional coordination and relevant policy tools must be considered. This study aimed at estimating CO_2 emissions from different vehicle categories during traffic congestion, using Uhuru Highway as a case study. The relationship between traffic congestion and CO_2 emissions was analyzed using qualitative and quantitative methods, through a bottom-up approach. 120 questionnaires were administered to vehicle owners, passengers and pedestrians to get individual vehicle characteristics and opinions on the best actions for reduction of CO_2 emissions along Uhuru Highway in Nairobi. The average annual daily traffic (AADT) for different vehicles from 2014 to 2019 was used to estimate the CO_2 emissions. Results showed that private cars predominate over other vehicle types, contributing 73% of the total CO_2 emissions in the Nairobi CBD. Private cars are the highest contributors of CO_2 emissions with a total of 25.3 million grams of carbon dioxide equivalent (gCO_2e), between 2014 and 2019. In comparison, public service vehicles, commonly referred to as Matatus, emitted 6.89 million gCO_2e, light commercial vehicles 1.82 million gCO_2e, heavy goods vehicles 251,683 gCO_2e and motorcycles 181,054 gCO_2e. To minimize CO_2 emissions, the study recommends the enforcement of strong mobility policies to control the high motorization rate. One of these policies is the prioritization of the development of a mass public transport system to achieve the potential health, economic and environmental gains within the CBD.

Keywords

greenhouse gas emissions, urbanization, traffic congestion, CO_2 emissions, policy

Introduction

The transport sector plays a vital role in contributing to the successful implementation of all Sustainable Development Goals (SDGs). According to Partnership on Sustainable Low Carbon Transport (SLoCaT), the sector is considered as the engine of the global economy that helps in accelerating human development (SLoCaT, 2019). However, as the global economy grows, transport greenhouse gas emissions continue to rise, impacting on the successful implementation of the SDGs. The increase in emission of greenhouse gases is as a result of burning of fossil fuels (IPCC, 2014) which ultimately alters the climate causing adverse effects such as floods, droughts and heatwaves, especially in cities (La Notte et al., 2018).

The International Energy Agency (IEA) estimates that the transport sector is responsible for 24% of total CO_2 emissions (IEA, 2020). Road transportation accounts for nearly three quarters of these emissions. This includes emissions from traffic congestion which has become a common occurrence in cities and urban areas around the world.

Traffic congestion is a serious problem in urban road networks, especially in developing cities (Rajé et al., 2018). It contributes to an increase in travel time and fuel consumption, causes environmental pollution, decreases productivity and thereby imposes huge economic, social and environmental costs on the economy (Bharadwaj et al., 2017). Also, it results in emissions of harmful substances such as particulate matter, volatile organic compounds (VOCs) and also greenhouse gases such as carbon dioxide (CO_2), nitrogen oxides (NO_x), methane (CH_4) and ozone (O_3) (Transportation Research Board, 2002). Among the greenhouse gases, CO_2 is the most significant because of its long-term impact on global warming (having a lifespan of hundreds of years) as compared to other gases. CO_2 has the capacity to absorb excess heat from the sun and redirect it back to the surface of the earth, making the planet habitable. Increased concentrations of CO_2 trap heat, thereby causing enhanced warming hence climate change.

Studies have shown that CO_2 emissions from traffic congestion are influenced by different factors. Pandian et al. (2009) investigated the effects of traffic on vehicle emissions and revealed that traffic conditions, driving speed and pattern, vehicle and road characteristics, and vehicle composition greatly influence emissions especially near road intersections. Other factors such as the slope, density and length of the road, travel time, distance and speed of each vehicle on the road and class of the road that determines the traffic conditions and driving patterns, have also been reported to influence CO_2 emissions (Li et al., 2014; Zhang & Zhu, 2017; Zhu, 2013).

According to Raje et al. (2018) traffic congestion poses a huge threat to the social, economic and environmental development in many cities of both developed and developing countries. Cities in the USA, UK, Poland, Slovakia and Spain have experienced congestion which has resulted in increased CO_2 emissions (Chang et al., 2017). Another study conducted in Chennai, India showed that the transportation sector was one of the largest emitters contributing 29.7% of the total CO_2 emissions (Kumar and Nagendra, 2016). China is the world's biggest emitter of CO_2 (Zheng et al., 2020) whose transportation sector accounts for 9.3% of the country's total emissions. More than 8% of the total carbon emissions are from traffic congestion (Zhang et al., 2019), in which private vehicles constitute to the majority of the traffic especially in metropolitan areas such as Beijing (Li and Jones, 2015; Zheng et al., 2020). Other cities such as Mexico City, Bangkok, Singapore, Jakarta, Manila, Delhi and Mumbai have reported drops in their average speed during peak hours, an indication of heavy traffic congestion (Chang et al., 2017). Sao Paulo, Brazil, has been widely known to experience traffic congestion on a daily basis that lasts for 2-3 hours (Chang et al., 2017).

The majority of African countries also experience traffic congestion which results in massive delays as well as a decrease in productivity (Agyapong and Ojo, 2018). This can be attributed to a rapid increase in vehicle ownership coupled with rapid urbanization in many African countries (Doumbia et al., 2018), second-hand vehicles from the developed nations and inefficiency of public transport (Wojuade, 2018). South Africa has the highest greenhouse gas emissions of the African countries with the transportation sector accounting for 30.8% (Rhikhotso et al., 2016). The city of Tshwane in South Africa has experienced heavy traffic congestion, contributing 7.2% of South Africa's total CO_2 emissions (Rhikhotso et al., 2016). In Ivory Coast, Abidjan has also experienced intense traffic congestion which has significantly contributed to the increase in the city's emission levels (Doumbia et al., 2018). Kenya has experienced traffic congestion in its major cities of Nairobi, Kisumu and Mombasa (Salon and Gulyani, 2019). This has caused a rise in emission levels within the transportation sector which is responsible for about 11% of the country's total greenhouse gas emissions (Government of Kenya, 2018).

Due to the rapid increase in vehicle ownership, the emission levels have been projected to increase to about 14.7% of total greenhouse gas emissions by 2030 (Government of Kenya, 2018). It is estimated that the road transport sector accounts for 99% of greenhouse gas emissions from non-aviation transport sector in Kenya (Cameron et al., 2012).

The problem: Nairobi City with Uhuru Highway as a case study

Nairobi is the most populated city in Kenya with a population of about 4.4 million people (Kenya National Bureau of Statistics (KNBS), 2019) and has the highest concentration of sources of industrial air pollutants and vehicles in Kenya (Gaita et al., 2014; Maroa, 2019). As the city's population continues to increase, air quality continues to deteriorate as a result of increased vehicle emissions, exposing both the citizens and visitors to health risks (Kinney et al., 2011; Odhiambo et al., 2010; Rajé et al., 2018a).

Heavy traffic congestion experienced within the city of Nairobi is mainly attributed to the high rates of motorization (Gachanja, 2015), as 60% of the total registered vehicles in Kenya operate in Nairobi (Madara et al., 2018). Nairobi City has one of the longest average trips to work compared to other African cities which is attributed to heavy traffic congestion (Rajé et al., 2018).

As of 2013, the total person trip generation by persons living inside Nairobi City was 6.8 million-person trips in a day. The most popular mode of transport is walking (over 40%), with 28% of commuters using public transport and 14% use private vehicles (JICA, 2014). In 2015, the city generated 7.8 million trips per day (JICA, 2018).

In Nairobi, vehicle emissions contribute about 39% of fine particulate matter (Gaita et al., 2014). Heavy traffic congestion worsens the situation as it consumes much fuel, increases travel time and leads to environmental pollution (Bharadwaj et al., 2017). Pedestrians walking along the busy streets of Nairobi are exposed to pollution emitted from stationary vehicles during times of traffic congestion. Street vendors and traffic police who spend much of their day on congested streets are also affected by the emissions from motor vehicles (Kinney et al., 2011).

Public transport, mainly dominated by matatus and private cars have been reported to be the major contributors to traffic congestion, especially in the CBD of Nairobi (Mitullah, 2020).

This is attributed to inefficiencies such as not operating on schedules within the public transport, making Nairobi residents opt for private means of transport. As a result, private vehicles become more dominant on the roads.

Traffic congestion has imposed a heavy economic burden on the Kenyan economy. In a recent report by the Nairobi Metropolitan Area Transport Authority (NMATA), it was estimated that traffic congestion costs Kenya almost $1 billion per year, with an average travel time of about 57 minutes in Nairobi, making Nairobi the fourth most congested city in the world (Mwakaneno, 2019). This implies that the economic and environmental costs of traffic congestion in Nairobi are at an acute stage and therefore need to be explored further to reduce these costs.

Regulatory guidelines such as the Kenya Standard Code of Practice for inspection of road vehicles (KS 1515 of 2000) (governs motor vehicle inspection and hence vehicle emissions in Kenya) have been put in place to control vehicular emissions. However, Nairobi lacks a regular air quality management system, which includes management of greenhouse gas emissions such as CO_2 (Japan International Cooperation Agency (JICA), 2018). Therefore, there is need to assess the city's CO_2 emissions from traffic congestion which contribute to the overall ambient air quality.

Materials and methods

The study adopted an exploratory case study considering Uhuru Highway in Nairobi to investigate the relationship between traffic congestion and greenhouse gas emissions. Four data sources were used: questionnaire surveys, traffic counts, scheduled interviews and secondary sources. Various actors were involved in the study including pedestrians along the street, passengers using public vehicles, drivers/owners/vehicle operators and key informants from the relevant authorities within the study area.

Study area

The study was carried out in Nairobi City which serves as an economic, administrative, political and cultural center. Nairobi is one of the largest and fastest-growing cities in Africa (Mastrota, 2019). Located at 1.32°S and 36.9°E, it covers an area of approximately 696 km^2, with an estimated population of 4.4 million (Kenya National Bureau of Statistics (KNBS), 2019). Nairobi is possibly the city most affected by vehicle emissions from traffic congestion in Kenya. The study examines how traffic congestion contributes to greenhouse gas emissions within the boundaries of the central business district of Nairobi City along Uhuru Highway (Figure 1).

The highway is a major artery into and out of the central business district of Nairobi and experiences heavy traffic volume compared to other roads. It is one of the busiest and most congestion-prone highways in Nairobi (Kenneth et al., 2020). The road segment connects major highways in the country (Mombasa Road, Thika Road and Nairobi-Nakuru highway), and there is a good mix of vehicles during traffic congestion. The

Figure 1: *Uhuru Highway study area map, Nairobi central business district (Source: Map Quest, 2019)*

strategic location of the Jomo Kenyatta International Airport (JKIA) along Mombasa Road gives an indication of major traffic flow into and out of the central business district on Uhuru Highway. Major cities (Nairobi, Mombasa and Kisumu) and towns (Nakuru, Eldoret and Malindi) in the country are also connected through this highway.

Sampling strategy for questionnaires

A quota sampling technique was used to obtain information from three groups of people: vehicle owners, passengers and pedestrians who were the main participants in the survey. To ensure that the survey was representative, vehicle owners were further divided into five different groups: passenger cars, matatus, heavy goods vehicles, light commercial vehicles and motorcycles.

Following the sampling procedure of Van Dessel (2013), a target sample of n=120 for the questionnaire survey was needed to obtain a 95% confidence level with a ±5% margin of error. 130 participants were invited to participate in the survey, out of which the researchers obtained 105 completed questionnaires. This represents a response rate of 80.7%, which according to Baruch and Holtom (2008) is within the acceptable range of 80-85% for face-to-face questionnaires.

Questionnaire surveys

A questionnaire survey that involved both quantitative and qualitative data was developed to collect information on vehicle characteristics, vehicle activity and opinions on emission reduction strategies from traffic congestion. These variables provided information on fleet characteristics, composition, infrastructural challenges, ease of movement and emission reduction measures.

Face-face questionnaire interviews were conducted from 10-14 June 2019 by two trained research assistants between 10:00 and 18:00. The targeted sites are high density areas, which made it easy for the researchers to obtain information relevant to the study. These areas are business parking lots and petrol filling stations located along Uhuru Highway.

Responses from the questionnaire survey were also used to determine the Travel Time Index (TTI), a congestion indicator. TTI is the ratio of the time that a vehicle takes to move from one point to another during free flow as compared to during peak hours.

Scheduled interviews

Scheduled interviews provided more information on strategies that can be used to reduce greenhouse gas emissions from traffic congestion. The key people to provide this information were representatives from relevant authorities in the transport sector (Kenya National Highway Authority, Nairobi City County Environment Department, National Transport and Safety Authority and the National Environment Management Authority). These interviews were important as they enabled the researchers to get an in-depth understanding of the problem.

Secondary sources

Secondary data was obtained from different sources from the literature review, government databases, scholarly articles and also reputable websites. Secondary data that described the population of registered vehicles in Kenya was used to provide a comparative analysis of estimated CO_2 emissions at a street level (Uhuru Highway), sub-national level (Nairobi) and national level (Kenya) as 60% of the total registered vehicles operate in Nairobi (Madara et al., 2018).The other source of secondary data was the Average Annual Daily Traffic (AADT) from the Kenya National Highways Authority (KENHA) database. These data were used to estimate traffic-related emissions of CO_2 between 2014 and 2019 by using IPCC Tier 3 methodology. However, only data for 2014 was available at the KENHA database. Data for the subsequent years was projected up to 2019 using the growth rate formula in Equation (1)

$$A = P(1 + r/100)^n \tag{1}$$

where:
A is the current average annual daily traffic
P is the Average Annual Daily Traffic of the previous year
r is the traffic growth rate of the vehicle fleet (%)
n is the number of years projected

The traffic growth rates were obtained from the Road Sector Investment Program-2 2015-2019 from the Kenya Roads Board Database. Due to limitations of AADT data availability, 2014 was used as a base year, from which trends and patterns of CO_2 emissions were estimated.

Estimation of CO_2 emissions

According to the 2016 report of the International Energy Agency, all CO_2 emissions associated with fuel combustion depend on the volume of fuel burned, the fuel density, the carbon content of the fuel and the carbon fraction that is oxidized to CO_2 (IEA, 2016). CO_2 emissions from fuel combustion depends on the amount of carbon in the fuel, which is specific to the type and grade of the fuel. To determine the carbon content in the fuel, a chemical analysis is carried out. As such, the information can be obtained from the fuel supplier. However, if the carbon content of the fuel is not known, the calculation of CO_2 emissions is based on the predetermined emission factors. The factors will help approximate the carbon content of the fuel to quantify the amount of CO_2 that will be released when the fuel is burned. IPCC Tier 3 methodology (IPCC, 2001) was used to calculate CO_2 emissions using the formula in the Equation (2).

GHG Emissions of a transport activity =
Transport Activity × (2)
GHG emission factor per transport activity

The formula was further modified by Adhi (2018), to suit the study as shown in Equation 3. The primary data requirements for this approach include:

- Traffic count/flow: this provides the total number of vehicles per hour/day/week along the chosen area of study
- The total length of the road segment under investigation
- Vehicle activity (fuel consumption/fuel economy)
- Emission factors are only based on fuel consumption.

The basis for these emission factors is the Handbook of Emission Factors for Road Transport (HBEFA Version 3.3). (Notter et al., 2019).

$$E = A \times f \times l \times ef \tag{3}$$

Where;
E is the total CO_2 emissions (gram CO_2 per day)
A is the average number of vehicles of type A per day
F is the specific fuel consumption per vehicle type A (litres/km)
l is the length of the road (km)
ef is the emission factor (grams CO_2/litre)

The specific fuel consumption per type of vehicle is the local fuel economy obtained from Mbandi et al. (2019), INFRAS (2017b) and INFRAS (2018), representing Kenya's specific emission factors. Tank-to-wheel CO_2 emission factors have been used to develop trends and patterns from 2014-2019 for road vehicle categories along Uhuru Highway.

Traffic counts

Traffic counts involved the use of both primary and secondary data. Primary data was collected from 18-23 June 2019, and was conducted with the help of two trained research assistants. The research assistants went through training to ensure that they have adequate knowledge on the subject matter. Secondary data comprising the Average Annual Daily Traffic was obtained from the Kenya National Highway Authority for 2014.

Descriptive and inferential analysis

While questionnaire surveys targeted members of the public and included both qualitative and quantitative information, traffic counts and secondary sources were mainly quantitative in nature. Interviews provided qualitative information since the researchers needed to have an in-depth understanding of traffic congestion and its impacts, and actions that the relevant authorities have/are taking to solve the problem.

Descriptive analyses were conducted to determine statistical parameters of primary data which formed the basis of quantitative analysis. Inferential analysis was used to deduce from the sample data the perceptions of the targeted population of the traffic congestion and its impacts on emissions. Overall and detailed conclusions were drawn from the analysis based on mean, mode, frequencies and percentages to describe the findings of the study. Questions with non-numeric responses were matched and an assumed opinion reached based on the frequency of the responses in which a codebook was developed with the help of SPSS software. The output has been presented in the form of graphs, charts and percentages. The estimated CO_2 emissions were then used to generate a traffic emissions inventory for different vehicle types.

Results

Factors that contribute to traffic congestion

Feedback from the questionnaire surveys on factors that contribute to traffic congestion was classified into six categories: i) traffic management; ii) attitude; iii) road network; iv) urbanization; v) vehicle fleet; and vi) Unforeseen (Figure 2). All issues related to poor traffic control by the police officers or poor functioning of traffic lights as the main cause of congestion were classified under traffic management. Issues of having too many roundabouts or the road capacity being too small to accommodate the growing number of vehicles were classified under the road network factor. Issues of ignorance of traffic rules by the drivers and careless driving were classified under attitude

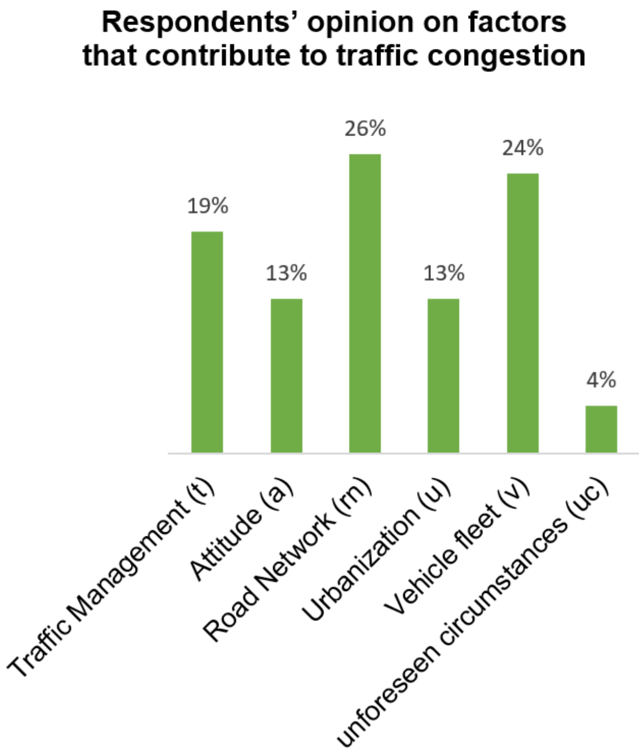

Figure 2: *Respondents' opinion on factors that contribute to traffic congestion along Uhuru Highway.*

while circumstances such as accidents or vehicle breakdown were classified under unforeseen circumstances. Vehicle fleet simply implies that congestion was caused by a specific type of vehicle. Lastly, the reasons given as due to increased motorization rates in Nairobi and too many vehicles using the highway at the same time resulting in congestion were classified under urbanization. In ranking these variables, it was observed that the inadequate road network was the most common reason given for congestion, followed by vehicle fleet and traffic management. Attitude and urbanization came in fourth while unforeseen circumstances came in last.

Congestion measures

Table 1 shows the average travel time and average TTI on Uhuru Highway for five vehicle categories. The varying distances travelled by different vehicle categories is attributed to the number of trips a particular vehicle takes. The average TTI for all vehicle classes on Uhuru Highway is 2.8, meaning that a vehicle

Table 1: *Travel time and travel time index for vehicles along Uhuru Highway, Nairobi*

Vehicle type	Average no. of daily trips	Distance travelled (km)	Average travel time (minutes)		Average Travel Time Index
			Peak hours (7-10am, 4-7pm)	Off-peak hours (10am-3pm)	
Passenger Cars	1	2.83	45	15	3
Matatus	5	14.15	30	10	3
Light Commercial Vehicles (LCVs)	2	5.66	45	15	3
Heavy Goods Vehicles (HGVs)	1	2.83	45	15	3
Motorcycles	8	22.64	10	5	2

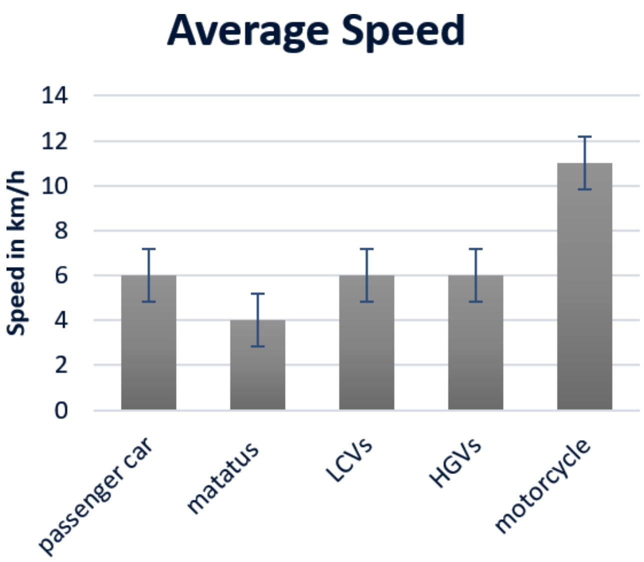

Figure 3: *Average speed for different vehicles on Uhuru Highway.*

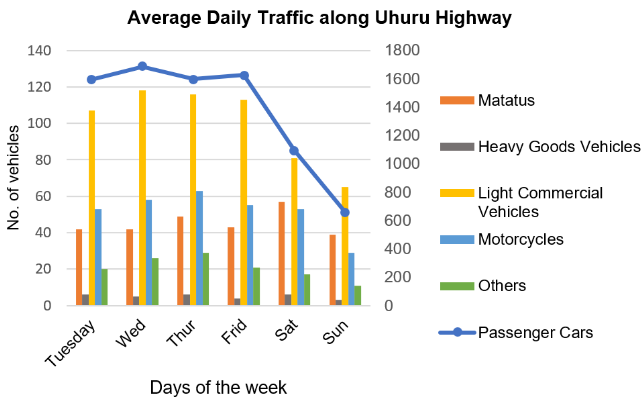

Figure 4: *Average daily traffic volume along Uhuru Highway.*

takes 2.8 times longer to do a certain trip in peak hours than in free flow traffic.

On average, passenger cars travel at an approximate speed of 6k m/h, *matatus* at 4 km/h, light goods vehicles at 6 km/h, heavy goods vehicles at 6 km/h and motorcycles at 11 km/h (Figure 3), regardless of the time (peak and off-peak hours).

A traffic count was carried out for six days in July 2019 (Figure 4). A similar pattern is observed on every day of the week. Passenger vehicles have the highest traffic count, recording an average number of more than 600 vehicles within a span of 15 minutes on a daily basis. The count also revealed that passenger cars have the highest traffic volume on Uhuru Highway on different days of the week. Wednesdays and Fridays had the highest traffic volume from the observed vehicle fleet, recording a total of 1934 and 1862 vehicles respectively. There was also a high volume of traffic on Tuesdays and Thursdays with 1821 and 1861 vehicles respectively. During the weekend, the traffic volume was moderate, averaging 1304 vehicles on Saturdays and 807 vehicles on Sundays.

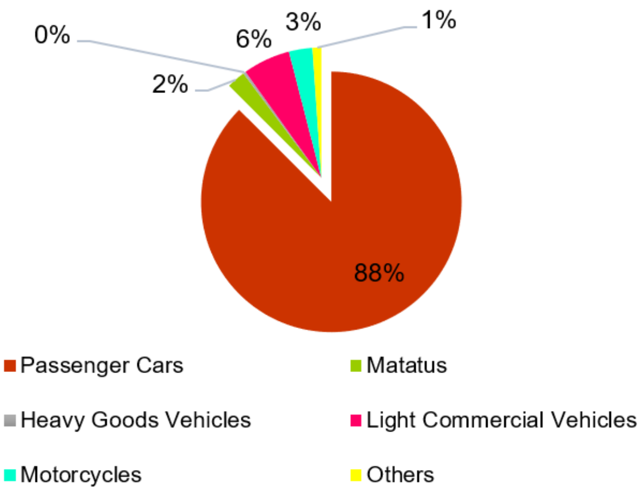

Figure 5: *Traffic volume composition along Uhuru Highway.*

Most vehicles on Uhuru Highway are passenger cars, comprising of 88% of the total traffic volume (Figure 5). This was followed by light commercial vehicles with 6% of the total traffic. Motorcycles came in the third place constituting 3%, followed by matatus with 2% while heavy goods vehicles comprised less than 1% of the total traffic volume.

Based on the responses from the questionnaire survey, the majority of residents use petrol rather than diesel. The fuel consumption rates were reported to be dependent on the usage of vehicles and number of trips made in a day. The majority of people with cars consume between 1-10 litres of fuel daily while some use more than 40 litres, depending on the number of trips.

Vehicle contribution to CO_2 emissions

Contribution of passenger cars

Passenger cars were the biggest contributors of CO_2 emissions, contributing cumulatively to a total of about 25 million grams of CO_2 equivalent per kilometer, from 2014-2019 (Table 2). This is because they are the most common vehicle on the highway. The increase in the total number of vehicles translates to the increase in CO_2 emissions. Private cars including station wagons and 4WD (four-wheeled drive) cars were all classified as passenger cars in this study.

Contribution of public service vehicles (PSV)

All public service vehicles (excluding taxis) providing public transport were categorized as matatus. This category of vehicles contributed the second most CO_2, about 6.9 million grams of CO_2 equivalent per vehicle kilometer (Table 2). The number of matatus on the highway has been on the rise since 2014 resulting in an increase in CO_2 emissions.

Contribution of light commercial vehicles (LCV)

This category of vehicle includes all medium-sized vehicles that are used for commercial purposes such as medium-sized lorries, pick-ups, vans and light buses. LCVs contribute a significant amount of CO_2 emissions and emissions have been increasing

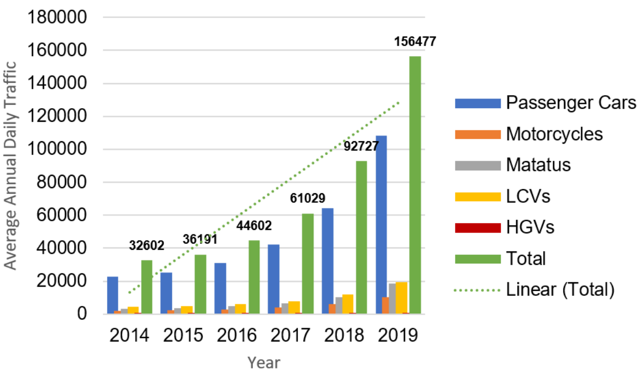

Figure 6: *AADT trend along Uhuru Highway between 2014 and 2019*

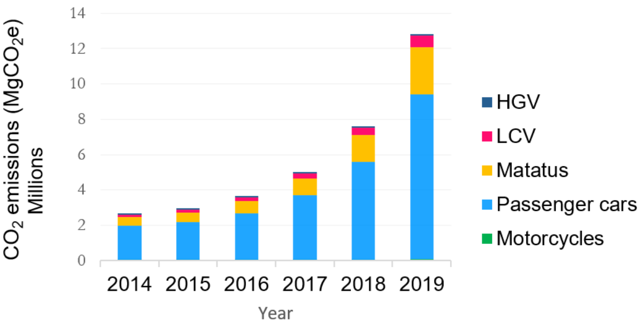

Figure 7: *Total estimated CO_2 emissions by vehicle category on Uhuru Highway between 2014 and 2019*

since 2014. Their emission factor is high, implying that they are among the worst polluters. LCVs have been estimated to contribute to about 650,000 gCO_2e/km by the end of 2019.

Contribution of heavy goods vehicle (HGV)

Heavy goods vehicles such as trucks and trailers have the lowest traffic volume on the highway. However, the number has been increasing gradually, with 2019 likely to have an average daily traffic of 160 trucks using the highway. Despite this group of vehicles having the least numbers, they are the worst polluters because of their high emission factor. By 2019, HGVs will have cumulatively contributed to about 250,000 gCO_2e/km, which is higher than that of motorcycles (181,000 gCO_2e/km).

Contribution of motorcycles

Motorcycles are the lowest contributors of CO_2 emissions. They also have the lowest emission factor. There has been a steady increase of CO_2 emissions by motorcycles on the highway. This trend is expected to increase owing to the increase in the number of motorcycles as a result of rapid urbanization, motorization and increase in population.

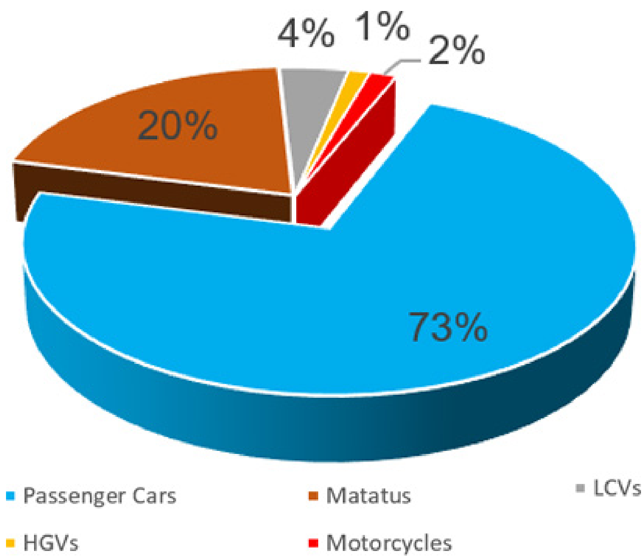

Figure 8: *Contribution of CO_2 emissions from Uhuru Highway by different vehicle categories*

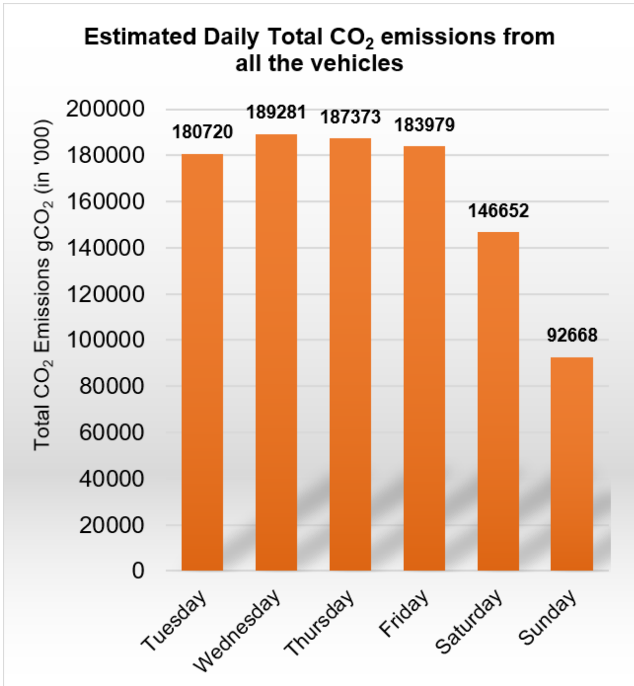

Figure 9: *Estimated daily CO_2 emissions along Uhuru Highway*

CO_2 emission levels

To estimate the CO_2 emissions from the different vehicle categories, a bottom-up approach was adopted. The primary data requirements were; the average daily traffic volume on

Table 2: *Cumulative estimated CO_2 emissions from 2014-2019 on Uhuru Highway*

Total estimated CO_2 emissions by vehicle category in 2014-2019					
Vehicle category	Motorcycles	Passenger cars	PSV	LCV	HGV
CO_2 emissions (Mt CO_2e/km)	0.18	25.35	6.89	1.82	0.25
Total vehicles	28072	293165	47296	54485	608

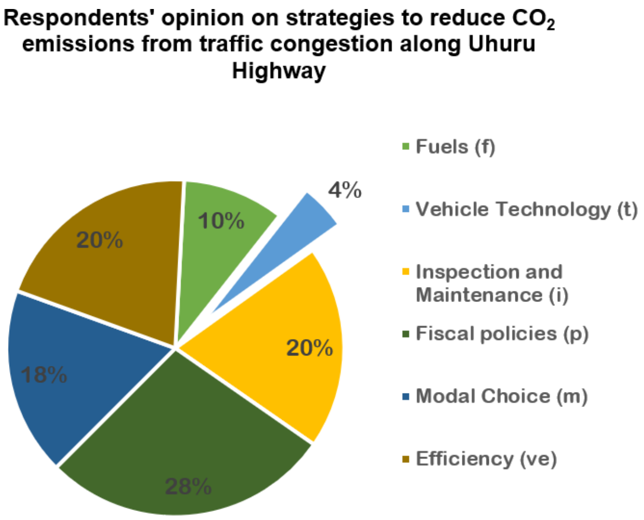

Figure 10: *Strategies to reduce CO_2 emissions from traffic congestion along Uhuru Highway*

Uhuru Highway, activity data of the vehicle (fuel consumption), emission factors and the total length of the road segment. The AADT has been increasing yearly with 2014 having a total of 32,602 vehicles of all types operating along the road (Figure 6). In 2015 there was also an increase in traffic volume recording a total of 36,191 vehicles, and 44,602, 61,029, 92,727 and 156,477 vehicles in 2016, 2017, 2018 and 2019 respectively.

There was a steady increase in CO_2 emissions from passenger cars, motorcycles, matatus, LCVs and HGVS between 2014 - 2019. By 2019, passenger cars were estimated to have contributed 9.3MgCO_2e, motorcycles 0.067 MgCO_2e, matatus 2.7 MgCO_2e, LCVs 0.65 MgCO_2e ang HGVs 0.067 MgCO_2e.

Passenger cars are the highest contributors to CO_2 emissions, comprising 73% of the total emissions followed by matatus (20%), LCVs (4%), Motorcycles (2%) and finally, the least contributors are the HGVs with 1% (Figure 8).

CO_2 emissions by day of the weekdays were highest on Tuesdays, Wednesdays, Thursdays and Fridays (180,720 gCO_2, 189,281 gCO_2, 187,373 gCO_2 and 183,979 gCO_2 respectively). On Saturdays and Sundays, the emission levels were lower at 146,652 gCO_2 and 92,668 gCO_2 respectively.

Interventions to reduce traffic-related CO_2 emissions

Figure 10 shows some of the interventions that could help in mitigating CO_2 emissions from traffic congestion as suggested by the questionnaire survey respondents. 28% of the respondents reported that enforcement of fiscal policies would be the best option to reduce CO_2 emissions from traffic congestion. This includes elimination of old and unroadworthy vehicles on the road, and imposing stricter rules on those that defy the already established regulations. The second strategy was ensuring proper inspection and maintenance, and improving vehicle efficiency of all vehicles which was supported by 20% of the respondents. Modal choice was proposed by 18% of respondents, use of quality fuels was supported by 10% and lastly was vehicle technology which was supported by 5% of the respondents.

Discussion

This study has demonstrated that traffic congestion contributes significantly to greenhouse gas emissions in cities such as Nairobi that experience heavy congestion during peak hours. Respondents acknowledged that congestion along Uhuru Highway is mainly caused by too many roundabouts and that the capacity of the road is too small to accommodate the growing number of vehicles. This means that the demand factors exceed the supply factors and as a result, vehicles move at sluggish speeds and greater speed variation, causing congestion. These findings conform with those of Koźlak and Wach (2018) that established demand-side factors to be more important than supply-side factors, hence contributing to congestion on urban roads.

Factors such as poor planning of the city, poor traffic management, increase in population, illegal parking, among others emerged to be the major causes of traffic congestion along Uhuru Highway. Studies by Rahane and Saharkar (2014) also conform to these findings, citing on-street parking as the major cause of congestion in cities. Additionally, Zhang et al. (2011) also noted that congestion mainly occurs as a result of rapid increase in car ownership and use, especially during rush hour periods in work zones. However, this study was only limited to specific types of vehicles during peak and off-peak hours on different days of the week in a specific work zone within the CBD of Nairobi City. Attitude, which includes arrogance, poor discipline and general driver behaviour, also emerged as a key contributing factor to traffic congestion along Uhuru Highway as reported by respondents during the questionnaire survey.

In Nairobi, passenger cars contribute the most to traffic congestion, and are the greatest contributor of CO_2 emissions. The calculations conform with those of Nejadkoorki (2008) who found out that passenger cars were the main source of greenhouse gas emissions accounting for 72.5% of all CO_2 emissions in Norwich. A similar study conducted in Tehran by Kakouei (2012) also revealed that private cars were the main sources of greenhouse gas emissions, contributing about 88% of CO_2 emissions from road transport.

Comparison with the Kenya National Greenhouse Gas Inventory

The Transport Inventory and Greenhouse Gas Emissions Reporting (TrIGGER) tool developed by GIZ is a bottom-up spreadsheet model that is used to calculate national transport greenhouse gas inventories for different countries, including Kenya (Scherer and Christoph, 2018). This tool was used to calculate CO_2 emissions for Kenya for the different vehicle categories in 2015 (Table 2).

Table 2: Total greenhouse gas emissions of different vehicle categories in Kenya in 2015 (Source: TriGGER, tool)

Vehicle Category	CO_2 emissions in million tonnes
Passenger cars	2.191
Light duty trucks	0.714
Heavy duty trucks & buses	3.48
Motorcycles	0.830

Uhuru highway contributes less than 1% of the total CO_2 emissions from transport in Kenya. According to Madara et al. (2018), 60% of the total vehicles registered in Kenya operate in Nairobi. This has been estimated by the National Transport and Safety Authority (NTSA) to have an average of 10% annual increase, and a total of 4-5 million vehicles by 2030 (Government of Kenya, 2015). The TriGGER tool provided a total of 7,199 Mt CO_2 emissions from the road transport sector in Kenya in 2015. Based on these results, vehicle emissions in Nairobi are likely to contribute 4.32 Mt CO_2, which is more than half of the total emissions from road transport in the country. Therefore, Nairobi needs to put in more efforts towards reducing emissions from road transport.

Policy implications

These results build on existing evidence that traffic congestion is a significant contributor to CO_2 emissions and air pollution. Previous research has focused on the contribution of traffic congestion to air pollution (Kinney et al., 2011; Gaita et al., 2014; Rajé et al., 2018; Singh et al., 2021). These results demonstrate that congestion is also a contributor to CO_2 emissions.

Traffic congestion in Nairobi increases fuel consumption and CO_2 emissions. This conforms with the findings of Bharadwal et al. (2017) in a study conducted in Mumbai Metropolitan Areas in which traffic congestion was reported to have a great impact on fuel consumption, travel time and CO_2 emissions. The stakeholder-led narratives reveal that the rapid growth rate of the economy in Nairobi city is a major driving force of traffic congestion. As the economy grows, there is increasing demand for a better lifestyle, including vehicle ownership (Brand et al, 2018). This could explain the steady increase in the number of vehicles, especially the passenger cars.

Nairobi City County Government (NCCG) is aware of these forces driving traffic congestion in the central business district and it is putting in a lot of effort to decongest the city, with the ultimate goal of improving air quality. Many innovative projects are underway to ease traffic such as the ongoing construction of the Nairobi Expressway which is expected to ease the flow of traffic through the city, reducing congestion along Mombasa Road, Uhuru Highway and Waiyaki Way, light rails, installation of intelligence traffic lights, traffic Marshalls and many more plans.

Enforcement of fiscal policies to eliminate old and unroadworthy vehicles, and imposing stricter rules on those that defy the already established regulations are key when proposing mitigative measures to reduce traffic congestion. Moreover, there should be effective stakeholder engagement for impactful interventions that benefit the public and promote sound environmental stewardship. The main lessons learnt from megacities such as Mexico City, Jakarta, Beijing, Singapore and Hong Kong highlight the importance of implementing policies such as driving restrictions, ownership restraints and other programmes that could limit growth of the vehicle fleet and improve the traffic conditions (Li and Jones, 2015; Song et al., 2018).

It must be acknowledged that this century offers a daunting challenge to control greenhouse gas emissions without affecting economic development, particularly in fast-urbanizing cities like Nairobi, where economic development, reducing unemployment and improving people's living standards are at the forefront. The steady growth of private vehicles in such cities will continue to have a significant impact on resource depletion and global warming if nothing is done to control the growth. It is important to have institutional coordination and appropriate policy tools in place so as to achieve CO_2 emission reduction in urban transportation. In addition, governance of urban transport must be strengthened, ensuring that transport policies are integrated into urban development plans. Also, to ensure that there is notable progress towards low-carbon transport, there must be involvement of a wide range of stakeholders as well as partners with a common goal and interests.

Conclusion

This study achieved its main objective by demonstrating that traffic congestion is a significant contributor to greenhouse gas emissions, particularly CO_2 emissions, in Nairobi.

Taking Uhuru Highway as a case study, the findings indicate that passenger cars are the highest contributors of CO_2 emissions because of their great numbers on the road segment. More efforts need to be put in place in to decongest the city and reduce CO_2 emissions. For example, adoption of car free days could help in lowering congestion in the city. Metropolises such as Kigali, Addis Ababa and Cape Town have already proved that car-free initiatives can work in sub-Saharan cities to help reduce congestion and encourage other alternative means of transport both during normal days and car-free days This goes beyond decongesting a city as people will claim their spaces in a friendly environment.

Nairobi city is growing fast and experiencing rapid growth in transport emissions. Allocation of the city's road space needs to be more equitable in order to accommodate people's needs. There is a strong desire for more sustainable mobility options within the city to reduce travel times and emissions, and create healthier and more livable cities. Most important is to implement effective policies that guide the changes in mobility within the city.

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Author contributions

Conceptualization, methodology, data collection and analysis, writing/revisions: CS; student supervision: CO, LO and AM.

References

Adhi, R.P. (2018) 'Top-Down and Bottom-Up Method on Measuring CO_2 Emission from Road-Based Transportation System (Case Study: Entire Gasoline Consumption, Bus Rapid Transit , and Highway in Jakarta, Indonesia)'. *Jurnal Teknologi Lingkungan*, 19(2), pp. 249–258.

Agyapong, F. and Ojo, T.K. (2018) 'Managing Traffic Congestion in the Accra Central Market, Ghana'. *Journal of Urban Management*, 7(2), pp. 85–96. DOI: 10.1016/j.jum.2018.04.002.

Baruch, Y. and Holtom, B.C. (2008) 'Survey Response Rate Levels and Trends in Organizational Research'. *Human Relations*, 61(8), pp. 1139–1160.

Bharadwaj, S., Ballare, S. and Chandel, M.K. (2017) 'Impact of Congestion on Greenhouse Gas Emissions for Road Transport in Mumbai Metropolitan Region'. *Transportation Research Procedia*, 25, pp. 3542–3555. DOI: 10.1016/j.trpro.2017.05.282.

Brand, C., Anable, J. and Morton, C. (2018) 'Lifestyle , Efficiency and Limits: Modelling Transport Energy and Emissions Using a Socio-Technical Approach'. Energy Efficiency. Available at: https://link.springer.com/article/10.1007/s12053-018-9678-9#citeas.

Cameron, L., Laura, W. and Seton, S. (2012) *Kenya's Climate Change Action Plan: Mitigation Chapter 7: Transportation*.

Chang, Y.S., Lee, Y.J. and Choi, S.S.B. (2017) 'Is There More Traffic Congestion in Larger Cities? -Scaling Analysis of the 101 Largest U.S. Urban Centers-'. *Transport Policy*, 59, pp. 54–63. DOI: 10.1016/j.tranpol.2017.07.002.

Van Dessel, G. (2013) *How to Determine Population and Survey Sample Size?*. Available at: https://www.checkmarket.com/blog/how-to-estimate-your-population-and-survey-sample-size/ (Accessed: 4 April 2019).

Doumbia, M. et al. (2018) 'Emissions from the Road Traffic of West African Cities: Assessment of Vehicle Fleet and Fuel Consumption'. *Energies*, 11(9), pp. 1–16. DOI: 10.3390/en11092300.

Gaita, S.M. et al. (2014) 'Source Apportionment and Seasonal Variation of $PM_{2.5}$ in a Sub-Sahara African City: Nairobi , Kenya'. *Atmospheric Chemistry and Physics*, (March). DOI: 10.5194/acpd-14-9565-2014.

Gastaldi, M. et al. (2017) 'On-Road Measurement of CO_2 Vehicle Emissions under Alternative Forms of Intersection Control'. *Transportation Research Procedia*, 27, pp. 476–483. DOI: 10.1016/j.trpro.2017.12.048.

Goh, M. (2002) 'Congestion Management and Electronic Road Pricing in Singapore'. *Journal of Transport Geography*, 10(1), pp. 29–38. DOI: 10.1016/S0966-6923(01)00036-9.

Government of Kenya. (2015) *Energy Regulatory Commission. Report on Global Fuel Economy Initiative Study in Kenya*.

De Grange, L. and Troncoso, R. (2011) 'Impacts of Vehicle Restrictions on Urban Transport Flows: The Case of Santiago, Chile'. *Transport Policy*, 18(6), pp. 862–869. DOI: 10.1016/j.tranpol.2011.06.001.

Hao, H., Wang, H. and Ouyang, M. (2011) 'Comparison of Policies on Vehicle Ownership and Use between Beijing and Shanghai and Their Impacts on Fuel Consumption by Passenger Vehicles'. *Energy Policy*, 39(2), pp. 1016–1021. DOI: 10.1016/j.enpol.2010.11.039.

IEA. (2016) *Key CO_2 Emission Trends. Excerpt from: CO_2 Emissions from Fuel Combustion*. Available at: http://www.iea.org/statistics/topics/CO2emissions/.

IEA. (2020) *Tracking Transport 2020*. Available at: https://www.iea.org/reports/tracking-transport-2020 (Accessed: 24 November 2020).

INFRAS. (2017) *Handbook Emission Factors for Road Transport (HBEFA). Version HBEFA 3.3*. INFRAS, Berne. Available at: www.hbefa.net/.

INFRAS. (2018) *Road Transport GHG Emission Factors for Kenya. Pilot Study for 2015*.

Japan International Cooperation Agency (JICA). (2018) *Republic of Kenya the Project on Detailed Planning of Integrated Trnasport System and Loop Line in the Nairobi Urban Core*. Nairobi.

Kakouei, A., Vatani, A. and Idris, A.K. Bin. (2012) 'An Estimation of Traffic Related CO_2 Emissions from Motor Vehicles in the Capital City of, Iran'. *Journal of Environmental Health Science and Engineering*, 9(1), pp. 1–5.

Kenneth, J.K., Mwea, S.K. and Dulo, S.O. (2020) 'Emissions of Vehicular Traffic along Uhuru Highway Corridor in Nairobi'. *Journal of Engineering and Architecture*, 8(2), pp. 22–27. DOI: 10.15640/jea.v8n2a4.

Kenya National Bureau of Statistics (KNBS). (2019) *2019 Kenya Population and Housing Census Volume I: Population By County and Sub-County*. Available at: http://www.knbs.or.ke.

Kinney, P.L. et al. (2011) 'Traffic Impacts on PM($_{2.5}$) Air Quality in Nairobi, Kenya.' *Environmental Science & Policy*, 14(4), pp. 369–378. DOI: 10.1016/j.envsci.2011.02.005.

Koźlak, A. and Wach, D. (2018) 'Causes of Traffic Congestion in Urban Areas. Case of Poland'. *SHS Web of Conferences*, 57, p. 01019. DOI: 10.1051/shsconf/20185701019.

Kumar, M.K. and Nagendra, S.M.S. (2016) 'Quantification of Anthropogenic CO_2 Emissions in a Tropical Urban Environment'. *Atmospheric Environment*, 125, pp. 272–282. DOI: 10.1016/j.atmosenv.2015.11.024.

Lakouari, N. et al. (2020) 'Modeling and Simulation of CO_2 Emissions in Roundabout Intersection'. *Journal of Computational Science*, 40, p. 101072. DOI: 10.1016/j.jocs.2019.101072.

Li, P. and Jones, S. (2015) 'Vehicle Restrictions and CO_2 Emissions in Beijing - A Simple Projection Using Available Data'. T*ransportation Research Part D: Transport and Environment*, 41(2015), pp. 467–476. DOI: 10.1016/j.trd.2015.09.020.

Li, Z. et al. (2014) 'Analytical Studies of CO_2 Emission in a Mixed Traffic Flow with Different Vehicles'. *Physica A: Statistical Mechanics and Its Applications*, 413, pp. 320–328. DOI: 10.1016/j.physa.2014.07.010.

Madara, O., James, N. and Rita, M. (2018) *Characteristics of the In-Service Vehicle Fleet in Kenya*. Nairobi, Kenya.

Maroa, J.C. (2019) 'An Assessment of the Contribution of the Corporate Taxi Industry to the Nairobi County Carbon Emission'. *International Journal of Current Business and Social Sciences*, 1(3), pp. 216–230.

Mastrota, E. (2019) *How Nairobi Became One Of The Fastest Growing Cities In The World. The National Digest in the World News*. Available at: https://thenationaldigest.com/how-nairobi-became-one-of-the-fastest-cities-in-the-world/ (Accessed: 1 May 2020).

Mbandi, A.M. et al. (2019) 'Estimating On-Road Vehicle Fuel Economy in Africa: A Case Study Based on an Urban Transport Survey in Nairobi, Kenya'. *Energies*, 12(6), pp. 6–8. DOI: 10.3390/en12061177.

Mitullah, W. V. (2020) *Kenya: Nairobi*. INC DOI: 10.1016/B978-0-12-815265-2.00007-8.

Mwakaneno, G. (2019) *JKIA-Westlands Expressway: The First Step Towards Solving Nairobi's Traffic Menace? The Kenyan Wallstreet*. Available at: https://kenyanwallstreet.com/expressway-between-jkia-and-westland-first-step-to-solving-nairobis-traffic-jam/ (Accessed: 25 January 2020).

Nejadkoorki, F. et al. (2008) 'An Approach for Modelling CO_2 Emissions from Road Traffic in Urban Areas'. *Science of the Total Environment*, 406(1–2), pp. 269–278. DOI: 10.1016/j.scitotenv.2008.07.055.

La Notte, A., Tonin, S. and Lucaroni, G. (2018) 'Assessing Direct and Indirect Emissions of Greenhouse Gases in Road Transportation, Taking into Account the Role of Uncertainty in the Emissions Inventory.' *Environmental Impact Assessment Review*, 69, pp. 82–93.

Odhiambo, G.O. et al. (2010) 'Motor Vehicles Air Pollution in Nairobi, Kenya'. *Research Journal of Environmental and Earth Sciences*, 2(4), pp. 178–187.

Pandian, S., Gokhale, S. and Ghoshal, A.K. (2009) 'Evaluating Effects of Traffic and Vehicle Characteristics on Vehicular Emissions near Traffic Intersections'. *Transportation Research Part D: Transport and Environment*, 14(3), pp. 180–196. DOI: 10.1016/j.trd.2008.12.001.

Rahane, S.K. and Saharkar, U.R. (2014) 'Traffic Congestion - Causes And Solutions: A Study Of Talegaon Dabhade City'. *Journal of Information, Knowledge and Research in Civil Engineering*, 3(1), pp. 160–163.

Rajé, F., Tight, M. and Pope, F.D. (2018) 'Traffic Pollution: A Search for Solutions for a City like Nairobi'. *Cities*, 82(June), pp. 100–107. DOI: 10.1016/j.cities.2018.05.008.

Salon, D. and Gulyani, S. (2019) 'Commuting in Urban Kenya: Unpacking Travel Demand in Large and Small Kenyan Cities'. *Sustainability (Switzerland)*, 11(14), pp. 1–22. DOI: 10.3390/su11143823.

Scherer, E. and Christoph, H. (2018) *User Manual TrIGGER Tool*. Available at: https://www.changing-transport.org/wp-content/uploads/2018_Manual_Technical_Report_TriGGER.pdf.

Singh, A. et al. (2021) 'Air Quality Assessment in Three East African Cities Using Calibrated Low-Cost Sensors with a Focus on Road-Based Hotspots'. *Environmental Research Communications*, 3(7). DOI: 10.1088/2515-7620/ac0e0a.

SLoCaT. (2019) *Sustainable Transport : A Critical Driver to Achieve the Sustainable Development Goals SUSTAINABLE TRANSPORT : A CRITICAL DRIVER TO ACHIEVE THE SUSTAINABLE*. Available at: https://slocat.net/wp-content/uploads/2020/02/SLOCAT_2019_transport_and_vnrs_2014-2019_final_version.pdf.

Song, Q. et al. (2018) 'Could Urban Electric Public Bus Really

Reduce the GHG Emissions: A Case Study in Macau?' *Journal of Cleaner Production*, 172, pp. 2133–2142. DOI: 10.1016/j.jclepro.2017.11.206.

Wojuade, C.A. (2018) 'Contribution of Automobile to Climate Change in Africa'. *JENRM*, 1, pp. 78–85.

Zhang, K., Batterman, S. and Dion, F. (2011) 'Vehicle Emissions in Congestion: Comparison of Work Zone, Rush Hour and Free-Flow Conditions'. *Atmospheric Environment*, 45(11), pp. 1929–1939. DOI: 10.1016/j.atmosenv.2011.01.030.

Zhang, L. et al. (2019) *A Review of China's Road Traffic Carbon Emissions*. Elsevier B.V. DOI: 10.1016/j.jclepro.2018.10.003.

Zhang, L.D. and Zhu, W.X. (2017) 'Heterogeneous Traffic Flow System Carbon-Dioxide Emission Control'. *Proceedings - 2017 Chinese Automation Congress, CAC 2017*, 2017-Janua(1), pp. 535–539. DOI: 10.1109/CAC.2017.8242826.

Zheng, J. et al. (2020) 'Comparative Analysis of the CO_2 Emissions of Expressway and Arterial Road Traffic : A Case in Beijing'. (2017), pp. 1–15. DOI: 10.1371/journal.pone.0231536.

Zhu, W.X. (2013) 'Analysis of CO_2 Emission in Traffic Flow and Numerical Tests'. *Physica A: Statistical Mechanics and Its Applications*, 392(20), pp. 4787–4792. DOI: 10.1016/j.physa.2013.06.024.

Research article

Exploring $PM_{2.5}$ variations from calibrated low-cost sensor network in Greater Kampala, during COVID-19 imposed lockdown restrictions: Lessons for Policy

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Abstract

Air pollution is considered a major public health risk globally, and the global South including sub-Saharan Africa face particular health risks, but there is limited data to quantify the level of pollution for different air quality contexts. The COVID-19 lockdown measures led to reduced human activities, and provided a unique opportunity to explore the impacts of reduced activities on urban air quality. This paper utilises calibrated data from a low-cost sensor network to explore insights from the diverse ambient air quality profile for four urban locations in Greater Kampala, Uganda before and during lockdown from March 31 to May 5 2020, highlighting the uniqueness of air pollution profiles in a sub-Saran Africa context. All locations saw year to year improvements in 24-hour mean $PM_{2.5}$ between 9 and 25µg/m^3 (i.e. 17-50% reduction from the previous year) and correlated well with reduction in traffic (up to approx. 80%) and commercial activities. The greatest improvement was observed in locations close to major transport routes in densely populated residential areas between 8 pm and 5 am. This suggests that the reduction in localised pollution sources such as nocturnal polluting activities including traffic and outdoor combustion including street cooking characteristic of fast-growing cities in developing countries, coupled with meteorological effects led to amplified reductions that continued well into the night, although meteorological effects are more generalised. Blanket policy initiatives targeting peak pollution hours could be adopted across all locations, while transport sector regulation could be very effective for pollution management. Likewise, because of the clustered and diffuse nature of pollution, community driven initiatives could be feasible for long-term mitigation.

Keywords

Ambient air quality, COVID-19, Low-cost sensors, Urban air quality, sub-Saharan Africa

Introduction

Air pollution is now considered one of the major public health risk factors for global morbidity and mortality, primarily associated with increased risk of respiratory illnesses, heart diseases, and there are growing links to mental health and cognitive impairment (Seaton et al. 1995; Brunekreef and Holgate 2002; Cohen et al. 2017; Pope III and Dockery 2006; Chen and Schwartz 2009; Xue et al. 2019). According to the World Health Organisation (WHO), more than 90% of the population in monitored urban centres worldwide are exposed to air pollution above WHO Air Quality Guideline (AQG) levels (World Health Organisation 2018; 2021). Populations in low- and middle-income countries such as those in sub-Saharan Africa with some of the highest urban population growth rates are among the most at risk of pollution exposure (United Nations 2018). In Africa, the socio-economic costs of air pollution are estimated to be much higher than malnutrition and unsafe sanitation (Roy 2016). Sub-Saharan Africa is home to over 475 million people (Lall, Henderson, and Vernon 2017), and the urban-settings face unique air quality challenges including; diffuse and clustered sources from increasing combustion emissions, increased informal settlements, lack of streamlined and efficient public transport systems, limited environmental regulations and urban planning deficiencies; in part arising from rapid urbanisation. (Petkova et al. 2013; Liousse et al. 2014). In the contemporary policy environment, diffuse pollution sources that often result into localised and clustered impacts present complexities for mitigation as the resulting pollution is from a conflation of activities within an air-shed as opposed to major point source situations. Ultimately, management of diffuse pollution cannot neatly fit within the conventional regulatory framework that allows for source-monitoring and permitting, and so spatio-temporal insights on pollution sources are essential to inform mitigation actions. This is the case for most of today's urban centres especially sub-Saharan Africa where diffuse activities continue to dominate the pollution profile (Liousse et al. 2014; Karagulian et al. 2015), but continuous monitoring datasets exploring different contexts remain extremely inadequate.

In this paper, we explore the impact of major disruptions to the scale of COVID-19 lockdown restrictions on ambient air quality for diverse physical environments within the same analytical unit/airshed considered. We hypothesize that the diversity and variations in the pollution profile in Greater Kampala, a typical sub-Saharan African context can provide unique insights not usually experienced in other geographic contexts during a major disruption, thus having important policy implications. The wide measure of variations in the restrictions adopted and implemented by different countries would equally be instructive in presenting each geographic context as a potential case study.

COVID-19 lockdown and air quality

There is growing body of literature on the relationship between air pollution and the COVID-19 pandemic. Evidence from recent preliminary studies suggests that exposure to high levels of particulate matter and fossil-related air pollution increases the risk of contracting COVID-19 and eventually mortality. Firstly, by raising the susceptibility of individuals by weakening lung function and secondly by particles providing a transmission mechanism for the spread of the coronavirus (Wu et al. 2020; Setti, Passarini, De Gennaro, Baribieri, et al. 2020; Setti, Passarini, De Gennaro, Barbieri, et al. 2020; Luigi Sanita di Toppi, Lorenzo Sanita di Toppi, and Bellini 2020; Travaglio et al. 2020), etc. Although still emerging, these linkages re-emphasise air pollution as an important public health risk. Since the WHO declared the pandemic on 12 March 2020, many countries introduced restrictions on mobility, social interactions and economic activities to contain the spread of the coronavirus and reduce the burden on health systems. The containment measures will undoubtedly have significant immediate and long-term impacts on the national and global economy, some of which are already being felt (Kabir et al. 2020; Nicola et al. 2020; Atkeson 2020; Baldwin and Tomiura 2020).

In almost equal measure, the restrictive measures have also had unintended consequences on the atmospheric environment including ambient air quality. Several specific case studies utilising data from satellite observations and ground station monitors have already been presented (Dantas et al. 2020; Mahato, Pal, and Ghosh 2020; Tobías et al. 2020). These studies indicate distinctive variations but largely significant reductions in air pollution in a range of environments. Susanta et al. (2020) conducted an analysis on the impact of air pollution during the lockdown period for New Delhi, India. The study shows an improvement in air quality by about 50% Nitrogen dioxide (NO_2), 30% Carbon monoxide (CO), and 50% particulate matter ($PM_{2.5}$ and PM_{10}) compared to the days immediately before (Mahato, Pal, and Ghosh 2020). A study in Barcelona, Spain showed a reduction of 45-51% for NO_2 and Black Carbon, 28-31% for PM_{10}, and an increase of 33-57% for Ozone (O_3) during the lockdown period (Tobías et al. 2020). Venter et al. (2020) found reduction of 60% and 31% for NO_2 and $PM_{2.5}$ respectively for population weighted concentrations with a 95% Confidence Interval. There are also several other studies that explored changes in air pollution for different locations worldwide (He, Pan, and Tanaka 2020; Venter et al. 2020; Ju, Oh, and Choi 2021; Mostafa, Gamal, and Wafiq 2021). However, to the best of our knowledge, a few studies (McFarlane et al. 2021) have published research investigating the impact of the COVID-19 lockdown on air pollution in urban sub-Saharan Africa while exploring the implications for air pollution mitigation. This is partly because ground monitoring remains extremely limited in Africa (Petkova et al. 2013; Liousse et al. 2014; World Health Organisation 2016) due to the prohibitive costs of establishing and maintaining traditional air monitoring networks that lead to sparse distribution of monitoring networks in resource-strained environments.

Improvements in low-cost air quality sensing technologies provide opportunities to characterise and measure air quality at micro-level and at a higher resolution, difficult to achieve with expensive grade reference monitors alone (Castell et al. 2017; Morawska et al. 2018; EPA 2020b; EPA 2020a). This approach is particularly important for low- and middle-income countries where the cost of monitoring remains a major inhibitor for air quality control and management programs. This paper leverages advances in low-cost measurement technologies to explore and quantify the variations in particulate matter ($PM_{2.5}$), a common measure of ambient air quality (Pope III and Dockery 2006; R. T. Burnett et al. 2014; World Health Organisation 2016), for selected diverse urban locations in Greater Kampala, Uganda, with regard to the lockdown restrictions, while discussing implications for air quality management.

Materials and methods

COVID-19 lockdown timelines in Uganda

The first case of COVID-19 in Uganda was recorded on March 22 2020, by May 15, Uganda had registered 203 cases. On March 18, the President announced the first measures to curb the spread of COVID-19. This began with the closure of schools and universities and suspension of public gatherings.

On March 21, borders were closed and all incoming and outgoing passenger aircraft and vehicles were prohibited. This was followed by suspension of all forms of public transport on March 25. On March 30 movement for all public and private vehicles with exception of cargo delivery and authorised essential activities was prohibited. Businesses were closed and outdoor movement was restricted to 06:30 to 19:00[1]. For the purposes of this paper, we define the period beginning March 31 2020 through to the end of the second extension on May 5 2020 as the 'lockdown period'.

[1] https://www.yowerikmuseveni.com/speeches

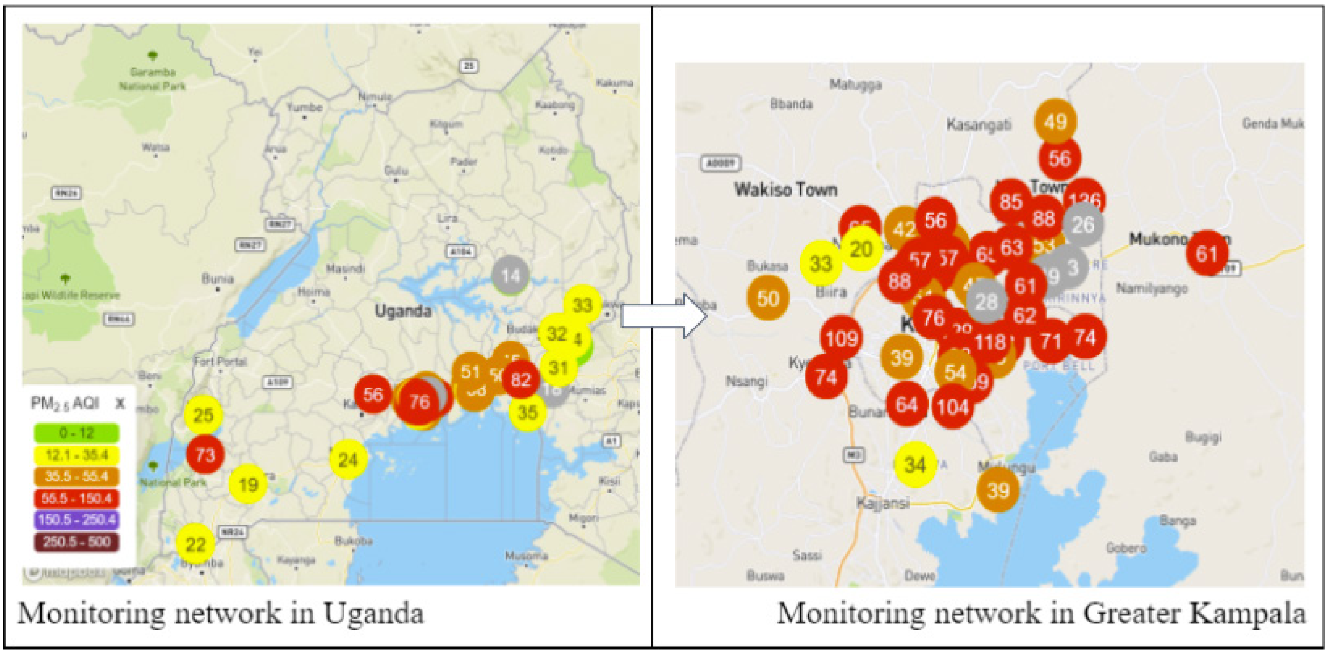

Figure 1: *Monitoring network in Uganda as at 01:04 EAT; 06-Oct-2021*

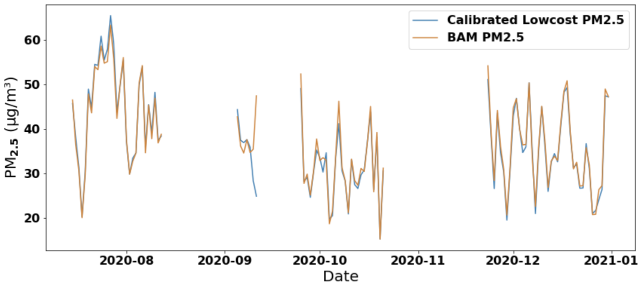

Figure 2: *$PM_{2.5}$ comparisons for AirQo device AQ88 vs BAM collocated at Makerere University*

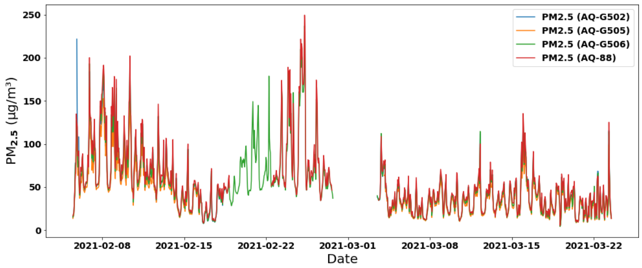

Figure 4: *: Comparison between hourly $PM_{2.5}$ values between AirQo devices collocated at Makerere University*

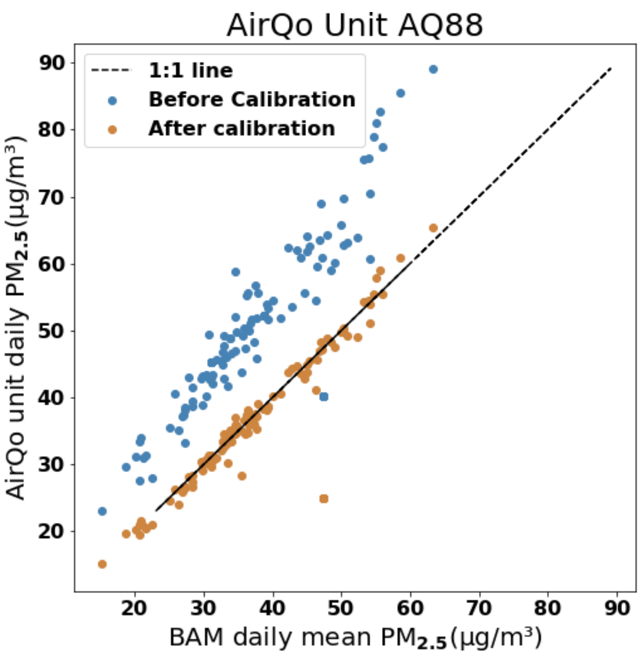

Figure 3: *A scatter plot showing the relationship between BAM and AirQo unit $PM_{2.5}$ before and after calibration.*

Measurement of air quality

$PM_{2.5}$ measurements were obtained from a network of low-cost sensors deployed across Greater Kampala (Figure 1), operated and managed by AirQo (www.airqo.africa) (AirQo 2020; Coker et al. 2021). Each AirQo device uses twin Plantower (PMS 5003) light scattering sensors and transmit averaged measurements every 90 seconds (for static installations) via local Global System for Mobile Communications (GSM) network to a cloud-based platform. The devices have a measurement range of 0-500µg/m^3 for both $PM_{2.5}$ and PM_{10} and are optimised to run on solar energy or mains to cater for limited power availability, typical of urban settings in Sub-Saharan Africa.

As part of data quality assurance, AirQo devices are collocated with a Met One Beta Attenuation Monitor (BAM 1022) Federal Reference Monitor approved to international standards and generally correlate well against the BAM with correlation coefficient (R) of more than 0.9. The collocation data is used to develop a calibration model that translates PM concentrations from AirQo devices to BAM equivalent. In this paper, we applied random model trained with data from an AirQo to BAM collocation site at Makerere University (coordinates: 0.333534, 32.568644) from 15th July 2020 to 23rd March 2021. Employing

(a) Bugolobi area

(b) Bweyogerere area

(c) Civic Center area

(d) Entebbe Kiwafu area

Figure 5: *Study locations in and around Kampala used to describe the variations of ambient air quality before and during the lockdown period.*

the calibration model increased BAM to low-cost correlation (R) and (R^2) values from 0.9 to 0.95 and 0.49 to 0.9 respectively; and decreased RMSE and MAE from 19.11µg/m^3 to 8.2 µg/m^3 and 14.99 µg/m^3 to 5.1 µg/m^3 respectively.

Figure 2 and 3 illustrate the comparison between calibrated $PM_{2.5}$ values from AirQo devices collocated with the BAM at Makerere University between July and December 2020. It can be seen that calibrated output from the low-cost sensors follow a similar trend with the BAM with slight upward and downward shifts, with R = 0.97 for AQ88[2]. AQ88. This is relatively comparable to other Plantower sensor evaluation studies (Kelly et al. 2017; Levy Zamora et al. 2018; Mukherjee et al. 2019; Liu et al. 2020) and gives a degree of confidence in the ability of the low-cost network to provide reliable insights on the spatio-temporal variations within a given study area.

We emphasise that data from AirQo to AirQo correlate strongly with both mean correlation coefficient of R^2 = 0.98 for four AirQo units collocated at Makerere university tween 15th July 2020 to 23rd March 2021 (see Figure 4).

Implicitly, we consider calibration model trained using field collocation datasets from one AirQo monitor to be largely applicable other AirQo monitors deployed within similar physical environmental setting and context. We therefore use the calibration model developed from collocation with the reference monitor at Makerere University to correct $PM_{2.5}$ concentrations from all devises used in this study. This data correction approach is not unique to this study and has previously been adapted for low-cost datasets from different settings e.g. Barkjohn et al. (2021), and McFarlane et al. (2021).

Study location (context)

The study area includes four sites, within the Greater Kampala Metropolitan Area (GKMA), Uganda (Figures 5 and 6), furthest point about 45 km apart.

Kampala City is the economic capital and administrative centre of Uganda with a resident population of over 1.5 million and an additional daytime transient population of over 2.5 million (Uganda Bureau of Statistics 2016; World Bank 2018).

GKMA has the highest population density in the country and hosts over 32% of manufacturing businesses, contributes more than one-third of the annual GDP, and ultimately hosts the greatest concentration of pollution generating activities in Uganda (Uganda Bureau of Statistics (UBOS) 2011; Uganda Bureau of Statistics 2016; World Bank 2018). Like many sub-Saharan African cities, Kampala is urbanising fast with one of the highest urban population growth rates in the world at about 5.6% (United Nations 2018; Vermeiren et al. 2012), leading to increased demand for resources and social services, and subsequently increased alterations of the natural environment.

[2] The data gaps in figure 2 between September & October and November & December is due to prolonged outage of the sensors

Table 1: *Sensor correlation (R) matrix for the low-cost AirQo network in Greater Kampala*

Monitoring sites	Nak II	Kas.	Nan. E	Lub.	Nan. W	Luk.	Bug.	Kya.	Seg.	Kiw.	Kwt.	CvC.	Mak.
Nakaseero II (Nak. II)	1.00	0.822	0.682	0.548	0.260	0.824	0.852	0.630	0.660	0.597	0.656	0.838	0.772
Kasanga (Kas.)	0.822	1.00	0.617	0.523	0.119	0.855	0.918	0.639	0.609	0.651	0.729	0.779	0.744
Nansana east (Nan. E)	0.682	0.617	1.000	0.472	0.407	0.725	0.684	0.579	0.541	0.499	0.501	0.709	0.707
Lubaga (Lub.)	0.548	0.523	0.472	1.000	0.257	0.521	0.509	0.432	0.494	0.462	0.488	0.549	0.492
Nansana west (Nan. W)	0.260	0.119	0.407	0.257	1.000	0.266	0.192	0.233	0.275	0.088	0.042	0.337	0.408
Lukuli (Luk.)	0.824	0.855	0.725	0.521	0.266	1.00	0.891	0.658	0.626	0.633	0.640	0.842	0.896
Bugolobi (Bug.)	0.852	0.918	0.684	0.509	0.192	0.891	1.00	0.673	0.623	0.631	0.696	0.857	0.796
Kyaliwajjala (Kya.)	0.630	0.639	0.579	0.432	0.233	0.658	0.673	1.00	0.496	0.469	0.531	0.625	0.616
Seguku (Seg.)	0.660	0.609	0.541	0.494	0.275	0.626	0.623	0.496	1.00	0.549	0.522	0.643	0.587
Kiwafu (Kiw.)	0.597	0.651	0.499	0.462	0.088	0.633	0.631	0.469	0.549	1.000	0.548	0.568	0.535
Kiwatule (Kwt.)	0.656	0.729	0.501	0.488	0.042	0.640	0.696	0.531	0.522	0.548	1.00	0.614	0.539
Civic Centre (CvC.)	0.838	0.779	0.709	0.549	0.337	0.842	0.857	0.625	0.643	0.568	0.614	1.000	0.817
Makindye (Mak.)	0.772	0.744	0.707	0.492	0.408	0.896	0.796	0.616	0.587	0.535	0.539	0.817	1.00

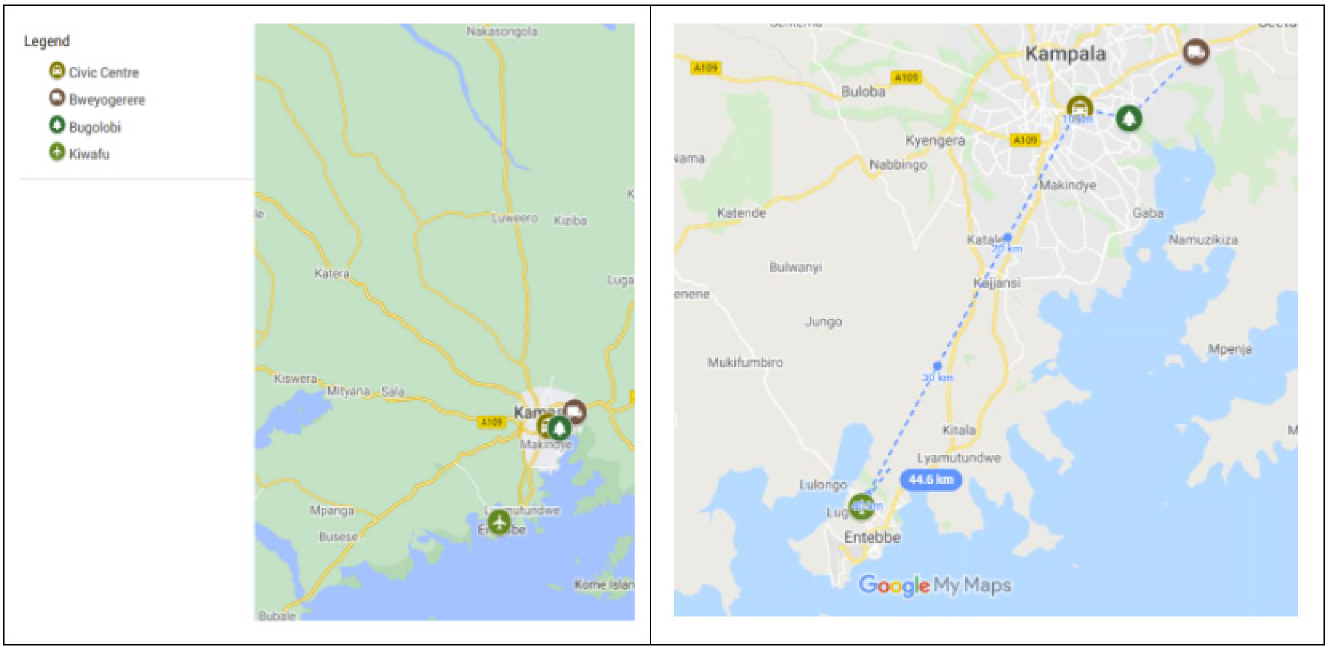

Figure 6: *Furthest distance between the monitoring locations*

Consequently, a large proportion of the population within GKMA live and work near pollution sources. The prevailing urban planning shortfalls have precipitated the growth of saturated informal settlement clusters (Richmond, Myers, and Namuli 2018) often intertwined with formal settlements and local pollution generating activities. In essence, local pollution levels tend to be largely influenced by the usually diverse and diffuse surrounding pollution generating activities coupled with the nature of settlements resulting into largely localised impacts (Okure et al. 2022). However, the unique variations in the air pollution profile is typical for many fast-growing urban areas in sub-Saharan Africa where pollution is dominated by multiple diffused sources (Marais and Wiedinmyer 2016; Pfotenhauer et al. 2019).

Table 1 shows the Greater Kampala pollution variations (clustered airsheds) using the sensor correlation matrix for 13 monitoring locations over a 6-month period, with an average correlation of 0.581, a function of the distance between the sensors and actual pollution levels in the monitoring locations/ intensity of immediate local sources. This socio-economic context suggests that the consequences of the disrupted socio-economic interactions with the ambient environment due to the lockdown restrictions will be more significant in this region, also consistent with novel findings on pollution progression and urbanisation (Mage et al. 1996). While in-depth investigation of the sensor variations highlighted in Table 1 is beyond the scope of this paper, we find that it reinforces the need for exploring air quality dynamics in a metropolitan city. Air quality insights from

Table 2: *Profiles of the study locations (Uganda Bureau of Statistics 2016)*

	Area km²	Population	Popn Density	Households per km²	Firewood/ Charcoal cooking households per km²	Domestic waste burning households per km²
Bugolobi	3.88	5023	1,295	327	103	23
Bweyogerere	11.27	58,679	5,207	1,363	1,153	575
Civic Centre	~1	375	375	130	24	1
Kiwafu Ward	5.30	22,243	4,194	1,081	238	242

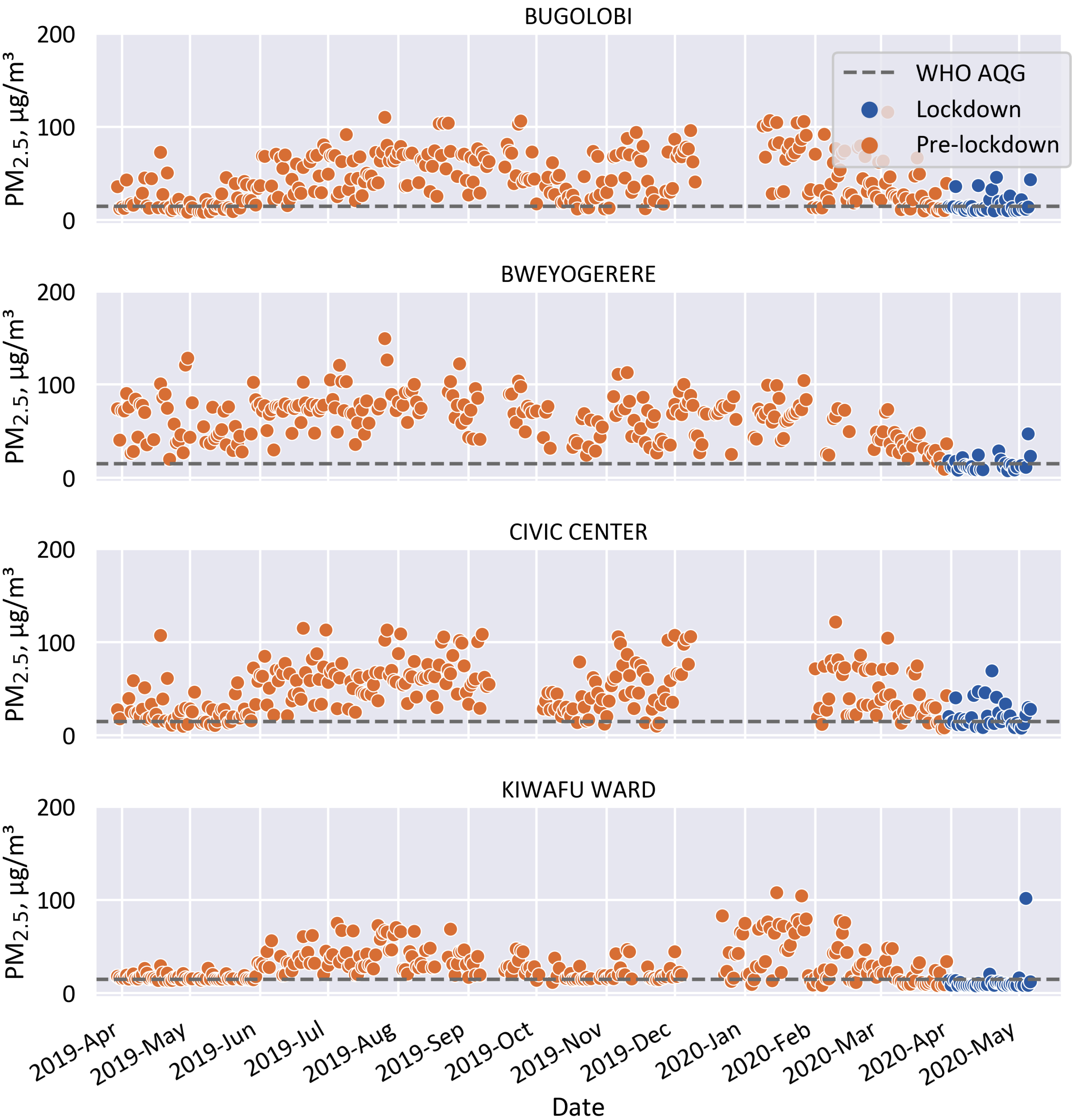

Figure 7: *Daily mean $PM_{2.5}$ concentrations, by location March 31 2019 - May 5 2020*

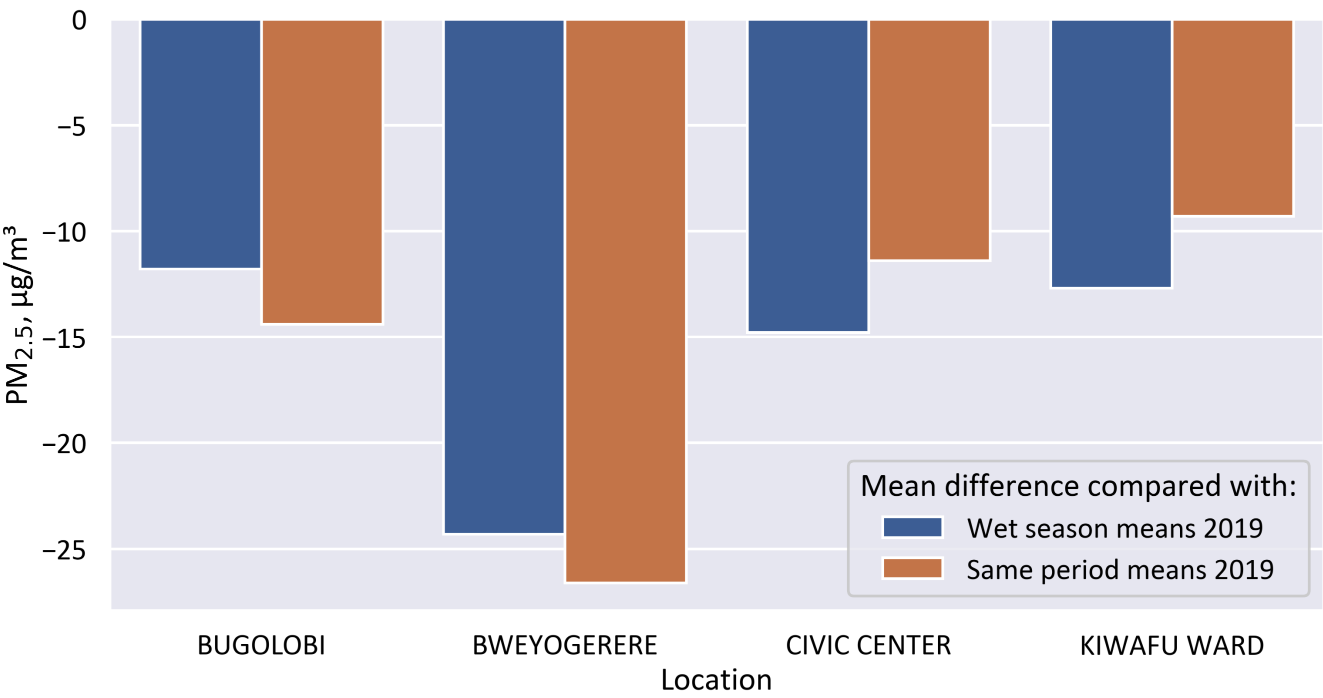

***Figure 8:** Mean reduction $PM_{2.5}$ values for lockdown compared to typical wet season 2019 and equivalent lockdown period prior year*

Greater Kampala during such an unprecedented circumstance would provide a rare opportunity for exploring mitigation policy insights for a sub-Saharan African setting.

In order to capture a cross section of the GKMA urban environment, we used parish level population distribution, observed satellite imagery from Google earth, knowledge of local context and availability of data for the 211 period of interest to identify four monitoring locations based on land use clusters i.e., residential/urban background and urban/traffic typically characteristic of land uses within the greater Kampala. This is also informed by the known pollution dynamics for various land uses (Harrison and Deacon 1998; Spangl et al. 2007; Alsahli and Al-Harbi 2018).

Inherently, diverse pollution profile would make it difficult to identify homogeneous land use clusters, which in theory, could introduce an analytical uncertainty. However, we attempted to obtain a fair spatial representation of the idealised location clustering which should provide spatially representative reference insights for the respective land uses taking into consideration that particulate matter is not always entirely constrained to immediate local sources like gaseous pollutants e.g. NO_2, and CO (Khuzestani et al. 2017), this does not present any significant limitations.

The locations represent a balance between densely and less populated areas with a wide measure of variations in pollution levels. The respective physical and demographic profiles for attributes with direct impact on local air pollution are presented in Table 2.

Results and discussion

Daily $PM_{2.5}$ levels

Figure 7 shows that for all locations there is substantial variation in daily mean $PM_{2.5}$ across the year. The blue section of the chart captures the lockdown period which records some of the lowest readings of the year for all locations. The cycles observed above vary in line with seasons with the two wet seasons (March-May/September-November) experiencing the lowest $PM_{2.5}$ levels. This corresponds to the influence of precipitation and unstable weather conditions on particulate suppression and decay (Chow et al. 1999; Yan et al. 2016).

Overall, Bweyogerere area which has greater population density, proximity to industries, with a major road network, and multiple diffuse sources, observes typically higher $PM_{2.5}$. Unsurprisingly, Kiwafu Ward in the smaller town of Entebbe with no major through-roads, less households using charcoal/firewood coupled with limited industrial and commercial activities experiences much lower levels. This suggests that both location-specific and meteorological factors impact on observed air quality during lockdown, with meteorological being more generalised (Cole and Neumayer 2004).

To establish the extent to which improvements in air quality coincide with lockdown measures, levels during lockdown are compared with wet season means for 2019, and the mean for the lockdown equivalent period in the previous year.

The results are as shown in Figure 8 with the bars representing the estimated differences in $PM_{2.5}$ between the two periods. Bweyogerere shows the greatest observed reduction in $PM_{2.5}$ at approximately 25µg/m³ or 50% lower than seasonal and

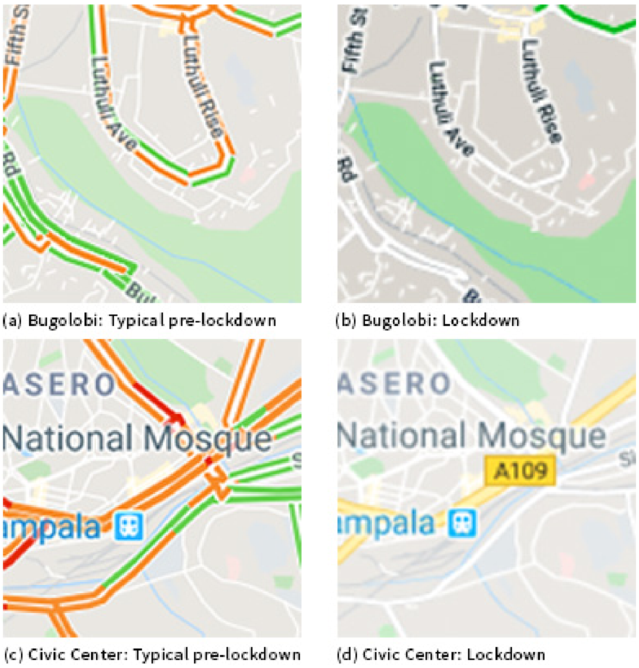

Figure 9: *Sample traffic activity images from Monday 8pm typical pre-lockdown and during lockdown*

the previous year means. Other locations saw reductions of around 9-15µg/m³. This is an indication that meteorology is not sufficient to explain the lower $PM_{2.5}$ levels during the lockdown period, but at the same time highlights the clustered nature of the pollution reduction. Modelling the meteorological impact on daily levels could provide precise estimates on the meteorological influence.

To further explore the extent of the impact of lockdown restrictions on the different physical environments, we used a working assumption that the impact of socio-economic factors i.e. population density, household energy use, will remain relatively constant during the lockdown period and possibly increase as people spend more time, cook and eat more at home than would normally be the case. We utilised data from the most recent census (Uganda Bureau of Statistics 2016) to obtain context for each of the land use areas. Bweyogerere, the most densely populated and probably the largest domestic emitter again experienced the greatest reduction in $PM_{2.5}$. Other parishes, from the densely populated Kiwafu Ward to the largely resident free Civic Centre experience comparable reductions. These insights provide little indication to suggest that the probable changes have led to a decrease in $PM_{2.5}$ that outweighs any benefits from other lockdown measures.

Localised influence of traffic activity

Because of the prohibitive logistical implications of conducting traffic counts; we employed Google Maps Traffic App that utilises smartphone data with activated location features, and recorded in motion to capture proxy data. 'Typical' traffic data was captured for day of the week between 6 am and 10 pm (the times available from Google Maps) for each location prior to lockdown. Hourly data from coloured pixels was collected throughout the lockdown period. Figure 9 demonstrates an example of the difference in activity after curfew, before and during lockdown. Figure 10 shows the level of traffic activity for each location at lock down levels compared to typical. Kiwafu Ward shows the greatest reduction in traffic activity with levels below 20% (about 80% reduction) of typical.

Access to the international airport and recreation are the main reasons for visiting the town and these options were no longer possible. Other locations show much higher activity, in the range of 20 to 40% of normal in the first two weeks of lockdown but rising to between 40 and 60% of normal in the second two weeks with Civic Centre as the most active.

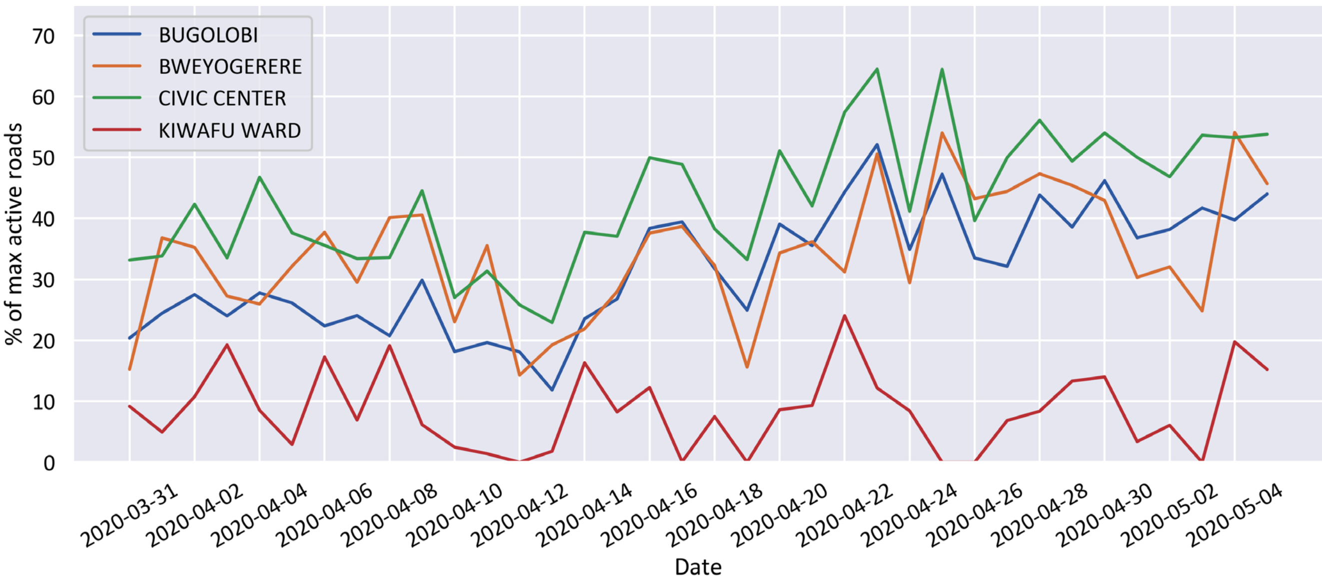

Figure 10: *Daily traffic activity during lockdown as a percentage of maximum typical levels*

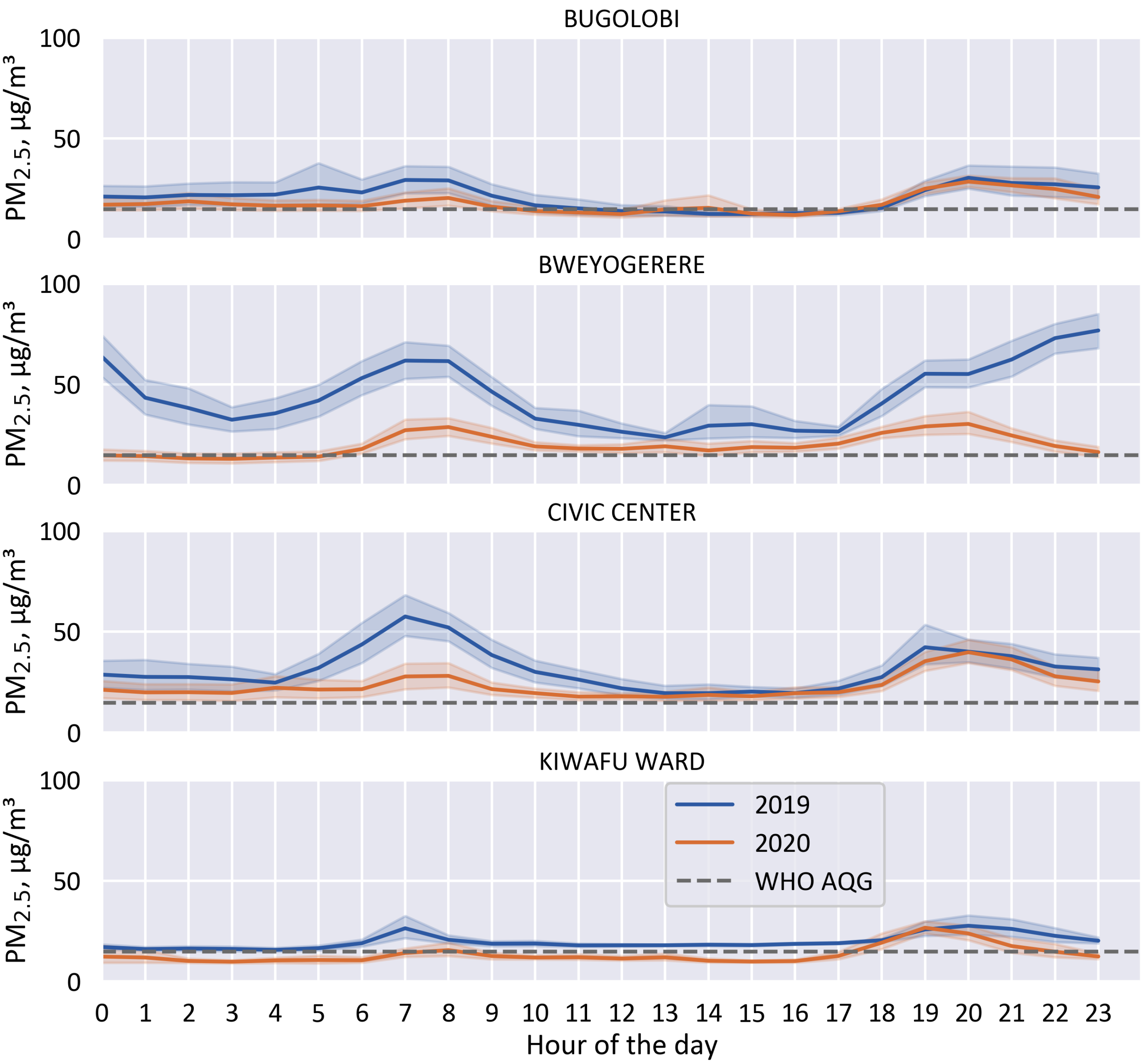

Figure 11: *Diurnal variations for lockdown period 2019 and 2020*

These locations, while varying in population density, remain on or close to major commercial through-routes across the city and so see higher sustained traffic volumes.

While Bugolobi has low population density and is beside wetland, it lies downwind from an informal settlement, an industrial area and a congested road which may explain higher activity rates.

Explanations for the increase over time may include; businesses adapting to the changing environment by investing in permissible transport such as motorcycles, home delivery services or increased mobility for essential workers and emergencies. This upward trend is not observed in daily $PM_{2.5}$ changes which are influenced more by daily weather variations. In summary, while the season of the year explains some of the improvement in daily air quality during the lockdown period, influence of lockdown restrictions appear to have been very significant. More detailed modelling of weather and pollution transport be required to identify the relationship between these features more precisely, while considering other emission parameters such as NO_2 (Mage et al. 1996; Watson and Chow 2002).

Diurnal variation

In Figure 11 we explore the difference between diurnal $PM_{2.5}$ levels during the lockdown period and the equivalent period in 2019. We observe that all locations carry the characteristic sinusoidal profile. A characteristic feature for pollution dispersion is the Boundary Layer Effect (BLE) (Ding et al. 2005) which creates the U-shape curve observed during daylight hours. As the sun rises around 7am, the ground warms due to radiation. This warms the air which rises lifting particulate matter with it creating

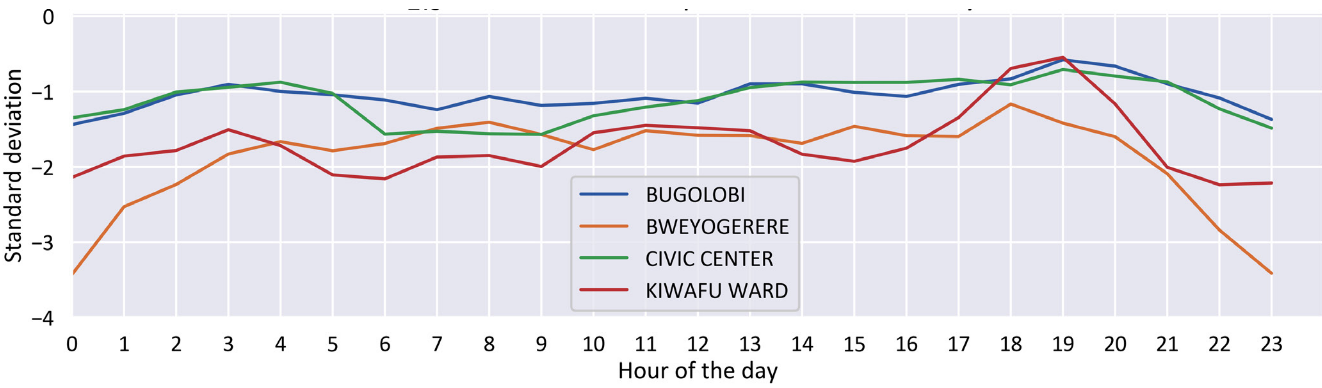

Figure 12: *Normalised $PM_{2.5}$ variation for the period March 31-May 5, 2019 to 2020*

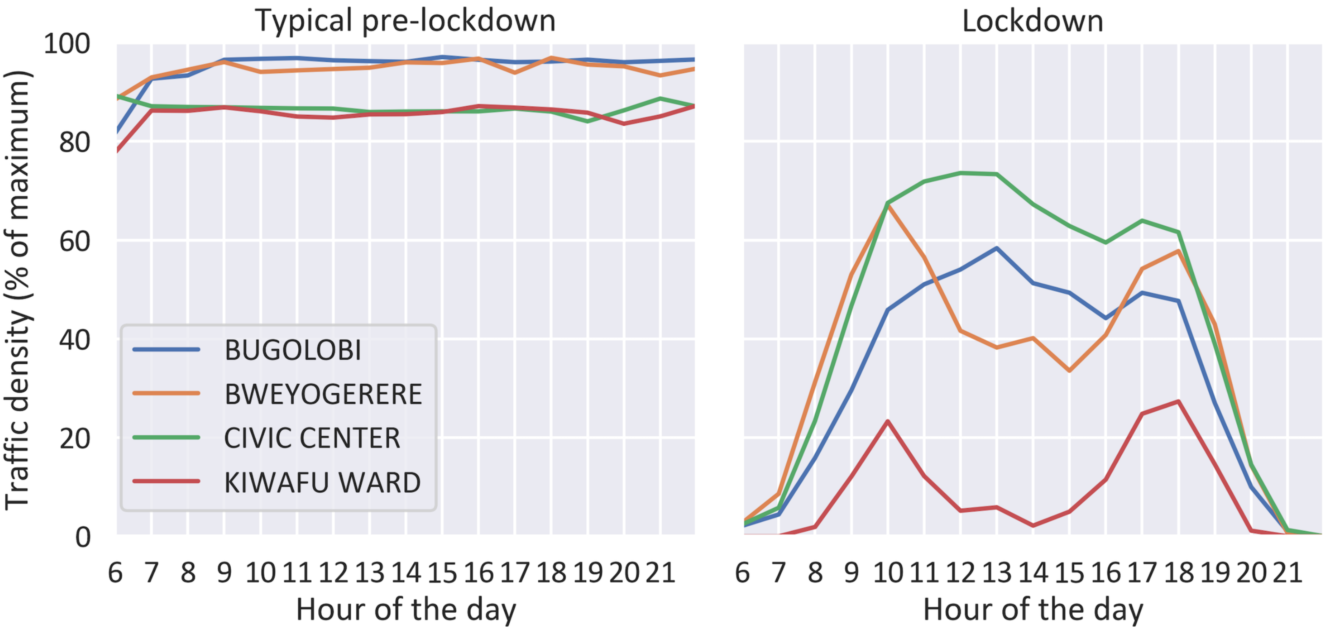

Figure 13: *Diurnal variation in traffic activity within 2.2km X 2.2km bounding box of the monitoring device for the pre-and during lockdown period*

turbulent ambient conditions. Pollution is now dispersed over a much greater vertical height and exposure levels reduce, being at their lowest mid-afternoon. During the evening conditions, cooler air is compressed into a smaller vertical space leading to higher concentrations during the night. These diurnal patterns are not unique to this study and have been replicated in other geographic settings (Chow et al. 1999; DeGaetano and Doherty 2004; Chen, Tang, and Zhao 2015), with levels dependent on location-specific activity patterns.

During the 2020 lockdown, mean $PM_{2.5}$ levels are consistently below the 2019 equivalent period. The morning and evening peaks are levelled out by reduced activity. We can see that while pre-lockdown levels (averagely) are always at or above new WHO daily AQG levels of 15µg/m^3 (World Health Organisation 2021), lockdown levels only exceed the threshold if only peak times are considered.

Investigating the difference further, Figure 12 shows the variation in mean hourly PM25 values during the lockdown period compared with the normalised values for the equivalent dates in 2019. All locations show a variation of greater than or equal to one standard deviation below equivalent period in the prior year. Similarly, all locations show a modest peak around 7 pm when levels are closest to prior year mean. Greater reductions are seen after 7 pm and continuing through until 3-5 am. This is in line with a substantial decrease in activity during the hours of curfew (7 pm-6:30 am). There is also variation between Civic Centre and Bugolobi, and Bweyogerere and Kiwafu with the latter pair seeing much greater reductions during the day and especially during night time. This difference can be explained by reduction in evening activity to be explored below, but also reinforced by the BLE which usually traps evening particulate matter close to the ground level, where it remains, reducing only slowly throughout the night. This is especially harmful for those living in open homes and in cities such as Kampala where polluting activity continues into the night.

As mentioned above we are assuming that cooking and waste burning activities remain the same or increase during the lockdown. Air quality improved at every stage of the day during lockdown and follows a similar pattern for all locations especially in the evening after curfew. It appears changes in domestic emissions are not a significant factor in explaining these observed patterns.

On traffic activity, the contrast between the typical and lockdown periods in figure 13 is striking. Typical levels show traffic activity at well above 80 percent of full activity at all times between 6 am and 10 pm. Under lockdown however there is very clear evidence that the curfew is being observed for all locations. All locations only begin to increase activity from 7 am. Civic Centre and Bugolobi locations continue to increase before falling steadily until 7 pm and then dropping sharply. For Bweyogerere and Kiwafu however, traffic activity drops away sharply at 10 am, remaining low through the day, before rising to a second peak at 6 pm and dropping away more sharply than others before curfew. This pattern of lower activity in Bweyogerere and Kiwafu is consistent with the lower $PM_{2.5}$ levels seen in Figure 12.

One possible explanation for why Kiwafu and Bweyogerere experience a greater drop after curfew is that being densely populated, lower income residential areas close to major roads they experience high levels of night-time combustion activities, typically charcoal and waste. Under lockdown scenario, traffic is reduced, but also polluting activities moves away from the main roads (and away from our sensors) possibly into homes. In the more affluent Bugolobi and the non-residential Civic Centre, only the traffic reduction which is already typically lighter in the evening is seen and limited outdoor combustion and reliance on street food much lower.

In summary, diurnal variation is largely explained by meteorological factors. Adjusting for this we observe that reduced traffic activity during the day leads to general reduction of 1 standard deviation across all regions, the greatest improvement comes at night for locations on major roads close to dense residential areas with heavy traffic and where outdoor combustion and street cooking is known to be prevalent. These findings indicate that policy initiatives that regulate transport activities would lead to immediate improvement on ambient air quality.

Conclusion and policy recommendations

This study explored the impact of COVID-19 lockdown restrictions on diverse air quality profiles for four distinctive locations in the Greater Kampala region of Uganda. We identify a reduction in $PM_{2.5}$ of between 17 and 50% compared with the same period in 2019 with the greatest increase coming in densely populated areas close to major roads. Investigation into diurnal variation reveals a broadly consistent improvement for all locations at all times. The greatest reductions occurred after the 7pm curfew and again, mostly in densely populated areas close to major roads. Implicitly, blanket policy interventions that target peak pollution periods could be proportionately adopted across the study area. Assuming that domestic fuel use is unlikely to have decreased, we identify traffic density, and street cooking and outdoor combustion as likely sources of the reduction. Policy initiatives in the transport sector such as vehicle emissions controls, traffic management, mass transit systems and pedestrianisation, in addition to regulation of outdoor open burning should be adopted to reduce the impact on air quality. Similarly, exploring the prospects of community driven initiatives could be essential to tackling diffuse and clustered pollution sources.

CRediT authorship contribution statement

Engineer Bainomugisha: supervision, conceptualization, review and editing. Paul Green: project administration, formal analysis, writing–original draft. Deo Okure: methodology, writing-original draft, writing-review and editing. Priscilla Adong: data curation, calibration, visualisation. Richard Sserunjogi: software, calibration, visualisation.

Declaration of competing interest

Authors declare no known competing financial and/or personal interests that could influence the findings of the paper.

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References

AirQo (2020). *AirQo, Africa's Air Quality Network*. Available at https: //www.airqo.africa (Accessed May 15, 2020).

Alsahli, Mohammad M and Meshari Al-Harbi (2018). "Allocating optimum sites for air quality monitoring stations using GIS suitability analysis". In: *Urban Climate* 24, pp. 875–886. doi: https://doi.org/10.1016/j.uclim.2017.11.001.

Atkeson, Andrew (2020). *What Will Be the Economic Impact of COVID-19 in the US? Rough Estimates of Disease Scenarios*. Working Paper 26867. National Bureau of Economic Research. doi: 10.3386/w26867. url: http://www.nber.org/papers/w26867.

Baldwin, Richard and Eiichi Tomiura (2020). "Thinking ahead about the trade impact of COVID-19". In: *Economics in the Time of COVID-19*. 59(2020).

Barkjohn, Karoline K, Brett Gantt, and Andrea L Clements (2021). "Development and application of a United States-wide correction for $PM_{2.5}$ data collected with the PurpleAir sensor". In: *Atmospheric Measurement Techniques* 14.6, pp. 4617–4637. doi: 10.5194/amt-14-4617-2021.

Brunekreef, Bert and Stephen T Holgate (2002). “Air pollution and health”. In: *The Lancet* 360.9341, pp. 1233–1242. doi: https://doi.org/10.1016/S0140-6736(02)11274-8.

Burnett, Richard T et al. (2014). “An integrated risk function for estimating the global burden of disease attributable to ambient fine particulate matter exposure”. In: *Environmental Health Perspectives* 122.4, pp. 397–403. doi: https://doi.org/10.1289/ehp.1307049.

Castell, Nuria et al. (2017). “Can commercial low-cost sensor platforms contribute to air quality monitoring and exposure estimates?” In: *Environment International* 99, pp. 293–302. doi: https://doi.org/10.1016/j.envint.2016.12.007.

Chen, Jiu-Chiuan and Joel Schwartz (2009). “Neurobehavioral effects of ambient air pollution on cognitive performance in US adults”. In: *Neurotoxicology* 30.2, pp. 231–239. doi: https://doi.org/10.1016/j.neuro.2008.12.011.

Chen, Wei, Hongzhao Tang, and Haimeng Zhao (2015). “Diurnal, weekly and monthly spatial variations of air pollutants and air quality of Beijing”. In: *Atmospheric Environment* 119, pp. 21–34. doi: 10.1016/j.atmosenv.2015.08.040.

Chow, Judith C. et al. (1999). “Temporal Variations of $PM_{2.5}$, PM_{10}, and Gaseous Precursors during the 1995 Integrated Monitoring Study in Central California”. In: *Journal of the Air & Waste Management Association* 49.9, pp. 16–24. doi: 10.1080/10473289.1999.10463909.

Cohen, Aaron J et al. (2017). “Estimates and 25-year trends of the global burden of disease attributable to ambient air pollution: an analysis of data from the Global Burden of Diseases Study 2015”. In: *The Lancet* 389.10082, pp. 1907–1918. doi: https://doi.org/10.1016/S0140-6736(17)30505-6.

Coker, Eric S et al. (2021). “A Land Use Regression Model using Machine Learning and Locally Developed Low Cost Particulate Matter Sensors in Uganda”. In: *Environmental Research*, p. 111352. doi: https://doi.org/10.1016/j.envres.2021.111352.

Cole, Matthew and Eric Neumayer (2004). “Examining the Impact of Demographic Factors On Air Pollution”. In: *Population and Environment*. doi: 10.1023/B:POEN.0000039950.85422.eb.

Dantas, Guilherme et al. (2020). “The impact of COVID-19 partial lockdown on the air quality of the city of Rio de Janeiro, Brazil”. In: *Science of The Total Environment* 729, p. 139085. issn: 0048-9697. doi: https://doi.org/10.1016/j.scitotenv.2020.139085.

DeGaetano, Arthur T. and Owen M. Doherty (2004). “Temporal, spatial and meteorological variations in hourly $PM_{2.5}$ concentration extremes in New York City”. In: *Atmospheric Environment* 38.11, pp. 1547–1558. doi: 10.1016/j.atmosenv.2003.12.020.

Ding, GA et al. (2005). “Vertical structures of PM_{10} and $PM_{2.5}$ and their dynamical character in low atmosphere in Beijing urban areas”. In: *Science in China*, Series D: Earth Sciences 48, pp. 38–54. doi: 10.1360/05yd0031.

EPA (2020a). *Aclima Project*. Available at https://www.epa.gov/air research/aclima-fact-sheet (Accessed on April 07, 2020)—(2020b). Village Green Project. Available at https://www.epa.gov/air research/village-green project (Accessed on May 14, 2020).

Harrison, RM and AR Deacon (1998). “Spatial correlation of automatic air quality monitoring at urban background sites: implications for network design”. In: *Environmental Technology* 19.2, pp. 121–132. doi: https://doi.org/10.1080/09593331908616664.

He, Guojun, Yuhang Pan, and Takanao Tanaka (2020). “The short-term impacts of COVID-19 lockdown on urban air pollution in China”. In: *Nature Sustainability* 3.12, pp. 1005–1011. doi: https://doi.org/10.1038/s41893-020-0581-y.

Ju, Min Jae, Jaehyun Oh, and Yoon-Hyeong Choi (2021). “Changes in air pollution levels after COVID-19 outbreak in Korea”. In: *Science of the Total Environment* 750, p. 141521.

Kabir, Mahvish et al. (2020). “COVID-19 pandemic and economic cost; impact on forcibly displaced people”. In: *Travel Medicine and Infectious Disease*, p. 101661. issn: 1477-8939. doi: https://doi.org/10.1016/j.tmaid.2020.101661.

Karagulian, Federico et al. (2015). “Contributions to cities’ ambient particulate matter (PM): A systematic review of local source contributions at global level”. In: *Atmospheric Environment* 120, pp. 475–483. doi: https: //doi.org/10.1016/j.atmosenv.2015.08.087.

Kelly, KE et al. (2017). “Ambient and laboratory evaluation of a low-cost particulate matter sensor”. In: *Environmental Pollution* 221, pp. 491–500. doi: https://doi.org/10.1016/j.envpol.2016.12.039.

Khuzestani, Reza Bashiri et al. (2017). “Quantification of the sources of long-range transport of $PM_{2.5}$ pollution in the Ordos region, Inner Mongolia, China”. In: *Environmental Pollution* 229, pp. 1019–1031. doi: https: //doi.org/10.1016/j.envpol.2017.07.093.

Lall, Somik Vinay, Henderson, and J. Vernon (2017). *Africa’s Cities: Opening Doors to the World*. http://hdl.handle.net/10986/25896. Washington, DC: World Bank. isbn: 978-1-4648-1045-9. doi: 10.1596/978-1-4648-1044-2.

Levy Zamora, Misti et al. (2018). “Field and laboratory evaluations of the low-cost plantower particulate matter sensor”. In: *Environmental Science & Technology* 53.2, pp. 838–849. doi: https://doi.org/10.1021/acs.est.8b05174.

Liousse, Catherine et al. (2014). "Explosive growth in African combustion emissions from 2005 to 2030". In: *Environmental Research Letters* 9.3, p. 035003. doi: https://doi.org/10.1088/1748-9326/9/3/035003.

Liu, Xiaoting et al. (2020). "Low-cost sensors as an alternative for long-term air quality monitoring". In: *Environmental Research* 185, p. 109438. doi: https://doi.org/10.1016/j.envres.2020.109438.

Mage, David et al. (1996). "Urban air pollution in megacities of the world". In: *Atmospheric Environment* 30.5, pp. 681–686. doi: https://doi.org/10.1016/1352-2310(95)00219-7.

Mahato, Susanta, Swades Pal, and Krishna Gopal Ghosh (2020). "Effect of lockdown amid COVID-19 pandemic on air quality of the megacity Delhi, India". In: *Science of The Total Environment* 730, p. 139086. issn: 0048-9697. doi: https://doi.org/10.1016/j.scitotenv.2020.139086.

Marais, Eloise A and Christine Wiedinmyer (2016). "Air quality impact of diffuse and inefficient combustion emissions in Africa (DICE-Africa)". In: *Environmental Science & Technology* 50.19, pp. 10739–10745. doi: https://doi.org/10.1021/acs.est.6b02602.

McFarlane, Celeste et al. (2021). "First Measurements of Ambient $PM_{2.5}$ in Kinshasa, Democratic Republic of Congo and Brazzaville, Republic of Congo Using Field-calibrated Low-cost Sensors". In: *Aerosol and Air Quality Research* 21, pp. 200619–200619. doi: https://doi.org/10.4209/aaqr.200619.

Morawska, Lidia et al. (2018). "Applications of low-cost sensing technologies for air quality monitoring and exposure assessment: How far have they gone?" In: *Environment International* 116, pp. 286–299. doi: https://doi.org/10.1016/j.envint.2018.04.018.

Mostafa, Mohamed K, Gamil Gamal, and A Wafiq (2021). "The impact of COVID 19 on air pollution levels and other environmental indicators-A case study of Egypt". In: *Journal of Environmental Management* 277, p.111496. doi: https://doi.org/10.1016/j.jenvman.2020.111496.

Mukherjee, Anondo et al. (2019). "Measuring spatial and temporal $PM_{2.5}$ variations in Sacramento, California, communities using a network of low-cost sensors". In: *Sensors* 19.21, p. 4701. doi: https://doi.org/10.3390/s19214701.

Nicola, Maria et al. (2020). "The socio-economic implications of the coronavirus pandemic (COVID-19): A review". In: *International Journal of Surgery* 78, pp. 185–193. issn: 1743-9191. doi: https://doi.org/10.1016/j.ijsu.2020.04.018.

Okure, Deo et al. (2022). "Characterization of Ambient Air Quality in Selected Urban Areas in Uganda Using Low-Cost Sensing and Measurement Technologies". In: *Environmental Science & Technology*. doi: https://doi.org/10.1021/acs.est.1c01443.

Petkova, Elisaveta P et al. (2013). "Particulate matter pollution in African cities". In: *Air Quality, Atmosphere & Health* 6.3, pp. 603–614. doi: https://doi.org/10.1007/s11869-013-0199-6.

Pfotenhauer, David J et al. (2019). "Updated Emission Factors from Diffuse Combustion Sources in Sub-Saharan Africa and Their Effect on Regional Emission Estimates". In: *Environmental Science & Technology* 53.11, pp. 6392–6401. doi: https://doi.org/10.1021/acs.est.8b06155.

Pope III, C Arden and Douglas W Dockery (2006). "Health Effects of Fine Particulate Air Pollution: Lines that Connect". In: *Journal of the Air & Waste Management Association* 56.6, pp. 709–742. doi: 10.1080/10473289.2006.10464485.

Richmond, Amy, Ian Myers, and Hafisa Namuli (2018). "Urban informality and vulnerability: A case study in Kampala, Uganda". In: *Urban Science* 2.1, p. 22. doi: https://doi.org/10.3390/urbansci2010022.

Roy, Rana (2016). "The cost of air pollution in Africa". In: *OECD Publishing*, Paris 333. doi: https://doi.org/10.1787/5jlqzq77x6f8-en.

Sanita di Toppi, Luigi, Lorenzo Sanita di Toppi, and Erika Bellini (2020). "Novel Coronavirus: How Atmospheric Particulate Affects Our Environ577 ment and Health". In: *Challenges* 11.1. issn: 2078-1547. doi: 10.3390/ challe11010006. url: https://www.mdpi.com/2078-1547/11/1/6.

Seaton, Anthony et al. (1995). "Particulate air pollution and acute health effects". In: *The Lancet* 345.8943, pp. 176–178.

Setti, Leonardo, Fabrizio Passarini, Gianluigi De Gennaro, Pierluigi Barbieri, et al. (2020). "The Potential role of Particulate Matter in the Spreading of COVID-19 in Northern Italy: First Evidence-based Research Hypotheses". In: *medRxiv*. doi: 10.1101/2020.04.11.20061713.

Setti, Leonardo, Fabrizio Passarini, Gianluigi De Gennaro, Pierluigi Baribieri, et al. (2020). "SARS-Cov-2 RNA Found on Particulate Matter ofcBergamo in Northern Italy: First Preliminary Evidence". In: *medRxiv*. doi: 10.1101/2020.04.15.20065995.

Spangl, Wolfgang et al. (2007). *Representativeness and classification of air quality monitoring stations*. Umweltbundesamt. 2007. Available at: https://www.umweltbundesamt.at/fileadmin/site/publikationen/rep0121.pdf

Tobías, Aurelio et al. (2020). "Changes in air quality during the lockdown in Barcelona (Spain) one month into the SARS-CoV-2 epidemic". In: *Science of The Total Environmen*t 726, p. 138540. issn: 0048-9697. doi: https://doi.org/10.1016/j.scitotenv.2020.138540.

Travaglio, Marco et al. (2020). "Links between air pollution and COVID-19 in England". In: *Environmental Pollution* 268, p. 115859. doi: doi.org/10.1016/j.envpol.2020.115859.

Uganda Bureau of Statistics (2016). *The National Population and Housing Census 2014 Main Report*.

Uganda Bureau of Statistics (UBOS) (2011). *Report on the Census of Business Establishments 2010/2011*.

United Nations (2018). World Urbanization Prospects: The 2018 Revision, *Online Edition*. Available at https://population.un.org/wup/Download/ (Accessed on May 10, 2020).

Venter, Zander S et al. (2020). "COVID-19 lockdowns cause global air pollution declines". In: *Proceedings of the National Academy of Sciences* 117.32, pp. 18984–18990.

Vermeiren, Karolien et al. (2012). "Urban growth of Kampala, Uganda: Pattern analysis and scenario development". In: *Landscape and Urban Planning* 106.2, pp. 199–206.

Watson, John G. and Judith C. Chow (2002). "A wintertime $PM_{2.5}$ episode at the Fresno, CA, supersite". In: *Atmospheric Environment* 36.3, pp. 465–475. doi: 10.1016/s1352-2310(01)00309-0.

World Bank (2018). *Coordinating Investments Across Greater Kampala Would Boost Growth for Uganda-World Bank Report*. Available at https://www.worldbank.org/en/news/press-release/2018/03/09/coordinating-investments-across-greater-kampala-wouldboost-growth-for-uganda-world-bank-report. (Accessed on May 10, 2020).

World Health Organisation (2016). *Air pollution levels rising in many of the world's poorest cities*. Available at https://www.who.int/news-room/detail/12-05-2016-air-pollution-levels-rising-in-many-of-the-world-s-poorest-cities (Accessed on May 28, 2020).

World Health Organisation (2018). "*9 out of 10 people worldwide breathe polluted air, but more countries are taking action, 2018*". In: Available at http://www.who.int/news-room/detail/02-05-2018-9-out-of-10-people-worldwide-breathe-polluted-air-but-more-countries-are-taking-action.

World Health Organisation (2021). *WHO global air quality guidelines: particulate matter ($PM_{2.5}$ and PM_{10}), ozone, nitrogen dioxide, sulfur dioxide and carbon monoxide*. World Health Organisation, xxi, 267

Wu, Xiao et al. (2020). "Exposure to air pollution and COVID-19 mortality in the United States: A nationwide cross-sectional study". In: *medRxiv*. doi: 10.1101/2020.04.05.20054502.

Xue, Tao et al. (2019). "Declines in mental health associated with air pollution and temperature variability in China". In: *Nature Communications* 10.1, pp.1–8.

Yan, Shujun et al. (2016). "Spatial and temporal characteristics of air quality and air pollutants in 2013 in Beijing". In: *Environmental Science and Pollution Research* 23.14, pp. 13996–14007. doi: 10.1007/s11356-016-6518-3.


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Research article

Air pollution abatement by selective nanoparticle deposition on filtration systems

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Abstract

Air pollution kills an estimated seven million people worldwide every year. The data from the World Health Organization (WHO) shows that almost all the global population (99%) breathe air that exceeds WHO guideline limits. The growing population and urbanization such as in Africa, which has the fastest growing population, may lead to substantial worsening of the air quality. Urbanization is also a powerful driver of the epidemiologic transition from traditional threats like infectious diseases and malnutrition to chronic, non-communicable diseases. Particulate matter less than 2.5 microns in size, $PM_{2.5}$, is the leading contributor to air pollution which results in such diseases like chronic obstructive pulmonary disease (COPD), bronchitis, and lung cancer. Recent studies have shown a strong correlation between ambient air pollution and COVID-19 cases, which has affected the lives of billions of people around the world. Abatement technologies such as ionic and other high efficiency filtration systems are quite expensive and hence unaffordable to communities with limited resources. The goal of this study was to develop an air pollution filtration method utilizing selective nanoparticle deposition in optimized concentrations, to maximize the entrapment of $PM_{2.5}$ particles. The experimental set-up consisted of a wind tunnel with incense sticks as the $PM_{2.5}$ source, measured by laser particle detectors upstream and downstream of the filters. Different nanoparticle coated filters were tested using the 'Design of Experiments' methodology and it was concluded that an optimized mixture of zinc oxide, titanium dioxide & graphene improved filtration efficiency of a baseline filter by 206% and was 70% cheaper than high efficiency filters. The versatility and cost-effectiveness of this design makes it applicable for personal masks & filters, air-conditioning and car-cabin filters, and fire-fighting equipment. The significant correlation between air pollution and fatalities from viral infections like COVID-19, makes such abatement technologies with innovative filtration systems critical to save human lives.

Keywords

Air Pollution, Filtration, Nanoparticle, Particulate Matter, $PM_{2.5}$

Introduction

Air pollution is the contamination of the air in the atmosphere by a physical, biological or chemical alteration. Particulate matter (PM) is one of the main contributors to air pollution, consisting of a mixture of solid and liquid particles suspended in the air. The most used health indicator related to PM refers to the mass concentration of particles with a diameter less than 2.5µm, $PM_{2.5}$ (Nolt-Helms et. al. 2018; Francesca et. al. 2006). Primary sources of $PM_{2.5}$ include anthropogenic sources such as automobile emissions, household fuel (Matawle et. al. 2017) and waste burning, energy production from fossil fuels (Kundu and Stone, 2014) and biomass, and industrial activities such as construction, mining, cement production, etc. 50% of the total PM emissions in urban areas is generated from traffic (Wrobel et. al. 2000). The most polluted areas around the world tend to be in developing countries in Africa, South-East Asia, India and China, due to their increased density of urban population, significant use of fossil fuels and relatively inadequate control measures and filtration systems (World Health Organization, WHO, 2022).

Africa has the fastest growing population in the world, predicted to surpass two billion by 2050 (UN 2011). Half of Africa's population is expected to live in urban areas by 2035, and Sub-Saharan Africa will host five of the world's 41 megacities by 2030 (Katoto et. al. 2019). Urbanization and increased industrialization, growing ownership of automobiles, and continued use of biomass as domestic energy source may lead to substantial worsening of air quality across the continent (Petkova et. al. 2013). Urbanization is also a powerful driver of the global demographic and epidemiologic transition, characterized by declining birth rates, increasing life expectancy, and a shift from traditional threats such as infectious diseases and malnutrition to chronic, non-communicable diseases like heart disease and diabetes (Omran 1971).

Almost all (99%) of the world's population lives in places exceeding WHO's air quality guidelines and 7 million people die every year because of air pollution (World Health Organization, WHO, 2022). The primary causes of such premature deaths are chronic obstructive pulmonary disease, heart disease, stroke, lung cancer and acute respiratory infections in children. $PM_{2.5}$, due to its small size, is capable of penetrating deep into lung passageways and entering the bloodstream causing cardio-vascular, cerebro-vascular, and respiratory impacts (Vijayan et. al. 2015; Xing et. al. 2016). Furthermore, long term exposure to air pollution has been found to increase the vulnerability to the most severe impacts of coronavirus outbreaks such as SARS in 2003 and COVID-19 in 2020 (Coker et. al. 2020; Comunian et. al. 2020; Zoran et. al. 2020; Wu et. al. 2020). An increase of only 1 μg/m^3 in $PM_{2.5}$ is associated with an 8% increase in the COVID-19 death rate in the United States (Wu et. al. 2020).

Indirect effects of ambient air pollution include acid rain caused by excessive amounts of sulphur oxides in the air. This can impact aquatic ecosystems, as well as man-made landscapes (Bhargava and Bhargava 2013; Banerjee and Sarkar 2019). Photochemical smog caused by the combination of nitrogen oxides and $PM_{2.5}$ can harm the ozone layer which in turn results in global warming and its associated social, economic, and geopolitical consequences (Davidson 2015).

Abatement technologies such as ionic and High Efficiency Particulate Air (HEPA) air filtration systems (Abatement Technologies, 2018) have been developed that can filter $PM_{2.5}$ particles significantly but remain quite expensive and hence unaffordable to communities with limited resources (Vyas et. al. 2016; Brook 2019). Therefore, a cost-effective and efficient abatement system is essential in helping to resolve the issue.

Nanoparticles have a high surface to volume ratio, which enhances the entrapment of particulate matter by adsorption. The surface adsorption energy is unique to the small size of nanoparticles with extremely high surface to volume ratios, where the unsaturated surface chemical bonds tend to adsorb other chemicals or biomolecules to reduce their surface energy (Xia et. al. 2011). The three nanoparticles used for this study were graphene, titanium dioxide (TiO_2), and zinc oxide (ZnO), which have been known to have filtration properties due to their high adsorption capabilities (Zhong et. al. 2015; Wongwatcharapaiboon et. al. 2019; Ruan et. al. 2020). Graphene, an allotrope of carbon consisting of a single layer of carbon atoms arranged in a hexagonal lattice structure, has high adsorption capacities mainly due to these unique nanostructures, and hence has been proven to be efficient in the capture of particulate matter (Szczęśniak 2017; Zhang et. al. 2018). TiO_2 nanoparticles, with their photocatalytic properties, absorb the ultraviolet component of sunlight and act as a catalyst to form reactive hydroxyl (•OH) radicals and the superoxide anion (O_2•-) from atmospheric moisture and oxygen. These radicals react with the $PM_{2.5}$ particles due to their strong oxidizing capabilities converting them into CO_2 and H_2O (Giovanetti et. al. 2017), hence have been used for air purification (Thanh Son Le et. al. 2015).

The current work is aimed to develop an efficient and cost-effective air-filtration system by the deposition of nanoparticles on a readily available filtration media. The nanoparticles are selected based on their air filtration capabilities, clinical safety, and non-toxicity. The filtration system needs to be versatile and effective at different pollution levels in different parts of the world and from different pollutant sources, as described above. The goal of this work is to also develop a simple application technique of the nanoparticles such that it can be easily applied to various filtration systems in different parts of the world, thus providing an affordable alternative to expensive high quality air filtration devices with comparable air filtration capabilities.

Materials and methods

Experimental set-up

A wind tunnel was designed and created to test the efficiency of the filters (Fig. 1), as a continuation of a previous study (Nag, 2021-22). A cardboard box (142x50x50 cm) was used as the body of the tunnel. A washer-dryer exhaust tube (diameter 10 cm) was used to connect the PM source to the inlet section of the tunnel. Incense sticks were used as the source of $PM_{2.5}$ (Lui et. al. 2016; Jilla and Kura 2017). A fan (Lasko, 50x50 cm) was placed on the inside of the tunnel to blow the $PM_{2.5}$ through the filter, which was placed downstream of the fan. $PM_{2.5}$ detectors (Temtop LKC-1000S) were hung inside the inlet and outlet sections of the wind tunnel, and plexiglass windows were installed into the tunnel for visualization of the detector readings. A manometer (PerfectPrime AR1890P2) was used to measure pressure drop, with the tubes placed upstream and downstream of the fan and filter; and a lamp (Hyperikon, 15W, 5000K) was used to simulate the effect of daylight to enhance the photocatalytic properties of the TiO_2-coated filters.

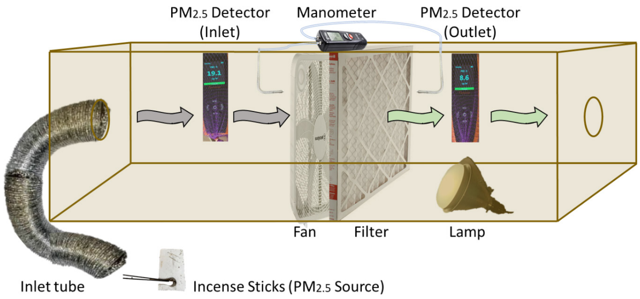


Figure 1: *A wind tunnel with a fan to create the draft, and incense sticks to simulate the $PM_{2.5}$, were used in this experimental set-up. Laser particle detectors at the inlet and outlet were used to measure the filtration efficiency and a manometer was used to measure the pressure drop.*

Nanoparticle deposition

The nanoparticles used for this study were Titanium Oxide (TiO_2 Anatase, 99.5% 40nm, US Research Nanomaterials, Inc.), Zinc Oxide (ZnO, 99+%, 35-45 nm, US Research Nanomaterials, Inc.) and Graphene (Alfa Aesar™ Graphene nanoplatelets aggregates, 500 m^2/g, Fisher-Scientific).

The combination of nanoparticles was mixed with ethanol (200 Proof (100%), USP/EP/ACS, Fisher-Scientific) to create a suspension which appears as a slurry. This suspension was then aerosolized using the pressurized sprayer system (Preval Airless Paint Sprayer, 70 psi, Home Depot), and the aerosolized spray was directed towards a typical Heating, Ventilation and Air Conditioning (HVAC) air filter (Rheem, 51x51x2.5 cm, wire-backed pleated fiberglass and paper media) with a Minimum Efficiency Reporting Value (MERV) rating of 8 (ASHRAE, 2020) while maintaining a spray-distance of about 15-18 cm. The filters were then air-dried for at least 8 hours and then tested for efficacy. A 'high quality' air filter (Rheem, 51x51x2.5 cm, MERV-14, Home Depot) was used for comparison and benchmarking the filtration efficiency.

Different combinations (Fig 4.) and concentrations (Fig. 7) of nanoparticles were experimented with, to determine the optimized mixture to maximize filtration efficiency. The nanoparticle loading was calculated at 2.33E-04 g/cm^2 for 2% coating with one nanoparticle, and at 4.65E-04 g/cm^2 for 2% coating with two nanoparticles. The maximum loading was 2.79E-03 g/cm^2 for the 8% coating with all three nanoparticles.

Spatial uniformity of nanoparticle deposition

The deposition method of nanoparticles onto the filtration media were also varied and tested for uniform spatial distribution as in the previous study by the same author (Nag, 2021-22). Different spray mechanisms were tested using pipettes, spray bottles and pressurized sprayers. Different zones of the air filter, in 9 locations, were tested for spatial variation in filtration efficiency (Fig. 2). The pressurized spray application resulted in the most uniform spatial distribution of the nanoparticles and was chosen as the preferred application method for its simplicity and effectiveness.

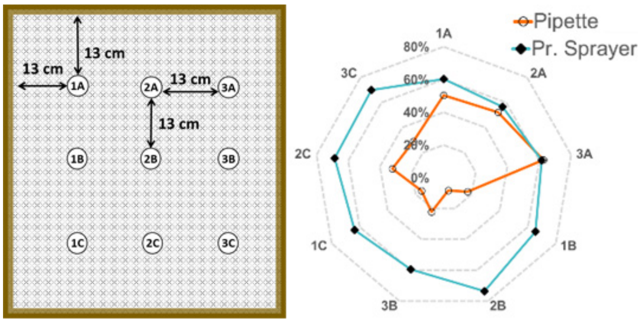


Figure 2: Spatial variation test to verify and optimize nanoparticle deposition uniformity. The spider-chart depicts the uniformity of the pressurized sprayer over the pipette deposition.

Surface morphology of coated filters

The surface morphology of the filters was characterized using the scanning electron microscope (SEM) imaging technique and confirmed the adhesion of the nanoparticles. A Zeiss Ultra-55 SEM with a Schottky field emission source and resolution of 1 nm @ 15 KV, 1.7 nm @ 1 KV was used. The images below are at 100X magnification. These SEM images (Fig. 3a) of the filters enabled the confirmation of the nanoparticle adhesion to the filters in the coated filters as compared to the baseline (uncoated). The

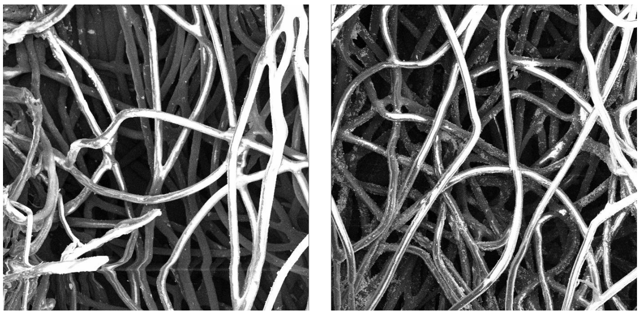


Figure 3a: *Scanning Electron Microscope (SEM) images of the baseline filter (left) and the TiO_2 coated one (right) confirm the adhesion of the nanoparticles to the fibres of the filtration media.*

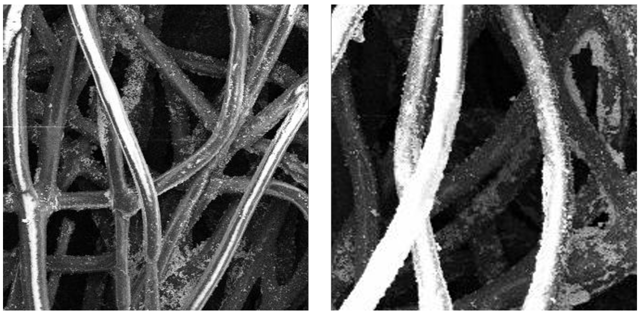
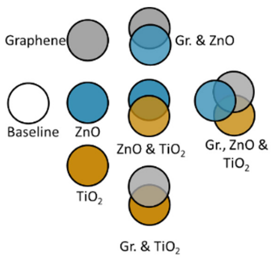


Figure 3b: *Scanning Electron Microscope (SEM) images of the TiO_2 coated filter before (left) and after (right) exposure to $PM_{2.5}$ particles, show the adsorption of the $PM_{2.5}$ particles to the nanoparticle adhesion areas of the filtration media.*

entrapment of particulate matter onto the nanoparticle surfaces in the 'after' images of the different nanoparticle coated filters are visualized in the SEM images (Fig. 3b), as previously reported (Nag, 2022). The embedded nanoparticles enhance the diffusion and electrostatic attraction mechanisms of filtration due to high surface to volume ratio and photocatalytic activation properties (N95 Respirators and Surgical Masks, CDC, 2009).

Design of experiments

A Design of Experiments (DOE) statistical analysis model helps with analysing and interpreting test results and the effects of multiple input variables and their interactions on an output variable (Minitab, 2020). A full factorial DOE design was used to collect and analyse the data to randomize the run order of the experiment, minimize bias and aid with the Analysis of Variance (ANOVA) study. Combinations of single nanoparticles (Graphene or ZnO or TiO_2), combination of two nanoparticles and finally all



StdOrder	RunOrder	Gr	ZnO	TiO2
2	1	1	-1	-1
3	2	-1	1	-1
8	3	1	1	1
1	4	-1	-1	-1
7	5	-1	1	1
4	6	1	1	-1
5	7	-1	-1	1
6	8	1	-1	1

-1: Absence, 1: Presence of nanoparticle

Figure 4: *Design of Experiments set-up using full-factorial design for ANOVA analysis. The table indicates the run order used to randomize the experimentation to avoid bias. The figure on the right illustrates the different combinations of the nanomaterials used for the experiment.*

three together were experimented in a random order, as shown in Fig. 4.

The $PM_{2.5}$ at the inlet was also varied significantly to test the versatility of the filters over different environmental conditions. The lower end of the inlet $PM_{2.5}$ at 80~100 µg/m^3 represents pollution levels in typical polluted cities in India and China. The mid-level at 150~200 µg/m^3 represents some polluted cities in Egypt and Niger, while the higher end at 250~300 µg/m^3 represents forest fires, heavy vehicle exhausts and smoke.

Validation of experimental results

To validate the results from this experiment, commercially available 'high quality' filters were tested in this set-up and compared to previously performed studies. HVAC filters rated with a Minimum Efficiency Reporting Value (MERV) of 14 (ASHRAE, 2020) were tested using this wind tunnel experimental set-up over several days as shown in Fig. 5 and were found to be consistent and comparable to a previous study (Zhao et. al. 2015) using MERV 14 (equivalent to 'Filter Performance Rating' FPR 10) type filters, thus confirming the accuracy of the experimental set-up.

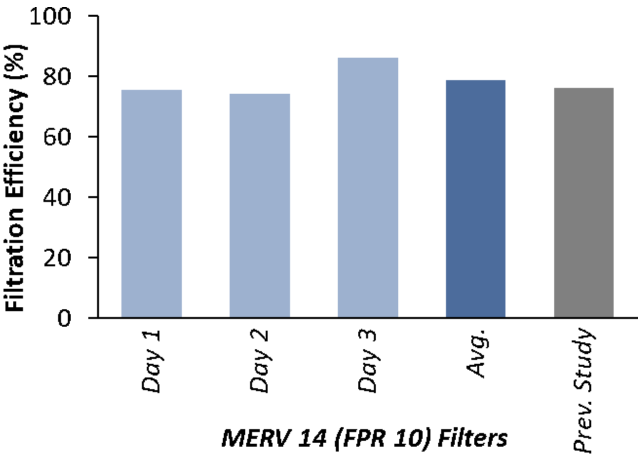


Figure 5: *Commercially available MERV14 filters' filtration efficiency was maintained over different days of testing and was found to be consistent to a previous study, thus confirming the validity of the experimental set-up.*

Measurement uncertainty

A statistical repeatability and reproducibility study (Gage R&R) was used using Minitab software (Minitab, 2020), to determine the measurement uncertainty of the experiment. The tests were repeated for 10 trials each and repeated over different days to measure repeatability and reproducibility. It was seen (Fig. 6) that 95% of the contribution was from 'part-to-part variation' (natural variability due to different parts) and 5% from the measurement process variation (variability due to measurement system uncertainties), which is deemed to be an acceptable measurement uncertainty, per statistical guidelines (Automotive Industry Action Group (AIAG), 2010). The reproducibility factor of this experiment (experiments conducted over different days) is very high since the uncertainty from the reproducibility is almost negligible.

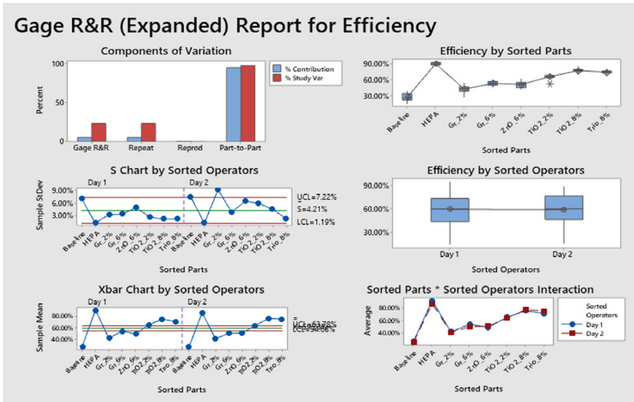


Figure 6: *The Gage Repeatability and Reproducibility analysis of the experimental set-up indicating acceptable levels (5% of total variation) of measurement uncertainty.*

Other sources of uncertainty

Sources of the remaining measurement uncertainty may include the calibration of the $PM_{2.5}$ detectors which may affect their absolute measurements, which can be improved by performing calibrations with mixtures of known $PM_{2.5}$ concentrations. The generation of $PM_{2.5}$ particles from incense sticks was inconsistent and may have also contributed to the measurement system uncertainty of the experiment. The sealing system (using insulation sealing materials) between the inlet and outlet sections of the wind-tunnel may have been another source of uncertainty.

Results and discussion

As explained in section 2.2, different concentrations and combinations of nanoparticles were evaluated for maximizing filtration efficiency. The results showed (Fig. 7) that the concentration of nanoparticles has a direct correlation to the filtration efficiency (1). The correlation between nanoparticle type and filtration efficiency was also observed, with titanium dioxide coated filters demonstrating the highest efficiency. The mixture of nanoparticles was more effective in reducing $PM_{2.5}$ as compared to the individual nanoparticles.

$$Filtration\ Efficiency\ (\%) = \frac{PM_{2.5_{inlet}} - PM_{2.5_{outlet}}}{PM_{2.5_{inlet}}} \times 100 \quad (1)$$

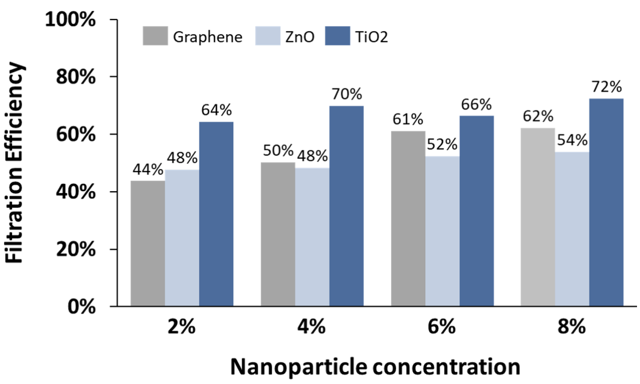


Figure 7: *Increase in nanoparticle concentration improves filtration efficiency. TiO_2 has the overall highest filtration efficiency, while Graphene demonstrates the strongest correlation in filtration efficiency and concentration.*

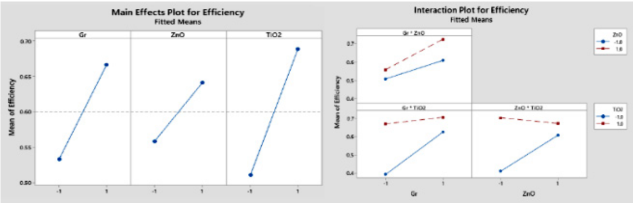


Figure 8: *Main and Interaction effect plots for nanoparticle coated filters indicating strong interaction effects of TiO_2 with Graphene and ZnO.*

The interaction plots from the ANOVA analysis (Fig. 8) indicate that although the main effects of the individual nanoparticles were significant, there was strong interaction between Graphene and TiO_2 and ZnO and TiO_2. However, the interaction between Graphene and ZnO was not as significant. Hence, it can be ascertained that TiO_2 has a key interaction effect and improves the efficiency of the other nanoparticles (ZnO and Graphene) in its presence.

The titanium dioxide coated filters had a 7% increase in filtration efficiency when placed under light, due to the activation of its photocatalytic properties. TiO_2 nanoparticles (NP), with their photocatalytic properties, absorb the ultraviolet component of sunlight which excites the electrons (e-) from Valence Band (VB) to Conduction Band (CB) and act as a catalyst to form the superoxide anion (O_2•-) and reactive hydroxyl (OH•) radicals from atmospheric moisture and oxygen (2). These radicals react with the $PM_{2.5}$ particles due to their strong oxidizing capabilities converting them into CO_2 and H_2O (Giovanetti et al. 2017).

a) NP(e-CB)+O_2 → (O_2•-)+NP *[The photo generated (e-) reacts with adsorbed O_2 to form superoxide radical(O2•-)]*
b) (O_2•-)+H_2O → HO_2• +OH-
c) HO_2•+H_2O → OH•+H_2O_2 (2)
d) H_2O_2 → 2OH• *[The (O_2•-) in turn reacts with moisture(H_2O) to form (OH•) hydroxyl radical]*
e) OH•+ air pollutant → CO_2+ H_2O *[The (OH•) degrades pollutants to CO_2 and H_2O]*

Although this mechanism is applicable to organic sources of $PM_{2.5}$, this author has also further evaluated the consistency of the filtration efficiency from different sources of $PM_{2.5}$ (Nag, 2022). Most of the $PM_{2.5}$ generated is from organic compounds such as fossil fuels and wood-burning (WHO, 2020).

A two-sample t-test (Fig. 9) with 95% confidence intervals, indicates the marked improvement of filtration efficiency with the application of light (p-value <0.05).

A significant parameter that affects the filtration system's energy consumption is its pressure drop. Pressure drop can be calculated by using the Bernoulli equation (3):

$$P_1 + \frac{1}{2}\rho v_1^2 + \rho g h_1 = P_2 + \frac{1}{2}\rho v_2^2 + \rho g h_2 \quad (3)$$

where P is the pressure, ρ is the density of the fluid, v is the velocity, g is the gravitational acceleration, h is the height, subscript[1] denotes upstream conditions and subscript[2] is for downstream conditions. Under steady, incompressible, and

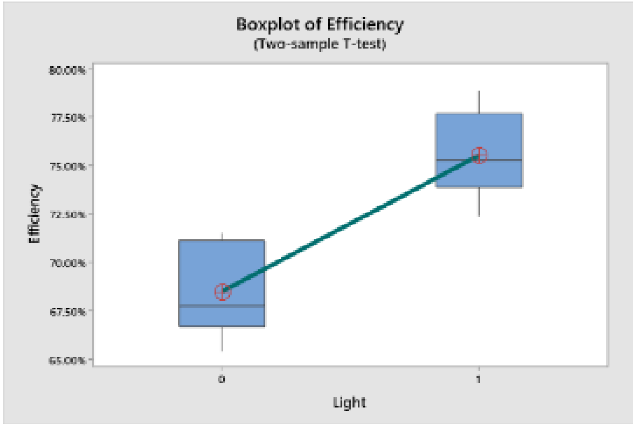


Figure 9: *A 2-sample t-test demonstrating impact of light on filtration efficiency of TiO_2.*

frictionless flow along a streamline assumption with the same horizontal height; (3) can be simplified to the pressure drop equation (4):

$$\Delta P = \frac{1}{2}\rho(v_1^2 - v_2^2) \quad (4)$$

A quantitative test of airflow was conducted by measuring the pressure drop (Δp) across the filter. As seen in Fig. 10, the pressure-drop (psi) was consistent across the baseline and the nanoparticle coated filters. This is because the size of these nanoparticles is considerably smaller (60-80 nm) than the size of the fibres in the filter (3-5 µm). This indicates that applying the nanoparticles to the air filters does not affect their energy consumption in a measurable way.

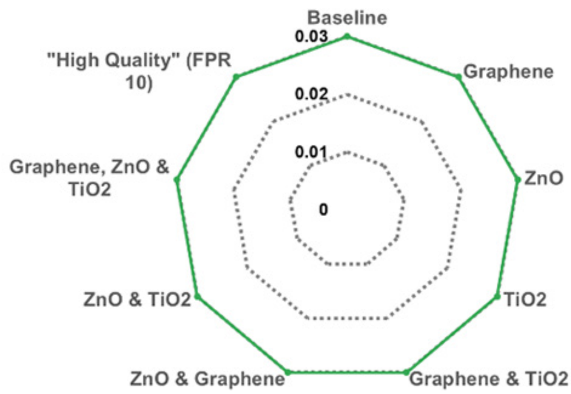


Figure 10: *Nanoparticle coated filters have no measurable impact on the pressure drop.*

An accelerated durability testing (5) was performed to test the effectiveness of the filters over longer usage periods. The durability testing was accelerated by increasing the volume flow rate of air by increasing the fan speed, and the mass flow rate of $PM_{2.5}$ was increased by increasing the incense stick count.

The samples were tested intermittently to monitor the decrease in filtration efficiency over time. It was seen that 90% of the filter's effectiveness was maintained after 50 equivalent operational days and 70% of its initial efficiency is maintained over 100 equivalent days of operation (Fig. 11).

$$\boldsymbol{H_{at}} = \frac{\mathrm{p \times a \times h \times FL}}{\mathrm{PM_i \times n \times a^{+}}} \qquad (5)$$

$p = \mathrm{PM}_{2.5}@polluted\ city = 150\left[\frac{\mu g}{m^3}\right]$ $\mathrm{PM_i} = PM_{2.5_{per\ incense\ stick}} = 300\left[\frac{\mu g}{m^3}\right]$

$\mathrm{a} = Air\ flow\ rate_{nominal} = 1\left[\frac{m}{s}\right]$ $\mathrm{n} = \#\ incense\ sticks = 20$

$\mathrm{h} = Hours_{Per\ day} = 24$ $\mathrm{a^{+}} = Air\ flow\ rate_{increased}$

$\mathrm{FL} = Filter\ Life = 90[\mathrm{days}]$ $\boldsymbol{H_{at}} = Hours_{accelerated\ \ test} = 18$

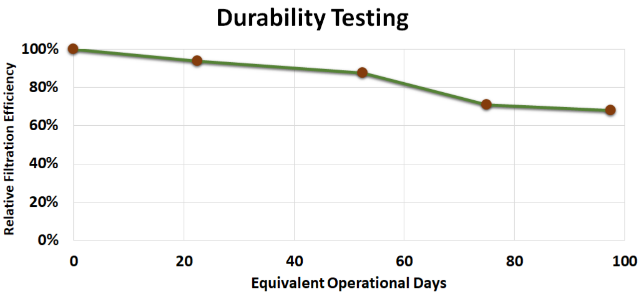


Figure 11: *Relative filtration efficiency maintained up to 90% of the initial efficiency during accelerated durability testing over 50 equivalent days.*

The nanoparticle coatings consistently demonstrated the ability to improve the filtration efficiency of a baseline filter (Fig. 12). The filters with single nanoparticle coatings (NP=1) with Graphene, ZnO or TiO_2, demonstrated an improvement of about 100% over the baseline uncoated filter. Coating the filters with two out of these three nanoparticles (NP=2) improved the efficiency further. However, the filter coated with a mixture of all three-nanoparticles (NP=3), had the highest filtration efficiency which was 3 times higher than the baseline filter. The filtration efficiency of this filter, at 77%, was quite comparable to the more expensive 'high-quality' MERV-14 filters. The filter coated with TiO_2 alone was also quite effective, but less versatile due to its dependence on light for activation of its photocatalytic properties.

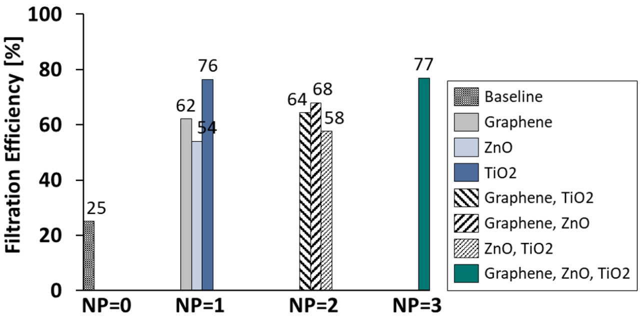


Figure 12: *Filtration efficiency of nanoparticle coated filters improved significantly relative to a baseline uncoated filter. The Graphene, ZnO, TiO_2 combination is 3 times more effective than a baseline uncoated filter.*

Cost-effectiveness was one of the main objectives of this experiment (Fig. 13) to make this technology available to societies with limited resources. Considering the baseline cost of a commercially available filter and the additional cost of nanoparticles and processing, the nanoparticle coated filters were 70% cheaper than the HEPA filters, and 99% cheaper than ionic filters. The cost assumptions behind this calculation are based on commercially available filters in a local hardware store. A regular air-conditioning filter with filter performance rating (FPR) 4 (equivalent to MERV 6/7) is considered as "baseline", priced at ~\$4. The nanoparticle coating content in each filter is approximately 2.5 g (8% concentration wt./wt.) at an average cost of ~\$400/kg thus a weighted total price of ~\$3 for the 3 nanoparticles. The processing cost of \$1 is assumed for consumables and labour. The FPR 10 (MERV equivalent 14) is the commercially available HEPA filter of the same size, priced at ~\$25 and an ionic filtration system is commercially available for ~\$500.

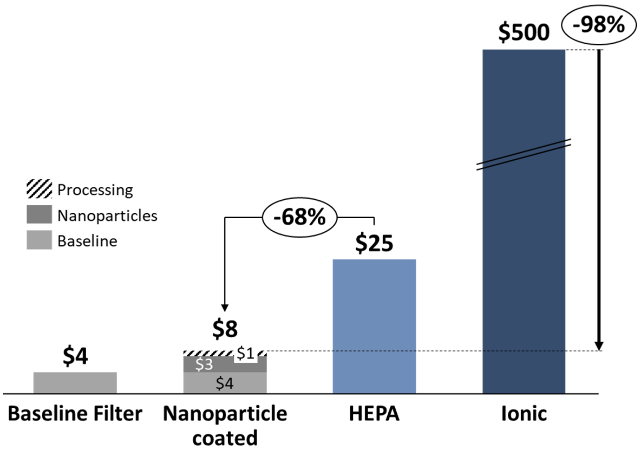


Figure 13: *Adding the costs of the nanoparticles and processing costs to a baseline filter, the coated filters are significantly less expensive than HEPA or ionic filtration systems.*

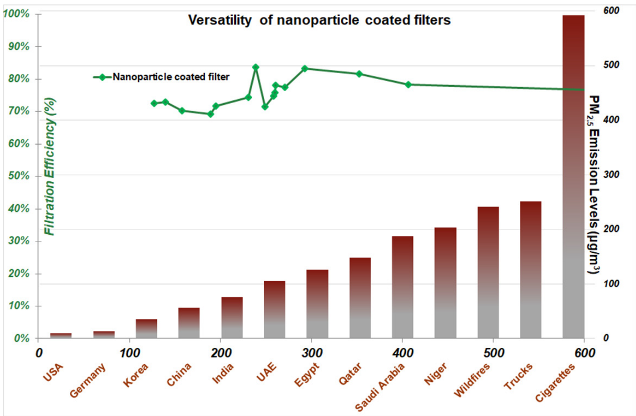


Figure 14: *Versatility of nanoparticle coated filters in different global pollution levels. The bar-chart indicates the different $PM_{2.5}$ levels in different countries and from different sources. The line graph indicates the filtration efficiency relative to the inlet $PM_{2.5}$ levels, indicating relatively consistent filtration efficiency over a wide range of $PM_{2.5}$ levels (100~600 µg/m³).*

The nanoparticle coated filters were also tested for their versatility at different inlet conditions and compared to the $PM_{2.5}$ pollution levels in different parts of the world and from different sources. In a further study, the filtration efficiency has been found to be consistent across different sources of $PM_{2.5}$, such as wood chips, paraffin and incense (Nag, 2022).

As illustrated in Fig. 14, the bar-chart indicates the $PM_{2.5}$ pollution levels found in different countries (2019 World Air Quality Report) like China, India, Niger etc. and from different pollutant sources (De Marco et. al. 2016) like wildfires, emissions from trucks etc. Typically, the $PM_{2.5}$ levels in countries like Niger are about 200 µg/m^3 or in India about 80 µg/m^3. The $PM_{2.5}$ levels from wildfires and emissions from trucks are in the range of 250-270 µg/m^3. The results of the filtration efficiency of the nanoparticle coated filters are plotted on the same graph relative to the inlet $PM_{2.5}$ conditions simulating the levels of pollution in different countries and from different sources. It can be seen in Fig. 14, that the filtration efficiency is relative consistent between 70-80% irrespective of the inlet levels of $PM_{2.5}$. This indicates that the nanoparticle coated filters are versatile in their usability – from different sources and levels of $PM_{2.5}$ pollution.

The safety of nanoparticle usage is of utmost importance and continues to be a subject of research worldwide. The nanoparticles chosen for this study are known for their clinical safety and non-toxicity. TiO_2, ZnO and Graphene are extensively used in commercial products like pill coatings, sunscreens, and biomedical applications (Shi et. al. 2015; Arvidsson 2018).

Conclusion

The results of this experiment can be used in several applications including personal protective equipment like air pollution masks, air-conditioning and car cabin filters, firefighting equipment, and industrial pollution control systems for power plants, incinerators, or automobiles. The versatility and effectiveness of this filtration system makes it applicable in different parts of the world with varying pollution levels. The simplicity of the application method and the cost-effectiveness make this process feasible to be used in low-income areas that have higher risks of air pollution and limited resources to combat their lethal effects. The nanoparticles selected for this application have been known for their non-toxicity and hence safe for human use. A significant correlation has been found between ambient air pollution and deaths caused by viral infections, such as COVID-19, which lead to respiratory diseases. This makes it essential for individuals to reduce their own pollution footprint, especially in rapidly urbanizing areas of the world, and adapt such novel, cost-effective and safe nanoparticle coated filtration technologies to reduce the deadly consequences of air pollution.

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References

"Abatement Technologies. Facts about true HEPA filtration", https://www.abatement.com/learning-center/patient-isolation/facts-about-hepa-filtration/, 2018.

2019 World Air Quality Report Region and City $PM_{2.5}$ Ranking. https://www.iqair.com/world-most-polluted-cities/world-air-quality-report-2019-en.pdf

Arvidsson, R., Boholm, M., & Montoya, L. D., "Just carbon: Ideas about graphene risks by graphene researchers and advisors", *Nanoethics*, 12(3), 199-210, 2018. DOI: 10.1007/s11569-018-0324-y
ASHRAE, American Society of Heating, Refrigerating and Air-Conditioning Engineers (ASHRAE), 2020. https://www.ashrae.org/technical-resources/filtration-and-disinfection-faq

Automotive Industry Action Group (AIAG), "Measurement systems analysis reference manual, 4th edition", *Chrysler, Ford, General Motors Supplier Quality Requirements Task Force*, 2010.

Banerjee, D., Sarkar, S., "Chemical and biochemical onslaught of anthropogenic airborne species on the heritage monument of the Taj Mahal", *Heritage*, 2(3), 2137–2159, 2019. https://doi.org/10.3390/heritage2030129

Bhargava, S., Bhargava, S., "Ecological Consequences of The Acid Rain", *IOSR Journal of Applied Chemistry (IOSR-JAC)*, e-ISSN: 2278-5736.5(4), PP 19-24, 2013. DOI:10.9790/5736-0541924

Brook, D., "What is an air ionizer? Are air ionizers good for you?", https://breathequality.com/ionizer/ (2019).

Coker, E. et. al., "The Effects of Air Pollution on COVID-19 Related Mortality in Northern Italy", *Environ Resour Econ (Dordr)*, 2020 Aug 4: 1–24. doi: 10.1007/s10640-020-00486-1

Comunian, S., et. al., "Air Pollution and COVID-19: The Role of Particulate Matter in the Spread and Increase of COVID-19's Morbidity and Mortality", *Int J Environ Res Public Health*, 2020 Jun; 17(12): 4487. doi: 10.3390/ijerph17124487

Davidson, A., "Photochemical oxidant air pollution: a historical perspective", *Studies in Environmental Science*, 72(1), 393-405, 1998.

De Marco, C., Ruprecht, A.A., Pozzi, P. et al. Particulate matters from diesel heavy duty trucks exhaust versus cigarettes emissions: a new educational antismoking instrument. *Multidiscip Respir Med* 11, 2 (2016). https://doi.org/10.1186/s40248-016-0042-7

Francesca Dominici, F., Peng, R., Bell, M., Pham, L., McDermott, A., Zeger, S., Samet, J., "Fine Particulate Air Pollution and Hospital Admission for Cardiovascular and Respiratory Diseases", *JAMA*, 295(10), 2006. doi:10.1001/jama.295.10.1127

Giovanetti, R., Rommozzi, E., Zannotti, M., & D'Amato, C. A., "Recent advances in graphene based TiO_2 nanocomposites ($GTiO_2Ns$) for photocatalytic degradation of synthetic dyes", *Catalysts*, 7(10), 305, 2017. doi:10.3390/catal7100305

Jilla, A., Kura, B., “Particulate matter and carbon monoxide emission factors from incense burning”, *Environmental Pollution Climate Change*, 1(4), 2017. doi: 10.4172/2573-458X.1000140

Katoto PDMC, Byamungu L, Brand AS, Mokaya J, Strijdom H, Goswami N, De Boever P, Nawrot TS, Nemery B. Ambient air pollution and health in Sub-Saharan Africa: Current evidence, perspectives and a call to action. *Environ Res. 2019* Jun;173:174-188. doi: 10.1016/j.envres.2019.03.029

Kundu, S., Stone, E., “Composition and sources of fine particulate matter across urban and rural sites in the Midwestern United States”, *Environ. Sci.: Processes Impacts*, 16, 1360-1370, 2014. doi: 10.1039/c3em00719g

Lui, K H, et.al., “Characterization of chemical components and bioreactivity of fine particulate matter ($PM_{2.5}$) during incense burning”, *Environmental Pollution*, ISSN: 0269-7491, 213, 524-532. https://doi.org/10.1016/j.envpol.2016.02.053

Matawle, J.L., Pervez, S., Shrivastava, A. et al., “$PM_{2.5}$ pollution from household solid fuel burning practices in central India:1. Impact on indoor air quality and associated health risks”, *Environ Geochem Health* 39,1045–1058. https://doi.org/10.1007/s10653-016-9871-8, 2017.

Minitab 20 Statistical Software (2020). [Computer software]. State College, PA: Minitab, Inc. (www.minitab.com)

N95 Respirators and Surgical Masks, CDC, 2009. https://blogs.cdc.gov/niosh-science-blog/2009/10/14/n95/

Nag, I. (2021-22). Enhancing Efficacy and Cost Effectiveness of Air Filtration Systems by Optimized Nanoparticle Deposition. Columbia Junior Science Journal. http://cjsjournal.org/s/Ishika-Nag-Article-164.pdf

Nag, I. (2022). Development of a combinatory filtration system for pollution and virus abatement. PLOS ONE 7(22): e0264991. https://doi.org/10.1371/journal.pone.0264991

Nolt-Helms, C., M. Breville., “Environmental Public Health Indicators Impact Report: Data and methods that support environmental public health decision-making by communities,” *U.S. Environmental Protection Agency*, Washington, DC, EPA/600/X-18/163, 2018.

Omran AR (1971) The epidemiological transition: a theory of the epidemiology of population change. Milbank Memorial Fund Q49(4):509–538. doi: 10.1111/j.1468-0009.2005.00398.x

Petkova, E.P., Jack, D.W., Volavka-Close, N.H. et al. Particulate matter pollution in African cities. *Air Qual Atmos Health* 6, 603–614 (2013). https://doi.org/10.1007/s11869-013-0199-6

Ruan, D., Qin, L., Chen, R. et al., “Transparent PAN:TiO_2 and PAN-co-PMA:TiO_2 Nanofiber Composite Membranes with High Efficiency in Particulate Matter Pollutants Filtration”, *Nanoscale Res Lett*., 15, 7. https://doi.org/10.1186/s11671-019-3225-2.

Shi, H., Magaye, R., Castranova, V. et al. Titanium dioxide nanoparticles: a review of current toxicological data. *Part Fibre Toxicol* 10, 15 (2013). https://doi.org/10.1186/1743-8977-10-15.

Szczęśniak, B., Choma, J., & Jaroniec, M., “Gas adsorption properties of graphene-based materials”, *Advances in Colloid and Interface Science*, 243(1), 46-59, 2017. DOI: 10.1016/j.cis.2017.03.007

Thanh Son Le et al., “Air purification equipment combining a filter coated by silver nanoparticles with a nano-TiO_2 photocatalyst for use in hospitals”, *Adv. Nat. Sci*, Nanosci. Nanotechnol. 6 015016, 2015. https://doi.org/10.1088/2043-6262/6/1/015016

United Nations (UN), Department of Economic and Social Affairs, Population Division (2011) World population prospects: the 2010 revision. Population Division of the Department of Economic and Social Affairs of the United Nations Secretariat, New York.

Vijayan, V.K., Paramesh, H., Salvi, S.S., & Dalal, A. A. K., “Enhancing indoor air quality – the air filter advantage”, *Lung India*, 33(6), 705, 2015. doi: 10.4103/0970-2113.164174

Vyas, S., Srivastav, N., Spears, D., “An experiment with Air Purifiers in Delhi during Winter 2015-2016”, *PLoS ONE* 11(12): e0167999. doi: 10.1371/journal.pone.0167999

Wongwatcharapaiboon, J., Gan, G., Riffat, S., “A new air $PM_{2.5}$ filtrative lamp with a combination of fabric filter and TiO_2 coating mop”, *International Journal of Low-Carbon Technologies*, 14 (3), 394–399, https://doi.org/10.1093/ijlct/ctz027, 2019.

World Health Organization, “Air pollution”, 2022. *https://www.who.int/health-topics/air-pollution#tab=tab_1*.

Wrobel, Andrzej & Rokita, E. & Maenhaut, Willy., “Transport of Traffic-Related Aerosols in Urban Areas”, *The Science of the Total Environment*, 257. 199-211, 2000. DOI:10.1016/S0048-9697(00)00519-2

Wu, X., Nethery, R., Sabath, M., Braun, D., Dominici, F., “Exposure to air pollution and COVID-19 mortality in the United States: A nationwide cross-sectional study” https://doi.org/10.1101/2020.04.05.20054502

Xia, X., Riviere, N., Mathur, S., Song, X., Xiao, L., Oldenberg, S., Fadeel, B., Riviere, J., “Mapping the surface adsorption forces of nanomaterials in biological systems”, *National Institute of Health*, 5(11), 9074-9081, 2011. doi: 10.1021/nn203303c

Xing, Y., Xu, Y., Shi, M., Lian, Y., “The impact of $PM_{2.5}$ on the human respiratory system”, *J Thorac Dis*., 2016 Jan; 8(1): E69–E74. doi: 10.3978/j.issn.2072-1439.2016.01.19

Zhang, S., Sun, J., Hu, D., Xiao, C., Zhuo, Q., Wang, J., Qin, C., Dai, L., "Large-sized graphene oxide/modified tourmaline nanoparticle aerogel with stable honeycomb-like structure for high-efficiency $PM_{2.5}$ capture", *J. Mater. Chem*. A, 2018, 6, 16139-16148. https://doi.org/10.1021/acsami.8b22382

Zhao. D., Parham. A., Stephens. B., "Evaluating the long-term health and economic impacts of central residential air filtration for reducing premature mortality associated with indoor fine particulate matter ($PM_{2.5}$) of outdoor origin", *Int. J. Environ. Res. Public Health*, 12(7), 8448-8479, 2015. doi: 10.3390/ijerph120708448

Zhong, Z., Xu, Z., Sheng, T., Yao, J., Xing, W., Wang, Y., "Unusual Air Filters with Ultrahigh Efficiency and Antibacterial Functionality Enabled by ZnO Nanorods", *ACS Applied Materials & Interfaces*, 7 (38), 21538-21544 https://doi.org/10.1021/acsami.5b06810

Zoran., M., et. al., "Assessing the relationship between surface levels of $PM_{2.5}$ and PM_{10} particulate matter impact on COVID-19 in Milan, Italy", *Sci Total Environ*, 2020 Oct 10;738:139825. doi: 10.3389/fpubh.2020.580057

Research article

Wet season chemical composition of atmospheric wet deposition at Cape Point

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Abstract
The measurement of precipitation chemistry enables the assessment of the temporal and spatial evolution of the chemical composition of the atmosphere associated with atmospheric physical and chemical mechanisms. The aims of this study were to report the chemical composition of rainwater collected at a marine environment, i.e. the Cape Point Global Atmosphere Watch (CPT GAW) station from 2004 to 2012. As expected, the volume weighted mean (VWM) concentrations of Na^+ (298.64 µEq.L^{-1}) and Cl^- (354.18 µEq.L^{-1}) were significantly higher compared to the VWM concentrations of other ionic species, as well as compared to the VWM concentrations thereof at the sites in the South African interior. The average pH of rainwater was slightly lower than the pH of unpolluted rainwater, mainly due to NO_3^- associated with the occasional influence of the Cape Town metropole. In contrast to the sites situated in the north-eastern South African interior, where anthropogenic SO_4^{2-} was the major constituent in rainwater, SO_4^{2-} at CPT GAW was entirely associated with marine air with no anthropogenic contribution. It was also indicated that 94% of the chemical content at CPT GAW can be attributed to the marine source.

Keywords
precipitation chemistry, atmospheric fluxes, inorganic ions, Global Atmosphere Watch (GAW), South Africa

Introduction
Atmospheric pollutants are emitted into the atmosphere from various natural (e.g. marine, biogenic and crustal sources) and anthropogenic sources (e.g. fossil fuel combustion, traffic emissions and household combustion) (Mphepya et al., 2004). One of the mechanisms by which pollutants are removed from the atmosphere is through wet deposition (Josipovic et al., 2011). Many atmospheric pollutants are water soluble and are dissolved in cloud water and rain droplets (Waldman et al., 1992). The chemical analysis of rainwater enables the assessment of the temporal and spatial evolution of the chemical composition of the atmosphere, which reflects the numerous atmospheric physical and chemical mechanisms (Mphepya et al., 2006). Rainwater chemistry also reveals changes in atmospheric composition attributed to variances in natural and anthropogenic source contribution and/or meteorology (Vet et al., 2014). In addition, wet deposition can be considered a source of nutrients (e.g. nitrogen, N and sulphur, S) to ecosystems or transport toxic species to the environment depending on its chemical composition (Duce et al., 2009).

Conradie et al. (2016) presented the chemical composition and fluxes of atmospheric wet deposition at four sites located in the interior of South Africa, which are considered to be regionally representative of semi-arid and savannah ecosystems. These sites form part of the South African component of the Deposition of Biogeochemically Important Trace Species (DEBITS) project, which was initiated in 1990 by the Global Atmosphere Watch (GAW) network of the World Meteorological Organisation (WMO) and currently endorsed by the International Global Atmospheric Chemistry (IGAC) programme. The main objectives of DEBITS entail long-term assessments of atmospheric biogeochemical species (mainly carbon (C), N and S species) in the tropics, as well as wet and dry deposition of these species (Lacaux et al., 2003). Also included in the South African DEBITS network is the globally significant Cape Point (CPT) Global Atmosphere Watch (GAW) station, which is a coastal site mainly being impacted by southern hemisphere marine air masses (Fig. 1). CPT GAW is, however, occasionally influenced by air masses passing over the urban-continental region (Fig.1). The aims of this study were to (i) assess the chemical composition of wet season rainwater collected at CPT GAW from 2004 to 2012, (ii) determine S and N wet deposition fluxes, (iii) establish the major sources of ionic species, and (iv) relate ionic composition and wet deposition fluxes at CPT GAW to the South African interior in order to complement the precipitation chemistry presented for the other South African sites (Conradie et al., 2016, Mphepya et al., 2004, Mphepya et al., 2006, Kok et al., 2021).

Methods

Site description

Detailed descriptions of the CPT GAW station are presented in a number of studies (Brunke et al., 2010, Labuschagne et al., 2018). As indicated in Fig. 1, the CPT GAW site (34°21'S, 18°29'E) is located at the southernmost tip of the peninsula on a cliff approximately 230 m above sea-level and approximately 60 km south of the Cape Town metropole (Brunke et al., 2004). The site is situated within a nature reserve within the Cape Floral Region Protected Areas (CFRPA), which has been afforded United Nations Educational, Scientific and Cultural Organization (UNESCO) world heritage status since 2004 (UNESCO, 2015).

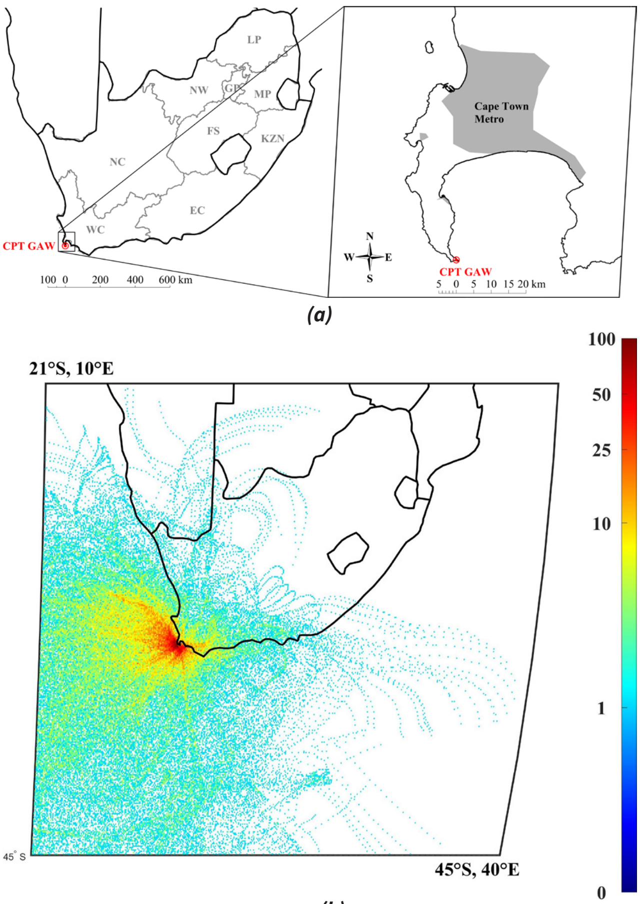

Figure 1: *(a) Regional map of South Africa indicating the location of the measurement station at Cape Point (34°21'S, 18°29'E) along with a zoomed-in map of the region around the site depicting the Cape Town metropole, and (b) normalised overlaid hourly-arriving 72-hour back-trajectories arriving at Cape Point on days in the wet season during which rain samples were collected between 2004 and 2012 with the colour bar indicating overpass intensity over 0.2° by 0.2° grid cells*

Back trajectory analysis

72-hour back trajectories of air mass movement prior to its arrival at the CPT GAW site on days in the wet season during which rain samples were collected at an arrival height of 100 m above ground level are overlaid in Fig. 1b. These trajectories were calculated using the Hybrid Single-Particle Langrangian Integrated Trajectory (HYSPLIT) model (version 4.8) that was developed by the National Oceanic and Atmospheric Administration (NOAA) Air Resources Laboratory (ARL) (Draxler and Hess, 2004). Meteorological data was obtained from the Global Data Assimilation System (GDAS) archive of the National Centre for Environmental Prediction (NCEP) of the United States National Weather Service. Back trajectories were overlaid with fit-for-purpose mathematical programming software on a map of southern Africa that was divided into 0.2° x 0.2° grid cells. The frequency with which trajectories pass over each grid cell is represented by a colour scale where dark blue indicates the lowest proportion, and dark red the highest. This overlay back trajectory map supports the general statement made earlier (in Section 1) that the site is predominantly impacted by marine air masses, and occasionally impacted by continental air masses passing over the Cape Town metropole.

Sampling procedures

Rainwater samples were collected during the wet season from January 2004 to December 2012 on an event basis with an automated wet-only sampler similar to those used at the western and central African DEBITS sites (Galy-Lacaux et al., 2009). The sampler was equipped with a precipitation sensor that controls the cover of the sampler, opening when rain was detected, and hermetically sealing a single-use polyethylene bag in which the rainwater was collected. The precipitation collection area was 225 cm^2. After a rain event, or as soon as practically possible, the collected rainwater was distributed between two 50 ml Greiner-type essay-tubes and frozen (-18°C) immediately. (Galy-Lacaux et al., 2009). Rain depth at CPT GAW was measured with a standard funnel rain gauge.

Although the field protocols of the WMO for precipitation chemistry measurements (WMO, 2004) were followed in general, rainwater sampling at CPT GAW did not entirely comply with WMO protocols. A limitation of the wet-only sampler used at the CPT GAW station was that the lid did not close automatically after a rain event. Furthermore, rainwater samples were only collected during scheduled visits to the CPT GAW site due to logistical restraints. Therefore, some of the advantages associated with using a wet-only sampler, which include minimising contamination of rain samples and delays related to manual operations, were not realised. In addition, some rain samples also comprised composite samples of more than one rain event, such as different rain events occurring on consecutive days. However, in the absence of any other precipitation chemistry measurements for a marine environment in South Africa, as well as within the logistical limitations, the rainwater samples collected at CPT GAW can be considered as the best available of rainwater chemistry for a southern-hemispherical marine background site.

Analysis

The same analytical procedures were followed, as presented by Conradie et al. (2016) and Kok et al. (2021) for the four South African DEBITS sites and Welgegund in the interior of South Africa. In short, samples were unfrozen and analysed immediately. Initial analysis entailed pH and conductivity measurements with an HI 255 combined meter (Hanna Instruments) utilising a low ionic strength electrode. A Dionex ICS 3000 ion chromatograph (IC) was used to determine cation and anion species, i.e. sodium (Na^+), ammonium (NH_4^+), potassium (K^+), calcium (Ca^{2+}), magnesium (Mg^{2+}), nitrate (NO_3^-), chloride (Cl^-), sulphate (SO_4^{2-}), as well as water-soluble organic acids (OA), including formic- (COO^-), acetic- (CH_3COO^-), propionic- ($C_2H_5COO^-$) and oxalic acid ($C_2O_4^{2-}$) in rainwater samples. Details of the IC analytical setup, as well as detection limits of these species were presented by Conradie et al. (2016).

Quality assurance / quality control

Data quality was ensured in accordance with the WMO Data Quality Objectives (DQO) stated in the WMO precipitation chemistry manual (WMO, 2004). All rainwater samples were visually inspected in order to identify visible contaminants, e.g. dust, insects and plant matter, which were removed by filtering the sample through a 0.2 µm filter (supplied by Sigma Aldrich) prior to chemical analysis. Any visible contamination was also recorded. The accuracy of all analytical methods utilised in rainwater analysis (IC, pH and conductivity measurements) were also verified bi-annually through participation in 11 inter-laboratory comparison study (LIS) of the WMO from 2007 to 2012. Example results of the laboratory performance are presented in Fig. 2. Although the analytical laboratory demonstrated a few unsatisfactory results for some measurements (either biased high or low), no continuous or systematic biases were shown throughout the period. The ionic balance of each rain sample was also considered by calculating the ion difference (ID) with the following equation (WMO, 2004):

$$ID(\%) = 100 \times \left({(CE - AE)} / {(CE + AE)} \right) \quad (1)$$

where AE is the total of the anions in μeqL^{-1} and CE is the total of the cations in μeqL^{-1}. Acceptance ranges for the ID, as indicated in the WMO (2004) report, were applied to all the rain samples collected in this study. Only samples that passed WMO ID% criteria are reported in this paper.

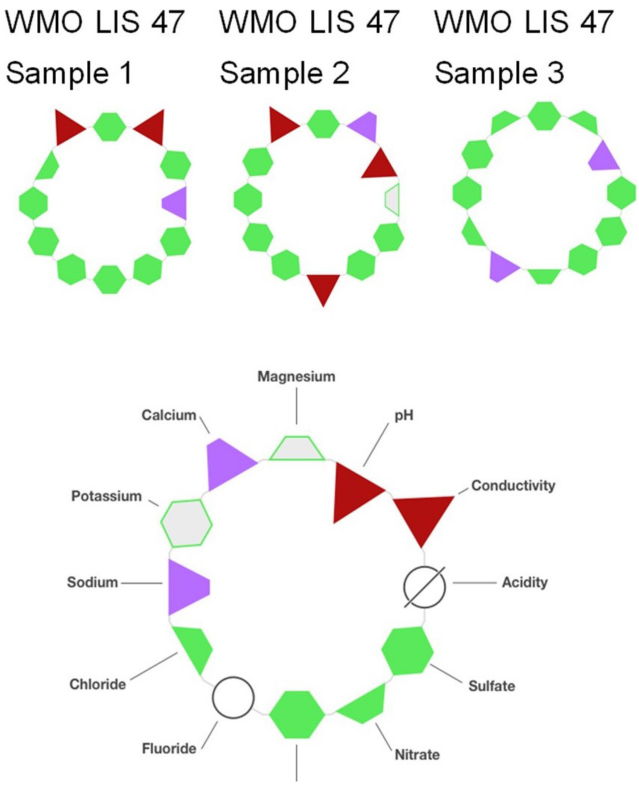

***Figure 2:** Results of the WMO LIS 47 study in 2012 indicated by ring diagrams with a legend for the ring diagram indicated. Green hexagons indicate good results (measurements are within the interquartile range (IQR), defined as the 25th to 75th percentile or middle half (50%) of the measurements), green trapezoids indicate satisfactory results (measurements are within the range defined by median ± IQR/1.349), purple trapezoids indicate results not within the satisfactory category, but within a range defined by the median ± 2(IQR/1.349), and red triangles indicate that the results are unsatisfactory (measurements are outside the range defined by the median + 2(IQR/1.349)). Measurements below the detection limit are indicated by an open circle, while an open circle with a slash through indicates that no measurement was reported. IQR/1.349 is the non-parametric estimate of the standard deviation, sometimes called the pseudo-standard deviation (Qasac-Americas, 2018)*

In Table 1, the total numbers of rainwater samples collected for chemical analysis are presented. All these samples passed the WMO ID% criteria, were of sufficient volume (> 0.2 mm) and were not associated with any analytical errors. As mentioned previously, due to limitations associated with the wet-only samplers utilised at CPT GAW, as well as logistical restraints related to sample collection, rainwater samples did

***Table 1:** Summary of wet deposition samples collected during the wet season at CPT GAW from 2004 to 2012*

	2004	2005	2006	2007	2008	2009	2010	2011	2012	Total
Number of samples collected	10	15	13	16	14	16	12	8	18	122
Collected rainfall (mm)	224	107	126	192	107	240	92	41	214	1341
Total wet season rainfall (mm)	367	186	235	319	191	304	182	167	288	2239
Wet season % TP	61	58	54	60	56	79	51	25	74	60
Total annual rainfall (mm)	484	366	304	422	264	393	254	256	378	3121
Average annual rainfall (mm)										347
% TP	46	29	41	45	40	61	36	16	57	43

not completely adhere to WMO protocols (e.g. some rainwater samples could contain an accumulation of several rain events). In addition, samples were only collected from May to October at CPT GAW, which correspond with the wet season in this part of South Africa. The percentage rainfall collected with valid precipitation chemistry data for all nine of the wet seasons at CPT GAW relative to the nine-year annual precipitation total, i.e. the percentage total precipitation (%TP), was 43%, with none of the nine sampling years reaching the WMO annual %TP acceptance range of 70%. In addition, seven of the nine years had a %TP < 50%. Although the dataset does not satisfy the WMO annual data completeness criterion of %TP ≥ 70%, it does meet the WMO seasonal data completeness criterion of TP ≥ 60%, having a multi-year wet season %TP = 60%. We therefore consider the dataset to be representative of wet season precipitation composition during the nine-year sampling period at the predominantly marine-influenced CPT site (Fig. 1b).

Calculations

The annual volume weighted mean (VWM) concentration of each ionic species was determined as follows (Conradie et al., 2016):

$$VWM(\mu eq.L^{-1}) = \frac{\sum_{i=1}^{N} C_i P_i}{\sum_{i=1}^{N} P_i} \quad (2)$$

where C_i and P_i representing the concentration ($\mu eq.L^{-1}$) of a given ion and the standard gauge rain depth (mm) of each precipitation event, respectively, while N is the total number of rain samples (Table 1). The H^+ concentrations were calculated from the measured pH values.

A general method utilised to estimate the contribution of sea salt to the ionic composition is to calculate the excess concentrations of K^+, Mg^{2+}, Ca^{2+}, Cl^- and SO_4^{2-} with respect to sea salt using Na^+ as a reference, i.e. assuming Na^+ was completely of marine origin. Reference ratios of these species in relation to Na^+ in seawater, as presented by (Keene et al., 1986), were used. The sea salt fractions (ssf) of any of these species, X, with respect to Na^+ are:

$$[X]_{ssf} = [Na^+]_{rain} \times \left(\frac{[X]}{[Na^+]} \right)_{seawater} \quad (3)$$

where $[X]_{ssf}$ is the sea salt contribution of X, $[Na^+]_{rain}$ is the Na^+ concentration in rain, and $[X / Na^+]_{seawater}$ is the seawater concentration ratio (Keene et al., 1986). The non-sea salt fraction (nssf) of X is then calculated by:

$$[X]_{nssf} = [X]_{rain} - ssf_X \quad (4)$$

where $[X]_{rain}$ is the concentration of species X in rainwater. The seawater enrichment factors (EF) of species X with regard to the reference ratio were also calculated as follows (Quiterio et al., 2004, Chao and Wong, 2002):

$$EF_X = \frac{(X / [Na^+])_{rain}}{(X / [Na^+])_{seawater}} \quad (5)$$

Water-soluble OA in wet deposition was considered as reference species for biomass burning (Helas and Pienaar, 1996, Conradie et al., 2016). Neutralisation of sulphuric and nitric acids by base cations can be evaluated by calculating the neutralisation factors (NF) with the following equation (Laouali et al., 2012, Possanzini et al., 1988):

$$NF_X = X / (SO_4^{2-} + NO_3^-) \quad (6)$$

where X is the base cation of interest, i.e. Mg^{2+}, Ca^{2+} and NH_4^+.

Results and discussion

Ionic composition

In Table 2, the wet season VWM concentrations of each ionic species, together with the averaged pH and electrical conductivity (EC) values determined at CPT GAW from 2004 to 2012, are listed. The water-soluble OAs are presented as the sum of VWM concentrations of COO^-, CH_3COO^-, $C_2H_5COO^-$ and $C_2O_4^{2-}$. All-year round VWM concentrations reported by Conradie et al. (2016) for the four South African DEBITS sites in the interior, i.e. Amersfoort (AF), Louis Trichardt (LT), Skukuza (SK) and Vaal Triangle (VT), from 2009 to 2014 are also presented.

It is evident from Table 2 that Na^+ and Cl^- were the most abundant ionic species at CPT GAW, with significantly higher wet season VWM concentrations compared to other ionic species. In addition, Na^+ and Cl^- concentrations were also substantially higher than levels thereof at the four sites in the interior of South Africa, as well as concentrations reported for other DEBITS sites in Africa (Galy-Lacaux et al., 2009). These higher recorded values for Na^+ and Cl^- can be expected for a site predominantly influenced by marine air masses. This is also

Table 2: *Wet season VWM concentrations (µEq.L^{-1}) of ionic species, as well as pH and EC at CPT GAW from 2004 to 2012. Also indicated are all-year round VWM, pH and EC at the four South African DEBITS sites from 2009 to 2014 (Conradie et al., 2016)*

	CPT GAW	AF	LT	SK	VT
pH	5.49	4.32	4.89	4.66	4.51
EC	80.98	42.6	13.1	22.9	33.6
H^+	7.16	61.18	15.24	22.24	44.64
Na^+	298.64	17.79	7.75	13.17	3.50
NH_4^+	13.41	28.50	10.85	12.80	29.06
K^+	11.28	7.35	5.12	2.08	1.41
Mg^{2+}	59.36	5.54	1.93	3.27	4.55
Ca^{2+}	18.57	16.39	6.25	4.69	16.18
NO_3^-	10.01	33.40	7.49	13.20	22.97
Cl^-	354.18	17.96	10.83	15.73	4.52
SO_4^{2-}	33.59	67.21	12.37	18.66	55.00
OA	3.47 (3.14)	14.64 (13.24)	12.14 (11.10)	9.69 (8.69)	12.51 (11.49)

*Dissociated fractions of the organic acids are indicated in brackets

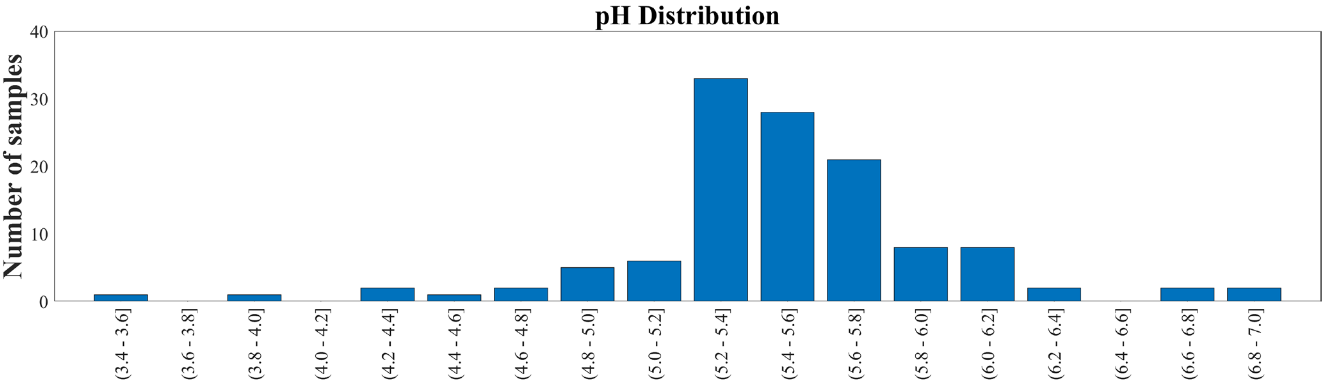

Figure 3: *pH distribution of precipitation samples collected during the annual wet season (May-October) at CPT GAW during the period 2004 to 2012.*

observed for other marine measurement sites such as Cape Grimm, Tasmania (Ayers and Ivey, 1988) and remote islands and coastal East Asian sites (Vet et al., 2014). The significantly higher Na^+ and Cl^- concentrations also contributed to noticeably higher EC of rainwater samples collected at CPT GAW compared to the other South African DEBITS sites. The second and third most abundant species were Mg^+ and SO_4^{2-}, respectively, which are also most likely mainly associated with marine air, as indicated in subsequent paragraphs. SO_4^{2-} was the most abundant species at the four DEBITS sites in the South African interior, which was attributed, by Conradie et al. (2016), to sulphur emissions from anthropogenic activities in this region.

Although CPT GAW is on occasion influenced by air masses passing over the Cape Town metropole (Fig.1), especially during winter, a significantly lower impact of anthropogenic emissions on rainwater chemistry is expected, which is also signified by the lower VWM concentration of NO_3^- compared to the two industrially impacted AF and VT sites.

Substantially lower values are also reported for OA at CPT GAW compared to the other South African DEBITS sites, which reflects a less significant influence of biomass burning on rain chemistry at CPT GAW. Swartz et al. (2020) also indicated that seasonal open biomass burning in the Overberg region did not contribute substantially to the NO_2 and O_3 concentrations at CPT GAW.

Acidity

The average pH of wet season rainwater at CPT GAW was 5.49, which is slightly lower than the pH of unpolluted rainwater, i.e. 5.60 (Eby, 2004). Average rainwater pH at GPT GAW was higher than the average pH at all the sites located in the South African interior, including the rural background sites LT and SK. Evidently, the resultant H^+ concentration was also lower at CPT GAW, especially being significantly lower than H^+ levels at AF and VT in proximity of anthropogenic emissions. In Fig. 3, the pH frequency distribution of rain events occurring during the wet season at CPT GAW is presented. It is evident that most of rain events had pH values ranging between 5.2 and 5.8. It was found that 85% of rain events had pH > 5.2, while 35% had pH > 5.6. The average pH value of rainwater at CPT GAW reflects the low frequency impacts of anthropogenic activities and biomass burning on rainwater chemistry at CPT GAW. However, the occasional influence of anthropogenic emissions is reflected by the largest number of rain events having pH values between 5.2 and 5.4 (Fig. 3), while 9.8% of rain events had pH values lower than 5.0.

Table 3: *Contributions of mineral and organic acids to the total acidity*

	µeq.L⁻¹	%
‡Sulphuric acid	0	0
Nitric acid	10.01	76.1
OA	3.14	23.9
Estimated total H^+ (pA)	13.15	100
Measured H^+	7.16	54.4

‡Anthropogenic sulphates

Table 4: *Acid neutralisation factors (NFX) of CPT GAW wet seasonal wet deposition for 2004 to 2012*

	NF_x
Ca^{2+}	1.85
Mg^{2+}	5.93
NH_4^+	1.34

The acidity potential (pA), presented in Table 3, is the sum of the potential acidic compounds, which include sulphuric acid (anthropogenic SO_4^{2-}), nitric acid (NO_3^-) and OA (Mphepya et al., 2004, Laouali et al., 2012). Empirical estimations indicated that SO_4^{2-} in rainwater at CPT GAW was entirely from marine origin, i.e. completely in the sea-salt fraction (Equation 4). Therefore, terrigenous and anthropogenic sources did not contribute to any SO_4^{2-} measured in rainwater. The measured acidity (H^+, measured) at CPT GAW is lower than the estimated acidity (total H^+, estimated from the pA), which can be ascribed to neutralisation by basic cation species such as Ca^{2+}, NH_4^+ and Mg^{2+}. Neutralisation factors were calculated (Equation 6) to evaluate the neutralisation of nitric acid by these bases, which indicated

Table 5: Pearson correlation for ionic species measured in CPT GAW wet deposition samples collected during the wet season for the period 2004 to 2012. Correlations ≥ 0.7 were considered significant and are highlighted in green (Sheskin, 2003), while correlations < 0.7 are highlighted in orange.

	H^+	Na^+	NH_4^+	K^+	Mg^{2+}	Ca^{2+}	NO_3^-	Cl^-	SO_4^{2-}	F^-	OA
H^+	1.00										
Na^+	-0.05	1.00									
NH_4^+	-0.02	0.51	1.00								
K^+	-0.10	0.77	0.34	1.00							
Mg^{2+}	-0.02	0.91	0.56	0.47	1.00						
Ca^{2+}	-0.10	0.94	0.56	0.72	0.88	1.00					
NO_3^-	0.02	0.12	0.09	0.00	0.14	0.13	1.00				
Cl^-	0.05	0.99	0.51	0.75	0.91	0.93	0.14	1.00			
SO_4^{2-}	0.03	0.86	0.51	0.73	0.77	0.84	-0.20	0.85	1.00		
F^-	-0.05	0.56	0.31	0.59	0.37	0.55	-0.10	0.55	0.63	1.00	
OA	-0.06	0.51	0.38	0.60	0.32	0.49	0.19	0.51	0.43	0.48	1.00

that Mg^{2+} is the major ionic species in rainwater that neutralises nitric acid (Table 4). The calculated potential contribution of the mineral acids (nitric acid) at CPT GAW is 76.1%, which forms the greatest part of free acidity. Therefore, the marginal acidity of rainwater at CPT GAW, as well as rain events with pH < 5, can be attributed to the influence of anthropogenic activities associated with the Cape Town metropole. In addition, Swartz et al. (2020) indicated that, especially during the wet season, CPT GAW is also impacted by the intermittent long-range transport of air masses passing over industrialised northern interior.

Sources

Explorative statistical analysis, i.e. Pearson correlation calculations, was conducted in order to establish relationships between the different ionic species, which could be indicative of similar sources of species. It is evident from Table 5 that good correlations are observed between all species considered to be associated with marine air masses, i.e. Na^+, Cl^-, Mg^{2+}, K^+, Ca^{2+} and SO_4^{2-}. However, correlations between some of these species could also be attributed to terrigenous sources (e.g. Ca^{2+} and Mg^{2+}), which will be explored in subsequent paragraphs. In contrast to Conradie et al. (2016), no correlation is observed between SO_4^{2-} and NO_3^-, which is consistent with different sources, i.e. marine and anthropogenic, respectively, of these species at CPT GAW. In addition, NH_4^+ is moderately correlated with SO_4^{2-} and other ionic species associated with marine air masses and weakly correlated to NO_3^-, which also reflects different sources of SO_4^{2-} and NO_3^-. Furthermore, it implies that marine emissions of NH_4^+ could also be a potential source of NH_4^+ measured in rain at CPT GAW. It is also of interest to note that moderate correlations are observed between OA and species associated with marine air masses (e.g. Na^+ and Cl^-), which are indicative of some of these species also being of marine origin. In addition, no correlations are observed between H^+ and acidic ions, which correspond to the low acidity of wet season rainwater.

The ssf and nssf were calculated for the wet season VWM concentrations of Cl^-, Mg^{2+}, K^+, Ca^{2+} and SO_4^{2-} with Equations 3 and 4, as discussed in the quality assurance / quality control section, in order to estimate the marine and non-marine contributions, while it was assumed that Na^+ was entirely from a marine source. The nssf of Cl^-, Mg^{2+}, K^+, Ca^{2+} and SO_4^{2-} was considered to be terrigenous. The sum of the VWM concentrations of the ssf and nssf of the species comprised the marine and terrigenous contributions, respectively. As indicated in the previous section, SO_4^{2-} was completely of marine origin and an anthropogenic fraction was not calculated. It can be assumed that NO_3^- at CPT GAW is predominantly associated with anthropogenic activities in the Cape Town metropole, e.g. vehicular emissions and household combustion. Swartz et al. (2020) attributed increased NO_2 concentrations at CP GAW to air masses passing over the Cape Town conurbation. NH_4^+ in precipitation is usually attributed to agricultural activities. However, guano from the sea birds and marine NH_4^+ emissions could potentially be a more significant source at CPT GAW. OA concentrations can be considered a proxy for the biomass burning contribution. In Fig. 4, a summary of the estimations of the source group contributions to the chemical composition of rainwater at CPT GAW is presented. It is evident from Fig. 4 that the chemical content of rainwater collected at CPT GAW was dominated by the marine contribution, i.e. 94%, while the other sources contributed ≤ 2% each. This source group distribution for CPT GAW is completely unique in comparison to the source group distribution reported for the four other South African DEBITS sites located in the north-eastern interior where SO_4^{2-} and NO_3^- dominated rainwater composition (Mphepya et al., 2004, Mphepya et al., 2006, Conradie et al., 2016). The main sources of ionic species in rainwater in the north-eastern interior of South African were the combustion of fossil fuels.

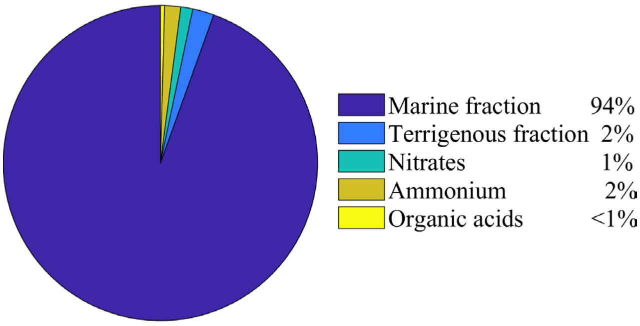

Figure 4: *Estimated source contributions to the chemical composition of rainwater at CPT GAW*

The ratios of Cl^-, Mg^{2+}, K^+, Ca^{2+} and SO_4^{2-} with regard to Na^+ are presented together with the EFs (Equation 5) in relation to the reference seawater ratios (Keene et al., 1986) in Table 6. It is evident that the Cl^-/Na^+, Mg^{+}/Na^+ and SO_4^{2-}/Na^+ ratios were similar to seawater ratios, with EFs very close to one. Calculation of nssf with Equation 4 indicated that Mg^{2+} and SO_4^{2-} were completely in the ssf, while Cl^- was almost entirely (98%) in the ssf. Comparison of rain- and seawater ratios of K^+/Na^+ and Ca^{2+}/Na^+ ratios, as well as EFs indicated that these were not only of marine origin. K^+ and Ca^{2+} were also estimated to be from terrigenous sources, which mainly comprised the terrigenous contribution to the chemical content of rainwater. Therefore, correlations between these species (Table 5) could also be attributed to terrigenous/crustal sources.

Table 6: *Comparison of rainwater ratios at CPT GAW with seawater ratios (Keene et al., 1986) and corresponding enrichment factors (EF)*

	Rain	*Sea-water*	*EF*
Mg^{2+} / Na^+	0.20	0.23	0.88
Cl^- / Na^+	1.19	1.16	1.02
Ca^{2+} / Na^+	0.06	0.04	1.41
K^+ / Na^+	0.04	0.02	1.72
SO_4^{2-} / Na^+	0.11	0.12	0.93

Conclusions

A total of 122 rainwater samples were collected during the wet season (May to October) at CPT GAW from 2004 to 2012. Although the WMO criteria for precipitation collection were generally followed, logistical and instrumental limitations did not allow for sample collection to adhere completely to the WMO guidelines. Therefore, only 43% of the total rain depth from 2004 to 2012 was collected. However, 100% of the samples passed the WMO ID% criteria and none of the samples were discarded due to analytical errors, while samples represented 60% of the wet season rainfall. Therefore, the samples were considered to satisfactorily represent the chemical composition of wet season rainfall over the nine-year sampling period.

Na^+ and Cl^- were the most abundant ionic species at CPT GAW, with wet season VWM concentrations significantly higher compared to other ionic species, as well as higher than all-year round VWM concentrations of Na^+ and Cl^- measured at other South African DEBITS sites in the interior. The average pH of rainwater at this marine site was slightly lower than the pH of unpolluted rainwater, while it is indicated that mainly NO_3^- contributed to the marginal acidity due to the occasional influence of air masses passing over the Cape Town metropole. It was also indicated that SO_4^{2-} in rainwater at CPT GAW was entirely associated with marine air mass with no anthropogenic contribution, which is in contrast to the DEBITS sites situated in the South African interior, where anthropogenic SO_4^{2-} was the major constituent in rainwater. Estimations of source contribution indicated that 94% of the chemical content at CPT GAW can be attributed to the marine source, which signifies that CPT GAW is representative of southern-hemispherical marine air masses.

Valuable conclusions could be drawn from rain samples collected for a region and ecosystem in Southern Africa, for which no precipitation chemistry has been reported. The significance of the influence of anthropogenic activities on precipitation chemistry in the South African interior is also highlighted by the rainwater chemistry for a clean marine background site. It is recommended that future precipitation chemistry studies should include more sites representative of different regions and ecosystems in Southern Africa, while precipitation collection should continue at the current South African DEBITS sites in order to reflect changes in source contributions and meteorology.

Acknowledgements

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References

Ayers, G. P. & Ivey, J. P. 1988. Precipitation composition at Cape Grim, 1977-1985. T*ellus. Series B: Chemical and Physical Meteorolog*y, 40B, 297-307.

Brunke, E. G., Labuschagne, C., Ebinghaus, R., Kock, H. H. & Slemr, F. 2010. Gaseous elemental mercury depletion events observed at Cape Point during 2007–2008. *Atmospheric Chemistry and Physics*, 10, 1121-1131.

Brunke, E. G., Labuschagne, C., Parker, B., Scheel, H. E. & Whittlestone, S. 2004. Baseline air mass selection at Cape Point, South Africa: application of ^{222}Rn and other filter criteria to CO_2. *Atmospheric Environment*, 38, 5693-5702.

Chao, C. Y. & Wong, K. K. 2002. Residential indoor PM_{10} and $PM_{2.5}$ in Hong Kong and the elemental composition. *Atmospheric Environment*, 36, 13.

Conradie, E. H., Van Zyl, P. G., Pienaar, J. J., Beukes, J. P., Galy-Lacaux, C., Venter, A. D. & Mkhatshwa, G. V. 2016. The chemical composition and fluxes of atmospheric wet deposition at four sites in South Africa. *Atmospheric Environment*, 146, 113-131.

Draxler, R. R. & Hess, G. D. 2004. Description of the HYSPLIT_4 modelling system. In: NOAA (ed.) *Technical Memorandum*. 7th ed. Silver Spring, Maryland: ERL ARL.

Duce, R. A., Galloway, J. N. & Liss, P. S. 2009. The impacts of atmospheric deposition to the ocean on marine ecosystems and climate'. *WMO Bulletin 58*.

Eby, G. N. 2004. *Principles of environmental geochemistry*, Australia ; Pacific Grove, CA : Thomson-Brooks/Cole, c2004.

Galy-Lacaux, C., Laouali, D., Descroix, L., Gobron, N. & Liousse, C. 2009. Long term precipitation chemistry and wet deposition in a remote dry savanna site in Africa (Niger). *Atmospheric Chemistry & Physics*, 9, 1579 - 1595.

Helas, G. & Pienaar, J. J. 1996. Chapter 3: "Biomass Burning Emissions. *In*: Held, G., Gore, B. J., Surridge, A. D., Tosen, G. R., Turner, C. R. & Walmsley, R. D. (eds.) *Air Pollution and its Impacts on the South African Highveld*. Cleveland: Environmental Scientific Association.

Josipovic, M., Annegarn, H. J., Kneen, M. A., Pienaar, J. J. & Piketh, S. J. 2011. Atmospheric dry and wet deposition of sulphur and nitrogen species and assessment of critical loads of acidic deposition exceedance in South Africa. *South African Journal Science*, 107, 10.

Keene, W. C., Pszenny, A. A., Galloway, J. N. & Hawley, M. E. 1986. Sea-salt corrections and interpretation of constituent ratios in marine precipitation. *Journal of Geophysical Research*, 91, 12.

Kok, L., Van Zyl, P. G., Beukes, J. P., Swartz, J.-S., Burger, R. P., Ellis, S., Josipov, M., Vakkari, V., Laakso, L. & Kulmala, M. 2021. Chemical composition of rain at a regional site on the South African Highveld. *Water SA*, 47, 12.

Labuschagne, C., Kuyper, B., Brunke, E.-G., Mokolo, T., Van Der Spuy, D., Martin, L., Mbambalala, E., Parker, B., Khan, M. A. H., Davies-Coleman, M. T., Shallcross, D. E. & Joubert, W. 2018. A review of four decades of atmospheric trace gas measurements at Cape Point, South Africa. *Transactions of the Royal Society of South Africa*, 73, 113-132.

Lacaux, J. P., Tathy, J. P. & Sigha, L. 2003. Acid wet deposition in the tropics: Two case studies using DEBITS measurements. *IGACtivities Newsletter of the International Global Atmospheric Chemistry Project*.

Laouali, D., Galy-Lacaux, C., Diop, B., Delon, C., Orange, D., Lacaux, J. P., Akpo, A., Lavenu, F., Gardrat, E. & Castera, P. 2012. Long term monitoring of the chemical composition of precipitation and wet deposition fluxes over three Sahelian savannas. *Atmospheric Environment*, 50, 314-327.

Mphepya, J. N., Galy-Lacaux, C., Lacaux, J. P., Held, G. & Pienaar, J. J. 2006. Precipitation Chemistry and Wet Deposition in Kruger National Park, South Africa. *Journal of Atmospheric Chemistry*, 53, 169-183.

Mphepya, J. N., Pienaar, J. J., Galy-Lacaux, C., Held, G. & Turner, C. R. 2004. Precipitation Chemistry in Semi-Arid Areas of Southern Africa: A Case Study of a Rural and an Industrial Site. *Journal of Atmospheric Chemistry*, 47, 24.

Possanzini, M., Buttini, P. & Di Palo, V. 1988. Characterization of a rural area in terms of dry and wet deposition. *Science of the Total Environment*, 74, 10.

QA/SAC - Americas. 2018. *Lab Intercomparison Study Data* [Online]. Available: http://www.qasac-americas.org/ringdiagram [Accessed 18 January 2019].

Quiterio, S. L., Da Silva, C. R. S., Arbilla, G. & Escaleira, V. 2004. Metals in airborne particulate matter in the industrial district of Santa Cruz, Rio de Janeiro, in an annual period. *Atmospheric Environment*, 32, 11.

Sheskin, D.J. 2003. Handbook of parametric and nonparametric statistical procedures. 3rd ed. Chapman and Hall/CRC Press, Boca Raton, 1193.

Swartz, J.-S., Van Zyl Pieter, G., Beukes Johan, P., Labuschagne, C., Brunke, E.-G., Portafaix, T., Galy-Lacaux, C. & Pienaar Jacobus, J. 2020. Twenty-one years of passive sampling monitoring of SO_2, NO_2 and O_3 at the Cape Point GAW station, South Africa. *Atmospheric Environment*, 222, 117128.

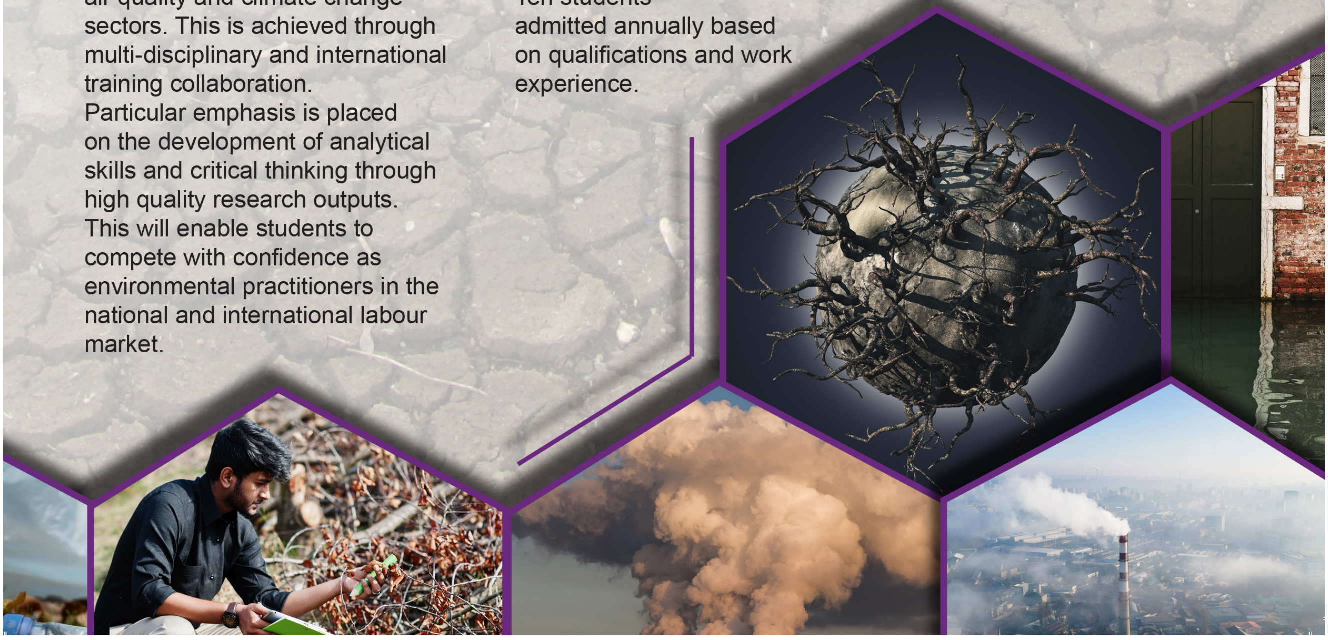
UNESCO. 2015. *Cape Floral Region Protected Areas* [Online]. Available: http://whc.unesco.org/en/list/1007/ [Accessed 20 June 2017].

Vet, R., Artz, R. S., Carou, S., Shaw, M., Ro, C.-U., Aas, W., Baker, A., Bowersox, V. C., Dentener, F., Galy-Lacaux, C., Hou, A., Pienaar,

J. J., Gillett, R., Forti, M. C., Gromov, S., Hara, H., Khodzher, T., Mahowald, N. M., Nickovic, S., Rao, P. S. P. & Reid, N. W. 2014. A global assessment of precipitation chemistry and deposition of sulfur, nitrogen, sea salt, base cations, organic acids, acidity and pH, and phosphorus. *Atmospheric Environment*, 93, 3-100.

Waldman, K. E., Munger, W. J. & Jacob, D. J. 1992. Measurement methods for atmospheric acidity and acid deposition. *In*: Radojević, M. & Harrison, R. M. (eds.) *Atmospheric acidity: Sources, consequences and abatement*. New York: Elsevier.

WMO 2004. WMO/GAW report no. 160, Manual for the GAW precipitation chemistry programme, WMO TD No. 1251.



Research article

A case study in the wintertime Vaal Triangle Air-Shed Priority Area on the utility of the nitrogen stable isotopic composition of aerosol nitrate to identify NO_x sources

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Abstract

In South Africa, the Highveld region and the Johannesburg-Pretoria megacity are known as global NO_x (NO_x = NO + NO_2) "hotspots" identified by satellite-based instruments. The ultimate sink for atmospheric NO_x is conversion to aerosol nitrate. However, measurements of aerosol nitrate concentrations do not provide information on which NO_x sources served as nitrate precursors at that location. This complicates efforts to reduce concentrations of particulate matter (PM) in these air quality priority areas. Here, we measured the nitrogen stable isotope composition of nitrate from daily wintertime collections of coarse mode $PM_{2.5\text{-}10}$ (PM ≤ 10 and >2.5 µm in diameter) at three air quality monitoring stations located in the Vaal Triangle Air-Shed Priority Area (VTAPA). The overall aim of this case study was to evaluate the use of the distinct stable isotope signatures of various NO_x sources to identify their relative contribution to aerosol nitrate across the Highveld. The nitrogen isotope ratios of aerosol nitrate were similar across the three sites, with greater day-to-day variability than site to site variability. Air mass history was the main driver of the variability in the nitrogen isotope ratios of aerosol nitrate, with significantly higher isotopic ratios observed for air masses originating from the southwest. Using an isotope mixing model we determined that NO_x from coal-burning is the dominant contributor to aerosol nitrate (66%), followed by biomass burning (16%), vehicles (12%), and soil emissions (6%).

Keywords

Aerosol Nitrate, Stable Isotopes, NO_x Sources, Highveld, Vaal Triangle

Introduction

High concentrations of ambient aerosols are associated with increased mortality risks. There is a wide range of evidence documenting the large health costs associated with high levels of air pollution (Pervin et al. 2008; Zhao et al. 2016; Altieri and Keen 2019). In many regions of South Africa, particulate matter (PM) is considered the pollutant of greatest concern (City of Cape Town 2008; Thabethe et al. 2014). Anthropogenic aerosol sources include fugitive dust, fires, mining, transportation, electricity generation, industrial activities, domestic fuel burning, and traffic. The latter two sources are typically considered the largest contributors to the South African PM burden (Feig et al. 2016; Venter et al. 2012). Natural sources are dominated by dust and sea spray. PM can be emitted to the atmosphere or formed through gas-to-particle conversion and is composed of a complex mixture of inorganic species, organic carbon, and black carbon with small amounts of trace metals (Seinfeld and Pandis 2012).

Globally, the emission of anthropogenic nitrogen oxides (NO_x = NO + NO_2) to the atmosphere has increased tenfold since preindustrial times (Galloway et al. 2004). NO_x itself is a local air pollutant, and it impacts regional air pollution by serving as a precursor to ozone and secondary PM formation. In South Africa, the Highveld region and the Johannesburg-Pretoria megacity are known as global NO_x "hotspots" identified by satellite-based instruments (Lourens et al. 2012). This region, along with the neighbouring Vaal Triangle, is responsible for > 90% of South Africa's anthropogenic NO_x emissions (Lourens et al. 2012). Similarly, there are large emissions of sulfur dioxide (SO_2) from industrial activities, coal mines, and power generation, particularly in winter (Feig et al. 2016; Lourens et al.

2011; Venter et al. 2012). NO_x and SO_2 emissions are the main precursors to secondary inorganic PM formation and highlight the coupling between gaseous emissions and the formation of secondary aerosols.

Air transported to the central plateau of South Africa originates primarily from the Atlantic and Indian Oceans, with a 25% contribution from the African continent (Freiman and Piketh, 2003). As oceanic NO_x sources are very low, the only external NO_x source to the Highveld/Vaal area would be seasonal biomass burning from neighbouring African countries. The fate of NO_x produced in the Highveld and Vaal regions includes dry and wet deposition and conversion to aerosol nitrate. The local recirculation time scale is 2 to 9 days (Freiman and Piketh 2003; Collett et al. 2010; Igbafe et al. 2015), allowing for complete NO_x conversion to nitrate, such that aerosol nitrate concentrations in the Highveld/Vaal area are primarily influenced by local NO_x emissions. Similarly, local industrial emissions of SO_2 make a considerable contribution to sulfate aerosols, the main component of the summer and winter haze layer in this region (Piketh et al. 1999).

NO_x negatively impacts human health and, as NO_2, is controlled by both minimum emission standards (MES) and national ambient air quality standards (NAAQS) in South Africa (Government Gazette, 24 December 2009, No. 32816). NO_x and volatile organic compounds lead to ozone formation, which is controlled by NAAQS due to its adverse impacts on human health and visibility. The ultimate sink for atmospheric NO_x is the conversion to aerosol nitrate. Nitrate can be present in PM aerosols as ammonium nitrate, sodium nitrate, magnesium nitrate, and calcium nitrate. Due to the adverse impacts on human health and visibility, PM is also controlled by NAAQS. The Highveld Priority Area (HPA) and the Vaal Triangle Air-Shed Priority Area (VTAPA) occasionally record NO_2 values above the NAAQS. However, PM and ozone are regularly out of compliance at stations across the HPA and VTAPA (saaqis.environment.gov.za). Although NO_x emissions rarely exceed the MES, emissions reductions would positively influence air quality in these regions by reducing ozone and PM. However, measuring the nitrate concentration in PM does not provide information on which specific NO_x sources lead to aerosol nitrate formation at that location. This complicates efforts to reduce concentrations of PM in these air quality priority areas.

The atmospheric cycle of NO_x and the conversion of NO_x to aerosol nitrate is complex, with different processes taking place during the day and night (Altieri et al. 2013). In addition to NO_x being a precursor to ozone, ozone can be consumed in the conversion of NO_x to nitrate aerosols, further complicating the influence of NO_x on the chemistry of local and regional air pollution. Previous studies have used nitrogen and oxygen isotopes of nitrate as a tool for distinguishing NO_x sources, as well as the chemical pathway through which NO_x was converted to nitrate in polluted environments (Freyer 1978; Elliott et al. 2007). As NO_x is converted to nitrate, the nitrogen atom is conserved; therefore the $\delta^{15}N$ of nitrate will reflect the $\delta^{15}N$ of the NO_x source. Indeed, studies of the isotopic composition of NO_x have shown that NO_x from vehicle emissions, power generation, industrial activities, lightning, soils, and biomass burning all have distinct isotopic compositions (Felix et al. 2012; Felix and Elliott 2014; Fibiger et al. 2014; Walters et al. 2015; Walters et al. 2015).

The fate of NO_x emissions are usually investigated using dispersion models, but these models lack ozone and secondary PM formation and cannot directly address the relationship between NO_x emissions and measured PM concentrations. Air mass back trajectory analysis is a useful approach for identifying which air masses are associated with higher/lower concentrations of a pollutant. However, the trajectory analysis does not account for loss processes, mixing, and secondary formation processes and only provides a qualitative connection between emission sources and ambient concentrations. The stable isotope approach is unique in that it enables us to quantitatively identify the NO_x sources contributing to aerosol nitrate. Despite the hotspot of NO_x emissions and the high levels of PM recorded at air quality monitoring stations in the HPA and VTAPA, to our knowledge, the nitrogen stable isotope approach has not been used in South Africa to attribute PM nitrate to local and regional NO_x sources.

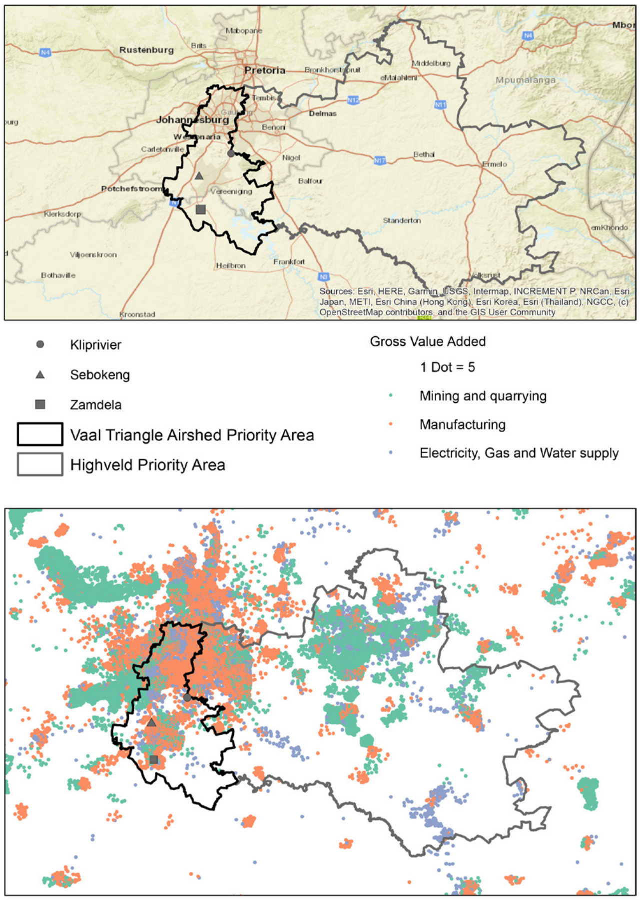

Figure 1: *Map showing the VTAPA and Highveld Priority Area and the three sampling sites in relation to a) the metropolitan area of Pretoria and Johannesburg with major roads indicated and b) the dot density of the gross value add (2009 Rands) of mining and quarrying, manufacturing, and electricity, gas and water supply (Naude et al. 2007).*

Methods

The VTAPA is a well-studied air pollution priority area in South Africa located near the Johannesburg-Pretoria megacity (Figure 1). Aerosol samples analysed for nitrogen stable isotope in this study were collected for a PM source apportionment study (Muyemeki et al. 2021). Briefly, continuous samples were collected at multiple sites using dichotomous low volume samplers with a flow rate of 15 L min^{-1} for the coarse size fraction ($PM_{2.5-10}$) on 47 mm quartz fiber filter membranes. Two consecutive continuous 12-hour samples were collected daily (10:00 – 22:00 and 22:00 to 10:00).

For the nitrogen isotope case study presented here, sixteen coarse mode samples (8 days and 8 nights) collected from 21 June 2018 to 5 July 2018 at three sites were analysed (n = 48). Two of the sites are in densely populated low-income settlements (i.e., Sebokeng (26.5879°S, 27.8410°E) and Zamdela (26.8449°S, 27.8551°E)) and the third is in a low-density area (i.e., Kliprivier (26.4203°S, 28.0849°E); Figure 1). The wintertime samples were the focus of this study as the highest median values for $PM_{2.5-10}$ occurred during winter (Muyemeki et al. 2021). The sampling and methodological approach used here results in a focus on the thermodynamically stable aerosol nitrate. Given the sampling was limited to coarse mode aerosols in the winter, there is little concern that fine mode ammonium nitrate volatilization is impacting the sampling of aerosol nitrate (Schaap et al. 2004). Furthermore, the isotopic approach is specific to inorganic nitrate such that organic nitrate molecules are excluded from analysis (Sigman et al. 2001).

Daytime and nighttime aerosol filters were combined to ensure sufficient sample nitrate for isotope analysis such that each combined sample represents 24-hours (e.g., Filter A = 21 June 2018 10:00 to 22:00 and Filter B = 21 June 2018 22:00 to 22 June 2018 10:00 were combined to generate extract 1 which is representative of 21 June 2018 10:00 to 22 June 2018 10:00).

Aerosol filters were extracted in 10 mL of ultrapure Milli-Q water, sonicated for one hour, stored at 4°C for 12-hours to maximize extraction efficiency, and then frozen for nitrate concentration and isotopic analysis (Gobel et al. 2013). The $^{15}N/^{14}N$ isotopic ratio of NO_3^- ($\delta^{15}N$) was determined using the denitrifier method following (Sigman et al. 2001) at Brown University. Briefly, natural strains of denitrifying bacteria (*Pseudomonas aureofaciens*) that lack the nitrous oxide (N_2O) reductase enzyme needed to convert N_2O to N_2 were used to convert 20 nmol of NO_3^- to N_2O. The N_2O was then analysed by Gas-Chromatograph Isotope Ratio Mass Spectrometry (Thermo-Scientific Delta V Plus) for isotopic determination of $^{15}N/^{14}N$ (Sigman et al. 2001). International reference materials IAEA-N-3 and USGS-34 were used to normalize isotopic values to N_2 in air. The pooled standard deviations of references IAEA-N3 and USGS-34 for $\delta^{15}N$ were 0.09‰ (n=7) and 0.12‰ (n=7), respectively. The pooled standard deviation of sample replicates and duplicates for $\delta^{15}N$ was 0.25‰.

Results and discussion

The coarse mode ($PM_{2.5-10}$) aerosol $\delta^{15}N\text{-}NO_3^-$ ranged from 1.1‰ to 16.6‰ (n=24) (Table 1). The aerosol $\delta^{15}N\text{-}NO_3^-$ values observed here in the Highveld are comparable to winter observations in other regions where coal is the dominant energy source. For example, $\delta^{15}N\text{-}NO_3^-$ in $PM_{2.5}$ in Beijing had a winter average of 11.9‰ ± 4.4 whereas in summer the average was 2.2‰ ± 2.5 (Song et al. 2019). A similar study in Beijing focused on winter pollution and found daily $PM_{2.5}$ $\delta^{15}N\text{-}NO_3^-$ to range from 1.0‰ to 19.6‰ (Zhang et al. 2020). Similar patterns have been observed in the coal-burning Midwest region of the United States, where winter aerosol $\delta^{15}N\text{-}NO_3^-$ was 7.6‰ higher than in summer (Elliott et al. 2009, 2019). For most locations where seasonal nitrate isotope data are available, winter is the season of highest $\delta^{15}N\text{-}NO_3^-$ (Song et al. 2019; Zhang et al. 2020, 2021; Zong et al. 2017, 2020). This is typically attributed to the use of fossil fuel sources for heating and residential combustion. A complete seasonal analysis of aerosol $\delta^{15}N\text{-}NO_3^-$ will reveal if similar patterns are evident in the Highveld region.

Table 1: *The range, average, and standard deviation (SD) of 24-hour coarse mode ($PM_{2.5-10}$) aerosol nitrate $\delta^{15}N$ samples collected at Zamdela (n=8), Sebokeng (n=8), and Kliprivier (n=8) from 21 June 2018 – 6 July 2018.*

Site	$\delta^{15}N\text{-}NO_3^-$ (‰) Range	$\delta^{15}N\text{-}NO_3^-$ (‰) Average (±SD)
Zamdela	3.6-13.0	6.2 (3.0)
Sebokeng	3.4-16.6	7.0 (4.3)
Kliprivier	1.1-14.1	7.1 (4.4)

The $\delta^{15}N$ was very similar across all three sites (Table 1; $p >> 0.05$), and the day-to-day variability was generally higher than the between site variability (Figure 2). In particular, the 4 July 2018 sample is higher in $\delta^{15}N$ at all sites than any of the other sample days (Figure 2). This suggests that the aerosol nitrate resulted from a different combination of NO_x sources, or that there was a change in atmospheric or chemical conditions such that isotopic fractionation occurred. The average NO_3^- concentration varied little across the last three sampling days, from 2.5 µg m^{-3} on the 2 July, 3.6 µg m^{-3} on the 4 July, and 3.1 µg m^{-3} on the 6 July. However, the across site average $\delta^{15}N\text{-}NO_3^-$ increased dramatically from the 2 to 4 July, i.e., from 3.5‰ to 14.6‰, and then decreased back to 3.8‰ on the 6 July (Figure 2). The air mass back trajectories for each of these days shows that there was indeed a change in the source region from the 2 to 4 July (Figure 3). This highlights the sensitivity of the $\delta^{15}N$ to changes in NO_x sources, as compared to the insensitivity of concentration changes with respect to changes in NO_x sources.

Although day-to-day variability is greater, there is still between site variability, particularly on the 26 June and 2 July. It is possible that the sites have different air mass histories on those days, but the coarse resolution of the meteorology input data (GDAS 1°) and back trajectory modeling limits our ability to run specific back trajectories for each site. Given how close the three

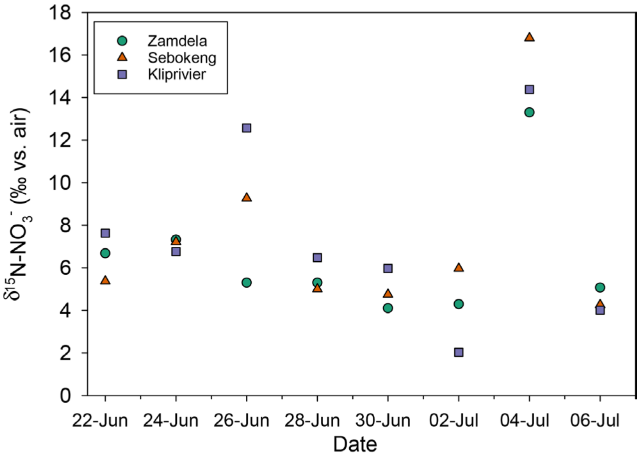

Figure 2: *Coarse mode ($PM_{2.5\text{-}10}$) aerosol nitrate $\delta^{15}N$ collected over 24-hours from 22 June 2018 to 6 July 2018 at Zamdela (green circles), Sebokeng (orange triangles), and Kliprivier (purple squares).*

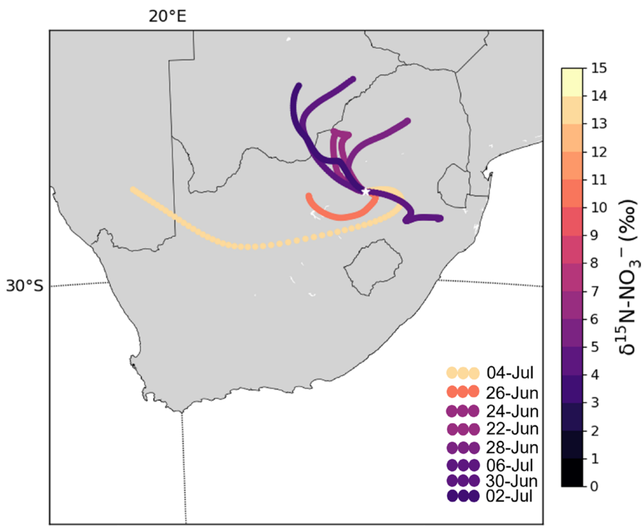

Figure 3: *72-hour air mass back trajectories clustered for each 24-hour aerosol sample and coloured by mass-weighted average $\delta^{15}N\text{-}NO_3^-$ (‰ vs air). The legend indicates the sample date and corresponding mass-weighted average $\delta^{15}N\text{-}NO_3^-$ (in order of descending $\delta^{15}N$) to facilitate comparison with Figure 1.*

sites are, very high-resolution meteorological data are needed to distinguish site-specific air mass back trajectories. Another possibility is that local emissions are influencing the sites on those days. Although they are all located in the Vaal Triangle and influenced by domestic fuel burning, the sites have different proximities to local industry. Zamdela is just south of the Sasol Chemical Industries Complex, Sebokeng is north of both metallurgical industries and mining activities, and Kliprivier is near steel and metalloy industries (Figure 1). Therefore, the between-site variability could be due to differing regional sources that are not evident with low-resolution back trajectory modeling, or site-specific local sources.

Source apportionment using a Bayesian isotope mixing model

Bayesian isotope mixing models are a commonly used approach for mathematically determining the contribution from multiple sources to a measured isotope value, in this case, the contribution of multiple NO_x sources to a measured aerosol nitrate sample. *simmr* is a stable isotope mixing model designed to solve mixing equations for stable isotope data within a Bayesian framework (Parnell et al. 2010, 2013). In this model, Markov chain Monte Carlo works by considering all possible solutions within the given space of uncertainty (i.e., 95% confidence interval). *simmr* requires a minimum of 3 input objects; the measured isotopic composition of the samples, the mean $\delta^{15}N$ of the possible sources, and the associated standard deviations of the source $\delta^{15}N$ (Figure 4).

simmr model inputs

The expected dominant NO_x sources in the Highveld include coal combustion, vehicles, biomass burning, and biogenic soil emissions (Collett et al. 2010). The utility of the nitrogen stable isotope composition is dependent on these NO_x sources having differing isotopic signatures. Below we present the most recent and community accepted $\delta^{15}N\text{-}NO_x$ values and discuss their relevance to the South African context.

The NO_x emitted from coal-burning power stations is formed from either the reaction of air N_2 with O_2 during combustion and/or the reaction of nitrogen in the coal with O_2. The $\delta^{15}N$ of NO_x emitted from coal-fired power plants is therefore dependent on the $\delta^{15}N$ of the nitrogen in the coal, and on the technologies used to reduce NO_x emissions (or lack thereof) (Felix et al. 2012). The $\delta^{15}N$ of coal does not vary considerably, with reported values ranging from 1.0‰ to 4.0‰ for coal from a diversity of sources (Felix et al. 2012). Furthermore, a local study of South African coal-$\delta^{15}N$ found values of 1.0‰ to 1.2‰ (Heaton 1990). A study in the USA of multiple power stations burning the same coal with different technologies found that the dominant control on the NO_x $\delta^{15}N$ was the presence (or absence) of NO_x reduction technologies (Felix et al. 2012). Coal-fired power stations in South Africa are not currently operated with NO_x or sulfur-reducing technologies such as over fire air, selective catalytic (or noncatalytic) reduction, low NO_x burners, or flue gas desulfurization. As such, the closest analogue from measured power station $\delta^{15}N\text{-}NO_x$ includes power stations in the USA measured with the selective catalytic reduction turned off ($\delta^{15}N\text{-}NO_x$ = 10.5‰ ± 2) (Felix et al. 2012; Walters et al. 2015), and a study of power plants in China with a range of emissions reductions ($\delta^{15}N\text{-}NO_x$ = 14.5‰ ± 4.4) (Song et al. 2019). Interestingly, the $\delta^{15}N$ of NO_x emitted from four South African power stations was measured in the 1980's and was found to range from 6‰ to 13‰ (Heaton 1990). However, these values should be treated with caution given recent studies on methodological challenges associated with capturing NO_x in these environments and the rigorous procedures that are now considered standard for capturing NO_x for isotopic analyses (Walters et al. 2015; Walters et al. 2015; Felix et al. 2012; Fibiger et al. 2014). In order to capture the variability in measured values, the $\delta^{15}N\text{-}NO_x$ for coal-fired power stations was set at 13.7‰ ± 4.6 (Figure 4), in line with other recent studies on air pollution in Chinese cities (e.g., Chang et al. 2018, 2019).

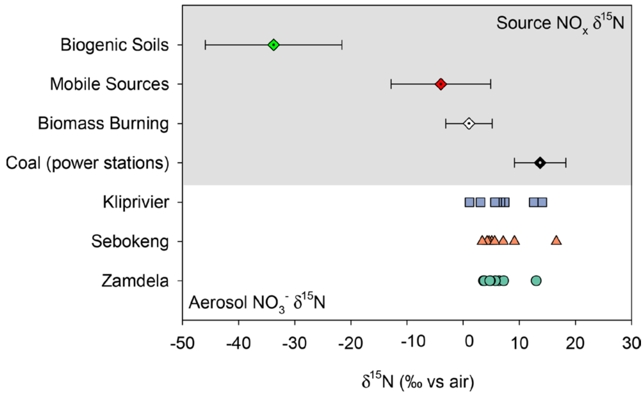

Figure 4: *Inputs to the* simmr *package include average and standard deviations of source NO_x $\delta^{15}N$ (grey shaded area) and the measured coarse mode aerosol $\delta^{15}N$ for Zamdela (green circles), Sebokeng (orange triangles), and Kliprivier (purple squares). Literature references used for the source NO_x $\delta^{15}N$ ranges are in the text.*

The $\delta^{15}N$ of vehicle emitted NO_x is well constrained with respect to the emissions from specific vehicles across driving conditions (Walters et al. 2015). The thermal production of NO_x in the combustion chamber of vehicles results from N_2 in air, which has a $\delta^{15}N$ of 0‰. In a study of 26 different petrol and diesel vehicles representing 12 manufacturers and varying in model year from 1995 to 2015, the $\delta^{15}N$-NO_x ranged from -19.1‰ to 9.8‰. Diesel vehicles had distinctly lower $\delta^{15}N$-NO_x values than petrol vehicles (-23.9‰ to -15.9‰ vs. -15.1‰ to 10.5‰, respectively) due to the different combustion conditions. The same South African study that measured power station NO_x in the 1980's also measured vehicle exhaust, and the $\delta^{15}N$-NO_x ranged from -2‰ to -13‰ (Heaton 1990), which is comparable to the more modern values. The vast majority of NO_x emissions result in the first few minutes when a cold engine is started, and as a result, vehicle run time is highly correlated with the $\delta^{15}N$-NO_x emitted.

Given these controls on the $\delta^{15}N$-NO_x from vehicle emissions, vehicle run time is commonly used to predict the $\delta^{15}N$ of vehicle emissions for petrol and diesel vehicles (Walters et al. 2015). Walters et al. (2015) apply this numerical approach to estimate $\delta^{15}N$-NO_x from vehicle emissions for the USA using commute times and assuming all vehicles are petrol (the diesel share is ~ 2% in the USA). Following their approach, we calculate an estimate of the range of $\delta^{15}N$-NO_x values for the South African vehicle fleet using local commute times and the market share of petrol and diesel vehicles. South African commute times are significantly longer than the OECD country average according to a comprehensive analysis of four surveys across a 20 year period (Kerr 2015). The total calculated commute times reported were 68 to 94 minutes per day, with driving times reported to range from 29 to 42 minutes and minibus times from 38 to 53 minutes. Vehicle fleet data suggest diesel vehicles are ~ 17% of the South African fleet (Posada 2018). Using commute times of 23 to 59 minutes, we calculate that the $\delta^{15}N$-NO_x for petrol vehicles ranges from -2.2‰ to -0.6‰ and for diesel vehicles ranges from -17.6‰ to -15.7‰. The mass-weighted average and standard deviation for South African vehicle emissions $\delta^{15}N$-NO_x, assuming diesel vehicles account for 17% of the total fleet, is calculated as -3.96‰ ± 8.9 (Figure 4). This is remarkably similar to the globally recommended value for mobile emissions of -3.71‰ ± 10.4 (Chang et al. 2018). Here, we used the South African estimate, although it is important to note that what is presented here is overly simplistic. It is recommended that an in-depth analysis of commute times as a function of transport mode, vehicle type, age, and fuel usage be conducted to constrain this end member input for future isotope studies.

The $\delta^{15}N$ of the NO_x released during biomass burning is primarily driven by the $\delta^{15}N$ of the biomass being burned. A comprehensive assessment of multiple biomass types burned in controlled conditions resulted in an average $\delta^{15}N$-NO_x of 1.04‰ ± 4.1 (Fibiger and Hastings 2016). The biomass species burned in Fibiger and Hastings (2016) were all North American, and there is evidence that climatic drivers influence plant and soil $\delta^{15}N$ with higher values observed in the mid-latitude southern hemisphere than in the mid-latitude northern hemisphere (Amundson et al. 2003). Indeed, a study of plant-$\delta^{15}N$ from a variety of species collected over three years from multiple habitats in Kruger National Park had values ranging from 2.3‰ to 6.7‰ (trees), 1.7‰ to 6.4‰ (grasses), and 2.3‰ to 8.4‰ (forb) (Codron et al. 2013). However, lower values were observed during the dry season than the wet season, e.g., dry season grasses had a $\delta^{15}N$ of 2.7‰ ± 2.0. There is significantly less rainfall in winter in the study region, which suggests the lower end of the isotopic range is most appropriate for this study. Until a more comprehensive assessment of the $\delta^{15}N$ of local biomass can be determined, the Fibiger and Hastings (2016) estimate for biomass burning $\delta^{15}N$-NO_x (i.e., 1.04‰ ± 4.1; Figure 4) is used in this study as it overlaps the values measured in at least one region of South Africa.

The $\delta^{15}N$ of biogenic NO emissions from soils is not dependent on the soil $\delta^{15}N$ or the $\delta^{15}N$ of fertilizer (i.e., if the soils are fertilized), but rather the biological processes that lead to NO release (Li and Wang 2008). The NO that is released is an intermediate that forms from nitrification of ammonium to nitrate and denitrification from nitrate to nitrogen gas. Both processes result in a strong kinetic preference for ^{14}N such that the products (in this case NO) are heavily depleted in $\delta^{15}N$ (Ludwig et al. 2001). As a result, NO from soils is very isotopically deplete (-50‰ to -30‰) and is isotopically distinct from all other NO_x emission sources. As the biological processes would be the same in South African soils, we use the commonly accepted $\delta^{15}N$-NO_x for biogenic soil emissions of -33.8‰ ± 12.2 (Li and Wang 2008) (Figure 4).

simmr model results

The *simmr* mixing model results were identical for the three sites when each dataset was input separately. Therefore, the results from the *simmr* model presented below are based on using the mass-weighted average for each sample day across the three sites. To summarize the above discussion, the source NO_x $\delta^{15}N$ values used were 13.7‰ ± 4.6 for coal-fired power stations, -3.96‰ ± 8.88 for mobile sources (i.e., vehicles), 1.04‰ ± 4.13 for biomass burning, and -33.77‰ ± 12.16 for biogenic soil emissions (Figure 4).

According to the *simmr* model output, coal combustion NO_x is the dominant source and accounts for 66% (± 8.1%) of the aerosol nitrate. This is in addition to the large direct contribution coal combustion makes to the fine (>60%) and coarse (up to 20%) PM fraction (Muyemeki et al. 2021). NO_x from biomass burning contributes 15.6% (± 10.3%) to aerosol nitrate. This is consistent with the seasonal patterns in biomass burning in South Africa with an expected peak in late winter and early spring (Hersey et al. 2015). The third largest NO_x source to aerosol nitrate was vehicles, which contributed 12.0% (± 7.5). Vehicles were also a large contributor to coarse PM concentrations (14%) (Muyemeki et al. 2021). The smallest NO_x source was soils, which contributed 6.4% (± 3.1%). Given that it was winter, it is expected that soil biological activity should be at a minimum.

The Monte Carlo analysis incorporates the uncertainty in the NO_x source signature $\delta^{15}N$. These estimates could be refined by directly measuring the source $\delta^{15}N\text{-}NO_x$ in South Africa. In particular, the emissions from coal fired power plants need to be measured directly due to the unique technology used in South African power stations as well as the lack of pollution control technologies. In addition, a more nuanced understanding of vehicle fleet composition and commute times would allow for a narrower estimate of vehicle emission $\delta^{15}N\text{-}NO_x$, and perhaps even site-specific source estimates. The results of this study suggest that future work on site-specific fleet modeling would be useful for understanding the impact of vehicle emissions on NO_x and aerosol nitrate.

Conclusions

Coarse mode ($PM_{2.5\text{-}10}$) aerosol samples were collected at three sites during winter in the VTAPA and the nitrogen isotope composition of the aerosol nitrate was determined. The range and variability in the $\delta^{15}N\text{-}NO_3^-$ was similar across all three sites. The sample days with elevated $\delta^{15}N\text{-}NO_3^-$ have different air mass back trajectories compared to the rest of the sample set, suggesting that those regions have a different combination of NO_x sources. A Bayesian isotope mixing model was used along with source NO_x $\delta^{15}N$ data from the literature to determine the NO_x source contributions to the observed aerosol $\delta^{15}N\text{-}NO_3^-$. Over two-thirds of the aerosol nitrate was formed from power station coal-burning NO_x emissions, while biomass burning and vehicle NO_x emissions contributed the remainder. Soil emissions made a minor contribution. This study highlights the utility of the nitrogen stable isotopes in constraining the relative importance of different NO_x sources to secondary PM, and in determining the NO_x sources that impact the nitrate component of PM at a given site. Ultimately this information can be used to design NO_x reduction strategies in a way that maximizes the impact on the PM burden.

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References

Altieri KE, Hastings MG, Gobel AR, Peters AJ, & Sigman DM (2013) 'Isotopic composition of rainwater nitrate at Bermuda: The influence of air mass source and chemistry in the marine boundary layer', *Journal of Geophysical Research: Atmospheres*, 118(19):11–304.

Altieri KE & Keen SL (2019) 'Public health benefits of reducing exposure to ambient fine particulate matter in South Africa', *Science of The Total Environment*, 684:610–620, doi:10.1016/j.scitotenv.2019.05.355.

Amundson R, Austin AT, Schuur E a. G, Yoo K, Matzek V, Kendall C, Uebersax A, Brenner D, & Baisden WT (2003) 'Global patterns of the isotopic composition of soil and plant nitrogen', *Global Biogeochemical Cycles*, 17(1), doi:10.1029/2002GB001903.

Chang Y, Zhang Y, Tian C, Zhang S, Ma X, Cao F, Liu X, Zhang W, Kuhn T, & Lehmann MF (2018) 'Nitrogen isotope fractionation during gas-to-particle conversion of NO_x to NO_3^- in the atmosphere – implications for isotope-based NO_x source apportionment', *Atmospheric Chemistry and Physic*s, 18(16):11647–11661, doi:https://doi.org/10.5194/acp-18-11647-2018.

Chang Y, Zhang Y-L, Li J, Tian C, Song L, Zhai X, Zhang W, Huang T, Lin Y-C, Zhu C, Fang Y, Lehmann MF, & Chen J (2019) 'Isotopic constraints on the atmospheric sources and formation of nitrogenous species in clouds influenced by biomass burning', *Atmospheric Chemistry and Physics*, 19(19):12221–12234, doi:10.5194/acp-19-12221-2019.

City of Cape Town (2008) *Khayelitsha Air Pollution Strategy Project (KAPS) Report*, City of Cape Town.

Codron J, Lee-Thorp JA, Sponheimer M, & Codron D (2013) 'Plant stable isotope composition across habitat gradients in a semi-arid savanna: implications for environmental reconstruction', *Journal of Quaternary Science*, 28(3):301–310, doi:10.1002/jqs.2614.

Collett KS, Piketh SJ, & Ross KE (2010) 'An assessment of the atmospheric nitrogen budget on the South African Highveld', *South African Journal of Science*, 106(5/6):1–9.

Elliott EM, Kendall C, Boyer EW, Burns DA, Lear GG, Golden HE, Harlin K, Bytnerowicz A, Butler TJ, & Glatz R (2009) 'Dual nitrate isotopes in dry deposition: Utility for partitioning NO_x source contributions to landscape nitrogen deposition', *Journal of Geophysical Research: Biogeosciences*, 114(G4), doi:10.1029/2008JG000889.

Elliott EM, Kendall C, Wankel SD, Burns DA, Boyer EW, Harlin K, Bain DJ, & Butler TJ (2007) 'Nitrogen isotopes as indicators of NO x source contributions to atmospheric nitrate deposition across the midwestern and northeastern United States', *Environmental Science & Technology*, 41(22):7661–7667.

Elliott EM, Yu Z, Cole AS, & Coughlin JG (2019) 'Isotopic advances in understanding reactive nitrogen deposition and atmospheric processing', *Science of The Total Environment*, 662:393–403, doi:10.1016/j.scitotenv.2018.12.177.

Feig GT, Naidoo S, & Ncgukana N (2016) 'Assessment of ambient air pollution in the Waterberg Priority Area 2012-2015', *The Clean Air Journal*, 26(1):21–28.

Felix JD & Elliott EM (2014) 'Isotopic composition of passively collected nitrogen dioxide emissions: Vehicle, soil and livestock source signatures', *Atmospheric Environment*, 92:359–366, doi:10.1016/j.atmosenv.2014.04.005.

Felix JD, Elliott EM, & Shaw SL (2012) 'Nitrogen Isotopic Composition of Coal-Fired Power Plant NO_x: Influence of Emission Controls and Implications for Global Emission Inventories', *Environmental Science & Technology*, 46(6):3528–3535, doi:10.1021/es203355v.

Fibiger DL & Hastings MG (2016) 'First Measurements of the Nitrogen Isotopic Composition of NO_x from Biomass Burning', *Environmental Science & Technology*, 50(21):11569–11574, doi:10.1021/acs.est.6b03510.

Fibiger DL, Hastings MG, Lew AF, & Peltier RE (2014) 'Collection of NO and NO_2 for Isotopic Analysis of NO_x Emissions', *Analytical Chemistry*, 86(24):12115–12121, doi:10.1021/ac502968e.

Freiman MT & Piketh SJ (2003) 'Air Transport into and out of the Industrial Highveld Region of South Africa', Journal of Applied Meteorology and Climatology, 42(7):994–1002, doi:10.1175/1520-0450(2003)042<0994:ATIAOO>2.0.CO;2.

Freyer HD (1978) 'Seasonal trends of NH+4 and NO−3 nitrogen isotope composition in rain collected at Jülich, Germany', *Tellus*, 30(1):83–92, doi:10.1111/j.2153-3490.1978.tb00820.x.

Galloway JN, Dentener FJ, Capone DG, Boyer EW, Howarth RW, Seitzinger SP, Asner GP, Cleveland CC, Green PA, Holland EA, Karl DM, Michaels AF, Porter JH, Townsend AR, & Vosmarty CJ (2004) 'Nitrogen Cycles: Past, Present, and Future', *Biogeochemistry*, 70(2):153–226, doi:10.1007/s10533-004-0370-0.

Gobel AR, Altieri KE, Peters AJ, Hastings MG, & Sigman DM (2013) 'Insights into anthropogenic nitrogen deposition to the North Atlantic investigated using the isotopic composition of aerosol and rainwater nitrate', *Geophysical Research Letters*, 40(22):5977–5982.

Heaton THE (1990) '$^{15}N/^{14}N$ ratios of NO_x from vehicle engines and coal-fired power stations', *Tellus* B, 42(3):304–307, doi:10.1034/j.1600-0889.1990.00007.x-i1.

Hersey SP, Garland RM, 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 modeling data', *Atmospheric Chemistry and Physics*, 15(8):4259–4278, doi:10.5194/acp-15-4259-2015.

Igbafe AI, Jewell LL, Piketh SJ, & Dlamini TS (2015) 'A Wintertime Impact of Dilution on Transformation Rate in the Planetary Boundary Layer', *Journal of Environmental Informatics*, 8(1):34–48.

Kerr A (2015) *Tax(i)ing the poor? Commuting costs in South Africa*, University of Cape Town, Cape Town.

Li D & Wang X (2008) 'Nitrogen isotopic signature of soil-released nitric oxide (NO) after fertilizer application', *Atmospheric Environment*, 42(19):4747–4754, doi:10.1016/j.atmosenv.2008.01.042.

Lourens AS, Beukes JP, Van Zyl PG, Fourie GD, Burger JW, Pienaar JJ, Read CE, & Jordaan JH (2011) 'Spatial and temporal assessment of gaseous pollutants in the Highveld of South Africa', *South African Journal of Science*, 107(1/2):1–8, doi:10.4102/sajs.v107i1/2.269.

Lourens ASM, Butler TM, Beukes JP, van Zyl PG, Beirle S, Wagner TK, Heue K-P, Pienaar JJ, Fourie GD, & Lawrence MG (2012) 'Re-evaluating the NO_2 hotspot over the South African Highveld', *South African Journal of Science*, 108(11–12):83–91.

Ludwig J, Meixner FX, Vogel B, & Förstner J (2001) 'Soil-air exchange of nitric oxide: An overview of processes, environmental factors, and modeling studies', *Biogeochemistry*, 52(3):225–257, doi:10.1023/A:1006424330555.

Muyemeki L, Burger R, Piketh SJ, Language B, Beukes JP, & Zyl PG van (2021) 'Source apportionment of ambient $PM_{10-2.5}$ and $PM_{2.5}$ for the Vaal Triangle, South Africa', *South African Journal of Science*, 117(5/6), doi:10.17159/sajs.2021/8617.

Naude A, Badenhorst W, Zietsman L, Van Huyssteen E, & Maritz J (2007) *Geospatial Analysis Platform - Version 2: Technical overview of the mesoframe methodology and South African Geospatial Analysis Platform.*, CSIR.

Parnell AC, Inger R, Bearhop S, & Jackson AL (2010) 'Source Partitioning Using Stable Isotopes: Coping with Too Much Variation', *PLOS ONE*, 5(3):e9672, doi:10.1371/journal.pone.0009672.

Parnell AC, Phillips DL, Bearhop S, Semmens BX, Ward EJ, Moore JW, Jackson AL, Grey J, Kelly DJ, & Inger R (2013) 'Bayesian stable isotope mixing models', *Environmetrics*, 24(6):387–399, doi:10.1002/env.2221.

Pervin T, Gerdtham U-G, & Lyttkens CH (2008) 'Societal costs of air pollution-related health hazards: A review of methods and results.', *Cost effectiveness and resource allocation: C/E*, 6:19, doi:10.1186/1478-7547-6-19.

Piketh SJ, Annegarn HJ, & Tyson PD (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, doi:10.1029/1998JD100014.

Posada F (2018) *South Africa's New Passenger Vehicle CO_2 Emission Standards: Baseline determination and benefits assessment*, The International Council on Clean Transportation, https://www.fiafoundation.org/media/461198/south-africa-pv-emission-stds_icct-white-paper_17012018_vf-1.pdf.

Schaap M, Spindler G, Schulz M, Acker K, Maenhaut W, Berner A, Wieprecht W, Streit N, Müller K, Brüggemann E, Chi X, Putaud J-P, Hitzenberger R, Puxbaum H, Baltensperger U, & ten Brink H (2004) 'Artefacts in the sampling of nitrate studied in the "INTERCOMP" campaigns of EUROTRAC-AEROSOL', *Atmospheric Environment*, 38(38):6487–6496, doi:10.1016/j.atmosenv.2004.08.026.

Seinfeld JH & Pandis SN (2012) *Atmospheric Chemistry and Physics: From Air Pollution to Climate Change*, Wiley.

Sigman DM, Casciotti KL, Andreani M, Barford C, Galanter M, & Böhlke JK (2001) 'A Bacterial Method for the Nitrogen Isotopic Analysis of Nitrate in Seawater and Freshwater', *Analytical Chemistry*, 73(17):4145–4153, doi:10.1021/ac010088e.

Song W, Wang Y-L, Yang W, Sun X-C, Tong Y-D, Wang X-M, Liu C-Q, Bai Z-P, & Liu X-Y (2019) 'Isotopic evaluation on relative contributions of major NO_x sources to nitrate of $PM_{2.5}$ in Beijing', *Environmental Pollution*, 248:183–190, doi:10.1016/j.envpol.2019.01.081.

Thabethe NDL, Engelbrecht JC, Wright CY, & Oosthuizen MA (2014) 'Human health risks posed by exposure to PM_{10} for four life stages in a low socio-economic community in South Africa', *Pan African Medical Journal*, 18:1–12, doi:10.11604/pamj.2014.18.206.3393.

Venter AD, Vakkari V, Beukes JPJP, van Zyl PG, Laakso H, Mabaso D, Tiitta P, Josipovic M, Kulmala M, Pienaar JJ, & Laakso L (2012) 'An air quality assessment in the industrialised western Bushveld Igneous Complex, South Africa', *South African Journal of Science*, 108(9–10):1–10.

Walters Wendell W., Goodwin SR, & Michalski G (2015) 'Nitrogen Stable Isotope Composition ($\delta^{15}N$) of Vehicle-Emitted NO_x', *Environmental Science & Technology*, 49(4):2278–2285, doi:10.1021/es505580v.

Walters W.W., Tharp BD, Fang H, Kozak BJ, & Michalski G (2015) 'Nitrogen Isotope Composition of Thermally Produced NO_x from Various Fossil-Fuel Combustion Sources', *Environmental Science and Technology*, 49(19):11363–11371, doi:10.1021/acs.est.5b02769.

Zhang Z, Guan H, Luo L, Zheng N, Xiao Hongwei, Liang Y, & Xiao Huayun (2020) 'Sources and transformation of nitrate aerosol in winter 2017–2018 of megacity Beijing: Insights from an alternative approach', *Atmospheric Environment*, 241:117842, doi:10.1016/j.atmosenv.2020.117842.

Zhang Z, Guan H, Xiao Hongwei, Liang Y, Zheng N, Luo L, Liu C, Fang X, & Xiao Huayun (2021) 'Oxidation and sources of atmospheric NO_x during winter in Beijing based on δ18O-$\delta^{15}N$ space of particulate nitrate', *Environmental Pollution*, 276:116708, doi:10.1016/j.envpol.2021.116708.

Zhao X, Yu X, Wang Y, & Fan C (2016) 'Economic evaluation of health losses from air pollution in Beijing, China', E*nvironmental Science and Pollution Research*, 23(12):11716–11728, doi:10.1007/s11356-016-6270-8.

Zong Z, Tan Y, Wang X, Tian C, Li J, Fang Y, Chen Y, Cui S, & Zhang G (2020) 'Dual-modelling-based source apportionment of NO_x in five Chinese megacities: Providing the isotopic footprint from 2013 to 2014', *Environment International*, 137:105592, doi:10.1016/j.envint.2020.105592.

Zong Z, Wang X, Tian C, Chen Y, Fang Y, Zhang F, Li C, Sun J, Li J, & Zhang G (2017) 'First Assessment of NO_x Sources at a Regional Background Site in North China Using Isotopic Analysis Linked with Modeling', *Environmental Science & Technology*, 51(11):5923–5931, doi:10.1021/acs.est.6b06316.

Research article

The use of dirty fuels by low-income households on the South African Highveld

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Abstract

Meaningful proportions of households on the South African Highveld regularly use energy carriers that result in the emission of significant quantities of particulate and gaseous pollutants. Dirty fuels are mostly used by lower-income households, with the exception of recreational wood use that is also prevalent in higher-income households. The dirty fuel use patterns and trends observed on the Highveld are the result of the unique combination of the utility, accessibility, affordability, availability, and desirability of the energy carriers and equipment, climatological factors, markets and infrastructure, as well as the inertia of historic energy use patterns. There are no systematic reviews and prognosis of the use of dirty fuels by low-income households on the South African Highveld that consider critical recent events such as the Covid pandemic and emerging dynamics such as the just transition movement. In this article we will use a literature review as well as our own research to describe dirty fuel use by low-income households on the Highveld, paying specific attention to changes over time. We will attempt to describe what is being used, who the users are, and for which utilities fuels are being used. From these descriptions, specific patterns emerge that shed light on possible avenues and prospects for ending dirty fuel use on the Highveld.

Keywords

Domestic solid fuel use, domestic coal use, domestic wood use, domestic paraffin use, dirty fuels, Highveld Priority Area, energy use patterns, energy use interventions, air quality, low-income settlements

In this article we will review available information on the use of dirty fuels (coal, wood, paraffin and dung) by low-income households on the Highveld. We focus particularly on coal and wood use in the Highveld Priority Area. We will review changes over time and offer a view on the prospects of ending dirty fuel use on the Highveld.

Background

Short chronology of important events

Significant societal changes occurred on the Mpumalanga Highveld in South Africa and in the world at large since Tyson, Kruger and Louw published their report "*Atmospheric pollution and its implications on the Eastern Transvaal Highveld*" in April 1988 (P. Tyson et al., 1988, see also Tyson et al., 1988).

The Berlin wall fell less than two years later in November 1989 (Langenbacher 2019). The end of the cold war ushered in a series of international and local transformations. South Africa's own political and social transformation gained irrevocable momentum with the release of Nelson Mandela in February 1990 (Mandela 2013). The Rio Earth Summit (formally known as the *United Nations Conference on Environment and Development*) was held in June 1992, where the UNFCCC was adopted (United Nations 1992). After South Africa's first democratic election in April 1994 local governments were re-organised, first into transitional councils and later into the current local municipalities and district municipalities.

The government of national unity implemented the *Reconstruction and Development Programme* (RDP) which intended, inter alia, the construction of a million houses for low-income households (Parliament, 1994). The appearance of these subsidy houses (colloquially known as 'RDP houses') and their accompanying services changed the character of townships on the Mpumalanga Highveld and in South Africa, in general.

The Kyoto Protocol was signed on 11 December 1997 and entered into force in February 2005, after ratification by Russia. The first implementation period took effect from 2008 to 2012. This marked the start of a growing awareness of, and actions to mitigate, anthropogenic climate change.

In the latter part of the 1990's, the then Department of Minerals and Energy (DME) implemented the Low-Smoke Fuel programme aimed at reducing air pollution caused by domestic coal use, particularly in townships. The programme sponsored basic research as well as a macro-scale experiment in Qalabotsha in 1997. At the same time, significant research was undertaken into the health effects of air pollution from domestic sources, notably by Petro Terblanche.

Following the macro-scale experiment, the DME formulated the *Integrated Household Clean Energy Strategy* which envisioned a suite of measures including, behaviour change measures such as improved top-down ignition of coal fires, low-smoke fuels, thermal insulation of houses, as well as cleaner fuels and stoves (Surridge et al. 2005).

On 19 November 1998, President Mandela signed the National Environmental Management (Act 107 of 1998, Government Gazette No. 19519 notice 1540). Within the framework of this act, the National Environment Management: Air Quality Act (NEM:AQA Act 39 of 2004) came into effect in February 2005. This act marked a change from only managing emissions to also managing air quality states. Under the NEM:AQA, National Ambient Air Quality Standards (NAAQS) were instituted and three air quality priority areas have since been declared.

The first priority area to be declared was the Vaal Triangle Airshed Priority Area (VTAPA). It was declared in terms of Section 18(1) of NEM:AQA under Notice No. 365 of 21 April 2006. Clarification that Heidelberg is not included in the VTAPA was later published in Government Gazette No. 30164 of 17 August 2007, Notice 711. The area includes parts of the City of Johannesburg and the Emfuleni, Midvaal and Metsimaholo Local Municipality.

The Highveld Priority Area (HPA) was declared on 23 November 2007 (Government Gazette No. 30518, notice 1123). The area includes the following municipalities: Ekurhuleni Metropolitan Municipality (MM), Lesedi Local Municipality (LM), Govan Mbeki LM, Dipaleseng LM, Lekwa LM, Msukaligwa LM, Pixley ka Seme LM, Delmas LM, Emalahleni LM and Steve Tshwete LM. The HPA will be the area of interest in the analyses that follow.

The Waterberg–Bojanala Priority Area (WBPA) was declared on 15 June 2012 (Government Gazette No. 35435, Notice 495). The WBPA encompasses the Waterberg District Municipality in Limpopo and parts of the Bojanala Platinum District Municipality in the North West Province. The WBPA is unique in that it was declared a priority area based on potential future development, rather than on the air quality prevalent at the time.

On 9 March 2010, minister Buyelwa Sonjica declared a *list of activities which result in atmospheric emissions which have or may have a significant detrimental effect* accompanied by minimum emission standards (MES) for each. (Government Gazette No. 33064, notice 248). The MES took effect on 1 April 2010. Section 5 stipulates that new plant had to comply with the new plant minimum emission standards on the date of publication. Existing plant had to comply with existing plant standards within 5 years and thereafter had another five years to comply with new plant standards (i.e., within 10 years). Section 6 allowed for an application to be made to the National Air Quality Officer for the postponement of the compliance timeframes in Section 5 for an existing plant under certain conditions. Many businesses opted to apply for postponement of the compliance timeframes for existing plant that were to take effect on 1 April 2015. In 2014, 34 such applications were lodged. A further 20 followed between 2015 and 2017 (Khumalo 2018).

From 2010 onwards, the idea of air quality offsets as a viable air quality management tool started to develop (Fischer and Pauw 2010). Eskom conducted an air quality offsets prefeasibility study in 2012 and 2013 (EScience Associates (Pty) Ltd and Nova Institute 2013). Sasol conducted a baseline study in eMbalenhle, Lebohang, eMzinoni and KwaDela together with a pilot implementation in KwaDela between 2013 and 2015. During the pilot implementation, 505 RDP houses were retrofitted with thermal insulation. Indoor temperature and solid fuel use were monitored in a sample of households (Sasol Limited, 2020:83).

DEA was already working on an overarching framework for environmental offsetting in 2013. (Department of Environmental Affairs, 2013). A draft Air Quality Offsets Policy was issued for public comment in January 2014 (Republic of South Africa, Department of Environmental Affairs 2014). After publishing a draft in 2015 (Government Gazette 38894, notice 597), Minister Edna Molewa published the Air Quality Offsets Guideline on 18 March 2016 (Government Gazette 39833, notice 333). From 2015 onwards, the requirement to implement air quality offsets often accompanied the granting of postponements of the compliance time frames for MES. This was the case for Eskom's coal-fired power stations as well as for Sasol's Secunda and Sasolburg operations as well as Natref.

On 25 September 2015, the General Assembly of the United Nations adopted a resolution entitled *Transforming our world: the 2030 Agenda for Sustainable Development*. The core of this resolution are 17 sustainable development goals (SDGs). Included in these and relevant for this analysis are: *Goal 7. Ensure access to affordable, reliable, sustainable and modern energy for all, and Goal 13. Take urgent action to combat climate change and its impacts*. South Africa produced a *SDG Baseline Report* in 2017, followed by a *Country Report* in 2019. In 2017, the reported value for the indicator *Proportion of population with access to electricity* (Indicator 7.1.1) was 95.3 %. It is worth noting that the 2019 General Household Survey specifies that 85 % of South African households were connected to the main electricity grid.

Shortly thereafter, in November and December 2015, the Paris Climate agreement was negotiated by the 196 parties to the United Nations Framework Convention on Climate. The final agreement was signed on 22 April 2016 and became effective on 4 November 2016. Article 2 of the agreement highlights the following objective, "…to strengthen the global response to the threat of climate change, in the context of sustainable development and efforts to eradicate poverty". This includes keeping the increase in the global average temperature to well below 2 °C above pre-industrial levels and to pursuing efforts to limit the temperature increase to 1.5 °C above pre-industrial levels. Enabling adaptation, climate resilience and low greenhouse gas emissions development was well as financing mechanisms for the above are further objectives. The parties will each undertake and communicate ambitious actions to reach these objectives. According to article 4 each party shall prepare, communicate and maintain successive nationally determined contributions (NDC) that it intends to achieve (Republic of South Africa 2015, 2021). In its first NDC (October 2015) South Africa committed to keep national greenhouse gas emissions between 398 and 614 Mt CO_2-eq by 2025 and 2030 and follow a peak, plateau and decline greenhouse gas emissions trajectory with greenhouse gas emissions peaking between 2020 and 2025. At the time of writing, the 2021 submission was open for public comment. It contained a more ambitious mitigation target of limiting annual greenhouse gas emissions to between 398-510 Mt CO_2-eq for the period 2021-2025 and between 398-440 Mt CO_2-eq for the period 2026-2030 (NDC draft p 14). The upper range of the proposed 2030 is 28% below the 2015 NDC targets.

On 15 March 2020, President Ramaphosa declared a national state of disaster in terms of the Disaster Management Act (Act 57 of 2002) following the first confirmed case of coronavirus disease 2019 (COVID-19). A national lockdown took effect on 27 March 2020 which led to a severe economic decline. A series of restrictions has been in effect up to the time of writing. At the time of writing, the official death toll was 87 052.

Demographics

The results of the 2011 census, the most recent national census conducted in South Africa, indicated that the HPA was home to 4.7 million persons or 1.4 million households. Based on historic population growth rates, we estimate the 2021 population at around 5.9 million persons and 2.0 million households.

This population includes vulnerable groups. In 1996, for example, the number of children in the HPA who were younger than five years of age was estimated to be 291 000. This number increased to 326 000 in the 2001 census and then to 470 500 in the 2011 census. At least 100 000 of the latter were less than one year of age at the time of the census. Informed by the General Household Surveys (GHSs) conducted between 2011 and 2019 (no GHS was conducted in 2020 due to the COVID-19 pandemic), the number of children younger than five years of age currently living in the HPA might be as much as 650 000.

In 2019, approximately half of the population (3.1 million people) in the Nkangala and Gert Sibande District Municipalities and the Ekurhuleni Metropolitan Municipality (the three district municipalities that make up the bulk of the HPA) lived below the upper bound poverty line of R 1 227 per person per month in 2019 (Republic of South Africa, Department of Cooperative Governance 2020c, 2020a, 2020b; Statistics South Africa 2021).

The impact of household use of dirty fuels on the Highveld

Environmental effects

There is extensive literature dealing with the effects of domestic solid fuel use on air quality. A bibliography of sources prior to 2008 can be found in Friedl et al. (2008). Notable studies since include Adesina et al. (2020), Chidhindi et al. (2019), Language et al. (2016), Moletsane et al. (2021) Muyemeki et al. (2021). An analysis of long-term trends in the annual average PM_{10}, $PM_{2.5}$ and SO_2 in the HPA using the Theil-Sen trend analysis by Feig et al. (2019) showed a very slight improving trend in annual average concentrations in the long-term over a number of sites in the HPA, including sites where solid fuel use plays a significant role.

Household solid fuel use leads to episodic and localised exceedances of air quality standards, specifically the 24-hour standard for PM_{10} and $PM_{2.5}$. A clear seasonal and diurnal pattern is present. The diurnal particulate concentrations follow the overlap of human activity patterns. Because solid fuel use is closely associated with space heating, higher emissions occur in winter. Combined with poor dispersion conditions in winter, this results in the bulk of exceedances of national ambient air quality standards occurring in winter. A peak in monthly average concentrations occur in July. Hourly averaged concentrations show a bimodal daily pattern with a morning peak at approximately 07:00 and an, often larger, evening peak at around 19:00. The pattern in more articulated in winter. The daily and annual pattern is well illustrated by Hersey et al. (2015).

The typical daily and seasonal pattern as well as the localised nature of the impact of domestic fuel use is demonstrated by Langerman et al. (2018) who analysed the difference in mean hourly PM_{10} concentrations between Hendrina and Kwazamokuhle for January and July 2015. Although the settlements are about three kilometres apart, the difference is stark. The winter diurnal profile for Kwazamokuhle, where solid fuel use is common, shows the characteristic bimodal pattern with a morning peak between 06:00 and 08:00 at about 150 $\mu g.m^{-3}$ and an evening peak between 17:00 and 18:00 in excess of 250 $\mu g.m^{-3}$. In summer the pattern is less articulated with peaks at approximately 50 $\mu g.m^{-3}$ and 75 $\mu g.m^{-3}$ respectively. In comparison, the mean hourly PM_{10} concentrations at Hendrina is much lower. A bimodal pattern is present in winter with peaks at approximately 75 $\mu g.m^{-3}$, but is totally absent in summer.

Source apportionment studies, for example those by Muyemeki et al. (2021) and Walton (2021), shows that domestic coal and wood use contribute proportionally more to fine particulate concentrations than to coarse particles.

Human health effects

The adverse human health effects of air pollution resulting from domestic dirty fuels have been extensively studied. A large body of work resulted from the efforts of Petro Terblanche during the 1990's. Once again, Friedl et al (2008) has an extensive bibliography for sources prior to 2008. In 2010, a study on ambient air quality, potential exposure to air pollution and air-related human health was conducted in KwaGuqa, Mpumalanga (Wright et al. 2011). Since then, two large-scale health studies have been conducted, one in the Vaal Triangle and one in the HPA.

Data sources

This section describes the data sources used in the description of dirty fuel use on the Highveld that is presented in section 3.

Official statistics

Data on dirty fuels are collected by Statistics South Africa as part of the *National Census*, the *General Household Survey* (GHS) and the *Income and Expenditure Survey* (IES).

Three national censuses have been conducted in South Africa to date, since the dawn of our democracy in 1994 – the first in 1996, the second in 2001 and the third in 2011. The National Census collects data on a person as well as household level, but the data are always aggregated to higher levels before publication (in the interest of the preservation of households' anonymity). The lowest level of aggregation on which the data sets are currently available from Statistics South Africa's SuperWEB2 platform is that of electoral ward. An anonymised 10 % sample of each data set is available to the public on household and person level, but the lowest spatial level to which these data sets can be aggregated is district municipality.

The question in the National Census questionnaire that is of primary interest to this study is about the energy carriers that household use for cooking, space heating and lighting. The question reads: "What type of energy/fuel does this household MAINLY use for cooking, heating and lighting?" The response options are electricity, gas, paraffin, wood, coal, candles, animal dung, solar, other and none . 'Candles' is not a valid option as a fuel for cooking or heating, while coal, wood and animal dung are not valid options for lighting.

The General Household Survey is conducted annually, albeit only on a sample of households. Like the National Census, it collects data at household- as well as at person level. The questionnaire used for the survey includes a verbatim copy of the census' question on main energy carriers, allowing cross-survey comparisons. The aggregated data sets of the GHS are, however, currently available to the public only on provincial and stratum ('urban' or 'non-urban') level.

The way in which the question is formulated in the census is poorly suited to the reality of multiple energy carrier use by households. This phenomenon is sometimes referred to as fuel stacking (Langerman 2018).

Significantly more households use solid fuels than is reported in the national census or the GHS. Pauw et al (2013) analysed data from surveys conducted by the Nova Institute in 2011 in 42 subplaces and compared results to the 2011 national census results. The subplaces in Mpumalanga and Gauteng fall within the HPA, the subplaces in the Free State are in the question in the surveys by the Nova Institute did not differentiate between primary and secondary use: It just asked: "Do you use coal in your house". The results are shown in Figure 1. The error bars show the 95 % confidence interval for the survey-based estimation of the proportion of households who use coal. For low-income subplaces where the national census reported coal use to be

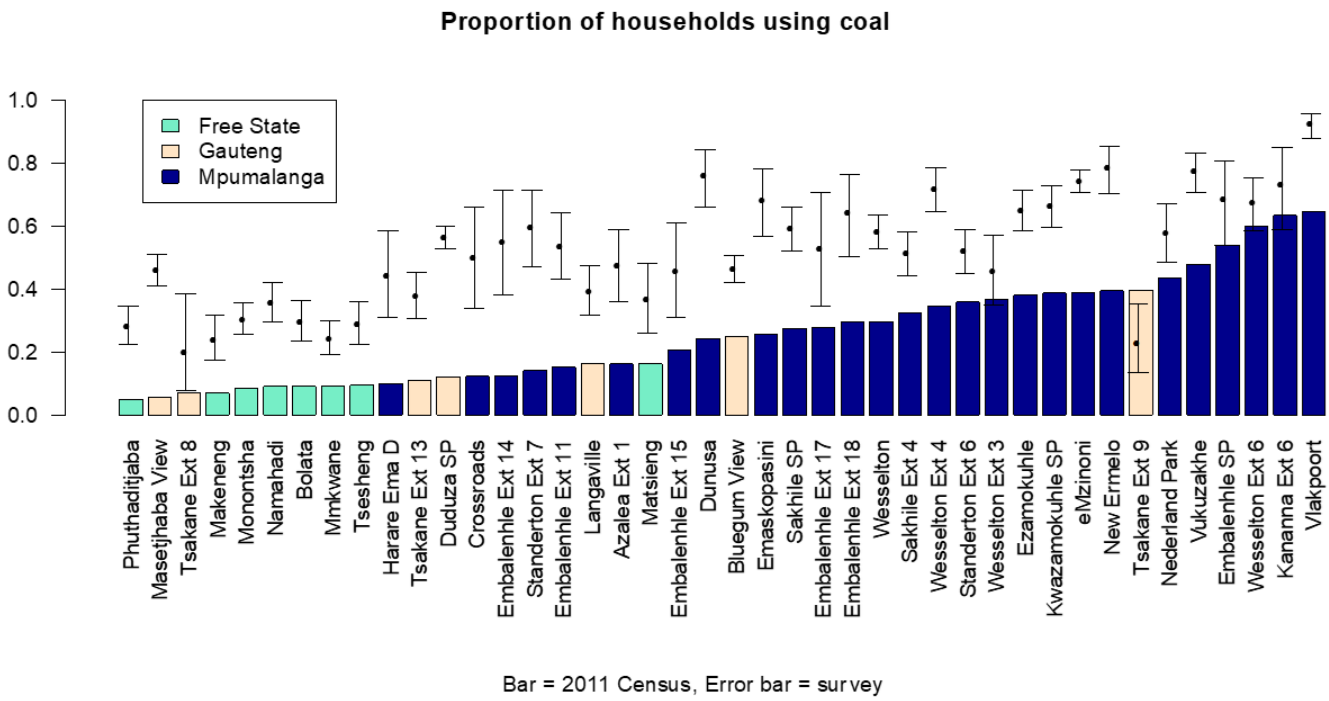

Figure 1: *Comparison of coal use in the 2011 Census with coal use surveys (from Pauw et al 2013)*

scarce, prevalence of coal-using households is systematically under-reported. The degree of under-reporting decreases as the proportion of primary coal users increases, especially above 50 %.

Industry-sponsored research

Sasol Secunda commissioned an extensive baseline assessment for its air quality offsets programme in 2013 in eMbalenhle, eMzinoni, Lebohang and Kwadela. This research included a general household survey as well as a detailed household energy use survey, direct monitoring of household fire making cycles and weighing of fuel.

In Kwadela, Sasol sponsored an experiment that compared two thermal insulation configurations and a reference group in terms of fuel consumption, stove use and end-user satisfaction.

In 2015, the Sasolburg operations of Sasol commissioned a baseline study for its air quality offset programme in Zamdela in the Free State. The study included a general household survey, a detailed household energy use survey, direct monitoring of household fire making cycles and weighing of fuel.

Eskom conducted a pilot study to test interventions to reduce air pollution from domestic coal burning in Kwazamokuhle in 2016. The study included a general household survey, a detailed household energy survey and direct monitoring of household fire making cycles and weighing of fuel in a sample of households. Coal samples from coal merchants in Kwazamokuhle was analysed for energy value, carbon content and ash content.

The study compared the effects of three potential interventions: thermal insulation, an electricity subsidy or a 'smokeless' coal stove. The study design included a control group, and results included fuel consumption, stove use and end-user satisfaction.

Data collected by the Nova Institute

Between 2007 and 2016, the Nova Institute implemented demonstrations of the improved top-down ignition technique (called *Basa Magogo!* or *Basa njengo Magogo*) in selected high coal-use areas in Gauteng, the Free State, Mpumalanga and KwaZulu-Natal. This programme was financed through the selling of verified emission reduction (VERs), mostly Gold Standard VERs. VERs are issued after submission of an annual monitoring report verified by a competent third-party. In total, 87 such reports were submitted and verified, each with a corresponding household and coal merchant survey (accessible on https://register.goldstandard.org). The annual household surveys monitored coal-use stove type and use of the alternative top-down ignition technique in the subplaces where implementation had taken place and where there were indications that there would still be a significant number of users. The coal merchant survey tracked the format in which coal was sold, the price of coal and, in some cases, the mine where the coal was sourced.

Dirty fuel use on the Highveld

Trends in the proportion of dirty fuel users

The proportional of households that use dirty fuels as primary energy carriers for cooking, space heating or lighting has been declining since at least the 1990's, but probably already since the 1980's.

According to the census data all municipalities within the HPA experienced a decrease between 1996 and 2011 in the percentage of households who used dirty fuels as their primary energy carriers for cooking or space heating[1]. The most remarkable decrease in primary dirty fuel use for cooking was seen in the Lekwa Municipality, Mpumalanga, where the percentage of households who mainly used coal, wood or paraffin for cooking dropped from 53.71% in 1996 to 16.69 % in 2011. The smallest decrease occurred in the Msukaligwa Municipality (also in Mpumalanga), which dropped from 60.57 % in 1996 to 47.77 % in 2011.

With regards to space heating, the Lesedi Municipality in Gauteng experienced the largest decrease between 1996 and 2011, with 48.39 % of households primarily using a dirty fuel for space heating in 1996, and less than half of that (20.93 %) in 2011. Msukaligwa, the most easterly of the HPA municipalities, was once again the municipality that experienced the smallest decrease, with the percentage of households who primarily used dirty fuels for heating still fairly high in 2011 at 49.23 %, after dropping from 60.26 % in 1996.

In 1996 the Pixley ka Seme Municipality, the municipality in the HPA extending furthest to the south, topped all the lists - it had the highest percentages of households using mainly dirty fuels for cooking (69.47%), space heating (67.83%) and paraffin for lighting (8.84%). In the other corner of the HPA, the Ekurhuleni Municipality (in Gauteng) – the municipality furthest to the west of the HPA- had the lowest percentages of households who primarily cooked (33.92%) and heated their homes (34.80%) with dirty fuels. However, it only had the third-lowest percentage of households (2.98 %) who used a dirty fuel for lighting. By 2011, the Pixley ka Seme Municipality still had the highest percentage of households who primarily heated their homes with a dirty fuel (50.17 %), but the Msukaligwa Municipality now had the largest percentage of households who primarily cooked with a dirty fuel (47.77 %), while the Ekurhuleni Municipality had the largest percentage of households who used paraffin for lighting (4.61 %). Interestingly, Ekurhuleni is the only HPA municipality who consistently saw an increase across the three census years in the percentage of households who primarily used paraffin for lighting: 2.98 % in 1996, 3.87 % in 2001 and 4.61 % in 2011.

Trends in the number of dirty fuel users

When looking at the absolute numbers of dirty fuel users in the HPA, a very different picture emerges. Ekurhuleni, one of the smaller municipalities in the HPA by area, and one of the only

[1]The response options differed slightly between the three censuses, but not in a manner that affected the use of the data for the present study.

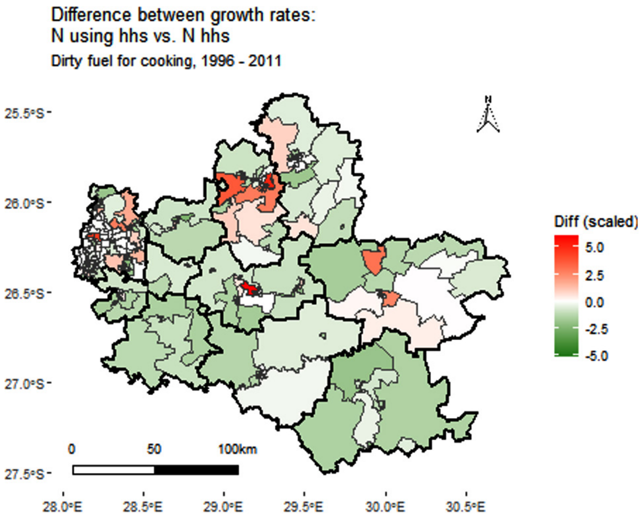

Figure 2: Comparison of growth in dirty fuel use to household growth

two HPA municipalities who form part of the Gauteng province, had vastly more households using dirty fuels than any of the other municipalities in the HPA. In 1996, Ekurhuleni had 184 047 households cooking primarily with dirty fuels, 188 805 households heating their homes primarily with dirty fuels, and 16 183 households using mainly dirty fuels for lighting. The Govan Mbeki Municipality in Mpumalanga - which ranked second on the list of HPA municipalities with the most users of dirty fuel in 1996 - had 25 631 households cooking, 25 248 households space heating, 2 940 households lighting with dirty fuels, i.e. only 13.9 %, 13.4 % and 18.2 % of Ekurhuleni's numbers.

While all municipalities in the HPA saw a decrease between 1996 and 2011 in the percentage of households cooking and space heating with dirty fuels (as discussed earlier), two HPA municipalities (Emalahleni and Msukaligwa) saw an increase in the absolute number of households cooking with dirty fuels, six HPA municipalities had an increase in the absolute number of households heating their homes with dirty fuels (Emalahleni, Msukaligwa, Steve Tshwete, Ekurhuleni, Victor Khanye and Pixley Ka Seme), and two HPA municipalities experienced an increase in the absolute number of households primarily using paraffin for lighting (Ekurhuleni and Emalahleni). At the time of the 2011 census results, however, Ekurhuleni still topped all the lists, with 178 088 households cooking, 189 572 households space heating, and 46 834 households mainly using paraffin for lighting. It is notable that, in absolute terms, dirty fuel use for cooking decreased only slightly, dirty fuel use for heating increased slightly and there was a substantial increase in the number of households who use paraffin for lighting. This indicates a growth in households living in unserviced informal houses.

The increases that these municipalities experienced are largely attributable to sheer population growth. A handful of "hotspots" within each municipality, however, can be identified where the growth rate in the number of households who use dirty fuels as their primary energy carriers (growth rate A) exceeds the growth rate in the absolute number of households in the area (growth rate B). The map below (Figure 2) shows where these hotspots are located in each HPA municipality when looking at dirty fuels as primary energy carriers for cooking. Shades of green indicate areas where growth rate A is slower than growth rate B; the darker the green, the slower A is compared to B - signalling a general move away from the use of dirty fuels as primary energy carrier for cooking. Shades of red indicate areas where growth rate A exceeds growth rate B; the darker the red, the greater A is compared to B - signalling a general move towards the use of dirty fuels as primary energy carrier for cooking.

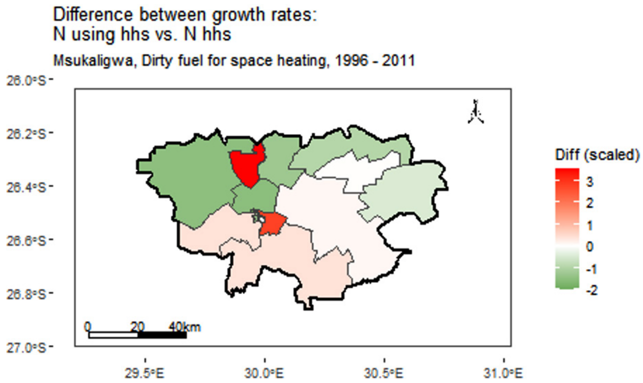

Figure 3: Growth in dirty fuel use and household growth in Msukaligwa

The Msukaligwa Municipality serves as a good example (the easternmost local municipality, demarcated in black, in the HPA in Figure 2, detailed by ward in Figure 3). Seven of the nineteen wards in the municipality had fewer households in 2011 who heated their homes mainly with dirty fuels than in 1996. A further six wards did experience an increase between 1996 and 2011 in the absolute number of dirty fuel space heating households, but the rate of the increase was in each case lower than the household growth rate of the ward in question. In the case of the remaining six wards, however, the number of households primarily heating with dirty fuels grew at a faster rate than the absolute number of households in the ward. This was especially pronounced in two wards: in the ward that encompasses Breyten and surrounds (such as KwaZanele) the number of households heating their homes with coal, wood or paraffin increased from 375 in 1996 to 2379 in 2011 (i.e. growth rate 13.11 % p.a.), while the absolute number of households in the ward increased over this period from 1536 to 4656 (growth rate of 7.66 % p.a.); in the ward that encompasses Ermelo and surrounds (such as Wesselton) the number of dirty fuel space heating households increased from 245 in 1996 to 1479 in 2011 (growth rate of 12.71 % p.a.), but the absolute number of households increased over this period from 857 to only 2379 (i.e. growth rate of 7.04 % p.a.).

Although it is useful to group coal, wood and paraffin together as "dirty fuel" it is important to also be aware of the trends and hotspots related to individual fuels - especially in cases where large-scale switches occurred between them - as the fuels have different impacts on air quality and different health risks associated with them.

On the scale of the HPA as a whole, coal user numbers decreased

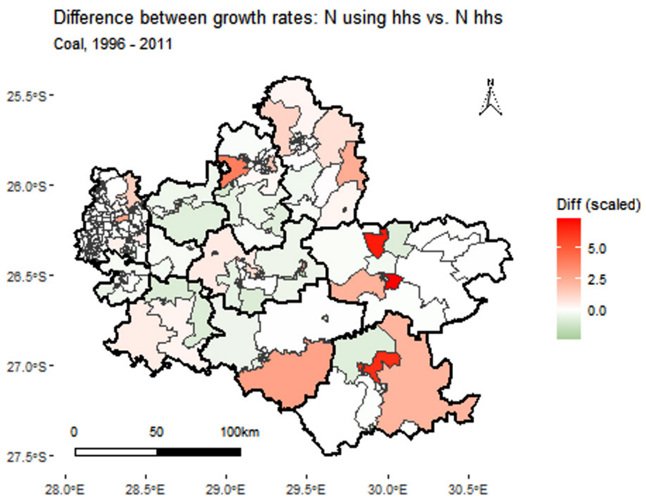

Figure 4: *Household growth and growth in primary coal use*

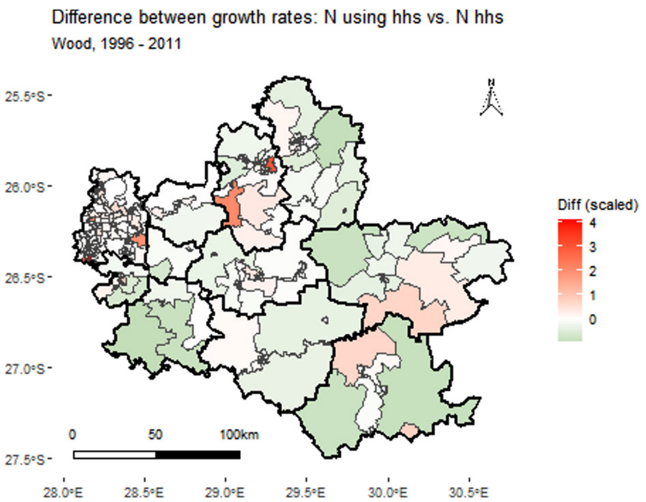

Figure 5: *Household growth and growth in primary wood use*

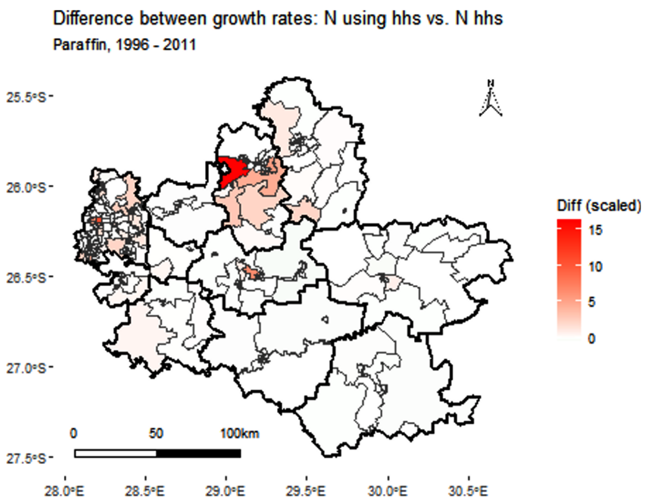

Figure 6: *Household growth and growth in primary paraffin use*

between 1996 and 2011 at a rate of 1.8 % p.a.; paraffin and wood use, on the other hand, increased at rates of 1.7 % and 2.95 % p.a. respectively. Where the majority of households in the HPA who primarily used dirty fuels in 1996 were coal users (62.8 %), the data from the 2011 census indicated that the majority were now paraffin users (66.1 %); coal using households had dropped to 46.8 % of dirty fuel using households, and wood use stood at 17.3 %. This increase in paraffin use is, however, mostly attributable to the dramatic rise in paraffin use in the municipalities of Ekurhuleni, Emalahleni and Steve Tshwete (see Figure 6). When these municipalities are excluded from considerations, it becomes clear that the rest of the HPA actually experienced a decrease in paraffin use and an increase in coal use between 1996 and 2011: at the time of the latter census, 69.5 % of dirty fuel using households in the rest of the HPA were coal users (up from 62.0 % in 1996), while only 16.5 % were paraffin users (down from 21.4 %); wood use experienced a slight decline by 0.03 % p.a. between 1996 and 2011, ending at 24.3 % of dirty fuel using households in 2011.

The most significant increases in coal use (still excluding the Ekurhuleni, Emalahleni and Steve Tshwete municipalities) occurred in specific wards spread throughout Lekwa, Msukaligwa and Pixley Ka Seme, although three of the four remaining municipalities also had at least one ward each where the growth rate in the number of households using coal was higher than the growth rate of the absolute number of households in the ward (Figure 4).

Wood use dynamics appear more complex as the prevalence of wood use decreased significantly in some areas, but also increased significantly in others (Figure 5). In the Lesedi Municipality, for example, one ward saw a negative growth rate among wood using households, while an adjacent ward experienced positive growth.

Although the dramatic uptake of paraffin in Emalahleni, Ekurhuleni and Steve Tshwete overshadow the trends in coal and wood use in these municipalities, it should be noted that some areas within these municipalities indeed also experienced an uptake of coal and wood.

With regard to dirty fuel switches, the census data for at least 13 wards across the HPA show signs of large-scale switches from coal to paraffin and/or wood between 1996 and 2011 (Figure 7). In Ekurhuleni ward 101, for example, the percentage of households using mainly coal for cooking dropped by 29.31 percentage points but that of paraffin increased by 38.58 percentage points; similarly, coal use for space heating decreased in the ward by 32.06 points, but paraffin and wood use increased with a collective total of 25.96 points. Comparable shifts are evident in eight other wards in Ekurhuleni, three wards in Emalahleni and one in Steve Tshwete.

Some wards in the Msukaligwa and Steve Tshwete municipalities seem to have experienced a switch from wood for cooking and space heating to coal and/or paraffin. In ward 3007, for example, the percentage of households who heat their homes primarily with wood decreased by 7.54 points while the percentage of those who use coal for the same task increased by 7.03 points. At least one ward in Lekwa and another in Emalahleni also showed signs of transitions away from wood towards coal or paraffin.

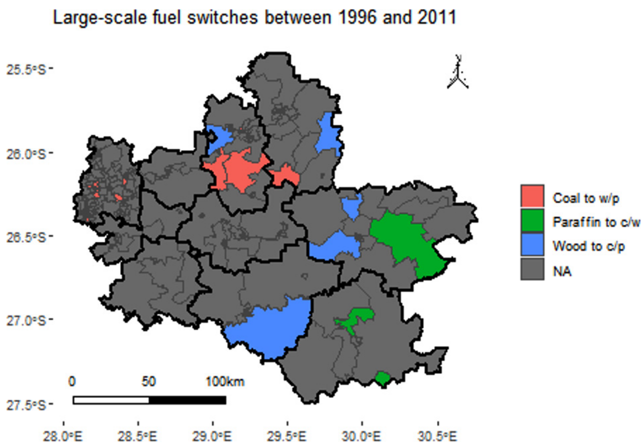

Figure 7: *Fuel switches between 1996 and 2011*

Lastly, at least two wards in the Pixley Ka Seme municipality seem to have had notable proportions of dirty fuel using households switching from paraffin to coal. Ward 4007 is a good example: the percentage of households using coal for space heating rose by 26.97 points between 1996 and 2011, while the percentage for paraffin fell with 24.82 points over the same period.

Fuel types and formats

Bituminous coal is commonly used by households on the highveld. The quality of coal used by households on the highveld varies – even within a single town. As part of the Eskom air quality offset pilot in 2015 in Kwazamokuhle, five bags of coal were obtained from five merchants each. The energy content of the coal was determined according to the ISO 1928 method, ash content according to ISO 1997 and carbon content according to ASTM 5373. The gross calorific value varied between 20.61 MJ/kg and 29.59 MJ/kg, ash content varied between 11.90 % and 34.80 % and the carbon content varied between 43.61 % and 73.30 %.

In all the areas where Nova conducted its coal merchant survey between 2008 and 2016, there were coal merchants who delivered door-to-door (especially large bags), often using tractors, small trucks, bakkies or, in some areas, horse-drawn carts. Coal is typically sold in large bags weighing between 50 kg and 70 kg, smaller bags (35 kg – 50 kg) or measured out in 20 L 'tins' (~17 kg). In Wesselton, Msugaligwa, we found coal being measured in drums of between 68 kg and 73.6 kg.

A small amount of paper, typically newspaper, and about a kilogram of wood is commonly used to ignite coal.

In urban areas, wood is often sold by the same merchants who sell coal or by entrepreneurs who obtain wood on farms or elsewhere and deliver bakkie-loads to households. Industrial waste wood, such as used shipping pallets are frequently sold by coal merchants (either chopped or whole). Although the Highveld is a grassland, there are areas with local wood sources that are harvested by households. Two examples that we have observed is at Tembisa, where Eucalyptus trees to the north of the township are exploited, and Bioketlong in Emfuleni, where a sudden expansion of informal houses led to cutting of green wood in the adjacent hills. It is here where we encountered households who mixed used motor oil with the green wood to make it burn better.

Wood ignites easier than coal and provides heat for cooking relatively fast. Wood can function as a substitute product for coal since wood can be burned in the same devices and provide the same utilities. Coal is the superior fuel for space heating and bulk water heating due to its higher energy value and the fact that a coal fire can burn much longer before it needs to be refuelled compared to wood. We observed a degree of substitution of wood for coal in Zamdela, Sharpeville, Bophelong and Boipatong. There was a steady increase in the price of coal in the Vaal Triangle between 2007 and 2014. This was accompanied by a decrease in the proportion of households who used coal and an increase in the proportion of households who use wood.

We observed an example of the substitution of wood for coal in Namahadi (near Frankfort, Free State). The fuel use pattern changed drastically when a large stock of eucalyptus wood became available, apparently as a result of nearby land-clearing. The wood was sold in Namahadi at competitive prices and large numbers of former coal users switched to using wood while it remained cheaply available.

Fuel burning devices

Three broad classes of fuel burning devices are commonly used to burn coal on the Highveld: cast iron stoves, locally made welded stoves and braziers (izimbaula). These devices are used for wood, but wood can also be used in an open fire on the ground (sometimes referred to in literature as a 'three stone fire') or in a open braai [barbecue] (which is occasionally used with coal as well).

Cast iron stoves are generally large stoves with four to six plates. The stoves currently still in use are generally old because the last large local manufacturer, Falkirk, closed their Newcastle plant at the end of the previous century.

Welded stoves come in a variety of forms and vary by region because they are made by local craftsman, sometimes from waste metal (off-cuts) obtained from local industries. These stoves are typically smaller than cast iron stoves and vary in sophistication of design.

Braziers are typically made from a 20-litre mild steel drum. The fact that paint is increasingly sold in plastic containers, means that such items are considerably harder to come by at zero cost than they were two decades ago. In the studies that Nova conducted between 2007 and 2017, the proportion of coal using households who used braziers was consistently lower than 10 %.

Wood use (without coal), specifically for thermal comfort in winter, are less common on the highveld. The proportion of wood users are highest where wood is cheaply or freely available, typically more in rural than urban or semi-urban settings. The same devices used for coal are also used indoors with wood, but wood may also be used in an open fire on the ground outside, in an outside kitchen or in a fireplace inside the house.

Makonese et al. (2017) determined emission factors of domestic coal-burning braziers under a variety of conditions in the laboratory. Nkosi at al. (2018) determined fine particulate matter emission factors from residential burning of solid fuels using traditional cast-iron coal stoves in field conditions. Both groups of authors found that emission factors vary. Makonese et al. found that the emissions varied according to fuel stacking and ignition technique (top-lit updraft [TLUD] or bottom-lit updraft [BLUD]), as well as ventilation (i.e. the density of holes in the brazier), The lowest $PM_{2.5}$ emission factor obtained was 0.3 g/MJ for the TLUD fire in a highly ventilated brazier. The highest $PM_{2.5}$ emission factor was 2.5 g/MJ for the BLUD fire in a brazier categorised as having low ventilation. Masondo et al. (2016) found that in addition to (stacking and) ignition methods and ventilation rates, the size of coal pieces influence emissions from brazier. They concluded that, in general, particulate and CO emission factors increase with an increase in the mean size of the coal pieces.

Nkosi found fine particulate emission factors ranging 6.8 $g.kg^{-1}$ and 13.5 $g.kg^{-1}$. The emission profiles varied depending on stove operation. Fire poking and refuelling lead to an increase in emissions. Like in other similar tests, emissions peaked shortly after ignition.

Utilities

Solid fuels are preferred for space heating rather than for cooking. This is visible by comparing the proportion of households who use coal and wood for space heating and cooking in all the Censuses (1996, 2001, 2011), General Household Surveys (between 2008 and 2020), and several surveys done by Nova.

Like space heating, solid fuels are also well suited for water heating. This is especially true of coal that has high energy content and burns for a long time. The fact that coal fires take long to start burning well and generate useful heat, makes coal less suited for cooking alone. In a coal stove, especially the cast iron variety, the thermal inertia of the device also contributes to the delay in the availability of useful heat to the pot. Wood fires ignite easier, providing flames under the plate much earlier than a coal fire, but wood burns out quicker.

For a simple task like boiling water for tea, electricity, LPG and paraffin are all cheaper and faster than both wood or coal (Graham and Dutkiewicz 1999). Cast iron coal stoves typically have four to six plates, meaning that it becomes efficient when cooking a large meal, or dishes that needs to cook for a long time (like samp [roughly cracked corn kernels]).

When energy is required for a combination of space heating for thermal comfort, water heating and cooking, at the same time, using a coal stove is very effective and efficient. This combined utility is the main reason for the strong seasonal use of coal, observed in most of the highveld household surveys.

Solid fuel use also has a social dimension, where families or friends often sit around the stove or the fire. Higher income groups also use solid fuels in this way. When Nova started work on energy in the 1990s, the stove and fire in general played an important part in bringing the family together in the evenings. We have in recent years observed a shift away from the gathering around coal stoves, as the hub of socialisation for the family in the evening has shifted towards the television in a number of highveld communities where Nova has been doing measurements.

Using solid fuels comes with certain trade-offs. Wood needs to be chopped and stacked, coal and wood take up space and need to be stored in a dry place, coal dirties the hands and solid fuels smoke, especially during ignition. In addition, wood and coal can leave a smell on one's clothes and ash needs to be cleaned out and disposed of. Some of these apparent disutilities are not necessarily experienced by end-users as such, for example, in surveys Nova recently conducted in more traditional wood using areas in Limpopo, households have mentioned that smoke is medicine to the eyes and a sign to the ancestors that the family is still alive. In other instances, our contention is that households accept disutilities with the utilities they require within the framework of what is economically, materially and symbolically possible for them.

Spatial distribution

The spatial distribution of solid fuel use on the Mpumalanga highveld is strongly determined by income distribution, climate and availability of fuels.

The role that income plays can be seen by comparing the prevalence of coal use in neighbouring areas differentiated by income. The results of the typical apartheid town planning (town -township) still persists throughout the region. The historically white towns have higher mean incomes and lower proportions of coal using households than that of township(s). It can also be seen inside the large urban townships in Gauteng such as Soweto, Tembisa, Tsakane where higher income areas have lower coal use. High-income households generally use clean modern energy carriers or sophisticated devices (such as high-end solid fuel heaters). Households with limited income appear to optimise the balance between utility, cost and trade-offs (such as pollution). The poorest (no income) households tend to make use of free energy sources (collecting wood, burning waste) or are forced to forego the utility.

The spatial distribution of solid fuel use is not only determined by income, but also by the availability of energy carriers. On the Highveld there is also clear urban – rural pattern in the distribution of solid fuel use. Coal tends to be used in low-income towns and urban areas, while wood (and sometimes dung) is used for similar utilities on farms and in more rural settings.

Coal use is highly dependent on proximity to coal mines because of the rapid increase in transport cost with increased distance.

A comparison of coal and paraffin use in the towns of the Free State clearly demonstrates this. The townships adjacent to the towns closest to the coal mines in Mpumalanga (Villiers, Frankfort, Vrede) still had significant coal use for heating in the 2011 census while the same socio-economic profile in the towns immediately adjacent (e.g., Heilbron, Petrus Steyn and Warden) had more paraffin use but virtually no coal use. The line representing cost-utility parity between paraffin and coal for space heating lies somewhere between Frankfort and Petrus Steyn. The same phenomenon, that paraffin is used for the same utilities as coal by the same socio-economic group is also apparent from a comparison of the East- and West Rand.

On the eastern side of the Mpumalanga coal fields, there is a similar line where cost-efficiency of wood use for space heating supersedes that of coal use, presumably due to the increase availability of wood from the plantations in the area.

The third important determinant of solid fuel use is minimum temperature. Spatially and temporally, solid fuel use correlates with low temperatures (Annegarn and Sithole 1999). Solid fuels are used more in application such as where bulk heat is needed. Coal is particularly suited to providing heat over a long time. The impact of minimum temperature may explain the difference in coal use between Mamelodi (Tshwane) and Tsakane (Ekurhuleni) where coal use is much more common. Mamelodi and Tsakane are both relatively close to the coal mine at Delmas, but Mamelodi is generally warmer than the higher-lying areas to its south where Tsakane is located. Very low winter temperatures explains why there is high coal use in the Maluti a Phofung Municipality despite its distance from the coal mines.

Temporal pattern

Predictably, solid fuel use has a marked seasonal pattern corresponding to its main uses for combined space heating, water heating and cooking with higher usage during winter, compared to summer. There are households who also cook with coal and wood in summer. Ownership of coal stoves and a certain behavioural inertia likely play a role here but often such households either do not have access to cheaper alternatives such as electricity or get their fuel cheap or at no cost (often wood).

Temporal profiles of stove use have been derived from data collected by Nova and North West University in eMbalenhle, Kwadela, Kwazamokuhle and Zamdela (Nkosi et al. 2017, 2018). The temporal stove use profile is typically bimodal or even trimodal with a morning peak at around 08:00, sometimes a slight increase in activity around 12:00 (representing cooking, especially on weekends) and an afternoon peek starting at around 16:00 and peaking at 18:00. The afternoon peak is generally higher since many households use electricity for warm water for bathing (typically from a kettle) and cooking in the morning when there is time pressure for household members who must go to school and work. In the afternoons, a coal stove (and to a lesser extent, a wood stove as well) provides enduring space heating, warm water for washing and heat of cooking. On cold nights it is not uncommon for households to refill the coal stove later in the evening (after 20:00).

Income and dirty fuel use

Coal and wood use differ in sensitivity to income. One needs some form of income to use coal or paraffin, but wood can sometimes be obtained for free.

Very high-income households can use clean energy sources for even energy intensive applications like space heating and water heating. As income declines, households switch to more cost-effective energy carriers which have more trade-offs for the most energy intensive applications (in the first place heating, but later also cooking). When income is extremely low, households have no choice but to either forego the utility (such as not to use any form of space heating) or to use very low-cost or zero-cost energy carriers which may have significant trade-offs.

There are very few cases where coal could be gathered for free. Coal is used by households with some discretionary income. Coal use for heating first increases slightly over the lower income deciles and then decreases thereafter. This means that an increase in income for the lowest income households living in the coal using areas will initially lead to an increase in coal use before it will lead to a decrease.

The proportion of households in South Africa that uses wood for space heating does not differ much by income decile in the first four income deciles but decreases as income rises above the fourth income decile (Friedl et al., 2008:20 with reference to Statistic South Africa 2006).

Electricity and dirty fuel use

Most dirty fuel using households in the HPA also use electricity. Households mix energy carriers based on considerations of availability, utility and cost. Solid fuels are cost-efficient sources of bulk heat. Coal stoves simultaneously function as cookstoves, heaters and geysers and can provide heat for hours on end. This utility is difficult to replace with separate electrical appliances in a cost-effective way, especially in low-income contexts where the bulk of houses often lacks proper thermal insulation to maintain thermal comfort. This utility is an important reason for the strong seasonal pattern in especially coal use seen in the HPA and elsewhere over the Highveld.

Dirty fuels are also used as a backup energy carrier where a preferred energy carrier fails, for example during power interruptions.

Association with structure type

Most solid fuel using households live in formal houses. However, analysis of the data from the national census and the GHS confirms the association between household dirty fuel use and structure type. Dirty fuel use is especially associated with free-standing informal houses and to a lesser extent with backyard shacks (informal structures on a formal stand) although dirty fuel use is by no means associated with informal houses only (see for example the most recent GHS, Statistics South Africa, 2020:141).

Paraffin use for lighting is a good indicator for absence of electricity since electricity is the most convenient, effective and cheapest energy carrier for lighting. Households who use paraffin for lighting can safely be assumed to be unelectrified. For obvious reasons this occurs more frequently in informal settlements.

The limited thermal protection offered by the structures themselves and the absence of alternatives mean that dirty fuel use will persist in unelectrified informal settlements in cold areas until either the structures themselves or the energy options available to residents changes.

Ending dirty fuel use on the Highveld

Historic attempts

An early example of a command-and-control approach to reducing air pollution from coal use in townships was the legislation to outlaw coal stoves not fitted with a secondary combustion chamber to ensure clean burning (Van Niekerk et al. 1999). At the time there were few stoves on the market (such as the Moderna) that were designed as smokeless stoves. The design of most stoves, such as those manufactured by Falkirk, were essentially not changed (see Ndebele Stoves 2022 for examples). These stoves were just fitted with a divider brick to divide the firebox in two and create a secondary combustion chamber where the smoke was burned off. These stoves complied with the law at the point when they left the factory, but due to the divider brick burning through fairly quickly or users actively removing the divider to enable them to load more coal, most stoves in use were not smokeless (Van Niekerk et al. 1999).

The Department of Minerals and Energy (DME) embarked on a *Low smoke fuels programme* after 1994 (Surridge et al. 2005). The objective of the program was to reduce pollution from township coal use by introducing low smoke fuels to replace coal. The programme generated significant research results, the most prominent being the Qalabotjha Low-Smoke Fuels Macro-Scale Experiment where three low-smoke fuels were tested during the winter of 1997 and it was found that the use of low-smoke fuels led to a significant improvement to the air quality (Engelbrecht et al. 1998). DME formulated an Integrated Household Clean Energy Strategy in which top-down ignition, low-smoke fuels, housing insulation and cleaner fuels and stoves had a place (Surridge et al. 2005).

The improved top-down ignition method was first developed in eMbalenhle by the Nova Institute and members of the community in a project sponsored by Sasol. The first successful implementations was undertaken by Sasol in eMbalenhle, Mpumalanga, in 2001 and 2003 and later in Zamdela, Free State, in 2003 (Wagner et al. 2005). Two large, government (DME) sponsored implementations took place in Gauteng: in Orange Farm in 2003 (Le Roux et al. 2009) and in Tembisa in 2005 (Palmer Development Consulting (Pty) Ltd 2005). All these projects employed a similar methodology of using demonstrations of the technique as the main form of dissemination although the government-sponsored projects tended to favour larger scale demonstrations. The Nova Institute used carbon finance to launch its Highveld Air Quality programme in 2007. In 2008, the (then) DEAT launched the Clean Fires Campaign (VTAPA AQM, 2008), which relied on mass media as a means of dissemination of awareness of the technique, presumably based on the assumption that this will lead to adoption of the technique. The mass media approach was not successful, and the campaign was abandoned after 2011.

One relatively recent development that mobilised new resources toward reducing air pollution from dirty fuel use is air quality offsets(Department of Environmental Affairs 2016; Langerman et al. 2018).

As far as we could determine, only one large air quality offsets programme have been successfully completed for the period 2015 to 2020, namely that of Sasol (with separate projects in Secunda and Sasolburg). Sasol successfully implemented a thermal insulation and stove swop intervention in eMbalenhle and Lebohang as well as a waste removal intervention in Zamdela. These were supplemented by smaller interventions aimed at air quality awareness and reducing veld fires (Sasol Limited 2020).

Eskom successfully completed baseline and preparation phases (Langerman et al. 2018), but failed to timeously implement its air quality offsets programme by 1 April 2020.

A further development is the *Strategy to address air pollution in dense low-income settlements* published in May 2019 by the then minister of Environmental Affairs, Nomvula Mokonyane (Government Gazette No. 42464). The goal of the strategy is to “map out the path that the country needs to take in reducing the impact of air pollution in dense low-income settlements” (Republic of South Africa 2019:1). The strategy sets three objectives: (1) ensuring that efforts to address air pollution in dense low-income settlements are undertaken in a coordinated and coherent manner, (2) facilitating through the National Coordinating Committee on Residential Air Pollution, the implementation of interventions aimed at reducing emissions from dense low-income settlements, and (3) ensuring continued monitoring, evaluation and reporting on the successes and failures of the proposed interventions and on air quality improvements. The strategy concludes with the statement: “It is important to note that the objectives of this strategy can only be achieved if there is an uncompromised coordination between the relevant national departments (DEA, DHS, DoE and DoH) together with the relevant provincial departments and municipalities” (Republic of South Africa 2019:40).

Determinants

The DANIDA-sponsored report on *Air Pollution in dense, low-income settlements in South Africa* (Friedl et al., 2008:265) identified factors that will determine the future trends in dirty fuel use namely fuel prices (since transport is an important determinant of coal prices), the rate of formal housing delivery compared to household formation, the electricity supply prospects and the economic growth rate in real terms.

Since the writing of that report in 2008, the price of petrol have increased from R7.18 (Unleaded 93, inland price) to R18.15 per litre in 2021 (https://aa.co.za/fuel-pricing), i.e. at approximately 6.38 % p.a..

In 2019, the General Household Survey showed that 12.7 %, or 2.1 million out of 17.2 million households, in South Africa lived in informal structures only (Statistics South Africa 2020). The proportions were higher than the national average in the cities of Johannesburg (19.1 %) and Ekurhuleni (18.4 %). While construction of subsidised houses by government is ongoing (the percentage of households that received any kind of government housing subsidy increased from 9.4% in 2009 to 18.7 % in 2019), there are no signs that informal settlements and backyard shacks as a feature of the South African landscape is disappearing. A stark reminder of this fact is found in the 2019 General Household Survey report where it is noted that compared to 2002 "…the percentage of households with access to mains electricity actually declined in Gauteng" by 10.6 % (Statistics South Africa, 2020:34).

Electricity prices have increased from 24.97 c/kWh in 2008 to 133.64 c/kWh as of 2021. The electricity supply remains under pressure with extensive load shedding still occurring during the first half of 2021 (Wright, 2021).

Economic growth remain low. Data from the World Bank (World Bank 2021) show that GDP per capita expressed in constant local currency units was the same in 2008 and in 2018. It declined in 2019, to the lowest levels in nine years lower (i.e., lowest level since the global financial crisis) and plummeted in 2020 after the outbreak of the SARS-CoV-2 pandemic.

A strategy to address dirty fuel use on the Highveld will have to take into account these economic realities, and cannot be built on the fantasy that the situation is not what it is.

Furthermore, new challenges have emerged that impact on the economic growth rate and as such, could also directly or indirectly impact the number of solid fuel users and quantities of solid fuel use on the Highveld in the years to come. Three macro phenomena worth mentioning are the COVID-19 pandemic, climate change and the drive to green the economy in a manner that also takes cognisance of the socio-economic realities of South Africa, also known as the *just transition* movement. It also remains to be seen how the Highveld will be impacted by the fourth and fifth industrial revolutions.

Economic development

The National Development Plan (NDP) identifies poverty and inequality, together with unemployment, as the triple challenge that is to be overcome by 2030 (World Bank, :xii). The NDP set the target to eradicate poverty (lower bound poverty line) by 2030. However, although the depth and severity of poverty has decreased overall between 2006 and 2015, it has worsened again from 2015 to 2020 (NDA, :8).

It is not yet clear what the long term impact of COVID-19 and the resulting lockdowns will be, but it can be expected that it will set back economic development markedly (International Food Policy Research Institute 2020).

It is evident that growth in per capita income of lower earning income households will be a crucial determinant for the future of dirty fuel use. For the poorest households, who out of desperation forego energy use (e.g. who live in cold areas but use no energy carrier for heating) or who use a combination of purchased and collected biomass and waste, an increase in income will mean an increase in dirty fuel use (especially coal and paraffin). For households with a somewhat higher income, increased income will give them the ability to move away from daily use of dirty fuels towards electricity or possibly LPG.

On the other hand, households that have to cope with reduced income could be obliged to revert back to dirty fuels as the cheapest viable option for space heating, especially.

Several communities in the Highveld will furthermore be impacted by South Africa's transition to a greener economy that includes the closure of several power stations in the next decade.

It has been clear for a number of years that the relative importance of coal mining is waning.

Ideological and policy developments

Ideology underlies policy and policies, to some extent, determines the actions of collective entities such as states and corporations. Ideological trends are therefore key determinants, not of what households do, but of how governments and corporations act with regards to households. Two interrelated ideological trends that are likely to influence action towards dirty fuel using households are the ideas of a *just energy transition* and the *end of coal*.

Velicu and Barca observe that labour and environmental justice organisations have different takes of the definition of justice: "The climate-justice movement has emphasized the values of self-determination through grassroots control over the use of resources, food sovereignty, energy democracy, reduction of overconsumption, recognition of climate debt, and respect for indigenous and peasant rights…Labour organisations, instead, have maintained a commitment to the green growth agenda as an unquestioned path toward a post-carbon society" (Velicu and Barca 2020:263).

The Groundworks report *The Destruction of the Highveld, Part 2: Burning Coal*, is an example of the former approach. In chapter 4, Hallows and Munnik (2017) propose a number of starting points for a more equal and ecologically sustainable economy. Their list includes: a new energy system based on socially owned renewables; new jobs in renewables; large scale restoration and detoxification of ecosystems injured by the fossil fuel economy on the Highveld; a new and healthier food economy; healthier and climate-wise housing; a new and healthier transport economy; a reorientation and expansion of municipal services; a basic income grant for all.

The shift from coal to renewable energy for electricity generation is a priority for the South African government. This is embedded in a broader movement away from fossil. The move away from coal is a starting point in South Africa (Halsey et al. 2019:1).

The transformative just energy transition sees the energy transition as a core part of the wider transformation of society (Halsey et al. 2019:6).

In July 2021, the Presidential Climate Commission hosted a high-level discussion on South Africa's Just Energy Transition, with a focus on socio-economic impacts, technology choices, and options for financing the transition (https://www.climatecommission.org.za/programs).

Ending solid fuel use on the Highveld

Considering the historic trends and considerations explicated in the preceding sections, the question arises as to what actions can be taken to end dirty fuel use in the Highveld.

It may be easier to start by expressing what is not likely to succeed: waiting for the poorest sectors of society to become richer, or for universal electricity provision at affordable prices, or for the disappearance of informal settlements will not work in the medium term. Economic development, universal electricity access and universal attainment of formal housing are doubtlessly important national priorities, but, judging by current trends, is a task that will take decades to complete.

Interventions to make coal illegal or hard to come by are bound to fail because households have fallen back to biomass where coal availability decreased, or where prices increased. If coal is unavailable, it will be replaced by wood and waste if need be.

What is needed is a series of solutions that are uncorrelated to national macro-economic trends, service delivery and infrastructure provision and do not rely on coercion or measures that limit the options of households.

We are of the opinion that dirty fuel use on the Highveld will end when households can be provided with clean alternatives that provide equivalent or better utility compared to what they currently derive from dirty fuel use at an equivalent or lower price relative to their income. The fact that solid fuels are used for energy-intensive applications such as space heating and water heating makes this a difficult task. This is because measures will likely be expensive. If improved stoves are to be introduced, these cannot be relatively cheap one-plate rocket stoves because the utility that must be replaced is multi-pots cooking as well as space heating and water heating, preferably all at the same time. An improved stove solution will necessarily imply a large, durable and therefore expensive stove.

An electricity subsidy did not prove to be an effective means to induce households to decrease their coal consumption (Eskom pilot mod 3 p21). This may be due to the income effect: a subsidy is a form of income and an increase in income leads to an increase in consumption of all normal goods. A subsidy does not change the economics of domestic energy use: electricity remains more expensive than coal for space heating. Households who receive an electricity subsidy will therefore only replace another energy carrier with electricity when the subsidy covers their current total electricity consumption, which in this case was approximately R400 (Langerman et al., 2015:3), as well as the additional expenditure of the energy carrier to be replaced.

Solutions based on provision of alternative fuels have not had historic success at scale and face the almost insurmountable challenge of a having to provide a perpetual fuel subsidy.

Thermal insulation is effective in reducing the need for space heating but is relatively expensive to retrofit (compared to the initial cost of the structure) and is currently only applicable to formal houses. The improved building standards implemented in recent *Breaking New Ground* (BNG) houses that also forms part of the *Strategy to combat air pollution in dense, low-income settlements* means that the problem of very poor thermal performance of houses will not be perpetuated in future (Republic of South Africa, Department of Environmental Affairs 2019).

Another limitation that is likely to become more important in future is the increasing resistance to the promotion of fossil fuel use, particularly to coal use. The increasing action to combat climate change and the movement to end coal use however presents an opportunity that can be exploited since this generates new financing avenues. The carbon tax regulations allow for only 10 % of the carbon tax liability to be offset with certified/verified emission reductions generated under the Clean Development Mechanism (CDM) of the Kyoto Protocol, Verra or Gold Standard. The carbon tax liability is currently relatively low at R127/t CO_2 emissions and will increase at 2 % above inflation until 2022 whereafter it will increase with inflation (South Africa Revenue Service 2021).

There are historic successes with using climate finance to address air quality on the Highveld. Nova generated 200 000 Gold Standard VERs between 2010 and 2016 with our programme to promote the alternative top-down ignition technique. The expansion of the programme was discontinued because the price of Gold Standard VERs did not justify further implementation. Top-down ignition is by far the cheapest intervention to reduce emissions from domestic coal use (Airshed Planning Professionals and Bentley West Management Consultants 2004). This remains true when the waning impact over time, due to reversion to polluting bottom-up ignition, is taken into consideration. Improved top-down ignition could scarcely be financed through climate finance. More costly interventions, like improved stoves and thermal insulation, cannot be funded through climate finance at current VER prices.

Air quality offsets have proven to be an effective policy tool in mobilising significant resources to combat air pollution from solid fuel use. Sasol Secunda has successfully implemented a durable intervention (LPG stove swop and thermal insulation) at the level of a large township (eMbalenhle), practically to saturation (i.e. all reachable households who are technically eligible have been given a chance to participate).

By 1 April 2020, nothing has come of Eskom's implementation of air quality offsets apart from a pilot project. This failure represents an important missed opportunity as well as a dangerous precedent.

In conclusion thus far: The permanent end of dirty fuel use is inherently linked to macroeconomic and societal dynamics that evolve on long timescales and where the prospects of large improvements in the short term appears slim. There are successful interventions to address coal use in the short term (top-down ignition) and long term (LPG stove swop and thermal insulation). There are also successful funding channels namely greenhouse gas offsets and air quality offsets. Air quality offsets have the potential to mobilise orders of magnitude more resources per project.

There is currently no implementation-ready intervention for households in informal houses who use solid fuels for space heating and water heating. We have shown that this is the category where solid fuel use is particularly persistent. The increase in paraffin use for lighting in Ekurhuleni between 1996 and 2011, shows that there are important areas where the number of households in informal, unserviced houses are growing, both in absolute terms and proportionally. Since informal structures are often not legal, nor meant to be permanent and vary greatly in size, design and material, passive measures focussing of the structure itself are likely to remain difficult. This implies that an active source of bulk thermal energy is the only viable avenue that can be pursued in the short term while the long-term political and economic solutions (hopefully) materialise.

Air quality offsets remain a viable funding mechanism but will depend on proximity to industries applying for new plant licences or for postponements for compliance to MES.

If climate finance is going to play a part, the (yet to be developed) active energy solution for informal houses must use renewable biomass as a fuel. Even if that is the case, the highest conceivable estimate for income for carbon credits is still less than R1000 per household p.a. (assuming heavy use of coal: a 50 kg bag of coal/week all year; 2.35t CO_2 per tonne coal; $10 per t$CO_2$). Households who use a bag of coal per week throughout the year are very rare. This means that for all practical purposes, climate finance will not enable a systematic eradication of dirty solid fuel use in informal households if the technology costs more than approximately R1500 (assuming R500 transaction cost and a three-year payback period).

What can be pursued in the short term are more modest but achievable goals. Where unserviced households use solid fuels for cooking, effective and affordable biomass or LPG cookstoves can be introduced and possibly financed with climate finance. Replacing the cooking utility with a clean-burning device will at least reduce emissions during specific seasons and times of the day when space heating is not required. Increasing the offsetable portion of the carbon tax to 100 % will greatly enhance the demand for carbon credits from South Africa but the number of projects will be limited by the low absolute value of the carbon tax liability.

Horizons

We can identify three directions from which potential new solutions may emerge: institutional innovation, new biomass and smart subsidies.

The end of informal settlements may be brought about by institutional innovation to enable large-scale urban land reform. Huchzermeyer et al. (2019) concluded that much of policies needed for urban land reform already exists but needs to be extended and put into practice. How that difficult task is to be undertaken in beyond the scope of this article, except to note that the strong association between dirty fuel use and informal housing means that a solution to formalise informal settlements will also to a large degree lead to a reduction in domestic solid fuel use.

Micro-scale biomass gasification for household cooking is a relatively new development with the first commercial units released in 2003 (Roth, 2011). The technology offers vast improvements over open fires and even over improved biomass cookstoves but maintaining control over the gasification process becomes more difficult as the device gets smaller. A technological refinement that produces a robust but affordable micro-gasification cookstove and water heater or even space heater may go a long way to replace the utility provided by coal and wood. If the fuel is from a renewable source and the char can be collected and sequestered, carbon finance (both from emission avoidance as well as carbon sequestration) may be available.

The other technological horizon that may yield new avenues for interventions are smart subsidies. Unlike a general electricity subsidy, that fails in theory and failed in practice, during Eskom's evaluation in Kwazamokuhle (Langerman et al. 2015), a smart subsidy targets the specific device, like a heater. There are examples of peer-to-peer electricity trading networks that leverage blockchain technology (like Solar Bankers, https://

solar-bankers.medium.com). It is conceivable that a similar technology can be used to subsidise the electricity consumption of a specific device (such as a space heater or a water heater) meant to replace the utility of a solid fuel burning device that is cheaper to operate (but not for anything else).

There are persistent and, in some cases, growing pockets of households that use dirty fuels on the Highveld but there are also interventions and financing mechanisms that are effective to some extent. There are also technologies that may develop in future that may accelerate the movement away from solid fuel use. However, the enduring end of dirty solid fuel use will come from large societal transformations related to income, acceleration of formalisation through land rights and provision of services.

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References

Adesina JA, Piketh SJ, Qhekwana M, Burger R, Language B, & Mkhatshwa G (2020) 'Contrasting indoor and ambient particulate matter concentrations and thermal comfort in coal and non-coal burning households at South Africa Highveld', *Science of The Total Environment*, 699:134403, doi:10.1016/j.scitotenv.2019.134403.

Airshed Planning Professionals & Bentley West Management Consultants (2004) *Study to examine the potential socio-economic impact of measures to reduce air pollution from combustion*. Final report, Trade and Industry Chamber / Fund for Research into Industrial Development, Growth and Equity (FRIDGE), Johannesburg.

Annegarn HJ & Sithole JS (1999) *Soweto air monitoring - project SAM: trend analysis of particulate pollution for the period 1992-1999*, Annegarn Environmental Research, Johannesburg.

Chidhindi P, Belelie MD, Burger RP, Mkhatshwa G, & Piketh SJ (2019) 'Assessing the impact of Eskom power plant emissions on ambient air quality over KwaZamokuhle', *Clean Air Journal*, 29(1), doi:10.17159/2410-972X/2019/v29n1a2.

Department of Environmental Affairs (2016) 'Air quality offsets guideline'.

Engelbrecht JP, Swanepoel L, Zunckel M, & Chow JC (1998) 'Modeling PM_{10} gravimetric data from the Qalabotjha low-smoke fuels macroscale experiment in South Africa' *Air & Waste Management Association. Bridging international boundaries - clean production for environmental stewardship: proceedings of the 91st Annual Meeting & Exhibition: San Diego, CA. June 14-18, 1998 [Conference proceedings on CD-ROM]* Air & Waste Management Association, Pittsburgh, PA.

EScience Associates (Pty) Ltd & Nova Institute (2013) *Household Emission Offset Pre-Feasibility Study for Eskom*, Eskom Holdings, Sunninghill, Sandton.

Feig G, Garland RM, Naidoo S, Maluleke A, & 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), doi:10.17159/caj/2019/29/2.7464.

Fischer T & Pauw CJ (2010) 'Household emissions offsetting process: toward the development of a verifiable and defensible process for the determination implementation and licensing of emission offsets' *NACA Conference 2010*.

Friedl A, Holm D, John J, Kornelius G, Pauw CJ, Oosthuizen R, & Niekerk AS van (2008) *Air Pollution in dense, low-income settlements in South Africa*, NOVA Institute for the DEAT on behalf of Royal Danish Embassy.

Graham JAN & Dutkiewicz RK (1999) 'Assessing the emissions and cost effectiveness of traditional and transitional household fuel burning appliances in South Africa', *Tydskrif vir Skoon Lug / The Clean Air Journal*, 10(3):13–21.

Hallowes D & Munnik V (2017) *The Destruction of the Highveld - Part 2: Burning Coal*, groundWork, Pietermaritzburg, https://groundwork.org.za/reports/gW_Report_2017.pdf.

Halsey R, Overy N, Schubert T, Appies E, McDaid L, & Kruyshaar K (2019) 'Remaking Our Energy Future: Towards a Just Energy Transition (JET) in South Africa', *Project 90 by 203066*.

Hersey SP, Garland RM, 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 modeling data', *Atmospheric Chemistry and Physics*, 15(8):4259–4278, doi:10.5194/acp-15-4259-2015.

International Food Policy Research Institute (2020) *Impact of Covid-19 on the South African economy: An initial analysis*, https://www.ifpri.org/publication/impact-covid-19-south-african-economy-initial-analysis, accessed 20 August 2021.

Khumalo T N (2018) 'Postponement of compliance with minimum emission standards of NEM: AQA (Act No. 39 0f 2004)', https://slideplayer.com/slide/14367756/.

Langenbacher E (2019) 'Introduction: The Thirtieth Anniversary of The Fall of the Berlin Wall and Unification', *German Politics and Society*, 37(3):1–3, doi:10.3167/gps.2019.370301.

Langerman K (2018) 'Household Energy Use Models and their Application to South African Energy Interventions', *South African Geographers*, 1:306.

Langerman K, Wernecke B, Mkhatshwa G, Herbst D, Piketh S, Burger R, Pauw C, Snyman H, Murray H, Fisher T, & Weston

M (2015) 'Domestic Fuel Burning Mission Reduction: Eskom's Kwazamokuhle Pilot Study' NACA, https://www.eskom.co.za/AirQuality/Documents/LangermanNACApaper2015.pdf.

Langerman KE, Pauw CJ, Smith HJ, & Piketh SJ (2018) 'Moving households to cleaner energy through air quality offsets' 2018 *International Conference on the Domestic Use of Energy (DUE)* 2018 International Conference on the Domestic Use of Energy (DUE), doi:10.23919/DUE.2018.8384405.

Language B, Piketh SJ, Wernecke B, & Burger R (2016) 'Household air pollution in South African low-income settlements: a case study' Air Pollution 2016 Crete, Greece, doi:10.2495/AIR160211.

Le Roux LJ, Zunckel M, & McCormick S (2009) 'Reduction in air pollution using the "basa njengo magogo" method and the applicability to low-smoke fuels', *Journal of Energy in Southern Africa*, 20:3–10.

Makonese T, Masekameni DM, Annegarn HJ, & Forbes PBC (2017) 'Emission factors of domestic coal-burning braziers', *South African Journal of Science*, Volume 113 (Number 3/4), doi:10.17159/sajs.2017/20160187.

Mandela N (2013) *Long walk to freedom: the autobiography*, Little, Brown and Company, New York, NY, https://zelalemkibret.files.wordpress.com/2012/01/the-autobiography-of-nelson-mandela.pdf.

Masondo L, Masekameni D, Makonese T, Annegarn H, & Mohlapi K (2016) 'Influence of coal-particle size on emissions using the top-lit updraft ignition method', *Clean Air Journal*, 26(1), doi:10.17159/2410-972X/2016/v26n1a8.

Moletsane S, Lindeque F, Language B, Nkosi C, Adesina J, Burger R, Mkhatshwa G, & Piketh S (2021) 'Intra-urban variability of PM 2.5 in a dense, low-income settlement on the South African Highveld', *Clean Air Journal*, Volume 31, doi:10.17159/caj/2020/31/1.9413.

Muyemeki L, Burger R, Piketh SJ, Language B, Beukes JP, & van Zyl PG (2021) 'Source apportionment of ambient $PM_{10-2.5}$ and $PM_{2.5}$ for the Vaal Triangle, South Africa', *South African Journal of Science*, 117(5/6), doi:10.17159/sajs.2021/8617.

Ndebele Stoves (2022) 'Union Stoves', https://www.ndebelestoves.co.za/product-category/refurbished-stoves/union-stoves/, accessed 6 May 2022.

Nkosi CN, Piketh JS, Burger PR, & Harrold AJ (2017) 'Variability of domestic burning habits in the South African Highveld: A case study in the KwaDela Township (April 2017)'2017 *International Conference on the Domestic Use of Energy (DUE)* 2017 International Conference on the Domestic Use of Energy (DUE), doi:10.23919/DUE.2017.7931820.

Nkosi NC, Piketh SJ, & Burger RP (2018) 'Fine PM emission factors from residential burning of solid fuels using traditional cast-iron coal stoves', *Clean Air Journal*, doi:10.17159/2410-972X/2018/v28n1a10.

Palmer Development Consulting (Pty) Ltd (2005) *Implementation of Basa Njengo Magogo technology. Tembisa. Final report. Submitted to: Department of Minerals and Energy. By Menyetla Projects (Pty) Ltd and Palmer Development Consulting (Pty) Ltd*, www.dme.gov.za/pdfs/energy/coal/.

Pauw C, Fischer T, Weston M, Preston-Whyte F, & Badenhorst H (2013) 'The elephant in the room: What do we know about urban wood use in South Africa' *16th IUAPPA World Clean Air Congress*, Cape Town.

Republic of South Africa (2015) 'South Africa's Intended Nationally Determined Contribution (INDC)', https://www4.unfccc.int/sites/ndcstaging/PublishedDocuments/South%20Africa%20First/South%20Africa.pdf.

(2021) 'South Africa's First Nationally Determined Contribution under the Paris Agreement', https://www.environment.gov.za/sites/default/files/reports/draftnationalydeterminedcontributions_2021updated.pdf.

Republic of South Africa, Department of Cooperative Governance (2020a) 'Nkangala District Mpumalanga - profile and analysis district develepment model', https://www.cogta.gov.za/ddm/wp-content/uploads/2020/07/Nkangala-District-Profile.pdf.

(2020b) 'Gert Sibande District Mpumalanga - profile and analysis district develepment model', https://www.cogta.gov.za/ddm/wp-content/uploads/2020/07/Final-Edited-Gert-Sibande-DM_26-June-2020-FINAL.pdf.

(2020c) 'City of Ekurhuleni metropolitan GAU - profile and analysis district development model', https://www.cogta.gov.za/ddm/wp-content/uploads/2020/08/Take2_DistrictProfile_EKURHULENI-2.pdf.

Republic of South Africa, Department of Environmental Affairs (2014) 'Air Quality Offsets Policy (Draft)'.

(2019) 'Strategy to address air pollution in dense low-income settlements', *Government Gazette*, 666(42464), https://saaqis.environment.gov.za/pagesfiles/Strategy_to_address_air_pollution_in_dense_low-income_settlements.pdf.

Sasol Limited (2020) *Sasol in Society Report*, https://www.sasol.com/sites/default/files/content/files/Sasol%20Report%20to%20Society%2019%20Nov%20Sopnhia.pdf.

South Africa Revenue Service (2021) *Carbon Tax*, https://www.sars.gov.za/customs-and-excise/excise/environmental-levy-products/carbon-tax/.

Statistics South Africa (2020) *General Household Survey 2019*, Pretoria,http://www.statssa.gov.za/publications/P0318/P03182019.pdf, accessed 26 September 2021.

(2021) *National Poverty Lines*, Pretoria, http://www.statssa.gov.za/publications/P03101/P031012021.pdf.

Surridge AD, Kgobane KB, & Chauke GR (2005) 'Strategy to combat the negative impacts of domestic coal combustion: basa njengo magogo', *Clean Air Journal = Tydskrif vir Skoon Lug*, 14(1):13–16, doi:10.10520/EJC27323.

Tyson PD, Kruger FJ, & Louw CW (1988a) *Atmospheric pollution and its implications in the Eastern Transvaal highveld*, Foundation for Research Development, CSIR, Pretoria, http://hdl.handle.net/10204/2376.

(1988b) 'Atmospheric pollution and its implications in the Eastern Transvaal Highveld', *Clean Air Journal*, 7(6):15–17, doi:10.17159/caj/1988/7/6.7209.

United Nations (1992) *United Nations Conference on Environment and Development, Rio de Janeiro, Brazil, 3-14 June 1992, United Nations*, https://www.un.org/en/conferences/environment/rio1992, accessed 19 August 2021.

Van Niekerk A, Swanepoel P, & Kornelius G (1999) 'New possibilities to improve air quality in low income housing and reduce greenhouse gasses', *Clean Air Journal*, 10(4).

Velicu I & Barca S (2020) 'The Just Transition and its work of inequality', *Sustainability: Science, Practice and Policy*, 16(1):263–273, doi:10.1080/15487733.2020.1814585.

Wagner NJ, Schoonraad P, Swanepoel P, Niekerk AV, Scholtz C, Kornelius G, Julies A, Pretorius O, Wasserman J, & Muller A (2005) 'Results of domestic smoke reduction programmes at eMbalenhle (Mpumalanga) and Zamdela (Free State)' *NACA Conference (Cape Town, 27-30 September 2005) [Conference proceedings on CD-ROM]* NACA, Johannesburg, http://www.airshed.co.za/documents/Recent/.

Walton NM (2021) *Aerosol source apportionment in southern Africa*, North West University, Potchefstroom.

World Bank (2021) *GDP per capita (constant LCU) - South Africa, World development indicators*, https://data.worldbank.org/indicator/NY.GDP. PCAP.KN, accessed 13 January 2022.

Wright CY, Oosthuizen R, John J, Garland RM, Albers P, & Pauw C (2011) 'Air quality and human health among a low income community in the Highveld priority area', *Clean Air Journal= Tydskrif vir Skoon Lug*, 20(1):12–20.

Research article

Analysis of the first surface nitrogen dioxide concentration observations over the South African Highveld derived from the Pandora-2s instrument

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Abstract

Anthropogenic emissions from industry, biomass burning and traffic are significant contributors to the atmospheric loading of nitrogen dioxide on the South African Highveld. These sources are dispersed across the region and emit nitrogen oxides (NO and NO_2) into the atmosphere at different elevations above the earth's surface. Additionally, atmospheric stability in the form of surface and elevated inversions decreases the dispersion of air pollutants and stratifies pollutants into distinctive layers above the surface. This study explores the Highveld near-surface nitrogen dioxide concentrations obtained using the ground-based Pandora-2s monitoring system. The Pandora-2s instrument retrieves surface NO_2 levels from clear sky measurements using a fully parameterised algorithm. We present the first near-surface concentration measurements of atmospheric NO_2 at Wakkerstroom, a site between Volksrust and Amersfoort, downstream of major source conglomerates, the Majuba power station and other industries. These data are explored in the presence and context of potential background NO_2 concentrations in the area derived from other ground-based sensors. The quasicontinuous data show elevated surface NO_2 levels in week 37 (September) of 2020 (7.3 ± 5.7 ppb), while the lowest levels were observed in week 15 (April) of 2020 (0.2 ± 0.04 ppb). The elevated surface NO_2 levels are driven by dominant emission sources and transport trajectories, while the accuracy in the measurements is based on the high temporal resolution of the ground-based Pandora-2s instrument.

Keywords

Nitrogen dioxide, Pandora-2s, plumes, Eskom, Highveld

Introduction

The South African Highveld region, characterised by elevations of ~700 – 2000 m above sea level, comprises portions of the inland provinces, including Mpumalanga (Balashov et al., 2014; Freiman and Piketh, 2003). Mpumalanga is home to approximately ninety-six registered stationary facilities that emit pollutants into the atmosphere, including most of South Africa's coalfired power stations (https://saaelip.environment.gov.za/ accessed 23rd June 2021). These emissions and others from anthropogenic activities such as domestic biomass burning, vehicular emissions, and solid fuel combustion result in elevated levels of pollutants, particularly sulphur dioxide (SO_2), carbon dioxide (CO_2), particle matter (PM) and nitrogen oxides (NO_x) (NO + NO_2) (Alade, 2011; Belelie et al., 2019; Hickman et al., 2021; Naiker et al., 2012; Shikwambana et al., 2020). As a result, there is a persistent pool of pollutants in the Highveld atmosphere (Lourens et al., 2012).

Coal-fired power stations produce electricity by converting thermal energy produced through coal combustion into electrical energy (Wang et al., 2020). During the coal combustion process, which accounts for ~65% of the global total NO_x emissions (Bauwens et al., 2020), nitrogen oxides are produced (Scorgie and Thomas, 2006), released into the atmosphere (Breeze, 2015), where they contribute to the atmospheric chemistry. Therefore, the plethora of emissions in the highly industrialised Highveld region contributes to the reported increased atmospheric levels of nitrogen oxides (NO_x) (NO + NO_2) (Celarier et al., 2008). As a result, the Mpumalanga Highveld is known as the major (> 80% of emissions in South Africa) emission region of nitrogen oxides (NO_x) (Collett et al., 2010).

Atmospheric NO emissions are highly reactive and have a distinctive diurnal cycle attributed to photolysis reactions (Celarier et al., 2008; Verhoelst et al., 2021). In the presence of solar radiation, they transform into NO_2. In the presence of volatile organic compounds (VOCs) and sunlight, the NO_2 forms ozone (O_3) (Hakkarainen et al., 2021; Wang and Hao, 2012), a secondary air pollutant. The rate at which O_3 is produced is determined by the NO_x emission time and location (Wedow et al., 2021). These emissions also play a role in forming secondary organic aerosols (SOAs), nitric acid and nitrates (Qi et al., 2020; Verhoelst et al., 2021). Therefore, these NO_x chemical transformations contribute to the formation of smog and acid rain (Qi et al., 2020; Verhoelst et al., 2021).

In addition to chemical processes, these emissions are subjected to various other processes linked with meteorological conditions, determining their dispersion and transport from the source region (Belelie et al., 2019). Ultimately, it has been deduced that principal pollutants such as NO_x play a critical role in indicating regional air quality (Behm and Haupt, 2020; Brancher, 2021; Wang and Hao, 2012; Zyrichidou et al., 2015).

Therefore, it is important to accurately quantify the distributions, trends and cycles of nitrogen dioxide levels in the atmosphere, particularly near the surface where most of the emissions originate. These have been measured from space using Low Earth Orbit (LEO) satellites since the 1970s (Verhoelst et al., 2021). However, the lack of continuous trace gas monitoring data is still a concern (de Lange et al., 2021; Omrani et al., 2020; Shabbir et al., 2016) in air quality research. The scarcity of data is also affected by the challenge in acquiring and extracting accurate atmospheric column trace gas measurements in cloudy conditions.

This study reports on the first near-ground (surface) nitrogen dioxide measurements from the Pandora-2s instrument in the Mpumalanga, South African Highveld. In contrast to the LEO satellites, the Pandora-2s is a ground-based instrument that derives measurements of atmospheric trace gases using one of three observation modes: direct sun, direct moon or MAXDOAS (multi-axis differential optical absorption spectroscopy) (Cede et al., 2021). In addition to that, it provides uninterrupted measurements instead of the once-a-day measurements from the current satellite instruments. This instrument was optimised to retrieve nitrogen dioxide column concentrations under cloudy conditions.

These first-time measurements will be used to assess the near-surface concentrations of NO_2 downwind the region of major Highveld emissions sources, in Wakkerstroom, Mpumalanga. We have used near-surface air quality data from three Eskom air quality monitoring sites (Kendal, Majuba and Elandsfontein) to compare the remotely derived NO_2 concentrations from the Pandora-2s instrument. Meteorological variables and kinematic backward trajectory analyses indicate near-ground nitrogen dioxide pollutant signatures.

Data and methodology

Four data sets have been used in this study: near-surface atmospheric column NO_2 concentrations from a Pandora-2s (Pan159) instrument; near-surface NO_2 gas concentrations from Eskom's Majuba, Kendal and Elandsfontein monitoring stations; surface meteorological data collected at Wakkerstroom; and backward kinematic trajectories calculated for Wakkerstroom. Details of the data and methods are provided below.

Instrumentation

The Pandora-2s instrument can obtain measurements in three observation modes; direct sun, direct moon or MAXDOAS (multi-axis differential optical absorption spectroscopy) (Cede et al., 2021). The MAXDOAS viewing geometry retrieves surface nitrogen dioxide concentrations (Herman et al., 2009) amongst other product outputs. The measurements are processed using the "L2 Air-Ratio Sky Algorithm (Cede et al., 2021). A fully parameterised algorithm (which does not require elaborative radiative transfer calculations) is then used to extract the real-time quasi-continuous surface concentration data (Cede, 2021; Tiefengraber et al., 2021).

Further details regarding the algorithm can be found in Cede (2021). Quasi-continuous data are discontinuous and are characterised by "events" (much higher than the average data) in the observations. These data are typical of atmospheric trace gas and meteorological variables measurements.

More characteristics of the dual spectrometer Pandora-2s instrument can be found in Herman et al. (2009) and Zhao et al. (2020, 2016). The extracted surface concentrations are from within the first 50 m above the ground. During winter and autumn, they are monitored between 06h00 and 17h00, and 05h00 to 18h00 in the summer and spring due to the instrument relying on sky radiance for measurements.

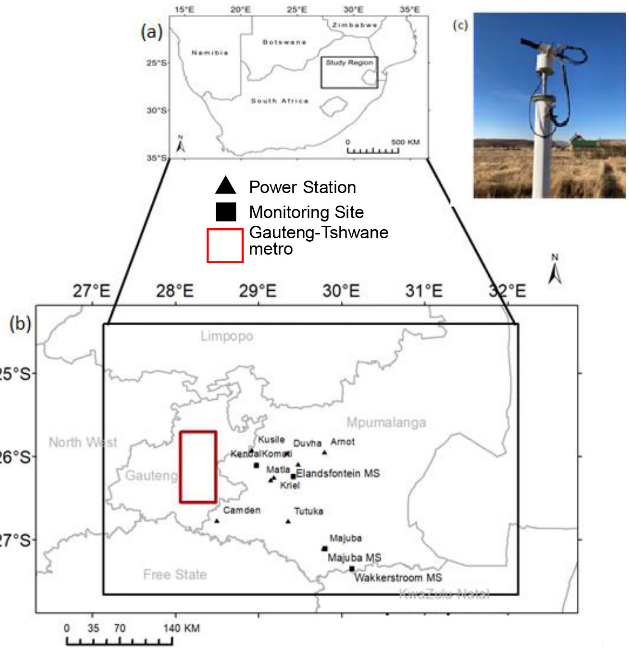

Figure 1: a) A map of Southern Africa and surrounding countries. b) Map of the Gauteng and Mpumalanga provinces of South Africa, indicating the site locations where this study's surface nitrogen dioxide concentrations were monitored. c) Inset: The Pandora-2s (Pan159) instrument in situ at the Wakkerstroom monitoring site.

Surface NO_2 data

The Pandora-2s instrument was primarily installed at Wakkerstroom to investigate the biogeochemical exchange of trace gases in ecosystem functions. Nitrogen biogeochemical flows, amongst others, contribute to the functioning of ecosystem services such as climate regulation (Watanabe and Ortega, 2011; Xu et al., 2021). The Wakkerstroom monitoring site is directly downwind of the Majuba Power station (PS) and the greater Highveld region and is ideally located to detect the high NO_x emissions and subsequent transport. The Pandora-2s instrument at Wakkerstroom, Mpumalanga, is part of the Pandora Global Network (PGN) ("NASA Pandora Project," last accessed: 18th September 2021) (Figure 1). The surface NO_2 data from the Wakkerstroom site are presented from 1st January to 31st December 2020. Although night-time measurements were taken using the moon as a reference, these data have not been included in this initial analysis. Weekly average surface concentrations have been calculated from the Pandora-2s data set. This provided the best data recovery, given missing data for several months.

Additionally, nitrogen dioxide data from three Eskom air quality monitoring sites (MS), namely Kendal, Majuba and Elandsfontein in Mpumalanga, are presented to provide comparative in situ surface measurements. The Elandsfontein MS is centrally located on the Highveld and represents an integrated plume from multiple significant NO sources. Kendal and Majuba monitoring sites are located downwind of the similarly named power stations to detect maximum surface concentrations (Thomas and Scorgie, 2006). The Kendal monitoring site is also ~30 km southwest of Emalahleni, with low-income townships where domestic coal is prevalent (Matimolane, 2019).

Data at the three Eskom monitoring sites above are continuous, with hourly averaging times collected between July 2015 and May 2020. In order to create comparable data sets, weekly average values were calculated from the continuous data sets.

NO_2 data description and acquisition

The Pandora-2s instrument data are processed to produce outputs; the total, tropospheric, and surface concentration. Data from the Pandonia global network (https://www.pandonia-globalnetwork.org/pgn-data/) were cleaned and analysed using Rstudio® and Excel® statistical software. Eskom provided the air quality monitoring data from Kendal, Majuba and Elandsfontein monitoring sites.

NO_2 data cleaning

The data from the Pandora instrument are filtered and flagged using data quality indicators, high, medium and low quality, which specify the confidence in the data. The quality of the data depend on the measurements' uncertainties during monitoring. Various factors contribute to the uncertainty in measurements; noise, permanent systematic effects, temporary systematic effect, calibration transfer uncertainty, transport uncertainty and drift correction uncertainty (Cede and Tiefengraber, 2013. The low-quality data should not be used for most purposes and are not presented in these observations. The first-time Pandora-2s instrument (Pan159) measurements at Wakkerstroom provided semi-continuous data that had gaps. The surface NO_2 concentrations are impacted by cloud cover as they depend on sky radiance for successful retrieval (Tiefengraber et al., 2021; Zhao et al., 2019).

Meteorological data

The dispersion and transport of trace gases such as nitrogen dioxide in the atmosphere depend on meteorological conditions (Goldberg et al., 2020). These conditions determine the atmosphere's stability and, therefore, the chemical and physical processes that affect the atmospheric accumulation of pollutants (Balashov et al., 2014). The ambient air temperature depends on solar irradiance, and they both affect the dissociation of NO_2 in the atmosphere (Balashov et al., 2014; Goldberg et al., 2020). The wind speed and direction contribute to the dispersion and transport of the pollutant around and to-and-from the source of emissions (Balashov et al., 2014). The removal rate of pollutants from the atmosphere is also dependent on rain and humidity through wet deposition (Seinfeld and Pandis, 2016).

Meteorological data from the Wakkerstroom monitoring site (Figure 1), measured using the HOBO® weather station and accessed through a CR300 logger, are used in this study. The measurements are near-surface/ground level values of temperature, rainfall, relative humidity, wind speed and wind direction. All variables were recorded as hourly averages. The wind direction data were averaged according to a technical note by Grange (2014) using Rstudio®. All variables data are available for the one year (2020) analysis period.

HYSPLIT backward trajectory analysis

The major atmospheric transport pathways to Wakkerstroom were computed using the HybridSingle-Particle Lagrangian Integrated Trajectory (HYSPLIT) model (https://www.ready.noaa.gov/HYSPLIT.php) at 850 hPa. The trajectory represents the temporal and spatial path of trace gases in masses of air in the atmosphere. Backward trajectories are used to identify the source region of atmospheric constituents (Bera et al., 2021).

Ten-day backward trajectories were used to determine the origin of the air masses to Wakkerstroom. The model uses a hybrid between the Lagrangian approach and the Eulerian methodology. The former uses a "moving frame of reference for the advection and diffusion calculations as the trajectories or air parcels move from their initial location", while the latter "uses a fixed three-dimensional grid as a frame of reference to compute pollutant air concentrations" (Stein et al., 2015).

Ten-day backward trajectories were calculated for every day of 2020, starting at the monitoring site at midday. The trajectories were visually classified into five distinctive transport pathways, and frequencies of each pathway were calculated as a percentage for the total of 366 days. Additionally, trajectory pathways were used to determine the likely atmospheric transport of the highest and lowest concentrations of NO_2 at Wakkerstroom.

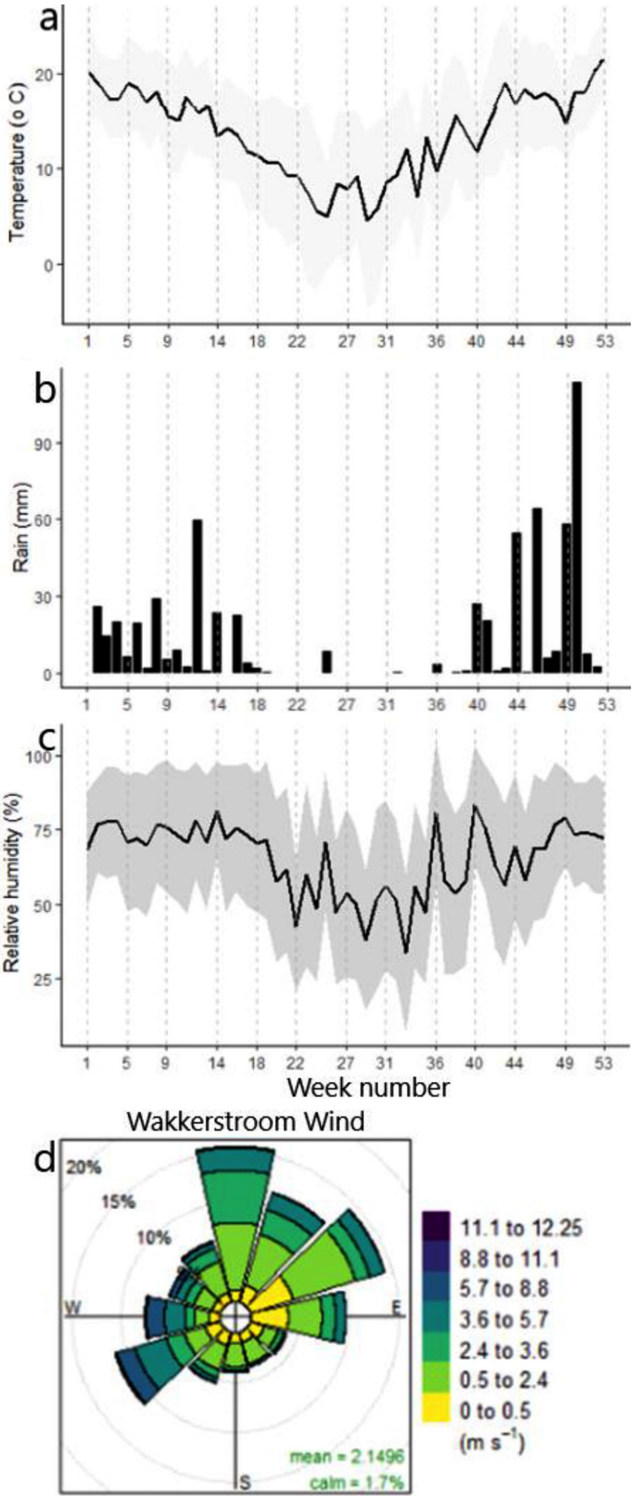

Figure 2: *Weekly averaged meteorological variables at Wakkerstroom, January to December 2020. a) temperature, b) rainfall, c) relative humidity, and d) wind direction and speed. The shaded areas (a and c) show the standard deviation, and the dashed vertical lines (a-c) the start of a new month.*

Results

Meteorology conditions at Wakkerstroom

The temperature data measured at Wakkerstroom are consistent with the seasonal variation in South Africa (Walt and Fitchett, 2020). Temperatures (Figure 2a) decrease in April, reaching an all-time low in July (week 29). The highest temperature was recorded in December 2020 (week 53).

The rainfall data (Figure 2b) shows less rainfall from May to early September 2020 with the wet season starting from October 2020. The highest rainfall was seen in week 50 (December 2020).

The relative humidity percentages (Figure 2c) were highest in weeks 36 (September 2020) and 40 (October 2020), while the lowest was seen in week 33 (August 2020).

The wind rose (Figure 2d) shows that the dominant wind directions for 2020 were from the northerly to easterly sectors (>40%) (Figure 2d). The wind also blew fairly frequently from the southwesterly and westerly sectors (~20%).

Atmospheric transport to Wakkerstroom

The transport pathways identified to Wakkerstroom are similar to the patterns obtained in previous studies for the Highveld (Freiman and Piketh, 2003) (Figure 3). Air transport is recirculated on the scale of approximately 500 km over the interior of South Africa for ~41% of days in 2020. Air originating from the south also represents an important pathway (30%). Approximately 5.2% (least) of the transport pathways originate from the southwest direction. Two other less dominant transport pathways were identified, namely, direct transport from the east coast of South Africa (~11) as well as recirculated air that passes over neighbouring countries to the North before reaching Wakkerstroom from the north-west (~13%). The trajectory

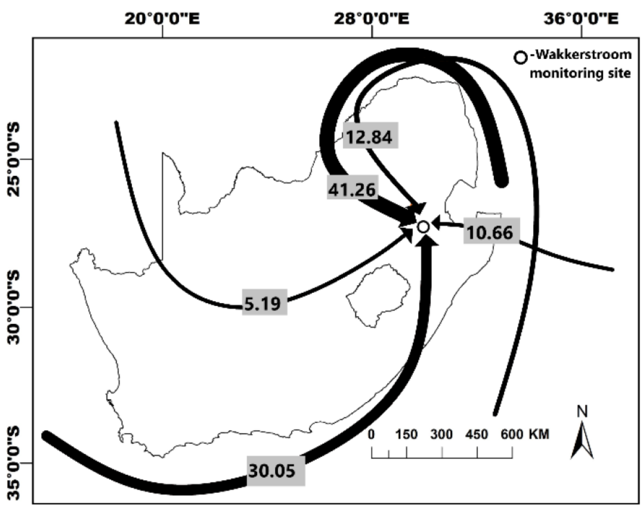

Figure 3: *Schematic diagram depicting a summary of HYSPLIT backward trajectory path frequencies at Wakkerstroom, Mpumalanga for the year 2020. The arrows are drawn relative to the pathway frequencies in the depicted direction. The numbers on the arrows show the percentage frequencies in the depicted direction.*

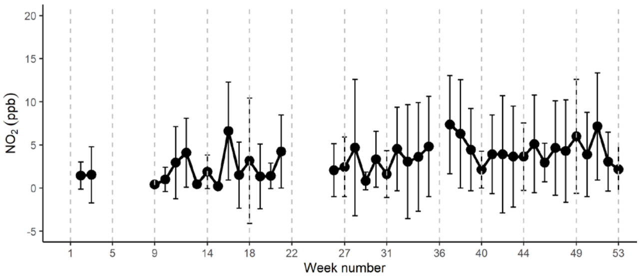

Figure 4: *Twelve months (53 weeks) of weekly Pandora-2s surface nitrogen dioxide concentration averages (diurnal averages) at the Wakkerstroom monitoring site in the Mpumalanga Highveld. The error bars show the standard deviation, and the dashed vertical lines show the beginning of a new month in 2020.*

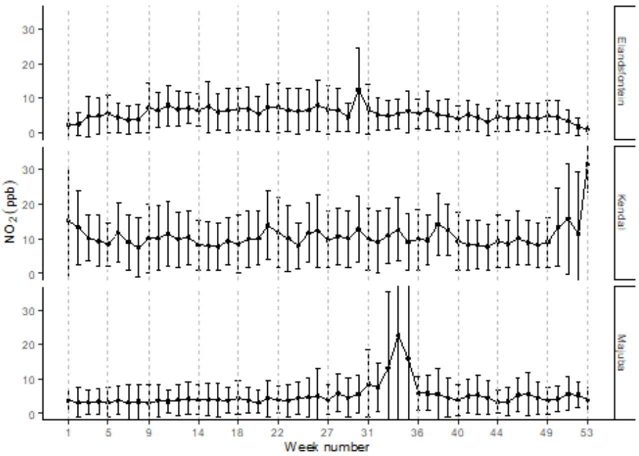

Figure 5: *Six years (2015 – 2020) of weekly averaged ambient air nitrogen dioxide concentrations from three Eskom monitoring sites (Elandsfontein, Kendal, and Majuba) in the Mpumalanga Highveld. The error bars show the standard deviation, and the dashed vertical lines show the beginning of a new month.*

analysis is consistent with the prominent wind pattern at Wakkerstroom shown in the wind rose (Figure 2d).

Surface nitrogen dioxide concentrations and meteorology over the Highveld

Surface level NO_2 concentrations measured by the Pandora-2s instrument are given in Figure 4. These average surface NO_2 concentrations fluctuate between 0.2 and 7.0 ppb (±0.04 and ±5.7) (Figure 4). These concentrations are lower than the South African National Air Quality Standards (NAAQS) of 40 ug/m^3 (21.3 ppb) per 1-year average. Maximum surface concentrations at Wakkerstroom only just exceeded 10 ppb. The highest concentrations were detected during weeks 37 (September 2020), 16 (April 2020) and 50 (December 2020), which fall in three different seasons. There is no distinct seasonal cycle visible in the Pandora-2s data at Wakkerstroom.

By comparison, the weekly variation of the surface nitrogen dioxide concentrations at the three Eskom monitoring sites did not show a great deal of variation throughout the year. Average weekly concentrations at Kendal, Majuba and Elandsfontein, were 10.4 (±9.1) ppb, 5.3 (±11.8) ppb and 5.8 (±5.6) ppb, respectively (Figure 5). The large standard deviation between the weekly averages is attributed to the high difference in the means ranging from 7.5 ppb to 31.47 ppb, 3.24 ppb to 27.62 ppb and 1.18 ppb to 12.53 ppb at Kendal, Majuba and Elandsfontein, respectively. Kendal MS showed the most weeks with higher NO_2 concentrations than the other monitoring stations. The Kendal MS is 2 km south south-east (almost directly below) of the Kendal PS and is aligned to capture the direct high concentration plumes from the Kendal PS. At the Kendal MS there is some indication of small increases during the initial summer months (November and December 2020). Concentrations at Elandsfontein and Majuba were lower overall. Majuba monitoring site showed the most significant evidence of a seasonal peak occurring from the end of July to the middle of September. At the Majuba MS, surface NO_2 concentrations exceeded 10 ppb during this season.

The comparison between the Pandora-2s data and the in-situ surface monitoring stations gives confidence that the algorithm for deriving surface concentrations from the column integrated data gives reasonable values at the Wakkerstroom site.

Atmospheric transport of highest and lowest NO_2 concentrations to Wakkerstroom

To better understand the drivers of the observations at Wakkerstroom, daily trajectories were calculated for the weeks with the highest (week 37, 51 and 16) and lowest (week 15 and 9) NO_2 concentrations.

The highest concentrations (7.5 ppb ±5.7) were detected from the 7th to the 13th September 2020 (week 37). Transport of air masses to the Wakkerstroom during this week was predominantly from the east and north-east. Four out of the seven days (Figure 6b-d and f) show trajectories that pass directly over eSwatini before arriving at the Wakkerstroom MS. The remaining three days have regional scale recirculation occurring (7th, 11th and 13th September 2020) (Figures 6a, 6e and 6g). The transport over eSwatini during this week is the most likely source of the elevated surface NO_2. Fire count data from the Moderate Resolution Imaging Spectro-radiometer (MODIS) confirms a high number of fire activity over the east coast of South Africa and in eSwatini during September 2020, particularly during week 37 (Figure 11).

The weeks with the second (7.1 ppb ±6.2) and third highest (6.6 ppb ±6.2) surface NO_2 weekly averaged concentrations (week 51 and week 16) from the Pandora-2s instrument show atmospheric transport dominated by regional scale recirculation, particularly week 16. The trajectories show transport over the eSwatini region, during week 51 (Figure 7 and 8). The regional scale recirculation transport patterns would certainly accumulate NO_2 concentrations over the source region of the Highveld as well as the greater Johannesburg (Gauteng-Tshwane metropolitan) region.

During the weeks with the lowest nitrogen dioxide concentrations (weeks 15 and 9), most of the air masses arriving

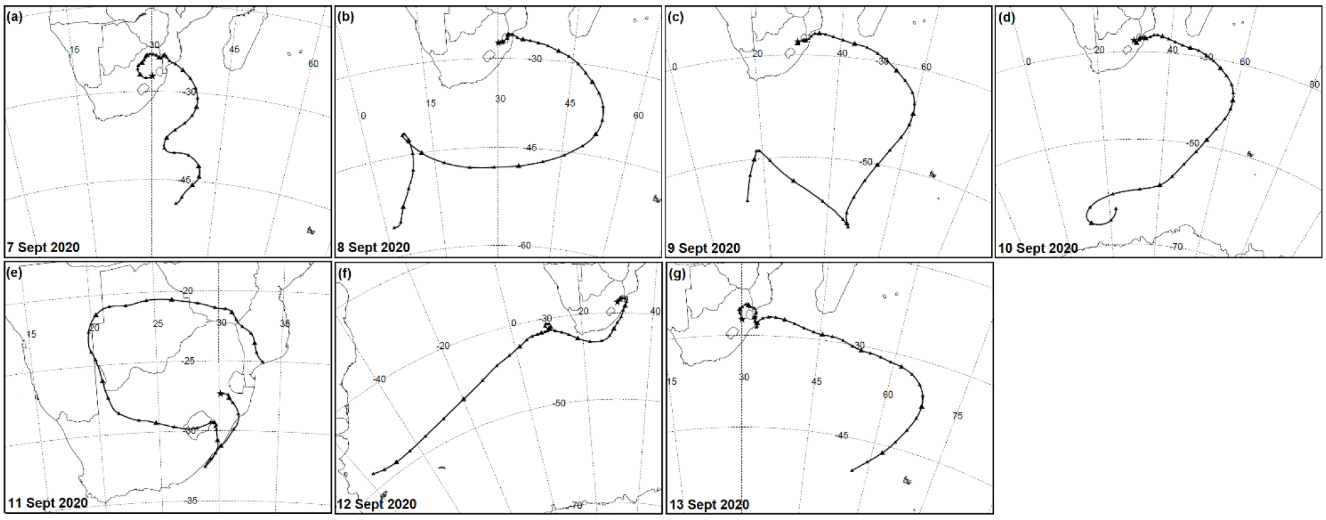

Figure 6: *The HYSPLIT backward trajectory paths to Wakkerstroom at 850 hPa during the week (week 37) (a-g) with the highest nitrogen dioxide concentration measurements from the Pandora-2s instrument at the Wakkerstroom monitoring site in 2020.*

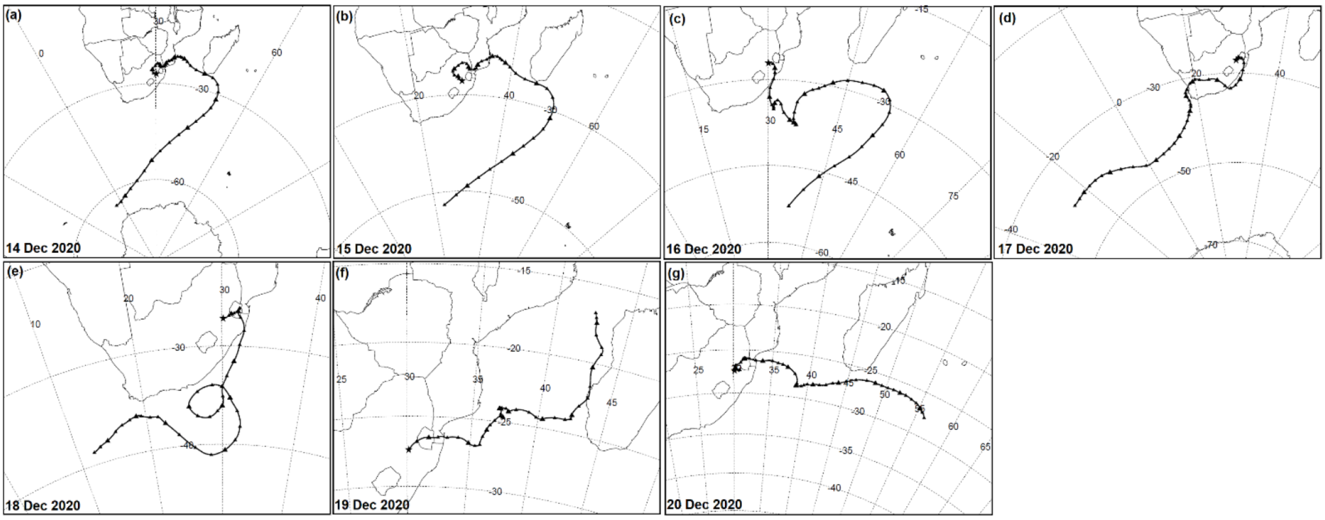

Figure 7: *The HYSPLIT backward trajectory paths at 850 hPa during the week (week 51) (a-g) with the second-highest nitrogen dioxide concentration measurements from the Pandora-2s instrument at the Wakkerstroom monitoring site in 2020.*

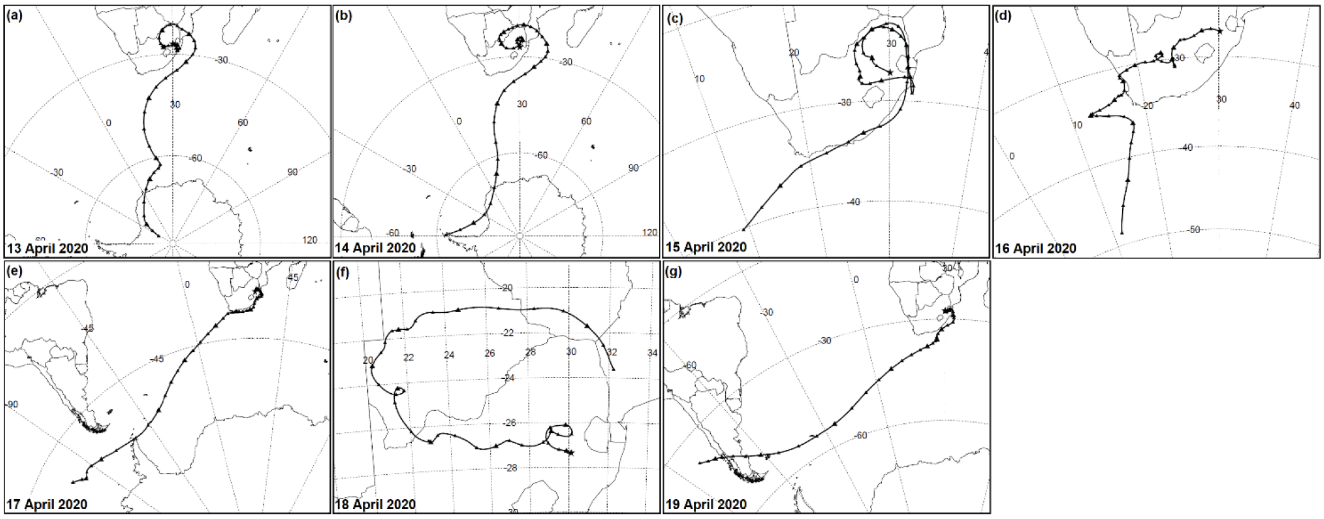

Figure 8: *The HYSPLIT backward trajectory paths at 850 hPa during the week (week 16) (a-g) with the third-highest nitrogen dioxide concentration measurements from the Pandora-2s instrument at the Wakkerstroom monitoring site in 2020.*

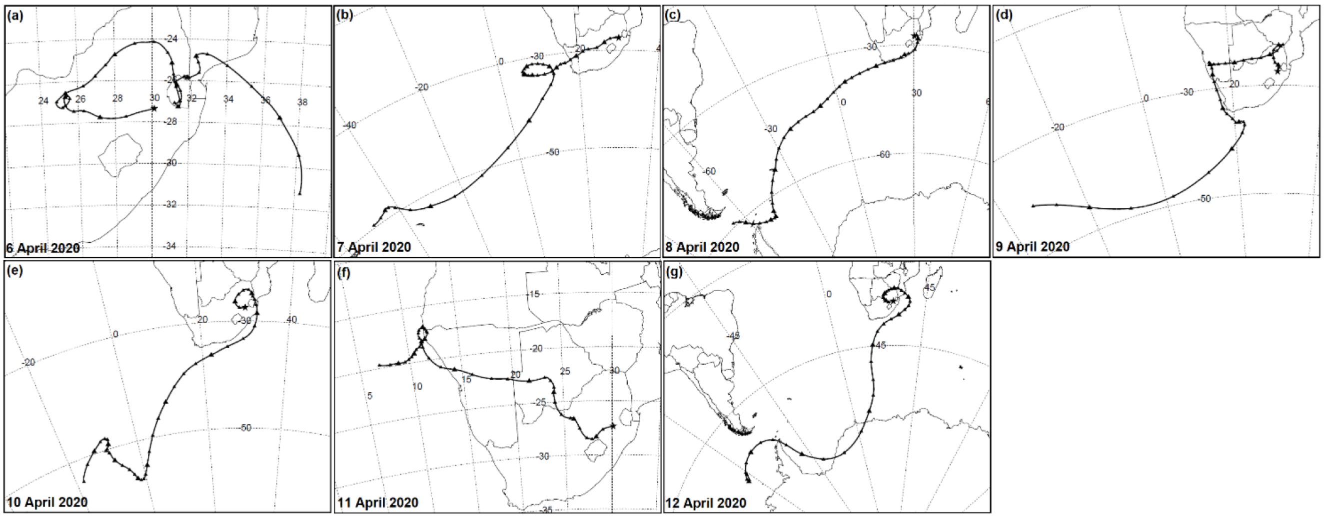

Figure 9: *The HYSPLIT backward trajectory paths at 850hPa during the week (week 15) (a-g) with the lowest nitrogen dioxide concentration measurements from the Pandora-2s instrument at the Wakkerstroom monitoring site in 2020.*

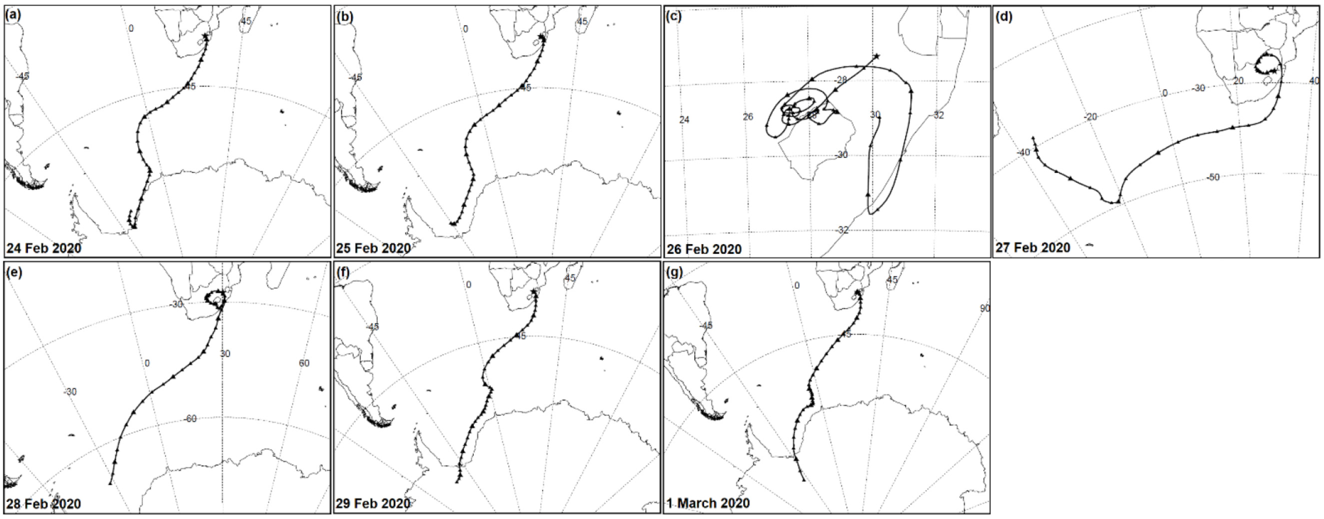

Figure 10: *The HYSPLIT backward trajectory paths at 850 hPa during the week (week 9) (a-g) with the lowest nitrogen dioxide concentration measurements from the Pandora-2s instrument at the Wakkerstroom monitoring site in 2020.*

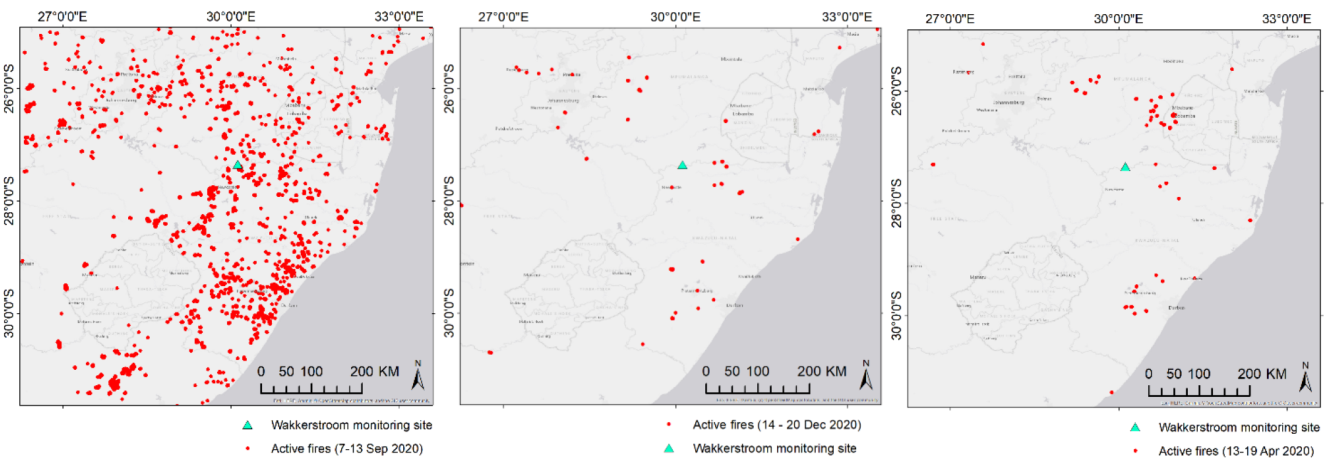

Figure 11: *The MODIS fire count (red dots) product for week 37 (left), 51 (centre) and 16 (right). The green triangle represents the site at Wakkerstroom.*

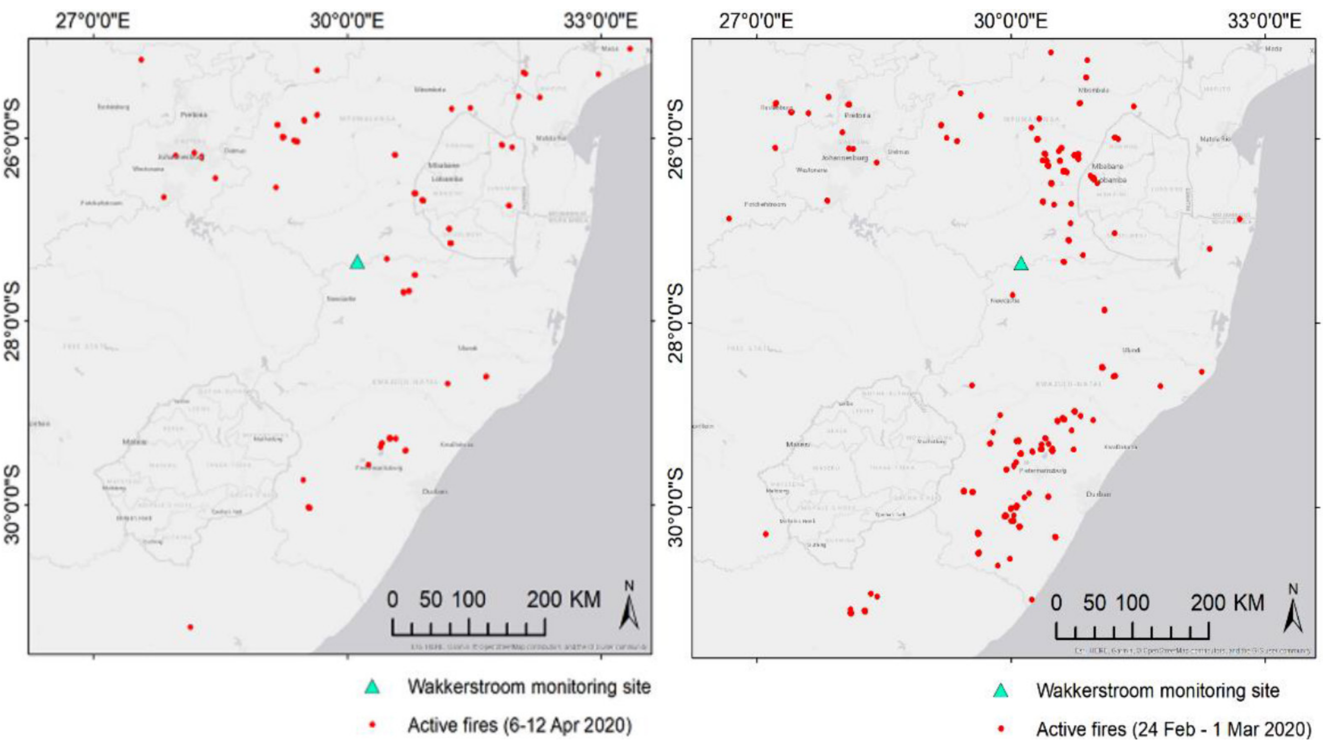

Figure 12: *The MODIS fire count (red dots) product for week 15 (left) and week 9 (right). The green triangle represents the site at Wakkerstroom.*

at the Wakkerstroom MS were directly from the south (Figures 9 and 10). This type of air mass trajectory is typically associated with cleaner air from the Southern Ocean. The trajectories spent very little time over the subcontinent before reaching the Wakkerstroom MS. Fire count data from MODIS shows that there was biomass burning activity during weeks 15 and 9 (Figure 12), however, since the airmass trajectories were dominantly from the Southern Ocean (Figures 9 and 10), these emissions had less effect on the levels of NO_2 concentrations to the Wakkerstroom MS during those weeks.

Discussion

Weekly NO_2 surface concentrations at the three conventional air quality monitoring stations and the Wakkerstroom MS with the Pandora-2s instrument are comparable, given their relative location to major sources. The sites closer to the power stations (Majuba and Kendal) have higher surface nitrogen dioxide levels than those further away, albeit downwind of the power stations (Figures 1, 4 and 5). Nitrogen dioxide has a short atmospheric lifetime and therefore does not get transported far from its source (Boersma et al., 2007). Interestingly, even though the Kendal and Majuba sites are both ca. 3 km south-southeast and south-east of direct emission sources, respectively, the Kendal site had higher weekly averaged surface nitrogen dioxide levels. This can be an indication of NO_2 loading; however, this study did not investigate the loading of NO_2 at the monitoring sites. No discernible seasonal cycle was observed at three of the four sites, including the Wakkerstroom site with the Pandora-2s instrument. However, the Majuba MS had slightly elevated concentrations in the spring. Distinctive atmospheric transport patterns associated with the highest and lowest concentrations of NO_2 have been identified. Although the regional scale recirculation was not surprising, high concentrations of NO_2 associated with biomass burning emissions transported from the east coast of South Africa and eSwatini were not expected and represented an important finding. The lowest concentrations were recorded during months when some biomass burning was observed; however, these concentrations were highly associated with air masses from the south coast of South Africa. It is encouraging that these data will indeed be instrumental in unpacking the biogeochemical cycling of trace gases over the study area. In the next phase of analysing Pandora-2s data from Wakkerstroom, the NO_2 concentrations in the middle and the upper troposphere will be extracted. These data will be invaluable at understanding the long-range transport of pollutants from the Highveld as well as providing important surface derived total column retrievals that can be compared to the many existing satellite retrievals. Until the installation of this instrument, there have been few attempts to validate the satellite retrieved concentration of NO_2 over the Highveld.

Conclusions

The first-time surface level NO_2 measurements retrieved from Wakkerstroom using the Pandora-2s instrument have introduced new confidence that the data products can be used in future to explore biogeochemical interactions in the Highveld area. This comes from the similarity in the Pandora-2s measurements to the already existing sensors in the Highveld and the instrument's capability to indicate elevated levels of surface NO_2 concentrations accurately. The Pandora-2s is a valuable instrument that promises to build and close the gap of high temporal resolution long-term data.

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References

Alade OL (2011) Characteristics of Particulate Matter over the South African Industrialized Highveld. MSc thesis, University of Witwatersrand, Johannesburg.

Balashov NV, Thompson AM, Piketh SJ, Langerman KE (2014) Surface Ozone Variability and Trends Over the South African Highveld from 1990 to 2007. Journal of Geophysical Research: Atmospheres, 119: 4323–4342. https://doi.org/10.1002/2013JD020555

Bauwens M, Compernolle S, Stavrakou T, Müller JF, Gent J van, Eskes H, Levelt PF, A R van der, Veefkind JP, Vlietinck J, Yu H, Zehner C (2020) Impact of Coronavirus Outbreak on NO_2 Pollution Assessed Using TROPOMI and OMI Observations. Geophysical Research Letters, 47: 11. https://doi.org/10.1029/2020GL087978

Behm S, Haupt H (2020) Predictability of Hourly Nitrogen Dioxide Concentration. Ecological Modelling. 42. https://doi.org/10.1016/j.ecolmodel.2020.109076

Belelie MD, Burger RP, Mkhatshwa G, Piketh SJ (2019) Assessing the Impact of Eskom Power Plant Emissions on Ambient Air Quality over KwaZamokuhle. Clean Air Journal, 29: 29–37. https://doi.org/10.17159/2410-972x/2019/v29n1a2

Bera B, Bhattacharjee S, Sengupta N, Saha S (2021) Variation and Dispersal of PM_{10} and $PM_{2.5}$ During COVID-19 Lockdown over Kolkata Metropolitan City, India Investigated Through HYSPLIT Model. Geoscience Frontiers. https://doi.org/10.1016/j.gsf.2021.101291

Brancher M (2021) Increased Ozone Pollution Alongside Reduced Nitrogen Dioxide Concentrations During Vienna's First COVID-19 Lockdown: Significance for Air Quality Management. Environmental Pollution, 284. https://doi.org/10.1016/j.envpol.2021.117153

Breeze P (2015) Coal Combustion and the Environment, in: Breeze, P. (Ed), Coal-Fired Generation, Chapter 6, 53-72. Academic Press, Boston. https://doi.org/10.1016/B978-0-12-804006-5.00012-5

Cede A (2021) Manual for Blick Software Suite 1.8, Version 1-8-3. Manual, 183-185 pp https://www.pandonia-global-network.org/wpcontent/uploads/2021/05/BlickSoftwareSuite_Manual_v1-8-3.pdf. Accessed 17 September 2021.

Cede PA, Tiefengraber M (2013) CEOS Intercalibration of Ground-Based Spectrometers and Lidars. Minispectrometer Intercalibration and Satellite Validation. Report. https://www.pandonia-global-network.org/wpcontent/uploads/2019/06/LuftBlick_CEOS_ICalMinispectrometers_IntercalRecommendations_RP_2013002_v5.pdf

Celarier EA, Brinksma EJ, Gleason JF, Veefkind JP, Cede A, Herman JR et al. (2008) Validation of Ozone Monitoring Instrument Nitrogen Dioxide Columns. Journal of Geophysical Research: Atmospheres, 113. https://doi.org/10.1029/2007JD008908

Collett KS, Piketh SJ, Ross KE (2010) An Assessment of the Atmospheric Nitrogen Budget on the South African Highveld. South African Journal of Science, 106: 35–43. DOI: 10.4102/sajs.v106i5/6.220

De Lange A., Naidoo M, Garland RM, Dyson LL (2021) The Sensitivity of Simulated Surface-Level Pollution Concentrations to WRF-ARW-Model PBL Parameterisation Schemes Over the Highveld of South Africa. Atmospheric Research, 254. https://doi.org/10.1016/j.atmosres.2021.105517

Freiman MT, Piketh SJ (2003) Air Transport into and out of the industrial Highveld region of South Africa. Journal of Applied Meteorology, 42: 994–1002. 10.1175/1520-0450(2003)042<0994:ATIAOO>2.0.CO;2

Goldberg DL, Anenberg SC, Griffin D, McLinden CA, Lu Z, Streets DG (2020) Disentangling the Impact of the COVID-19 lockdowns on urban NO_2 from natural vvariability. Geophysical Research Letters, 47. https://doi.org/10.1029/2020GL089269

Grange S (2014) Technical note: Averaging wind speeds and directions. https://doi.org/10.13140/RG.2.1.3349.2006

Hakkarainen J, Szeląg ME, Ialongo I, Retscher C, Oda T, Crisp D (2021) Analysing nitrogen oxides to carbon dioxide emission ratios from space: A case study of Matimba Power Station in South Africa. Atmospheric Environment: X, 10. https://doi.org/10.1016/j.aeaoa.2021.100110

Herman J, Cede A, Spinei E, Mount G, Tzortziou M, Abuhassan N (2009) NO_2 column amounts from ground-based Pandora and MFDOAS spectrometers using the direct-sun DOAS technique: Intercomparisons and application to OMI validation. Journal of Geophysical Research: Atmospheres, 114. https://doi.org/10.1029/2009JD011848

Hickman JE, Andela N, Tsigaridis K, Galy-Lacaux C, Ossohou M, Bauer SE (2021) Reductions in NO_2 burden over north equatorial Africa from decline in biomass burning in spite of growing fossil fuel use, 2005 to 2017. Proceedings of the National Academy of Sciences, 118:7. https://doi.org/10.1073/pnas.2002579118

Judd LM, Al-Saadi JA, Janz SJ, Kowalewski MG, Pierce RB, Szykman JJ et al. (2019) Evaluating the impact of spatial resolution on tropospheric NO_2 column comparisons within urban areas using highresolution airborne data. Atmospheric Measurement Techniques, 12: 6091–6111. https://doi.org/10.5194/amt-12-6091-2019

Lourens ASM, Butler TM, Beukes JP, Van Zyl PG, Beirle S, Wagner TK, et al. (2012) Re-evaluating the NO_2 hotspot over the South African Highveld. South African Journal of Science, 108. https://doi.org/10.4102/sajs.v108i11/12.1146

Matimolane M (2019) Air Quality Offsets Implementation Plan for Gert Sibande District Municipality: Tutuka, Majuba, Camden and Grootvlei Power Stations: March 2019 update (No. ENV19-R059). https://www.eskom.co.za/AirQuality/Documents/AQoffsetImplPlanGertSibandePubMeetingPres.pdf

Naiker Y, Diab RD, Zunckel M, Hayes ET (2012) Introduction of local air quality management in South Africa: overview and challenges. Environmental Science and Policy, 17: 62–71. https://doi.org/10.1016/j.envsci.2011.11.009

NASA Pandora Project. https://pandora.gsfc.nasa.gov/Research/RNA/ Accessed 16 September.

NASA Pandora Project. https://pandora.gsfc.nasa.gov/PGN/ Accessed 18 September 2021.

Omrani H, Omrani B, Parmentier B, Helbich M (2020) Spatio-temporal data on the air pollutant nitrogen dioxide derived from Sentinel satellite for France. Data in Brief, 28. https://doi.org/10.1016/j.dib.2019.105089

Qi X, Zhu S, Zhu C, Hu J, Lou S, Xu L et al. (2020) Smog chamber study of the effects of NO_x and NH3 on the formation of secondary organic aerosols and optical properties from photo-oxidation of toluene. Science of Total Environment, 727:138632. https://doi.org/10.1016/j.scitotenv.2020.138632

Scorgie Y, Thomas R (2006) judd Mpumalanga Highveld cumulative scenario planning study: air pollution compliance assessment and health risk analysis of cumulative operations of current, RTS and proposed Eskom Power Station located within the Mpumalanga and Gauteng Provinces (No. APP/06/ESKOM-05 Rev 1.0). https://cer.org.za/wpcontent/uploads/2018/08/Airshed_Infotox_MpumalangaCumulativeReport_Rev1.pdf

Seinfeld JH, Pandis SN (2016) Atmospheric Chemistry and Physics: From Air Pollution to Climate Change. John Wiley & Sons.

Shabbir Y, Khokhar MF, Shaiganfar R, Wagner (2016) Spatial variance and assessment of nitrogen dioxide pollution in major cities of Pakistan along N5-Highway. Journal of Environmental Science, 43: 4–14. https://doi.org/10.1016/j.jes.2015.04.038

Shikwambana L, Mhangara P, Mbatha N (2020) Trend analysis and first time observations of sulphur dioxide and nitrogen dioxide in South Africa using TROPOMI/Sentinel-5 P data. International Journal of Applied Earth Observation and Geoinformation, 91. https://doi.org/10.1016/j.jag.2020.102130

Stein AF, Draxler RR, Rolph GD, Stunder BJB, Cohen MD, Ngan F (2015) NOAA'S HYSPLIT Atmospheric Transport and dispersion modeling system. Bulletin of the American Meteorological Society, 96: 2059–2078. https://doi.org/10.1175/BAMS-D-14-00110.1

Thomas R, Scorgie Y (2006) Air quality impact assessment for the proposed new coal-fired power station (Kendal north) in the Witbank area (No. APP/06/NMS-01 Rev 0.2). https://www.eskom.co.za/OurCompany/SustainableDevelopment/EnvironmentalImpactAssessments/Documents/Air_quality_part_1.pdf

Tiefengraber M, Cede A, Gebetsberger M, Müller M (2021) Fiducial Reference Measurements for Air Quality. New Algorithm and Product Development Plan (No. version 5.1). https://www.pandoniaglobal-network.org/wpcontent/uploads/2021/07/LuftBlick_FRM4AQ_NewAlgorithmPlan-ATBD_RP_2019005_v5.1.pdf

Verhoelst T, Compernolle S, Pinardi G, Lambert JC, Eskes HJ, Eichmann KU et al. (2021) Ground-based validation of the Copernicus Sentinel-5P TROPOMI NO_2 measurements with the NDACC ZSL-DOAS, MAX-DOAS and Pandonia global networks. Atmospheric Measurement Techniques, 14: 481–510. https://doi.org/10.5194/amt-14-481-2021

Walt AJ van der, Fitchett JM (2020) Statistical classification of South African seasonal divisions on the basis of daily temperature data. South African Journal of Science, 116. https://doi.org/10.17159/sajs.2020/7614

Wang G, Deng J, Zhang Y, Zhang Q, Duan L, Hao J et al. (2020) Air pollutant emissions from coal-fired power plants in China over the past two decades. Science of the Total Environment, 741. https://doi.org/10.1016/j.scitotenv.2020.140326

Wang S, Hao J (2012) Air quality management in China: Issues, challenges, and options. Journal of Environmental Sciences, 24: 2–13. https://doi.org/10.1016/S1001-0742(11)60724-9

Watanabe MDB, Ortega E (2011) Ecosystem services and biogeochemical cycles on a global scale: valuation of water, carbon and nitrogen processes, Environmental Science & Policy,14 (6). https://doi.org/10.1016/j.envsci.2011.05.013

Wedow JM, Ainsworth EA, Li S (2021) Plant biochemistry influences tropospheric ozone formation, destruction, deposition, and response. Trends in Biochemical Sciences, 1813. https://doi.org/10.1016/j.tibs.2021.06.007

Xu G, Xiao J, Oliver DM, Yang Z, Xiong K, Zhao Z, Zheng L, Fan H, Zhang F (2021) Spatio-temporal characteristics and determinants of anthropogenic nitrogen and phosphorus inputs in an ecologically fragile karst basin: Environmental responses and management strategies. Ecological Indicators, 133:108453. https://doi.org/10.1016/j.ecolind.2021.108453

Zhao X, Fioletov V, Cede A, Davies J, Strong K (2016) Accuracy, precision, and temperature dependence of Pandora total ozone measurements estimated from a comparison with the Brewer triad in Toronto. Atmospheric Measurement Techniques, 9: 5747–5761. https://doi.org/10.5194/amt-9-5747-2016

Zhao X, Griffin D, Fioletov V, McLinden C, Davies J, Ogyu A et al. (2019) Retrieval of total column and surface NO_2 from Pandora zenith-sky measurements. Atmospheric Chemistry and Physics, 19: 10619–10642. https://doi.org/10.5194/acp-19-10619-2019

Zhao X, Griffin D, Fioletov V, McLinden C, Cede A, Tiefengraber M et al. (2020) Assessment of the quality of TROPOMI high-spatial-resolution NO_2 data products in the Greater Toronto Area. Atmospheric Measurement Techniques, 13: 2131–2159. https://doi.org/10.5194/amt-13-2131-2020

Zyrichidou I, Koukouli ME, Balis D, Markakis K, Poupkou A, Katragkou E et al. (2015) Identification of surface NO_x emission sources on a regional scale using OMI NO_2. Atmospheric Environment, 101: 82–93. https://doi.org/10.1016/j.atmosenv.2014.11.023

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Technical article

Integrating project-based infrastructures with long-term greenhouse gas observations in Africa

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Abstract

There is a lack of long-term greenhouse gas (GHG) measurement infrastructures in Africa. This limits our understanding of the temporal dynamics of the biosphere-atmosphere exchange of carbon in response to climate change. Where relevant infrastructures have been established in externally funded research projects, they have often not been successfully transferred to local institutions at project termination, nor maintained in the long term. This leads to loss of capacity and continuity in primary data. We describe a collaborative approach where eddy-covariance (EC) towers for continuous long-term observation of carbon dioxide and energy fluxes were constructed under two consecutive German-funded research projects and designed to complement existing South African infrastructures. They will be transferred to partner institutions at project termination, supported by deliberate capacity building actions for long term sustainability. Joint activities were implemented to i) strengthen technical expertise for infrastructure maintenance, ii) introduce a new generation of academic scientists to the topic, iii) co-develop a training concept to enhance local capacity to continue teaching the topic, iv) improve the uptake and use of data by the research community, v) improve data use and access by stakeholders, and vi) facilitate knowledge exchange between institutions. Co-designed activities included training, apprenticeships and knowledge exchange, student exchange, co-supervision, and public outreach. Following a similar model in international research projects could significantly benefit 1) national capacity for emission inventories, 2) development of long-term GHG observation networks, and 3) the global scientific community via improved availability of data. While we specifically focus on a network of GHG observations, the principles are applicable for the infrastructure to observe other surface/atmosphere exchange processes or other long term observational infrastructure.

Keywords

capacity development, eddy covariance, research collaboration, climate change, South Africa

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Authors' contributions

All authors contributed to the study conception, design and methodology. The first draft of the manuscript was written by MB and all authors participated in revisions. In addition, JdT, GF, NEM, BM, MM, GFM, SM, GvM and CB were responsible for funding acquisition, and MB and CB for project administration. CB acted in a supervisory role and as project leader.

Introduction

The African continent makes a relatively small contribution towards the global carbon emissions from fossil fuels, however its emissions from land use and land-cover change are significant (Davis-Reddy and Vincent 2017; Kutsch et al. 2011; Valentini et al. 2014), and its vast tropical and subtropical ecosystems contribute significantly to the global carbon and water cycle (Ciais et al., 2011; Hickler et al. 2005; Scheiter and Higgins 2009). Africa was suggested to act as an overall small sink for atmospheric carbon (Valentini et al. 2014), but the estimations are uncertain due to a lack of long-term greenhouse gas (GHG) observations in many of the important ecosystems (López-Ballesteros et al. 2018; Quansah et al. 2015). Eddy Covariance (EC) towers are used to measure fluxes of trace gases, water vapour, and energy between the land surface and the atmosphere on a continuous basis. In combination with data collected by ecologists, remote sensing scientists, hydrologists, and other disciplines, flux measurements can be used to validate vegetation or ecosystem models and to understand the patterns of carbon and water fluxes between the biosphere and atmosphere. At the larger scale, coordinated tower networks can be used in quantifying the response of biomes to the impacts of climate change and land use, and in the estimation of regional carbon budgets when coupled to larger-scale climate or ecosystem models.

Most EC measurements in Africa have been associated with field campaigns of research projects, and as such, have been of limited duration (Ago et al. 2014; Brümmer et al. 2008; Quansah et al. 2015; Tagesson et al. 2015; Tagesson et al. 2016; Veenendaal et al. 2004). Collaborative international research initiatives on land-atmosphere interactions include for example the Southern African Fire-Atmosphere Research Initiative 1992 (SAFARI 92), the Southern African Atmosphere Research Initiative 1994 (SAFARI 94), and the Southern Africa Regional Science Initiative (SAFARI 2000), along with the Miombo network (Desanker et al. 1997) and several collaborative efforts by e.g. the Max Planck Institute. These collaborations have contributed significantly to the generation of new knowledge and helped to spin off further initiatives. Still, a very limited number of the established EC towers have continued long past the original project that established them. A prime example of a successfully continued, active site is the Skukuza flux tower; this is one of several sites established in 2000 as part of the SAFARI 2000 campaigns (Scholes et al. 2001; Swap et al. 2003). It is still operational and maintained by the Council for Scientific and Industrial Research (CSIR) (Khosa et al. 2019).

López-Ballesteros et al. (2018) recorded several inactive EC stations associated with past projects in their recent assessment of GHG monitoring infrastructures in Africa. This could be partially attributed to the costly maintenance of the towers and the associated demand of specialist technicians, but also to the fact that projects are often not co-designed with local partners, and sustainable capacity building activities are not sufficiently incorporated in project planning.

The abandonment of project-related EC towers leads to loss of opportunity to strengthen national infrastructures in target countries, and also loss of globally valuable long-term data. Long-term data records through networks of measurements are important for understanding complex ecosystem processes including the extent to which the land contributes towards being a carbon source or sink (Baldocchi 2014). The lack of long-term data is particularly problematic in African ecosystems, such as shrublands and savannas, because these systems are highly dependent on rainfall and periodic fires. However, these environments experience large interannual differences in precipitation and disturbance regime, and hence require long timeseries to understand trends (e.g. Ahlström et al. 2015; Brümmer et al. 2008; Brümmer et al. 2009; Davis-Reddy and Vincent 2017; Merbold et al. 2009; Veenendaal et al. 2004).

We describe one route by which an externally funded research project in Africa has attempted to address these limitations by better implementing long-term capacity building and partnerships with EC tower infrastructures. We present our approach in which site selection was designed to complement national infrastructure and align with national level processes and in which strengthening the relevant technical and academic capacities received equal attention along with the production of research outputs and suggest that such an approach could be used by other similar, externally funded research projects as well. We describe the specific South African-German research project and the main collaborating institutions, the evolution of research, knowledge generation and capacity building activities, and their value and impact within the South African applied scientific landscape. Finally, we briefly discuss the limitations of the approach and suggest general operational steps for projects that may be of use in similar future initiatives.

Project collaboration and design

South Africa hosts the majority of currently operational EC towers within Africa (López-Ballesteros et al. 2018) (Table 1). The Skukuza tower in the Kruger National Park (KNP) is the longest running African EC tower, in operation since 2000 (Feig et al. 2017). The Skukuza tower, along with another EC tower within the KNP, the Malopeni tower at Phalaborwa, operational since 2008, are maintained and operated by the CSIR. These towers have been used for numerous studies from local level physiological studies (e.g. Kutsch et al. 2008) to facilitating model (Khosa et al. 2020; Martínez et al. 2020) and remote sensing development (Khosa et al. 2019; Ramoelo et al. 2014) as well as emission inventory development (Feig 2008).

The South African Environmental Observation Network (SAEON) also runs operational towers with long-term stations established in 2016 at Cathedral Peak in the Drakensberg and in 2019 at Jonkershoek - both part of the Long-Term Ecological Research (LTER) network. SAEON is currently engaged with the development of a long-term landscape level research infrastructure (RI) in the process of establishing six long-term RI sites across South Africa (Feig 2018). The RI, termed the

Extended Freshwater and Terrestrial Observation Network (EFTEON) follows a landscape-based approach that attempts to link flux measurements at selected points representing utilized and relatively less utilized sites to a wide range of environmental and social measures, with a special focus on links to freshwater systems.

The South African-German research project 'Adaptive Resilience in Southern African Ecosystems' (ARS AfricaE) (2014-2018) installed three additional EC towers in South Africa through a collaboration of the German Johann Heinrich von Thünen Institute and local partners, the University of Venda and the Grootfontein Agricultural Development Institute (GADI) (Figure 1; Table 1). Furthermore, one additional tower, Agincourt, was installed by the CSIR during project implementation. The location and site design were chosen to complement the existing tower network in South Africa and appear to have informed the developing strategy for the EFTEON RI, given that several local partners were variously engaged in this cluster of activities (see Feig, 2017).

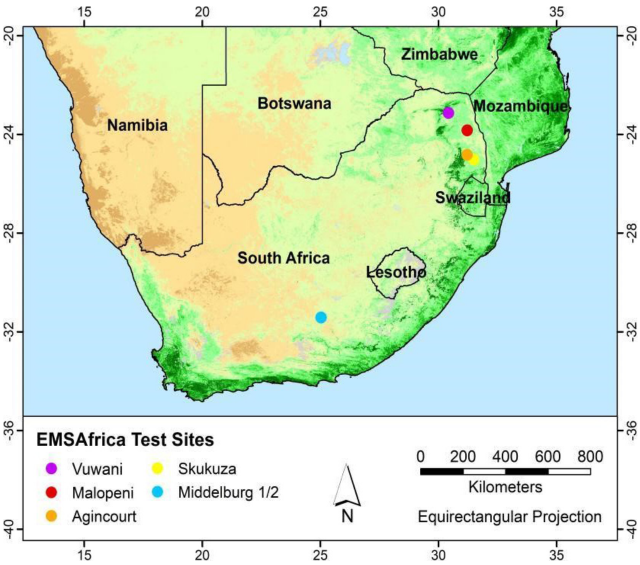

Figure 1: *The field design of the ARS AfricaE/EMSAfrica projects. Skukuza and Malopeni represent the towers that were part of the South African monitoring infrastructure, Agincourt, Vuwani and Middelburg towers were built during the ARS AfricaE project.*

The field design of ARS AfricaE and its follow-up project 'Ecosystem Management Support for Climate Change in Southern Africa' (EMSAfrica, 2018-2021) is based on six research sites, each with an EC tower as the central infrastructure. The sites are situated along a precipitation gradient, with paired sites allowing comparison between a natural-like environment with a human-modified site. This makes it possible to begin quantifying the separate and combined impacts of land use and climate on the structure and function of ecosystems. Importantly, this approach complements the previously existing tower network, where most sites were located on natural ecosystems.

Following the multidisciplinary, multi-scale approach of EMSAfrica, EC flux data are linked with a variety of on-site measurements, such as ecophysiological experiments, and coupled with remote sensing data to produce models on Southern African vegetation patterns and carbon balance (for a more detailed description of the approach, see Berger et al. 2019).

Capacity building approach and activities

In their recent assessment of an integrated strategy for an African GHG observation infrastructure, Ndisi et al. (2020) emphasise that investments in infrastructure form only one part of the process. The development of human resources capacity and long-term infrastructure maintenance strategies are of equal importance for successful operation (Ndisi et al. 2020). In this context, we define capacity building as an approach to develop

Table 1: *Summary of the currently operational long-term eddy covariance flux towers in South Africa.*

Name	Location	Operational since	Approach	Operated / established by
Skukuza	-25.02°, 31.49°	2000	Savanna / conservation area	CSIR
Malopeni	-23.83°, 31.21°;	2008	Savanna / conservation area	CSIR
Welgegund	-26.56°, 26.93°;	2010	Grassland / commercial agriculture	North-West University
Cathedral Peak	-28.98°, 29.24°	2012	High-altitude grassland	SAEON
Middelburg (1 and 2)	-31.52°, 25.01°	2015	Nama Karoo / paired setup, heavy and lenient grazing	GADI/ ARS AfricaE through Thünen Institute
Agincourt	-24.82°, 31.21°	2016	Savanna / communal area	CSIR
Vuwani	-23.14°, 30.43°	2016	Savanna / communal area	University of Venda / ARS AfricaE through Thünen Institute
Benfontein Savanna	-28.89°, 24.86°	2020	Kimberly Thornveld, paired setup, transition from Savanna-like to Nama-Karoo-like vegetation	SAEON
Benfontein Nama-Karoo	-28.86°, 24.84°	2020	Kimberly Thornveld, paired setup, transition from Savanna-like to Nama-Karoo-like vegetation	SAEON

long-term skills and commitment towards the sustainable employment of the EC infrastructures, data, and applications. Ika and Donnelly (2017) define 'conventional' means of capacity building as training, workshops, and technical advice. Capacity building conducted in association with research projects has been criticized for overemphasising the conventional view, often reduced to just training (e.g. Potter and Brough 2004).

Following Ika and Donnelly's (2017) classification, advanced forms of capacity building relate to fostering increased engagement and dialogue between stakeholders; facilitation of processes and networking belong to this type of capacity building. The key conditions to successful capacity building were proposed to include well-established commitment from stakeholders, successful collaboration, and aims based on stakeholder needs (Ika and Donnelly 2017).

Our attempt was to design the project and its capacity building activities as a collaborative effort, designed primarily to address locally identified priorities regarding training needs, data accessibility and use, and the strengthening of international collaborations and development of new projects. We used a range of conventional and advanced forms of capacity building, with strong collaborations between institutions and individuals at the centre of the approach. Our aims were to:

1. Strengthen the technical expertise required for the maintenance of the EC towers in the long term;
2. Introduce a new generation of academic students to the topic;
3. Strengthen the local capacity to teach and coach the topic;
4. Improve the uptake and use of EC data by researchers working on relevant fields;
5. Improve the access and use of the infrastructures and data by the various stakeholders;
6. Facilitate knowledge exchange between institutions in Germany and South Africa, including co-supervision of students and the development of joint scientific publications.

In the following subsections, the specific activities are described in more detail.

Training workshops on Eddy Covariance Flux Measurements

The concept of a five-day intensive course on EC flux measurements was initially based on demand defined at a 2018 meeting of the South African 'Carbon Connections' community, i.e. researchers and stakeholders working on the topic of carbon exchange. Following the recommendations, the development of the technical capacity to operate EC towers, as well as an improved capacity of researchers and data scientists to manage and utilise data were defined as priorities. The course concept was further developed based on experiences gained via a land-atmosphere interactions training workshop organised by the global research initiative 'Integrated Land Ecosystem Atmosphere Processes Study' (iLEAPS).

Funding for the three planned courses was secured via the German Ministry of Education and Research (BMBF) programme for training and knowledge exchange (coordinated by the EMSAfrica project) with significant in-kind support in personnel, equipment and the provision of facilities by the South African partners. The course aims were to enhance participants' skills and understanding of: 1) EC theory including fetch area requirements and experimental design, via lectures and practical examples; 2) operation and function principles of equipment, such as the gas analyzer and sonic anemometer, via hands-on demonstrations; 3) how to set up and operate an EC system, via hands-on sessions and demonstrations as well as Q&A sessions at a tower site; 4) technique and relevance of ancillary measurements, such as soil chamber measurements and meteorological measurements, via hands-on sessions; 5) how to process raw flux data, via demonstrations and hands-on sessions; and 6) various ways of applying EC systems and data in research projects and models. The aim was to link the mechanistic understanding to the drivers of ecosystem and atmospheric processes and drivers at a larger scale, and to increase the participants' understanding of how they can take advantage of the data in their own projects.

The course advertisement was distributed online (www.spaces-training.org) and via the networks of the collaborating institutes, as well as the project partners and stakeholders. Early-career researchers and technicians applying for the training were asked to describe how they used or were planning to use EC measurements or data in their project, and this information was used in tailoring course content. The aim was to have a combination of students planning their research career, technicians needing to strengthen their skills, and young researchers of different fields needing to understand the production of the data, to be able to apply it in their research. All course materials, background materials and additional resources were made available to the participants, many acting as lecturers and teachers at their home institutes in southern Africa.

The first of the three workshops was realised as a Winter School in 2019 under a shared academic leadership of SAEON and the Thünen Institute, with strong input and several co-lecturers from the CSIR, and contributions from a variety of collaborating universities and research institutes in southern Africa. It was hosted by the University of Venda at the Vuwani Science Resources Centre. The course was attended by 25 students from a number of southern African countries, including Angola, Botswana, Namibia, Zimbabwe and South Africa. It was realised in collaboration with the EU-African project 'Supporting EU-African Cooperation on Research Infrastructures for Food Security and Greenhouse Gas Observations' (SEACRIFOG) and the regional initiative 'Southern African Science Service Centre for Climate Change and Adaptive Land Management' (SASSCAL). Consequently, the course incorporated a strong overview of the status of overall GHG measurements in the African continent and climate adaptation and mitigation.

The second workshop was initially postponed due to the COVID-19 pandemic, and finally organised as an online intensive course on the 24.-26.11.2021. The course was again co-designed by the Thünen Institute, SAEON and CSIR partners, with invited lectures from a variety of South African and German colleagues. A total of 25 participants, representing eight different countries, attended the course, which had an even stronger emphasis on data processing and analysis. The third course (drafted for 2022) intends to incorporate a stronger input from the multidisciplinary research community, ecosystem modellers, earth observation scientists and others applying EC data into their research.

Knowledge and staff exchange

The collaboration between the Thünen Institute and partners from CSIR, SAEON and the University of Venda benefited from regular knowledge exchange and joint field visits. For the three EC towers built by the Thünen Institute together with South African partners during the project ARS AfricaE, maintenance and data transfer were conducted by the local partners with regular visits from the German team. This was in conjunction with on-site training, maintenance manuals and regular exchange, and in the case of the University of Venda, also student apprenticeships.

Southern African researchers' and students' exchanges were made possible during the EMSAfrica project via a grant programme CaBuDe (Capacity Building and Development) of the DAAD (German Academic Exchange Service), funded by the BMBF. These included four-year grants given to doctoral researchers to complete their degree in Germany, sandwich degrees for co-supervised doctoral programmes, as well as shorter grants for BSc and MSc students and varying lengths of research visits. Despite delays in the programme caused by the global COVID-19 pandemic, we used some grants for collaboratively supervised doctoral and MSc projects, as well as to strengthen collaborations and specific capacities of individuals through visits to German academic institutes. For example, two doctoral students working full-time on project-relevant topics in South Africa and Germany were both co-supervised by a South-African-German supervision committee, and field trips and research exchanges were organised both ways (see www.emsafrica.org). In addition, several research and grant proposals were initiated as a direct result of the collaborations and expanded networks between Germany and southern Africa.

Infrastructures such as EC towers can act as valuable anchor points to attract further research initiatives and projects. The Middelburg site has gained significant attention from collaborating institutions during the EMSAfrica project, both locally and internationally, and is now hosting several short- and long-term projects as a 'regional hub' of experimental research. The University of Stellenbosch has graduated one MSc student and has two further MSc students working on the site and has assigned two post-doctoral staff to provide analytical support over the past four years, funded by the South African National Research Foundation (NRF). The EMSAfrica project is also working together with local land-users through stakeholder workshops to explore ways in which its data and products can be useful to either the livestock and/or game industry, or the conservation organisations and researchers in the area. The data collected via the EC towers helps to assess the impact of livestock grazing on carbon balance in the semi-arid ecosystems (Rybchak et al. 2020) and is of long-term interest to the planning of sustainable management practices. First results, including broader discussion on land-use history and management implications, are published as a case study in an up-and-coming handbook on Southern African ecosystems, especially aimed at policy makers and those working on the science-policy interface (Rybchak et al. 2022).

All data from the EC towers hosted by the Thünen Institute are made accessible via the Open Access Data Centre (OADC) of SASSCAL (https://www.sasscal.org/prototype-oadc-open-access-data-center/) and FLUXNET (https://fluxnet.org/). Ancillary data measured by the EC towers, for example radiation and soil moisture, are used in various student projects at South African universities.

Stakeholder engagement and long-term sustainability

EMSAfrica is a multidisciplinary project, with a diverse group of stakeholders in research, governmental and non-governmental institutions in South Africa. The stakeholders are incorporated into steering the project's outputs to better contribute to sustainable land management and climate mitigation efforts and processes in South Africa via workshops and discussions. The main aim of stakeholder work at this level is to enhance knowledge exchange, to strengthen the use of existing infrastructures and resources, and to better integrate the data products into policy and decision-making frameworks.

As form of public outreach, EMSAfrica also engages school and community educators via the collaboration with the University of Venda. Vuwani Science Resource Centre, where one of the EC towers is located, is a flagship community outreach project of the University of Venda. Climate change and renewable energies are the centre's focus areas, and the EC tower adds to its uniqueness as an educational site to both students and the public. At this site, a request was presented by the site manager to produce additional EC tower information materials, for increasing the general awareness of the rural learners and community in the Limpopo province in climate change impacts and research. As a result, information posters explaining the EC tower's principles of function and links to climate change research were produced. Furthermore, a GHG demonstrator – a glass chamber with miniature landscape and an interactive "blow-in CO_2" component demonstrating the impacts of increasing GHGs – along with an explanation board, was constructed by the German team engineers as part of the physical demonstrations at the Vuwani exhibition hall.

The EMSAfrica project preceded the development of the EFTEON Landscapes, a National Network of environmental research infrastructure sites, where the site selection was based on proposals put together by research consortia (https://efteon.saeon.ac.za/landscapes/). The connections made during the EMSAfrica program acted as nodal points for the development of strong proposals, and while not all of these were successful, the networks, and thinking that went into the development of these proposals is likely to facilitate further research engagement in the future. When EMSAfrica ends in 2022, two of the recently built EC towers in Middelburg are planned to be transferred to the newly launched (2021) Stellenbosch University School for Climate Studies. This interdisciplinary academic structure seeks to link specialist skills and approaches across multiple departments and faculties to provide a platform for integrated research and training at post graduate level that is relevant to climate variability and change. An institution such as this School could integrate the infrastructures and equipment into its program and continue to develop the data-intensive, long-term research in landscape level functioning and its relevance to landscape management and policy development. The Vuwani EC tower will be transferred to the ownership of the University of Venda, remaining at the Vuwani Science Resources Centre, where it will strengthen the existing climate change measurement infrastructures, with planned research collaboration with the Stellenbosch University School for Climate Studies and technical maintenance support from EFTEON.

It is expected that the legacy from these projects will be felt in the South African Environmental Research community for the foreseeable future. The flux towers that have either been established or supported through these projects will continue providing essential long-term data in a data sparse region; the impetus for their establishment was strongly driven by these projects. In terms of the training, many of the technical staff that will continue the management of these infrastructure will

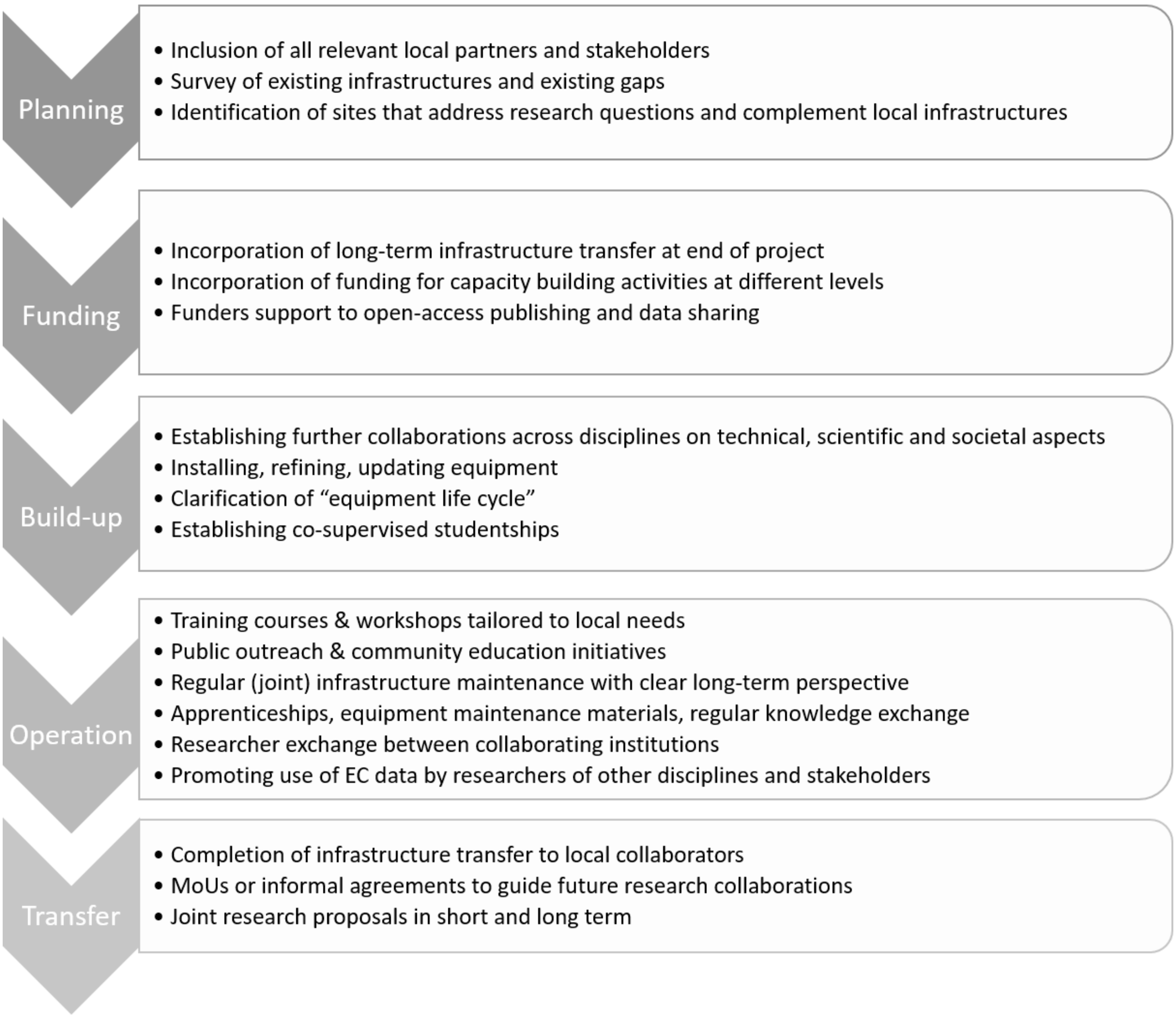

Figure 2: *Summary of the key activities at each project stage from project planning to the transfer of infrastructures to local collaborators at project end.*

have gone through the EMSAfrica training workshops, and we are beginning to see the establishment of a cohort of technically capacitated researchers able to utilise these data in South Africa. This will feed into the long-term research direction in South Africa.

The connections that have been made through these projects continue. A Memorandum of Understanding was signed between SAEON and the Thünen Institute; it facilitated exchange of students and staff, as well as the submission of joint proposals for funding calls, with plans to continue such activities also in the future.

Limitations

It must be noted that the structures and organisations present in South Africa are already well developed, which greatly improves the likelihood of success of efforts such as those described here. A key issue in maintaining tower operability after project end is the continuity of funding. As described under Section Stakeholder engagement and long-term sustainability, South Africa's recently established long-term research platforms, their associated technical personnel and further research collaborations, will be crucial in the long-term sustainability of the legacy EMSAfrica project infrastructure.

Generally, little information is available on the success of capacity building for GHG observation infrastructures in the least developed countries with poor infrastructures (see e.g. Umemiya et al. 2020). Recent initiatives, such as the integrated strategy for African GHG observation infrastructure (Ndisi et al. 2020) can help guide collaborative efforts. In the case of the EMS Africa efforts, early planning of the larger research program (SPACES) did involve technical German-South African discussions at inter-ministerial level, as well as engagement with academic leaders at least in South Africa, as the agendas were shaped. It remains to be seen if this will permit the legacy EMS Africa infrastructure to continue to serve its purpose.

Conclusions and recommendations

There is a clear urgency to develop long-term GHG observation infrastructures and technical and academic capacities related to climate change in Africa (Niang et al. 2014; Ndisi et al. 2020). We suggest that international research projects should better align their aims with building national capacities to conduct emission inventories, even when this is not the primary aim of the project. Long-term planning of infrastructures such as EC flux towers as part of regional observation networks would have a major role in helping the scientific community answer the large unanswered questions on the fluxes of carbon and water in African ecosystems. Open sharing of data and building collaborative partnerships would increase the impact and sustainability of projects and could best spur the careers of young researchers in all collaborating countries.

We summarize our collaborative approach in Figure 2, by identifying a "check-list" of key activities at the different project stages. We suggest that such a list could help other third-party funded research projects with the combining of short-term aims with sustainable outcomes.

References

Ago E.E., Agbossou E.K, Galle S., Cohard J.-M., Heinesch B., Aubinet M. 2014, Long term observations of carbon dioxide exchange over cultivated savanna under a Sudanian climate in Benin (West Africa). *Agric For Meteorol* 197: 13–25. https://doi.org/10.1016/j.agrformet.2014.06.005

Ahlström A., Raupach M.R., Schurgers G., Smith B., Arneth A., Jung M., Reichstein M., Canadell J.G., Friedlingstein P., Jain A.K., Kato E., Poulter B., Sitch S., Stocker B.D., Viovy N., Wang Y.P., Wiltshire A., Zaehle S., Zeng N. 2015, The dominant role of semi-arid ecosystems in the trend and variability of the land CO_2 sink. *Science* 348: 895–899. https://doi.org/10.1126/science.aaa1668

Baldocchi D. 2014, Measuring fluxes of trace gases and energy between ecosystems and the atmosphere – the state and future of the eddy covariance method. *Glob Chang Biol* 20: 3600–3609. https://doi.org/10.1111/gcb.12649

Berger C., Bieri M., Bradshaw K., Brümmer C., Clemen T., Hickler T., Kutsch W.L., Lenfers U.A., Martens C., Midgley G.F., Mukwashi K., Odipo V., Scheiter S., Schmullius C., Baade J., du Toit J.C.O., Scholes R.J., Smit I.P.J., Stevens N., Twine W. 2019, Linking scales and disciplines: an interdisciplinary cross-scale approach to supporting climate-relevant ecosystem management. *Clim Change* 156: 139–150. https://doi.org/10.1007/s10584-019-02544-0

Brümmer C., Falk U., Papen H., Szarzynski J., Wassmann R., Brüggemann N. 2008, Diurnal, seasonal, and interannual variation in carbon dioxide and energy exchange in shrub savanna in Burkina Faso (West Africa). *J Geophys Res Biogeosci* 113, G02030. https://doi.org/10.1029/2007JG000583

Brümmer C., Papen H., Wassmann R., Brüggemann N. 2009, Fluxes of CH4 and CO_2 from soil and termite mounds in south Sudanian savanna of Burkina Faso (West Africa). *Global Biogeochem Cycles* 23, GB1001. https://doi.org/10.1029/2008GB003237

Ciais P., Bombelli A., Williams M., Piao S., Chave J., Ryan C., Henry M., Brender P., Valentini R. 2011, The carbon balance of Africa: synthesis of recent research Studies. *Philos Trans R Soc A* 369:1–20. https://doi.org/10.1098/rsta.2010.0328

Davis-Reddy C.L., Vincent K. 2017, C*limate Risk and Vulnerability: A Handbook for Southern Africa* (2nd edition). CSIR, Pretoria, South Africa.

Desanker P.V., Frost P.G.H., Frost C.O., Justice C.O., Scholes R.J., (eds.). 1997, *The Miombo Network: Framework for a Terrestrial Transect Study of Land-Use and Land-Cover Change in the Miombo Ecosystems of Central Africa*, IGBP Report 41, The International Geosphere-Biosphere Programme, Stockholm, Sweden, 109 pp.

Feig G.T., Mamtimin B., Meixner F.X. 2008, Soil biogenic emissions of nitric oxide from a semi-arid savanna in South Africa. *Biogeosciences* 5, 1723-1738 https://doi.org/10.5194/bg-5-1723-2008

Feig G. 2018, The Expanded Freshwater and Terrestrial Environmental Observation Network (EFTEON). *Clean Air J* 28. https://doi.org/10.17159/2410-972x/2018/v28n2a14

Feig G., Joubert W.R., Mudau A.E., Monteiro P.M.S. 2017, South African carbon observations: CO_2 measurements for land, atmosphere and ocean. *S Afr J Sci* 113, 11/12. https://doi.org/10.17159/sajs.2017/a0237

Hickler T., Eklundh L., Seaquist J.W., Smith B., Ardö J., Olsson L., Sykes M.T., Sjöström M. 2005, Precipitation controls Sahel greening trend. *Geophys Res Lett* 32: L21415. https://doi.org/10.1029/2005GL024370

Ika L.A., Donnelly J., 2017, Success conditions for international development capacity building projects. *Int J Proj Manag* 35: 44–63. https://doi.org/10.1016/j.ijproman.2016.10.005

Khosa F.V., Feig G., van der Merwe M.R., Mateyisi M.J., Mudau A.E., Savage M.J. 2019, Evaluation of modelled actual evapotranspiration estimates from a land surface, empirical and satellite-based models using in situ observations from a South African semi-arid savanna ecosystem. *Agric For Meteorol* 279, 107706. https://doi.org/10.1016/j.agrformet.2019.107706

Khosa F.V., Mateyisi M.J., van der Merwe M.R., Feig G., Engelbrecht F.A., Savage M.J. 2020, Evaluation of soil moisture from CCAM-CABLE simulation, satellite-based models estimates and satellite observations: a case study of Skukuza and Malopeni flux towers. *Hydrol Earth Syst Sci* 24: 1587–1609. https://doi.org/10.5194/hess-24-1587-2020

Kutsch W.L., Hanan N., Scholes R.J., Mchugh I., Kubheka W., Eckhardt H., Williams C. 2008, Response of carbon fluxes to water relations in a savanna ecosystem in South Africa. *Biogeosciences Discussions* 5: 2197–2235. https://doi.org/10.5194/bg-5-1797-2008

Kutsch W.L., Merbold L., Ziegler W., Mukelabai M.M., Muchinda M., Kolle O., Scholes R.J. 2011, The charcoal trap: Miombo forests and the energy needs of people. *Carbon Balance Manag* 6:5. https://doi.org/10.1186/1750-0680-6-5

López-Ballesteros A., Beck J., Bombelli A., Grieco E., Lorencová E.K., Merbold L., Brümmer C., Hugo W., Scholes R.J., Vačkář D., Vermeulen A., Acosta M., Butterbach-Bahl K., Helmschrot J., Dong-Gill K., Jones M., Jorch V., Pavelka M., Skjelvan I., Saunders M. 2018, Towards a feasible and representative pan-African Research Infrastructure network for GHG observations. *Environ Res Lett* 13. https://doi.org/10.1088/1748-9326/aad66c

Martínez B., Gilabert M.A., Sánchez-Ruiz S., Campos-Taberner M., García-Haro F.J., Brümmer C., Carrara A., Feig G., Grünwald T., Mammarella I., Tagesson T. 2020, Evaluation of the LSA-SAF gross primary production product derived from SEVIRI/MSG data (MGPP). *ISPRS J Photogramm Remote Sens* 159: 220–236. https://doi.org/10.1016/j.isprsjprs.2019.11.010

Merbold L., Ardö J., Arneth A., Scholes R.J., Nouvellon Y., De Grandcourt A., Archibald S., Bonnefond J.M., Boulain N., Brueggemann N., Bruemmer C., Cappelaere B., Ceschia E., El-Khidir HaM., El-Tahir B.A., Falk U., Lloyd J., Kergoat L., Le Dantec V., Mougin E., Muchinda M., Mukelabai M.M., Ramier D., Roupsard O., Timouk F., Veenendaal E.M., Kutsch W.L. 2009, Precipitation as driver of carbon fluxes in 11 African ecosystems. *Biogeosciences* 6: 1027–1041. https://doi.org/10.5194/bg-6-1027-2009

Ndisi M.S., Kasurinen V., Kutsch W. 2020, *An integrated strategy for sustainable Africa-Europe research cooperation on greenhouse gas observations and food security*. Supporting EU-African Cooperation on Research Infrastructures for Food Security and Greenhouse Gas Observations (SEACRIFOG) Deliverable 7.1. https://www.seacrifog.eu/fileadmin/seacrifog/Deliverables/D7.1_Intergrated_concept_29.10.20_FINAL_Sameblue.pdf

Niang I., Ruppel O.C., Abdrabo M.A., Essel A., Lennard C., Padgham J., Urquhart P. 2014, *Climate Change 2014: Impacts, Adaptation, and Vulnerability. Part B: Regional Aspects*, in: Fifth Assessment Report of the Intergovernmental Panel on Climate Change. Cambridge University Press, Cambridge, UK and New York USA, pp. 1199–1265.

Potter C., Brough R. 2004, Systemic capacity building: a hierarchy of needs. *Health Policy and Planning* 19: 336–345. https://doi.org/10.1093/heapol/czh038

Quansah E., Mauder M., Balogun A.A., Amekudzi L.K., Hingerl L., Bliefernicht J., Kunstmann H. 2015, Carbon dioxide fluxes from contrasting ecosystems in the Sudanian Savanna in West Africa. *Carbon Balance Manag* 10. https://doi.org/10.1186/s13021-014-0011-4

Ramoelo A., Majozi N., Mathieu R., Jovanovic N., Nickless A., Dzikiti S. 2014, Validation of Global Evapotranspiration Product (MOD16) using Flux Tower Data in the African Savanna, South Africa. *Remote Sens* 6: 7406–7423. https://doi.org/10.3390/rs6087406

Rybchak, O., du Toit, J., Maluleke, A., Bieri, M., Midgley, G., Feig, G., Brümmer, C. 2022, A fine line between carbon source and sink – potential CO_2 sequestration through sustainable grazing management, in: Sustainability of southern African ecosystems under global change: Science for management and policy interventions (Ed. von Maltitz, G., Midgley, G.F., Brümmer, C., Rötter, R., Veste, M., Veitch, J., Viehberg, F.). Springer Ecological Studies. [Accepted]

Rybchak O., du Toit J., Delorme J.-P., Jüdt J.-K., Mukwashi K., Thau C., Feig G., Bieri M., Brümmer C. 2020, Multi-year CO_2 budgets in South African semi-arid Karoo ecosystems under different grazing intensities, *Biogeosciences Discuss*. [preprint], https://doi.org/10.5194/bg-2020-420.

Scheiter S., Higgins S.I. 2009, Impacts of climate change on the vegetation of Africa: an adaptive dynamic vegetation modelling approach. *Glob Chang Biol* 15: 2224–2246. https://doi.org/10.1111/j.1365-2486.2008.01838.x

Scholes R.J., Gureja N., Giannecchinni M., Dovie D., Wilson B., Davidson N., Piggott K., McLoughlin C., Velde K., van der Freeman A., Bradley S., Smart R., Ndala S. 2001, The environment and vegetation of the flux measurement site near Skukuza, Kruger National Park. *Koedoe* 44: 73–83. https://doi.org/10.4102/koedoe.v44i1.187

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: https://doi.org/10.1029/2003JD003747

Tagesson T., Fensholt R., Cappelaere B., Mougin E., Horion S., Kergoat L., Nieto H., Mbow C., Ehammer A., Demarty J., Ardö J. 2016, Spatiotemporal variability in carbon exchange fluxes across the Sahel. *Agric For Meteorol* 226–227: 108–118. https://doi.org/10.1016/j.agrformet.2016.05.013

Tagesson T., Fensholt R., Cropley F., Guiro I., Horion S., Ehammer A., Ardö J. 2015, Dynamics in carbon exchange fluxes for a grazed semi-arid savanna ecosystem in West Africa. *Agric Ecosyst Environ* 205: 15–24. https://doi.org/10.1016/j.agee.2015.02.017

Umemiya C., Ikeda M., White M.K. 2020, Lessons learned for future transparency capacity building under the Paris Agreement: A review of greenhouse gas inventory capacity building projects in Viet Nam and Cambodia. *J Clean Prod* 245, 118881. https://doi.org/10.1016/j.jclepro.2019.118881

Valentini R., Arneth A., Bombelli A., Castaldi S., Cazzolla Gatti R., Chevallier F., Ciais P., Grieco E., Hartmann J., Henry M. 2014, A full greenhouse gases budget of Africa: synthesis, uncertainties, and vulnerabilities. *Biogeosciences* 11: 381–407. https://doi.org/10.5194/bg-11-381-2014

Veenendaal E.M., Kolle O., Lloyd J. 2004, Seasonal variation in energy fluxes and carbon dioxide exchange for a broad-leaved semi-arid savanna (Mopane woodland) in Southern Africa. *Glob Chang Biol* 10: 318–328. https://doi.org/10.1111/j.1365-2486.2003.00699.x