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Contents

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Editorial
South Africa's electricity disaster is an air quality disaster, too

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South Africa's electricity supply constraints were declared a State of Disaster on 9 February 2023, coinciding with the 102nd consecutive day of power cuts, euphemistically called 'load shedding'. Load shedding dates back to late 2007, but increased more than fourfold in 2022 relative to 2021, occurring on 43% of hours in the year (Pierce and le Roux, 2023).

Eskom, the national electricity utility, is the highest profile emitter of air pollution in South Africa (Euridídou et al., 2022), but other sources of air pollution increase when coal-fired power stations are down. Generators running on diesel or petrol are fired up by businesses and higher-income households during load shedding. In many lower-income residential areas, load shedding is exacerbated by the tripping of overloaded distribution networks and the slow response time of the utilities that de-prioritise restoration in municipalities with dubious payment histories. Such communities, in which large proportions of households already use dirty fuels (e.g. wood, coal, paraffin) to meet some of their energy needs, now become even more reliant on these. The use of these fuels greatly contributes to poor ambient and indoor air quality (e.g., Pauw et al., 2022; Walton et al., 2021; Wernecke et al., 2021; Wernecke et al., 2015, Hersey et al., 2015, Naidoo et al., 2014; Moletsane et al., 2021). These generator and residential burning emissions are of course released in very close proximity to people, have a high inhalation intake fraction and represent an increased health risk to humans and the environment. Thus, during load shedding, the air pollution from energy generation shifts to local sources which can lead to short-term peaks in ambient pollution.

Returning to the power stations, the shortage and more recently low availability of generating capacity have long been grounds to keep coal-fired units running with emissions above the licence limits. Since 2008, Eskom's power stations have been applying to the licensing authorities for short-term exemptions from some emission limits to allow them to continue operating in the event of an equipment malfunction or breakdown. The lack of sufficient capacity and shortage of funds integral to the electricity crisis then formed the main justification for Eskom's applications for postponement and then the suspension of the Minimum Emission Standards. Now in 2023, none of Eskom's coal-fired power stations are fully compliant with the emission limits that came into effect in 2020 and were first gazetted in 2010. Power stations are emitting SO_2 at levels 3-6 times higher than the Minimum Emission Standards limit.

The declaration of the State of Disaster means that even the more lenient emission limits and controls that have not been waived due to exemptions and postponements are no longer in effect. The Electricity National State of Disaster regulations published on 28 February 2023 allow the Minister to exclude 'repairs … and existing generation … facilities from the provisions of the National Environmental Management Act, or any specific environmental management Act … for the duration of the national state of disaster' (paragraph 5(1)(i)). While the relaxing of environmental controls no doubt has the potential to assist with short-term improvements in plant availability and consequently electricity supply, the neglect permitted will probably lead to longer-term declines in the state of the plant, which is in direct conflict with one of the stated objectives of the State of Disaster to 'protect property', and will increase air pollution in the region. It is critical that this relaxation is only temporary.

The current inoperation of units 1-3 at Kusile Power Station, Eskom's newest and "cleanest" coal-fired station, illustrates how running a unit too hard can contribute to a plant failure that can result in a much longer outage time and much greater cost of repairs than would have been the case had the unit been taken down when problems were first detected. The three units at Kusile have been out of commission since 23 October 2022, when the flue duct exiting unit 1's flue gas desulphurization (FGD) plant to the stack failed and at the same time compromised the flue ducts for units 2 and 3 (Creamer, 2023). Eskom intends to build temporary stacks that will bypass the FGD and operate the three units using these flues for a year or so while the permanent flues and chimney are repaired. These stacks are likely to be considerably shorter than the existing 220-metre stacks at

Kusile. The combination of the SO_2 emissions that are an order of magnitude higher without the FGD and the shorter stacks resulting in poorer dispersion will greatly increase ambient SO_2 concentrations in the vicinity of the power station. This is an area with already unacceptably high SO_2 concentrations, as was pointed out in the Air Quality Impact Assessment conducted prior to the construction of Kusile (Thomas and Scorgie, 2006).

So where to from here? It seems the only thing worse than having an electricity supply dominated by coal generation (80% in 2022; Pierce and le Roux, 2023) is having coal-fired power stations that do not supply electricity and perform badly. The availability of Eskom's fleet has declined from around 85% in 2008 (Eskom, 2011) to 58% in 2022 due to 30% of the fleet being on unplanned outages during the last year (Pierce and le Roux, 2023). Relative particulate (ash) emissions, a good indication of the performance of the abatement technology, more than doubled between 2008 (0.21 kg/MWh sent out) and 2020 (0.47 kg/MWh sent out), although they declined again by 2022 to 0.34 kg/MWhSO (Eskom, 2022; 2011). Prioritising production over the integrity of the plant for the last 15 years is one of the reasons that the coal-fired fleet is so unreliable today, and the State of Disaster will only exacerbate this practice. It is very well established that air pollution leads to thousands of premature deaths in South Africa a year (Altieri and Keen, 2019; Marais et al., 2019 ; Langerman and Pauw, 2018). The cost of the emissions from both residential energy use and Eskom's power stations cannot be ignored. While South Africa still depends on a fleet of coal-fired power stations, it is crucial it be properly maintained and responsibly operated to avoid exacerbating the already poor air quality in the Highveld.

References

Altieri, K.E. and Keen, S.L. (2019). Public health benefits of reducing exposure to ambient fine particulate matter in South Africa. *Sci Total Environ*. Sep 20;684:610-620. https://doi.org/10.1016/j.scitotenv.2019.05.355

Creamer, T. (2023). Temporary Kusile stacks set to bypass FGD for a year while remediation works are completed. *Engineering News*. https://www.engineeringnews.co.za/article/temporary-kusile-stacks-set-to-bypass-fgd-for-a-year-while-remediation-works-are-completed-2023-01-30

Eskom. (2011). Integrated Report 2011. Eskom Holdings SOC Limited, Sunninghill. https://www.eskom.co.za/wp-content/uploads/2021/10/eskom-ar2011.pdf

Eskom. (2022). Integrated Report 2022. Eskom Holdings SOC Limited, Sunninghill. https://www.eskom.co.za/wp-content/uploads/2022/12/2022_integrated_report.pdf

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. *Atmos. Chem. Phys*., 15, 4259–4278. https://doi.org/10.5194/acp-15-4259-2015

Langerman, K. E., & Pauw, C. J. (2018). A critical review of health risk assessments of exposure to emissions from coal-fired power stations in South Africa. *Clean Air Journal*, 28(2). https://doi.org/10.17159/2410-972X/2018/v28n2a19

Marais, E.A., Silvern, R.F., Vodonos, A., Dupin, E., Bockarie, A.S., Mickley, L.J. and Schwartz, J. (2019). Air quality and health impact of future fossil fuel use for electricity generation and transport in Africa. *Environmental Science & Technology*, 53(22), 13524-13534. https://doi.org/10.1021/acs.est.9b04958

Moletsane, S. D., Lindeque, F., Language, B., Ncobile C.Nkosi, Adesina, J. A., Burger, R. P., Mkhatshwa, G., & Piketh, S. J. (2021). Intra-urban variability of $PM_{2.5}$ in a dense, low-income settlement on the South African Highveld. *Clean Air Journal*, 31(1). https://doi.org/10.17159/caj/2021/31/1.9413

Naidoo, S., Piketh, S., & Curtis, C. (2014). Quantification of Emissions Generated from Domestic Burning Activities from Townships in Johannesburg. *Clean Air Journal*, 24(1), 34–40. https://doi.org/10.17159/caj/2014/24/1.7047

Pauw, C., Murray, H. M. ., & Howard, M. A. (2022). The use of dirty fuels by low-income households on the South African Highveld. *Clean Air Journal*, 32(1). https://doi.org/10.17159/caj/2022/32/1.12508

Pierce, W. and le Roux, M. (2023). Statistics of utility-scale power generation in South Africa 2022. CSIR Energy Centre. https://www.csir.co.za/sites/default/files/Documents/Statistics%20of%20power%20in%20SA%202022-CSIR-%5BFINAL%5D.pdf

Thomas, R. and Scorgie, Y. (2006). Air quality impact assessment for the proposed new coal-fired power station (Kendal North) in the Witbank area, Report No.: APP/06/NMS-01, Airshed Planning Professionals.

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

Wernecke, B., Burger, R. P., Language, B., Wright, C. Y., & Piketh, S. J. (2021). Quantifying potential particulate matter intake dose in a low-income community in South Africa. *Clean Air Journal*, 31(2). https://doi.org/10.17159/caj/2021/31/2.9426

Wernecke, B., Language, B., Piketh, S., & Burger, R. (2015). Indoor and outdoor particulate matter concentrations on the Mpumalanga highveld – A case study. *Clean Air Journal*, 25(2), 12. https://doi.org/10.17159/2410-972X/2015/v25n2a1

Commentary
Air quality risks pertaining to tailings storage facilities within the Witwatersrand Goldfields

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President Nelson Mandela at the 104th Annual General Meeting of the South African of the Chamber of Mines on the 8th of November 1994 announced that: "*South Africa is blessed with a special geological heritage. As such, the mining industry has been the bedrock of the South African economy for more than a century*".

Mining also creates residuals (tailings). Research has shown that tailings particles showed inherent in vitro toxicity based on physicochemical properties (Andraos & Gulumian, 2021).

Mine tailings are stored in Tailings Storage Facilities. Tailings Storage Facilities (TSFs) are engineered structures that are designed to contain tailings (residue following the extraction of valuable material from metal ore processing) and to manage associated water. TSFs are among the largest dams and structures in the world and will stand in perpetuity.

The Witwatersrand has been mined for more than a century. It is the world's largest gold and uranium mining basin with the extraction, from more than 120 mines, of 43 500 tons of gold in one century and 73 000 tons of uranium between 1953 and 1995, which led to a legacy of some 400 km^2 of TSFs and 6 billion tons of pyrite tailings containing 600 000 tons of uranium (Chevril et al, 2008).

Pollution related to Witwatersrand TSFs poses a number of hazards to surrounding communities. The major primary pathways by which contamination can enter the environment from TSFs are:

- the airborne pathway, where radon gas and windblown dust disperse outwards from mine sites,
- the waterborne pathway, either via ground or surface water or due to direct access, where people are contaminated,
- or externally irradiated after unauthorized entry to a mine site,
- by living in settlements directly adjacent to TSFs or in some cases, living in settlements on the contaminated footprints of abandoned mines (Sutton, 2007).

The Department of Agriculture and Rural Development in its 2011 Report, titled "*Feasibility Study on Reclamation of Mine Residue, Areas for Development Purposes: Phase II Strategy and Implementation Plan*" also identified air-quality, with particular reference to dust pollution from TSFs (including radioactive dust) as one of the three main issues relating to TSFs within the Witwatersrand (Department of Agriculture and Rural Development, 2011).

Dust concentrations of up to 3 700 mg per m^3 of air were reported from areas adjacent to TSFs of the East Rand during a windy day (Coetzee et al, 2006).

Hazards relating to the airborne pathway include particulate matter (PM_{10} and $PM_{2.5}$), total suspended particulates, dust fall, silica quartz, iron pyrite and elements with concentrations higher than the average crustal soil such as gold, arsenic, lead, mercury, cyanide, etc. (Annegarn, 2021).

With reference to dust fall, the National Dust Control Regulations of 1 November 2013 prescribe a dust fall rate for residential areas of no more than 600 mg/m^2 per day, with a permitted frequency of two exceedances within a year but not in sequential months. Dust fall within non-residential areas may not exceed 1200 mg/m^2 per day, with a permitted frequency of two exceedances a year, not in sequential months. A residential area is defined as an area classified for residential use in terms of the local town planning scheme while a non-residential area is defined as an area not classified for residential use in terms of the local town planning scheme. The National Dust Control Regulations do not include any penalties for exceedances. Penalties only apply to a failure to prepare, submit and implement an air quality management plan.

The proposed amendments to the National Dust Control Regulations of 2023 will require mining operations to develop and submit a dust management plan to an Air Quality Officer for approval **prior** to commencement of the operation or activity and not only if there were exceedances. Furthermore, the proposed amendments now include in its definition of "residential area", informal settlement areas, where no zoning is in place (Department of Forestry, Fisheries and Environment, 2023).

Notwithstanding Regulations 17(6) to 17(10) of the Mine Health and Safety Act Regulations, under the heading "*Safety Precautions*", which prescribe a horizontal distance (buffer zone) of 100 metres from "*dams, waste dumps or any other structure whatsoever including structures beyond mining boundaries*" and the Department of Mineral Resources and Energy's rule

to extend the prescribed 100 metres to 2 000 metres, housing developments, both formal and informal, continue to encroach onto land close to TSFs (Department of Mineral Resources and Energy, 2015). The housing and population sprawl near TSFs in the Witwatersrand is exacerbating human exposure to windblown dust (Kneen et al, 2015).

Analysis of monitoring campaign data has confirmed multiple occurrences of quartz rich inhalable dust in residential settings at levels that exceed occupational health standards. Research by Kneen et al. (2015) indicates that the finer milling used for modern gold extraction results in aeolian dust emanating from the TSFs, which contributes to a higher proportion of inhalable particles in the source material. Air quality dispersion modelling, validated by ambient aerosol monitoring campaigns, indicates that episodic dust events generate particulate matter (PM_{10}) and, specifically, quartz dust concentrations that are unhealthy at distances of up to 2 km downwind from TSFs (Kneen et al, 2015).

As a consequence of the uraniferous nature of the ore, Witwatersrand tailings often contain significantly elevated concentrations of uranium and its daughter radionuclides, with the decay series of U^{238} being dominant. Mining has resulted in the dispersal of radioactive material into the environment via *inter alia* windblown dust (Coetzee & van Tonder, 2008).

The eastern catchment of the Mooi River, also known as the Wonderfonteinspruit, has been identified in a number of studies as the site of significant radioactive and other pollution, generally attributed to the mining and processing of uraniferous gold ores in the area. An assessment of radiological impacts of mining activities in the Wonderfonteinspruit Catchment Area (WCA)[1] was carried out on behalf of the National Nuclear Regulator (NNR)[2] by BS Associates in 2007.

Although the potential radiation exposures caused by emissions of radon and contaminated dust from mining legacies were outside the scope of the investigations, it was found that "*significant radiation exposure can occur in the surroundings of mining legacies, due to:*

- *Inhalation of Rn-222 daughter nuclides from radon emissions of desiccated water storage dams and slimes dams.*
- *The inhalation of contaminated dust generated by wind erosion from these objects, and*
- *The contamination of agricultural crop (pasture, vegetables) by the deposition of radioactive dust particles, which can cause considerable dose contributions via ingestion.*

"Due to the small particle size of the slimes, particulate matter can be transported over relatively long distances to agriculturally used land in the surroundings… the deposition of radioactively contaminated dust on leaves of vegetable and forage plants can cause radiation exposures exceeding those from the "inhalation of contaminated dust" substantially, being in the order of dose contribution of the so-called 'water pathways'" (National Nuclear Regulator, 2007).

In any pollution scenario, it is important to understand the risk posed by the pollution to the local human population hence it begs the question: how dangerous is uranium and its radioactive progeny to human health? High confidence scientific research has found that uranium is capable of spontaneously giving off energy and particles that can break chemical bonds and damage living cells – this property is termed radio-toxicity. Alpha emitters, once deposited inside the body can irreparably damage adjacent tissue and result in mutagenic defects and other malign transformations. In the light of new findings, uranium apart from kidneys, attacks the brain, acts as an endocrine disruptive compound by mimicking oestrogen (with possible consequences for the foetal development), compromises the immune system and damages the DNA (Brugge & Buchner, 2011; Institute de Radioprotection et de surete nucleaire, 2005; Zaire et al, 1997).

With reference to the health effects of the inhalation of uranium particles, research has found that small particles are carried by the inhaled air stream all the way into the alveoli. Here the particles can remain for periods from weeks up to years depending on their solubility. Highly insoluble uranium compounds may remain in the alveoli, whereas soluble uranium compounds may dissolve and pass across the alveolar membranes into the bloodstream, where they may exert systemic toxic effects. In some cases, insoluble particles are absorbed into the body from the alveoli by phagocytosis into the associated lymph nodes. Insoluble particles may reside in the lungs for years, causing chronic radiotoxicity to be expressed in the alveoli (Coetzee et al, 2004).

Stormwater drainage systems, into which windblown dust from adjacent slimes dams is flushed by run-off from sealed surfaces are also likely to constitute a major source of potential water pollution. Based on conservative assumptions regarding the affected surface area and average deposition rates of dust from adjacent slimes dams, it was estimated that approximately 10 tons of particle-bound uranium per year are flushed by stormwater into receiving watercourses (Coetzee et al, 2006).

In the light of the abovementioned findings, it logically follows that tailings storage facilities and tailings within the Witwatersrand goldfields represent a significant risk to local communities and ecosystems, especially in downwind and

[1]The Wonderfonteinspruit Catchment Area is a densely populated area and is extensively used for irrigation of edible crops, watering of cattle, spiritual rituals such as baptisms, recreational and domestic use and at times for drinking purposes due to the erratic supply of water (Liefferink, 2015)

[2]The NNR is mandated to provide for the protection of persons (the public and workers), property and the environment against nuclear damage as the competent authority for nuclear regulation in South Africa

downstream environments. It is therefore incumbent upon mining companies within the Witwatersrand to prevent the airborne risks associated with tailings and tailings storage facilities; to implement best practices in the design, construction, operation, maintenance, monitoring and management of the tailings storage facilities through all phases of a facility's lifecycle, including care and maintenance, decommissioning, closure and post-closure; and to disclose relevant information to communities at risk to support public accountability.

During the South African Human Rights Commission's (SAHRC) National Hearing on the Underlying Socio-Economic Challenges of Mining Affected Communities in South Africa in 2016 and the SAHRC's consultations with affected communities one of the most crucial factors raised with respect to environmental impacts was the increased levels of dust and the impact of these factors on food security, health, and overall conditions of well-being.

Pursuant to the SAHRC's National Hearing, the Commission issued the following directives to the relevant organs of state:

- The Department of Environmental Affairs (DEA) (now the Department of Forestry, Fisheries and Environment), in cooperation with the Department of Cooperative Governance and Traditional Affairs (COGTA) and the South African Local Government Association (SALGA), is directed to conduct an audit of all provincial governments and municipalities to confirm:

a. Whether all municipalities have developed and incorporated an air quality management plan into their Integrated Development Plans; and

b. Whether all provincial Members of the Executive Council and municipalities have appointed an air quality officer in line with the National Environmental Management Air Quality Act (NEMAQA).

- Noting the reported lack of certainty around the applicability of NEMAQA to mining activities, the Department of Mineral Resources (DMR) (now the Department of Mineral Resources and Energy) together with the DEA are directed to issue a formal notice clarifying the requirements. A copy of this public notice must be submitted to the SAHRC within three months from the release of this Report and must be accompanied by a report outlining measures taken to ensure that all industry role players are adequately made aware of the requirements.
- The DEA and DMR must jointly report on the measures taken to streamline the control of the cumulative air pollution impacts of mining operations. This report must outline the mechanisms that have been put in place for collation, verification and dissemination of information between stakeholders in relation to impacts reported and/or interventions undertaken in relation to air quality.

It is imperative that the SAHRC, whose Constitutional Mandate requires it to promote respect for human rights and a culture of human rights; promote the protection, development and attainment of human rights; and monitor and assess the observance of human rights in the South Africa, to ensure the compliance of the above-mentioned directives by the relevant organs of state, and in the absence of which, to legally enforce non-compliances. After all, Section 24 of the Constitution of the Republic of South Africa enshrines the right of "*everyone … to an environment that is not harmful to health and well-being*" (The Constitution, 1996).

References

Andraos, C & Gulumian, M. National Institute for Occupational Health. "Hazard Identification and Exposure Assessment of Dust Emissions from South African Gold Mine Tailings Sites". 2021.

Annegarn, H, School of Geo and Spatial Sciences, North West University, Potchefstroom. Presentation on "Monitoring and measurement of mine dust emissions, with a focus on windborne dust from gold tailings". GCRF Mine Dust & Health Network: Mine Dust and Gold Tailings Thursday, 7 October 2021 Virtual Seminar as part of the NACA 2021 Annual Conference.

Brugge, D, & Buchner, V. Department of Public Health and Family Medicine, Tufts University School of Medicine, "Health effects of uranium: new research findings". 2011.

Chevrel, S, Croukamp, L, Bourguignon, A & Cottard, F 2008, A Remote Sensing and GIS Based Integrated Approach for Risk Based. Prioritization of Gold Tailings Facilities — Witwatersrand, South Africa, in A Fourie, M Tibbett, I Weiersbye & P Dye (eds), Proceedings of the Third International Seminar on Mine Closure, Australian Centre for Geomechanics, Perth, pp. 639–650.

Coetzee, H, Winde F & Wade, PW 2004, An Assessment of Sources, Pathways, Mechanisms and Risks of Current and Potential Future Pollution of Water and Sediments in Gold-mining Areas of the Wonderfonteinspruit Catchment, Water Research Commission Report No 1214/1/06, pp. iv, 21 & 119-165.

Coetzee, H & van Tonder, D 2008, Draft Regional Mine Closure Strategy for the West Rand Goldfield, Council of Geosciences, report no. 2008-0253, Pretoria.

Department of Agriculture and Rural Development 2011, Feasibility Study on Reclamation of Mine Residue. Areas for Development Purposes: Phase II Strategy and Implementation Plan, Technical report no. 788/06/01/2011, Gauteng.

Department of Forestry, Fisheries and the Environment. Government Notice. 2023. National Environmental Management: Air Quality Act, 2004 (Act No. 39 Of 2004). Proposed Amendments to the National Dust Control Regulations.

Department of Mineral Resources and Energy. Mine Health and Safety Regulations under heading "Safety Precautions". 10 July 2015.

Institute de Radioprotection et de surete nucleaire, 2005. Envirhom: Bioaccumulation of radioncuclides in situations of chronic exposure of ecosystems and members of the public. Progress report 2 covering the period June 2003 – September 2005. Report DRPH 2005-07 & DEI 2005-05. pp. 64

Kneen, MA, Ojelede, ME & Annegarn, HJ 2015, "Housing and population sprawl near tailings storage facilities in the Witwatersrand: 1952 to current", South African Journal of Science, vol. 111.

Liefferink, S, 2015. "Determining attainable ecological quality requirements for the Upper Wonderfonteinspruit Catchment, based on human community requirements: The case of Bekkersdal". Dissertation submitted in fulfilment of the requirements for the degree Magister Scientiae in Environmental Sciences at the Potchefstroom Campus of the North-West University.

National Nuclear Regulator, 2007. "Radiological Impacts of the Mining Activities to the Public in the Wonderfonteinspruit Catchment Area", report TR-RRD-07-0006.

Reinhard Z, Notter, M, Riedel, W & Thiel, E. 1997. "Unexpected rates of chromosomal instabilities and alterations of hormone levels in Namibian uranium miners". Radiation Research 147, 5, 579 – 584.

South African Human Rights Commission. "National Hearing on the Underlying Socio-economic Challenges of Mining-affected Communities in South Africa". 13-14 September; 26 and 28 September; 3 November 2016.

Sutton, MW 2007, "An environmental risk assessment of gold mine residue deposits (MRDs) and reclaimed footprints in the East Rand, South Africa – with recommendations for safe land use options". Dissertation submitted in fulfilment of the requirements for the degree Magister Scientiae in Environmental Sciences at the University of the Witwatersrand.

The Constitution of the Republic of South Africa, Act 108 of 1996.

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Sustainable Development Goals

News

Initiating activities to tackle the health impacts from air pollution in East Africa: bringing together research, policy and practice

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Air pollution is a global threat to human health, especially in low- and middle-income countries such as those on the African continent. In more than half of the countries across Africa, annual average concentrations of fine particulate matter ($PM_{2.5}$) are over 35 µg/m^3, the least stringent interim target set by the World Health Organization (WHO, 2021) (Figure 1). This indicates the drastic problem facing human health across Africa in relation to air pollution exposure and subsequent adverse health impacts.

Countries in East Africa experience high levels of air pollution as well as significant health impacts from both outdoor and household air pollution. According to the *State of Air Quality and Health Impacts in Africa* (2022) report, published by the Health Effects Institute (HEI) in partnership with the Institute for Health Metrics and Evaluation (IHME), Eastern Africa has the highest proportion of the population (95%) relying on solid fuels for cooking. Furthermore, the levels of ozone in the region are steadily increasing (HEI, 2022). In good news, deaths linked to ambient and household $PM_{2.5}$ exposure in Ethiopia and Rwanda have been decreasing and steady over the past 5 years, respectively (Fisher et al., 2021). Public and governmental interest in the topic is growing in the region, and there is also a greater demand for data and evidence on air pollution levels and trends as well as associated health effects. There is also a growing focus on vulnerable groups such as unborn children, infants, children under 5 years of age, pregnant women, older people, people with pre-existing diseases and minority groups.

In this context, targeted interventions can play an important role in improving air quality and alleviating the associated public health impacts.

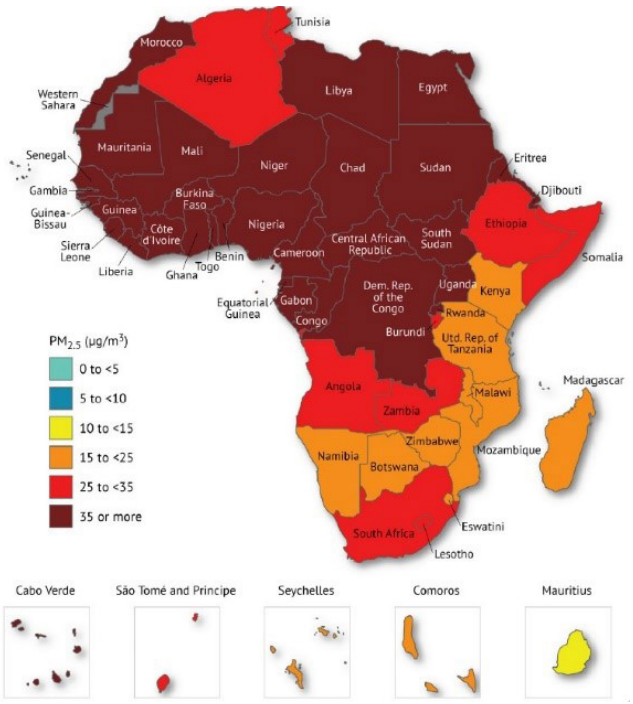

Figure 1: *Annual average $PM_{2.5}$ exposures in Africa in 2019 (HEI, 2022).*

Recent workshop held in Nairobi

Convened in March 2023, the *Workshop on Air Pollution and Health* in East Africa aimed to review the status of current data and evidence on air quality and associated health effects in the region and its interlinkage to current policy debate and actions. Discussions also aimed at coming up with concrete strategies for collaboration and strengthening technical expertise on air pollution and health in East Africa. The workshop was organized by Health Effects Institute in partnership with the Stockholm Environment Institute – Africa Centre (SEI Africa), World Resources Institute (WRI Africa), Eastern Africa GEOHealth Hub (Kenya) and AirQo.

The workshop was attended by more than 50 researchers, policy-makers, and other key researchers and practitioners from across the East African region (Figure 2). Dr. Alice Kaudia from the Climate and Clean Air Coalition spoke about the need for an integrated approach to addressing air pollution and climate change with a focus on improvements in public health and engagement across sectors.

Some pertinent messages were drawn out during the two-day workshop. Cost estimates of air pollution exposures on human health are needed to drive air pollution action; increased public understanding of air pollution and its health risks is a priority; and identifying viable, cost-effective pathways to meeting the WHO Air Quality Guidelines for criteria pollutants is important for the region.

The group also highlighted the need to include health as a central pillar in decision-making related to air pollution. Almost all organs, systems and processes in the human body are affected by air pollution. Air pollution-related health effects include respiratory, cardiovascular, and cardiorespiratory diseases, as well as trachea, bronchus and lung cancers, pre-term birth and low birth weight, type 2 diabetes, and dementia, among others (Schraufnagel at el., 2019). Short-term exposure to air pollution has been associated with aggravated asthma, ear, nose and throat irritations, and

Figure 2: *Workshop group photo taken in Nairobi, Kenya. Credit: Paul Kaberere*

increased emergency room visits (Manisalidis, 2020). Long-term exposure can lead to a range of outcomes including stroke, cataract, obesity, Alzheimer's disease and also death (Manisalidis, 2020).

Through group discussions as well as a breakout session, participants discussed themes and topics that are aligned to short- and long-term research and policy priorities in air pollution and health research in East Africa as well as what would it take to address them. For example, one suggestion for a medium-term goal was to establish an East Africa air pollution and health repository where both air pollution and health data are housed to facilitate easier access to data for assessing the situation against the East Africa Standard (EAS) 1047 (UNEP, 2022) which all Eastern African countries have adopted.

During the workshop, it was also evident that policymakers in the region are engaged on issues related to air quality and are keen to use research evidence for decision-making. A key theme in this context was the need for implementation-based science, and opportunities to address key questions being faced by policymakers rather than science for the sake of science.

Next steps

The deliberations at the workshop will feed into a research agenda for the region and partnerships that will deliver cleaner for all living in these countries. We hope that the connections and conversations lead to a sharper focus on air quality policies and actions on those most beneficial to public health in the region. Materials from the workshop including slides, recordings and a workshop summary are available on the HEI website. HEI also invites suggestions and ideas from the broader community.

Conflicts of interest / Declaration

Dr. Caradee Wright serves on the HEI Global Health Oversight Committee which provides advice and input into the work undertaken by HEI's Global Health program.

References

Fisher S, Bellinger DC, Cropper ML, Kumar P, Binagwaho A, Koudenoukpo JB, Park Y, Taghian G, Landrigan PJ. Air pollution and development in Africa: impacts on health, the economy, and human capital. Lancet Planet Health. 2021 Oct;5(10):e681-e688. DOI: 10.1016/S2542-5196(21)00201-1

Health Effects Institute. 2022. The State of Air Quality and Health Impacts in Africa. A Report from the State of Global Air Initiative. Boston, MA:Health Effects Institute. Manisalidis I, Stavropoulou E, Stavropoulos A, Bezirtzoglou E. Environmental and Health Impacts of Air Pollution: A Review. Front Public Health. 2020 Feb 20;8:14. doi: 10.3389/fpubh.2020.00014.

Manisalidis I, Stavropoulou E, Stavropoulos A, Bezirtzoglou E. Environmental and Health Impacts of Air Pollution: A Review. Front Public Health. 2020;8:14. DOI: 10.3389/fpubh.2020.00014.

Schraufnagel DE, Balmes JR, Cowl CT, De Matteis S, Jung SH, Mortimer K, Perez-Padilla R, Rice MB, Riojas-Rodriguez H, Sood A, Thurston GD, To T, Vanker A, Wuebbles DJ. Air Pollution and Noncommunicable Diseases: A Review by the Forum of International Respiratory Societies' Environmental Committee, Part 2: Air Pollution and Organ Systems. Chest. 2019 Feb;155(2):417-426. doi: 10.1016/j.chest.2018.10.041.

UNEP. 2022. East Africa Sub-Region becomes second Sub-Region in Africa to adopt Euro 4/IV Equivalent Vehicle Emission Standards. Available at: https://www.unep.org/events/workshop/east-africa-sub-region-becomes-second-sub-region-africa-adopt-euro-4iv-equivalent (Accessed 7 April 2023)

WHO. 2021. Air Quality Guidelines. September 2021, World Health Organization, Geneva, Switzerland.

Erratum
The launch of the first-ever Integrated Assessment of Air Pollution and Climate Change for Sustainable Development in Africa

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There was an omission of an author in this News piece. The lead author (Alice Kaudia) wishes to add Cynthia Sitati to the author list. The correct author list for this News piece is above.

Research article

Studies into the reduction of domestic fuel burning emissions by means of facile catalytic abatement technology

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Abstract

The negative health and socio-economic impacts of emissions associated with domestic fuel burning are widely recognized. Although there has been much progress in the provision of electricity to households in South Africa, many still rely on solid fuel sources such as wood and coal. While various investigations have been done on reducing household emissions by reducing the use of polluting fuels and improvements in combustion efficiency, comparatively fewer studies have been conducted on the reduction of emissions through use of abatement technology. Catalytic oxidation could be utilized to oxidize particulate matter precursors such as volatile organic compounds and soot particles to reduce secondary particulate formation. Although catalytic methods have not been effectively utilized in practical domestic applications, studies have shown effective soot reduction during laboratory testing. This study investigated the synthesis and use of a manganese oxide based catalyst to reduce particulate matter from domestic fuel burning stoves. The catalyst was synthesized onto a mesh substrate and inserted into the flue of the stove. During field testing, the presence of the catalyst increased the mass of particulate matter collected onto PTFE filters used for gravimetric analysis, with Scanning Electron Microscopy (SEM) analysis showing spherical particles in the pores of the filters used during the catalytic runs. The baseline runs had very few of these particle clusters. Energy Dispersive X-Ray (EDX) analysis of the catalyst run filters did not detect manganese, revealing that increased particulate concentrations were not as a result of macroscopic particles of the catalyst being dislodged from the support. Dislodgement of very small metal particles from the catalyst could, however serve as nucleation nodes for particle growth which would have a non-metal coating leading to the non-detection of manganese. The increase in particulate matter could also be caused by the impingement of particulate matter precursors on the catalyst followed by particle growth and dislodgement into the flue gas. The testwork showed that an active catalyst can be synthesized onto a mesh catalyst support in a relatively simple and cost-effective manner, which can be utilized in domestic fuel burning devices. It is recommended that a range of optimized, potentially active catalysts be tested to improve the oxidation of particulate matter precursors to carbon dioxide.

Keywords

household air pollution; particulate matter; domestic fuel burning; catalytic reduction

Introduction

Domestic fuel burning

The negative health and socio-economic impacts of emissions associated with domestic fuel burning are widely recognized. The Global Burden of Diseases Study estimated that household air pollution was responsible for 2.8 million deaths and 85.6 million disability adjusted life years (DALYs) globally in 2015 (Cohen et al 2017). The study concluded that household fuel burning contributed significantly to mortality in low- and middle-income countries. The 1996 and 2011 census data indicate that the proportion of households making use of solid fuels decreased in South Africa during this period, however there remain pockets of households that are still reliant on solid fuel (Pauw et al 2020). Areas that experience the highest impact of domestic fuel burning would be densely populated with a high density of emission sources. The negative impacts of household emissions are exacerbated by the fact that they are released

in the breathing zone of individuals (DEA 2019). Globally, the greatest negative health impact of domestic energy use is from incomplete combustion of fuels in low efficiency stoves and lighting devices (WHO 2014).

Although there has been much progress in the provision of electricity to households in South Africa, many still rely on solid fuel sources such as wood and coal (Israel-Akinbo et al 2018). A study by Israel-Akinbo et al. (2018) examined how poor households transition from traditional energy carriers such as wood to modern energy carriers such as electricity, biofuels and liquefied petroleum gas. In the case of energy usage for heating purposes, households would not necessarily switch to modern fuels as income rises, in line with the energy stacking model, where households use a combination of energy carriers on the upper and lower stages of the energy ladder depending on their needs. An earlier study conducted by the then Department of Energy (presently the Department of Minerals and Energy) came to a similar conclusion as their results indicated that poorer households relied on multiple sources of energy regardless of electrification status, which further points to an energy stacking model rather than the energy ladder theory (DOE 2012).

A South African study analyzing the economic impact of various air quality initiatives found that technology interventions in the domestic sector would be the most efficient way to reduce healthcare costs associated with urban air pollution (Leiman et al 2007). The interventions that have been investigated in South Africa include the electrification of households, the use of alternative fuels, improved household energy efficiency and encouraging the use of top down fire ignition methods, amongst others (Matimolane 2017, Msibi and Kornelius 2017, Mdluli et al 2010, Wagner et al 2005, Scorgie 2012, Makonese 2015).

While various investigations have been done on reducing household emissions by reducing the use of polluting fuels and improvements in combustion efficiency, comparatively fewer studies have been conducted on the reduction of emissions using abatement technology.

The aim of this study was to investigate the potential of particulate matter emission reduction in the domestic sector using an appropriate abatement technology. The study thus investigated the emissions that are likely to arise from domestic fuel burning and the available catalytic and non-catalytic abatement technologies for the reduction of the particulate emissions reported in the literature. The study sought to identify, synthesise and test a suitable abatement technique to decrease particulate matter emissions from domestic fuel burning devices.

Implementation of abatement technologies

Previous studies

Investigations and research into the reduction in emissions from household appliances appears to have been largely focused on improving combustion efficiency and little information seems to be available on secondary or end of pipe type measures to reduce emissions. The use of secondary measures to reduce emissions from small combustion sources in Europe was found to be limited due to the high cost and maintenance requirements of such installations which were limited to the use of electrostatic precipitators (ESPs) (Amann et al 2018).

A review by Lim et al (2015) investigated the use of emission reduction technologies for small scale installations. The study evaluated several technologies that had been tested such as the use of additives, catalytic filters, electrostatic precipitators as well as technologies from the automotive industry. It was concluded that most technologies were still under development and had experienced challenges on implementation.

Cheng et al (2018) investigated the use of high gradient magnetic separation in capturing airborne particles but it achieved low capture efficiency for particles smaller than 50 µm.

In another study, Yamamoto et al (2013) investigated the use of a heated carbon fiber filter to remove soot particles from diesel combustion. Without heating the filter, particles would accumulate on the filter, reducing the porosity significantly. By increasing the filter wall temperature using an electric heater, the study found that most of the particulates could be burned off, resulting in continuous filter regeneration. The study indicated that, while most of the particulates were burned off in the filter, new ultra-fine particles of less than 30 nm were formed, which may have an impact on human health.

Hukkanen et al (2012) investigated the use of a catalytic combustor in reducing emissions from a wood fired boiler. The catalyst utilized consisted of metal wire mesh covered with a platinum and palladium catalyst. For the entire combustion cycle, reductions of 21% for CO, 14% for organic gaseous carbon and 30% of PM_1 was achieved. In this case, the flue gas temperatures were high enough to activate the catalyst, but the study notes that some residential heaters may not have sufficiently high flue gas temperatures to achieve this. A major disadvantage of this technology is the high cost of the precious metals used as catalysts.

Ozil et al (2009) investigated the use of two catalysts, a cordierite honeycomb monolith support and a metallic corrugated structure impregnated with an alumina washout. The study showed that the temperature of the flue gas of a wood fired boiler was too low during start up and shut down for the catalyst to be effective and the use of a heating system was found to dramatically reduce the emissions of CO and VOCs, with a CO reduction of between 80 and 90% being achieved.

Emissions from domestic fuel burning

Flue gas from domestic solid fuel combustion typically has a low particle loading and the particles are small in size (Xu 2014); specifically, the particulates are expected to be 95% $PM_{2.5}$ (Zhang et al 2018).

Previous studies have indicated that the particulate matter emissions from solid fuel burning comprised mainly of organic particles, soot and inorganic fly ash (Zhang et al 2018, Torvela et al 2014, Nystrom 2016, Makonese et al 2019). These particulates are also expected to react with each other and with gaseous emissions in the combustion chamber and flue. The temperature of the flue gas, which is likely to depend on the positioning of the abatement device, will influence the catalyst efficiency as well as both the aging of the particulates and their properties. Closer to the combustion chamber, sticky, tarry carbonaceous particles may form (Makonese et al 2019). Various other chain-like or cluster-like particulates are also likely to be present (Forbes 2012). In addition, organic particles with inorganic inclusions may form. A smaller fraction of inorganic fly ash emissions is expected, which may be larger in size than the organic particles (Zhang et al 2018, Makonese et al 2019).

Catalytic abatement

The collection efficiency for particles smaller than 5 µm, as is expected in flue gas streams, is low and non-catalytic methods are unlikely to achieve a meaningful reduction in particulate matter concentration (Perry et al 1997). Catalytic oxidation could be utilized to oxidize particulate matter precursors such as volatile organic compounds and soot particles to reduce secondary particulate formation. Catalytic abatement allows users to utilise the available fuel, with reduced negative health outcomes. Although catalytic methods have not been effectively utilized in practical domestic applications, studies have shown effective soot reduction during laboratory testing (Gao et al 2020).

Volatile organic matter and soot can be oxidized using supported noble metal catalysts and metal oxide catalysts. Noble metal catalysts generally are more active than metal oxide catalysts, but metal oxide catalysts are more resistant to certain catalyst poisons such as halogens, As and Pb. Metal oxide catalysts importantly have a substantially lower cost and can be sufficiently reactive for some applications (Huang et al 2015) and were therefore selected for this study. The most effective single metal catalysts have been found to be oxides of V, Cr, Mn, Fe, Co, Ni and Cu (Spivey 1987). Manganese was selected for this study due to the promising results achieved with laboratory testing (Gao et al 2020). The results indicated that the soot conversion percentage was temperature dependent and increased at temperatures above 250°C. Furthermore, manganese oxide catalysts have low toxicity, low raw material cost and a diversity of crystalline structures which determine the catalytic activity (Huang et al 2015). In this study, a manganese oxide based catalyst was prepared and characterized. It was then tested for potential local application, utilizing a commercial domestic stove.

Methods

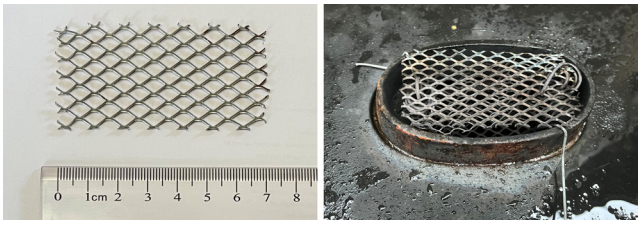
Catalyst Requirements

In order to be considered effective, the designed catalyst should conform to various safety and performance criteria such as acceptable emission reduction, limited clogging, ease of installation, heat resistance, cost effectiveness and the catalyst should be non-toxic. Installation should be possible on non-standardised units, should be self-cleaning or easy to clean with little or no electricity required for operation. The catalyst must further be acceptable to the end user.

Emissions are also expected to vary during the different stages of the combustion process. As the abatement will be in operation during the entire combustion cycle, it should accommodate the emissions from the combined combustion cycle.

Catalyst Synthesis

Three catalyst preparation techniques were tested namely precipitation, wet deposition and dry deposition. The choice of the catalyst support was based on the risk of clogging, ease of installation, minimizing pressure drop, high air flow through the catalyst, and cost, amongst other parameters. A coarse 0.5 mm thick steel mesh was thus used as the catalyst support for direct use in domestic flues as shown in Figure 1.



a) *b)*

Figure 1: *0.5 mm thick steel mesh used as catalyst support (a) for installation at the base of the flue (b)*

The synthesis utilizing precipitation was based on the polyol method, which is well-known for producing metallic nanoparticles (Augustin et al 2015, Li et al 2011, Liu et al 2010, Sukhdev et al 2020). A glycol solvent such as ethylene glycol functions as a solvent for the metal precursors as well as functioning as the reducing agent (Benseeba 2013, Rao et al 2017). The glycol further controls particle formation by controlling nucleation, growth and agglomeration of the particles. The method described by Augustin et al (2015) utilizes mild reaction conditions to produce manganese oxide particles with varying structures and compositions and was therefore used as a basis for the synthesis of the manganese oxide catalyst in this project.

Manganese (II) acetate tetrahydrate (>99%), tetraethylene glycol (99%), iron (III) nitrate nonahydrate (>98%) and ethylene glycol (>99%) were obtained from Sigma-Aldrich (South Africa). All reagents were used as received.

Typical test conditions were as follows: The reaction solution was prepared using 0.5 g of magnesium acetate dissolved in 60 mL of ethylene glycol and 6 mL of tetraethylene glycol (TEG). The catalyst support mesh (commercially available 0.5mm thick steel mesh obtained from Leroy Merlin Hardware Store) was washed using acetone prior to synthesis. The mesh was then

suspended in the reaction mixture using wire hooks secured to the beaker using rubber-based putty adhesive. The mixture was then gradually heated on a magnetic stirrer hot plate (Heidolph, Germany) and stirred using a magnetic stirrer. The temperature of the reagent mixture was measured manually using a thermometer. The reagent mixture was gradually heated to a temperature of 170°C. Samples were removed from the reaction mixture after precipitation occurred at 170°C and were washed using ethanol (>98%, Sigma-Aldrich). The samples were dried under argon (99.9%, Afrox) and were then transferred to a muffle furnace (Lenton, South Africa) at 550°C for five hours to calcine under stagnant air.

The wet deposition catalyst was synthesized by dipping the catalyst support into a concentrated manganese acetate tetrahydrate solution prior to calcining. A mesh sample was prepared using 8 g of manganese acetate tetrahydrate in 30 mL of water. The sample was dipped in the solution, removed and calcined without drying at 550°C for two hours.

The dry deposition catalyst was covered in dry manganese acetate tetrahydrate powder prior to calcining at 550°C for two hours.

Catalyst Testing Methodology

The experimental setup consisted of a commercial one plate combustion device with a flue as shown in Figure 2 (Ndebele Appliances and Coal Stoves, Bronkhorstspruit, South Africa), which is representative of cooking stoves available in South Africa.

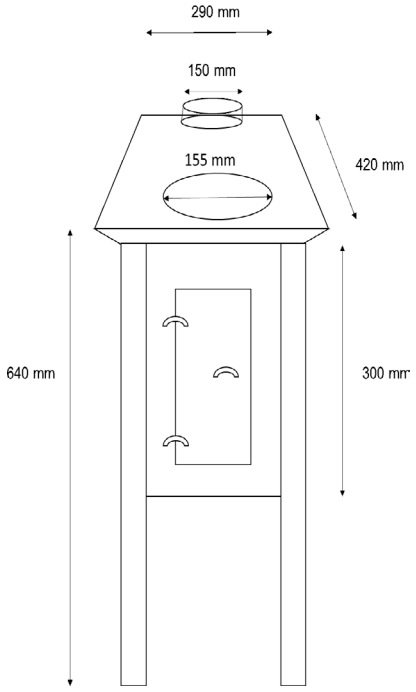


Figure 2: *Diagram of the one-plate stove employed in testing of the catalyst*

Bulk catalysts were produced using precipitation and wet impregnation. The catalyst was positioned at the base of the flue where the highest flue temperatures were expected, as laboratory testing had indicated that the catalyst was only effective temperatures above 250°C (Gao et al, 2020).

For each test run, 100 g of soft wood sticks approximately 150mm in length and 20 x 20 mm diameter were utilized and the combustion process was started using a single wax based firelighter ball. The soft wood was selected as fuel to improve the reproducibility of the test runs as the wood is more homogenous in size and composition when compared to hard wood or coal. The test runs were conducted under similar ambient conditions at the same time each day to improve reproducibility. The fuel used for the runs was sized, weighed and bagged to reduce fuel variability and to ensure consistent fuel moisture content.

Measurement Methodology

The flue gas flow rate was measured using a vane type digital anemometer (Benetech, China). The flue temperature was measured using an infrared thermometer (Shenzen FLUS Technology Company, China). Measurements were taken at 290mm from the flue exit point to allow for the cooling of the flue gas.

Particulates were collected for gravimetric analysis using 37 mm polystyrene cassettes, with 37 mm polytetrafluoroethylene (PTFE) filters with a pore size of 2 µm (Environmental Express, South Carolina, USA). PTFE filters were selected as they are temperature resistant up to 260°C. A Gilian (GilAir Plus) personal sampling pump was used to collect particulates onto the filters at a sampling flow rate of 600 mL/min for 10 min. The positioning of the sampling probe could potentially influence the size distribution of the particulate matter sampled, favouring the capture of finer particles. For this study non-isokinetic sampling was considered adequate as the study was interested in whether the catalyst affected the particulate matter formation in a relative manner, and not to quantify the emissions or derive emission factors. Sample filters were weighed using a Mettler Toledo microgram analytical balance in a temperature (21±1°C) and humidity (50±5%) controlled environment.

Catalyst and PTFE Filter Analysis

The catalyst samples produced were examined using Scanning Electron Microscopy (SEM) using a Zeiss Crossbeam 540 SEM at 20 kV to determine the morphology and particle size of the catalyst and to determine the catalyst coverage on the mesh substrate.

The catalyst samples were analysed using Energy Dispersive X-Ray (EDX) (OXFORD Link-ISIS-300 Zeiss, Germany) analysis to determine the semi-quantitative composition of the catalysts.

The PTFE filter samples were examined using the same SEM instrument at 2 kV. A section of the center part of each filter was removed using a steel blade, mounted on an aluminum stub and sputter coated with Au to improve conductivity.

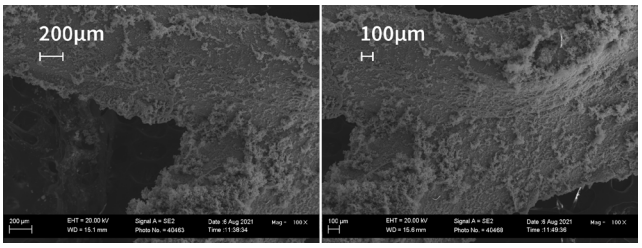
Results and Discussion

Catalyst Synthesis

Synthesis using precipitation

The synthesis was optimized by changing the initial concentrations of manganese acetate tetrahydrate and TEG used to prepare the reaction mixture. The test sample was produced utilizing a 5 x 5 mm piece of mesh suspended in the reagent mixture. Although the precipitate formed on the wire mesh, the precipitate was patchy and did not cover a large percentage of the mesh surface. The Energy Dispersive X-Ray (EDX) analysis confirmed that the precipitates formed during the precipitation runs were manganese oxides with carbon inclusions.

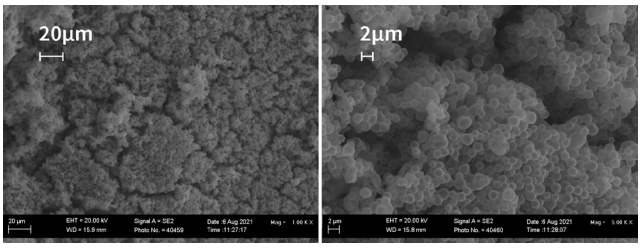
The test run was repeated with an increased manganese acetate tetrahydrate mass of 1 g to improve the catalyst coverage on the mesh. The mesh was calcined directly after synthesis and the calcining time was reduced to 2 hours to ensure that the catalyst was calcined directly after synthesis. The precipitate on the mesh could clearly be seen at 100 X magnification as shown in Figure 3. The precipitate covered the mesh well and formed clusters of precipitate in places. The utilization of higher concentrations of manganese acetate tetrahydrate greatly improved the precipitate coverage on the mesh.



a) *b)*

Figure 3: *SEM micrographs of the catalyst synthesized using increased manganese acetate tetrahydrate concentrations showing precipitate on the mesh at 100 X magnification*

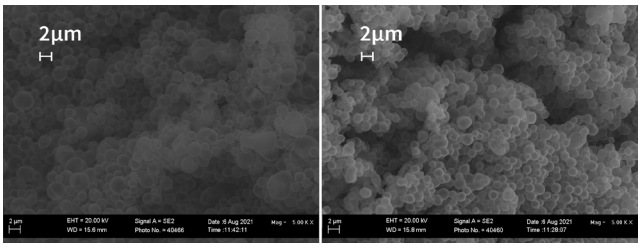
To investigate the impact of the addition of TEG on the coverage and morphology of the catalyst, the previous test was repeated without the addition of TEG. The precipitate formed had good coverage, as shown in Figure 4.



a) *b)*

Figure 4: *SEM micrographs showing the precipitate comprised of spherical particles at 1000 X (a) and 5000 X (b) magnification for the catalyst produced without the addition of TEG*

The addition of TEG did not appear to improve coverage of the mesh with catalyst particles and the precipitates appeared to have similar morphologies as shown in Figure 5.

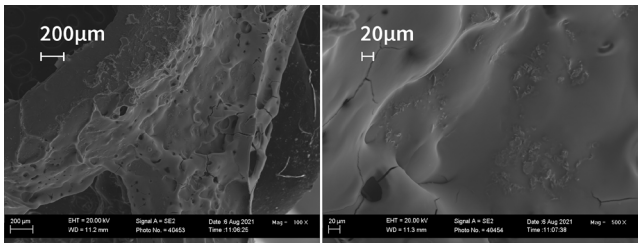


a) *b)*

Figure 5: *SEM micrographs at 5000 X magnification comparing the samples prepared with the addition of TEG (a) and samples prepared without TEG (b)*

Synthesis using wet and dry deposition

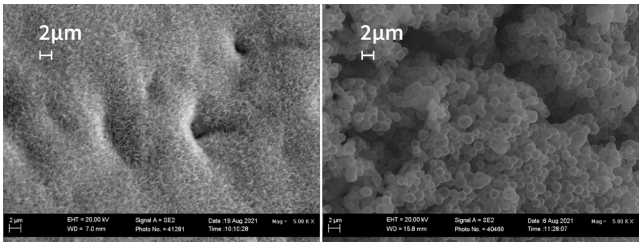
A relatively smooth manganese oxide deposit was formed using wet precipitation and good coverage of the catalyst on the mesh was achieved as shown in Figure 6.



a) *b)*

Figure 6: *SEM micrographs showing the catalyst synthesized using wet impregnation at 100 X (a) and 500 X (b) magnification*

The morphology of the catalyst changed significantly depending on the synthesis method used. At higher magnifications, the surface of the wet impregnation sample had a plate-like structure compared to the spherical, agglomerated particle type structure of the sample prepared using precipitation as shown in Figure 7.



a) *b)*

Figure 7: *SEM micrographs comparing the surface of samples prepared using wet impregnation (a) and precipitation (b) at 5000 X magnification*

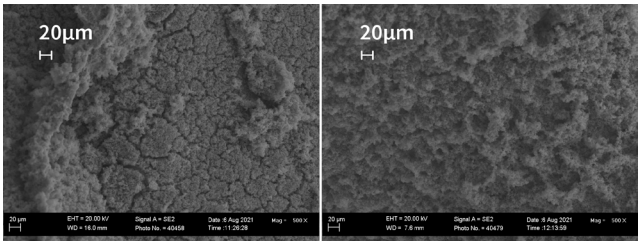
The catalyst prepared using dry impregnation did not achieve good coverage and the mesh substrate was still clearly visible.

Bulk catalyst preparation

Both the optimized precipitation synthesis and wet impregnation techniques achieved good coverage of the mesh substrate. The wet impregnation technique is a simpler process

involving dipping the mesh into a concentrated solution of manganese acetate tetrahydrate prior to calcination, whereas the precipitation technique involves the gradual heating of the reagents to form a precipitate. The morphologies of the samples prepared were significantly different, as shown in the paragraph 'Synthesis using wet and dry deposition'.

The sample was analysed using SEM. The results indicated that that the procedure can be successfully scaled up and that similar coverage and morphology was achieved to that obtained in the smaller test scale tests as shown in Figure 8.



a) *b)*

Figure 8: *SEM micrographs at 500 X magnification comparing the catalyst synthesized from the optimized test run (a) and synthesis of the bulk sample (b)*

Catalyst Testing

Bulk catalyst samples were prepared for testwork using both techniques and were then field tested.

The gravimetric analysis results indicated that relatively higher mass concentrations of particulate matter were recorded for the catalyst runs compared to the baseline runs as shown in Figure 9. It was expected that due to the oxidation of VOCs over the catalyst, secondary particulate matter would be reduced during the catalyst runs.

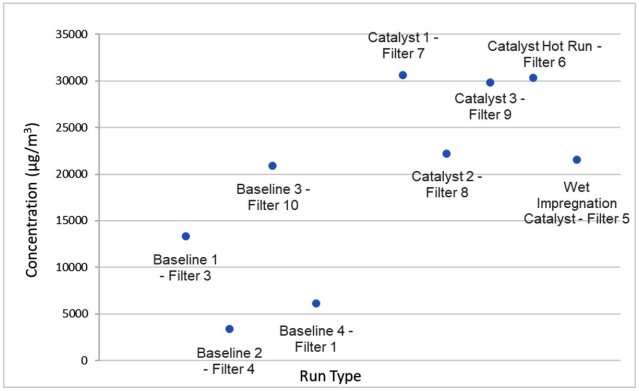
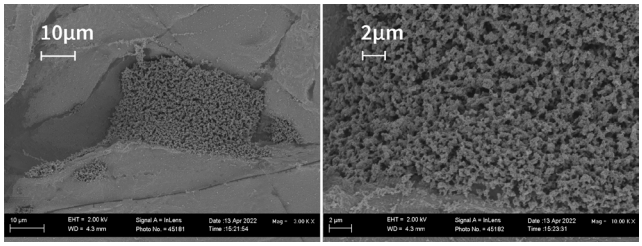


Figure 9: *Comparison of the gravimetric results for the catalyst and baseline runs*

To investigate the cause of the increased particulate matter, Scanning Electron Microscopy (SEM) analysis was conducted on five of the samples. Two baseline samples (Filters 3 and 10), two catalyst samples (Filters 6 and 9) and the field blank sample (Filter 2) were selected for analysis.

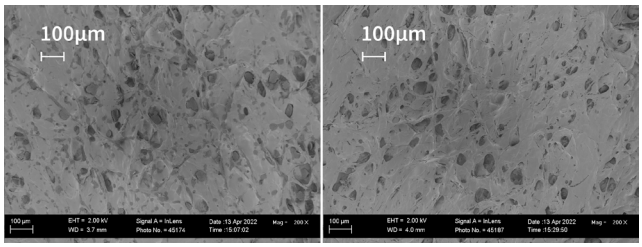
For the catalyst runs, clusters of particles could be seen within the pores of the filters, as shown in Figure 10. Both filters had particle clusters within the pores, but the Catalyst Hot Run which was sampled only during the flaming phase of the run and therefore for a shorter period (Filter 6) had fewer of these clusters.



a) *b)*

Figure 10: *SEM micrograph at 3000 X (a) and 10000 X (b) magnification showing spherical particles smaller than 1 µm in size for the Catalyst Hot Run (Filter 6)*

Particulate clusters can clearly be seen in the pores of the filter for the catalyst run. Very few similar clusters could be seen on the baseline sample filter as shown in Figure 11.



a) *b)*

Figure 11: *SEM micrographs comparing the filters from catalyst Run 3 (Filter 9) (a) to baseline Run 3 (Filter 10) (b) at 200 X magnification*

Energy-dispersive X-ray spectroscopy (EDX) analysis was conducted at 15 kV on the samples to determine whether there was any detectable catalyst lost to the flue gas which would contribute to the PM load on the filters. Carbon and fluoride were detected by the EDX analysis due to the composition of the filter, therefore it was not possible to definitively determine whether the particulates formed were carbon-based due to the background carbon content of the filter. The EDX analysis did not detect any manganese and the increased particulate mass on the filters used for the catalyst runs were therefore not as a result of macroscopic particles of the catalyst detaching from the support.

Conclusions and Recommendations

The testwork showed that an active catalyst can be synthesized in a facile and cost effective manner onto a mesh catalyst support which can be utilized in domestic fuel burning devices. The catalyst is easy to install and can be customised to fit non-standard domestic combustion units. The manganese based catalyst is heat resistant and does not release toxic materials. The catalyst is light in weight and requires no electricity to operate. During the limited test runs conducted, the catalyst

did not clog, the airflow was not restricted and the stove vented as per the baseline runs. It is recommended that the catalyst be tested over a prolonged period to determine whether the catalyst remains self-cleaning or whether it clogs over time. The number of consecutive cycles it can be used over should also be determined (in this study four cycles were successfully tested in this regard).

The total particulate concentrations were measured gravimetrically and the particle mass concentration of the catalyst runs were found to be higher than for the baseline runs. EDX analysis indicated that the increased particulate concentration on the catalyst runs was not likely to be as a result of macroscopic particles of the catalyst being dislodged from the support, as no manganese was detected. The catalyst therefore had good adherence to the catalyst support.

SEM analysis showed that the catalyst run filters had particulate clusters comprised of spherical particles in the pores of the filters. The baseline runs had very few, if any, of these particle clusters. The increased particulate matter could result from the dislodgement of very small metal particles from the catalyst which served as nucleation nodes for particle growth. The metallic nuclei may have had a non-metal coating which may lead to non-detection by EDX. The increase in particulate matter could also be caused by the impingement of particulate matter precursors on the catalyst. Particle growth occurs on the catalyst surface. If the catalyst does not oxidize the impinged pollutant efficiently, particle growth can occur until the particles formed are dislodged from the catalyst by the flue gas, resulting in chain like aggregates. Measuring total particulate matter mass concentrations does not provide information on the morphology, surface chemistry, composition and size characterization of the particulates. It is thus recommended that more detailed analysis be conducted on the particulate matter to provide further insight into secondary particulate formation.

Although the reaction mechanism of oxidation over a manganese oxide based catalyst is generally understood as being through the supply of oxygen from the crystal lattice, a detailed understanding of the elemental reactions are not well understood, particularly for multicomponent systems (Khaskheli et. al. 2022). An improved understanding of the reaction mechanism could lead to the optimisation of the catalyst activity.

Various studies have shown that manganese oxide catalysts are able to oxidize CO, however previous studies investigating the use of manganese oxide catalysts for soot, CO and VOC reduction have mostly been conducted at laboratory scale and not under typical practical (real world) conditions. During laboratory testing, the soot or gas mixture has sufficient time to react and sufficient contact is achieved between the catalyst and the soot or gas (Khaskheli et. al. 2022). In a practical application, there will be much less contact time between the catalyst and the flue gas, which may impact on the efficiency of the catalyst. It is therefore recommended that a range of optimized, potentially active catalysts be tested to improve the oxidation of particulate matter precursors to CO_2. In addition, it is recommended that further testwork include the analysis of gaseous components such as CO, VOCs and CO_2 to determine the extent of oxidation of the particulate matter precursors.

The testwork showed that an active catalyst can be synthesized onto a mesh catalyst support which can be utilized in domestic fuel burning devices. The potential use of an optimised, cost effective catalyst to reduce domestic fuel burning emissions by oxidising particulate matter precursors would enable the continued use of locally available solid fuel with reduced health and environmental impacts.

References

Amann M, Cofala J, Klimont Z, Nagl C, Schieder W (2018) *Measures to address air pollution from small combustion source*s, Report produced under Specific Agreement 11 under Framework Contract ENV.C.3/FRA/2013/ 00131 of DG-Environment of the European Commission.

Augustin M, Fenske D, Bardenhagen I, Westphal A, Knipper M, Plaggenborg T, Kolny-Olesiak J and Parisi J (2015) 'Manganese oxide phases and morphologies: A study on calcination temperature and atmospheric dependence', *Beilstein Journal of Nanotechnology*, vol. 6, pp. 47 - 59.

Benseeba F (2013) *Nanoparticle Technologies*, From Lab to Market, First Edition, Elsevier, ISBN: 9780080919270.

Cheng M.G, Murphy B, Moon J.W, Lutdka G.M and Cable-Dunlap P.R (2018) 'On the use of high-gradient magnetic force field in capturing airborne particles', Journal of Aerosol Science, vol. 120, pp. 22-31.

Cohen A.J, Brauer M, Burnett R, Ross Anderson H, Frostad J, Estep K, Balakrishnan K, Brunekreef B, Dandona L, Dandona R, Feigin V, Freedman G, Hubbell B, Jobling A, Kan H, Knibbs L, Liu Y, Martin R, Morawska L, Arden Pope III C, Shin H, Straif K, Shaddick G, Thomas M, Van Dingenen R, Van Donkelaar A, Vos T, Murray C.J.L, Forouzanfar M.H (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', *The Lancet*, vol. 389, pp. 1907–1918.

Department of Environmental Affairs (2019) *National Environmental Management Air Quality Act 39 of 2004: Strategy to address air pollution in dense low-income settlements*, Government Notice GN666, 14 May 2019.

Department of Energy (DOE 2012) *A survey of energy-related behaviour and perceptions in South Africa*, Human Sciences Research Council (HSRC)

Fletcher TH, Ma J, Rigby J, Bown AL and Webb BW (1997) 'Soot in coal combustion systems', *Progress in Energy Combustion Science*, vol. 23, pp. 283-301.

Forbes PBC (2012) 'Particle emissions from household fires in South Africa', *WIT Transactions on Ecology and the Environment*, vol. 157, pp. 445-456.

Gao Y, Wang Y, Ciu C, Wang B, Liu W, Liu W and Wang L (2020) 'Amorphous manganese oxide as a highly active catalyst for soot oxidation', *Environmental Science and Pollution Research*, vol. 27, pp. 13448-13500.

Huang H, Xu Y, Feng Q and Leung DY (2015) 'Low temperature catalytic oxidation of vol-atile organic compounds: a review', *Catalysis Science and Technology*, vol. 5, pp. 2649-2669.

Hukkanen A, Kaivosoja T, Sippula O, Jokiniemi J and Tissari J (2012) 'Reduction of gaseous and particulate emissions from small-scale wood combustion with a catalytic combustor', *Atmospheric Environment*, vol. 50, pp. 16 - 23.

Israel-Akinbo S, Snowball J and Fraser G (2018) 'The energy transition patterns of low-income households in South Africa: An evaluation of energy programme and policy', *Journal of Energy in Southern Africa*, vol. 29, no. 3, pp. 75 - 85.

Leiman A, Standish B, Boting H, Van Zyl H (2007) 'Reducing the healthcare costs of urban air pollution: The South African experience', *Journal of Environmental Management*, vol. 84, pp. 27–37.

Li Y, Tan H, Yang XY, Goris B, Verbeeck J, Bals S, Colson P, Cloots R, Van Tendeloo G, and Su BL (2011) 'Well Shaped Mn_3O_4 Nano-octahedra with Anomalous Magnetic Behavior and Enhanced Photodecomposition Properties', *Nano, Micro, Small*, vol. 7, no. 4, pp 475–483.

Lim MT, Phan A, Roddy D and Harvey A (2015) 'Technologies for measurement and mitigation of particulate emissions from domestic combustion of biomass: A review', *Renewable and Sustainable Energy Reviews*, no. 49, pp. 574-584.

Liu L, Yang Z, Liang H, Yang H, Yang Y (2010) 'Shape-controlled synthesis of manganese oxide nanoplates by a polyol-based precursor route', *Materials Letters*, vol. 64, pp. 891–893.

Makonese T, Forbes P, Mudau L and Annergarn HJ (2014) 'Aerosol particle morphology of residential coal combustion smoke', *Clean Air Journal*, vol. 24, no. 2, pp. 24-28.

Nystrom R (2016) 'Particle emissions from residential wood and biodiesel combustion', Doctoral Thesis, Umea University, Sweden.

Ozil F, Tschamber V, Haas F and Trouve G (2009) 'Efficiency of catalytic processes for the reduction of CO and VOC emissions from wood combustion in domestic fireplaces', *Fuel Processing Technology*, vol. 90, pp. 1053-1061.

Perry R and Green DW (1997) *Chemical Engineers Handbook*, 7th Edition, Mcgraw Hill Company Inc.

Pauw CJ, Murray HM and Howard MA (2022) 'The use of dirty fuels by low-income households on the South African Highveld', *Clean Air Journal*, vol. 32, no. 1, pp. 81-97.

Rao B.G, Mukerjee D.M and reddy B.M (2017) *Nanostructures for Novel Therapy, Chapter 1: Novel appraoches for prepartion of nanoparticles*, Elsevier, pp 1-36.

Spivey JJ (1987) 'Complete Catalytic Oxidation of Volatile Organics', *Industrial Engineering Chemical Research*, vol. 26, pp. 2165-2180.

Sukhdev A, Challa M, Narayani L, Manjunatha AS, Deepthi PR, Angadi JV, Kumar PM Pasha M (2020) 'Synthesis, phase transformation, and morphology of hausmannite Mn_3O_4 nanoparticles: photocatalytic and antibacterial investigations', *Heliyon*, vol. 6.

Torvela T, Tissari J, Sippula O, Kaivosoja T, Leskinen J, Viren A, Lahde A and Jokiniemi J (2014) 'Effect of wood combustion conditions on the morphology of freshly emitted fine particles', *Atmospheric Environment*, vol. 89, pp. 65-76.

World Health Organization (2014) *WHO guidelines for indoor air quality: household fuel combustion*, WHO Press, World Health Organization, Geneva, Switzerland.

Xu Z (2014) *Fundamentals of Air Cleaning Technology and Its Application in Cleanrooms*, Springer, Berlin, Heidelberg.

Yamamoto K, Fujikake F and Matsui K (2013) 'Non-catalytic after-treatment for diesel particulates using carbon fibre filter and experimental validation', *Proceedings of the Combustion Institute*, no. 34, pp. 2865 - 2875.

Zhang R, Liu C, Zhou G, Sun J, Liu N, Hsu PC, Wang H, Qiu Y, Zhao J, Wu T, Zhao W and Cui Y (2018) 'Morphology and property investigation of primary particulate matter particles from different sources', *Nano Research*, vol. 11, no. 6, pp. 3182-3192.

Research article

Tropospheric ozone (O_3) pollution in Johannesburg, South Africa: Exceedances, diurnal cycles, seasonality, O_x chemistry and O_3 production rate

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Abstract

Ground-level ozone (O_3) is an air pollutant of major health and environmental concern. The Johannesburg-Pretoria megacity in South Africa is the industrial and economical capital of the country with more than 10 million inhabitants experiencing poor air quality. In 2004, the City of Johannesburg (CoJ) began monitoring trace gases to assess ground-level O_3 pollution. Here, we use CoJ's publicly available air quality data, and present the first long-term data analysis of O_3, nitric oxide (NO), nitrogen dioxide (NO_2), NO_x and carbon monoxide (CO) in the City from 2004 to 2011 at three air quality monitoring sites: Buccleuch, Delta Park and Newtown. We quantified CoJ's South African National Ambient Air Quality Standards (NAAQS) exceedances for O_3 and NO_2, and demonstrate the City's substantial O_3 and NO_2 air pollution problem. O_3 mixing ratios peak in the early afternoon as expected due to photochemical production. To estimate O_3 production rates, we summed O_3 and NO_2 diurnal profiles to obtain O_x mixing ratios at each site. This analysis provided insight into missing volatile organic compound (VOC) reactivity as well as primary NO_2 emissions information necessary for developing tropospheric O_3 pollution mitigation strategies. Furthermore, CoJ experiences high O_3 mixing ratios on weekends due to lower NO_x traffic emissions titrating the O_3, thereby providing evidence of a VOC-limited regime for O_3 production. Seasonal peak O_3 occurs in the austral spring, a maximum that we link to increases in water (H_2O) concentrations which in turn increases radical chemistry leading to O_3. In addition, wintertime VOC and aerosol emissions from biomass burning over the winter add important precursors for O_3 formation once radical chemistry is initiated during the first rain events in early spring. In all, this study will help inform air quality modelling and policy work on air pollutants in the City of Johannesburg, South Africa.

Keywords

ozone, nitrogen oxides, Ox, air quality, air pollution, monitoring, Johannesburg, South Africa

Introduction

Ground-level ozone (O_3) is a major component of photochemical smog (Jacobs, 1999). It can negatively impact human health and the environment by causing oxidative stress in the human lungs and/or in plant stomata (Monks et al., 2015; Sillman, 2003). O_3 is a stressor on agricultural crops, and consequently on the world's food supply (Mills et al., 2018; Wilkinson et al., 2012). O_3 has been labelled as the most difficult pollutant to bring into compliance with air quality standards (Jacobs, 1999).

O_3 is termed a secondary pollutant because it is formed in the atmosphere from precursor gases such as nitrogen oxides (NO_x) and volatile organic compounds (VOCs) in the presence of sunlight (Jacobs, 1999; Pusede et al., 2015). However, O_3 has a non-linear dependence on the concentrations of its precursors and thus is notoriously difficult to manage and mitigate. NO and NO_2 interconvert rapidly in the atmosphere and their sum is denoted as NO_x (Figure 1). Daytime photolysis of NO_2 leads to the production of an O atom (Reaction 1) which then goes on to

react with abundant oxygen molecules to form O_3 (Reaction 2) and involves a third body molecule, most likely N_2 (Figure 1). O_3 is also destroyed by NO to regenerate NO_2, thus forming a null cycle (Reaction 3), traditionally termed the photo-stationary state (Jacobs, 1999). We subsequently refer to Reaction 3 as O_3 titration. However, if NO is converted back into NO_2 by peroxy radicals (RO_2) (Reaction 5), the latter originating from oxidation of VOCs often by OH radicals or direct photolysis (Reaction 4), O_3 production ensues (Figure 1). It is the regeneration of NO_2 from Reaction 5 instead of from Reaction 3 that leads to O_3 production $P(O_3)$ and therefore to ground-level O_3 pollution.

Understanding the termination reactions that remove RO_2 and NO_2 radicals from this cycle are important in assessing O_3 production. In an area with elevated NO_2 mixing ratios, NO_2 can irreversibly react with OH to form nitric acid, HNO_3 (Reaction 6), which is soluble and will be washed out by wet deposition (Figure 1). The other termination pathway occurs in low NO_x regions when RO_2 (including HO_2) radicals self-react to from ROOR (including H_2O_2) and O_2 (Reaction 7) (Figure 1). The latter pathway is negligible in urban areas where considerable NO_x mixing ratios are expected. Furthermore, we define the sum of O_3 and NO_2 as O_x, used as proxy for total oxidants in the atmosphere (Geddes et al., 2009). NO_x and VOC emissions can vary considerably depending on location, yet O_3 pollution is typically a regional problem due to the transport of precursors from their emission points, further complicating its source apportionment.

Tropospheric O_3 mixing ratios in southern Africa remain high, often exceeding air quality standards. A lack of spatiotemporal monitoring as well as elevated precursor emissions (Mills et al., 2018) hinders abilities to mitigate O_3 pollution. South Africa is one of the few countries on the African continent to have comprehensive air quality legislation (Schwela, 2012). The National Environmental Management: Air Quality Act (Act 39 of 2004) (AQA) enacted in 2004 identifies near-surface O_3 as a criteria pollutant. The National Ambient Air Quality Standards (NAAQS) for O_3 was set in 2009 at 61 ppb (120 µg/m^3) for an 8-hour running average, with 11 allowable hourly exceedances per year (Sonjica, 2009). In South Africa, air quality priority areas can be declared for specific regions which consistently exceed standards, or are expected to exceed in the near future, and which require specific air quality management actions from a national level to improve air quality (Legislation, 2005). There are currently three air quality priority areas within South Africa, namely the Vaal Triangle Airshed Priority Area (VTAPA) (van Schalkwyk, DEAT, 2006), the Highveld Priority Area (HPA) (van Schalkwyk, DEAT, 2007) and the Waterberg-Bojanala Priority Area (WBPA) (DEA, 2012). A southern portion of the city of Johannesburg falls within the VTAPA, while the HPA borders the city to the east (Figure 2). The WBPA is approximately 75 km to the north of the city.

The City of Johannesburg (CoJ) is a rapidly growing urban area within the Gauteng Province of South Africa and is also the most populous city within the province (Figure 2). It is a noticeably higher altitude city at an elevation of 1750 m a.s.l. It is the most economically active in the country, contributing 17% to the country's GDP (Parks Tau and Fowler, 2016). Amidst this economic activity and development, CoJ experiences degraded air quality due to emissions from sources such as a rapidly increasing commercial and private vehicle fleets, household fuel combustion of coal, wood, liquefied petroleum gas and paraffin, mine tailing dumps, and various other industries (Figure 3) (CSIR and Airshed Professionals, 2019). Additionally, emissions emanating outside the city boundaries can also impact the urban air quality, such as a 420 MW pulverized coal fired power plant (Kelvin Power Station) outside the north-eastern edge of the city, as well as pollutants emitted in the adjacent priority areas. Emissions from adjacent priority areas include those from 13 of the country's 15 coal-fired power stations with an installed nominal capacity of 38.5 GW. In addition, CoJ is impacted by large-scale biomass burning that occurs in southern Africa in the austral winter and early spring, from June to October (Archibald et al., 2010; Giglio et al., 2006). NO_x is emitted through high temperature combustion from sources such as vehicles and coal-fired power stations; as well as natural sources such as lightning strikes (Maseko et al., 2021). VOC emissions have a variety of sources to the atmosphere, which may be natural (biomass burning or biogenic) or anthropogenic, including from transport, industrial and residential sectors. Indeed, an air quality study in South Africa attributed continental O_3 to biomass and residential burning (Laban et al., 2018). From the Irene O_3 sondes launch station 15 km north-east of CoJ, biomass burning and long-range transport are known to have important impacts on the variability and magnitude of column O_3 (Diab et al., 2004; Raghunandan et al., 2007; Thompson et al., 2007; Witte et al., 2017). However, there has been less research on ground-level O_3 mixing ratios in Johannesburg and surrounding regions; though there have been some long-term measurements of O_3 in South Africa (Balashov et al., 2014; Krohm, 1993; Rorich and Galpin, 1998).

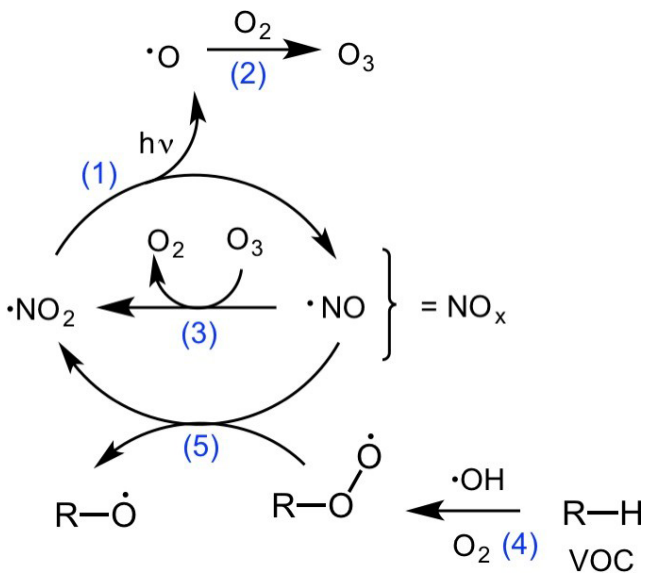

Figure 1: *The mechanism for O_3 production involves NO_x, VOCs and sunlight. The individual reactions are labelled according to the reactions in the text. Importantly, Reaction 3 is the O_3 titration reaction (Jacobs, 1999).*

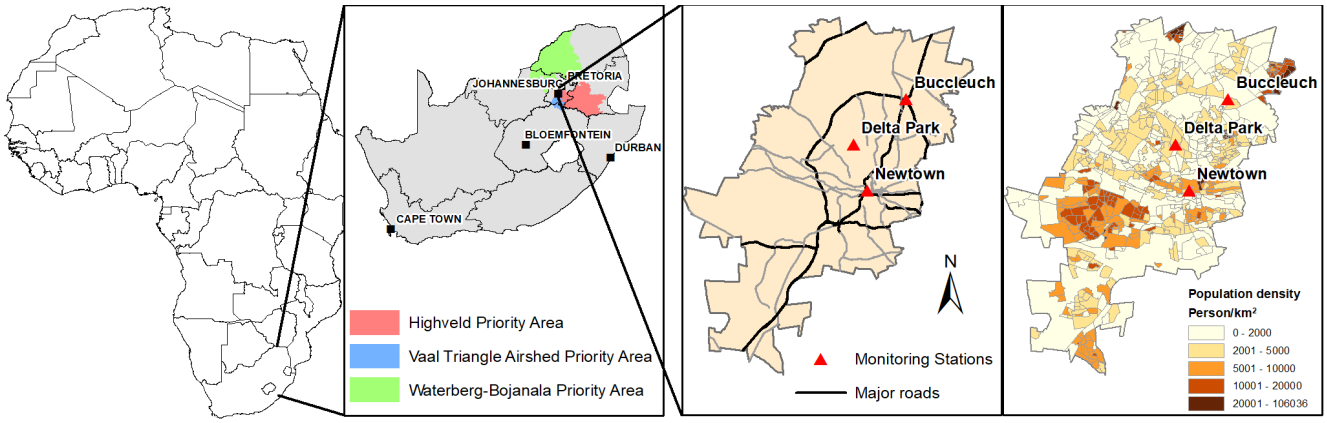

Figure 2: *Maps of the study area within the context of the African continent, of South Africa and the Highveld Area as well as of Johannesburg. The three air quality monitoring stations, Buccleuch, Delta Park and Newtown are depicted as red triangles within Johannesburg. The population density (2015) is represented by the color scale.*

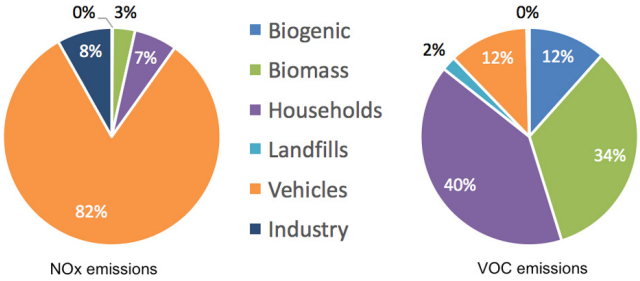

Figure 3: *Johannesburg emissions share by sector and O_3 precursor of total tonnage per year (CSIR and Airshed Professionals, 2019).*

Literature on ground-level O_3 mixing ratios in and around CoJ is primarily based on previous passive sampling measurements of monthly O_3 mixing ratios with some continuous monitoring (Zunckel et al., 2004). Indeed, in the 1980s, O_3 levels were already known to be unusually high, with a recorded peak of 302 ppbv in the spring of 1984 in Johannesburg (Stevens, 1987). Furthermore, there were 84 exceedances of the previous US EPA hourly standard (80 ppbv) for the time period between 1984-winter 1985 (Stevens, 1987). In that study, emissions from traffic were identified as a key contributor to NO_x pollution. However, between 2005–2007, monthly averages of O_3, measured by passive sampling, ranged between 0 and 43 ppbv at a rural site (Josipovic et al., 2010). Lourens et al. in 2011 saw few exceedances of the NAAQS in O_3 and NO_x from passive sampling between 2007 and 2008, but did observe an anti-correlation between high spring O_3 mixing ratios and low spring NO_x, likely indicative of a VOC-limited regime. A study of ground-level O_3 data of 1990-2007 in the Mpumalanga Province (east of Johannesburg) found no statistically significant trend in annual O_3, and only one station exhibited a slight negative trend (-0.92 ppb/yr) in spring O_3 averages (Balashov et al., 2014). In addition, four out of five stations studied in Mpumalanga Province showed sensitivity to ENSO in December-May, with El Niño amplifying O_3 formation (Balashov et al., 2014). Measurement sites in the VTAPA, which include the Diepkloof site in the southern part of CoJ, recorded exceedances of the NAAQS as well as a seasonal increase in springtime O_3 (Govender and Sivakumar, 2019). Most of these studies conclude that further research in the Johannesburg-Pretoria Megacity area is needed.

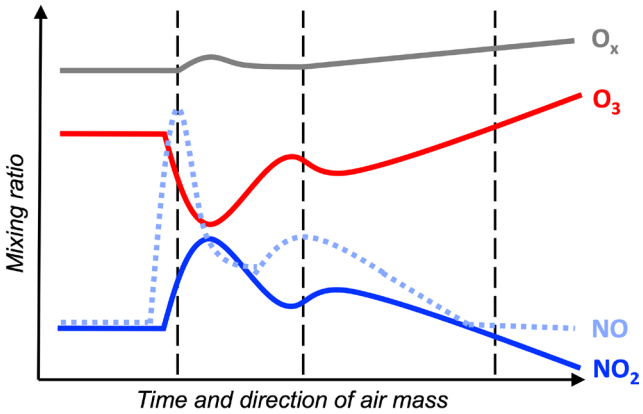

Figure 4: *Mixing ratios of O_x (O_3 + NO_2) as a function of air mass transport of a plume containing NO, NO_2 and O_3. As a polluted air mass travels downwind of its emission source, along the x-axis, O_3 and NO_2 concentrations interconvert, and so O_x calculations allow the visualization of the sum of gas phase oxidants. Note that point source increases in NO_2 can also lead to increases in O_x.*

CoJ installed an air quality monitoring network in 2004 to quantify the extent of its air quality problem in its metropolitan area. Since then, air quality data including ozone (O_3), nitrogen oxides (NO and NO_2) and carbon monoxide (CO) have been measured in real time. In this study, we looked at a dataset from 2004 to 2011 of hourly O_3 mixing ratios and its precursors, namely NO, NO_2, NO_x and CO, measured at three different monitoring sites within CoJ. Importantly, we quantify O_x, the sum of NO_2 and O_3, to estimate the photochemical production of O_3 on a regional scale (see Figure 4). To mitigate tropospheric O_3 pollution, we must develop strategies to lower O_3 production rates, and O_x represents a good proxy for this rate (Geddes et al., 2009; Sokhi et al., 2021). To be clear, high O_3 mixing ratios do not equate to high O_3 *production rates* because of the non-linear formation of this secondary pollutant (see Figure 9 for examples). Since O_3 and NO_2 interconvert rapidly between each other and form a null cycle (R1, R2 and R3 in Figure 1), changes in either one of these pollutants do not necessarily translate into changes in ozone production (see Figure 4). Furthermore, as a polluted air mass travels downwind of its emission source, O_3 and NO_2 concentrations continue to interconvert, and so O_x calculations allow the visualization of the sum of gas phase

oxidants (Figure 4). We recommend considering O_x chemistry for O_3 pollution mitigation strategies. Through this analysis, we aim to better understand O_3 pollution within the city to better characterize the pollution and its drivers, and thus inform air quality management.

Methods

Description of studied area

The City of Johannesburg (CoJ) is surrounded by other populous cities (e.g. City of Tshwane), as well as heavily industrialized areas. Specifically, Ekurhuleni Metropolitan Municipality and Mpumalanga Province lie to the east, and the Vaal Triangle lies to the south (Figure 2). The southern part of Johannesburg falls within the Vaal Triangle Airshed Priority Area, and the Highveld Priority Area borders the city to the east (Figure 2). The recently declared Waterberg-Bojanala Priority Area is to the north and northeast of Johannesburg. Thus, transboundary pollution from these priority areas is a concern for the city.

Monitoring stations

Dataset length and availability

The hourly air quality and meteorological data from four monitoring sites, Buccleuch, Delta Park, Newtown and Alexandra (Figure 2) were acquired through the South African Air Quality Information Systems (SAAQIS; https://saaqis.environment.gov.za) with permission from CoJ. O_3 was measured at the Alexandra site from 2004 to 2008, but the data collection ended in 2008. Since the Alexandra time-series represented only 3.5 years of data, we omitted this site from our analysis. Therefore, only three air quality monitoring sites, Buccleuch, Delta Park, and Newtown, were chosen for this study for their O_3 measurement availability from 2004 to 2011. After 2011, the data availability dropped significantly, and we therefore focus our analysis on the continuous time-series of pollutants between 2004-2011.

Relative humidity (RH) and dewpoint temperature data for the seasonal analysis were not available at Buccleuch, Delta Park or Newtown, and so we used RH, temperature and dewpoint temperature data from the Global Hourly Integrated Surface Database hosted by NOAA. We used weather data from 2004-2011 at the nearby location of OR Tambo International Airport considered part of the broader CoJ metropolitan area.

Site descriptions

The Buccleuch, Delta Park and Newtown sites are influenced by traffic, residential, and urban environments, respectively. The Buccleuch site (26.0453°S; 28.0991°E) is located at the interchange of three major highways (4 – 6 lanes per direction), the N1, N3 and M1, in the northern part of the city (Figure 2). According to the South African National Roads Agency Limited (SANRAL) vehicle counts, over 146 million vehicles passed through the interchange in 2016, with peak volumes seen at either 08:00 or 16:00 (The South African National Roads Agency LTD, 2016). This amounts to an average daily traffic of 403 590 vehicles per day which is comparable for major metropolitan areas around the world. For context, the Springfield Interchange on the Capital Beltway around Washington DC in the United States saw 430 000 vehicles per day in a 2008 estimate (Washington Post, 2007). Additionally, the interchange between Highway 401 and Highway 400 in Toronto, Canada is considered to be Canada's busiest interchange, with a peak flow over 400 000 vehicles per day in 2004 (Nikolic et al., 2005). Thus, the Buccleuch interchange may be considered to have particularly high volumes of vehicles.

The Delta Park site (26.125°S; 28.0086°E) is situated in the middle-class residential suburb of Blairgowrie. The station itself is located in a semi- to sparsely vegetated park with an approximately 1 km^2 area. According to the South African National Land Cover 2018 dataset, the dominant vegetation types in the park are dense forest, woodland and natural grassland (GeoTerraImage, 2018). The nearest busy road is a four lane arterial road (Jan Smuts Avenue) 1.6 km to the north-east.

The Newtown site (26.2052°S; 28.0321°E) is located in downtown Johannesburg in the central business district at an office parking lot, 145 m to the east of the double decker section of the M1 highway.

Instrumentation for trace gas analysis

O_3 measurements were made with Thermo 49C UV photometric O_3 analyzer (Thermo Fisher Scientific, Franklin, MA, USA), and NO, NO_2 and NO_x data were from measurements made with the Thermo 42C chemiluminescent gas analyser (Thermo Fisher Scientific, Franklin, MA, USA)). These instruments both use U.S. EPA Designated Methods and show equivalency to the South African monitoring standards.

We further verified our dataset for the reported BTEX (benzene, toluene, ethylbenzene and xylene) interferences to O_3 measurements (Xu et al., 2018). We conclude that this interference is unlikely in this study for 2 reasons. (1) BTEX compounds are co-emitted with CO but in our study, CO was anti-correlated with O_3 in CoJ's VOC-limited regime for O_3 production. In other words, the higher recorded mixing ratios of O_3 occurred when CO, and thus BTEX, were lower, thereby avoiding a bias in high O_3 mixing ratios due to interference. (2) O_3 measurements at Buccleuch, the traffic site, show a clear photochemical diurnal profile, further supporting the validity of the O_3 measurements despite a high BTEX environment which would likely have peaked during morning and evening traffic (see CO diurnal profile in Figure 8A).

We also note that alkyl nitrates, such as peroxyacetyl nitrate PAN, and HONO were also likely measured by the NO_x analyzer, and thus these measurements are likely an overestimate of NO_x (Wooldridge et al., 2010). Meteorological measurements at each site included wind speed, wind direction, temperature, relative humidity, solar radiation and atmospheric pressure data, although at times these datasets were incomplete. Buccleuch and Newtown also had available intermittent CO data measured

by a Thermo Model 48C CO analyser. Finally, Buccleuch had incomplete BTEX data and it was deemed unusable for data analysis. This study looks at the available data from July 2004 to December 2011, after which time, the data completeness decreases dramatically and thus is unreliable.

Data analysis

Data quality control

These monitoring stations are intended to be continuous; however numerous issues relating to instrumentation errors, technical problems and power-failures have resulted in incomplete datasets throughout the 2004-2011 study period. Although the data can be publicly requested, we unfortunately do not have access to instrument logs, logbooks or flagged data. Consequently, our data quality control focused on obvious invalid data such as multi-day identical values (unrealistic for short lived species), negative values (nonsensical) and infinity values ("inf", nonsensical). Note that we did first look for baseline drifts which could have occurred due to lack of calibrations, but there was no obvious drift to account for (see Figures S1 and S2). The hourly concentrations were rounded to the nearest integer for O_3 and NO_x, and to the nearest tenth of a decimal for CO.

To further substantiate the data quality control analysis, diurnal plots for all pollutants were generated with raw versus quality controlled data, to ensure no significant differences were introduced during our quality control. For example, diurnal profiles of CO at Buccleuch showed no difference in mean, concluding that our quality control methods have not altered the overall dataset (Figure S3).

Data completeness

Data completeness varied between years and months but did not show a significant difference between weekdays and weekends. Furthermore, no clear difference in data completeness was observed between seasons and hours of the day, indicating that despite incompleteness, the quality-controlled data showed good representativeness. In all, data availability over the 8-year period across sites binned hourly ranged between 58.8–77.3% for O_3, 54.3–64.1% for NO_x and 40.5–66.4% for CO, all with standard deviations of less than 5.2% (Table S6).

Statistical analysis

The quality-controlled datasets were checked for normal vs lognormal distributions. All inspected pollutant data showed lognormal distribution, indicating that the data was skewed to lower concentrations, which is typically observed in environmental and air quality observations (Limpert et al., 2001). Indeed, high values were observed, although at lower frequencies. Diurnal cycle plots are used to demonstrate daily profiles of O_3 and its precursors.

Statistics for the seasonal analysis were conducted in R using the Simple linear regression method, with the lm() function from the stats package. We report R^2 values of the regression models for quantifying how much variability of the monthly mixing ratios of the trace gases can be explained by the change of time. We also report the p-values of the trend slopes, with p-value < 0.01 considered to represent a significant trend.

We attempted to look at trends in O_3, NO_2 and O_x over our study period, but the dataset is too short to draw conclusions (Figure S11).

Air quality exceedance calculations

O_3 and NO_2 exceedances were calculated based on the NAAQS for South Africa. An O_3 exceedance is an 8-hour running average concentration above 61 ppbv, with an annual allowance of 11 hourly exceedances. The prescribed O_3 exceedance is not limited to one exceedance per day, like the WHO (World Health Organization, 2021) and US EPA standards (US EPA, 2020), but rather any 8-h running average that exceeds 61 ppbv. Therefore, multiple exceedances per day can occur in South Africa. NO_2 exceedances are defined as those above 106 ppbv and 21 ppbv for hourly and yearly averages, respectively. 88 NO_2 hourly exceedances per year and zero NO_2 yearly exceedances are allowed by the NAAQS.

H_2O concentrations calculations

The partial pressure of H_2O was calculated using the dew point temperature as input into the Tetens equation (mbar):

$$p_{H2O} = 0.61078 \times \exp(17.27 \times \text{dewpointT}/(\text{dewpointT} + 243.04)) \times 10$$

The saturation vapour pressure of H_2O was also calculated using the ambient recorded temperature within the Tetens equation and subsequently converted into H_2O partial pressure using the RH data. Both methods corroborate the partial pressure of H_2O. The concentration of H_2O was then calculated using:

$$N_{H2O} = \frac{P_{H2O} A_V}{RT}$$

where A_v is Avogadro's number, R is the gas constant and T is the ambient temperature in K. H_2O concentrations are reported in molecules/cm^3.

Data analysis software

Finally, plots were generated using either R version 3.4.1 or IGOR version 7.

Results and discussion

O_3 exceedances in Johannesburg

O_3 mixing ratios from three air quality monitoring sites show exceedances according to South Africa NAAQS (Figure 5). The number of yearly O_3 exceedances from 2004-2011 at Buccleuch, Delta Park and Newtown ranged between 0 – 121, 20 – 427 and 6 – 75, respectively, out of a possible 8 760 hours (Figure 5 and Table S1). Note that since NAAQS exceedances are calculated on an hourly basis in South Africa, multiple exceedances per day are possible. Because of variable data completeness, the number of O_3 exceedances likely represent a lower bound (Figure 5). Clearly, CoJ has a substantial and quantifiable ground-level O_3 pollution problem, particularly at Delta Park.

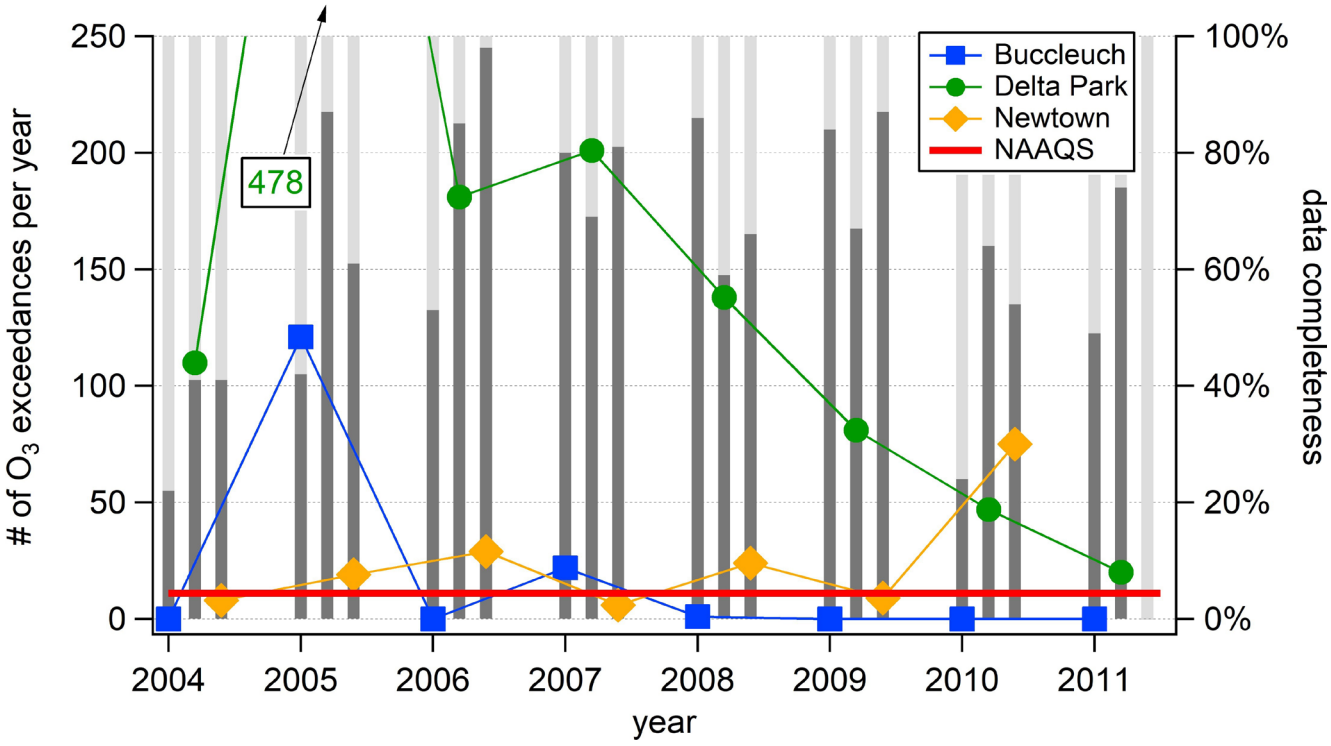

Figure 5: *Number of O_3 exceedances per year at Buccleuch, Delta Park and Newtown over the study period. The O_3 exceedances were calculated using 8-h running averages of O_3 at each monitoring station. The data completeness percentage is plotted as the grey bars and helps interpret the number of exceedances.*

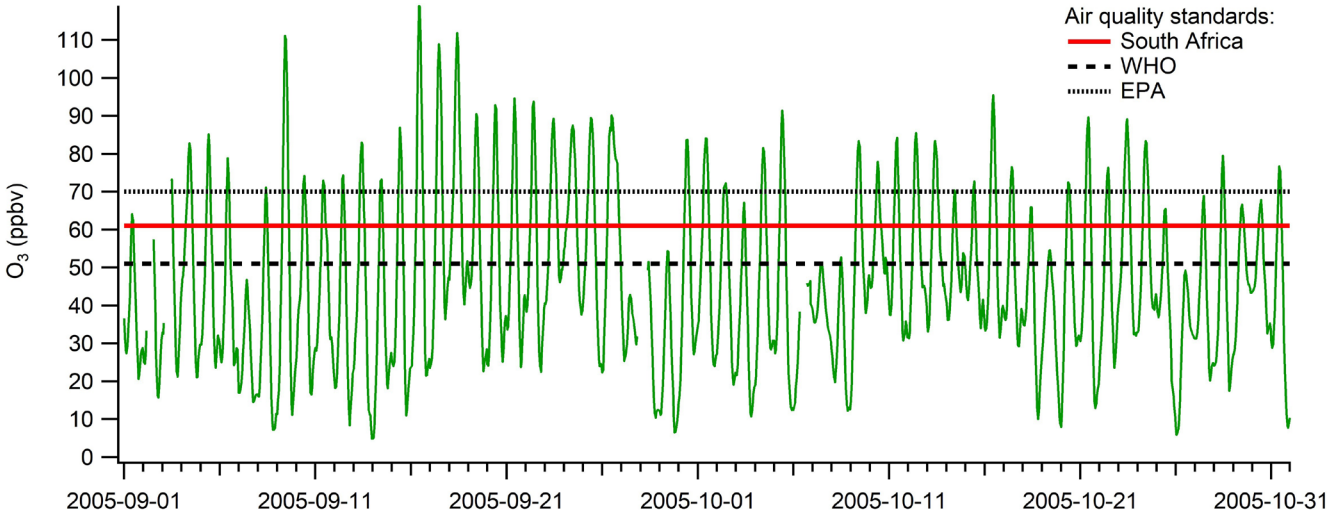

Figure 6: *Time series of 8-h running average of O_3 mixing ratios at Delta Park from September-October 2005 during an excessively high O_3 pollution period. Red line represents the South African NAAQS. The 2015 US EPA standards (70 ppbv as the fourth-highest daily maximum 8-hour concentration, averaged across three consecutive years) and 2021 WHO guidelines (100 µg/m³, or 50 ppbv, 8-hour daily maximum) are presented as references.*

There are clear differences in the number of O_3 exceedances per site. O_3 exceedances occurred at Delta Park every year, with 478 recorded exceedances in 2005. Delta Park is a residential site and was initially intended to be an urban background site, however, it clearly experiences the highest O_3 pollution, concurrently with lower NO_x concentrations. On the other hand, at Buccleuch, a heavily traffic influenced site, few annual O_3 exceedances were observed. This low frequency of exceedance at Buccleuch is attributed to NO titrating O_3 mixing ratios through Reaction 3 in Figure 1. At Delta Park, this titration does not occur to the same extent, leading to higher O_3 mixing ratios. Newtown is an inner-city site near a busy road and experiences O_3 exceedances near the NAAQS; however not as many as at Delta Park. Of note, a concerning sustained high O_3 pollution period occurred at Delta Park from September to October 2005 (Figure 6, Table S1). 8-h running average O_3 exceeded the 61 ppbv set by NAAQS, 51 out of 61 days during the spring of 2005 (Figure 6). Delta Park is in a residential area within Johannesburg, and we expect these high O_3 mixing ratios had non-negligible effects on the health of the residents in the spring of 2005.

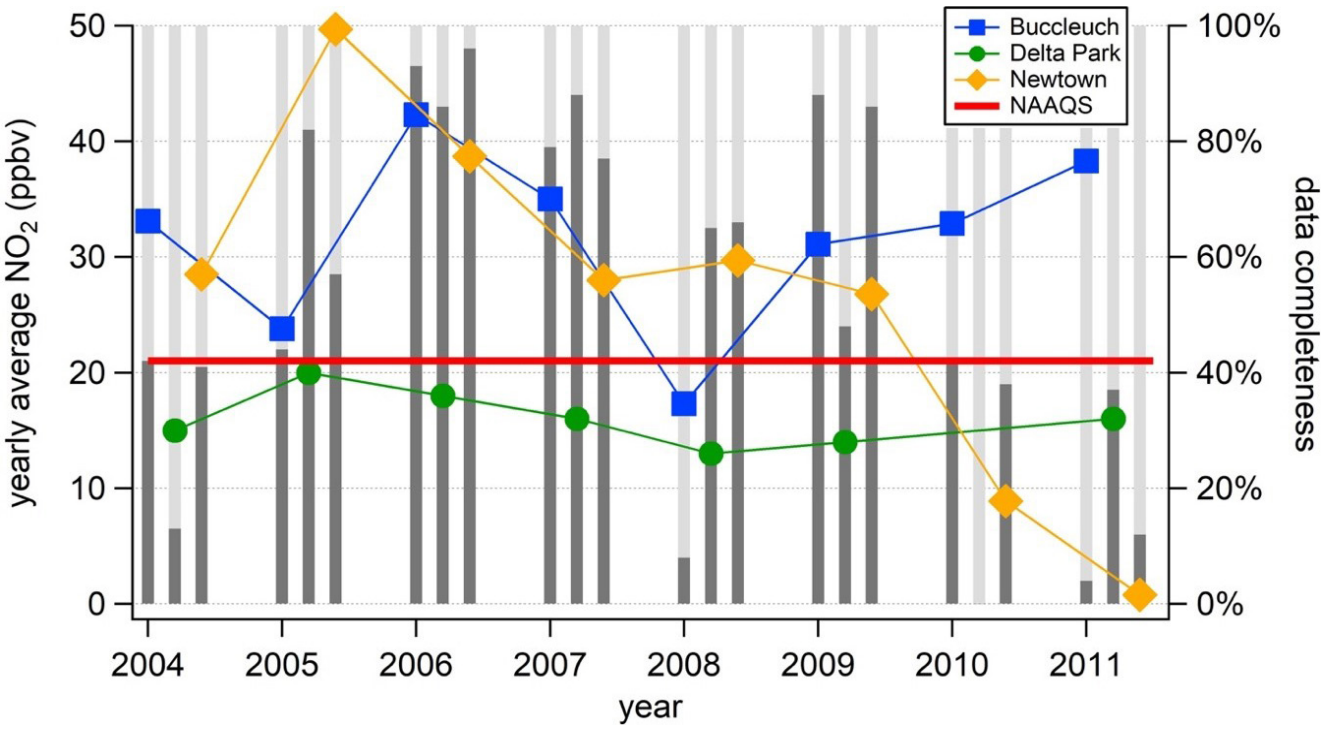

Figure 7: *Annual average NO_2 per year, indicating years in exceedance of the NAAQS at Buccleuch, Delta Park and Newtown over the study period. The yearly NO_2 averages were calculated using hourly NO_2 data from each monitoring station. Data completeness percentage is plotted as the dark grey bars and helps interpret the number of exceedances.*

CoJ is not the only city in South Africa to experience large numbers of O_3 exceedances of the NAAQS. At the Marikana monitoring station located in a mining area northwest of CoJ, O_3 exceedances of NAAQS more than 322 times per year were measured between February 2008 and May 2010 (Venter et al., 2012). These exceedances were attributed to regional air masses bringing O_3 precursors to the monitoring site. In the Mpumalanga Province monitoring sites in the Highveld Priority Area (see Figure 2), annual O_3 exceedances across five sites for 2012-2014 were between 17-761 per year (Lukey, Peter et al., 2011). The Vaal Priority Area also experienced notable exceedances over the period of 2007-2017, but were not numbered (Govender and Sivakumar, 2019). This large number of exceedances in the areas surrounding Johannesburg highlights that O_3 pollution is an issue across the region.

NO_2 exceedances in Johannesburg

Newtown and Buccleuch have high exceedances of the NO_2 annual NAAQS (21 ppbv), with yearly averages of NO_2 mixing ratios up to 49.7 ppbv and 42.3 ppbv, respectively, over the 8-year study period (Figure 7 and Table S2). The available data ranged from 0% to 96% completeness, and thus is important to consider when analysing the annual averages and number of exceedances calculated (Figure 7). Nonetheless, Buccleuch and Newtown appeared to consistently exceed the annual NAAQS for NO_2, whereas Delta Park showed no exceedances in NO_2 regardless of the data fraction available. Since the lifetime of NO_2 in the boundary layer is relatively short (approximately 6 hours in summer time), it is often co-located with its sources, which in this case are majorly traffic emissions (see Figure 3) (Pusede et al., 2015).

Diurnal profiles of O_3, NO_x and CO

Hourly diurnal profiles of O_3, NO_x and CO display strong time-of-day dependence at the sites (Figure 8 and refer to Tables S3, S4, S5 and S6). O_3 peaked in the afternoon as expected, coinciding with peak solar irradiance necessary for its photochemical production. The lowest O_3 mixing ratios were recorded during morning rush hour around 7:00AM, concurrent with high NO traffic emissions titrating O_3 from R3 in Figure 1 (Figure 8). Increasing O_3 mixing ratios overnight were perhaps due to the entrainment of residual O_3 above the shallow nocturnal boundary layer. O_3 mixing ratios were consistently anti-correlated with traffic emitted NO_x, as expected from the formation mechanism of O_3 from the photolysis of NO_2 (R1 and R2 in Figure 1). At all sites, the evening peak in NO_x coincided with slightly higher NO_2, indicative of a shallower boundary layer trapping pollutants and/or of a change in emission sources. Despite these similarities, Buccleuch, Delta Park and Newton had important differences in diurnal profiles of these criteria pollutants.

Buccleuch's diurnal profile showed remarkably high NO_x mixing ratios, peaking around 250 ppbv every morning from traffic emitted NO (Figure 8A). Over 400 000 vehicles pass by the Buccleuch interchange on average per day (See Figure S10 for diurnal variation in hourly traffic counts) (The South African National Roads Agency LTD, 2016). NO clearly remains the major component of NO_x throughout the day as expected for this traffic site (Figure 8A). Furthermore, the CO/NO_x ratio is slightly lower in the evening from 7:00 PM to 11:00 PM, indicating a change in source emissions from traffic to other types of incomplete combustion sources (Figure 8A). In addition, domestic coal

combustion in nearby low-income settlements for cooking and heating could be contributing to the nighttime (from 11:00 PM to 5:00 AM) increased ratio of CO/NO_x at Buccleuch.

Delta Park O_3 mixing ratios are distinctively high with an average of 48 ppbv at 2:00 PM over the 8-year period (Figure 8B). Lower NO_x mixing ratios contribute to higher O_3 levels at Delta Park, because less O_3 titration is occurring (R3). Of note, the morning rush hour peak of NO_x at this station saw similar contributions from NO and NO_2, suggesting localized NO traffic emissions are low, reaching only 25 ppbv in the morning (Figure 8B). Average O_3 mixing ratios peaked an hour later at Delta Park in comparison to the other stations. We hypothesize that in the presence of less NO_x compared to Buccleuch and Newtown, O_3 production can peak later in the day at Delta Park (Figure 8).

Air pollutants at Newtown have intermediate mixing ratios between Buccleuch, a highly traffic influenced site, and Delta Park, a residential site (Figure 8). Newtown is located in downtown CoJ in an office parking lot, and experiences influences from medium to heavy traffic on the M1, 145 m to the west and light traffic from the immediate street. The morning rush hour signal at Newtown is similar in timing and composition to Buccleuch's, albeit in lower absolute concentrations (Figure 8). Newtown's diurnal profile also supports intermediate exceedances in both O_3 and NO_x (Figure 5 and S1). Finally, CO mixing ratios follow an anthropogenic emission profile related to traffic emissions similar to Buccleuch.

Ox (NO_2 + O_3) chemistry to estimate O_3 production

The observed anticorrelation between O_3 and NO_2 in Figure 8 is difficult to interpret due to the dual role of NO_x (1) as an O_3 precursor (R1 and R2 in Figure 1) and (2) as a temporary O_3 reservoir (R3 in Figure 1). In other words, since O_3 and NO_2 interconvert rapidly between each other and form a null cycle (R1, R2 and R3 in Figure 1), increases in O_3 are not necessarily proportional to chemical O_3 production (Figure 4). Nonetheless, mitigation of regional O_3 pollution depends strongly on controlling and understanding O_3 production rates (Geddes et al., 2009). We therefore use O_x as the sum of NO_2 and O_3 to address this rapid interconversion and, assuming negligible direct sources of NO_2, we can use changes in O_x over time as a proxy for chemical O_3 production from VOC oxidation (R4 and R5 in Figure 1 and Figure 4) (Clapp and Jenkin, 2001; Mazzeo et al., 2005; Sokhi et al., 2021).

The multi-year average diurnal profile of O_x is different at each location (Figure 9). During daytime, we can assume that the rise in O_x is solely driven by chemical O_3 production, since NO_2 mixing ratios are relatively constant (Figure 8). We can therefore approximate average O_3 production rates from the change in O_x over time ($\Delta[O_x]/\Delta t$). In other words, the steeper the slope of the O_x between 8:00 AM and 1:00 PM, the faster the rate of O_3 production (Figure 9). At Buccleuch, Delta Park and Newtown, the estimated multi-year average O_3 production rates were 3.3 ppb h^{-1}, 3.5 ppb h^{-1} and 2.7 ppb h^{-1}, respectively from 2004 to 2011 (Figure 9). We hypothesize that at the traffic site, there must be high VOC reactivity to drive high O_3 production rates in the presence of peaks of 250 ppbv of NO_x (Figure 8). We can further infer that the oxidative capacity at Buccleuch is highest.

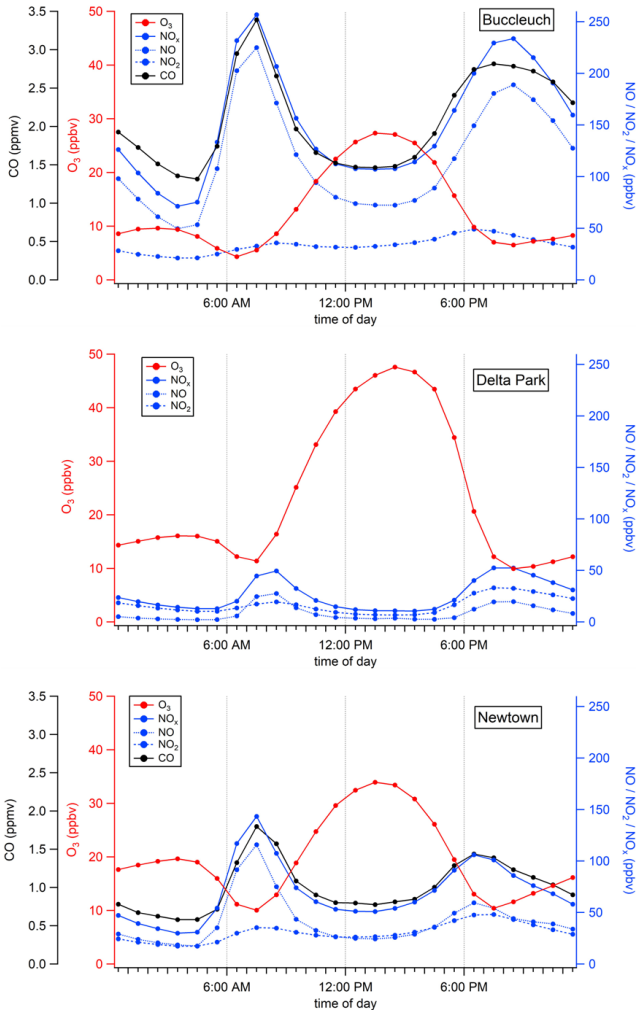

Figure 8: *Multi-year averaged diurnal profiles at Buccleuch (A), Delta Park (B) and Newtown (C) with available data presented in Figure 5 and S1. Of note, axes on the three graphs are set to identical values for comparison. Standard deviations are not included here for clarity, but are presented in Tables S3, S4 and S5 for Buccleuch, Delta Park and Newtown, respectively. The data completeness for this figure is presented in Table S6 and shows consistent data availability throughout all the hours of the day. The data points are presented at the half hour as the average of the full hour.*

Weekend O_3 effect and evidence of a VOC-limited regime

At all three sites, higher O_3 mixing ratios were observed during the weekend, a previously documented observation in cities termed the "weekend effect" (Figure 10) (Murphy et al., 2007, 2006). During the weekend, lower NO mixing ratios are reported at Newtown (blue arrow in Figure 10), concurrent with higher O_3 mixing ratios (red arrow in Figure 10). Govender and Sivakumar, (2019) also noticed that O_3 mixing ratios were higher on Saturdays and Sundays in the Vaal Priority Area. This observation at the same site may be evidence of a VOC-limited regime for O_3 production, but requires accounting for a decrease in the temporary titration effect of near-field NO sources.

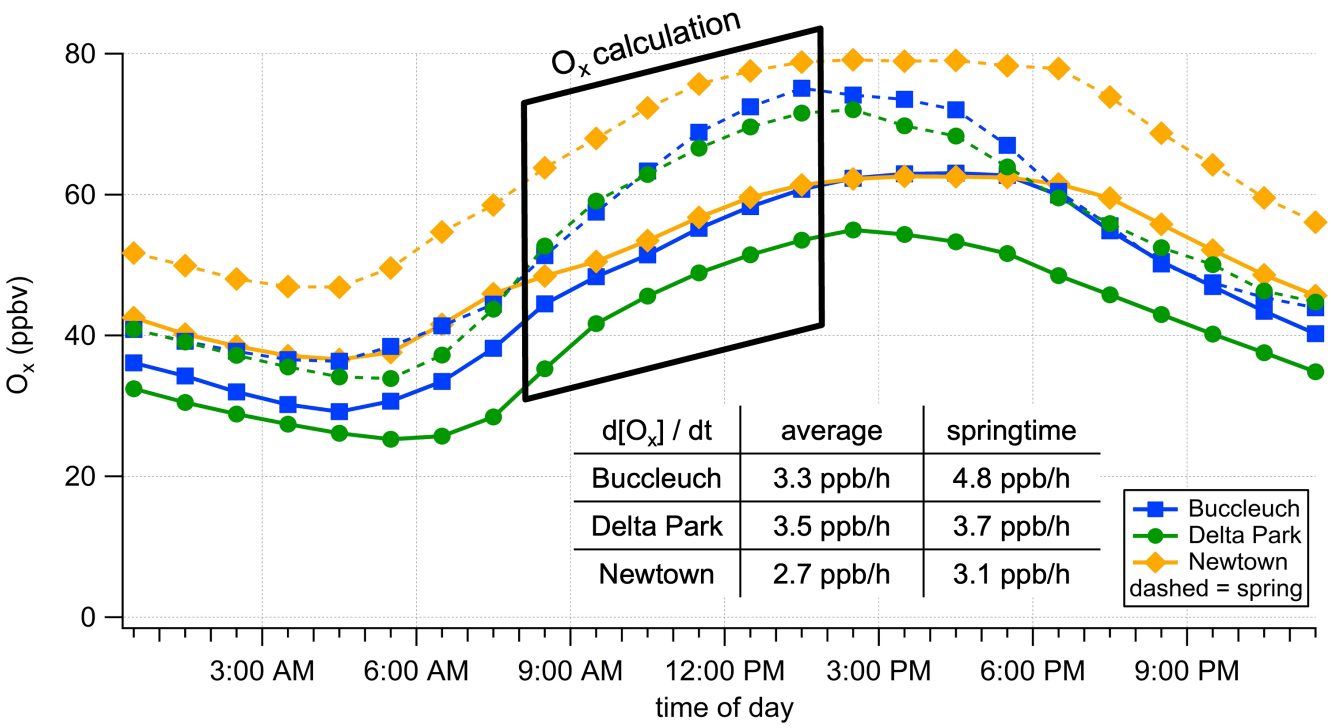

Figure 9: *Multi-year average O_x diurnal profiles as a function of time-of-day for each sites provides the rate of change of O_x, which can be used to approximate O_3 production rates. The peak O_x production in the springtime is earlier and steeper than the multi-year average, which also includes springtime data. $P(O_x)$ is estimated to be equivalent to $P(O_3)$ during the linear increase between 8:00 AM and 1:00 PM.*

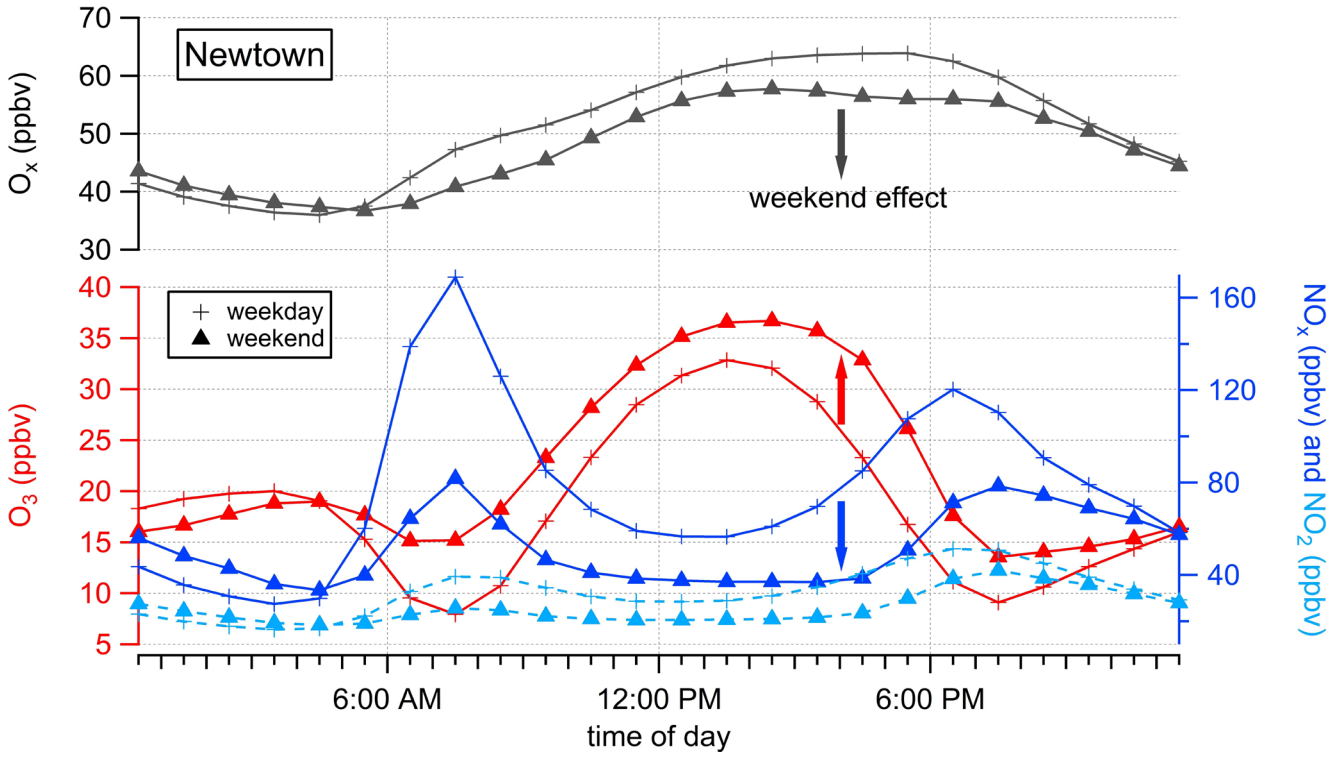

Figure 10: *O_3 (in red), NO_2 (in blue) and O_x (in grey) data at Newtown over the study period plotted as a function of day and separated into weekday (crosses) and weekend (triangles) diurnals. O_x is shown to highlight the absence of a titration mechanism. We conducted this analysis for Newtown only for consistency with our O_3 production analysis in Section 8.*

We looked at O_x (NO_2 + O_3) diurnal profiles during the weekday and weekend at Newtown as a proxy for O_3 production rates. Interestingly, O_x mixing ratios are lower on weekends than on weekdays by 8 ppbv (grey arrow in Figure 10). Assuming that VOC reactivity is not strongly dependent on day of the week (given local emissions are dominated by household activities as seen in Figure 3), this weekend O_x decrease can be explained by two scenarios. First, O_3 production rates in the region are

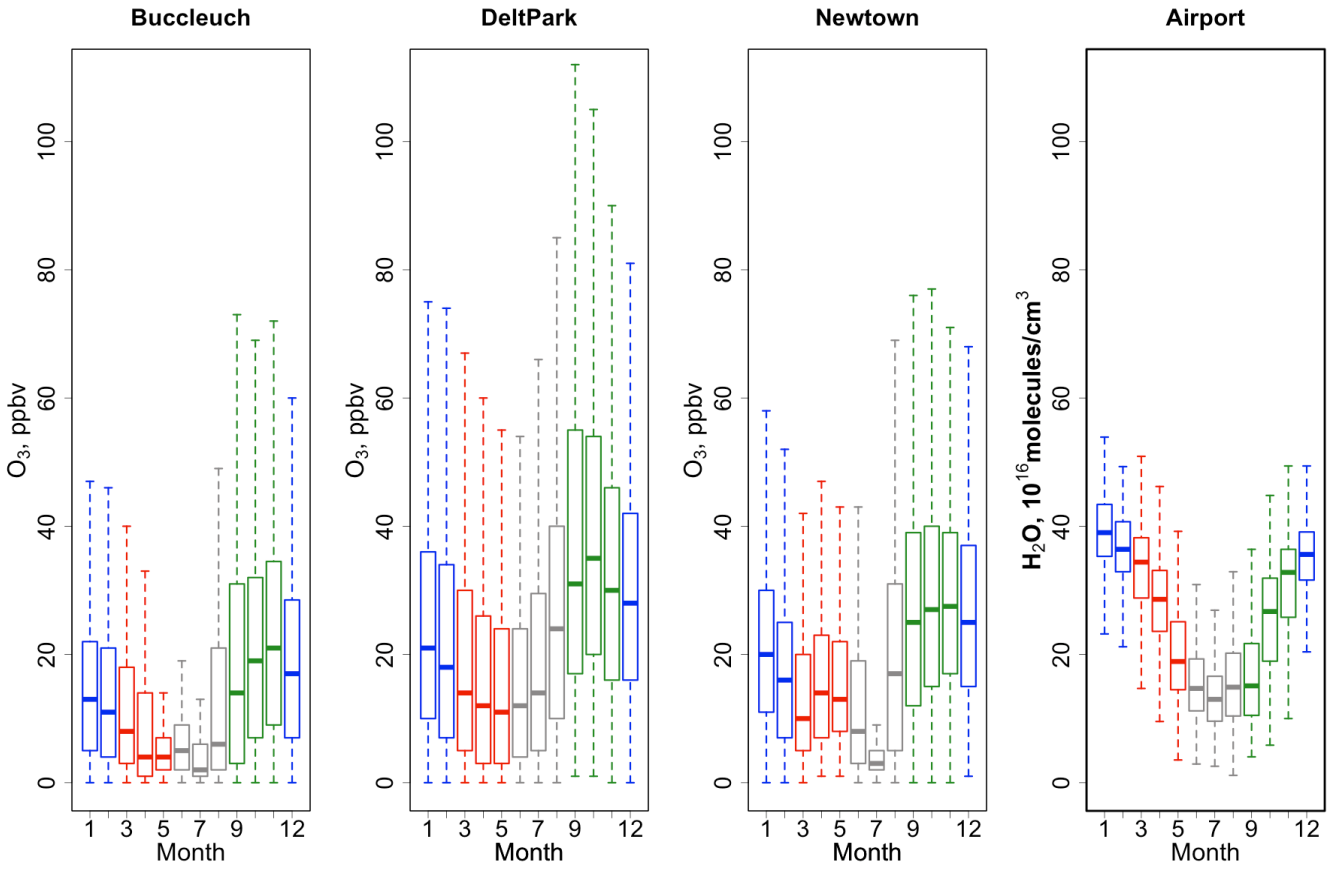

Figure 11: *Box and whisker plots of monthly O_3 mixing ratios at Buccleuch, Delta Park and Newtown as well as monthly H_2O concentrations at OR Tambo airport, colour coded by season where blue is summer (DJF), red is autumn (MAM), grey is winter (JJA) and green is spring (SON). The line is the median and the box limits show the 25th and 75th percentile and the lower and upper whiskers the 1st and 99th percentile. Corresponding monthly and seasonal solar irradiance data, although with incomplete datasets for Delta Park and Newtown, show an austral springtime and summer maxima, indicating that solar irradiance is not the only driving factor in the clear springtime O_3 high in Johannesburg (Figure S5).*

NO_x-limited, leading to O_x decreasing with decreasing in NO_x emission. Or second, there is a significant direct source of NO_2 in the NO_x emissions which confound the use of O_x as a proxy for O_3 production. In the following section, we use a mathematical model to argue that O_3 production rates in this region are highly VOC-limited and we therefore reject the first scenario. The most likely culprit for the weekend O_x behaviour is direct emissions of NO_2 from diesel fuels (Carslaw, 2005). In situations of NO_2 direct emissions, we advise caution in interpreting O_x changes over time as indicative of O_3 production rates. It is noteworthy that the weekend effect analysis which looked at changing O_3 and NO_2 mixing ratios, provided insight into likely NO_2 direct emissions at Newtown.

Seasonality of O_3

O_3 mixing ratios displayed a clear seasonality in CoJ (Figure 11), consistent with findings from the 2000 SAFARI field campaigns (Swap et al., 2002a) as well as from sites in north-eastern South Africa (Govender and Sivakumar, 2019; Laban et al., 2018). Buccleuch, Delta Park and Newtown experienced the highest ground-level O_3 mixing ratios in the springtime, and notably the highest values were recorded at Delta Park (Figure 11). The seasonality of NO at all sites peaked in the wintertime, consistent with a shallow winter boundary layer and increased burning of domestic fuel for heating (Figure S6). NO_2 mixing ratios did not have as clear of a seasonality, although they peaked slightly during the wintertime (Figure S7). However, the magnitude of the NO_2 mixing ratios is different between each site with implications for O_3 production (Figure 8, S7, S8 and S9). Indeed, O_x mixing ratios also peaked in the spring at all sites (Figure S9), indicative of higher O_3 production rates.

We also isolated average springtime O_x mixing ratios as a function of time-of-day to estimate changes in O_3 production in the spring (Figure 9). Clearly, springtime O_x increased at all stations and during each hour of the day compared to annual means (Figure 9, dashed lines). At Buccleuch, Delta Park and Newtown, the average estimated O_3 production rates from the calculated instantaneous O_x production rates during the spring are 4.8 ppb h^{-1}, 3.7 ppb h^{-1} and 3.1 ppb h^{-1}, respectively (Figure 9). These values are all higher than the calculated $P(O_x)$ values using the data from all seasons, indicating that O_3 production rates may be faster in the spring. Springtime O_3 highs are therefore consistent with (1) increased O_3 production and with (2) increased O_x even during the nighttime. The following section explores potential mechanisms leading to increased O_3 production rates.

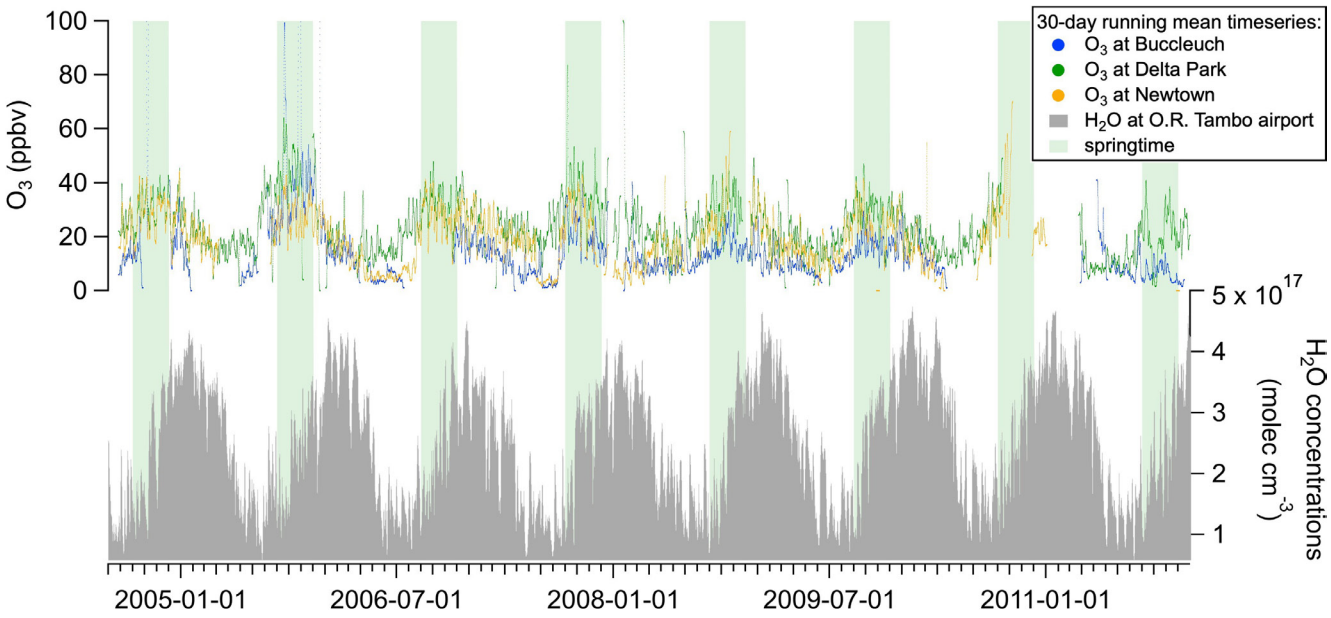

Figure 12: *Timeseries of 3-day running means of O_3 mixing ratios at Buccleuch, Delta Park and Newtown as well as H_2O concentrations at OR Tambo airport (JNB) east of CoJ. The H_2O concentrations were calculated using the dew point temperature reported hourly at OR Tambo airport in the broader Johannesburg area to the east of the three stations. The H_2O concentrations were also corroborated by the RH and T data at the same station. There is a remarkable connection between the onset of increasing H_2O concentrations in the late winter and high O_3 mixing ratios, highlighted by the springtime green shaded regions.*

Mechanism for springtime increased O_3 production rates and the role of RH

High O_3 mixing ratios in the spring over CoJ may be attributed to meteorological effects, such as solar irradiance, relative humidity, temperature, stratospheric intrusions, etc. In addition to enhanced photochemical production, large synoptic effects may also impact springtime O_3 in CoJ by bringing in air masses with different precursors, such as biomass burning events (Laban et al., 2018; Swap et al., 2002b). In comparison to South Africa, North American and European cities have documented peak seasonal O_3 in summertime correlated to peak solar irradiance (Petetin et al., 2018; Pusede et al., 2015). Peak summertime O_3 across the United States has also been shown to correlate with low relative humidity through a mechanism involving dry deposition to leaf stomata (Kavassalis and Murphy, 2017). On the other hand, springtime O_3 has recently been shown to correlate with relative humidity in the interior of South Africa (Laban et al., 2020). Finally, there is evidence of springtime O_3 highs in the western US due to stratospheric intrusions of high altitude cites (Lin et al., 2012). There is the possibility that springtime O_3 caused by stratospheric O_3 intrusions could also be relevant for CoJ, which lies on the Gauteng plateau at 1750 m a.s.l. Mkololo et al., (2020) identified these phenomena using sonde data at a site approximately 30 km northeast of Delta Park, indicating the potential of O_3 intrusions in CoJ. Here, we further explore the mechanisms behind the seasonality of O_3 with data available at the monitoring sites.

CoJ has a dry winter season and a wet summer season, although summertime cloud cover often occurs in the evenings accompanied by thunderstorms. We had access to solar irradiance (Figure S5) and temperature at Buccleuch, Delta Park and Newtown as well as dew point temperature data from a nearby location which we used to calculate H_2O concentrations (Figure 12) to investigate a seasonal meteorological effect on O_3.

Minor role of solar irradiance and temperature

The seasonality of the solar irradiance shows that springtime irradiance is similar to summertime, and slightly higher than in wintertime (Figure S5, where the Buccleuch data is the most complete and reliable). This observation suggests that increased irradiance is not the primary driver of increased O_3 mixing ratios in the spring. Furthermore, O_3 exceedances at Buccleuch were not found to correlate with temperature (Figure S4), and thus temperature is unlikely to be responsible for seasonal O_3 variability (Wilkinson et al., 2012).

Intermediate role of O_3 precursor emissions

Next, we considered O_3 precursor emissions. Veld fires are common during the dry winters in CoJ and emit large concentrations of aerosols (Hersey et al., 2015; Segakweng et al., 2022). In fact, biomass burning has been identified as the instigator for high O_3 events such as those over Pretoria 12 Sept 1985 (Pillay et al., 1994) and in the Vaal Priority Area on 2 June 2013 (Feig et al., 2014). There is also recent evidence of an aerosol-inhibited regime where O_3 mixing ratios are reduced due to heterogeneous chemistry (Ivatt et al., 2022). Particulate matter concentrations in CoJ are highest in the wintertime (Hersey et al., 2015) concurrently with the lowest O_3 mixing ratios and thus this aerosol-inhibited mechanism would be worth exploring for South Africa. However, if biomass burning was responsible for increased O_3 production, we would have expected the seasonality of O_3 to start peaking in mid-winter, which is clearly not observed. So why is the onset of high O_x and high O_3 only in August-September?

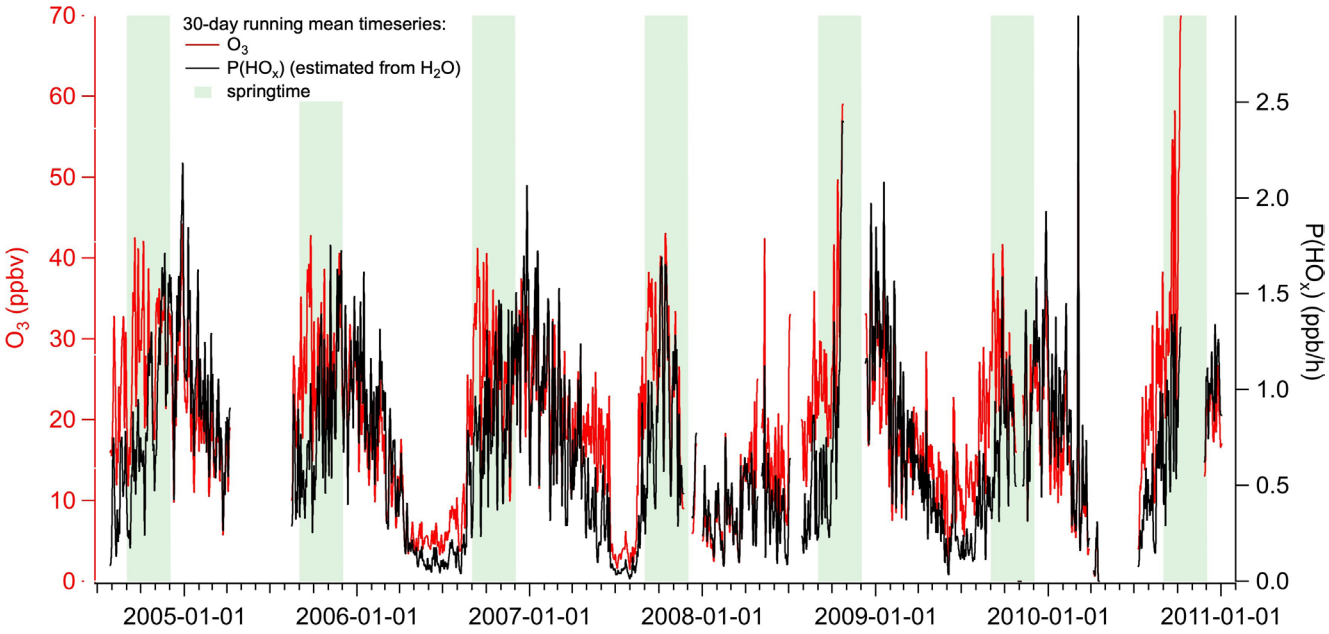

Figure 13: *Timeseries of 30-day running means of O_3 and of estimates of $P(HO_x)$ based on H_2O concentrations. The shaded green areas indicate springtime (SON).*

Major role of H_2O concentrations

We found the most interesting meteorological phenomena in the H_2O data, where the H_2O concentrations drop by a factor of 6 in the springtime (Figure 12). We used RH and dew point temperature data from OR Tambo International Airport situated just east of the CoJ, and available by the Global Hourly Integrated Surface Database hosted by NOAA. CoJ experiences dry winters, with seasonal H_2O concentrations dropping to ~5 x 10^{16} molec/cm^3 in August (Figure 12). Since water vapour is necessary for OH radical production and consequently for the initiation of VOC oxidation leading to O_3 formation, it is likely that wintertime O_3 production is suppressed by slow initiation chemistry. To verify this hypothesis, we estimated the production of HO_x ($P(HO_x)$) radicals using Eq.1 (see SI), and subsequently used this value to estimate OH radical concentrations (Eq. 2 in the SI) (Figure 13). The calculation of OH radicals require CO and NO data and so we did this analysis for the Newtown station only, using the H_2O concentrations from OR Tambo. We find a clear correlation between the onset of $P(HO_x)$, the increase in OH radicals and the increase O_3 in the spring (Figure 13).

Therefore, despite the presence of VOC and NO_x precursors, low O_3 production rates occur in the wintertime due to the low oxidative capacity, including low OH radical concentrations, of the atmosphere to initiate VOC oxidation and to drive R4 in Figure 1. However, when the first spring rain events occur in September, and average H_2O concentrations increase, the rate of R4 increases, kickstarting O_3 production rates. All the necessary precursors are then present and abundant; VOC and NO_x concentrations are high in the wintertime polluted shallow boundary layer, and facilitate radical chemistry propagation leading to O_3. Thus, we suggest that springtime O3 production rates increase in CoJ because of sharp increases in RH in the spring, leading to increases in OH radical products and thus to increases in radical initiation rates (R4 in Figure 1).

Laban et al., 2020 observed an anti-correlation between RH and O_3 by Pearson correlations over the year at sites in the interior of South Africa. In our dataset, RH and O_3 are also anti-correlated (Figure S12). We further emphasize the importance of measuring H_2O concentrations rather than RH in order to estimate $P(HO_x)$.

Conclusion and outlook

Exceedances

We conducted a study on ground-level ozone (O_3) and its precursors in the City of Johannesburg looking specifically at three different sites, Buccleuch, Delta Park and Newtown, in traffic-, residential- and urban-influenced areas, respectively, from 2004-2011. O_3 and NO_2 yearly exceedances showed inverse correlations; stations with large O_3 exceedances showed low NO_2 exceedances and vice versa. Indeed, Delta Park station is located furthest away from NO point emissions in a residential area and experienced a high number of O_3 exceedances. A clear weekend effect was also observed at all locations where NO_x concentrations, associated with transport, were significantly reduced by up to 80 ppbv on weekends, which led to higher O_3 mixing ratios. The hourly continuous data of O_3 mixing ratios presented in this study are informative for potential health impacts of O_3 pollution in South Africa, an impact only qualitatively captured by previous monthly passive sampling data.

VOC-limited regime

The air quality monitoring data at the three distinctively different sites suggest a VOC-limited regime for O_3 production across the entire city. Identifying Johannesburg's O_3 production rate and regime allows us to speculate on the most effective mitigation strategies for air quality. Reducing NO_x concentrations by mandatory catalytic converters on engine exhausts for example

would in fact increase O_3 production kinetics (assuming CO and VOC emissions are unaffected) near the highways. Rather, a more effective short-term strategy in reducing O_3 in the city may be to reduce anthropogenic CO and VOC emissions. Yet, VOC measurements in Johannesburg do not exist in the literature; a clear gap to be addressed for effective air quality management in Johannesburg. Decreasing VOC emissions alongside NO_x emissions would lead to a more effective strategy to reducing O_3 production in Johannesburg. As the urban plume moves further away from CoJ, less NO_2 will be converted to NO and thus less O_3 will be formed (R1 in Figure 1). Additional measurements at rural receptor sites in the City's outflow are needed to improve our understanding on the City's regional impact.

Springtime O_3 and the role of H_2O

We are proposing that increases in H_2O concentrations during the first springtime rains in CoJ initiate spring-time ozone air pollution episodes. Radical chemistry is slow during dry winters despite the presence of O_3 precursors such as VOCs from biomass burning and NO_x from traffic emissions in a low boundary layer. There is a clear correlation between the onset of rainfall in CoJ and the onset of $P(HO_x)$ production (Figure 13), despite the presence of an anti-correlation between RH and O_3 as observed here (Figure S12) and by Laban et al. 2020. We therefore hypothesize that changes in climate in CoJ in a warming future could lead to significant changes in ozone pollution in CoJ and in South Africa.

In all, this study further highlights the importance of long-term monitoring data in general to better understand and hence address air pollution in the hope of improving air quality, and human exposure to airborne pollutants.

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Author contributions

N.B.D and R.G. conceptualized and led the study. N.B.D and R.G. collected the data and N.B.D analysed and interpretated the data with help from R.G., M.N., B.Z and J.G.. N.B.D. wrote the manuscript and all authors contributed to the revisions.

References

Archibald, S., Scholes, R.J., Roy, D.P., Roberts, G., Boschetti, L., Archibald, S., Scholes, R.J., Roy, D.P., Roberts, G., Boschetti, L., 2010. Southern African fire regimes as revealed by remote sensing. Int. J. Wildland Fire 19, 861–878. https://doi.org/10.1071/WF10008

Balashov, N.V., Thompson, A.M., Piketh, S.J., Langerman, K.E., 2014. Surface ozone variability and trends over the South African Highveld from 1990 to 2007. J. Geophys. Res. Atmospheres 119, 4323–4342. https://doi.org/10.1002/2013JD020555

Carslaw, D.C., 2005. Evidence of an increasing NO_2/NO_x emissions ratio from road traffic emissions. Atmos. Environ. 39, 4793–4802. https://doi.org/10.1016/j.atmosenv.2005.06.023

Clapp, L.J., Jenkin, M.E., 2001. Analysis of the relationship between ambient levels of O_3, NO_2 and NO as a function of NO_x in the UK. Atmos. Environ. 35, 6391–6405. https://doi.org/10.1016/S1352-2310(01)00378-8

CSIR, Airshed Professionals, 2019. City of Johannesburg Air Quality Management Plan. https://saaqis.environment.gov.za/Pagesfiles/City%20of%20Johannesburg%20%20AQMP%202019.pdf

Diab, R.D., Thompson, A.M., Mari, K., Ramsay, L., Coetzee, G.J.R., 2004. Tropospheric ozone climatology over Irene, South Africa, from 1990 to 1994 and 1998 to 2002. J. Geophys. Res. Atmospheres 109, D20301. https://doi.org/10.1029/2004JD004793

Feig, G., Ncipha, X., Naidoo, S., Mabaso, D., Ngcukana, N., Tshehla, C., Masuku, N., 2014. Analysis of a period of elevated ozone concentration reported over the Vaal Triangle on 2 June 2013. Clean Air J. 24, 10–16. https://doi.org/10.17159/caj/2014/24/1.7051

Geddes, J.A., Murphy, J.G., Wang, D.K., 2009. Long term changes in nitrogen oxides and volatile organic compounds in Toronto and the challenges facing local ozone control. Atmos. Environ. 43, 3407–3415. https://doi.org/10.1016/j.atmosenv.2009.03.053

GeoTerraImage, 2018. South African National Land-Cover 2018 Report and Accuracy Assessment. https://egis.environment.gov.za/sa_national_land_cover_datasets

Giglio, L., Csiszar, I., Justice, C.O., 2006. Global distribution and seasonality of active fires as observed with the Terra and Aqua Moderate Resolution Imaging Spectroradiometer (MODIS) sensors. J. Geophys. Res. Biogeosciences 111. https://doi.org/10.1029/2005JG000142

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

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

Ivatt, P.D., Evans, M.J., Lewis, A.C., 2022. Suppression of surface ozone by an aerosol-inhibited photochemical ozone regime. Nat. Geosci. 1–5. https://doi.org/10.1038/s41561-022-00972-9

Jacobs, D.J., 1999. Introduction to Atmospheric Chemistry, Introduction to Atmospheric Chemistry. Princeton University Press, Princeton, New Jersey.

Josipovic, M., Annegarn, H., Kneen, M., Pienaar, J., Piketh, S., 2010. Concentrations, distributions and critical level exceedance assessment of SO2, NO_2 and O_3 in South Africa. Environ. Monit. Assess. 171, 181–196. https://doi.org/10.1007/s10661-009-1270-5

Kavassalis, S.C., Murphy, J.G., 2017. Understanding ozone-meteorology correlations: A role for dry deposition. Geophys. Res. Lett. 44, 2922–2931. https://doi.org/10.1002/2016GL071791

Krohm, H.J., 1993. 10 Years of photo-chemical smog monitoring in Johannesburg. Proc. Natl. Assoc. Clean Air NACA 8, 8–18. http://www.cleanairjournal.org.za/download/hkrohm_1993.pdf

Laban, T.L., Van Zyl, P.G., Beukes, J.P., Mikkonen, S., Santana, L., Josipovic, M., Vakkari, V., Thompson, A.M., Kulmala, M., Laakso, L., 2020. Statistical analysis of factors driving surface ozone variability over continental South Africa. J. Integr. Environ. Sci. 17, 1–28. https://doi.org/10.1080/1943815X.2020.1768550

Laban, T.L., Zyl, P.G. van, Beukes, J.P., Vakkari, V., Jaars, K., Borduas-Dedekind, N., Josipovic, M., Thompson, A.M., Kulmala, M., Laakso, L., 2018. Seasonal influences on surface ozone variability in continental South Africa and implications for air quality. Atmospheric Chem. Phys. 18, 15491–15514. https://doi.org/10.5194/acp-18-15491-2018

Legislation, 2005. National Environmental Management: Air Quality Act No. 39 of 2004. https://cer.org.za/virtual-library/legislation/national/air-quality/national-environmental-management-air-quality-act-2004

Limpert, E., Stahel, W.A., Abbt, M., 2001. Log-normal Distributions across the Sciences: Keys and CluesOn the charms of statistics, and how mechanical models resembling gambling machines offer a link to a handy way to characterize log-normal distributions, which can provide deeper insight into variability and probability—normal or log-normal: That is the question. BioScience 51, 341–352. https://academic.oup.com/bioscience/article/51/5/341/243981

Lin, M., Fiore, A.M., Cooper, O.R., Horowitz, L.W., Langford, A.O., Levy II, H., Johnson, B.J., Naik, V., Oltmans, S.J., Senff, C.J., 2012. Springtime high surface ozone events over the western United States: Quantifying the role of stratospheric intrusions. J. Geophys. Res. Atmospheres 117. https://doi.org/10.1029/2012JD018151

Lourens, A.S.M., Beukes, J.P., Van, P.G., Fourie, G.D., Burger, J.W., Pienaar, J.J., Read, C.E., Jordaan, J.H., 2011. Spatial and temporal assessment of gaseous pollutants in the Highveld of South Africa. South Afr. J. Sci. 107, 1–8. https://doi.org/10.4102/sajs.v107i1/2.269

Lukey, Peter, Mdluli, Thulie, Phoshoko, Mathabo, Songxaba, Bathabile, Mafumo, Humbu, Molokwane, Gift, 2011. Highveld Priority Area Air Quality Management Plan. http://www.saaqis.org.za/documents/HIGHVELD%20PRIORITY%20AREA%20AQMP.pdf

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

Mazzeo, N.A., Venegas, L.E., Choren, H., 2005. Analysis of NO, NO_2, O_3 and NO_x concentrations measured at a green area of Buenos Aires City during wintertime. Atmos. Environ. 39, 3055–3068. https://doi.org/10.1016/j.atmosenv.2005.01.029

Mills, G., Pleijel, H., Malley, C.S., Sinha, B., Cooper, O.R., Schultz, M.G., Neufeld, H.S., Simpson, D., Sharps, K., Feng, Z., Gerosa, G., Harmens, H., Kobayashi, K., Saxena, P., Paoletti, E., Sinha, V., Xu, X., 2018. Tropospheric Ozone Assessment Report: Present-day tropospheric ozone distribution and trends relevant to vegetation. Elem. Sci. Anthr. 6, 47. https://doi.org/10.1525/elementa.302

Mkololo, T., Mbatha, N., Sivakumar, V., Bègue, N., Coetzee, G., Labuschagne, C., 2020. Stratosphere–Troposphere Exchange and O_3 Variability in the Lower Stratosphere and Upper Troposphere over the Irene SHADOZ Site, South Africa. Atmosphere 11, 586. https://doi.org/10.3390/atmos11060586

Molewa, B.E.E., 2012. Declaration of the Waterberg National Priority Area. https://www.dffe.gov.za/sites/default/files/gazetted_notices/nemaqa_waterberg_declaration_g35435gen495_0.pdf

Monks, P.S., Archibald, A.T., Colette, A., Cooper, O., Coyle, M., Derwent, R., Fowler, D., Granier, C., Law, K.S., Mills, G.E., Stevenson, D.S., Tarasova, O., Thouret, V., von Schneidemesser, E., Sommariva, R., Wild, O., Williams, M.L., 2015. Tropospheric ozone and its precursors from the urban to the global scale from air quality to short-lived climate forcer. Atmos Chem Phys 15, 8889–8973. https://doi.org/10.5194/acp-15-8889-2015

Murphy, J.G., Day, D.A., Cleary, P.A., Wooldridge, P.J., Millet, D.B., Goldstein, A.H., Cohen, R.C., 2007. The weekend effect within and downwind of Sacramento – Part 1: Observations of ozone, nitrogen oxides, and VOC reactivity. Atmos.Chem.Phys. 7, 5327–5339. https://doi.org/10.5194/acp-7-5327-2007

Murphy, J.G., Day, D.A., Cleary, P.A., Wooldridge, P.J., Millet, D.B., Goldstein, A.H., Cohen, R.C., 2006. The weekend effect within and downwind of Sacramento: Part 2. Observational evidence

for chemical and dynamical contributions. Atmos Chem Phys Discuss 2006, 11971–12019. https://doi.org/10.5194/acpd-6-11971-2006

Nikolic, G., Chan, M., Pringle, R., 2005. Planning and Implementing a Full Closure of the Highway 401 Express Lanes with the Help of Micro-simulation. Presented at the 2005 Annual Conference of the Transportation Association of CanadaTransportation Association of Canada (TAC). https://trid.trb.org/view/798210

Parks Tau, M., Fowler, T., 2016. 2012-2016 Integrated Development Plan "Turning Challenges into Opportunities." City of Johannesburg. https://www.joburg.org.za/documents_/Documents/Intergrated%20Development%20Plan/IDP%20REVIEW.pdf

Petetin, H., Sauvage, B., Smit, H.G.J., Gheusi, F., Lohou, F., Blot, R., Clark, H., Athier, G., Boulanger, D., Cousin, J.-M., Nedelec, P., Neis, P., Rohs, S., Thouret, V., 2018. A climatological view of the vertical stratification of RH, O_3 and CO within the PBL and at the interface with free troposphere as seen by IAGOS aircraft and ozonesondes at northern mid-latitudes over 1994–2016. Atmospheric Chem. Phys. 18, 9561–9581. https://doi.org/10.5194/acp-18-9561-2018

Pillay, Y., Diab, R.D., Sokolic, F., 1994. Ozone maxima off the east coast of South Africa: The role of biomass burning. Clean Air J. 9, 25–28. https://doi.org/10.17159/caj/1994/9/1.7115

Pusede, S.E., Steiner, A.L., Cohen, R.C., 2015. Temperature and Recent Trends in the Chemistry of Continental Surface Ozone. Chem. Rev. 115, 3898–3918. https://doi.org/10.1021/cr5006815

Raghunandan, A., Mahumane, G., Diab, R., 2007. Elevated ozone events over Johannesburg based on analysis of tropospheric ozone partial columns: research letters. South Afr. J. Sci. 103, 248–252. http://reference.sabinet.co.za/webx/access/electronic_journals/sajsci/sajsci_v103_n5_a18.pdf

Rorich, R.P., Galpin, J.S., 1998. Air quality in the Mpumalanga Highveld region, South Africa. South Afr. J. Sci. 94, 109–114. https://journals.co.za/content/sajsci/94/3/AJA00382353_133

Schwela, D., 2012. Review of urban air quality in Sub-Saharan Africa region - air quality profile of SSA countries (No. 67794). The World Bank. http://documents.worldbank.org/curated/en/936031468000276054/Review-of-urban-air-quality-in-Sub-Saharan-Africa-region-air-quality-profile-of-SSA-countries

Segakweng, C.K., van Zyl, P.G., Liousse, C., Beukes, J.P., Swartz, J.-S., Gardrat, E., Dias-Alves, M., Language, B., Burger, R.P., Piketh, S.J., 2022. Measurement report: Size-resolved chemical characterisation of aerosols in low-income urban settlements in South Africa. Atmospheric Chem. Phys. Discuss. 1–58. https://doi.org/10.5194/acp-2021-1026

Sillman, S., 2003. Tropospheric ozone and photochemical smog, in: Holland, H.D., Turekian, K.K. (Eds.), Treatise on Geochemistry. Elsevier-Pergammon, Oxford, pp. 407–432.

Sokhi, R.S., Singh, V., Querol, X., Finardi, S., Targino, A.C., Andrade, M. de F., Pavlovic, R., Garland, R.M., Massagué, J., Kong, S., Baklanov, A., Ren, L., Tarasova, O., Carmichael, G., Peuch, V.-H., Anand, V., Arbilla, G., Badali, K., Beig, G., Belalcazar, L.C., Bolignano, A., Brimblecombe, P., Camacho, P., Casallas, A., Charland, J.-P., Choi, J., Chourdakis, E., Coll, I., Collins, M., Cyrys, J., da Silva, C.M., Di Giosa, A.D., Di Leo, A., Ferro, C., Gavidia-Calderon, M., Gayen, A., Ginzburg, A., Godefroy, F., Gonzalez, Y.A., Guevara-Luna, M., Haque, Sk.M., Havenga, H., Herod, D., Hõrrak, U., Hussein, T., Ibarra, S., Jaimes, M., Kaasik, M., Khaiwal, R., Kim, J., Kousa, A., Kukkonen, J., Kulmala, M., Kuula, J., La Violette, N., Lanzani, G., Liu, X., MacDougall, S., Manseau, P.M., Marchegiani, G., McDonald, B., Mishra, S.V., Molina, L.T., Mooibroek, D., Mor, S., Moussiopoulos, N., Murena, F., Niemi, J.V., Noe, S., Nogueira, T., Norman, M., Pérez-Camaño, J.L., Petäjä, T., Piketh, S., Rathod, A., Reid, K., Retama, A., Rivera, O., Rojas, N.Y., Rojas-Quincho, J.P., San José, R., Sánchez, O., Seguel, R.J., Sillanpää, S., Su, Y., Tapper, N., Terrazas, A., Timonen, H., Toscano, D., Tsegas, G., Velders, G.J.M., Vlachokostas, C., von Schneidemesser, E., Vpm, R., Yadav, R., Zalakeviciute, R., Zavala, M., 2021. A global observational analysis to understand changes in air quality during exceptionally low anthropogenic emission conditions. Environ. Int. 157, 106818. https://doi.org/10.1016/j.envint.2021.106818

Sonjica, B.P., 2009. National Environmental Management: Air Quality Act, 2004 (ACT NO. 39 OF 2004) National Ambient Air Quality Standards 4. https://www.iea.org/policies/13185-national-environmental-management-air-quality-act-39-of-2004

Stevens, C.S., 1987. Ozone formation in the greater Johannesburg region. Atmospheric Environ. 1967 21, 523–530. https://doi.org/10.1016/0004-6981(87)90035-7

Swap, R.J., Annegarn, H.J., Otter, L., 2002a. Southern African Regional Science Initiative (SAFARI 2000): summary of science plan. South Afr. J. Sci. 98, 119–124. http://journals.co.za/content/sajsci/98/3-4/EJC97440

Swap, R.J., Annegarn, H.J., Suttles, J.T., Haywood, J., Helmlinger, M.C., Hely, C., Hobbs, P.V., Holben, B.N., Ji, J., King, M.D., Landmann, T., Maenhaut, W., Otter, L., Pak, B., Piketh, S.J., Platnick, S., Privette, J., Roy, D., Thompson, A.M., Ward, D., Yokelson, R., 2002b. The Southern African Regional Science Initiative (SAFARI 2000): overview of the dry season field campaign. South Afr. J. Sci. 98, 125–130. http://journals.co.za/content/sajsci/98/3-4/EJC97439

The South African National Roads Agency LTD, 2016. https://www.arrivealive.mobi/The-South-African-National-Roads-Agency-LTD.

Thompson, A.M., Witte, J.C., Smit, H.G.J., Oltmans, S.J., Johnson, B.J., Kirchhoff, V.W.J.H., Schmidlin, F.J., 2007. Southern Hemisphere Additional Ozonesondes (SHADOZ) 1998–2004 tropical ozone climatology: 3. Instrumentation, station-to-station variability, and evaluation with simulated flight profiles. J. Geophys. Res. Atmospheres 112, D03304. https://doi.org/10.1029/2005JD007042

US EPA, O., 2020. Ozone National Ambient Air Quality Standards (NAAQS) [WWW Document]. URL https://www.epa.gov/ground-level-ozone-pollution/ozone-national-ambient-air-quality-standards-naaqs (accessed 8.9.22).

van Schalkwyk, M., 2007. National Environmental Management: Air Quality Act, 2004 (Act No. 39 of 2004), Declaration of the Highveld Priority Area in terms of Section 18(1) of the National Environmental Management: Air Quality Act, 2004 (Act No. 39 of 2004) (No. No. 1123). https://www.gov.za/sites/default/files/gcis_document/201409/30518.pdf

van Schalkwyk, M., 2006. Declaration of the Vaal Triangle Air-shed Priority Area in terms of Section 18(1) of the National Environmental Management: Air Quality Act, 2004 (Act No. 39 of 2004) (No. No. 365). https://www.gov.za/sites/default/files/gcis_document/201409/28732b.pdf

Venter, A.D., Vakkari, V., Beukes, J.P., Zyl, P.G.V., Laakso, H., Mabaso, D., Tiitta, P., Josipovic, M., Kulmala, M., Pienaar, J.J., Laakso, L., 2012. An air quality assessment in the industrialised western Bushveld Igneous Complex, South Africa. South Afr. J. Sci. 108, 10 Pages. https://doi.org/10.4102/sajs.v108i9/10.1059

Washington Post, 2007. At a Glance 1. http://www.washingtonpost.com/wp-dyn/content/article/2007/07/17/AR2007071701826.html

Wilkinson, S., Mills, G., Illidge, R., Davies, W.J., 2012. How is ozone pollution reducing our food supply? J. Exp. Bot. 63, 527–536. https://doi.org/10.1093/jxb/err317

Witte, J.C., Thompson, A.M., Smit, H.G.J., Fujiwara, M., Posny, F., Coetzee, G.J.R., Northam, E.T., Johnson, B.J., Sterling, C.W., Mohamad, M., Ogino, S.-Y., Jordan, A., da Silva, F.R., 2017. First reprocessing of Southern Hemisphere ADditional OZonesondes (SHADOZ) profile records (1998–2015): 1. Methodology and evaluation. J. Geophys. Res. Atmospheres 122, 2016JD026403. https://doi.org/10.1002/2016JD026403

Wooldridge, P.J., Perring, A.E., Bertram, T.H., Flocke, F.M., Roberts, J.M., Singh, H.B., Huey, L.G., Thornton, J.A., Wolfe, G.M., Murphy, J.G., Fry, J.L., Rollins, A.W., LaFranchi, B.W., Cohen, R.C., 2010. Total Peroxy Nitrates (Sigma;PNs) in the atmosphere: the Thermal Dissociation-Laser Induced Fluorescence (TD-LIF) technique and comparisons to speciated PAN measurements. Atmospheric Meas. Tech. 3, 593–607. https://doi.org/10.5194/amt-3-593-2010

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, Geneva. https://apps.who.int/iris/handle/10665/345329

Xu, Zheng, Nie, W., Chi, X., Huang, X., Zheng, L., Xu, Zhengning, Wang, J., Xie, Y., Qi, X., Wang, X., Xue, L., Ding, A., 2018. Ozone from fireworks: Chemical processes or measurement interference? Sci. Total Environ. 633, 1007–1011. https://doi.org/10.1016/j.scitotenv.2018.03.203

Zunckel, M., Venjonoka, K., Pienaar, J.J., Brunke, E.-G., Pretorius, O., Koosialee, A., Raghunandan, A., van Tienhoven, A.M., 2004. Surface ozone over southern Africa: synthesis of monitoring results during the Cross border Air Pollution Impact Assessment project. Atmos. Environ. 38, 6139–6147. https://doi.org/10.1016/j.atmosenv.2004.07.029

Supplementary material

Supplementary material can be accessed at https://cleanairjournal.org.za/article/view/15367

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BACKGROUND

Climate change and air quality in environmental management faces significant current and future challenges, especially in South Africa (being one of the developing countries in the world). It is recognized that dealing with climate change and air pollution challenges will require a holistic approach from stakeholders from various disciplines.
The learning outcomes of this programme will allow graduates to successfully quip themselves with environmental management and air quality management skills especially those in the sector responsible for management, governance and strategic decision making.

MISSION STATEMENT

The programme aims to build capacity and skills within the air quality and climate change sectors. This is achieved through multi-disciplinary and international training collaboration.
Particular emphasis is placed on the development of analytical skills and critical thinking through high quality research outputs.
This will enable students to compete with confidence as environmental practitioners in the national and international labour market.

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Review article

Establishing a baseline of published air pollution and health research studies in the Waterberg-Bojanala Priority Area

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Abstract

Though originally the Waterberg-Bojanala Priority Area (WBPA) was strategically declared as an air quality priority area due to potential future air pollution risks, it is now a confirmed air pollution hotspot. More research is needed to assess the health impacts of air pollution in the WBPA. The aim of this study was to conduct an umbrella review to establish a baseline of the peer-reviewed research which has been conducted and published to assess the health outcomes associated with air pollution exposure, specifically in the WBPA. Just over seventy peer-reviewed research studies were included, based on the systematic search criteria. Fewer than ten studies considered air quality and health in the WBPA (as opposed to only air quality) and of these studies, only a few collected human health data in relation to air pollution exposure. Identified studies together showed that poor air quality is a problem in the WBPA, with ambient air quality levels often exceeding national ambient air quality standards. Based on the findings, we recommend that more focused health studies be conducted in the WBPA to advance our understanding of the air pollution-related health burden at the population and the individual level. Such studies will help bolster the baseline evidence of the impacts of air pollution on human health and wellbeing in the WBPA and support decision-making in the future.

Keywords

air quality, environmental health, epidemiology, umbrella review, South Africa

Introduction

In September 2021, the World Health Organization (WHO) released revised ambient air quality guidelines (AQGs) which are more ambitious than those proposed in 2005 (WHO, 2006; WHO, 2021a). Based on the latest scientific evidence, the new AQGs reinforce the need for urgent action to address air pollution and improve the health and wellbeing of people, especially for vulnerable populations. Around 90% of the global population is exposed to air pollution at concentrations above the WHO AQGs (DFFE 2019). Globally, there are an estimated 7.4 million premature deaths from air pollution exposure every year (WHO 2021b; WHO 2022).

Air pollution is a longstanding environment and health problem in South Africa (DFFE 2019). There are numerous sources of air pollution including from coal-fired power stations, mines, industry, residential burning of fuels, vehicles, biomass burning / veld fires and unpaved roads (DEA 2014). These emissions have environmental and health impacts, hence a network to monitor criteria pollutant concentrations is run to monitor air quality around the country. These concentrations are compared with the South African National Ambient Air Quality Standards (NAAQSs) which were set to indicate what levels of exposure to pollution are generally "safe" for most people's health.

South Africa has a long history in air quality management to ensure all its citizens the right to a clean, healthy environment, officially instituted in The Constitution (RSA 1996). One of the mechanisms that South Africa (specifically the Department of Forestry, Fisheries and Environment) has implemented, is the declaration of air pollution priority areas. These have been put in place to highlight geographical areas in which elevated concentrations of criteria pollutants are, or could be, present and where the concentrations are likely to exceed the NAAQS, and where air quality management activities are required to address the air pollution problem. The Vaal Triangle Airshed Priority Area was declared in 2007 and the Highveld Priority Area was later

declared in 2008 (DEAT 2006; DEAT 2007). Several studies have been conducted in these two priority areas (Terblanche et al., 1992; Zwi et al., 1990; Shirinde et al., 2014; Albers et al., 2015; Wright et al., 2018). The Department of Environmental Affairs commissioned air pollution-related health baseline assessment studies in each of these priority areas between 2010 and 2020. Results of the Vaal report have recently been written up into a manuscript (Phaswana et al., 2022), while the Highveld report is not yet publicly available.

The Waterberg-Bojanala Priority Area (WBPA) was declared as the third national air quality Priority Area on 15 June 2012 in line with the precautionary principle of the National Environmental Management Act (Act No. 107 of 1998) due to planned developments in the area (DEA 2012). The Priority Area's main emission sources are mining, industry, residential areas, motor vehicles and biomass burning (DEA 2014). The WBPA Air Quality Management Plan: Baseline Characterisation Report describes the state of air two years after the Priority Area was declared. The report listed twelve government-owned ambient air quality monitoring stations and two industry-owned stations. Sulphur dioxide (SO_2), nitrogen dioxide (NO_2) and benzene (C_6H_6) concentrations were deemed to be relatively low in the Priority Area. While the concentrations of particulate matter (PM_{10} and $PM_{2.5}$) were also generally low, there was evidence of exceedances of the both the annual and the daily 2015 and 2016 NAAQS, respectively (DEA 2014).

The report also identified important gaps in knowledge (DEA 2014, p141) for example -

'...health impacts as a result of air pollution in the Waterberg-Bojanala Priority Area are not understood and therefore not prioritised' … 'There is no health baseline with respect to air pollution in the WBPA and ecological impacts are not understood, i.e., with modelling and monitoring, efforts focus on industry, mining and residential fuel burning. Emissions from small boilers, biomass burning, waste management and transport were excluded…' and 'There is generally a poor understanding of air quality and potential impacts on human and ecological health…'

According to the District Health Barometer published in 2019/2020 (Massyn et al., 2020), many of the leading causes of death (e.g., respiratory diseases, preterm birth complications, cerebrovascular disease, hypertensive heart disease and even diabetes) listed in the WBPA District Municipalities have been linked to air pollution exposure in previous research studies around the world (Burkart et al., 2022; Stafoggia et al., 2022).

More research is needed on the health impacts of air pollution in the WBPA. To the best of our knowledge, no study has drawn together the peer-reviewed evidence reporting on air pollution (ambient and / or household air pollution) and health outcomes associated with exposure to air pollution in the WBPA. The aim of this study was to conduct an umbrella review of published literature to fill this gap and to present evidence to inform future studies.

Methods

Study area

This study's geographical area of focus is the WBPA, and the provinces within which it falls i.e., North West Province and Limpopo Province. Figure 1 illustrates the location of the WBPA in South Africa, in relation to the two other Priority Areas. It includes parts of North-West and Limpopo Provinces and covers an area of about 67 000 km^2 and comprises two District Municipalities (i.e., Waterberg and Bojanala) and nine Local Municipalities (i.e., Thabazimbi, Modimolle, Mogalakwena, Bela-Bela, Mookgopong, Lephalale, Moses Kotane, Rustenburg and Madibeng) (DEA 2014).

Ambient air quality monitoring network

The South African Air Quality Information System was used to identify the location (in the form of GPS coordinates) of all ambient air quality monitoring stations located in the WBPA that reported to SAAQIS at the time during which this manuscript was conceptualised and written. Knowing where ambient air pollution is measured on the ground helps us identify areas in which ambient air quality standards are not being met, and consequently where air pollution exposure may be harmful to the health of humans and biodiversity. Though ambient air quality monitoring stations are sparsely located, and their position is mainly focused on industrial air pollution hot spots, they are essential in helping us measure what air pollution concentrations people on the ground in the WBPA may be experiencing. In this study, their location was referenced for contextual purposes to discuss the literature reviewed.

Review methods

An umbrella review, broadly following the PRISMA guidelines (Moher et al., 2009), of published and peer-reviewed research studies considering air pollution and air pollution-related health outcomes and symptoms in the WBPA was conducted. Health was considered broadly to include health and well-being (including mental health, for example).

Table 1: *Search strategy applied to retrieve published articles reporting on air pollution (ambient and / or household) AND respiratory health outcomes associated with exposure to air pollution in (or near to, i.e., in North-West or Limpopo Provinces) the Waterberg-Bojanala Priority Area.*

Term Group 1	Term Group 2	Term Group 3	Combinations
Air pollution OR air quality AND	Health OR Respiratory health OR respiratory health outcomes AND	Waterberg OR Waterberg-Bojanala OR Bojanala OR North-West OR Limpopo	Term from Group 1 plus Term from Group 2 plus Term from Group 3 until all combinations exhausted

The following databases were searched for articles published up until 31 August 2022: Pubmed, Web of Science, ScienceDirect and Google Scholar. The term groups listed in Table 1 were used for the separate searches and in various combinations. The reference lists of included papers were searched to ensure no studies were omitted. Over and above this, the Clean Air Journal's archives from all articles available online to 2022 were systematically checked for applicable studies which may have been missed in the systematic online search. This was done as the CAJ was deemed most likely to have published studies relevant to the aim of this study.

Any studies which took place outside of these geographical areas (Table 1, column 3) and were not written in English were excluded. Municipality-specific Air Quality Management Plans or Specialist Air Quality Reports, which were conducted as part of Environmental Impact Assessments, were not included, as these do not fall under peer-reviewed and published research articles. All epidemiological study designs were considered and there was no limit set for the number of studies included.

Once studies which met our broad search criteria were identified, these were classified as "inside" or "outside" the WBPA and as "having conducted" or "not having conducted" a health-related study. Indirect health assessments, defined as studies which only evaluated their air quality findings against the NAAQS or the WHO Guidelines, but which did not consider actual health data, were not classified as "health studies". Results are discussed descriptively.

Results

Sample description

A total of 51 studies in North-West Province and 18 studies in Limpopo Province were identified as eligible for inclusion in the study (see Table 2 and Supplementary Material). Over and above this, there were three studies, where the geographical scope of the research included both provinces.

Based on the search criteria, 72 studies were identified. Of these 72 studies, 58 were located in the WBPA, and most of these studies were based in North West Province (Figure 1). A fifth of the identified studies included health outcomes in their scope. Only seven studies were both located in the WBPA and considered health outcomes in their research objectives and findings.

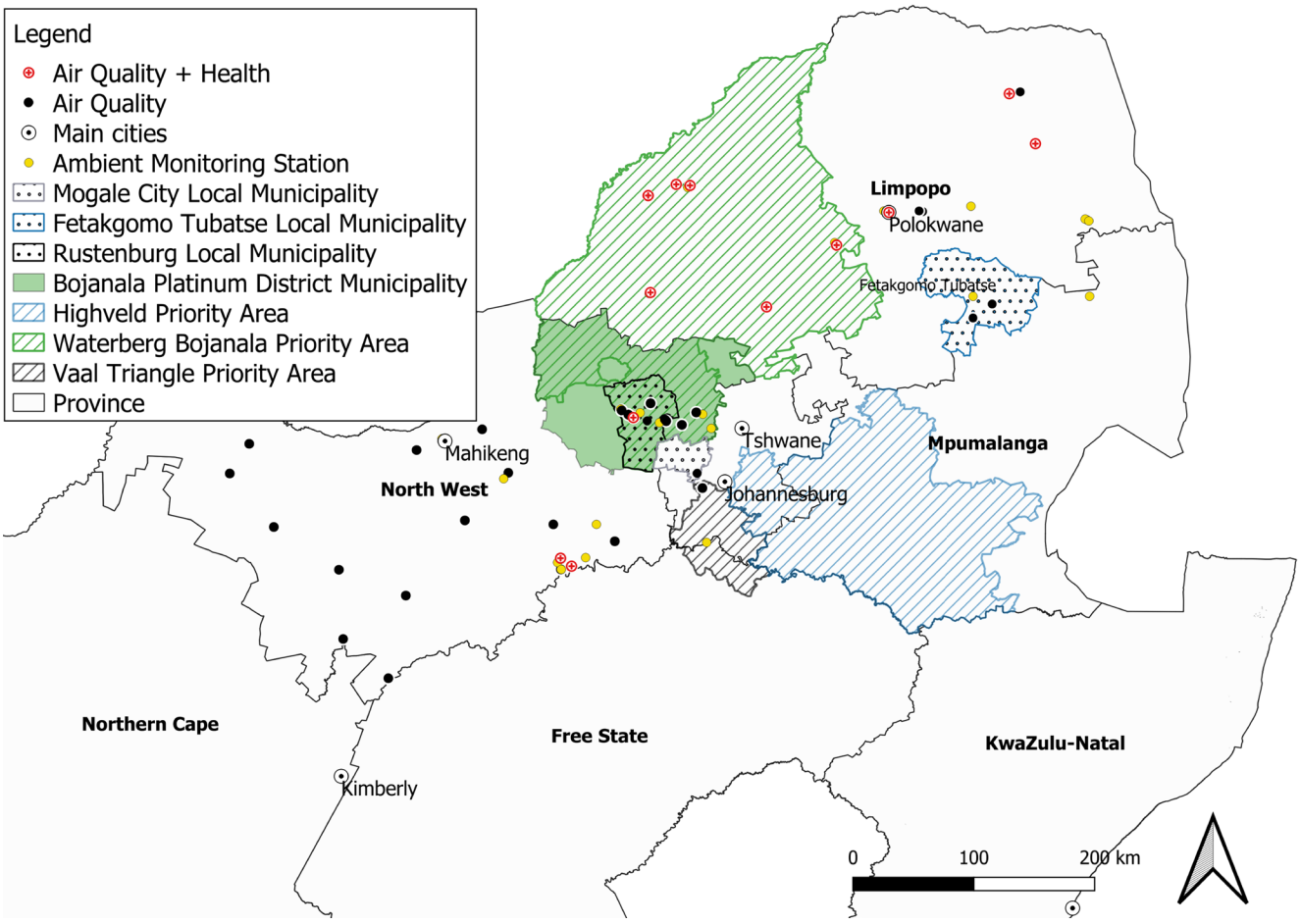

Figure 3: *Spatial overview of "air quality" and "air quality and health" studies in and around the Waterberg-Bojanala Priority Area, Limpopo and North West Province*

Table 2: *Overview of studies identified from systematic literature search*

	Limpopo Province	North West Province	Limpopo and North West	Total
Total number of studies (N)	18	51	3	72
Number of studies in the Priority Area (n)	8	48	3	59
Studies including health in both *provinces* *	5	5	3	13
Studies including health and air quality in the *Priority Area* (n) *	2	2	3	7
Air quality "parameters" considered in all studies re-viewed	Smoke, Dust, Indoor PM_4 Ambient PM_{10}, $PM_{2.5}$, NO_x and BTEX, emission rates, for various pollutants, hydrogen sulphide (H_2S), non-methane hydrocarbons (NMHCs), and volatile organic compounds (VOCs). Indoor and ambient CO, CO_2, O_3, SO_2, NO_2 and H_2S were measured	Indoor PM_{10}, Indoor PM_4, Indoor CO, Personal CO, Ambient $PM_{2.5}$, PM_{10}, SO_2, NO_2, CO and O_3 Biogenic and anthropogenic VOCs, wind-blown dust, Aerosol optical thickness and Ångstrom exponent	Ambient $PM_{2.5}$, Black carbon, Emission inventories	
Details of health data collected	- Respiratory health status (defined in terms of respiratory illness, past and present as well as self-reported respiratory symptoms and spirometry lung function tests) - Health perceptions were collected via means of a questionnaire asking about the presence of breathing disorders, coughing and tuberculosis, asthma and other health issues - Admissions to hospitals for gastrointestinal illnesses including diarrhoea, pneumonia-related diagnosis, malaria and asthma cases - Questionnaire (frequency of medical examinations) - Self-reported respiratory-related health outcomes	- Asthma in school children - The incidence/ community burden of influenza infection - The health risks of air pollution stemming from mining practices - Self-reported results outlining the nuisance of dust and air pollution exposure near the mines, as well as self-reported health impacts which include asthma, sinusitis, eye problems - Psychological responses associated with air pollution exposure	- Number of premature mortalities in South Africa (from coal fired power stations) 2019 population counts per ward were derived from the observed population change between 2011 and 2019 on a district municipal level - Health data were based on district-level health plans and health barometers, supplemented by data from Arrive Alive for road traffic accidents and the National Statistics Service, including the latest national mortality report for South Africa	

Note: The number of studies in the table will not necessarily represent the number of studies indicated on the map, as numerous studies may have taken place in the same area or in a larger area as a whole, instead of a single location (e.g., Rustenburg vs. Rustenburg Local Municipality). Additionally, some studies have included numerous study sites in their scope, so multiple locations on the map may indicate one study.

* Excludes studies which explicitly assessed "health risk" by considering NAAQS compliance only (i.e., they did not measure health impacts directly)

Studies and study findings

Many studies were conducted around large industrial centres (in particular, in Rustenburg and surrounds) where major industrial activities, mining and power generation occur and where ambient air quality monitoring stations are located (Figure 1). Many air quality studies were also conducted in the Welgegund measurement site, approximately 25km north-west of Potchefstroom.

Overall, the air pollution-related parameters which were measured across the studies spanned all primary pollutants (PM_{10}, $PM_{2.5}$, SO_2, NO_2, O_3 and CO) and considered ambient, indoor and personal air pollution levels (See Supplementary Material). Particulates were measured especially frequently in study sites closest to mining activities (e.g., in Rustenburg, see Kgabi et al., 2006) and power generation activities (e.g., Langerman and Pauw 2018). Air pollutants were not only measured using *in situ* air quality instruments, but they were also modelled using a Land Use Regression Model (Simelane and Langerman 2020), as well as dispersion modelling tools (Bryszewski and Visser 2004; DEA 2014; Tshehla and Wright 2019). Remote sensing and satellite data were also used to identify air quality concentrations for various pollutants (Barnes 2015). Amongst the monitored gaseous pollutants, over and above the primary pollutants mentioned, were TVOCs (BTEX), H_2S, CO_2 and non-methane hydrocarbons.

A combination of health outcomes and symptoms were considered in relation to air pollution exposure (See Supplementary Table). Most of the studies focused on respiratory health. This included self-reported and quantitatively measured health parameters. Asthma was the most frequently considered respiratory health outcome (Zwi et al., 1991; Cairncross and Kisting 2016; Njoku et al., 2019; Kapwata et al., 2021). One study considered psychological consequences and responses associated with air pollution exposure (Barnwell 2021).

Air quality and health in the WBPA

We found seven studies located in the WBPA which considered air quality and health (Table 3). Of these studies, four studies used self-reported information as the source of their health data. Only one study, which was published in 1991 conducted physical health measurements (spirometry) in combination with self-reports to identify associations between air pollution and health. Studies considered health data from existing databases/ research to conduct health risk assessments (e.g., Simelane and Langerman 2020). Only one of the seven studies physically measured air pollution levels as part of the study design (i.e., this study did not rely on existing data/data collected by third parties) (Zwi et al., 1991). The remaining studies either used self-reports on air quality as proxy data or sourced their air quality data from pollution sources (e.g., stack emissions), existing ambient monitoring networks or other studies.

Discussion

A review of the available, published and peer-reviewed literature of air quality-related health studies in the WBPA illustrated a paucity of such studies despite the WPBA having been declared ten years ago.

Air quality in the WBPA

Considering the air quality findings from the reviewed material, several studies found that ambient air pollution levels in the WBPA were high and at times exceeded relevant NAAQS. High-stack industry emissions (including from power generation activities) and mining activities as well as combustion activities in semi-formal and informal communities (including domestic fuel burning and waste burning activities) were most quoted as notable pollution sources. A principal component analysis also identified soil dust and traffic as abundant air pollution sources (Kgabi et al., 2006).

A study conducted in an area just south-east of Rustenburg in North-West Province recorded an average of 322 exceedances / annum of the O_3 8-h moving average NAAQS and an average of 42 exceedances / annum of the daily PM_{10} NAAQS (Venter et al., 2012). The remaining pollutants measured (i.e., CO, NO_2 and SO_2) did not exceed their relevant NAAQS. High-stack emissions were identified as the main source of ambient SO_2 concentrations, while household combustion from semi-formal and informal settlements were identified as the predominant sources of PM_{10}, NO_2 and CO. The influx of regional precursor pollutants contributed to the high O_3 concentrations (Venter et al., 2012).

Similarly, in the WBPA, in Limpopo Province, Feig et al., (2016) conducted a study which assessed the ambient air pollution in Lephalale, Thabazimbi and Mokopane. Peak SO_2 concentrations were also attributed to industrial activities, and high morning and evening PM peaks were attributed to domestic burning practices. At times, the daily $PM_{2.5}$ NAAQS were exceeded. High O_3 concentration events were associated with periods with strong winds from other regions (Feig et al., 2016).

More qualitative studies focused on the visible wind-blown dust from mining activities in the WBPA (e.g., platinum, gold, or coal mining). Images of dust blowing off tailing dams in the direction of residential mining communities, as well as self-reports about dust levels in houses by dwelling occupants paint a picture that air quality is poor and represents a health risk in the affected communities.

Though there have been studies measuring indoor air pollution in Limpopo and in North West Province, only one peer-reviewed and published research article identified here measured indoor air pollution levels within the WBPA (Barnes et al., 2011). The study considered behavioural change interventions to improve indoor levels of PM_{10} and CO in solid fuel-using households. The control site, in which indoor pollutants were also measured, was located in the WBPA. Due to indoor fires for heating and / or cooking, daily average indoor PM_{10} and CO concentrations were high, exceeding 'safe' values.

Air pollution exposure and health in the WBPA

Of the two studies (Cairncross and Kisting 2016; Barnwell 2021) which focused on the North West Province section of the WBPA and included air quality and health data, none collected empirical data for both air quality exposure and health outcomes to conduct a rigorous analysis of the associations between the risk and the health outcomes in their respective studies. For air pollution exposure data, these studies relied on self-reported responses, images or previous research findings in the area. The health outcomes were determined by self-reported questionnaire data, self-reported data gathered in focus group meetings and previous study findings of air pollution impacts on human health, including mental health. Cairncross and Kisting (2016) reported on the multiple health problems faced by those who partake in mining activities or are exposed to their emissions (e.g., people reportedly suffered from asthma, sinusitis and eye problems to name a few). Barnwell (2021) highlights the severe health burden placed on physical health and psychological wellbeing when people are exposed to air pollution, especially in highly polluted and poor communities.

One of the two air quality and health studies conducted in the Limpopo Province section of the WBPA (Itzkin 2015) collected self-reported health data (i.e., self-reported respiratory health outcomes at a household level combined with ambient air quality data). An older study conducted by Zwi et al. (1991) collected self-reported respiratory health data, and also conducted lung function tests to find associations between ill health symptoms and air pollution exposure. Zwi et al. (1991) found that respiratory symptoms (e.g., wheezing or coughing) were significantly more common in children who went to school in polluted communities, than in children who went to schools in less polluted areas.

One study developed a Land Use Regression Model to estimate ambient $PM_{2.5}$ concentrations from coal-fired power stations and

Table 3: *Overview of the studies which included health and air quality in the WBPA*

	Article Title	Location of research	Study Population	Study Aim	Study findings
1	Platinum and Gold Mining in South Africa: The Context of the Marikana Massacre (Cairncross and Kisting 2016)	Wonderkop, Marikana, North West Province and Tudor Shaft, Mogale City, Gauteng Province	Mining communities and mining workers	To determine the impact of mining on communities and workers using two case studies of platinum and gold mining in South Africa.	These testimonials captured the extreme poverty as well as multiple co-morbidities caused by mining activities and the social conditions surrounding mines. People reportedly suffered from asthma, sinusitis and eye problems.
2	Expert Report: The Psychological and Mental Health Consequences of Climate Change in South Africa (Barnwell 2021)	South Africa as a whole, with specific mention of Rustenburg and the Bojanala Platinum District	Poor people living in highly polluted communities	This expert report focuses on the considerable health and mental health consequences of climate change due to its impacts on (among others) environmental and planetary health.	Climate change will have insurmountable negative repercussions for South Africa. Air pollution is quoted to have a severe burden on health and psychological wellbeing. Poor people living in highly polluted communities are stressed, as they are, for example, unable to reduce their exposure to air pollution by moving, due to their socio-economic status.
3	Respiratory Health Status of Children in the Eastern Transvaal Highveld (Zwi et al., 1991)	Numerous schools in today's Mpumalanga Province and Limpopo Province (including schools in the now WBPA). The schools which are in the now WBPA were the control schools for this study (as they were deemed to be areas unlikely to be polluted). The study sites were primarily in the now Highveld Priority Area	Primary school children	To determine whether there were detectable effects of the respiratory health status of children as a consequence of exposure to air pollution.	Respiratory symptoms (e.g., wheezing or cough) were significantly more common in exposed children. Of the risk factors tested, attendance at school in the exposed area was the most important risk factor for the development of respiratory symptoms.
4	Health in the Waterberg, Up in Smoke? (Itzkin 2015)	Lephalale, Marapong and Steenbokpan towns in Limpopo Province	Schools and clinics and households	To assess the perceived state of air quality because of fossil fuel burning and domestic activities in the Waterberg using stakeholder surveys and scenario analysis.	Air pollution perceptions and self-reported health outcomes were found to vary with socio-economic status. Qualification of respondents, subscription to medical aid and presence of a ceiling on the home came up as associated with 'air pollution understanding', as well as 'overall rating of household health over the last two years (Itzkin 2015). Annual average concentrations for all measured pollutants were within the respective NAAQS, except for PM_{10} in Marapong, which exceeded the annual NAAQS. Hourly SO_2 exceedances and daily $PM_{2.5}$ and PM_{10} exceedances were measured in Marapong.
5	A critical review of health risk assessments of exposure to emissions from coal-fired power stations in South Africa (Langerman and Pauw 2018)	The air quality priority areas of South Africa, including the WBPA	NA	This paper investigates the reasons for the large discrepancies calculated in five comprehensive health risk assessments of South African coal-fired power station emissions.	Four health risk assessments of emissions from coal fired power stations in South Africa were analysed and classified as over-or underestimating health effects of coal fired power stations. Suggestions are made for improved health risk assessments. A more accurate estimate of health effects would be obtained by applying integrated exposure-response functions to quantify health risks at actual exposure levels, and then apportioning the health effects relative to the contribution made by each source to total exposure levels (Langerman and Pauw 2018).
6	Improving health risk assessments of $PM_{2.5}$ from coal-fired power stations (Simelane and Langerman 2020)	Areas affected by emissions from coal fired power stations, including the WBPA	Population in South Africa exposed to $PM_{2.5}$ from coal-fired power stations	To use a new "proportional log-linear approach" to calculate health outcomes from one component of $PM_{2.5}$. Using this new approach, total premature deaths from exposure to ambient $PM_{2.5}$ levels are first calculated, and then the proportion attributable to the coal-fired power stations assigned.	Emissions from coal-fired power stations contribute between 1.8% and 5.6% of all deaths attributable to $PM_{2.5}$ exposure in the study area (Simelane and Langerman 2020). Coal-fired power station emissions contribute a relatively higher proportion of premature deaths where power stations have the highest contribution to ambient $PM_{2.5}$ concentrations.
7	Health and wellbeing needs and priorities in mining host communities in South Africa: a mixed-methods approach for identifying key SDG3 targets (Rice et al., 2022)	Rustenburg, Thabazimbi and Mogalakwena Local Municipality/ Waterberg and Bojanala District Municipalities	Host communities of 15 mining operations	To identify local needs and priorities relating to SDG3 targets in host communities through stakeholder workshops and key informant interviews.	Poor housing, air quality, and ventilation in clinics, transport, and homes were put forward as key factors in relation to TB (by community members) and air pollution was mentioned as a priority for action by community members living in mining host communities.

their associated health risks on human health (Simelane and Langerman 2020). District municipality-level population data were used to derive population-weighted $PM_{2.5}$ concentrations. The study found that emissions from coal-fired power stations contribute between 1.8% and 5.6% of all deaths attributable to $PM_{2.5}$ exposure in the study area identified (RSA as a whole, in areas affected by $PM_{2.5}$ from power stations). They also found that coal-fired power station emissions contribute to a relatively higher proportion of premature deaths where power stations have the highest contribution to ambient $PM_{2.5}$ concentrations (Simelane and Langerman 2020).

Study limitations

The scope of the umbrella review focused solely on peer-reviewed and published research articles, theses, dissertations and book chapters, but excluded reports which would have been written by air quality specialists as a legal requirement as part of an Environmental Impact Assessment process, or by consultants appointed to conduct an assessment for the government (e.g., for an Air Quality Management Plan or cost-benefit analysis).

There are several such studies which have been conducted within the WBPA due to the high number of industrial activities taking place there, and which have an impact on air quality. These studies will typically have considered the ambient air quality impacts of the industrial activity for which the environmental impact assessment / study was conducted. Air quality specialist studies usually include the results of rigorous dispersion modelling exercises which take into account the emissions created by the proposed activity (e.g., they would use a dispersion model to determine how the activities would influence the ambient air quality in the proximity of the activity, including in any surrounding residential areas). While these are important studies, which should receive special mention here as work which highlights the health risks associated with poor air quality, these are not peer reviewed and published research articles, and were thus not included in this study. This is an acknowledged limitation of this article, and a future study could expand on this research by including such work. Additionally, studies which may have considered air quality and/ or health in the North West Province or Limpopo Province, but did so indirectly, and not as part of their main scope/ research aim of the study, may have been missed (e.g., if a study inadvertently included parts of the study area, as it was located on the periphery of the main research area).

Recommendations for future research

Based on the evidence outlined in this umbrella review, evidence suggests poor air quality in the WBPA represents a human health risk. We need to learn about the health impacts of criteria air pollutants at a population-level, but also at the individual level, so that we can answer questions like: How do chronic and acute ambient and indoor air pollution levels impact on the health of those living in the WBPA? How does air pollution exposure influence the genome of those exposed? How does air pollution exposure influence the unborn children of those mothers who are exposed? What types of interventions can be implemented to reduce air pollution exposure and to improve health?

Conclusions

This review set out to establish a baseline of published and peer-reviewed air pollution and health research studies in the WBPA. Just over 70 studies were identified as relevant based on the systematic search criteria. Only seven studies considered air quality and health in the Priority Area (as opposed to only air quality or only health) and of these, only one actively collected human health data in relation to air pollution exposure. All of the reviewed articles identified air quality as a problem with ambient air quality levels often exceeding relevant NAAQSs. We recommend that well-designed epidemiological health studies be conducted in the WBPA to enhance our understanding of the air pollution-related health burden in the WPBA population.

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References

Albers P, Voyi K, Wright CY and Mathee A. 2015, 'Household fuel use and child respiratory ill health in two South African towns, Mpumalanga', *South African Medical Journal*, 205(7): 573-577. https://doi.org/ 10.7196/SAMJnew.7934.

Aurela, M., Beukes, J.P., Van Zyl, P.G., Vakkari, V., Teinilä, K., Saarikoski, S., Laakso, L. 2016 'The composition of ambient and fresh biomass burning aerosols at a savannah site, South Africa', *South African Journal of Science*, 112(5/6):55-62. http://dx.doi.org/10.17159/sajs.2016/20150223.

Backman, J., Virkkula, A., Vakkari, V., Beukes, J. P., Van Zyl, P. G., Josipovic, M., Piketh, S., Tiitta, P., Chiloane, K., Petäjä, T., Kulmala, M., and Laakso, L. 2014 'Differences in aerosol absorption Ångström exponents between correction algorithms for a particle soot absorption photometer measured on the South African Highveld', *Atmospheric Measurement Techniques*, 7:4285-4298. https://doi.org/10.5194/amt-7-4285-2014.

Barnes B., Mathee A. and Thomas E. 2011, 'The impact of health behaviour change intervention on indoor air pollution indicators in the rural North West Province, South Africa', *Journal of Energy in Southern Africa*, 22:35-44. https://doi.org/10.17159/2413-3051%2F2011%2FV22I3A3220.

Barnes, B.C. 2015, 'The Spatial Distribution of Haze Over the Bojanala District', *MSc dissertation, University of the Witwatersrand, Johannesburg, South Africa*. Available at: https://core.ac.uk/download/pdf/188768999.pdf. [Accessed 25 September 2022].

Barnwell, G. 2021, 'The Psychological and Mental Health Consequences of Climate Change in South Africa', Available at:

https://cer.org.za/wp-content/uploads/2021/09/CER-Expert-Report-Garret-Barnwell-31-August-2021-Public-1.pdf. [Accessed on 25 September 2022].

Bird, T., Liebenberg-Enslin, H., v Gruenewaldt, R., Modisamongwe, D., Thivhafuni, P. And Mphahlele, T. 2012, 'Developing an Air Quality Management Plan: Lessons from Limpopo'. Available at: https://docplayer.net/101051882-Developing-an-air-quality-management-plan-lessons-from-limpopo.html [Accessed 10 December 2021].

Booyens, W., Van Zyl, PG., Beukes, JP., Ruiz-Jimenez, J., Kopperi, M., Riekkola, M-L., Josipovic, M., Venter, A., Jaars, K., Laakso, L., Vakkari, V., Kulmala, M., Pienaar, J.J. 2015 'Size-resolved characterisation of organic compounds in atmospheric aerosols collected at Welgegund, South Africa', *Journal of Atmospheric Chemistry*, 72:43-64. https://doi.org/10.1007/s10874-015-9304-6.

Booyens, W., Beukes, J.P., Van Zyl, P.G. Ruiz-Jimenez, J., Kopperi, M., Riekkola, M.-L., Josipovic, M., Vakkari, V., Laakso, L. 2019, 'Assessment of polar organic aerosols at a regional background site in southern Africa', *Journal of Atmospheric Chemistry*, 76,89–113. https://doi.org/10.1007/s10874-019-09389-y.

Booyens, W., Van Zyl, G. P.,Beukes, P.J.,Ruiz-Jimenez, J., Kopperi, M., Riekkola, M.-L., Vakkari, V., Josipovic, M., Kulmala, M., Laakso, L. 2019, 'Characterising Particulate Organic Nitrogen at A Savannah-Grassland Region in South Africa', *Atmosphere*, 10(9), 492. https://doi.org/10.3390/atmos10090492.

Bryszewski W. and Visser J. 2004, Air quality management in the North West Province of South Africa–A successful partnership with industry. *In Proceedings: Tenth International Ferroalloys Congress'*, 1:4. Available at: oai:CiteSeerX.psu:10.1.1.616.4941. [Accessed on 10 December 2021].

Burkart, K., et al. 2022, 'Estimates, trends, and drivers of the global burden of type 2 diabetes attributable to $PM_{2.5}$ air pollution, 1990-2019: an analysis of data from the Global Burden of Disease Study 2019', *The Lancet Planetary Health*, 6(7):e586-e600. https://doi.org/10.1016/S2542-5196(22)00122-X.

Cairncross, E. and Kisting S. 2016, 'Platinum and Gold Mining in South Africa: The Context of the Marikana Massacre', NEW SOLUTIONS: *A Journal of Environmental and Occupational Health Policy*, 25(4):513-534. https://doi.org/10.1177/1048291115622027.

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

Cohen C., Kleynhans J., Moyes J., McMorrow M.L., Treurnicht F.K., Hellferscee O., Mathunjwa A., von Gottberg A., Wolter N., Martinson N.A. and Kahn K. 2021, Asymptomatic transmission and high community burden of seasonal influenza in an urban and a rural community in South Africa, 2017–18 (PHIRST): a population cohort study, *The Lancet Global Health*, 9:e863-e874. https://doi.org/10.1016/S2214-109X(21)00141-8.

Cronje, F. and Chenga, C. 2007, 'Health issues in a mining community in South Africa. In HIV/AIDS, Illness, and African Well-Being', Editors Falola, T. and Heaton, M.M., Rochester, University of Rochester Press.

Cronjé, F., Reyneke, S. and Van Wyk, D., 2013, 'Local communities and health disaster management in the mining sector', *Jàmbá: Journal of Disaster Risk Studies* 5(2). http://dx.doi. org/10.4102/jamba.v5i2.78.

Dambisya, Y.M. and Modipa, N.B. 2007, 'Risk of exposure to silica dust at some dust-generating workplaces in the Limpopo Province: a survey and recommendations', *Occupational Health Southern Africa*. Available at: https://www.occhealth.co.za/index.php/files/info@sasom.org?/viewArticle/860. [Accessed 01 February 2022].

Department of Environmental Affairs (DEA). 2012, '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. [Accessed 15 September 2015].

Department of Environmental Affairs (DEA). 2014, 'The Waterberg-Bojanala Priority Area Air Quality Management Plan: Baseline Characterization, October 2014', Available at: https://saaqis.environment.gov.za/Pagesfiles/Annexure%20A1%20-%20Waterberg%20Bojanala%20Priority%20Area%20AQMP%20Baseline%20assessment_24-04-2015.pdf [Accessed 15 September 2022].

Department of Environmental Affairs and Tourism DEAT, 2006, 'Declaration of the Vaal Triangle Air-Shed Priority Area', Available at: https://www.gov.za/sites/default/files/gcis_document/201409/28732b.pdf. [Accessed 15 September 2022].

Department of Environmental Affairs and Tourism (DEAT). 2007, 'Declaration of the Highveld as Priority Area', Available at: https://www.gov.za/sites/default/files/gcis_document/201409/30518.pdf. [Accessed 15 September 2022].

Department of Forestry, Fisheries and the Environment (DFFE). 2019, 'The state of our global air report', Available at: https://soer.environment.gov.za/soer/CMSWebSite/Reporting.aspx?menuId=4323# [Accessed on 15 September 2022].

Feig, G., Naidoo, S., & Ncgukana, N. 2016, 'Assessment of ambient air pollution in the Waterberg Priority Area 2012-2015', *Clean Air Journal* 26(1):21–28. https://doi.org/10.17159/2410-972X/2016/v26n1a9.

Gierens, R., Laakso, L., Mogense, D., Vakkari, V., Beukes, J.P., va Zyl, P.G., Hakola, H., Guenther, A., Pienaar, J.J., Boy, M. 2014 'Modelling new particle formation events in the South African savannah', *South African Journal of Science*, 110(5/6). http://dx.doi.org/10.1590/sajs.2014/20130108.

Hirsikko, A., Vakkari, V., Tiitta, P., Manninen, H. E., Gagné, S., Laakso, H., Kulmala, M., Mirme, A., Mirme, S., Mabaso, D., Beukes, J. P., and Laakso, L. 2012, 'Characterisation of sub-micron particle number concentrations and formation events in the western Bushveld Igneous Complex, South Africa', *Atmospheric Chemistry and Physics*, 12:3951-3967. http://dx.doi.org/10.5194/acp-12-3951-2012.

Hirsikko, A., Vakkari, V., Tiitta, P., Hatakka, J., Kerminen, V.-M., Sundström, A.-M., Beukes, J. P., Manninen, H. E., Kulmala, M., and Laakso, L. 2013, 'Multiple daytime nucleation events in semi-clean savannah and industrial environments in South Africa: analysis based on observations', *Atmospheric Chemistry and Physics*, 13:5523-5532. http://dx.doi.org/10.5194/acp-13-5523-2013.

Itzkin, A. 2015, 'Health in the Waterberg, Up in Smoke?' *MSc Thesis, University of the Witwatersrand, South Africa*. Available at: 188770545.pdf (core.ac.uk) [Accessed on 01 February 2022].

Jaars K. 2012, Temporal assessment of volatile organic compounds at a site with high atmospheric variability in the North–West Province, PhD Thesis, *North-West University, Potchefstroom, South Africa*. Available at: https://repository.nwu.ac.za/handle/10394/9513 [Accessed on 28 January 2022].

Jaars, K., Beukes, J. P., van Zyl, P. G., Venter, A. D., Josipovic, M., Pienaar, J. J., Vakkari, V., Aaltonen, H., Laakso, H., Kulmala, M., Tiitta, P., Guenther, A., Hellén, H., Laakso, L., and Hakola, H. 2014 'Ambient aromatic hydrocarbon measurements at Welgegund, South Africa', *Atmospheric Chemistry and Physics*, 14:7075-7089. https://doi.org/10.5194/acp-14-7075-2014.

Jaars, K., van Zyl, P. G., Beukes, J. P., Hellén, H., Vakkari, V., Josipovic, M., Venter, A. D., Räsänen, M., Knoetze, L., Cilliers, D. P., Siebert, S. J., Kulmala, M., Rinne, J., Guenther, A., Laakso, L., and Hakola, H. 2016 'Measurements of biogenic volatile organic compounds at a grazed savannah grassland agricultural landscape in South Africa', *Atmospheric Chemistry and Physics*, 16(24):15665-15688. https://doi.org/10.5194/acp-16-15665-2016.

Jaars, K., Vestenius, M., van Zyl, P. G., Beukes, J. P., Hellén, H., Vakkari, V., Venter, M., Josipovic, M. and Hakola, H. 2018, 'Receptor modelling and risk assessment of volatile organic compounds measured at a regional background site in South Africa', *Atmospheric Environment*, 172(1):133–148. https://doi.org/10.1016/j.atmosenv.2017.10.047.

Kaonga, B. and Ebenso, E.E. 2011, 'An Evaluation of Atmospheric Aerosols in Kanana, Klerksdorp Gold Mining Town, North-West Province, South Africa'. In Mazzeo, N.A. (ed.) Air Quality Monitoring, Assessment and Management, *Rijeka: InTech*, pp285 - 304. https://www.intechopen.com/chapters/16226. https://doi.org/10.5772/17826.

Kapwata T., Language, B., Piketh S. and Wright C.Y. 2018, Variation of indoor particulate matter concentrations and association with indoor/outdoor temperature: a case study in rural Limpopo, South Africa, *Atmosphere*, 9:124. https://doi.org/10.3390/atmos9040124

Kapwata T., Piketh, S. and Wright, C.Y. 2019, Indoor particulate matter concentration variations and associations with indoor/outdoor temperature in rural Limpopo, *Clean Air Journal*, 29:17-17. http://dx.doi.org/10.17159/2410-972x/2019/v29n1a46.

Kapwata T., Wright C.Y., du Preez D.J., Kunene Z., Mathee A., Ikeda T., Landman W., Maharaj R., Sweijd N., Minakawa N. and Blesic S. 2021, Exploring rural hospital admissions for diarrhoeal disease, malaria, pneumonia, and asthma in relation to temperature, rainfall and air pollution using wavelet transform analysis, *Science of The Total Environment*, 791():148307. https://doi.org/10.1016/j.scitotenv.2021.148307.

Kgabi N.A., Pienaar J.J. and Kulmala M. 2006, Sources of atmospheric pollutants in the North West province of South Africa: a case of the Rustenburg municipality, *WIT Transactions on Ecology and the Environment*, 99. https://doi.org/10.2495/rav060581.

Kgabi, N.A., Pienaar, J.J. and Kulmala, M. 2005, 'Trace Metal composition of atmospheric aerosols in the North West province of South Africa,' *Geophysical Research Abstracts*, 7():03229. Available at: http://repository.nwu.ac.za/bitstream/handle/10394/1504 [Accessed on 28 January 2022].

Kok, L., van Zyl, P.G. Beukes, J.P., Swartz, J-S., Burger, R.P., Ellis, S., Josipovic, M., Vakkari, V., Laakso, L., and Kulmala, M. 2021, 'Chemical composition of rain at a regional site on the South African Highveld', *Water SA*, 47 (3). https://doi.org/10.17159/wsa/2021.v47.i3.11861.

Korhonen, K., Giannakaki, E., Mielonen, T., Pfüller, A., Laakso, L., Vakkari, V., Baars, H., Engelmann, R., Beukes, J. P., Van Zyl, P. G., Ramandh, A., Ntsangwane, L., Josipovic, M., Tiitta, P., Fourie, G., Ngwana, I., Chiloane, K., and Komppula, M. 2014, 'Atmospheric boundary layer top height in South Africa: measurements with lidar and radiosonde compared to three atmospheric models', *Atmospheric Chemistry and Physics*, 14:4263-4278. https://doi.org/10.5194/acp-14-4263-2014.

Kuik, F., Lauer, A., Beukes, J. P., Van Zyl, P. G., Josipovic, M., Vakkari, V., Laakso, L., and Feig, G. T. 2015, 'The anthropogenic contribution to atmospheric black carbon concentrations in southern Africa: a WRF-Chem modeling study', *Atmospheric Chemistry and Physics*, 15: 8809-8830. https://doi.org /10.5194/acp-15-8809-2015.

Laakso, L., H. Laakso, P.P. Aalto, P. Keronen, T. Petäjä, T. Nieminen, T. Pohja, E. Siivola, M. Kulmala, N. Kgabi, D. Phalatse, M. Molefe, D. Mabaso, K. Pienaar, Kerminen, V.-M. 2008, 'Basic characteristics of atmospheric particles, trace gases and meteorology in a relatively clean Southern African Savannah environment', *Atmospheric Chemistry and Physics*, 8: 4823-4839. https://doi.org /10.5194/acp-8-4823-2008.

Laakso, L., Vakkari, V., Virkkula, A., Laakso, H., Backman, J., Kulmala, M., Beukes, J. P., van Zyl, P. G., Tiitta, P., Josipovic, M., Pienaar, J. J., Chiloane, K., Gilardoni, S., Vignati, E., Wiedensohler, A., Tuch, T., Birmili, W., Piketh, S., Collett, K., Fourie, G. D., Komppula, M., Lihavainen, H., de Leeuw, G., and Kerminen, V.-M. 2012, 'South African EUCAARI measurements: seasonal variation of trace gases and aerosol optical properties', *Atmospheric Chemistry and Physics*, 12:1847-1864. https://doi.org/10.5194/acp-12-1847-2012.

Laakso, L., Merikanto, J., Vakkari, V., Laakso, H., Kulmala, M., Molefe, M., Kgabi, N., Mabaso, D., Carslaw, K. S., Spracklen, D. V., Lee, L. A., Reddington, C. L., and Kerminen, V.-M. 2013, 'Boundary layer nucleation as a source of new CCN in savannah environment', *Atmospheric Chemistry and Physic*s, 13:1957-1972. https://doi.org /10.5194/acp-13-1957-2013.

Laban, T. L., van Zyl, P. G., Beukes, J. P., Vakkari, V., Jaars, K., Borduas-Dedekind, N., Josipovic, M., Thompson, A. M., Kulmala, M., and Laakso, L. 2018, 'Seasonal influences on surface ozone variability in continental South Africa and implications for air quality', *Atmospheric Chemistry and Physics*, 18(1):15491-15514. https://doi.org/10.5194/acp-18-15491-2018.

Laban, T. L., van Zyl, P. G., Beukes, J. P., Mikkonen, S., Santana, L., Josipovic, M., Vakkari, V., Thompson, A. M., Kulmala, M. and Laakso, L. 2020, 'Statistical analysis of factors driving surface ozone variability over continental South Africa', *Journal of Integrative Environmental Sciences*, 1–28. https://doi.org/doi:10.1080/1943815X.2020.1768550.

Langerman, K. E., & Pauw, C. J. 2018, 'A critical review of health risk assessments of exposure to emissions from coal-fired power stations in South Africa', *Clean Air Journal*, 28(2). https://doi.org/10.17159/2410-972X/2018/v28n2a19.

Lethoko M.X. 2016, Inclusion of climate change strategies in municipal Integrated Development Plans: A case from seven municipalities in Limpopo Province, South Africa, *Jàmbá: Journal of Disaster Risk Studies*, 8:1-6. http://dx.doi.org/10.4102/jamba.v8i3.245.

Mafusire, G., Annegarn, H. J. Vakkari, V., Beukes, J.P., Josipovic, M., van Zyl, P.G. and Laakso,L. 2016 'Sub-micrometer aerosols and excess CO as tracers for biomass burning air mass transport over southern Africa', *Journal of Geophysical Research: Atmospheres*, 121(10):262-282. https://doi.org/10.1002/2015JD023965.

Massyn, N., Day, C., Ndlovu, N. and Padayachee, T. 2020, 'District Health Barometer 2019/2020. Health Systems Trust;, Available at: https://www.hst.org.za/publications/District%20Health%20Barometers/DHB%202019-20%20Complete%20Book.pdf . [Accessed 26 September 2022].

Moher, D., Liberati, A., Tetzlaff, J., Altman, D. G. 2009,' Preferred reporting items for systematic reviews and meta-analyses: the PRISMA statement', *British Medical Journal*, 339:b2535. https://doi.org/10.1136/bmj.b2535.

Mokgoetsi, K.J. 2015, 'The environmental effects of population growth: a case study of the Rustenburg core area (Bojana Region) in North West Province (South Africa)', *MSc dissertation. North-West University*. Available at: https://repository.nwu.ac.za/handle/10394/38644?show=full. [Accessed 25 September 2022].

Momoh A., Mhlongo S.E., Abiodun O., Muzerengi C. and Mudanalwo M. 2013, Potential implications of mine dusts on human health: A case study of Mukula Mine, Limpopo Province, South Africa, Pakistan *Journal of Medical Sciences*, 29:1444-1446. http://dx.doi.org/10.12669/pjms.296.3787.

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.

Mundackal A. and M Ngole-Jeme V. 2020, Evaluation of indoor and outdoor air quality in university academic buildings and associated health risk, *International Journal of Environmental Health Research*, 0(0): 1-19. https://doi.org/10.1080/09603123.2020.1828304.

Mundackal A.J. 2020, An Assessment of Indoor and Outdoor Air Quality in a University Environment: A Case of University of Limpopo, South Africa', *PhD Thesis. University of Pretoria, Hatfield, South Africa*. https://uir.unisa.ac.za/handle/10500/27534.

Ncipha, X.G. 2011, 'Comparison of air pollution hotspots in the Highveld using airborne data', *MSc dissertation. University of Witwatersrand*. Available at: https://core.ac.uk/download/pdf/39669036.pdf. [Accessed 25 September 2022].

Ngoasheng M., Beukes J.P., van Zyl P.G., Swartz J.S., Loate V., Krisjan P., Mpambani S., Kulmala M., Vakkari V. and Laakso L. 2021, Assessing SO_2, NO_2 and O_3 in rural areas of the North West Province, *Clean Air Journal*, 31:1-14. http://dx.doi.org/10.17159/caj/2021/31/1.9087.

Njoku P.O., Edokpayi J.N. and Odiyo J.O. 2019, Health and environmental risks of residents living close to a landfill: A case study of Thohoyandou Landfill, Limpopo Province, South Africa, *International Journal of Environmental Research and Public Health*, 16:2125. https://doi.org/10.3390/ijerph16122125.

Nkosi V., Wichmann J. and Voyi K. 2017, Indoor and outdoor PM 10 levels at schools located near mine dumps in Gauteng and

North West Provinces, South Africa, *BMC Public Health*, 7:1-7. https://doi.org/10.1186/s12889-016-3950-8.

Paasonen,P., Asmi, A., Petäjä, T., Kajos, M. K., Äijälä, M., Junninen, H., Holst, T., Abbatt, J.P.D., Arneth, A., Birmili, W., van der Gon, H.D., Hamed, A., Hoffer, A., Laakso, L., Laaksonen, A., Leaitch, W.R., Plass-Dülmer, C. Pryor, S.C., Räisänen, P., Swietlicki, E., Wiedensohler, A., Worsnop, D.R., Kerminen, V.-M. and Kulmala, M. 2013, 'Warming-induced increase in aerosol number concentration likely to moderate climate change', *Nature Geoscience*, 6:438-442. https://doi.org/10.1038/ngeo1800.

Phasawa, S., Wright Y.C., Garland, R.M., Khumalo, N.T. and Naidoo, R.N. 2022, 'Lagged acute respiratory outcomes among children related to ambient pollutant exposure in a high exposure setting in South Africa', Environmental Epidemiology, (in press).

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.

Rice, B., Boccia, D., Carter, D.J. et al. 2022, 'Health and wellbeing needs and priorities in mining host communities in South Africa: a mixed-methods approach for identifying key SDG3 targets', *BMC Public Health*, 22(68). https://doi.org/10.1186/s12889-021-12348-6.

Shirinde J, Wichmann J, Voyi K. 2014, 'Association between wheeze and selected air pollution sources in an air pollution priority area in South Africa: a cross-sectional study', *Environmental Health*, 6;13(1):32. https://doi.org/10.1186/1476-069X-13-32.

Simelane, S.P. and Langerman, K.E. 2020, 'Improving Health Risk Assessments of $PM_{2.5}$ from Coal-fired Power Stations', *Proceedings of the NACA Conference 2020*. Available at: https://www.naca.org.za/uploads/2020_NACA_Conference_Proceedings_Full_Papers.pdf. [Accessed 25 September 2022].

Stafoggia, M., et al. 2022, 'Long-term exposure to low ambient air pollution concentrations and mortality among 28 million people: results from seven large European cohorts within the ELAPSE project', *The Lancet Planetary Health*, 6(1): e9-e18. https://doi.org/10.1016/S2542-5196(21)00277-1.

Steyn S. 2006, The management of aerial particulate pollution: the case of Platinum Industry Smelters in the Rustenburg region of North West Province, South Africa, *PhD Thesis, University of Pretoria, Hatfield, South Africa*. Available at: https://repository.up.ac.za/handle/2263/30332 [Accessed on 28 January 2022].

Terblanche AP, Opperman L, Nel CM, Reinach SG, Tosen G, Cadman A. 1992, 'Preliminary results of exposure measurements and health effects of the Vaal Triangle Air Pollution Health Study', *South African Medical Journal*, 6;81(11):550-6. PMID: 1598646.

Tiitta, P., Vakkari, V., Croteau, P., Beukes, J. P., van Zyl, P. G., Josipovic, M., Venter, A. D., Jaars, K., Pienaar, J. J., Ng, N. L., Canagaratna, M. R., Jayne, J. T., Kerminen, V.-M., Kokkola, H., Kulmala, M., Laaksonen, A., Worsnop, D. R., and Laakso, L. 2014, 'Chemical composition, main sources and temporal variability of PM_1 aerosols in southern African grassland', *Atmospheric Chemistry and Physics*, 14:1909-1927. http://dx.doi.org/10.5194/acp-14-1909-2014.

Tshehla C.E. 2020, Determination of heavy metal composition of particulate matter in a typical chrome and platinum mine area in the Limpopo Province, South Africa, *PhD Thesis, University of Pretoria, Hatfield, South Africa*, Available at: https://repository.up.ac.za/handle/2263/77890 [Accessed on 04 November 2022].

Tshehla C.E. and Wright C.Y. 2019, Spatial and temporal variation of PM_{10} from industrial point sources in a rural area in Limpopo, South Africa, *International Journal of Environmental Research and Public Health*, 16:3455. https://doi.org/10.3390/ijerph16183455.

Tshehla, C. and Djolov, G., 2018, Source profiling, source apportionment and cluster transport analysis to identify the sources of PM and the origin of air masses to an industrialised rural area in Limpopo, *Clean Air Journal*, 28:54-66. http://dx.doi.org/10.17159/2410-972X/2018/v28n2a18.

Vakkari, V., Laakso, H., Kulmala, M., Laaksonen, A., Mabaso, D., Molefe, M., Kgabi, N. and Laakso, L. 2011, 'New particle formation events in semi-clean South African savannah', *Atmospheric Chemistry and Physics*, 11:3333-3346. https://doi.org/10.5194/acp-11-3333-2011.

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

Vakkari, V., Kerminen, V.-M., Beukes, J.P., Tiitta, P., van Zyl, P.G., Josipovic, M., Venter, A., Jaars, K., Worsnop, D., Kulmala, M., and Laakso, L. 2014 'Rapid changes in biomass burning aerosols by atmospheric oxidation', *Geophysical Research Letters*, 41. http://.doi.org/10.1002/2014GL059396.

Vakkari, V., Tiitta, P., Jaars, K., Croteau, P., Beukes, J.P., Josipovic, M., Kerminen, V.-M., Kulmala, M., Venter, A.D., Van Zyl, P.G., Worsnop, D.R., Laakso, L. 2015, 'Reevaluating the contribution of sulfuric acid and the origin of organic compounds in atmospheric nanoparticle growth', *Geophysical Research Letters*, 42(10):486-493. http://dx.doi.org/10.1002/2015GL066459.

Vakkari, V., Beukes, J. P., Josipovic, M. and van Zyl, P. G. 2020, 'Observations of ozone formation in southern African savanna and grassland fire plumes', *Atmospheric Environment* 223, 117256. http://dx.doi.org/10.1016/j.atmosenv.2019.117256.

Van Zyl P.G., Beukes J.P., Du Toit G., Mabaso D., Hendriks J., Vakkari V., Tiitta, P., Pienaar, J.J., Kulmala, M., Laakso, L. 2014, 'Assessment of atmospheric trace metals in the western Bushveld Igneous Complex, South Africa', *South African Journal of Science*, 110(3/4). http://dx.doi.org/10.1590/sajs.2014/20130280.

Venter A.D., Beukes J.P., Van Zyl P.G., Tiitta P., Josipovic M., Pienaar J.J., Laakso L., Vakkari V., Laakso H. and Kulmala M. 2012, An air quality assessment in the industrialised western Bushveld Igneous Complex, South Africa, *South African Journal of Science*, 108;1-10. Available at: http://www.scielo.org.za/scielo.php?script=sci_arttext&pid=S0038-23532012000500017&lng=en&nrm=iso. [Access on 23 January 2022].

Venter, A.D., Jaars, K., Booyens, W., Beukes, J.P., Van Zyl, P.G., Josipovic, M., Hendriks, J., Vakkari, V., Hellén, H., Hakola, H., Aaltonen, H., Ruiz-Jimenez, J., Riekkola M-L., and Laakso, L. 2015 'Plume characterisation of a typical South African braai', *South African Journal of Chemistry*, 68:181–194. https://doi.org/10.17159/0379-4350/2015/v68a25.

Venter, A. D., Beukes, J. P., Gideon van Zyl, P., Josipovic, M., Jaars, K. and Vakkari, V. 2016 'Regional atmospheric Cr(VI) pollution from the Bushveld Complex South Africa', *Atmospheric Pollution Research*, 7(5):762–767. https://doi.org/10.1016/j.apr.2016.03.009.

Venter, A.D., van Zyl, P.G. , Beukes, J.P. , Swartz, J—S., Josipovic, M., Vakkari, V., Laakso, L., Kulmala, M. 2018 'Size-resolved characteristics of inorganic ionic species in atmospheric aerosols at a regional background site on the South African Highveld', *Journal of Atmospheric Chemistry*, 75(1):285-304. https://doi.org/10.1007/s10874-018-9378-z.

Venter, M., Beukes, J. P., van Zyl, P. G., Vakkari, V., Virkkula, A., Josipovic, M., Kulmala, M. and Laakso, L. 2020, 'Six-year observations of aerosol optical properties at a southern African grassland savannah site', *Atmospheric Environment*, 230,117477. http://dx.doi.org/10.1016/j.atmosenv.2020.117477.

World Health Organization (WHO) 2006, 'Air Quality Guidelines. Global Update 2005', Available at: https://www.euro.who.int/__data/assets/pdf_file/0005/78638/E90038.pdf [Accessed 25 September 2022].

World Health Organization (WHO). 2021a, 'Key facts: Ambient (outdoor) air pollution', Available at: https://www.who.int/news-room/fact-sheets/detail/ambient-(outdoor)-air-quality-and-health [Accessed 15 September 2022].

World Health Organization (WHO). 2021b, 'WHO global air quality guidelines: particulate matter ($PM_{2.5}$ and PM_{10}), ozone, nitrogen dioxide, sulfur dioxide and carbon monoxide'. Available at: https://apps.who.int/iris/handle/10665/345329. [Accessed 04 November 2022].

World Health Organization (WHO). 2022, 'Key facts: Household air pollution and health', Available at: https://www.who.int/news-room/fact-sheets/detail/household-air-pollution-and-health [Accessed 15 September 2022].

Wright C.Y. and Wernecke B. 2020, 'Using Microsoft© Power BI© to visualise Rustenburg Local Municipality's Air Quality Data, *Clean Air Journal*, 30:1-5. http://dx.doi.org/10.17159/caj/2020/30/1.7512.

Wright CY, Nkosi V and Wichmann J. 2018, 'Respiratory health symptoms among school children in relation to possible food-related risk and protective factors', *International Journal of Environmental Research and Public Health*, 15(3):502. https://doi.org/10.3390%2Fijerph15030502.

Zwi S, Davies JC, Becklake MR, Goldman HI, Reinach SG, Kallenbach JM. 1991, 'Respiratory health status of children in the eastern Transvaal highveld', *South African Medical Journal*, 78(11):647-53. PMID: 2251608.

Supplementary material

Supplementary material can be accessed at https://cleanairjournal.org.za/article/view/14887