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Editorial
Celebrating the successes of the Clean Air Journal over the past 6 years and looking forward

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Science in South Africa has a triple role to play. First and foremost, it must serve society. It must contribute to the growing knowledge-based economy both nationally and globally and it must be the springboard for young people to find opportunities to begin their careers.

The Clean Air Journal has for almost 50 years been the premier local journal on atmospheric science, air pollution and its impacts, and air quality management. Topics featured in the journal have varied widely over the years, from fundamental air pollution science to the complexities of offsetting, making the Clean Air Journal the largest single repository of South African air quality-related literature.

Since 2013, with an exchange of more than 1 500 email, dozens of meetings coupled with laughs, sweat and tears, I have been a proud Co-Editor of the Clean Air Journal together with Assoc/Prof Rebecca Garland and Dr Gregor Feig. It was a formidable task taking on this role from Past Editor of the Journal, Dr Gerrit Kornelius, and I am pleased to say that together the three of us have done our very best to advance the presence and standard of the Journal both locally and worldwide.

Simultaneous to publishing 11 issues of the Journal between 2013 and 2018, we worked very hard to have the Clean Air Journal indexed in multiple databases, including *Google Scholar*, *Ulrich's ProQuest* and *SHERPA/RoMEO*. Recently, we were also accepted into the *Directory of Open Access Journals* and *Scopus*, although for the latter we are indexed, yet waiting to meet a minimum target of published articles before appearing live on their website. We also remain indexed in *Sabinet*, however, now as an open access journal. The enormous effort that was needed to be included in these databases entailed overhauling the website, archiving all previous Journal publications, writing new Journal policies, adding Digital Object Identifiers to all new articles and mounds of paperwork. But, it has been worth the effort and we are hopeful for two additional exciting developments in the near future.

The Clean Air Journal has for the past 6 years had a 'front section' providing news, commentaries, research briefs and other material about the latest happenings in air quality in South Africa and globally. It has been fascinating to read the topics proposed by Journal contributors. About three-quarters of all published articles include at least one postgraduate student as a co-author highlighting the important role of the Journal in human capital development in South Africa. A very big thank you to all our reviewers for their patience and dedication to the review process in assisting all authors to improve on the science and quality of their submitted manuscripts.

Thank you to our Editorial team for sharing the workload, to the *National Association for Clean Air* for their continued support, to the authors who commit to publishing in the Journal and helping us to improve the quality of publications. I also acknowledge the critical role of our Editorial Board and thank each member for the contributions to the Journal.

I have thoroughly enjoyed my 6-year term as Co-Editor of the Clean Air Journal. I welcome Dr Kristy Langerman into her new role in the editorial team. I will never be more than a phone call away and I will continue to support the journal with submissions and commentaries. I am very excited about the Journal's future and I encourage all who work in the fields of air quality to continue to support the Journal as it transforms from a premier local journal to one of international prestige.

Company Profile

an EOH company

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

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

SI Analytics is currently a B-BBEE level 2 contributor.

Being a founding member of Europa Environmental gives us the expertise to supply and support advanced environmental monitoring technology.

Our aim is to bring you the world's best instrumentation, spare parts, service, technical support and training - at the most affordable pricing - to meet your environmental monitoring needs.

Services

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

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

- Environmental monitoring
- Certified USEPA, European EN and UK MCerts equipment sales
- Data management and reporting
- Service and maintenance contracts
- Equipment rentals
- Indoor air quality assessments
- Short courses (e.g. Operator monitoring assessment training).

Products

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

Instrumentation from the most popular products includes:

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

Clients

an EOH company

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

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

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Letter to the Editor
Response to: "An economic assessment of SO_2 reduction from industrial sources on the highveld of South Africa" by Steyn and Kornelius

Prof. Eugene Cairncross
Emeritus Professor, Chemical Engineering, CPUT

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The article, 'An economic assessment of SO_2 reduction from industrial sources on the highveld of South Africa' (Steyn and Kornelius, 2018) is of considerable interest in the context of Eskom's ongoing applications for "rolling postponements" of the Minimum Emissions Standards. (DEA, 2013)

Analysis of the article indicates that certain abatement costs not related to the emission sources are included in the cost-benefit estimates, and benefits related to sulphate exposure and a larger domain are excluded. These factors and other inconsistencies in the article place doubt on the paper's unequivocal conclusion that

"The results of the study indicate that costs of the implementation of the category 1.1 new plant (2020) SO_2 standard exceed the likely quantifiable benefits due to the high capital and operating cost associated with the implementation of FGD."

1. Sources included in the emissions inventory

In 'Methodology', the authors state that "All category 1.1 sources operational at the time of study within the study area expected to have a significant impact on ambient SO_2 concentrations were included in the study." Figure 3 shows the model domain, and the location of the 'major sources', including the Lethabo Stations 1 and 2 and Secunda Stations 1 and 2 (presumably the Sasol-Synfuels plants) respectively. Sasol Stations 1 and 2, and the Kelvin power plant are also shown. Figures 4 and 5 (modelled SO_2 concentrations) do not include the Lethabo or the Sasol-Synfuels plants and the isopleths do not show the concentration imprints of these two plants. The authors state that Sasol Stations 1 and 2 and Kelvin are ***not*** included in the dispersion modelling.

a) Could the authors please clarify why Sasol Stations 1 and 2 and Kelvin are not included in the modelling though they are within the modelling domain?
b) The Sasol-Synfuels plants ('Secunda' Stations 1 and 2) are clearly within the model domain shown in Figure 3. Please clarify if emissions from these plants are included in the dispersion modelling?

2. Are secondary sulphates included in health impact estimates?

Secondary sulphate formation appears to be included in the modelling but sulphate results are not shown, nor are sulphate benefits included in the health benefits.

Several statements infer that sulphates are considered: page 6, "Chemical transformations were modelled using the Mesopuff II chemical transformation model, included in the Calpuff model."; "The changes in ambient concentration of SO_2 and sulfates between the baseline (current operations) and compliance scenario (emissions at 500 mg/Nm^3) were extracted from the dispersion modelling results for short-term (daily average) and long-term (annual average) impacts."; Tables 1 and 2 include mortality and morbidity response values for sulphates; "Discussion" "The methodology for the health impact assessment was as follows: The changes in ambient concentration of SO_2 and sulfates between the baseline (current operations) and compliance scenario (emissions at 500 mg/Nm3) were extracted from the dispersion modelling results for short-term (daily average) and long-term (annual average) impacts."

In contrast, the statement that "The associated health benefit associated with ***SO_2 mortality impacts*** calculated using the base data was R50 billion, compared to the Asian estimate of R26 billion and the South African estimate of R53 billion ***for the SO_2 only impact.***" [emphasis added] unequivocally states that sulphates are not included in the health benefits. Modelled sulphate concentrations are also not reported.

Are sulphates included in the analysis of the impacts and benefits reflected in Table 5 and Figure 3?

3. The influence of a confined domain on the estimation of health benefits

Figures 4 and 5 appear to reflect a significantly smaller domain compared with Figure 3, the "Model Domain". This excludes the populated areas surrounding the Lethabo power station. Confining the model domain effectively assumes a threshold at its boundaries, particularly pertinent for emissions from Lethabo and Sasol Stations 1 and 2 plants. This results in an underestimate of the exposed and impacted population, and the health benefits. A larger domain would account for the known long-range impacts of industrial emissions from tall stacks. Figure 4 reveals that the domain boundaries are curtailing the estimate of impacts and health benefits.

a) Was the larger model domain shown in Figure 3 the basis for estimating ambient concentrations and population health benefits?
b) Which criteria are used for defining domain boundaries?

4. Capital costs of Flue Gas Desulphurisation (FGD)

In 'Capital costs', "The overall calculated direct capital cost was R187 billion (2020 costs) for all the Eskom stations currently in operation within the study area as well as the Sasol facility at Secunda."

Total FGD capital costs include the Sasol-Secunda plants but the estimate of health benefits does not appear to include the Sasol emissions (Question 1b).

Please clarify if this is the case?

5. Water consumption rates and costs

Water requirements are stated as "The operation of FGD on all the facilities will require an estimated 98 million m^3 of water per annum (Sasol, 2014; Eskom, 2014).". Eskom estimated (Eskom, 2014) that implementing FGD ***on all its plants*** (including Medupi) would increase its water consumption by 70 million m^3 (or 67 million m^3 per annum in Table 4.) per annum. The estimate of a total of 98 million m^3 per annum therefore appears to be high.

a) What is the estimate of water consumption for Eskom's plants that are included in this study?
b) Have water consumption and cost data for Sasol plants been included while their emissions and health benefits were excluded?
c) Are water consumption figures based on wet FGD, wet FGD with inlet cooler or semi-dry FGD or a mix of these technologies? What is the water consumption split, per technology?

6. The sorbent (lime or limestone) consumption and cost estimates

The estimates of sorbent consumption are: "Sasol requires approximately 180 000 tons of lime per annum, while Eskom will require an estimated 5 000 000 tonnes per annum of lime (Sasol, 2014; Eskom, 2014)."

Medupi proposes to use limestone rather than lime in its proposed wet FGD process (Eskom, 2014. Page 32), and limestone appeared to be the sorbent of choice in Eskom's postponement applications. In 2014, Eskom estimated that "Up to almost 5 million tons of sorbent (limestone) per annum is required to operate the FGD ***across the generating fleet***." (Eskom, 2014. Page 14) [emphasis added] The 5 million tonnes per annum of limestone includes sorbent for Medupi and Matimba. The limestone requirements for the Eskom plants within the study area appear to have been overestimated. Could the authors please clarify the basis of their figure?

The total (30-year lifecycle) limestone cost estimates is given as R50 billion in Table 5 and as R63 billion on page 3. The per tonne cost is R300 and the total limestone consumption rate is 5,18 million tonnes/year. For 2020, the annual limestone cost would be 5.18 million x R300 = R1,554 billion. If total limestone consumption rates remain constant over the 30 year period, the cumulative cost would be R46,6 billion. However, the total annual SO_2 emission rates, would decrease as plants are decommissioned. Limestone annual consumption rates, directly related to SO_2 emissions and removal rates, should therefore decline proportionately resulting in a significantly lower 30-year cumulative cost.

Please clarify the calculation of the 30-year total limestone cost, for example, with a year-on-year schedule of consumption (and cost), based on the assumed decommissioning schedule.

7. Clarification of the emissions scenarios used in the modelling

The year-on-year difference in baseline and compliance emissions scenarios is not clear. The assumptions were: ".. all retrofits could be completed by 2020" (page 7); ".. the plants would be decommissioned according to schedule and that their lifetime will not be extended."; "Costs and benefits were only calculated for the remaining life of each facility." The Eskom decommissioning dates in their postponement applications, are: "Camden: 2020-2023; Hendrina: 2020-2026; Arnot: 2021-2029; Komati: 2024-2028; Grootvlei: 2025-2028; Kriel: 2026-2029." (Eskom, 2014. Page 17) Therefore the full cost of retrofitting these plants would be incurred by 2020 but the benefits of reduced SO2 emissions would reduce as these plants are decommissioned. For Camden and Hendrina, retrofitting would be completed in 2020 and decommissioning also begin in 2020. It would clearly be preferable to not retrofit units scheduled for immediate decommissioning, thus saving FGD capital costs. For Camden, Hendrina and Arnot, compliance could be achieved through a combination of accelerated decommissioning of some units and retrofitting of the remainder. This could significantly shift the balance of costs and benefits.

Please clarify year-on-year costs and benefits by providing, for example, the assumed year-on-year baseline and compliance emissions scenarios, and corresponding costs.

Yours sincerely
Prof. Eugene Cairncross
Emeritus Professor, Chemical Engineering, CPUT

8. References

DEA (2013) Department of Environmental Affairs National Environmental Management: Air Quality Act (39/2004): List of Activities which result in atmospheric emissions which have or may have a significant detrimental effect on the environment, including health, social conditions, economic conditions, ecological conditions or cultural heritage. Government Notice 893 of 2013.

Eskom (2014) Applications for postponement of the Minimum Emissions Standards (MES) for Eskom's Coal and Liquid Fuel- Fired Power Stations, February 2014.

Steyn M. and Kornelius G. 2018, 'An economic assessment of SO_2 reduction from industrial sources on the highveld of South Africa'. *Clean Air Journal*. 28:23-33.

Authors' Response to Letter to the Editor "An economic assessment of SO_2 reduction from industrial sources on the highveld of South Africa" by Steyn and Kornelius

Marilize Steyn[1] and Gerrit Kornelius[2]

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Dear Editor

Thank you for the opportunity to respond to the Letter to the Editor sent by Prof Cairncross as a response to the Paper "An Economic assessment of SO_2 reduction from industrial sources on the highveld of South Africa" by Steyn and Kornelius, that was published in the Clean Air Journal Vol 28 No 1.

In the response letter Prof Cairncross raised seven points for clarification, which are highlighted in the response.

1. Sources included in the emissions inventory, 3. impacts of confined domain and 4. capital costs of flue gas desulphurisation

The study focused on the Highveld Priority Area (HPA) and as such included only the sources located in the HPA that were operational at the time of the study. As the Sasol Synfuels plants are located within this area, the emissions thereof were included in the model and the benefits associated with emission reduction from that facility were included in the calculations (point 1 and 4). Category 1.1 (in terms of the Air Quality Act section 21 regulations) sources that are not located in the Highveld Priority Area were not included in this study. The HPA was chosen as the area for analysis as it is a declared priority area with defined borders which hosts the majority of the sources that fall under category 1.1. The model domain was larger than the analysis area, but benefits were only calculated for populations residing in the priority area. We must emphasize that that analysis was done on concentration ***differences*** for the options, which would obviate or reduce to a minimum the effect of sources outside the modelling domain.

2. Are secondary sulphates included in health impact estimates?

The study considered both direct SO_2 impacts as well as the impacts associated with the resulting secondary sulfate formation. The results that are referred to in point 2 of the letter extracted only the SO_2 impacts for purposes of comparing the impacts when concentration response functions are used that are derived in different parts of the world to ascertain whether the concentration response functions derived in the United States were comparable with those derived in South Africa and Asia. Since the South African study that was used only considered the impacts associated with SO_2 and not sulfate, these impacts were discussed only in the context of that comparison. The analysis indicated that the concentration response functions derived in the United States compared well with a study conducted in South Africa. All other benefit calculations include the impact of sulfates.

6. The sorbent (lime or limestone) and water (point 5) consumption and cost estimates

Apportioning the 5 000 000 tons of limestone required per annum and deducting the usage by facilities not included in the study does result in an estimated limestone usage lower than the amount used in the study. It would be preferable to use the Eskom estimate, resulting in a 30% reduction in the calculated limestone cost. The limestone costs account for 20% of the net present value and therefore using the Eskom figure, the total cost will be adjusted by 6%. The limestone and water costs do reduce as the facilities are decommissioned. All costs and benefits were escalated using CPI to determine net present values, which would increase the NPV beyond merely multiplying by the number of years. The study assumed that wet FGD would be the preferred option, as discussed in the article.

7. Clarification of the emissions scenarios used in the modelling

It is indeed correct to state that it would not be preferable to retrofit facilities that are due to be decommissioned. The study considered the case of retrofitting ***only*** the station with the largest health impact. The results indicated that the costs still outweigh the benefits, albeit at a reduced ratio. The study only considered the retrofitting of the facilities and not benefits associated with accelerated decommissioning. The study indicates that retrofitting is associated with high operating costs

and alternative solutions such as accelerated decommissioning may well be preferable. While these alternatives fell outside the scope of our study, it would be quite informative to determine such benefits.

The study did not take into consideration the impact of an increase in electricity tariffs, which may have a significant broader economic impact. The costs associated with increased greenhouse gas emissions were similarly not included. The costs can therefore be seen to be conservative. On the benefit side, the largest benefit was the reduction in premature mortality. The increased risk associated with premature mortality used the Value of a Statistical Life (VSL) to monetise the benefit. The VSL used was based on values used by the United States Environmental Protection Agency as values for South Africa are not available. One of the areas for further research identified by the study is the need for the development of appropriate VSL values that are reflective of the economic consequences of premature mortality in South Africa. This value is the single most important factor in determining whether the costs of compliance outweigh the benefits.

Yours faithfully

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uMOYA-NILU

Service Offering:

- **Air quality baseline & impact assessments**
- **Air quality management planning**
- **Air quality monitoring**
- **Stack emission testing**
- **Multi-point calibrations**
- **Emissions inventory compilations**
- **Leak detection and repair**
- **Listed actiity compliance with AEL conditions**
- **Air quality manaement training**

Some Flagship Projects:

- **Development of NAEIS**
- **Development of Highveld and Waterberg-Bojanala Priority Area AQMPs**
- **Development of Gauteng, Western and Eastern Cape AQMPs**
- **Development of Transport Sector GHG Emission Inventory for SA**
- **Supply, Operation & Maintenance of 8 AQ Monitoring Stations for TNPA**

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News
Air Pollution in Emerging Mega-Cities: Sources, Evolution and Impacts Workshop

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A tri-lateral workshop on "Air Pollution in Emerging Megacities: Sources, Evolution and Impacts" was held from 16-20 April 2018 in Nairobi, Kenya, bringing together early career researchers (ECRs) from the United Kingdom (UK), Kenya and South Africa. The workshop had 51 participants and was coordinated by Dr Kirsti Ashworth, Lancaster University (UK), Dr Nicholas Ozor, African Technology and Policy Studies Network (ATPS, Kenya) and Prof Paul Beukes, North-West University (South Africa). It was sponsored by the British Council Newton Fund (UK), National Research Fund (Kenya) and National Research Foundation (South Africa). The workshop was moderated by Dr Akan Odon, Lancaster University and Dr Rebecca Garland, Council for Scientific and Industrial Research (CSIR), Dr David Odee, Kenya Forestry Research Institute (KEFRI), Dr Paul Young (Lancaster University), and Dr Aderiana Mbandi of Stockholm Environment Institute (SEI) were leaders and mentors.

The main aims of the workshop were to bring together ECRs and experienced researchers to:

- establish the current state of the science and the current and future needs of the atmospheric research community to address the growing issues of air pollution, climate change and their impacts on society in and around emerging mega-cities, particularly in sub-Saharan Africa;
- introduce ECRs to a wide range of techniques and skills required to pursue cutting-edge, transformative research in atmospheric composition and air quality;
- raise awareness of the need for transdisciplinary co-produced research to holistically tackle the challenges of the UN Sustainable Development Goals; and
- facilitate cross-boundary networking and identify common experience on which to build future collaborative partnerships with fellow ECRs, established scientists, policy makers and stakeholders.

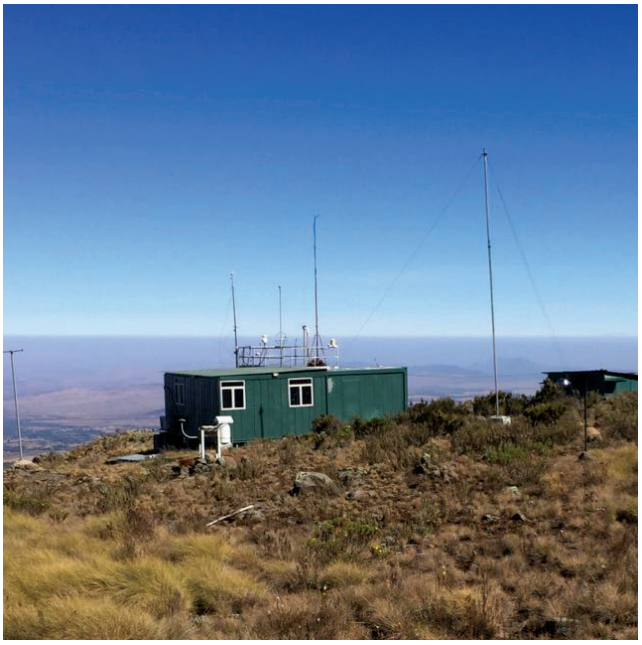
Air pollution is rapidly increasing in emerging mega-cities especially in Africa, affecting both health and natural and managed ecosystems, yet it is a field that has received little study. Air quality is an issue of increasing concern to governments and intergovernmental organisations world-wide, exemplified by a recently-passed resolution from UN Environment, signed by all parties, to tackle air pollution. A key goal of that resolution is to establish global and regional knowledge platforms to bring together all stakeholders with expertise in the field to jointly act.

Participants of the trilateral workshop, Nairobi Kenya

This workshop was a highly interactive 5-day event including presentations from local experts from the policy, governance and academic sectors, intensive group activities and discussions, and a highly informative field trip to key sites around Nairobi. Invited guest speakers included Dr Ozor of ATPS who spoke about policy and strategy to combat air pollution, and Profs Gitari and Madara, from the University of Nairobi (UON), who addressed the current state of the science regarding air pollution measurement techniques. They both noted that AQ data in Africa were sparse with little previous or on-going research work. Other guest speakers included Soraya Smaoun (UN Environment) who spoke of current UN initiatives to tackle air pollution across the globe, Drs Osano and Mbandi from SEI who presented several case studies of community participatory research in Nairobi and further afield, and Mr Nthusi (UN Environment) who recounted details of his work of AQ measurements with the UN. High quality posters were presented by the ECRs with Cassilde Muhonja and William Appondo (joint poster) of SEI winning the peer-judged prize for best poster.

The participants enjoyed a field trip to Kenya Meteorological Department (KMD) to learn of the air pollution measurements and sampling techniques employed in Kenya. KMD has three urban air quality monitoring stations at Dagoretti Corner, Jomo Kenyatta international Airport and Chiromo Campus, UON. KMD also operates Mt Kenya Global Atmosphere Watch station

for long-term measurements of both chemical and physical composition of the earth's atmosphere for early detection of climate variability and change in the region. The station is in a data-sparse region within Africa and provides a unique opportunity to monitor background air pollution as well as to conduct research in a pristine continental environment. KMD also operates a mobile air pollution monitoring laboratory van and low-cost AQ station, which has been deployed to monitor air pollution on major roads and industrial areas in Nairobi City. The data from this campaign showed episodes of extremely poor AQ especially in the morning and evening which coincided with known vehicle traffic jams in Nairobi.



Mt Kenya Global Atmosphere Watch station, Kenya

The participants also visited Mukuru Viwandani, an informal settlement in Nairobi city to learn how SEI were successfully engaging "community champions" to raise awareness of and monitor indoor and outdoor air pollution within the settlement. SEI Africa office is actively engaged in research, policy engagement and capacity building in the region. SEI involves citizens in AQ measurements through citizen science initiatives that aim at involving the community in making measurements and helping them understand the importance of the data collected. The participants were addressed by Wajukuu Project artists and community champions Mr Joseph Waweru, Ms Freshia Njeri and Ms Elizabeth Njoroge. Knowledge and experience gained through citizen science enabled them to link the widespread respiratory diseases and illness with poor AQ.

On their way back to the conference hotel, participants experienced the Nairobi traffic and associated air pollution first hand.

By the close of the workshop, the participants had shared and gained knowledge and expertise regarding passive, active and continuous monitoring of O_3, CO, PM_{10} and $PM_{2.5}$, BC, SO_2 NO_x, VOC and POPs. ECRs learned how these AQ measurements were used for health, environment, and agriculture impact assessments, as the basis of early warning systems and advisories, and for the fulfilment of multilateral conventions and agreements. The participants proposed that integration of all measurements of AQ to an open access portal, availability of low cost and easily accessible devices that can be connected online in near real time, and a combination of super sites and smaller sites would constitute an ideal AQ monitoring situation. The ECRs were also introduced to the potential of AQ modelling for supplementing sparse data coverage, downscaling satellite data, conducting rigorous impact assessments and exploring possible intervention and mitigation strategies to support policy. ECRs discussed the clear links between many of the Sustainable Development Goals (SDGs) and air quality, particularly in the context of emerging mega-cities. Participants also highlighted the need to identify and engage with other stakeholders; i.e. government, academia, industrialists and civil society, and to use research and innovation to deliver a genuinely transformative science agenda.

The workshop participants will follow up the activities in Nairobi by compiling a newsletter, producing an academic review paper of the current knowledge, gaps and needs of air pollution research in Africa, and developing a joint proposal for collaborative research projects. The participants also aim to make this workshop a regional rotational annual event.

Acknowledgments

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For further information, please visit www.newtonfund.ac.uk.

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News

Joint iCACGP-IGAC 2018: 'Atmospheric Chemistry - from molecules to global impacts' in Kagawa, Japan

Brigitte Language
Climatology Research Group, Unit for Environmental Science and Management, North-West University, South Africa

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Joint iCACGP-IGAC Early Career Short Course 2018: ECSC participants on Shodoshima Island, Kagawa, Japan

The international Commission on Atmospheric Chemistry and Global Pollution (iCACGP) was founded in 1957 and celebrated 60 year of support for research in atmospheric chemistry which contribute to solving societal issues. The iCACGP is a sponsor of the International Global Atmospheric Chemistry (IGAC) project provides a platform that encourages the development of atmospheric chemistry research in relation to developing a sustainable world.

The Early Career Short Course (ECSC) was held on Shodoshima Island in Kagawa, Japan in the days prior to the conference (22-24 September) and brought together forty (40) promising early career atmospheric scientist from twenty-eight (28) countries. The main aim of the short course was to encourage knowledge sharing, network building, and collaboration- among those who will become future leaders in the atmospheric chemistry research community. This was done through four sessions themed around i) connecting modelling, observations and laboratory studies; ii) the future of atmospheric chemistry; and iii) science-policy engagement presented by several prominent scientist:

- Dr James Crawford, NASA, USA
- Prof Mei Zheng, Peking University, China
- Dr Christian George, Institut de recherches sur le catalyse et l'environnement de Lyon, CNRS, France
- Prof Mark Lawrence, Institute for Advance Sustainability Studies, Germany
- Dr Manish Naja, Aryabhatta Research Institute of Observational Sciences, India
- Dr Deborah Stein Sweers, Koninklijk Nederlands Meteoroligisch Instituut, Netherlands
- Dr Todd Sanford, Polygon Sun Research & Consulting, USA
- Dr Rebecca Garland, Council for Scientific and Industrial Research, South Africa

The last session included a World Café which was an open discussion on global issues connected to the topics discussed over the two day period. The course was very successful in connecting scientist from across the globe and making clear the important roles that we as early career scientists have in the future of atmospheric chemistry research in order to create a sustainable world.

The joint 14th iCACGP Quadrennial Symposium and 15th IGAC Science Conference took place in Takamatsu, Kagawa, Japan (25-29 September) following the ECSC. There were single session in atmospheric chemistry and '-people', '-fundamentals', '-ecosystems', '-climate/weather', and 'challenging the future'. These sessions were led by distinguished speakers. There were more than 500 posters presented during the conference. The ECSC was not the only opportunity for early career scientist to learn, as the conference itself hosted a talk by Prof Yaun T. Lee a Nobel Laureate in Chemistry (1986) and various soft skills talks by other experiences scientist.

Acknowledgements

We acknowledge iCACGP-IGAC and all of its sponsors for bringing together students and early career scientist to participate and contribute towards building and fostering a community of atmospheric scientist that will continue to flourish in the years to come. BL received the iCACGP-IGAC 2018 Early Career Travel Grant to attend both the 2018 iCACGP-IGAC Early Career Short Course and joint 14th iCACGP Quadrennial Symposium and 15th IGAC Science Conference.

"Being part of the world air quality reinforcement with a common goal gave me assurance that we are not fighting a losing battle and we are not alone" – Ncobile Nkosi (Unit for Environmental Science and Management, North-West University, South Africa)

"…helped me to find my interest in research." – Tanzina Akther (Department of Chemistry, University of Dhaka, Dhaka, Bangladesh)

"One of the most meteoric journey of my life, it is the platform where rising suns from different land, met, interacted, learnt and try find their role as an atmospheric scientist in the society" – Renu Sakura (Environmental Sciences & Biomedical Metrology Division, CSIR National Physical Laboratory, India)

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Research brief
Outdoor temperature and its effect on mortality in South Africa

Scovronick N, Sera F, Acquaotta F, Garzena D, Fratianni S, Wright CY and Gasparrini A
Department of Environmental Health, Emory Rollins School of Public Health, Emory University

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Researchers from the USA, UK, Italy and South Africa recently conducted the first long-term, nation-wide study of the association between outdoor temperature and mortality in South Africa. Overall, the authors identified a U-shaped relationship, indicating that the risk of mortality increases from exposure to both heat and cold. Mortality risks were highest in children and the elderly, whereas no relationship was found in people aged 25-44 years. Over the full study period (1997-2013), roughly 3.4% of all deaths in the country were attributable to non-optimum temperatures, with much of the burden occurring on days with relatively moderate outdoor temperatures.

Studies exploring temperature-related mortality have ballooned in recent years due in large part to concerns about climate change. Researchers from around the world have reported elevated mortality risks from high and low temperatures – as in this study – and have also suggested that the associated health burdens are likely to increase in the future. Nevertheless, there have been few studies of this type focusing on South or Southern Africa (or Africa in general), even though warming is expected to occur at a substantially faster rate compared to the global average. A key reason for the lack of research has been data availability, as this type of study requires information on the number of deaths per day in a given location, as well as daily temperatures.

In this study, the researchers combined data from three sources to conduct their analysis. The mortality dataset, provided by Statistics South Africa[1], included all reported deaths in the country from 1997 to 2013 for each of the 52 district municipalities, which amounted to about 8.8 million deaths overall. The temperature data was obtained from South Africa's Agricultural Research Council as well as the National Oceanographic and Atmospheric Association of the USA. The focus of the study was on daily maximum temperature, as this was considered the most reliable of the measures. After compiling the data, the researchers performed a two-step time-series analysis. First, temperature-mortality associations were estimated for each district. And second, the district-level results were pooled to identify an overall country-level relationship. Analyses were also conducted separately for different age groups and for different causes of death.

Understanding the temperature-mortality relationship may be particularly important in South Africa, as there are unique opportunities for intervention compared to many other places. The country is rapidly developing and there are explicit government programs that may have the ability to modify the risk of temperature-related mortality or morbidity, including through the provision of housing, water and electricity. There have also been steps towards climate change adaptation planning. However, it should be noted that the results of this study do not fully clarify whether South Africans can expect a worsening of the temperature-mortality relationship in the future; of the 3.4% of total mortality attributable to non-optimum temperatures, nearly 90% was from exposure to cold.

Reference

Scovronick N, Sera F, Acquaotta F, Garzena D, Fratianni S, Wright CY and Gasparrini A. The association between ambient temperature and mortality in South Africa: A time-series analysis. Environmental Research 2018;161:229-235.

[1]Statistics South Africa had no role in the design, analysis or interpretation of the study.

Research brief

Size-resolved characteristics of inorganic ionic species in atmospheric aerosols at a regional background site on the South African Highveld North-West University's research published in Journal of Atmospheric Chemistry

Andrew D. Venter[1], Pieter G. van Zyl[1], Johan P. Beukes[1], Jan-Stefan Swartz[1], Miroslav Josipovic[1], Ville Vakkari[2], Lauri Laakso[1,2] and Markku Kulmala[3]

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Atmospheric aerosols or particulate matter (PM) are important components of the atmosphere with high temporal and spatial variability, which can have significant impacts on air quality and climate change. Detailed physical and chemical characterisation are crucial in establishing the impacts of atmospheric aerosol. Aerosols consist of organic and inorganic species, and the composition and concentration of these species depends on their sources, chemical transformation and sinks. In this study an assessment of major inorganic ions determined in three aerosol particle size ranges, i.e. PM_1, $PM_{1-2.5}$ and $PM_{2.5-10}$ collected for one year at Welgegund was conducted.

Sulphate (SO_4^{2-}) and ammonium (NH_4^+) dominated the PM_1 size fraction, while SO_4^{2-} and nitrate (NO_3^-) dominated the $PM_{1-2.5}$ and $PM_{2.5-10}$ size fractions. SO_4^{2-} had the highest contribution in the two smaller size fractions, while NO_3^- had the highest contribution in the $PM_{2.5-10}$ size fraction. SO_4^{2-} and NO_3^- levels were attributed to the impacts of aged air masses passing over major anthropogenic source regions. Comparison of inorganic ion concentrations to levels thereof within the western Bushveld Igneous Complexes – a source region influencing Welgegund – indicated higher levels of most inorganic species within the source region. However, the comparative ratio of SO_4^{2-} was significantly lower due to SO_4^{2-} being formed distant from SO_2 emissions and submicron SO_4^{2-} having longer atmospheric residencies. Aerosols at Welgegund were determined to be generally acidic, which was mainly attributed to high concentrations of SO_4^{2-}.

PM_1 and $PM_{1-2.5}$ fractions revealed a seasonal pattern, with higher inorganic ion concentrations measured from May to September. Higher PM concentrations were attributed to decreased wet removal, more pronounced inversion layers trapping pollutants, and increases in household combustion and wild fires during winter. Back trajectory analysis also revealed higher concentrations of inorganic ionic species corresponding to air mass movements over significant anthropogenic activities.

Research brief

Receptor modelling and risk assessment of volatile organic compounds measured at a regional background site in South Africa
North-West University's research published in Atmospheric Environment

Kerneels Jaars[1], Mika Vestenius[2], Pieter G. van Zyl[1], Johan P. Beukes[1], Heidi Hellén[2], Ville Vakkari[2], Marcell Venter[1], Miroslav Josipovic[1] and Hannele Hakola[2]

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Volatile organic compounds (VOCs) can have significant impacts on climate and human health. In order to develop climate change reduction strategies and to assess the impacts of VOCs on human health, it is crucial to determine the sources of VOCs, which can be emitted from biogenic and anthropogenic sources. In this study source apportionment was performed using positive matrix factorisation (PMF) analysis on VOC data collected at Welgegund – a regional background station located on a commercial farm approximately 100 km west of Johannesburg that is influenced by the major sources in the interior of South Africa – for more than two years. In addition, a risk assessment study was also performed in view of the major source regions affecting Welgegund in order to quantify the impacts of anthropogenic VOCs measured at Welgegund on human health.

PMF analysis indicated ten meaningful factor solutions, with five factors being associated with biogenic emissions and five with anthropogenic sources. Three of the biogenic factors were characterised by a specific biogenic species, i.e. isoprene, limonene and 2-methyl-3-buten-2-ol (MBO), while the other two biogenic factors comprised mixtures of biogenic species with different tracer species. The temporal factor contribution for the isoprene, limonene and MBO factors correlated relatively well with the seasonal wet pattern. One anthropogenic factor was associated with emissions from the Johannesburg-Pretoria megacity, the Vaal Triangle and the Mpumalanga Highveld source regions, while another anthropogenic factor could be related to coal combustion. An anthropogenic factor was also identified that reflected the influence of solvents on atmospheric VOC concentrations, while two anthropogenic factors were determined that indicated the influence of farming activities in proximity to Welgegund.

A lifetime cancer risk (LCR) and non-cancer hazard ratio (HR) assessment study conducted for VOCs in relation to three source regions indicated that the non-cancerous influence of VOCs measured in the source regions is significantly lower compared to the cancerous influence of these species on human health, which raises concern. However, LCR values were within an acceptable range. Factor analysis also identified sources that could be targeted to minimise VOC-related LCRs and HRs, which included benzene-related cancers that can be reduced by targeting incomplete combustion sources and coal combustion.

Research brief
A review of four decades of atmospheric trace gas measurements at Cape Point, South Africa

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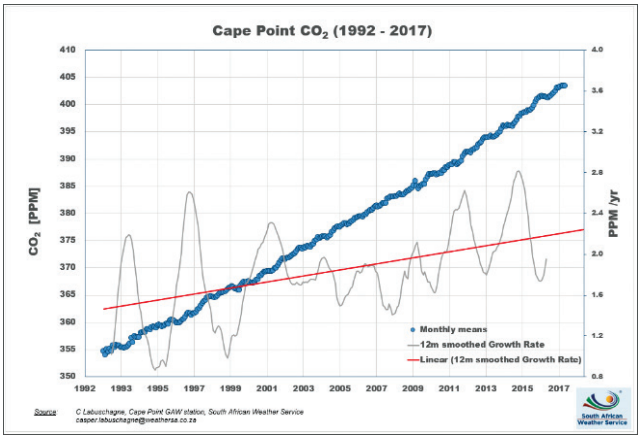
In the late 1970's the need to start monitoring the composition and changes within the clean atmosphere was raised. Since then, the Cape Point (CPT) stations' unique, strategic position has filled a crucial latitudinal gap in atmospheric trace gas monitoring in the overall sparsely sampled Southern Hemisphere (SH). Local meteorological conditions typically consists of clean marine air originating from deep within the southern Atlantic Ocean, making the Cape Point station an ideal baseline facility to monitor key indicators of changes and trends in the atmosphere of the SH. The site has two dominant, but distinctly different air-mass regimes. During austral spring to autumn (November to April), clean marine air predominates when the Cape Peninsular is buffeted by strong south easterly to south westerly winds. This is usually contrasted with winter months (June–August), when the South Atlantic High Pressure system retreats towards the equator relative to its summer position, and results in the intensification and advancement polar frontal systems (in the form of westerly winds) across the southern tip of the African sub-continent.

A recent article, published by the *Transactions of the Royal Society of South Africa*, highlighted the activities and achievements of the CPT GAW program. The measurement program, now in its 4th decade of operation, targets a wide spectrum of air chemical species, which are known to either enhance the anthropogenic greenhouse effect, e.g. CO_2, and CH_4, and various nitrogen oxides represented as NO_x, or can induce global cooling, e.g. regional aerosol measurements. Instrument calibrations are performed through analysis of gas cylinders, having known greenhouse gas mole fractions, which are obtained from the WMO GAW Central Calibration Laboratory (CCL) maintained by NOAA ESRL in Boulder, USA. After internal quality checks have been assured, the Cape Point greenhouse gas data are annually submitted to the World Data Centre for Greenhouse Gases (WDCGG, http://ds.data.jma.go.jp/gmd/wdcgg/) in Tokyo, Japan, as well as our local South African Air Quality Information System (SAAQIS: http://www.saaqis.org.za/). Recently, the archiving responsibility for reactive gases (other than CO) has been moved to the newly established GAW World Data Centre for Reactive Gases (WDCRG) hosted by the Norwegian Institute for Air Research (NILU).This enables the wider scientific community to utilize the data sets for their own research objectives.

Due to Cape Point's air-mass footprint being mostly from the pristine Southern Ocean marine environment, GAW measurements are highly valued within the United Nations



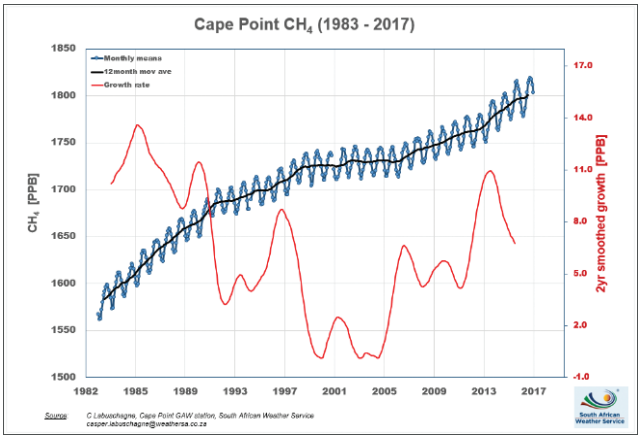


Figure 1 (left): *CO_2 time series (1993–2017) showing background monthly means and smoothed growth rate.* ***Figure 1 (right):*** *CH_4 time series (1983–2017) for background data. Red line highlights the 2-yr smoothed growth rate.*

Environment Programme (UNEP)/WMO Ozone and IPCC Climate Assessments) and serve as a bellwether for regional changes in southern African atmospheric composition. One of the aims of the Cape Point GAW station is to fulfil a mandate from the scientific and policy communities by providing regional trace gas background data for utilization in future climate change scenarios. Moreover, it also honours environmental treaties such as the United Nations Framework Convention on Climate Change (UNFCCC; http://unfccc.int/2860.php) and provides sound scientific information to policy makers as well as the Intergovernmental Panel for Climate Change (IPCC; http://www.ipcc.ch/).

Figure 1 shows the time series of the two most prominent greenhouse gases measured at Cape Point since the early 1980's (CH4) and 1990's (CO_2). When short-term variations attributed to local influences have been removed by data filtering techniques, both of these climatically relevant gases display atmospheric growth rates which have intensified substantially over the past few years and currently shows no signs of slowing down.

Figure 1 (left): CO_2 time series (1993–2016) showing background monthly means and a moving average (red line). Figure1 (Right): CH4 time series (1983–2016) for background data. Red line highlights the moving average.

For CO_2 a 5-year smoothing of the growth rate for the entire data record shows an annual fluctuation, ranging between 1.65 (for 1993) and 2.80 ppm/year^{-1} (for 2017). These estimations for CPT in concurrence with the globally observed CO_2 growth rates. Atmospheric CH_4 mixing ratios at CPT highlights the need for long-term, continuous measurements, by having had various periods of strong growth rates, which was followed by zero or negative growth rates, and then most recently again record high growth rates (2016 -2018). Interestingly, in contrast, after two decades of minimal change in CO mixing ratios measured at Cape Point, the concentration of atmospheric CO has steadily decreased since 2005. A similar decline in CO mixing ratios has been reported from a number of global atmospheric monitoring sites and also from satellite observations.

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Research brief
Radon-222 measurements at Cape Point: A characterization of a 15-year time series

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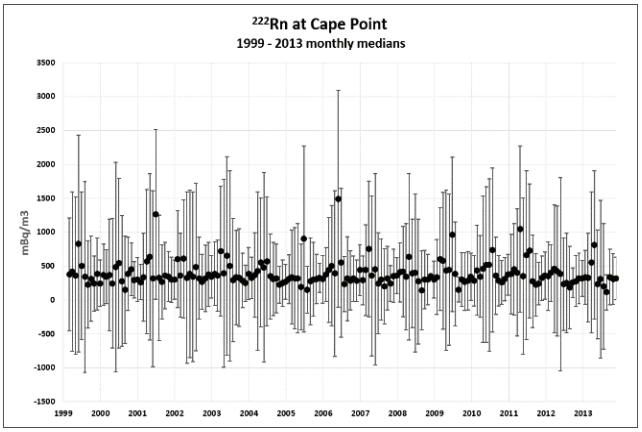
The Cape Point (CPT) Global Atmosphere Watch (GAW) research station have been monitoring climatically significant trace gases for four decades. Among these is radon, a naturally occurring noble gas with a large continental source, which has proven very useful for atmospheric tracer studies. ^{222}Rn, the radioactive decay daughter product of radon gas, forms part of the long-term exposure of radiation dosages that humans are continuously exposed to in the environment. In a first of its kind for the African continent, a radon climatology, based on a 15-year measurement record at CPT, was published in the Atmospheric Environment journal (www.elsevier.com/locate/atmosenv).

The authors found the mean continental air mass (1 004 ± 6 mBq/m^3) radon concentration to be a factor of 2–3 times higher, compared to oceanic air masses (479 ± 3 mBq/m^3), and that "deep baseline" oceanic air masses (279 ± 3 mBq/m^3) are factor of 3–4 times smaller than continental air masses. In addition, the authors described in detail an annual peak (Figure 1b) which reflects major seasonal changes in the patterns of offshore versus onshore flow - associated with regional/hemispheric circulation patterns, as well as diurnal and semi-diurnal peaks which together reflect the influence of local nocturnal radon build-up over land, and the interplay between mesoscale sea/land breezes.

A detailed percentile analysis of the entire date set was performed and several sub-sets were generated and investigated. These included seasonal subsets, diurnal subsets and fetch region (based on wind directions) subsets. An interesting feature of these analyses was the observed slow decline within the higher radon percentiles (75th and 95th) for the winter and spring seasons which was found over the 15-year data set. It was established that most of the observed changes occurred within the first 9 years (1999–2007) of observations, and particularly prominent within the north wind sectors.

The observed inter-annual decline appears to be largely associated with changes in the frequency of air masses having originated from over the African continental surfaces, and no significant trend is found in the lower radon percentiles associated with oceanic air masses. The general observed decrease of atmospheric radon - associated with continental air-masses at Cape Point could be attributed to changing meteorological conditions, possibly driven by climate change. Factors which could potentially contribute to the observed long-



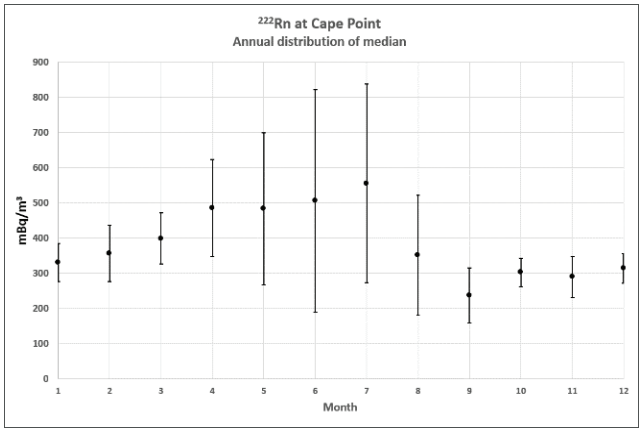


Figure 1 (left): *15-year ^{222}Rn concentration distribution and 1sigma standard deviation showing inter-annual variation.* ***Figure 1 (right):*** *15-year composite monthly median radon concentration showing the seasonal distribution of ^{222}Rn.*

term decreasing higher radon percentiles in the continental sectors (see Figs. 9 and 10 in the full paper) at the Cape Point Station include changes in (1) airflow patterns; (2) surface air temperature; (3) rainfall patterns affecting water-table depth and, consequently, soil-moisture content and therefore radon emanations; as well as (4) mixing depths and removal of radon from the boundary layer by deep convection and frontal systems. Furthermore, large-scale regional wind run decreases of more than 25% have also been observed from a 30-year study (1974–2005) at 20 climatic stations of the Western Cape.

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Commentary
Hackathons and TrainUps: Engaging New Users of Air Quality Data

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Introduction

Hackathons have become a popular approach for sourcing technology-oriented ideas to help address social issues. In 2017 there were at least 30 air quality-oriented hackathon events on different continents and in many of these participants worked with air quality data from a variety of sources. For environmental and community activists, the growing interest in air quality data and associated hackathons can have benefits; but there is also caution that deeper social, political and legal processes can be undermined by technology hype and commercial interests.

During 2018, Open Data South Africa[1] collaborated with the Department of Environmental Affairs (DEA), the Council of Land Informal Residence and Family Development South Africa (CULISA) and Geekulcha to run three hackathon (GK 2018) and TrainUp (2018) events exploring the use of air quality data for social impact. These activities introduced a large number of young graduates, ICT students and civic activists to air quality data. The team also learned a number of lessons, so it is a good time to reflect on these experiences and consider how engaging new users of air quality data can be done more effectively and with more sustainable impact.

Five days with air quality data

Open Data South Africa is a pilot project which aims to raise awareness and encourage use of open government data amongst civil society, public servants and young people across the country. Much of the work is influenced by the principles, methods, technologies and people connected to the Open Government Partnership (OGP), of which South Africa was a founding member in 2011.

In the collaboration between Open Data South Africa, DEA, Geekulcha and CULISA, three events were held between March and June 2018. During these events, participants were introduced to the new South African Air Quality Information System (SAAQIS 2018) and then given a chance to build new use cases or stories related to data on the platform. As summarised in the figure below, each of the events (H1, H2 and T1) had a slightly different format, orientation and outputs. The events and outputs have been mapped along two axes:

- Y-axis: The tone of the event was either oriented to *contesting* air quality issues with government and industry and holding them accountable for air quality problems, or it was more oriented to *collaborating* with industry and government to address these issues together.
- X-axis: The event was either led by t*echnology development/ developers*, or it was more *community* or *user-led*.

H1: #AirQualityHack Soshanguve: Over 70 individuals, mainly

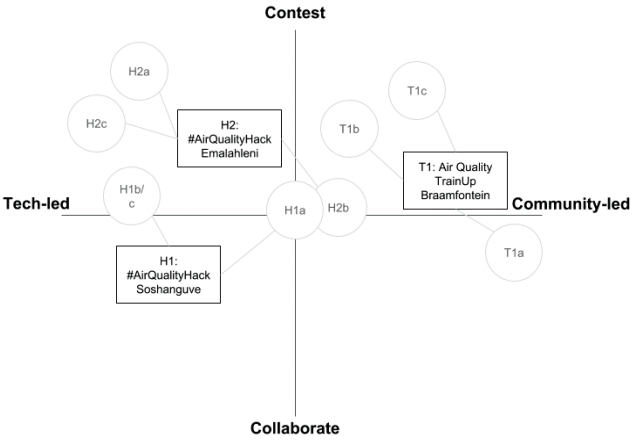

Figure 1: *Orientation of air quality data events and key outputs (the circles represent specific outputs of each event and are discussed below) (Adapted from Hirschheim and Klein 1989)*

ICT students from the Tshwane University of Technology (TUT) Computer Systems Department, participated in a 1-day hackathon in teams of 4 or 5.

The hackathon started with a short introduction to air quality management and data by the DEA, after which participants formed teams and started working on ideas for mitigating air quality issues using available data. At the end of the hackathon,

[1]Open Data South Africa is a joint initiative by South Africa's Department of Public Service and Administration, Open Up, the Centre for Public Service Innovation and The Innovation Hub Management Company.

each of the 10 teams presented their concept and 3 winning teams were selected by a panel of judges. The tone of the hackathon was oriented to collaborating or supporting DEA with their monitoring activities, and the participants were focused on building technology solutions that could increase the coverage or efficiency of DEA monitoring and alerting.

Two teams built Arduino-based sensing devices and web-dashboards with a view to filling gaps in ambient monitoring, whilst another team simulated drone-based data collection as a more cost-effective way of covering a wider area. Two teams developed apps to alert residents or DEA of unhealthy conditions. The winning concept (H1a) focused more on community participation by encouraging residents to take photos of air pollution on their smartphones and tag them with the nearest ambient air quality measurements.

T1: Air Quality TrainUp Braamfontein: Eight individuals participated in a 2-day TrainUp workshop in groups of 2 or 3. The workshop introduced participants to storytelling from a data journalism perspective, including how to develop a persona of the target audience you are looking to reach, building a story structure, working through the data pipeline (from sourcing to verification, cleaning to analysis and visualisation) and thinking about what channels are appropriate for sharing the story with your target audience.

The participants included a mix of civic tech, entrepreneur and local civil society representatives from CULISA who directed the workshop more towards empowering community members to take action and hold industry (and public officials) accountable for reducing pollution in the area. The workshop was also grounded in design thinking principles, which encouraged a more community-driven perspective.

The first concept that came from the workshop (T1a) sought to communicate air quality risks to mothers of children in a more engaging way so that they could manage exposure, and to enrol the Department of Health in prevention and treatment of air quality-related issues. The second concept attempted to assess whether ambient air quality monitoring stations were located correctly. The third concept was aimed at raising awareness in the community about industry-municipality non-compliance with SO2 thresholds, with the aim of encouraging community action on this issue.

H2: #AirQualityHack Emalahleni: Over 35 people were involved in the 2-day hackathon, with a roughly 50/50 split between a new group of TUT Computer Systems students and environmental activists linked to CULISA, and other local community organisations in Emalahleni.

As a hackathon, the tone still tended toward technology development, but the influence of local civil society attendees was significant in encouraging a more activist atmosphere, aimed at ensuring stronger industry and government accountability. Most of the concepts sought to confront industry about their pollution activities by alerting residents when air quality levels were above thresholds (from ambient monitoring or South African Atmospheric Emission Licensing and Inventory Portal (SAAELIP)/ real-time industry reporting data) or by supplementing DEA monitoring with additional community observations.

From solutions to movements

Our experience with the two air quality data hackathons and a TrainUp reflects a lot of what has been observed in other communities and sectors about technology and communities.

The involvement of CULISA and partners had an influence on the orientation of events, and the nature of this influence is partly captured by their work in another environmental training course in which:

- "The starting point for the course is people's own experience and own work [but] not something that they want to do as individuals but a concern of their communities or organisations
- [The course is] based on a working together and working away model [in which participants] come together to deepen skills and explore ways of building cases and building networks
- The course has been designed to support movement building [in which participants] work continually with their organisations and networks and present their work to these organisations and networks" (EMG 2018, pp. 8)

Much of what CULISA does is about building awareness in the community and networks of support to ensure industry accountability for pollution in Mpumalanga (Komane and Mahlangu, 2018). This thinking permeated a number of the concepts coming out of the Emalahleni hackathon and TrainUp, and suggests that open data engagement and technology ideas should look more towards support the growth of networks and movements rather than on developing specific 'solutions' to problems.

A highlight for facilitators, ICT students and community activists was the interaction between diverse groups. Their reflections included:

- Activists recognising that ICT students (who may be perceived as well-informed on most things) have limited awareness of air quality issues and data, and that they have an important role to play in any 'solution' development process. At the same time, they can find their own solutions to their own problems with a helping hand from like-minded stakeholders, including the government. However, the knowledge of the ICT students and the potential of emerging technologies for reaching a wider audience was also clear.
- ICT students seeing the importance of working with environmental and community activists to better

understand issues and processes on the ground in developing concepts that would be relevant to users.

- Programme managers and facilitators recognising that hackathons and TrainUps can benefit from a more honest, even confrontational dialogue between stakeholders. Further, that these events and the opening of data form part of an ongoing process that (1) helps participants articulate "what it means to be a resourceful and legitimate" (Björgvinsson, et al. 2010, pp. 49) resident of Emalahleni and South Africa and (2) supports the growth of technology-oriented (such as through the Geekulcha student societies, GKSS 2018) or environmental movements that can have a positive impact on social issues.
- Finally, for government departments and agencies, it is exciting to see the interest in air quality data and that engaging with young people through these solution-oriented events can generate constructive inputs (and ongoing interest) in the design of air quality information systems. The ICT students at hackathons are future partners or staff in government departments, and exposing them to practical issues during their studies can be transformative for their work over the long term. It is also important to note how students and civil society organisations are increasingly testing low-cost Arduino-enabled air quality measurement devices, and to explore how data from these citizen sensors could be referenced to DEA or municipal monitoring stations, as part of an ongoing efforts to deepen understanding and participation in air quality management.

Going forward

The hackathons and TrainUp uncovered some interesting ideas and were an important learning experience. To improve on what was done and to build longer term sustainability and impact around air quality data use, there are some smaller and larger actions needed:

- Using ambient air quality monitoring data from monitoring stations meant very little unless it could be referenced to regulated pollutant thresholds and license conditions. This means that accessing textual data in legislation or reports (such as through Open By-laws South Africa 2018) and SAAELIP is as important as accessing sensor data from SAAQIS.
- Most teams made limited use of actual data, partly because understanding and using data is a steep learning curve for a 1 or 2-day event, but also because both technical and community participants need additional training on basic data sourcing and analysis.
- Access to a suitable computer or laptop and data connection is still a major issue for ensuring effective participation by students and community members.
- Training on (data) storytelling and is just as important as learning core data skills.
- CULISA has proposed a locally-driven "Air Quality Data and Innovation Hub" that can help to coordinate ongoing development of data stories with community members and sharing of information with a wider audience. This would require some physical resources, but can be supported by a virtual network of partners, including academics and the CAJ community.

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Commentary
The need for open data on air quality monitoring in logistically difficult environments

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Background

Reports from the World Health Organization indicate that approximately seven million people died in 2012 due to the consequences of poor air quality (WHO, 2014). Air pollution is partly linked to lung and heart diseases, asthma and bronchitis. Efforts have been made in terms of policy and data availability in developed nations to help curb air inequality. Though this has not completely eradicated problems of air inequality, remarkable progress seemed to have been made to reduce air pollution in these countries. The same cannot be said for under-developed economies, especially countries in the Sub Saharan Africa, where Ghana belongs. The adverse effects of air pollution on health and general well-being in developing economies is alarming; sadly, availability of open data on air quality to enable scientists and individuals to take proactive measures to mitigate the effects of air pollution has become problematic.

The government of Ghana has enacted several policies to ensure the safety of the environment and to make quality health care a must have for every Ghanaian. Designated state institutions responsible for protecting the environment have been working to monitor air quality and to address environmental health problems, though they are under-resourced. These agencies, departments and institutions usually have a few number of air quality monitoring devices which are sparsely distributed in urban centers. A large part of the country is not monitored to assess the level of pollution in these areas. Scientists and researchers need air quality data, both historical and real time, to solve the challenges of poor air quality. Open data on air quality seem to be lacking, and access to reliable air quality data from state agencies by scientist and researchers is an arduous task as there are no clear guidelines to data access. This situation makes scientists and researchers handicapped in their quest to address the issue of poor air quality.

Air pollution and public awareness

More alarming is the fact that majority of the citizens are not aware of the dangerous effects of poor air quality, hence, they continue engaging in harmful practices like road-side cooking, burning of biomass in residential areas, use of fire wood and charcoal for cooking, and use of kerosene lanterns, which exposes the majority of the population to diseases related to air pollution. The question that begs for an answer is, will citizens engage in these practices if they were aware of the dangers associated with them?

More sensitization is needed to make citizens aware of the harmful effects of air pollution and the measures they need to adopt to mitigate it. Furthermore, if awareness is created on the importance of air quality, and air quality data is made open and easily accessible to the citizenry, there will be a positive behavioral change and they will demand policy makers to address the challenges associated with poor air quality.

Practical steps and the way forward

Commendable steps have been taken by government and other agencies to ensure the nation gets clean ambient air. Notable among these measures are the Akoben Rating to serve as an incentive for adhering to air quality standards by industries (Michaud, 2013). That notwithstanding, more needs to be done in terms of making air quality data accessible to individuals and institutions, and increasing the number of monitors to cover all parts of the country; especially areas that are more prone to air pollution (mining communities, lumbering sites). There is also the need to create awareness and educate citizens on the damaging consequences of air pollution. Beyond the provision of access to air quality data, the available data should be presented to the citizenry in a form and language they can easily understand to make them take the necessary steps to curb air pollution.

The critical issue of open AQ data in the African context

Open data on air quality requires the development of some form of infrastructure. This includes, but is not limited to, reliable internet access, real time air quality monitors integrated with general packet radio services (GPRS) for data telemetry, file transfer protocol (FTP) server, reliable power source and skilled human capital. For example, the Ghana Environmental

Protection Agency (GhEPA) uses gravimetric method for collecting data on air quality species, namely PM2.5 and PM10, which limits its ability to report the measurement data to cloud-based platforms, even though this is not impossible. The traditional air quality monitoring approaches, coupled with ad-hoc studies on air quality in Africa without uniform environmental sensing tools, are considerable obstacles in the fight against air inequality in Africa.

Relatively low-cost emerging cutting-edge high-resolution sensor networks are capable of collecting fast, reliable, real-time and in situ data on air quality (AQ) when properly operated (Brauer et al., 2012; Evans et al., 2012; Amann et al., 2013). These sensors are integrated with GPRS for data telemetry and a GPS for timestamp and location service. These low-cost sensors can bridge the gap in terms of routine air quality monitoring and facilitate the work of the responsible agencies to make air quality data readily available.

Awareness ignites actions

Air quality is treated as a very sensitive subject in Africa and most often a no-go area to talk about. Even staff from governmental agencies responsible for developing and enforcing air quality standards in many countries across Africa finds it difficult to talk about it. A personal communication with the staff of the GhEPA has shown that the misinterpretation of information from few experts in the governmental agencies by the public makes it difficult for disseminating information on air quality. An explicit example is when an officer during a presentation indicated that "most of the public vehicles on our roads pollute a lot and must be given a critical attention." This information was read by the general public as banning all public vehicles from operating in Ghana.

It is therefore critical for governments in Africa to find a way to communicate air quality data to their citizens bearing in mind everyone has the right to breathe clean air and therefore need to know what they breathe. In Macedonia for example, open data was initially introduced in 2012 through the National Action Plan for Open Government Partnership. This action plan legally binds the government to open data in a machine-readable format. This commitment led to a follow-up study on mapping of citizen interest in the type of government collected data, of which air quality data was part (OGP Macedonia, 2018). As such, the Ministry of Environment and Physical Planning (MEPP) began a project to update a dedicated website with hourly information on air quality which is auto-generated by the State Automatic System for Ambient Air Quality (Irena Bojadzievska, personal communication). The case of Macedonia is a critical example to put pressure on governmental institutions to address air pollution. Open data is key to addressing many of the challenges in the context of Ghana and wider Africa of which air pollution is critical area.

Link to the community Statement

https://medium.com/@openaq/community-statement-on-air-quality-in-ghana-time-to-take-action-21fb8fb2e769

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Commentary
The Expanded Freshwater and Terrestrial Environmental Observation Network (EFTEON)

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The Expanded Freshwater and Terrestrial Environmental Observation Network (EFTEON) is a research infrastructure that is being developed under the South African Research Infrastructure Roadmap (SARIR) program of the Department of Science and Technology. EFTEON is conceived as a modular, highly-networked, research infrastructure to support studies on coupled ecological social systems in South Africa.

The design concept is based on distributed nodes, each with responsibility for a core landscape representing an important South African Ecosystem/Human complex. The nodes are intended to include representatives of major biomes in South Africa and human transformed ecosystems such as urban areas and agricultural systems. The nodes are supported by a central co-ordination and data management facility (shared with the Shallow Marine and Costal Research infrastructure (SMCRI) and general SAEON operations). Each of the nodes is planned to have a heavily instrumented core site for fresh water and terrestrial observations and a network of instrumented subsidiary sites, to provide supporting data at a broader spatial scale.

Each of the landscape sites will have:

- A standard set of automated instruments, measuring:
  - The exchange of carbon, water and energy through the use of Eddy covariance measurements
  - The water cycle (flow and quality) at connected freshwater monitoring locations within the landscape
  - Meteorological measurements
- A suite of standard repeated manual measurements, covering:
  - Biodiversity, productivity, ecosystem condition
  - Ecosystem service provision and use, and
- A systematic collection of a comprehensive set of remotely sensed data and
- Socio-ecological data for each landscape

Potentially one of the EFTEON sites will have a “catchment-to-coast” approach and link to a coastal long term ecological research site in South Africa or one to the sites of the Shallow Marine and Coastal Research Infrastructure (SMCRI) which is also being developed under the SARIR program.

EFTEON is expected to offer a number of free and discoverable datasets and data products for the scientific community, such as:

- Long term, time series measurement data of:
  - The fluxes of energy, carbon dioxide and water,
  - Measurements of meteorology, soil moisture, soil temperature
  - River flow, daily groundwater recharge, continuous stream chemistry (Electro-conductivity, pH, dissolved N, P and O_2 monitoring)
  - Documentation of vegetation, soil and disturbance parameters.
- Landscape Scale (100km x 100km) observations of land use and land cover at 30m resolution
- Socioeconomic studies of human population, livelihoods, health and use of resources,
- Inputs and disturbances, such as deposition or fires
- Population dynamics of representative and important species in the landscape for both terrestrial and freshwater ecosystems.

EFTEON contributes to global research infrastructures by providing a terrestrial research infrastructure for socially-relevant ecosystems research based in Africa and the Southern Hemisphere, strongly linked to coastal and marine ecosystems research and global environmental data systems.

Commentary
The Rwanda Climate Observatory: Developing Climate Science in East Africa

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Kigali, Rwanda will forever be associated with climate change with the Kigali Amendment to the Montreal Protocol. This hilly East African capital city, with a population of approximately one million, was where plans to reduce and phase out hydrofluorocarbons (HFCs) emissions were negotiated. As a class of compounds, long-lived HFCs are used as refrigerants and as foam-blowing agents, among other industrial uses, and are the fourth-highest anthropogenic greenhouse gas in terms of radiative forcing (scaled by atmospheric concentration) according to the 2013 Intergovernmental Panel on Climate Change (IPCC) report.

Rwanda itself is a small country not known for its climate forcing emissions. The majority of citizens are subsistence farmers and, while economic development and industry is increasing, greenhouse gas emissions are still minimal compared to other countries in North America, Europe, and Asia. However, Rwanda is a country ripe for the impacts of climate change. IPCC reports find that Rwanda's rain and rainfall intensity will increase, and Rwanda's hilly countryside, often deforested for agriculture, is prone to landslides. Changing rain patterns will impact local farmers and crop yields, and the Rwandan government is concerned about food security, as Rwanda is the second most densely populated country in Africa.

In countries like Rwanda, adaptation to a changing climate and mitigation of climatic effects, rather than reducing greenhouse gas emissions, is the main focus of policy. To really understand local climate change and thus develop adaptation policies, however, local understanding of the changing climate and greenhouse gas concentrations is essential. While many initiatives are starting to emerge in Africa to monitor the atmosphere (e.g., Mt Kenya's Global Atmospheric Watch Station, South Africa's network of air quality monitors), time-resolved and continuous measurements are much sparser than in Europe, North America, and much of Asia. There is a gap in knowledge in this area of the world that needs to be filled.

Rwanda's government is concerned with environmental policies. They have enacted several Green Growth initiatives (http://www.greengrowthknowledge.org/country/rwanda), have banned the import of plastic bags (https://plasticoceans.org/rwanda-plastic-bag-ban/), and have a thriving conservation effort in their mountain gorilla tourism. Rwanda is also seeking to move its population from subsistence agriculture to new, higher-income generating industries, particularly the STEM sector. To do this, STEM entities need to be brought into the country. In 2009, Rwanda's president, President Paul Kagame, sought partnership with the Massachusetts Institute of Technology (MIT) for the development of STEM in Rwanda. With thoughts of both STEM and green growth, the government of Rwanda partnered with MIT to establish the Rwanda Climate Observatory (RCO).

Since 1978, MIT, under the supervision of Professor Ron Prinn, has run a global network of greenhouse gas measurements, currently known as the AGAGE (Advanced Global Atmospheric Gases Experiment) network. One of the key components of this network is the measurement of trace greenhouse gas species, such as HFCs, with a GC/MS using a customized sample collection and inlet system (known as the Medusa). This instrument delivers hourly concentration information of HFCs and other trace greenhouse gases. These data can be analyzed and ingested into models to increase understanding of sources, sinks, and long-range transport of these species. Before the establishment of the RCO, no AGAGE site existed in Africa and few HFC measurements were made.

It would be fitting for Rwanda, as the site of the ratification of the Kigali Amendment, to host the first continuous and hourly time-resolved measurements of HFCs in sub-Saharan Africa. And that day is close: a custom-built instrument has been delivered to Kigali for testing in 2018. While waiting the construction of this instrument, much other work has been done to establish the RCO. Potential sites were evaluated for their ability to capture long-range transport (through atmospheric modeling) and for their ease of access for technician visits, remoteness from urban centers, and access to infrastructure (through site visits by MIT scientists). MIT raised funds from donors to make instrument purchases, with the understanding that these would be donated to the government of Rwanda. By 2013, the initial installation of instrumentation at the RCO began on the summit of Mt Mugogo, a 2540 m peak in western Rwanda. Currently, the RCO hosts regular measurements of black carbon particles (an instrument paid for by a grant from COMESA), carbon dioxide, carbon monoxide, nitrous oxide, ozone, meteorological

parameters (such as temperature, wind speed, wind direction), and solar intensity. Nitrous oxide has been measured since 2017 and the other measurements have been ongoing since 2015. In 2017, in a partnership with Carnegie Mellon University (which has a campus in Kigali), a Real-Time Multi Pollutant (RAMP) sensor measuring typical air quality criteria pollutants was also deployed at the RCO. Carbon monoxide and black carbon data have been used to understand the impact of seasonal large-scale biomass burning versus more local pollution on Rwanda, and the carbon dioxide measurements have been used in models to understand emissions and sinks in this understudied region of the world.

Most of the AGAGE network sites are run by MIT or its AGAGE partners, the Scripps Institute of Oceanography and the University of Bristol (with other participating universities). The RCO is a different model: while MIT donors paid for initial instrument purchases, the government of Rwanda owns and manages the Observatory and the station is run by an MIT-trained PhD scientist from Rwanda, Dr. Jimmy Gasore. The day-to-day operation of the station is maintained by four station technicians, trained by MIT scientists, and a technical coordinator and financial coordinator who sit in the Rwandan Ministry of Education. A new University of Rwanda Master's Program in Climate Science, which just graduated its first class in 2018, uses the data generated from the RCO for their research projects. While still able to tap into the scientific expertise of the AGAGE community, embedding the RCO within the Rwandan government ensures the longevity of the project. Local technicians are able to answer any routine maintenance issues, local scientists are able to direct new research, and local students are able to drive the research forward. Government officials like the Director General of Science and Technology, Dr. Marie-Christine Gasingriwa, were involved in the RCO construction process and are able to advocate for the RCO within the government.

The model of providing instrumentation and training, but leaving the infrastructure and project continuation to the observatory country location, has worked well in Rwanda. From this project other associated projects have sprung up, including the establishment of a countrywide air quality monitoring network. In early 2018, the RCO was accepted as a World Meteorological Organization atmospheric watch station. This will connect the RCO, already part of the global AGAGE network, to another global network and help ensure data quality control and data decimation.

Climate change is a global problem, and one likely to impact those who have contributed least to it. These areas of the world are also rapidly developing, but often lack data to understand how their development could affect their emissions of climate forcers. Local scientists who may wish to work in their home country on climate and air quality issues may leave to find better opportunities in countries with established research centers. Beginning to establish more measurements in data poor regions of the world will increase these regions' abilities to enact and enforce emissions policy and increase accuracy and local applicability of climatic impact models by inserting real, spatially resolved data into climate models. The RCO will hopefully provide (along with Mt. Kenya) long-term data and analysis on climate change in equatorial East Africa.

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In Memoriam
Remembering the late Minister Edna Molewa

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The National Association for Clean Air (NACA) council and its members wish to remember and pay tribute to the late Minister for Environmental Affairs, Dr Edna Molewa, for her great contributions to air quality and environmental management at large.

She will be remembered for her many successful initiatives and especially for pioneering and championing environmental causes in South Africa and beyond, such as the global fight against climate change, strategic management of rhinos, stratospheric ozone protection, and recycling enterprises, amongst others.

NACA recognises her tireless efforts and contributions, especially her support in promoting clean air in South Africa.

Photo: Department of Environmental Affairs website

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Research article

Trace metal enrichment observed in soils around a coal fired power plant in South Africa

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Abstract

A site assessment was conducted at a coal fired power plant in South Africa to determine whether surrounding soils were being enriched with trace metals resulting from activities at the power plant. It was found that deposition of fly ash from the flue stacks and the ash dump along with deposition of coal dust from the coal stock yard were the activities most likely to lead to such enrichment. Eighty topsoil samples were gathered and analysed for total metal content. Results were interpreted within the context of background values. It was found that concentrations of As, Cu, Mn, Ni and Pb exceeded local screening levels, but only As and Pb could be confidently attributed to anthropogenic intervention and actual enrichment.

Keywords

Trace metals; soil pollution; coal fired power plant

Introduction

Coal fired thermal power plants are one of the largest anthropogenic sources of trace metals in the natural environment (Dragovic et al., 2013). Changes to the concentrations of heavy metals in soils is a very important indicator of contamination as soil can act as a sink for these metals (Freudenschuss et al., 2001). In this study, the nature and extent of possible enrichment of trace metals in soils that might arise from the various operations at such a power plant were investigated through an in-situ site assessment.

Due to the complexities of the operations at a coal fired power plant, including coal handling, coal combustion and subsequent disposal of ash, it is important to holistically consider such a power plant's interaction with soil within the context of these various operations. Subsequently a holistic view of soil pollution at a coal fired power plant can be formed. Due to national and international similarities in the designs of coal fired power plants, it is expected that the contamination profiles and patterns observed within this study, would to some extent be mirrored at other sites depending, however, on the composition of coal burnt and emission controls in place.

A study of fly ash from the Matla power plant, which is in the same region as the power plant being investigated here, showed that the fly ash contained toxic heavy metals such as Arsenic (As), Antimony (Sb), Cadmium (Cd), Chromium (Cr) and Lead (Pb) (Ayanda et al., 2012).

Trace metal enrichment has been observed in soils around various sites exposed to the deposition of fly ash and coal dust. A few studies have been conducted in South and southern Africa which provide some indication on what the expected impact of coal power station emissions are likely to be. A study in Bloemfontein examined the impact of a coal fired power station on heavy metal deposition and showed elevated levels of Cd, Sb, Mercury (Hg) and localised contamination of As (Clark, Tredoux and van Huyssteen, 2015). Similarly, a study at the Morupule Power station in Botswana showed increases in the concentration of Cr, Nickel (Ni), Zinc (Zn) and As in the surface soils for a distance of approximately 9 km downwind of the power plant (which at 132 MW capacity is much smaller than the standard coal power stations ~3.5 GW in the South African Highveld region) (Zhai et al., 2009). Various studies outside of the southern Africa region have assessed the impact of coal power stations on soil concentrations of heavy metals. In studies conducted by Singh et al. (1995) and Praharaj et al. (2003) on soils around coal fired power plants in India, enrichment of trace metals in soils were observed and Raja et al. (2015) found high concentrations of Cd, Pb Cr and Ni. In both studies, a correlation was observed between the most heavily contaminated soils and the mean wind vectors. In Slovakia the

levels of As in the soil in the near vicinity of the power station are raised in comparison to the surrounding environment (Keegan et al., 2006). Similarly, Dragović et al. (2013) observed an enrichment of trace metals in soils around a large coal fired power plant in Serbia. Within the Eordea Basin in Greece, it was found that the most enriched elements in the local soil were S, Chlorine (Cl), Copper (Cu), As, Selenium (Se), Bromine (Br), Cd and Pb which were attributed to power plant activities (Petaloti et al., 2006). Within the vicinity of a lignite burning power station in southern Greece AS, Molybdenum (Mo), Se, Sb, Uranium (U) and Zn were found to be enriched in the samples influenced by power station emissions (Papaefthymiou, 2008). The specific trace metals expected to be enriched in soils is dependent on the type of coal being burnt, however, enrichment of some or all of the following notable trace metals are often observed in soils around coal fired power plants: As, Cd, Cobalt (Co), Cr, Cu, Hg, Manganese (Mn), Ni, Pb and Zn (Singh et al., 1995; Praharaj et al., 2003; Dragović et al., 2013; Okedeyi et al., 2014).

South Africa's electricity sector is heavily reliant on coal, with approximately 92% of the country's electricity generation coming from coal (IEA, 2015). South African coals are typically bituminous coals that have higher ash content (i.e. non- combustible mineral content) than their counterparts in the northern hemisphere (Hancox and Götz, 2014). The results of this study will be discussed within the context of local legislative regulations pertaining to contaminated land.

Materials and Methods

The Study Area

Lands were assessed on and around a large coal fired power plant in the Mpumalanga province. The power plant has an installed capacity of 3654 MW and annually sends out approximately 19000 GWh (2013-2016 average) onto the national grid. The first of the power plant's 6 turbine units became commercially operational in 1986. Approximately 11 x 10^6 Mg of coal is burnt annually resulting in approximately 2.8 x 10^6 Mg of ash being produced. Coal gets delivered to the power plant via truck or rail and subsequently gets stored at a 28-ha coal stockyard which is 3.4 km to the north of the main station precinct. From the stockyard, coal gets transported via conveyor systems to the boilers. Most ash produced by the power plant is captured by electrostatic precipitators and subsequently disposed via overland conveyor at its ash disposal facility. The ash disposal facility is located approximately 3 km east of the main station precinct. The size of the dump, including the active deposition area and the rehabilitated area, is about 320ha. Fly ash not captured by the electrostatic precipitators is emitted from the power plant's flue stacks. The annual average particulate matter emissions (from 2013-2016) is approximately 1.5x10^3 Mg per year.

Sampling

Stratified random sampling (STSI) was used as the sampling pattern. The STSI is a systematic design-based approach whereby the area is divided into several sub-regions, or strata, after which simple random sampling is conducted within each of the strata (Brus and De Gruijter, 1997). The stratification process typically divides the site into smaller strata based on specific properties of the stratum whereby the variance of a parameter within a stratum should, if accurately applied, be smaller than the variance between strata (Edwards, 2010). Primarily STSI was employed to reduce the error associated with simple random sampling or bias introduced with other traditional sampling schemes such as following an X or W pattern (Edwards, 2010).

Prior to the sampling phase, a detailed preliminary site assessment was conducted to decide on the most efficient stratification, based on identification of activities at the coal fired power plant that were deemed most likely to lead to trace metal enrichment of adjacent soils. From the preliminary site assessment, the primary hazards and potential pathways of trace elements to soil as observed were:

- Deposition of windblown ash and coal dust and subsequent particle deposition onto adjacent land;
- Coal and ash handling processes within the main station precinct, notably the emergency ashing area and various associated activities that could lead to agitation, entrainment and subsequent deposition coal and ash dust.
- Deposition of particulate matter from the flue stack onto adjacent land – particularly farmland further to the east of the ash dump.

The study area was divided accordingly into four strata as seen in Figure 1: stratum 1 – farmland further to the east of the ash dump; stratum 2 – land directly adjacent to the ash dump; stratum 3 – land directly adjacent to the coal stock yard and stratum 4 – the main station precinct. In determining the dimensions and the location of stratum 1, deposition of particulate matter from the flue stacks was considered using air quality modelling studies which were previously conducted at the power plant and made use of the CALPUFF dispersion model. From the modelling studies, it was evident that the highest predicted ambient concentrations – for both emissions scenarios considered – were generally found to the east of the power plant's ash disposal facility. The assumption was made that these areas of maximum ground-level impact would correlate to areas of maximum deposition of pollutants. Therefore, isopleths indicating maximum ambient concentrations of particulates, as obtained from the modelling run, were used to establish the geographical area of stratum 1.

Figure 1: *Overview of division of strata within study area*

Dust fallout figures obtained from the power plant, based on monthly independent monitoring being conducted around the ash dump and coal stock yard, along with mean wind vectors, were used to determine the dimensions of strata 2 and 3. Pertaining to stratum 2, maximum dust fallout was observed

at sites located 1600m to the east and 2300m to the west of the ash dump's eastern face. Monitoring sites directly to the north and south of the ash dump experienced comparatively little dust fallout. These observations determined the horizontally elongated ellipsoid shape of stratum 2. The dimensions of stratum 3 were similarly established, though maximum dust fallout was measured at sites closer to the coal stock yard which is likely due to mean differences between coal dust and fly ash particle size distribution. Stratum 4 was established by taking into consideration the various activities being conducted in the main station precinct – particularly to the south of the unit houses – that could lead to the agitation and entrainment of particulates. These activities include the emergency ash storage area, electrostatic precipitators and dust hoppers, coal silos and ash conveyors.

Twenty samples were randomly gathered within each stratum, resulting in 80 samples across the study area. To ensure randomness, Microsoft Excel's random number generator function was used to select the intra-strata coordinates at which samples were gathered. Areas such as fence lines, roads, water channels, field edges, conveyor belts, buildings, areas directly beneath power lines, large rocks, and water bodies were avoided. If the randomly selected co-ordinates within a stratum fell upon such a location, coordinates were reselected. Samples of approximately 100g were collected in April 2017, at a sampling depth of 0-15cm. The 0-15cm sampling depth was selected as the highest concentration of contaminants were expected within the first diagnostic horizon (A-horizon) as nutrient status diminishes with depth whereby dilution with deeper submerged nutrient-poor soil could occur (Herselman et al., 2005).

To avoid cross-contamination between samples the sampling equipment was cleaned between each sampling point. Prior to the collection of a sample, the area to be sampled was cleared of surface debris such as twigs, rocks and dried leaves. To avoid sample deterioration, samples were stored in dark and cooled conditions (below 5°C, but not allowed to freeze) on site and during transportation. This was achieved using cold boxes and wet ice. For all samples, pre-cleaned glass bottles were used as sample containers. Upon completion of taking a sample, the bottles were filled to the brim and closed to allow minimum airspace.

Analytical Methods

A portion of the soil sample (>20g) was transferred to a weighing dish and the wet weight was recorded after which the sample was dried at 60°C. Subsequently the sample was ground, using a ring and puck mill to achieve homogeneity. For analysis of total metals, a 0.5 g portion of the dried, homogenized sample was digested with dilute aqua regia (2.5 ml deionised water, followed by 7mL HNO_3, 0.5ml Hydrogen peroxide and 5mL HCL) in screw capped vessels and heated to approximately 95°C using a hotblock. The sample extract was diluted to 50ml and analysed by inductively coupled plasma optical emissions spectrometry (ICP-OES). The method for digestion was based on the US.EPA method 200.2 and the US.EPA method 200.7 and APHA 3120 were used for the analysis. The same extraction procedure was followed for Hg analysis. The sample was, however, not dried due to the volatility of Hg. The analysis for Hg content was conducted by coupled plasma mass spectrometry (ICP-MS) based on US.EPA 200.8. Instrument detection limits for selected priority metals were as follows: As - 1mg/kg; Cd - 0.1mg/kg; Cr(III) - 0.2mg/kg; Co - 0.5mg/kg; Cu - 2mg/kg; Hg - 0.1µ/kg; Pb - 1mg/kg; Mn - 1mg/kg; Ni - 0.5mg/kg; Pb - 1mg/kg and Zn- 1mg/kg.

For quality assurance purposes each batch of 20 samples would contain a minimum of 1 quality control sample which must meet pre-set criteria during analysis.

Background Values and Soil Screening Values

Background values for selected trace metals were used as an indication of natural concentrations of elements that could be expected prior to contamination through anthropogenic activity. To establish a baseline for selected trace metals (Cd, Co, Cr, Cu, Pb, Ni and Zn) across South Africa, Herselman (2007) analysed soil samples that were collected during the Natural Resources Land Type mapping project (consisting of approx. 4500 samples taken across the country) that was conducted in South Africa during the mid-1970s. The Natural Resources Land Type mapping project was conducted prior to the construction of the coal fired power plant under consideration, thus making these samples ideal for determining baseline concentrations. An arithmetic average of trace metal content (Cd, Co, Cr, Cu, Ni, Pb and Zn) of three samples taken near the study area during the Natural Resources Land Type mapping project, are seen as uncontaminated background samples. For other metals of importance that were not included in the analysis of Herselman (2007), notably As and Mn, the median concentrations for rangeland in Mpumalanga, based on 514 samples, is used (Steyn and Herselman, 2006). These median concentrations for rangeland in Mpumalanga are considered acceptably representative as they are closely correlated to the arithmetic averages of the three samples taken during the Natural Resources Land Type mapping project. Background concentration as used along with their respective arithmetic standard deviations (a.s.d.) are: As-1.45mg/kg; Cu-34.01±11.94mg/kg; Co-21.16±16.80mg/kg; Cr-87.65±39.08mg/kg; Mn-538mg/kg; Ni-64.41±54.10mg/kg; Pb-11.08±4.60mg/kg; and Zn-49.74±16.67mg/kg. The background values for As and Mn were obtained from a study by Steyn and Herselman (2006) that did not state the a.s.d.

As evident from the standard deviations expressed, soil across the study area is heterogeneous and highly variable as most of the study area is underlain by dolerite and arenite (Lidwala Consulting Engineers, 2013). In instances where dolerites served as parent material for soils, an increase in background trace element concentration for most elements can be expected. In instances where arenite or shale served as parent material for residual soils, lower concentrations of trace elements are expected. Another reason for high background trace metal concentrations observed is the high incidences of clayey soils observed across the study area. This reasoning is based on the metal binding properties of clayey soils (Tack et al., 1997).

Results are discussed with reference to local legislation. The National Norms and Standards for Contaminated Land and Soil

Quality (hence called the 'Norms and Standards') (DEA, 2014), published in terms of the National Environmental Management: Waste Act (59 of 2008) provides soil screening values (SSVs) for various trace metals. The conservative SSV1 for all land use protective of water resources was used to contextualize the results. SSV1s provided are: As-5,8mg/kg; Co-300mg/kg; Cu-16mg/kg; Pb-20mg/kg; Mn-740mg/kg; Hg-0.93mg/kg; Ni-91mg/kg; Sn-150mg/kg; Zn-240mg/kg. It should be noted that the background concentrations of Cu and Ni exceed the SSV1 thresholds.

Statistical Methods

In determining whether enrichment of trace metals has occurred in soils, the mean difference (Δ) between observed and background concentrations of trace metals were determined. Secondly, hypothesis testing was conducted whereby the null hypothesis was that trace metal concentrations observed in soils were equal to or less than their respective background concentrations. Prior to conducting a hypothesis test, the data distribution had to be determined, as certain methods for hypothesis testing such as the Student's t-test assumes normal distribution. For testing the data distribution across strata, the D'Agostino test (two-sided) was used for departures from normality, as advocated by the US.EPA (2002) for sample sizes greater than 50. For intra-stratum normality, the Shapiro-Wilks test was used as proposed by the US.EPA (2002) when the sample size is less than or equal to 50. The Shapiro-Wilk test calculates a W value which is dependent on the correlation between the measured data set and their corresponding normal values. To determine whether the difference between the sample concentrations and background concentrations were consistently, and significantly, larger than the background values, the Wilcoxon Rank Sum (WRS) test was used, as proposed by the US.EPA (2002) for non-normally distributed data. The assumption was made that any differences between the site concentrations and background values are attributable to anthropogenic intervention. The WSR test is preferable for this kind of analysis as it is considered robust with respect to outliers because analysis is done in terms of ranks of the data and it does not assume that the data is normally distributed. All statistical analysis was done using the R Foundation for Statistical Computing software (R Core Team, 2013).

For display purposes, results of STSI were interpolated across the strata by way of the Inverse Distance Weighted (IDW) function on ArcGIS software (ESRI, 2011). IDW is a deterministic and nonlinear interpolation technique that uses values from surrounding measured data points to predict values in unmeasured areas. The use of IDW is motivated by its robustness and simplicity in favour of variants of kriging as discussed by Babak and Deutsch (2009). With IDW, it is assumed that a data point has a localized influence on the predicted area, and that influence diminishes with distance in accordance to Tobler's first law of geography (i.e. simply that things that are closer together are more alike than those that are further apart) (Tobler, 1970). Subsequently, greater weights are assigned to data points closer to an interpolated location, than those further away.

In terms of the power parameter - which determines the significance of measured points on interpolated values - the default value of 2 was used, therefore an inverse distance-squared relationship is established. Four pseudo points were added to the date set using the mean value for each element so that the raster would cover the entire area of interest. In terms of the search radius, twenty points were specified - which was deemed appropriate considering the relatively small surface areas of the respective strata. Furthermore, as IDW is a weighted distance average, it cannot create peaks or valleys as interpolated values cannot be lower than the lowest value in the observed data set, or higher than the highest value in the observed data set.

Results and Discussion

Numerous exceedances of the conservative SSV1 were observed for As, Cu, Mn, Ni, Pb – which was considered the first indicator of anthropogenic enrichment for these metals. Mn concentrations in soil also exceeded the more stringent SSVs for informal (740mg/kg) and standard residential (1500mg/kg) areas, which is indicative of a risk to human health. In terms of Hg, no SSV exceedances were observed. The mean difference (Δ) between the metal concentrations observed in samples and their respective background concentrations were determined. A positive difference was found for As, Mn and Pb across the study area, which is indicative of enrichment. Average measured concentrations of Co, Cu, Cr(total), Ni and Zn were, however, found to be less than their background values across the study area, as can be seen in Table 1. Additionally when considering delta values normalized against the mean background data, as in Table 2, fairly large negative values were obtained for Zi, Cr and Ni, but positive values were obtained for As - further indication of enrichment. Interestingly, a positive Δ was found for Co in stratum 1 which is indicative of enrichment specific to that stratum if not necessarily across the entire study area.

In using the D'Agostino skewness test for the 80 surface samples taken across strata during STSI, and when considering selected trace metals, it was found that As, Co, Cr(total), Cu, Mn, Ni and Zn were not normally distributed. The only metal considered to be normally distributed across strata was Pb with a skewness of 0.309 and a p-value of 0.24, signifying the H_0 that data is not significantly different from the normal could not be rejected. In terms of inter-stratum distributions the Shapiro-Wilk test for normality was used. It was found that the data was not normally distributed and therefore the non-parametric Wilcoxon Rank Sum test at 0.95 confidence level was used to determine whether the difference between the site data and the background data was in fact significant. Subsequently it was found that the positive difference between sample concentrations and background concentrations as noted for As, Mn and Pb, were in fact highly significant with p-values of 109×10^{-14}. 121×10^{-9} and 7.83×10^{-3} respectively. This is deemed to indicate enrichment of these trace metals in soils.

When considering distributions of Cu and Ni concentrations in soils across the study area, as displayed in Figures 2-3, no clear dispersion pattern that could be linked to activities at the power plant is evident. Concentrations of Cu and Ni – though deemed naturally high – do not increase in areas around the ash dump or further east into adjacent farmland. Similarly, despite a positive Δ

Table 1: *Summary of Δ across study area and within specific strata ± the a.s.d.*

	Δ Total (mg/kg)	Δ Stratum 1 (mg/kg)	Δ Stratum 2 (mg/kg)	Δ Stratum 3 (mg/kg)	Δ Stratum 4 (mg/kg)
As	2.03±1.17	1.98±1.64	2.40±1.12	1.68±0.57	2.04±1.01
Co	-1.53±9.40	4.66±12.72	-1.24±8.93	-7.02±3.36	-3.06±5.95
Cr	-49.32± 26.19	-20.11±34.57	-49.66±9.57	-67.27±5.38	-62.05±9.63
Cu	-15.03±13.28	-13.24±10.13	-15.10±14.06	-18.59±12.33	-13.52±16.13
Mn	362.59± 471.31	527.15± 578.36	347.45± 527.33	182.50± 303.18	375.25± 381.79
Ni	-42.77± 20.95	-36.71±28.07	-46.10±17.05	-49.92±9.38	-39.07±22.54
Pb	1.73±5.78	0.50±6.57	2.14±4.88	1.90±3.50	2.39±7.42
Zn	-32.20± 11.18	-33.21±6.48	-37.03±4.83	-31.65±8.96	-26.87±17.71

Table 2: *Delta values normalised against the mean background data*

	Total (z-value)	Stratum 1 (z-value)	Stratum 2 (z-value)	Stratum 3 (z-value)	Stratum 4 (z-value)
As	0.50	0.32	0.84	0.40	0.58
Co	-2.41	-1.30	-2.51	-8.39	-4.07
Cr	-5.23	-3.12	-14.35	-28.79	-15.55
Cu	-3.69	-4.67	-3.49	-4.27	-2.95
Mn	-0.37	-0.02	-0.36	-1.17	-0.43
Ni	-5.12	-3.60	-6.48	-12.18	-4.59
Pb	-1.62	-1.61	-1.83	-2.63	-1.50
Zn	-7.33	-12.80	-17.95	-9.08	-4.33

value being obtained for Mn, no identifiable distribution patterns were evident across the study area, notably adjacent to the ash dump where maximum deposition of fly ash would occur (Figure 4). Additionally, the concentrations of Mn in the power plant's ash (315mg/kg) are much lower than the mean Mn concentrations found in soils across the study area of 898.55±471.31mg/kg.

It could therefore be postulated that observed SSV exceedances of Cu, Mn and Ni concentrations in soils are in fact not anthropogenically caused but is rather a reflection of naturally high background concentrations. This postulation would however be contradictory to various studies done pertaining to trace metal concentrations in soils around coal fired power stations, both locally and internationally (Praharaj et al. 2003, Dracovic et al. 2013, Okedeyi et al., 2014). Conclusions made by such studies are, however, frequently dependant on calculation of an 'enrichment factor'. Fe is often used as a normalizer element (Neto et al., 2006; Mediolla et al., 2008; Okedey et al., 2014) though in certain instances Al, total organic compounds or fractions of grain size could also be used (Liu et al., 2003). For this study, however, a good fit normalizer element could not be found, and as such enrichment factor calculations were not used.

It is argued that the reason for this enrichment not being evident in the results of this study is twofold. Firstly, any detectable signal of enrichment – whether in terms of Δ values or in terms of visible distribution pattern in IDW maps – was impossible to detect due to the already very high background values for these elements of Cu-34.01±11.94mg/kg, Mn-538mg/kg and Ni-64.41±54.10mg/kg, whereby incremental increases in these elements could not be distinguished. Secondly, the naturally heterogeneous distribution of these elements in soils across the study area made detecting a signal for anthropogenic enrichment impossible with the data garnered from the number of samples collected.

When considering the distribution of As and Pb across the study area, as seen in Figures 5-6, it seems evident that higher concentrations of As and Pb were observed near the ash dump (stratum 2). This observation would be expected if the source of As and Pb in soils is deposition of windblown ash. With reference to As, the perceived enrichment thereof in soil around the ash dump is deemed as partial confirmation of the statement made by Kazakis et al., (2017) that As can be seen as an indicator

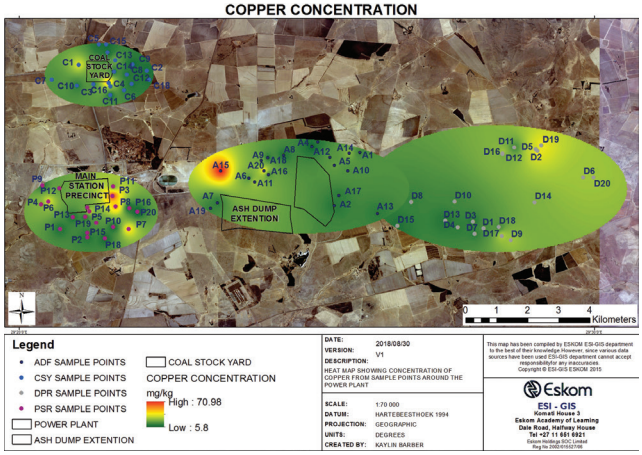

Figure 2: *Overview of copper concentrations across the study area*

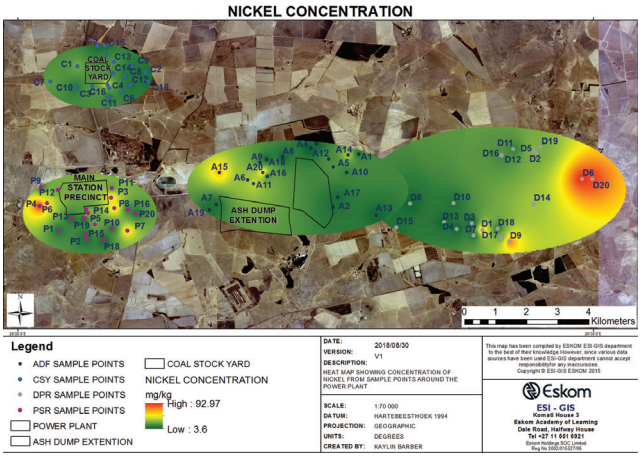

Figure 3: *Overview of nickel concentrations across the study area*

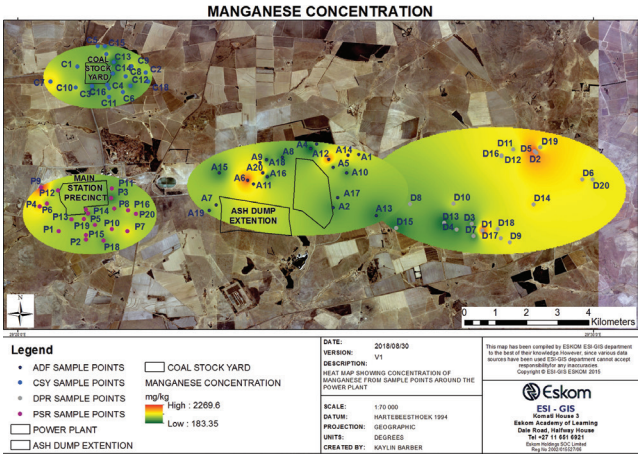

Figure 4: *Overview of manganese concentrations across the study area*

element for coal ash in soils. High concentrations of As and Pb are similarly evident towards the centre and south of stratum 1 further east of the ash dump. Based on modelling studies as discussed, it is believed that deposition from the flue stacks of the power plant would have been a contributing factor to trace metal concentrations in soils in Stratum 1. However, this relative hotspot of As and Pb concentrations may also in part be contributed to by agricultural practices. Though information about specific

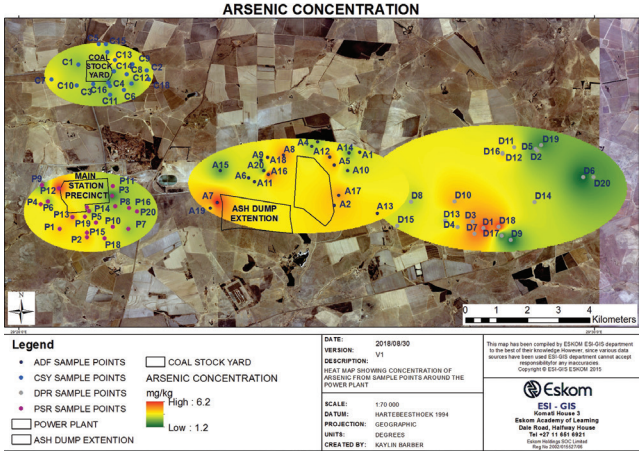

Figure 5: *Overview of arsenic concentrations across the study area*

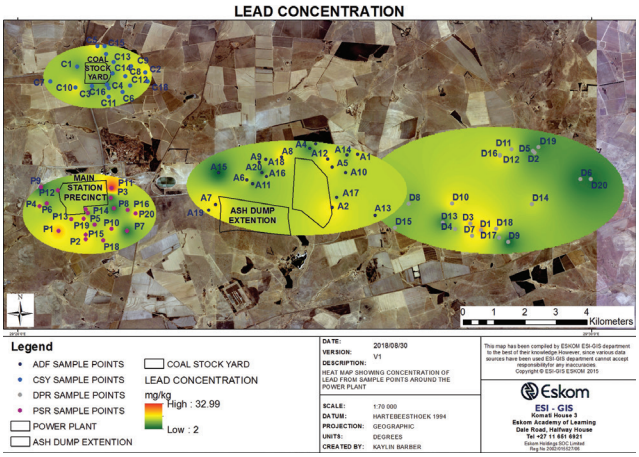

Figure 6: *Overview of lead concentrations across the study area*

agricultural practices around the study area was not available, various studies have observed trace metal enrichment in agricultural soils which could have resulted from application of various agrochemicals such as pesticides, fertilizers, herbicides, defoliants, inclusions to animal feeds and fungicides (Chen et al., 1997; Steyn and Herselman, 2006; Nziguheba and Smolders 2008). An alternative contributing factor to high trace metal concentrations observed in Stratum 1 is the underlying geology and soil type, notably instances where intrusive dolerites served as parent material and frequent clayey soils observed, whereby an increase in trace metal concentrations are expected (Herselman, 2007). The elevated concentrations of As and Pb in soils in Stratum 1 is not solely attributed to deposition from the flue stacks as its localized nature is not aligned to the disperse distribution patterns that is expected had deposition from flue stacks, or the ash dump, been the primary source of As and Pb enrichment.

It was found that As and Pb concentrations in soils are highly significant and positively correlated at 0.95 confidence level, with a correlation coefficient value of 0.65 ($p=1.15\times10^{-10}$) across the study area, based on the Pearson Correlation Coefficient. The inter-stratum correlation between Pb and As was found to be the strongest in Strata 1 and 2 with values of 0.93 ($p=4.648\times10^{-9}$) and 0.73 ($p=2.37\times10^{-4}$) respectively. In Stratum 3 a correlation

of 0.55 (p=0.018) was observed and in Stratum 4 a correlation coefficient of 0.34 (p=0.13) was observed. Therefore, across the study area Stratum 4 was the only area where As and Pb were not significantly positively correlated if taking an alpha value of 0.05 as the cut off for significance. It is deemed that these strong correlations between As and Pb is indicative of these metals partially originating from the same source. Though correlation does not necessarily imply causation, within the specific context of As and Pb concentrations in soils, it was similarly found by Chen et al. (1999) that correlation between these elements in soils may be attributed to anthropogenic intervention, specifically atmospheric deposition.

Conclusions

An integrated and holistic approach to a site assessment for contaminated land was taken at a coal fired power plant in the Mpumalanga province of South Africa. Various operations from which possible enrichment of trace metals could result were considered in designing a sampling strategy. Results were interpreted within the context of enrichment of trace metals when compared to respective background values and SSVs as provided by local environmental legislation. As far as could be determined from the literature consulted, this integrated method for a site assessment has never been conducted at the site of a coal fired power plant. Based on the results of this study, the following conclusions can be made.

Various SSV exceedances of metal concentrations in soils, within the context of local legislation, were observed across the study area, particularly As, Cu, Mn, Ni and Pb. When considering the SSV1 exceedances of Cu, Mn and Ni concentrations it is concluded that these exceedances are primarily due to high background concentrations of these elements in soils - particularly due to underlying dolerite and high clay content of soils (Herselman, 2007). It was, however, noted from various peer-reviewed sources on trace metal enrichment in soils around coal fired power plants that enrichment of Cu, Mn and Ni were to be expected. The postulated reason for this enrichment not being evident in this study is due to the already very high background values of these elements in soils which would disguise the incremental enrichment that is expected from activities at the power plant. Therefore, though some anthropogenic enrichment of these elements in soils is not discarded, it is concluded that observed SSV1 exceedances of Cu, Ni and Mn concentrations are not primarily attributable to activities at the power plant.

It is further concluded SSV exceedances of As and Pb concentrations in soils across the study area are due to anthropogenic enrichment – particularly adjacent to and towards the east of the ash dump. Lesser enrichment of As and Pb concentrations was also evident in the farmland further to the east of the ash dump and around the coal stock yard. It is therefore postulated that observed enrichment of As and Pb is primarily due to deposition of windblown fly ash, which would be aligned with a significant body of literature on the subject (Sing et al., 1995; Praharaj et al., 2003; Dragovic et al., 2013; Okedeyi et al., 2014). Within the station precinct and around the coal stock yard, deposition of coal dust and other industrial activities could also be contributing factors. In the adjacent farmland further east of the power plant, observed enrichment that is mostly attributed to ash deposition from the flue stacks and in part attributed to active agricultural practices which has been found in various studies to lead to an enrichment of trace metals in soils (Köleli, 2004; Steyn and Herselman, 2006).

It is clearly shown that the background concentrations of certain heavy metals in parts of South Africa are naturally high without the additional impact of anthropogenic inputs (Morrey et al., 1989; Herselman et al., 2005; Steyn and Herselman, 2006). Considering that some of these background concentrations are in exceedance of the legislated SSVs (DEA, 2014), determining active contamination of a site can be problematic. It is thereby suggested that further refinement of South African legislative tools is required whereby background concentrations, bioavailability of elements, a concise risk-based methodology to site assessment and a partitioning coefficient that is representative of South African soils are to be used (Eijsackers et al., 2014; Papenfus et al., 2015).

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Research article

Assessment of heavy metals and radionuclides in dust fallout in the West Rand mining area of South Africa

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Abstract

Windblown dust can contain radioactive materials from mining. These radionuclides when inhaled in dust produce ionizing radiation which damages the cells and tissues in the body. The aim of this study was to assess dust fallout radionuclides in the West Rand mining area of South Africa. Dust fallout monitoring was done using the method (ASTM) D-1739 of 1998 employing multi-directional buckets. Results show that all 9 locations investigated in the different seasons recorded dust fallout rates within stipulated residential limit according to the National Dust Control Regulations (NDCR) (Government Gazette 36974, 1 Nov 2013). Locations 3 in spring (301.93 mg/m^2/day) and 6 in winter (589.8 mg/m^2/day) recorded dust fallout rates with values above the target value of 300 mg/m^2/day according to guidelines by South African Standards (SANS 1929:2011). The ANOVA tests (p-value < 0.05) indicate that the mean dust fallout rate is significantly different across the seasons, mean dust fallout rate in summer is significantly (p-value < 0.05) more than the autumn by 103.15 mg/m^2/day. Moderate, positive correlations ($0.4 \leq r < 0.7$) exist between average dust fallout rate and wind speed. A weak, negative correlation exist between average dust fallout rate and rainfall with a value of -0.393. All the investigated elements had Enrichment Factor (EF) greater than 1. Cadmium (Cd) and lead (Pb) were extremely enriched (68.36 and 43.48, respectively), whereas Thorium (Th) (7.26) and Chromium (Cr) (9.79) had significant enrichment. Activity concentrations obtained for ^{226}Ra, ^{238}U, ^{232}Th and ^{40}K were 53.59 ± 20.45 Bq/kg, 15.20 ± 6.74 Bq/kg, 6.62 ± 2.76 Bq/kg and 278.51 ± 84.39 Bq/kg, respectively. Activity concentrations were within world averages except ^{226}Ra which exceeded the 32 Bq/kg world average. Statistically significant (p-value < 0.05), strong ($0.7 \leq r < 0.9$) and positive correlation exists between ^{226}Ra and ^{40}K. A moderate, positive correlation ($0.4 \leq r < 0.7$) exist between ^{238}U and ^{232}Th. Since EF values showed that the elements in the dust had anthropogenic influence, further studies should look at contributions of different sources to the elements found in dust.

Keywords

dust, radionuclides, enrichment factor, correlation coefficient, activity concentrations

Introduction

Mining activities pose a great threat to the environment including contamination of the environment. Some contaminants produced from mining contain toxic substances for example radioactive elements which can be trapped in dust particles (Csavina et al. 2012). These radionuclides may be carcinogenic although it is considered that there is no threshold below which adverse effects occur (Cothern 1990, Fadol et al. 2015, Taskin et al. 2009). Naturally Occurring Radioactive Materials (NORMs) are present in soils, water, rocks, food and humans are exposed to normal background levels of these NORMs which do not vary remarkably from place to place (Odumo et al. 2011, Sýkora et al. 2017). Technologically Enhanced Naturally Occurring Radioactive Materials (TENORMs) are a result of human activities (such as mining, burning of coal among others) which have increased the relative concentration of the radionuclides (Odumo et al. 2011). While radionuclides occur naturally in soils and rocks as a result of radioactive decay, most environmental releases are due to industrial processes such as mining and processes. Examples of these radionuclides include

uranium (U), potassium (K), thorium (Th) and radium (Ra). Radioactive environmental contamination related to mining is one of global concerns (Černe et al. 2011). The most affected areas will be those in close proximity to the mine tailings due to large concentrations of radioactive material in dust. Research in South Africa has shown that many residential areas near mining areas are at high risk of radioactivity contamination (Makgae 2011, Maseki 2013). Major pathways of exposure to radiation include inhalation, food contamination and occupational exposure (Mahur et al. 2008, McCartor and Becker 2013, Miah et al. 2012). For inhalation the major risks are airborne radon and windblown dust (Giannadaki et al. 2014, Makgae 2011).

Dust is composed of small, dry and solid particles released into the atmosphere by natural or anthropogenic activities and it settles slowly from the atmosphere. Anthropogenic dust is the major contributor of the total dust in the atmosphere (up to 50%) (Csavina et al. 2012, Peng et al. 2016). As observed by Langmann (2013) and Papagiannis et al. (2014) dust emissions are complex and are dependent on meteorological conditions (wind friction velocity and precipitation) and properties (particle size distribution of surface soils, surface roughness length, and soil moisture content).

To understand the potential hazardous content in ambient dust and dust fallout, these need to be monitored.

Dust fallout monitoring includes measurement of dust deposition in ambient air as part of air quality monitoring. Dust deposition data in an area shows monthly, seasonal and inter-annual variability and this can explain the pre- or operational phase of an activity (Maseki 2013, Wright et al. 2014).

Dust from mines can be blown into the communities affecting their health and this can have serious long term consequences (Abdul-Wahab and Marikar 2012, Castillo et al. 2013). Contaminants such as radionuclides from mining operations are usually associated with the finer particles (< 2µm in diameter). These particles can travel long distances and cause health problems to humans as they penetrate through the respiratory system (Csavina et al. 2012). Airborne radiometric mapping surveys over mining areas (e.g. Coetzee 1995, Sutton and Weiersbye 2008, Tutu et al. 2003) have revealed that high gamma activities from immobile daughters of the Uranium decay series such as ^{226}Ra in tailings dams can pose serious threat to the environment due to dust dispersion. There is need to monitor and quantify anthropogenic pollution resulting from dust fallout.

The enrichment factor (EF) method is one of the ways in which anthropogenic pollution on a given site is quantified. According to Chen et al. (2007), the EF is a good tool which can be used to distinguish between naturally occurring and anthropogenic sources of dust pollution. According to Papadopoulos et al. (2014), assessment of gamma radiation dose from natural sources is important since natural radiation is the largest contributor to external dose of the world population. Assessment of radiation dose from natural radiation is very important as it can help in assessing the health effects emanating from exposure to ionizing radiation.

Direct determination of ^{238}U using gamma spectrometer is very difficult since ^{238}U does not have intensive gamma lines of its own (Mehra and Singh 2011). Hence daughter products with more intensive lines when in secular equilibrium with their parents were used specifically: ^{214}Bi and ^{214}Pb.

The EF is widely used to determine the origin of pollutants in air, dust, precipitation and other environmental samples (Reimann and Caritat 2000, Sucharovà et al. 2012). The basic assumption behind the use of EF is that the ratio is approximately 1. So any change in the ratio requires an external source which is human input (Chen et al. 2007). The equation for calculating the EF is as follows:

$$EF = \frac{(X/Al)\ sample}{(X/Al)crust} \quad \text{Eqn 1}$$

where X/Al is the ratio of the element of interest (X) to aluminium (Al) (used as the reference material in this study) (Bourennane et al. 2010, Chen et al. 2007, Reimann and de Caritat 2005). The choice of aluminium is because it is not related to other metals. Usually Al has relatively high natural concentrations and hence is not expected to be substantially enriched from anthropogenic sources (Abrahim and Parker 2008). The categories for EF values are described as follows according to (Latif et al. 2015, Tholkappian et al. 2017):

EF < no enrichment
EF = 1-2 low enrichment
EF = 2-5 moderate enrichment
EF = 5-20 significant enrichment
EF = 20-40 very high enrichment
EF > 40 extremely high enrichment

Although there have been various studies on dust fall out in mining areas of South Africa (e.g. Oguntoke et al. 2013), very few studies have focussed on radionuclides in the dust from soil and mine tailings in the West Rand mining area (e.g. Maseki 2013). Hence the objectives of the study were to:

- measure dust fallout rate at various locations in the study area
- determine the long lived radionuclides (^{238}U, ^{226}Ra, ^{232}Th and ^{40}K) present in the dust and their source using enrichment factors
- assess the level of compliance to regulatory limits using National Dust Control Regulations (NDCR) of 2013.

The importance of monitoring dust fallout in an area is to check whether legislative requirements are met and to generate or maintain awareness of the dust generating activities on site. The results can be used in health risk assessments for example to evaluate population exposures. The research can provide complementary data which can be used in improving air quality management of the area.

Methodology

Description of the study area

The West Rand mining area consist of sedimentary rocks with the gold bearing conglomerates composed of 70-90% quartz (SiO_2) and 10-30% phyllosilicates. The phyllosilicates are mainly made up of sericite, $KAl_2(AlSi_3O_{10})(OH)_2$ and minor minerals such as uranite (UO_2), chromite ($FeCr_2O_4$), pyrite (FeS_2) and rutile (TiO_2) (Robb and Meyer 1995, Tutu et al. 2003).

The climate in the highveld consists of short cold winters from June to August and long hot summers usually from October to March. The rainfall occurs mostly in summer ranging from 600 - 732 mm per annum. The period October-March is often characterised by heavy storms and the average annual temperature is 16 °C (Tutu et al. 2003). The sampling locations are shown in Figure 1. The coordinates of the sampling locations shown in Appendix A were recorded with a Global Positioning System (GPS) receiver. Ethical clearance was granted by the North-West University. Permission to access the mining area and communities was sought from the mine and granted.

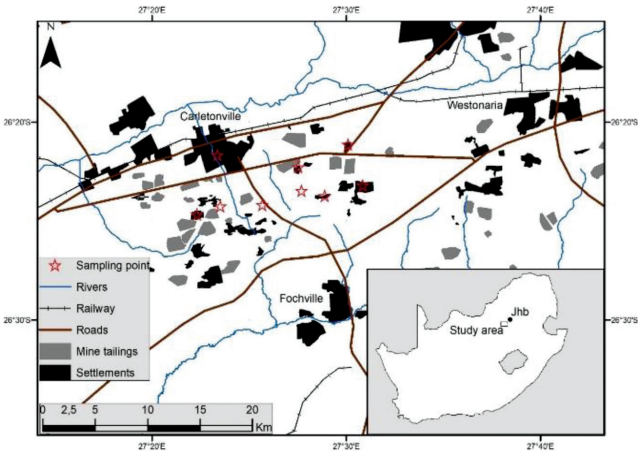

Figure 1: *Map showing the study area. The sampling points are indicated by a star. Coordinates are in Appendix A.*

Method of data collection

Dust collection was done using multidirectional dust fall buckets with wind shield, following the American Standard Test Method (ASTM) D-1739 of 1998 (2004). Four buckets were deployed to collect dust from different directions (north, south, west and east) depending on the wind direction. Samples were collected throughout the year to cater for seasonal variations from January 2016 to December 2016. The exposure time during the monitoring period complied with the standard operating procedure of 30 ± 2 days of the (NDCR) of 2013 (NEMA: AQA 2013). The calculation of the dust deposition rate was then given by the equation as cited by (Kornelius and Kwata 2014):

$$Dust\ Fallout\ rate\ D\ (mg/m^2/day) = \frac{W \times 1}{A \times days} \quad \text{Eqn 2}$$

where W = collected mass (mg); A = cross sectional area over which fall out collection has been made and has standard constant value of 0.02545 m².

An electronic mass balance RADWAG Model PS750/C/2 with precision 0.0001 g from LASEC South Africa was used for weighing the samples. For gamma spectroscopy the dust samples were left sealed for four weeks in labelled airtight vials to prevent the escape of radiogenic gases such as radon (^{222}Rn) and thoron (^{220}Rn), and to achieve secular equilibrium of the ^{238}U and ^{232}Th and their respective progenies (Frostick et al. 2011, Mahur et al. 2008). Measurements were done at the Centre for Applied Radiation, Science and Technology (CARST) at the North-West University using an HPGe well detector. The radionuclides of interest were identified at the following energies: ^{238}U (186 KeV for ^{226}Ra, 351.9 KeV for ^{214}Pb, 609.2 KeV ^{214}Bi), ^{232}Th (238.6 KeV for ^{212}Pb, 583.1 KeV for ^{208}Tl, 911 KeV for ^{228}Ac) and 1460 KeV for ^{40}K. For quality assurance, standard procedures for energy and efficiency calibration were done using LabSOCS software. GENIE 2000, Gamma Acquisition V.3.2.1 and Gamma Analysis Software V.3.2.3 were used for data acquisition and analysis.

Since gamma spectroscopy is a non-destructive technique, the samples from gamma spectroscopy were then sent for Inductively Coupled Plasma Mass Spectroscopy (ICP-MS) for elemental analysis. Microwave assisted digestion using EPA method 3052 on Anton Paar Multiwave 3000 was used in preparing the samples for ICP-MS analysis. The dust was weighed and mixed with 3 ml of 65% concentrated nitric acid (HNO_3), and 9 ml of 32% concentrated hydrochloric acid (HCl) in a reaction vessel. 2 ml of hydrogen peroxide (H_2O_2) was added slowly to the mixture. The acid volumes were measured using a graduated measuring cylinder. The Hydrochloric acid was used as a complexation reagent because of the presence of silicates and precious metals. Hydrogen peroxide was used to enhance the oxidation of nitric acid in the digestion of organics. The mixture was allowed to react for one minute prior to sealing the vessels. The vessels were placed in the rotor and then placed in the microwave. The mixture was then digested at a temperature of 120 °C for one hour and then allowed to cool down for 15 minutes. After that the digested samples were transferred to 100 ml volumetric flasks with 2% HNO_3. Deionised water was used to top up to the 100 ml mark (Nogueirol et al. 2013). The extracts were then allowed to sediment overnight and then filtered with Whatman filter paper 110 mm in diameter in readiness for ICP-MS using Total Quant method. A blank digest was prepared in the same way as the samples. Standard and blank solutions were used to correct for the analytical and instrumental drifts. The calibration was achieved using Multi-Element Internal Standard (Perkin Elmer Pure Plus) with a concentration of 10 µg/ml and the elements present were Bi, Ho, In, Li^6, Sc, Tb and Y. All chemicals and reagents used were of certified analytical grade and procured from Merck (South Africa).

Statistical analysis

SPSS Version 23 was employed for statistical analysis of the data. Two-way ANOVA was used to test for significant differences between variables since there are two treatments (location and season) and one dependent variable (dust fallout rate) (Ireland 2010). Pearson correlation coefficient tests were used to test the relationship between average dust fallout rate and meteorological parameters. To determine the correlation

between activity concentrations of the different radionuclides, the Pearson correlation coefficient test was also employed

Results

Variation in dust fallout rates across locations and seasons

Figure 2 shows that all locations (except for location 3 in spring (301.93 mg/m²/day) and location 6 in winter (589.8 mg/m²/day) were within the target of 300 mg/m²/day according to National Dust Control Regulations (NEMA: AQA 2013). Location 6 had the highest recorded value in winter of 589.75 mg/m²/day and this is the only location which had a value close to the National Dust Control Regulations of below 600 mg/m²/day for residential areas (NEMA: AQA 2013). Locations 6, 4 and 3 in winter had the highest dust fall rates compared to all other sites. However, the autumn season had the lowest dust fallout rates in all locations. Concentrations of dust at all sites per season decreased in the following order: spring > summer > winter > autumn. Spring is characterised by strong winds, whereas in autumn the wind speeds are low. The wind speeds during this period were recorded and the range was 2.9 m/s - 3.7 m/s.

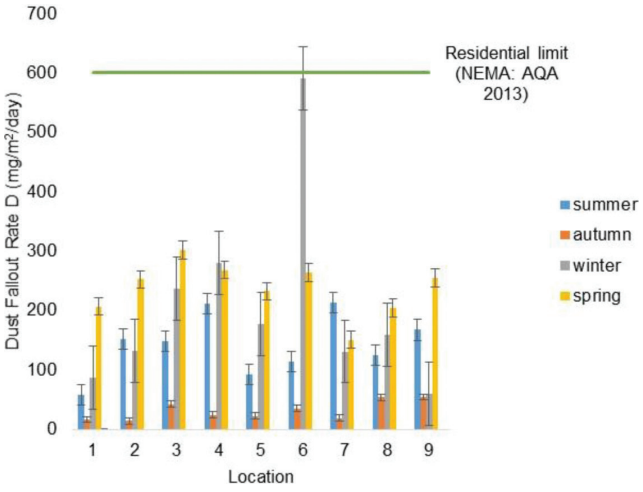

Figure 2: *Seasonal variation of average dust fallout rate per site (± standard error represented by error bars).*

Figure 3 shows the following locations: 1, 5, 6 and 8 have the average dust fallout rate below that for summer season with an average of 128.05 mg/m²/day. The average dust fallout rate for all other locations 2, 3, 4, 7 and 9 is above the average dust fallout rate for summer. In winter the average dust fallout rate was 125.86 mg/m²/day. Only locations 1 and 9 were below the average for the winter season. All other locations (2, 3, 4, 5, 6, 7 and 8) surpassed the average dust fallout rate of 125.86 mg/m²/day. The average dust fallout in autumn for locations 1, 2, 3, 4, 5, 6 and 7 is below the average dust fallout rate of 44.91 mg/m²/day for the season autumn. The average dust fallout rate for other locations 8 (53.70 mg/m²/day) and 9 (54.30 mg/m²/day) is above the average for the corresponding season (autumn) with 44.91 mg/m²/day. Only the average dust fallout for location 3 (309.10 mg/m²/day) is above the average dust fallout rate for the corresponding season (spring) (268.13 mg/m²/day).

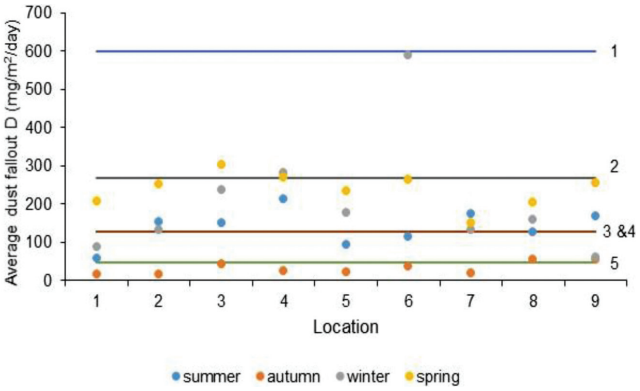

Figure 3: *Average dust fallout rates across seasons and locations. The lines are:*
1 = Residential limit
2 = Mean dust fallout rate (spring)
3 = Mean dust fallout rate (summer)
4 = Mean dust fallout rate (winter)
5 = Mean dust fallout rate (autumn)

Results for the ANOVA test (Appendix B) for significant difference across locations and seasons indicate that the mean dust fallout rate is significantly different across the seasons (p-value < 0.05). Dust fallout rate does not differ significantly across the locations. The mean dust fallout rate differs significantly in summer and autumn (p-values <0.05) as shown in Table 1.

Table 1: *Test of the difference in the mean dust fallout rate across the seasons*

Parameter Estimates				
Dependent Variable: Dust Fallout				
Parameter	B	Std. Error	t	Sig.
Intercept	235.442	27.334	8.613	.000
[Summer]	-102.013	38.656	-2.639*	.010
[Autumn]	-205.164	38.656	-5.307*	.000
[Winter]	-35.966	38.656	-.930	.355
[Spring]	0[a]	.	.	.
a. This parameter is set to zero because it is redundant.				

*significant at 5%
B represents regression coefficient (estimate)
t is the t-value

The significance and magnitude of the difference in the mean dust fallout rate between summer and autumn shows that the mean dust fallout rate in summer is significantly different to that of autumn (p-value < 0.05) and has a magnitude of 103.15 mg/m²/day.

Dust fallout rates variation with meteorological parameters

Dust fallout rate in different months of the year were plotted against meteorological parameters to show the trends exhibited as shown in Figure 4. The effect of temperature on dust fallout

rate was not apparent when judging by looking at the graph. Hence, it was necessary to carry out a statistical test to determine whether there was any association and the strength of the association between dust fallout rate and the variables.

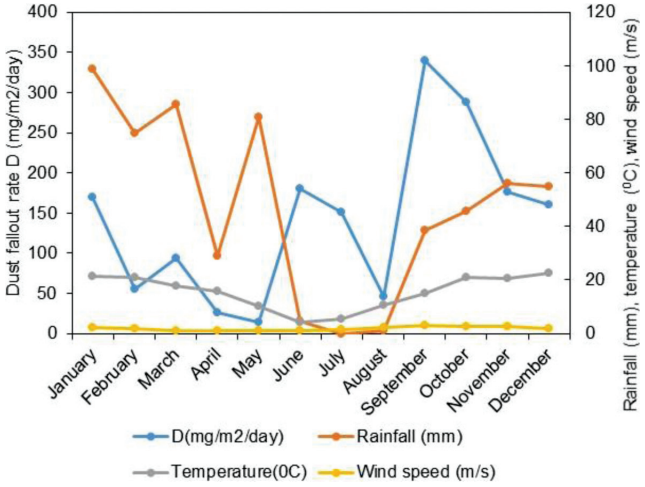

Figure 4: *Average dust fallout rate (D) variation with meteorological parameters.*

Table 2: *Test of the relationship between dust fallout rate and meteorological parameters (Pearson correlation coefficient (r)) across the locations.*

Dust fallout location	Average Dust Fallout rate D (mg/m²/day)	Average Rainfall (mm)	Average Temperature (°C)	Average Wind speed (m/s)
1	.984*	-0.263	0.192	0.916
2	.970*	-0.17	0.33	.992**
3	0.903	-0.542	-0.068	0.865
4	0.733	-0.557	-0.11	0.77
5	0.899	-0.571	-0.109	0.842
6	0.278	-.963*	-0.749	0.217
7	0.672	-0.117	0.312	0.804
8	0.929	-0.461	0.027	0.908
9	0.89	0.301	0.715	0.93

**significant at 1%, *significant at 5%

The Pearson's correlation coefficient test was done to test the relationship between dust fallout rate and meteorological parameters as well as across locations. Moderate, positive correlation with a value of 0.588 exist between average dust fallout rate and wind speed. During different seasons, the predominant winds come from different wind regimes. However, the dominant winds in the area come from the north and northwest directions as shown on the wind rose in Appendix C (two years average for 2014 - 2015) with wind speeds of 1.5 - 3.0 m/s. According to the weather data from South African Weather Service higher wind speed in the areas seldom reach > 8.0 m/s but the range 5.0 - 8.0 m/s are sometimes recorded.

There is a weak, negative correlation between average dust fallout rate and rainfall with a value of -0.393.

Table 2 shows statistically significant (p-values < 0.05) and very strong ($0.9 \leq r < 1$) (Soh 2016) positive correlation between the dust fallout rate of location 2 and average wind speed. In addition, a statistically significant (p-values < 0.05), very strong negative correlation was observed between the dust fallout rate of location 6 and average rainfall.

Concentration and Enrichment Factors (EF) of metals and radionuclides

Table 3 provides results of elemental analysis done using ICP-MS and calculated enrichment factors for selected elements. Elements associated with anthropogenic emissions include Hg, Cr, Cd and Pb. As seen in Table 3 from the heavy metals investigated Hg has the lowest concentration (0.16 ± 0.066) mg/kg followed by Cd (0.94 ± 0.38) mg/kg and Cr has the highest concentration with a mean value of 125.61 ± 7.92 mg/kg. When considering the radionuclides, Pb has the highest concentration with a mean value of 39.84 ± 3.36 mg/kg followed by Th (3.99 ± 0.75) mg/kg and U (2.94 ± 0.68) mg/kg. The world average values for U and Th in soils are 2.64 mg/kg and 11.1 mg/kg respectively (Dragović et al. 2006). World average refers to mean values obtained from other locations in different countries in the world. In this study the Th concentration was below the world average whereas the U concentration was in line with the world average. According to (Papadopoulos et al. 2014), the geochemical behaviour of the Th and U radionuclides is expected to be the same as they have similar distribution within the soils.

Table 3: *Mean concentration and Enrichment Factor (EF) of metals and radionuclides in dust.*

Element	Mean concentration ± SE (mg/kg)	Enrichment Factor
U	2.94 ± 0.68	17.85
Th	3.99 ± 0.75	7.26
Pb	39.84 ± 3.36	43.48
Hg	0.16 ± 0.066	26.27
Cd	0.94 ± 0.38	68.36
Cr	125.61 ± 7.92	9.79

SE represents standard error
NB U, Th and Pb are referred to as radionuclides in this paper whereas Hg, Cd and Cr are heavy metals

There were no elements with mean EF values less than 1. Th and Cr had the least values of EF with 7.26 and 9.79 respectively and had significant enrichment according to the classification of categories (Latif et al. 2015, Tholkappian et al. 2017). Uranium also had significant enrichment as the EF value was in the range 5-20 according to the classification. Mercury (Hg) with mean EF value of 29.22 had very high enrichment according to the

classification of the categories (Latif et al. 2015, Tholkappian et al. 2017). Cd and Pb have extremely high enrichment as the EF value is greater than 40.

Activity concentrations of radionuclides

Table 4 shows the measured mean activity concentrations of the radionuclides in the dust samples.

Table 4: *Activity concentrations of radionuclides.*

Radionuclide (Bq/kg)	Mean ± SD	Median	Min-Max
^{226}Ra	53.59 ± 20.45	51.08	23.50 - 101.20
^{238}U	15.20 ± 6.74	14.90	5.06 - 26.56
^{232}Th	6.62 ± 2.76	6.20	3.67 - 13.39
^{40}K	278.51 ± 84.39	277.90	133.60 - 486.10

SD represents standard deviation

^{232}Th had the least activity concentration and the maximum observed was for ^{40}K. The order of the activity concentration from the least to the highest was ^{232}Th < ^{238}U < ^{226}Ra < ^{40}K.

Pearson's correlation coefficient test was used to assess the relationships of the activity concentrations of the radionuclides. Table 5 shows the results obtained.

Table 5: *Pearson's correlation matrix of activity concentrations of radionuclides.*

	^{226}Ra	^{238}U	^{232}Th	^{40}K
^{226}Ra	1			
^{238}U	0.53	1		
^{232}Th	-0.012	.566*	1	
^{40}K	.730**	0.229	-0.383	1

**significant at 1%
*significant at 5%

There is statistically significant (p-value < 0.05) and strong ($0.7 \leq r < 0.9$) (Soh and Soh 2016) positive correlation between Ra-226 and K-40. A moderate, positive correlation ($0.4 \leq r < 0.7$) (Soh and Soh 2016) exist between ^{238}U and ^{232}Th due to similar distribution of Th and U within the soils and this could be attributed to their similar geochemical characteristics. According to (Papadopoulos et al. 2014), U and Th are primarily associated with heavy minerals such as the monazite, zircon and allanite and secondarily associated with epidote, titanite and apatite. On the other hand ^{40}K is negatively correlated with ^{232}Th showing that different mechanisms control the ^{40}K and ^{232}Th concentrations in soil (Papadopoulos et al. 2014).

Discussion

Seasonal variation of dust fallout rates and relationship with meteorological parameters

All 9 locations in summer, autumn, winter and spring did not exceed the permissible residential limit of 600 mg/m^2/day according to National Dust Control Regulations (Figure 2). This is a good trend considering that there are residential areas near the sampling locations where the dust was collected. Hence the nuisance impact significance is low. It is also important to note that the study area is situated in an area which is just a few hundred metres away from where mining is the major activity. Mining releases dust to the surrounding environment and if no proper control methods are in place, the dust can exceed limits as per National Dust Control Regulations. In all locations as depicted on Figure 3, dust deposition rates were below 600 mg/m^2/day which is the National Dust Control Regulations limit for the 30 day dust fallout. Compared to other countries a research conducted in Jamaica and Namibia (Kgabi et al. 2015) showed that both the residential and non-residential limits of dust rates of 1200 mg/m^2/day set by National Dust Control Regulations were exceeded by Kingston site in Jamaica (1557 mg/m^2/day). Whereas the Windhoek site in Namibia exceeded the residential limit only with an average of 1034 mg/m^2/day.

During spring in 2016 there were high amounts of dust. High dust fall out rates in spring was due to lack of precipitation during this period and possibly higher wind speeds. According to Van Zyl et al (2014), lower wind speeds are in the months of May to August (winter) whereas in summer more unstable tropospheric conditions occur (Van Zyl et al. 2014). The month of August has strong prevailing winds in the north-westerly direction and this could have contributed to the high dust fall out rates during winter in the study area. Dominant winds were from the north and northwest directions in the study area though during the different seasons the predominant winds were coming from different wind directions.

Wind speed is an important factor as it influences the dispersion of pollution from a source. Precipitation reduces wind erosion by increasing moisture content of materials. This is an important factor for removal of pollutants and is regarded as very important in air pollution studies. The observed trends of lower dust fallout rates during autumn can be related to above average precipitation which was received in the area.

Ambient air quality is influenced by regional atmospheric movements, local climatic and meteorological conditions. The descriptive statistics show that there is some variation in the dust fallout rates across the locations in the study area and the seasons. The results presented in this study show that dust deposition rates vary with seasonal fluctuations in wind speed patterns (Appendix D).

According to Wright et al. (2014) in summer dust is less due to rain and in winter it is more because of the wind and dry

conditions. Favourable conditions such as high wind speed and lower humidity are conducive to the spread of pollutants.

Concentration and Enrichment Factors (EF) of metals and radionuclides

Results from ICP-MS analysis showed that the uranium concentration in this study (2.94 ± 0.68 mg/kg) was similar to the values obtained in Serbia Montenegro (mean 2.76 mg/kg) and world average of 2.64 mg/kg. However, the mean Th concentration in this study was approximately 2.7 times less than the world average of 11.1 mg/kg. In Serbia Montenegro the highest concentrations of U, Th and K were found in soil samples that originated from sedimentary rocks and magmatic rocks with silica oversaturation (Dragović et al. 2006). Hence the differences observed in this study to those in other areas may be probably due to differences in geological origin. Also, in our study, this might be an indication that Th is not easily transported via dust.

According to Pekey (2006) As, Cr, Cu, Mn, Ni and Fe are used to show existence of metal industries whereas Cd and Pb are markers of the paint industry. In Table 3 the Cd concentrations are low 0.94 ± 0.38 mg/kg compared to Cr (125.61 ± 7.92) mg/kg. It is possible that these metals could be coming from industries nearby as shown on the map. For Cd, sources can be both natural and anthropogenic hence it was necessary to go further with the study to determine sources of the elements as either being natural or anthropogenic. That is the reason for using EF values in this study.

The trend of radionuclides in decreasing order of concentrations is Pb > Th > U (Table 3). This could be explained in several ways. The Pb in dust could be coming from a mixture of natural and anthropogenic sources. Secondly, Pb can also come from sintering of ore containing ^{238}U and the burning of coal (Bollhöfer et al. 2006). Considering that the dust samples were collected in a geographical area where mining is dominant, it is probable that some source of the Pb is coming from the industrial activities (mining and coal burning) although we lack precise information on the exact percentage of the contribution from the different sources. Pb pollution from mining activities is a relatively localized problem (Naja and Volesky 2009) hence in this case our assumption that Pb is from mining can be true. A certain contribution of Pb can also be coming from vehicular emissions. According to Wuana and Okieimen (2011), the most hazardous source of exposure to soil Pb is through ingestion of contaminated soil or dust. However very few plants if any can absorb or accumulate lead (Pb) thus leaving exposure through dust inhalation as the most serious exposure route (Wuana and Okieimen 2011).

The trend shown in Table 3 shows that of all the six elements investigated there is none with EF value less than 1. Accordingly, all the elements presented in Table 3 have EF greater than 1 and are influenced by anthropogenic emissions (Latif et al. 2015, Tholkappian et al. 2017) though the degree of contributions to the different metals differ. The order of the mean EF values of the elements in the study area from the highest to lowest is Cd > Pb > Hg > U > Cr > Th. According to Reimann and Caritat (2000), elements such as Hg and Pb always give high enrichment values for example from 100-1000 regardless of where the samples have been collected in the world. This is explained by a large and chemically similar anthropogenic contaminatio of the atmosphere by the highly volatile elements which top the EFs values on a worldwide scale. A certain contribution of Pb can also be coming from vehicular emissions, yet lead (Pb) in petrol was phased out in South Africa in January 2006. Thus, sources of Pb can be multiple in anthropogenic nature apart from the natural source thus confirming the high EF values. Another process besides pollution must also be responsible for the extremely high EFs for Cd and Pb. According to Reimann and de Caritat (2005) the elements easily form organo-metallic compounds thus formation of methyl metals might be the process enriching these elements. However, some authors have argued that the EFs can exaggerate some results (Reimann and Caritat 2000). The reason being that crustal values are based on total element concentrations determined by x-ray fluorescence, neutron activation analysis, multi-acid dissolution and many other methods but results presented in environmental researches are obtained by weaker extractions methods. These include aqua regia method whereby element extraction differs from element to element because different elements bind differently to the mineral component (Reimann and Caritat 2000). Historically Pb was used as an additive to gasoline therefore released in huge quantities over large areas by vehicles.

Radionuclide activity concentrations and correlation coefficients

The order of the activity concentration from the least to the highest was $^{232}Th < ^{238}U < ^{226}Ra < ^{40}K$. In literature granite rocks are known to display high levels of total potassium (Júnior et al. 2010). The parent rock has some granite characteristics thus the corresponding increase in ^{40}K concentrations.

As reported by UNSCEAR Radiation (2000), ^{238}U, ^{232}Th and ^{40}K are within the world averages of 35, 45 and 420 Bq/kg for soil respectively whereas ^{226}Ra exceeds the 32 Bq/kg world average by approximately 1.67 times orders of magnitude. The activity concentration of ^{238}U is within the range of values obtained from other studies carried out previously (Radiation 2000, Taskin et al. 2009) and is about 2.3 times orders of magnitude below the world averages. The activity concentrations for radionuclides obtained in this study are much lower than activities obtained from other countries such as India and Greece (Chakrabarty et al. 2009, Papadopoulos et al. 2014). In the study area in Table 5, ^{238}U with ^{232}Th and ^{226}Ra with ^{40}K are positively correlated indicating the radionuclides may have originated from the same source and their concentrations are affected by the same geological processes (Arafat et al. 2017). However, the radionuclides ^{238}U with ^{226}Ra show no significant correlation. This is contradictory to what is expected because ^{226}Ra and ^{238}U belong to the same decay chain. According to Navas et al. (2002) low correlations between members in the uranium decay series can be due to differences in mobility and transfer of the radionuclides.

Conclusion and Recommendations

Concentrations of dust at all sites per season decreased in the following order: spring > summer > winter > autumn. Highest dust fallout rate was recorded in location 6 during winter season whereas the lowest dust flux was in autumn in location 2. Winter should have lower deposition rates because lowest wind speeds are between May and end of August, this is when more stable tropospheric conditions occur (caused by a persistent high pressure system over South Africa). On the other hand, the month of August has strong prevailing winds in the north-westerly direction and possibly this could have contributed to the high dust fallout rates during winter in the study area.

The fallout rate for all the locations throughout all seasons were below National Dust Control Regulations for residential and light commercial areas (NEMA: AQA 2013). All locations except for location 3 in spring and location 6 in winter were within the target of 300 mg/m^2/day as specified by the South African guidelines (SANS 1929:2011).

The mean dust fallout rate is significantly different across the seasons. Lower dust loadings were observed in the rainy season and during autumn and this was in agreement with similar studies in literature. The higher dust fallout rates in winter and spring can be attributed to higher wind speeds.

The activity concentrations of ^{226}Ra, ^{238}U, ^{232}Th and ^{40}K were similar to those obtained in various studies in the world. However, the ^{226}Ra activity concentration exceeded the world average value hence caution needs to be taken as long term exposure to the dust containing the radionuclides may pose a health risk due to ionizing radiation. In our study we can conclude that all the elements investigated were influenced by anthropogenic sources. However, further study should look at contributions of different sources to the elements found in dust in the study area.

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Appendix A

Sampling locations in the study area.

Location	GPS coordinates	
	Latitude	Longitude
1	26°23'43.7''S	27°28'53.8''E
2	26°23'12.1''S	27°30'50.9''E
3	26°22'15.6''S	27°27'32.1''E
4	26°24'39.9''S	27°22'19.2''E
5	26°21'8''S	27°30'5.7''E
6	26°23'28''S	27°27'41.9''E
7	26°24'15''S	27°23'30.9''E
8	26°21'39.4''S	27°23'21.6''E
9	26°24'11.1''S	27°25'40.9''E

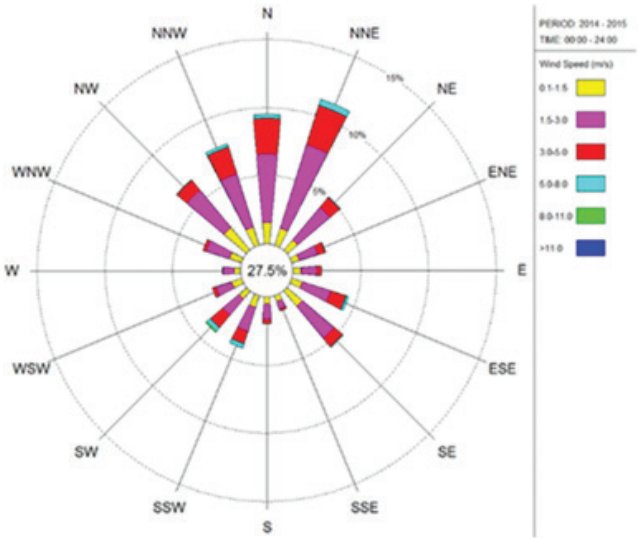
Appendix B

Test of the difference in the mean dust fallout rate across the locations and seasons

Tests of Between-Subjects Effects					
Dependent Variable: Dust Fallout rate					
Source	Type III Sum of Squares	df	Mean Square	F	Sig.
Corrected Model	960316.420	35	27437.612	2.092	.013
Intercept	1747367.601	1	1747367.601	133.245	.000
Location	162802.460	8	20350.307	1.552	.171
Season	466466.135	3	155488.712	11.857*	.000
Location * Season	334756.517	24	13948.188	1.064	.421
Error	524556.529	40	13113.913		
Total	3187036.348	76			
Corrected Total	1484872.949	75			

Appendix C

Wind pattern year average for the study area for the period 2014 – 2015 (South African Weather Services)



***significant at 1%*
**significant at 5%*

Appendix D

[1]Description of the data

Descriptive statistics for Dust Fall out rates in the study area

Descriptive Statistics				
Dependent Variable: Dust Fallout rate				
Location	Season	Mean (mg/m²/day)	Std. Deviation	N
1.00	Summer	58.0173	31.74256	3
	Autumn	16.2757	5.50055	3
	Winter	85.9660	5.68822	3
	Spring	206.7357	103.35538	3
	Total	91.7487	87.26832	12
2.00	Summer	151.0475	115.62822	2
	Autumn	13.7215	3.04692	2
	Winter	132.3870	19.49918	2
	Spring	252.2325	10.68792	2
	Total	137.3471	100.89748	8
3.00	Summer	148.7460	13.62171	2
	Autumn	42.1775	16.52862	2
	Winter	236.5915	12.34113	2
	Spring	301.9315	86.17722	2
	Total	182.3616	109.59434	8
4.00	Summer	211.5455	19.02754	2
	Autumn	23.8870	10.76641	2
	Winter	280.6070	127.65540	2
	Spring	267.7635	9.00642	2
	Total	195.9508	120.23830	8
5.00	Summer	92.0900	13.40250	2
	Autumn	22.0930	3.22865	2
	Winter	176.7940	22.95551	2
	Spring	232.1055	148.29797	2
	Total	130.7706	102.88492	8
6.00	Summer	113.4345	75.83791	2
	Autumn	34.9565	16.42397	2
	Winter	589.7510	606.15739	2
	Spring	263.4745	142.69486	2
	Total	250.4041	328.37712	8
7.00	Summer	172.6025	57.28201	2
	Autumn	18.3685	7.03783	2
	Winter	130.8805	13.29431	2
	Spring	150.7040	40.17074	2
	Total	118.1389	69.08348	8
8.00	Summer	123.9900	84.39319	2
	Autumn	53.7005	11.48412	2
	Winter	158.6290	21.46493	2
	Spring	204.0435	101.11132	2
	Total	135.0908	77.50631	8
9.00	Summer	167.0875	108.30542	2
	Autumn	54.3220	47.37191	2
	Winter	60.4285	73.23434	2
	Spring	254.3385	58.04569	2
	Total	134.0441	105.14051	8
Total	Summer	133.4284	68.34669	19
	Autumn	30.2779	20.24956	19
	Winter	199.4755	212.32794	19
	Spring	235.4418	81.36169	19
	Total	149.6559	140.70645	76

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Research article

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

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Abstract

The Greater Tubatse Municipality in Limpopo is home to three ferrochrome smelters and over fifteen operational mines which are mining chromium, platinum or silica. Source apportionment in this study was performed by combining air mass back trajectories and receptor modelling. The particulate matter (PM) samples at six sites were collected using the University of North Carolina Passive Samplers. The monthly samples were collected for a period of 4-5 weeks, except for August-September and September-October 2015 where the samples were collected for up to 6 weeks. The sampling was carried out from July 2015 to June 2016. PM chemical analysis was performed using Computer Controlled Scanning Electron Microscopy coupled with Energy-Dispersive X-ray Spectroscopy (CCSEM-EDS). The PM chemical analysis indicated the presence of elements such as carbon (C), calcium (Ca), chromium (Cr), iron (Fe), aluminium (Al), silicon (Si), magnesium (Mg) and lead (Pb). All the six sites except site 1 exceeded the WHO annual guidelines for PM_{10} concentration of 20 µg/m^3. The annual chromium concentrations exceeded the New Zealand limits of 0.0001 µg/m^3 and 0.11µg/m^3 Cr (VI) and Cr (III), respectively. The back trajectory clusters computed by the HYSPLIT model identified 5 transport clusters for each site. The main transport patterns were northerly to north-easterly, easterly to south-easterly, and south-westerly to north-westerly. The US EPA PMF model version 5.0 used in source profiling and source apportionment identified agriculture/wood combustion, coal combustion, crustal/road dust, ferrochrome smelters, and vehicle emissions as the main sources in the area. The source contributions varied across all sites indicating the existence of different microenvironments within the airshed and that the pollution can originate from either local or regional sources as indicated by back trajectory clusters.

Keywords

source apportionment, back trajectories, particulate matter, chemical characterization

Introduction

South Africa is one of the developing countries in the world, and as such the country has considered economic growth, social and educational development and industrialization as key development priorities. In the Greater-Tubatse Municipality (GTM) in Limpopo Province, South Africa, mining is viewed as one of the important economic activities which has the potential of contributing to the development of the area's economy (Community Empowerment Impact Assessment Report, 2007). Though the contribution of mining activities to economic development of GTM is well acknowledged, this might be achieved at a significant environmental, health and social costs to the region due to urbanization, high traffic volumes and higher industrial and domestic waste production.

Environmental pollution due to heavy metals from mining activities, vehicular emissions, agricultural and biomass burning are a major concern in many parts of the world (UNEP, 2006). Extensive mining of chromite, platinum and silica in the GTM mining belt may pose a serious threat to the environment. Mining of these ores may release toxic metals such as hexavalent chromium and platinum group metals which are carcinogenic and mutagenic to human health (Zayed and Terry, 2003; Ravindra et al., 2001). Studies worldwide have found that ambient levels of PM_{10} are associated with adverse health effects including increase in premature deaths, hospital admissions and emergency attendances for respiratory and cardiovascular disease and exacerbation of asthma (Pope III, 2000; Dockery, 2001). $PM_{2.5}$ which is smaller in aerodynamic diameter than PM_{10},

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can penetrate deeper into the lungs and reach the blood system and it can also impact on visibility and climate change (Chen et al., 2014) The potential of adverse health effects of particulate pollution has triggered extensive research on PM chemical composition and source apportionment in many countries over the past decade (Liu et al., 2018). Understanding the chemical components of PM is crucial to adequately assess the impacts of these chemicals on the receiving environment. A number of ferrochrome smelters are found in the GTM. Ferrochrome is a major chromium source used as raw material for the production of stainless steel and ferro-metal alloys. The morphology of the chromite spinel ore can be described stoichiometrically as (Fe, Mg)(Cr, Al, Fe)$_2$O$_4$ and they often contain gangue compounds of SiO_2 and MgO. Submerged-arc furnaces are commonly used to smelt chromite ores by using carbonaceous reductants such as coke, bituminous coal and char. The ferrochrome slag created during the smelting process mainly consists of SiO_2, Al_2O_3 and MgO in different proportions but also smaller amounts of CaO, chromium and iron oxides (Nkohla, 2006; Hockaday and Bisaka, 2010). These elements may find their way into the atmosphere during the smelting operations and they can cause serious harm to human health.

In South Africa, the National Environmental Management: Air Quality Act (AQA, Act No. 39 of 2004) was promulgated in 2005 as an approach to manage air quality in the country. The Act requires national, provincial and local authorities to identify substances or mixtures of substances in ambient air which may reasonably be anticipated to endanger public health, and to establish air quality standards to limit emission of such substances. However, to date no ambient metal standards (with the exception of Pb) have been promulgated to define the level of air quality that is necessary to protect the public welfare from known or anticipated adverse effects of these pollutants on the receiving environment in South Africa. There is also little information on PM source apportionment studies to assist in developing effective policies to mitigate the impacts of PM in South Africa.

Numerous methods have been developed over the years to collect and analyze air pollutant samples, using both active and passive techniques. Easy to use passive samplers are available in the market and are less expensive than conventional samplers. Therefore, a large number of passive samplers can be deployed at a given time to capture representative spatial measurements in an airshed (Lagudu et al., 2011).

There are two types of models (source-oriented models and receptor models) used to identify sources of pollution in the environment (Schauer et al., 1996). Source-oriented (dispersion) models require knowledge of all emissions from the contributing sources (Pant and Harrison, 2012). Receptor modes are statistical analysis tools used to identify contributions from different sources using multivariate measurements from different receptor locations. These models use ambient data and the chemical components in source emissions to quantify contributions, unlike the source models that use emissions and meteorological parameters to estimate concentrations at the receiving environment (Watson, 2002). Positive Matrix Factorization (PMF) is one of the recent models developed by the United State (U.S) Environmental Protection Agency (EPA). PMF has been used widely in source apportionment of ambient PM because of its ability to account for the uncertainty variables that are often associated with sample measurements and also the output values in the solution profiles and contributions are nonnegative (Reff and Eberly, 2007). The key output of PMF is the percentage contributions of different sources to ambient pollutant concentration at specific receptors (Pant and Harrison, 2012). The PMF model has been used by many researchers across the world for source identification and profiling. A study by Harris and Davidson (2005) characterized Ca, Al, Mg, Si, K, Fe, Mn and Zn as elements emitted from metal smelters. Soil was found to contain elements such as Fe, Al, K, Ca, Ti in a study by Watson and Chow (2001a; 2001b), while road dust contributed to Fe, Al, K, Ca, Si, Mg, P, S, Na and BC (Bhave et al., 2001; Ho et al., 2003). However, these profiles can vary from one area to the other due to varying soil types. A source profiling study by Kim et al., (2003b) and Begum et al., (2004) identified motor vehicle emissions as the source of Mg, Al, Fe, Si, S and BC (black carbon). Biomass burning emissions are a mixture of both organic carbon and elemental carbon and their ratio depends on the type of fuel used (Karanasiou et al., 2015).

The quantitative assessment of sources contributing to the ambient PM on the receiving environment is required to develop sound and effective control strategies to address the scourge of PM pollution in South Africa. However, the bottom-up approach based on emission inventories is hindered by poor availability of the emissions data particularly from industries. The objective of this study is to identify sources of PM in a mountainous terrain in Limpopo, South Africa and estimate their contributions through receptor modeling (PMF) application. The sampling data obtained during the sampling campaign from July 2015 to June 2016 will be used.

Methods

Study area

Ambient PM sampling was undertaken in a rural area of the Greater Tubatse Municipality (GTM) in Limpopo Province, South Africa (Fig. A1). The main towns in the area are Steelpoort and Burgersfort which are sustained through economic activities such as mining and smelting of chromium ores. Furthermore there are agricultural and forestry activities and transportation that also add to the economic activities in the area. Most of the households in the area are dependent on wood burning for space heating and cooking (Community Empowerment Impact Assessment Report, 2007). The GTM has a complex terrain with high mountains and steep inclinations. The elevation of the surface area is approximately 740 m above sea level with the surrounding mountains extending to a height of approximately 1200-1900 m above sea level. The area is located in the subtropical climate zone where the maximum average

temperature reaches 35 °C with minimum average temperature of 18 °C in summer. In winter the maximum average temperature reaches 22 °C with average minimum of 4 °C (Schulze, 1986). The annual rainfall for the area ranges between 500 and 600mm (DWAF, 2005). Figure A1 in appendix should the map of the study area showing passive sampler locations (red place-marks) with smelter locations shown as green place-marks.

Site selection

The location of the monitoring sites was selected to optimize spatial sampling for exposure assessment. A sequential sampling technique (Goovaerts, 1997; Van Groenigen et al., 1997) was used to design an optimal sampling network of 6 sites in the GTM. This technique is based on extended knowledge of the area to be sampled and factors controlling the distribution of pollutants. These factors amongst others were terrain and various phenomena like meteorological conditions and the chemistry of pollutants (Frączek et al, 2009). The sites were located at private residences, a church, a hospital and a school for security reasons and easy access during site visits.

Sampling and sample analysis

The University of North Carolina (UNC) passive samplers designed by Wagner and Leith (2001) and housed in a protective shelter designed by Ott and Peters (2008) were deployed at six sites for PM sampling. Ott and Peters (2008) designed the shelter to shield the passive sampler from precipitation and to minimize the influence of wind speed on particle deposition (Sawvel, 2015). The samplers consist of a scanning electron microscopy (SEM) stub, a collection substrate, and a protective mesh cap (Lagudu et al., 2011). The samplers were deployed for sequential periods of approximately 30 days from July 2015 to June 2016, except for the month of August and September when they were deployed for a period of approximately 40 days. The longer sampling periods of 3-4 weeks were selected to ensure that there was sufficient particle loading on the samplers as suggested by Sawvel (2015).

The $PM_{2.5}$, PM_{10} concentrations and the elemental composition of individual particles deposited on the passive sampler were determined by CCSEM-EDS (Tescan Vega 3 model). Before sample analysis with Personal Scanning Electron Microscopy (PSEM) (method by Hopke and Casuccio, 1991), the samples were coated with a thin layer of graphitic carbon under vacuum to bleed off the charges induced by the electron beam in the SEM. The PSEM was operated with a 20-kV beam (Sawvel 2015). Lagudu et al., (2011) gave a brief description of the CCSEM analysis. The elements obtained from the analysis were Carbon (C), Chromium (Cr), Calcium (Ca), Iron (Fe), Silicon (Si), Aluminium (Al), Magnesium (Mg), Lead (Pb), and other miscellaneous elements. Pb and miscellaneous elements were not included in further analysis due to their insignificant weight. The concentrations of $PM_{2.5}$, PM_{10} and each particle was determined using the method outlined in Ott and Peters (2008). Data obtained from CCSEM is semi-quantitative, and therefore, in order to use the data in PMF for source contributions the particles needs to be classified into homogenous groups by applying cluster analysis (Song and Hopke, 1996b; Kim and Hopke, 2008; Lagudu et al., 2011).

Cluster analysis

Buhot et al., (1999) described cluster analysis in detail. In this study, hierarchical cluster analysis was performed on single particle data from CCSEM using the open source clustering software (Cluster 3) developed by the Institute of Medical Science (IDS) at the University of Tokyo. Once the analyses were completed, all the cluster groups with less than 4 (excluding Pb and miscellaneous elements) particles were chosen as potential homogenous classes. This is because as the number of classes created for each source sample increases, the number of particles assigned to each class decreases (Kim and Hopke, 1988). The particle classes obtained from cluster analysis include Carbon-rich (C-rich), Chromium-rich (Cr-rich), Iron-rich (Fe-rich), Iron/Chromium-rich (FeCr-rich), Silicon (Si-rich), Silicon/Aluminium/Iron-rich (SiAlFe-rich), Calcium-rich (Ca-rich), Silicon/Magnesium-rich (SiMg-rich) and Silicon/Aluminium-rich (SiAl-rich).

Positive matrix factorization

PMF 5.0 (USEPA, 2014) was used to apportion the contribution from emission sources (Lee et al., 1999; Reff et al., 2007). The guidelines specified in the user manual were closely followed in this study. Two input files are required by the model: the sample species concentration values and the sample species uncertainty values or parameters for calculating uncertainty. In this study, the sample species uncertainties were obtained during CCSEM analysis of samples.

The receptor modelling in principle relies on the observed concentrations of chemical species in the atmosphere and these species must be conserved during transport between the source and the receptor. The conserved mass is then used during the analysis in the identification and apportionment of these species (Pant and Harrison, 2012). The PMF model identifies the sources by applying the following mass balance equation:

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

Where x_{ij} is the concentration of the j^{th} species in the i^{th} sample, g_{ik} the contribution of k^{th} source to the i^{th} sample, f^{kj} the concentration of the j^{th} species in the k^{th} source, and e_{ij} is the difference between the measured and fitted value. If the number and sources in the area being modelled are known, (f_{kj}), then the mass contribution of each source to each sample, g_{ik}, in equation (1) is known (European Commission, 2014). However, the objective is to calculate values of g_{ik}, f_{kj}, and p that can reproduce x_{ij}. An adjustment is then made to g_{ik} and $f_{k}j$ until the minimum value of Q for a given *p* is found. Q is defined as:

Where σ_{ij} is the uncertainty of the j^{th} species concentration in sample I, *n* is the number of samples, and *m* is the number

$$Q = \sum_{i=1}^{n} \sum_{j=1}^{m} \left(\frac{e_{ij}}{\sigma_{ij}}\right) 2 \quad (2)$$

of species (Reff et al., 2007). In most cases, a given chemical

constituent will have multiple sources and the program performs correlation analysis to generate chemical profiles of 'factors' characteristic of the sources. Past knowledge of source chemical profiles is then used to assign factors to sources (Pant and Harrison, 2012). However, in South Africa there are few source chemical profiles available. Therefore, the international source chemical profiles (Central Pollution Control Board Parivesh Bhawan, East Arjun Nagar Delhi; Pant and Harrison, 2012; Barrera et al., 2012) were used as a guide.

HYSPLIT model

Backward air trajectories arriving at the six sampling sites were calculated using the Windows PC version of the Hybrid Single Particle Integrated Trajectory (HYSPLIT-4) model. This model is a system for computing air mass trajectories and complex dispersion and deposition simulations (Draxler and Hess, 1997; 1998). Meteorological data fields to run the model are available from routine archives. In this study, 24-hour back trajectories were calculated at a height 500 meters above ground level using reanalysis data from National Centre for Environmental Prediction (NCEP) and National Centre for Atmospheric Research (NCAR) available from the National Oceanic and Atmospheric Administration's (NOAA) Air Resources Laboratory (ARL) archives. The reanalysis data covers the globe from 1948 to the present with a horizontal resolution of about 2.5 x 2.5 degrees latitude-longitude and with an output every 6- hours.

Table 1: *Model parameters used for all runs.*

Model parameter	Setting
Meteorological dataset	NCEP/NCAR Reanalysis, 2.5 degree latitude-longitude
Trajectory direction	Backward
Trajectory duration	24hr
Site 1	(-24.825222, 30.093418)
Site 2	(-24.753453, 30.153452)
Site 3	(-24.726582, 30.205004)
Site 4	(-24.614130, 30.172281)
Site 5	(-24.541220, 30.149056)
Site 6	(-24.497841, 30.064537)
Start time	00:00 UTC
Start height1	500m AGL

The isosigma vertical motion method was selected for computing trajectories. This method follows the internal terrain following coordinates systems. A review on computation and applications of trajectories was provided by Stohl (1998). The length of the back trajectories is restricted in many ways by the distances between source regions and the destination zone. The choice of 24-hour back trajectory duration is a compromise between the objective to identify local and distant sources and sink regions and to limit the uncertainties in the trajectories. Stohl (1998) affirmed that errors of 20% of the distance travelled seem to be typical for trajectories computed from analysed wind fields.

Trajectory cluster analysis

In this study trajectory cluster analysis was performed using the HYSPLIT_4 model. The model uses Ward's method described by Romesburg (1984), Moody and Galloway (1988) and Stunder (1996). The HYSPLIT clustering method is described in detail by Draxler et al., (2018). Five clusters were chosen for each set of trajectory end point files because this number was sufficient to identify all major flow patterns, as well as several less common but nonetheless important patterns. Once the number of clusters has been decided, 'Special Runs' was chosen as the technique to produce standard clusters for all trajectory end points for the period starting from July 2015 to June 2016. A practical advantage of 'Special Runs' it's its ability to accommodate large volumes of data. The variables clustered were the latitude and longitude of trajectory segment endpoints at 1-hour intervals along each 24-hour back trajectory. Individual trajectories were further averaged to produce "cluster-mean" trajectories. Thus, our large data base (annual trajectories) was reduced to a number of cluster-mean plots that can be interpreted in terms of known mesoscale and synoptic features. For each cluster, ensemble plots of all individual trajectories belonging to that cluster were produced. These ensembles or "cluster membership" plots were used to validate the mean and to assess the variability within the cluster (HYSPLIT4, 2018).

Results and discussion

The mean annual concentrations and the range between minimum and maximum concentrations for $PM_{2.5}$, PM_{10}, and particle classes obtained through cluster analysis are shown in Table 2. The $PM_{2.5}$ and PM_{10} annual mean concentrations were below the South Africa National Ambient Air Quality Standard (NAAQS) of 20 µg/m^3 and 40 µg/m^3, respectively. However, all the sites exceeded the WHO annual guideline of 20 µg/m^3 for PM_{10} and only site 1 exceeded the WHO annual guideline of 10 µg/m^3 for $PM_{2.5}$ (WHO, 2017). All the sites with the exception of site 4 and 6 recorded highest maximum PM_{10} concentration above 50 µg/m^3 with the minimum concentration of 10.3 µg/m^3 recorded at site 5. For chromium, all sites exceeded the New Zealand annual chromium limits (Ministry for the Environment, 2009) of 0.0011 µg/m^3 and 0.11 µg/m^3 for Cr (VI) and Cr (III), respectively. This is an indication that the area may experience negative impact on human health, and because the pollutants' toxicity differs so should be the environmental regulation strategies to mitigate their impacts. The existence of SiAl-rich particles with concentrations ranging between 3.3 µg/m^3 and 42.9 µg/m^3 in the study area may result in cardio metabolic effects on animals and humans (Sun et al., 2012) because they contribute 15%-55% of the total PM in the study area.

Table 2: *Species mean concentrations and range.*

	Mean						Range					
Species	Site1	Site2	Site3	Site4	Site5	Site6	Site1	Site2	Site3	Site4	Site5	Site6
PM_{10}	32.02	31.28	38.11	24.10	24.65	20.98	14.9-71.2	16.3-56.6	18.2-53.9	12.2-44.7	10.3-64.9	12.5-33.5
$PM_{2.5}$	11.8	4.7	4.8	3.0	3.2	2.5	2.3-65.6	2.1-8.3	2.4-9.4	1.8-3.8	1.2-7.5	1.4-3.2
Ca-Rich	1.9	3.1	2.7	1.7	1.8	3.4	0.4-3.5	0.9-6.3	0.8-4.4	0.7-5.6	0.3-9.9	0.9-6.3
C-Rich	5.4	3.2	2.6	2.5	2.1	1.9	2-16.1	2-4.6	1.6-4.8	1.6-4.5	1.3-2.9	0.9-6.3
Cr-Rich	1.3	1.0	1.4	0.2	1.3	0.2	0.8-1.9	0.3-2.5	0.4-3.4	0-0.7	0-6.9	0-1.2
Fe-Rich	0.8	0.4	0.6	0.3	0.2	0.5	0.1-1.1	0.2-1.1	0.4-1.2	0.1-1.4	0.1-0.5	0.1-0.6
FeCr-Rich	1.7	1.5	2.8	0.5	2.8	0.4	0-3.4	0.7-2.8	0.7-6.7	0.1-1	0.2-16.7	0-1.9
Si-Rich	1.5	1.8	2.6	1.3	1.0	0.9	0.8-2.5	1-3.1	1.1-4.1	0.5-3	0.4-2.6	0.2-1.5
SiAl-Rich	11.6	11.4	11.8	7.8	7.3	8.2	3.3-42.9	5-23	4.9-16.9	3.3-17.1	2.3-17.9	4-16.2
SiAlFe-Rich	3.8	5.1	7.0	6.7	4.6	3.2	1.3-7.8	2.7-10.2	3-12.6	3.1-13.8	2.4-7.5	1.2-5.4
SiMg-Rich	1.0	1.3	3.9	1.0	1.7	0.8	0.4-2.1	0.5-2.7	0.6-8.8	0.4-1.7	0.5-6.8	0.4-1.8

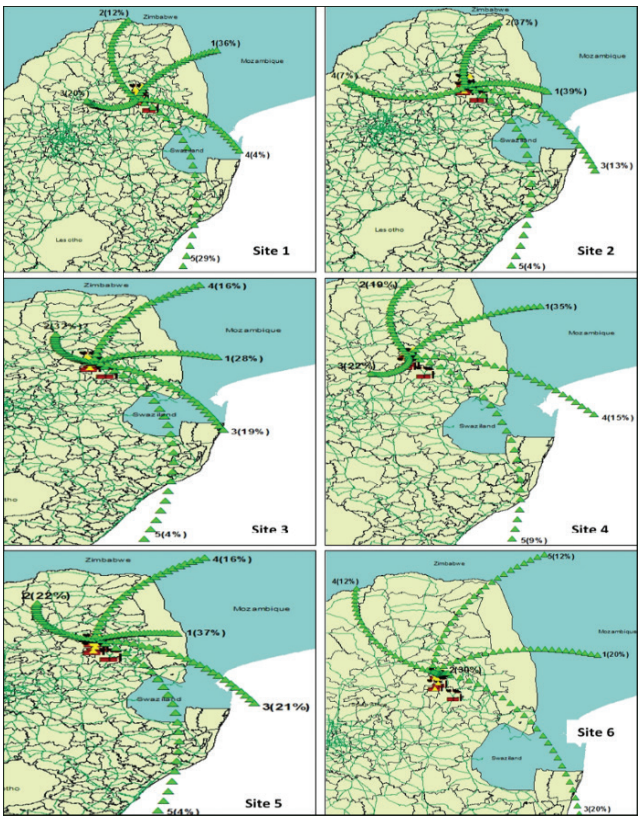

Figure 1: *Five transport pathways (clusters) arriving at the six sampling sites.*

HYSPLIT transport clusters

Figure 1 shows five clusters per receptor site. Clusters for site 1 to site 4 have similar pattern with varying trajectory percentage. For site 1, cluster 1 with 36% of trajectories originates from Mozambique and arrives at the receptor site by passing through mining areas and a smelter northeast of the sampling site. Cluster 2 with 12% of the trajectories originates from Zimbabwe and arrives at the receptor site from the northwest and away from the mines and smelters in the area. Cluster 3 with 20% of trajectories originates from the west while clusters 4 and 5 originate from the Indian Ocean. Only cluster 1, 4 and 5 pass through areas with mines and smelters within the study area. This makes them potential transporters of heavy metals to the receptor site. For site 2, all clusters with the exception of cluster 4 passes over the mines and smelters and as such have the potential of carrying elemental particles to the site. For site 3, all the clusters pass over the mining areas and only cluster 3 and 5 passes over the smelters. Hence all these clusters can transport elemental particles to the receptor. For site 4, all the clusters pass over mining areas and only cluster 2 and 3 pass over the smelters before arriving at the receptor site. For site 5, only cluster 1 is not passing over any industrial facility in the area. For site 6, only cluster 2, 3 and 5 pass over industrial facilities. Cluster 2 with 30% of trajectories is recirculating within the vicinity of the receptor site and this makes it the most likely contributor of local pollutants. However, it should be noted that even though some trajectories are not passing through the industrial facilities, they can also transport pollutants from a variety of sources such as transport, agriculture, domestic, crustal and oceans, and that these pollutants also contribute to the observed concentrations as shown in Table 2.

Source profiling by PMF

The most important step in PMF is to determine the number of factors which correspond to potential particle sources. After applying bootstrap and displacement tests on the data sets, three to five factors were deemed to be appropriate for a five source solution. The profile graph (Fig. A2.1-A2.6) displays the mass of each species apportioned to the factor (blue bar) and the percentage of each species apportioned to the factor (red bar). The factors were identified according to the type of elements dominating in percentage in that factor. Agricultural/ wood combustion was associated with the dominance of C as

Table 3: *Source contributions to the PM particles.*

		Vehicular emissions	Ferrochrome smeltering	Coal combustion	Agricultural/Wood combustion	Crustal/Road dust
Site 1	C-rich		35%		63%	2%
	Ca-rich			25%	44%	30%
	SiAl-rich		43%	21%	36%	
	Si-rich		21%	19%	26%	35%
	SiAlFe-rich		49%	32%		19%
	SiMg-rich		10%	61%	26%	3%
	Cr-rich		28%	13%	26%	33%
	Fe-rich		7%	40%	35%	19%
	CrFe-rich		2%	28%	36%	34%
Site 2	C-rich	45%	0%	18%	6%	31%
	Ca-rich	4%	3%	35%	50%	9%
	SiAl-rich	13%	10%	29%	33%	15%
	Si-rich	0%	28%	7%	15%	51%
	SiAlFe-rich	19%	25%	13%	23%	22%
	SiMg-rich		17%	11%	50%	22%
	Cr-rich	19%	55%	26%		
	Fe-rich		7%	38%	23%	32%
	CrFe-rich	34%	26%	30%	8%	2%
Site 3	C-rich	52%		45%	3%	
	Ca-rich	6%	21%	60%		14%
	SiAl-rich	18%	14%	26%	7%	36%
	Si-rich	13%	10%	20%	15%	42%
	SiAlFe-rich	36%		2%		62%
	SiMg-rich		1%	44%	41%	14%
	Cr-rich	1%	52%	31%	7%	9%
	Fe-rich	15%	6%	39%	6%	34%
	CrFe-rich	12%	40%	36%	12%	0%
Site 4	C-rich	55%	10%	6%	23%	7%
	Ca-rich	5%	7%	9%	45%	34%
	SiAl-rich		10%	28%	42%	21%
	Si-rich		6%	15%	23%	55%
	SiAlFe-rich	17%	3%	24%	16%	41%
	SiMg-rich	19%	28%	12%	38%	3%
	Cr-rich	7%	62%			31%
	Fe-rich	21%	27%	1%	24%	28%
	CrFe-rich	12%	35%	5%	13%	35%
Site 5	C-rich	17%	4%	13%	66%	
	Ca-rich	14%	10%	76%		
	SiAl-rich	15%	22%	30%	15%	19%
	Si-rich	23%	1%	14%	2%	61%
	SiAlFe-rich	19%	5%	15%	21%	41%
	SiMg-rich	3%	44%	13%	10%	30%
	Cr-rich	20%	65%	2%	6%	7%
	Fe-rich	12%	8%	23%	2%	55%
	CrFe-rich	2%	75%	4%	7%	13%
Site 6	C-rich	21%		4%	62%	13%
	Ca-rich	12%		8%	23%	57%
	SiAl-rich	16%	9%	8%	23%	44%
	Si-rich	25%	22%	23%	8%	22%
	SiAlFe-rich	9%	23%	22%	10%	37%
	SiMg-rich	32%	22%	5%	39%	3%
	Cr-rich		64%		36%	
	Fe-rich	45%	30%	3%	23%	
	CrFe-rich	20%	55%	11%		15%
Average contributions		18.61%	23.52%	22.00%	24.72%	26.33%

a primary species and Fe elements. Ferrochrome smelter was identified by the domination of Cr and Fe elements. Crustal/road dust factor was associated with Si, Al, Ca and Mg as the primary elemental species. Industrial coal combustion factor was identified by Ca, C, Al and Si as the dominating elements. Vehicular emission factor was associated with Fe as the primary species, C, Al, Si, and Ca. The most interesting outcome is that Cr was also associated with crustal/road dust and agricultural/wood burning which is an indication that once this element is emitted from the source it is then deposited on to the soil and water bodies before it is absorbed by plant material and released into the atmosphere during burning. Figures A2.1-2.6 in the appendix show the factors fingerprints for the species.

Source contributions to PM

PMF analysis determines the number of factors which correspond to potential particle sources. Source categories that potentially have contributed to ambient PM in the GTM rural area were identified as crustal/road dust, coal combustion fly ash, vehicle exhaust dust, agricultural/wood burning and industrial sources (Table 3). The average PM source contributions across all sites indicate that geological material (crustal/road dust) accounts for 26.33%. The crustal material could be mainly from resuspended dust from mine tailing in the area or transported from regional sources as indicated by the HYSPLIT transport pathways. Vehicular emissions are associated with tail pipe, emissions, tire and brake wear, road surface abrasion, wear and tear of other vehicle components such as the clutch, and resuspension of road surface dusts and accounted to 18.69% of the PM sources. Fly ash from industrial coal combustion contributed 22% to the PM in the study area. Most coal ash contain aluminium oxide (Al_2O_3), calcium oxide (CaO) and silicon dioxide (SiO_2) (Coal Ash, 2016). And regardless of the by-product produced, there are many toxic substances that are present in coal ash (such as arsenic, chromium, lead, mercury and uranium) that can cause major health problems in humans (Lockwood and Evans, 2016).

Agricultural and wood (cooking and space heating) burning accounts to 24.7% of the total PM emissions. The three ferrochrome smelters contributed 23.52% of the total PM in the airshed. The distribution of Cr elements across all source types indicate that ferrochrome smelters have a on the ambient air and have the potential to pollute the water bodies, which can impact on the human health since some of the communities in the area depend on water from the river streams for cooking and bathing.

Both PMF results and HYSPLIT clusters can be combined to make informed decisions on the sources and origin of PM sources. Crustal/road and soil dust can be transported by trajectories from both local and regional anthropogenic sources as shown in Fig. 1. However, it should be noted with caution that the trajectories are not accurately terrain following. Therefore, high-ending trajectories were chosen to represent more accurate boundary layer flow above the local terrain.

Emission reduction strategies

Source apportionment results can be useful for the review or development and implementation of strategies to reduce the impacts of pollution due to industrialization. Some of the immediate interventions to be considered in GTM are;

- Address fugitive emissions from industrial activities.
- Review of the separation process of silica and chrome from milling process.
- Review of the handling and transportation of milled chrome by road (as some of this material is spilled on the road).
- Effective enforcement of the Air Quality Act by authorities.
- Reduction of fugitive emissions from smelters during material handling.
- Electrification of households to reduce wood burning.
- Reduction of dust from mine tailings and haul roads.
- Re-look at the transportation of ground milled chromium to address the current scourge of spillages on the roads.
- Initiate waste collection from rural households to prevent waste burning.

These interventions can also be copied to other areas in South Africa.

Conclusion

The study investigated a selection of the chemical components of PM collected over a period of a year in the Greater Tubatse Municipality in Limpopo province of South Africa. The chemical components were used in PMF analysis for source profiling and source identification. HYSPLIT model was used in the identification of possible pollution source locations using backward clusters. The combination of the two models assisted in concluding that the sources can be either of local or regional origin.

These results show that the composition and levels of PM in GTM varied significantly among the six sites, however, systematic sampling and characterization of PM is needed in order to increase the sampling numbers and improve PMF results.

The PMF analysis identified a mixture of tracer elements as markers for source identification. However, differentiating these markers for different sources in the area has proved to be very difficult. This is because some of the chemical components that are solely industrial can find their way into other source categories such soil and road dust through deposition, and wood and agricultural plantation through absorption from the soil. The average source contributions were dominated by crustal/road dust, industrial activities such as ferrochrome smelters, and wood burning for space heating, with industrial coal burning, agricultural activities and vehicle emissions being other sources identified in the area. The contribution of this sources varied across the study area with chromium pollution being concentrated closer to the sources. These results show that the composition and levels of PM in GTM varied significantly

among the six sites indicating the existence of varying microenvironments within the airshed, however, systematic sampling and characterization of PM is needed in order to increase the sampling numbers and improve PMF results.

This study can serve as a base for siting of AQ monitoring stations to ensure harmonized AQ assessments throughout the GTM. It can also be used as a guiding document for strengthening intervention strategies in the industrialized areas across South Africa.

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Appendix A

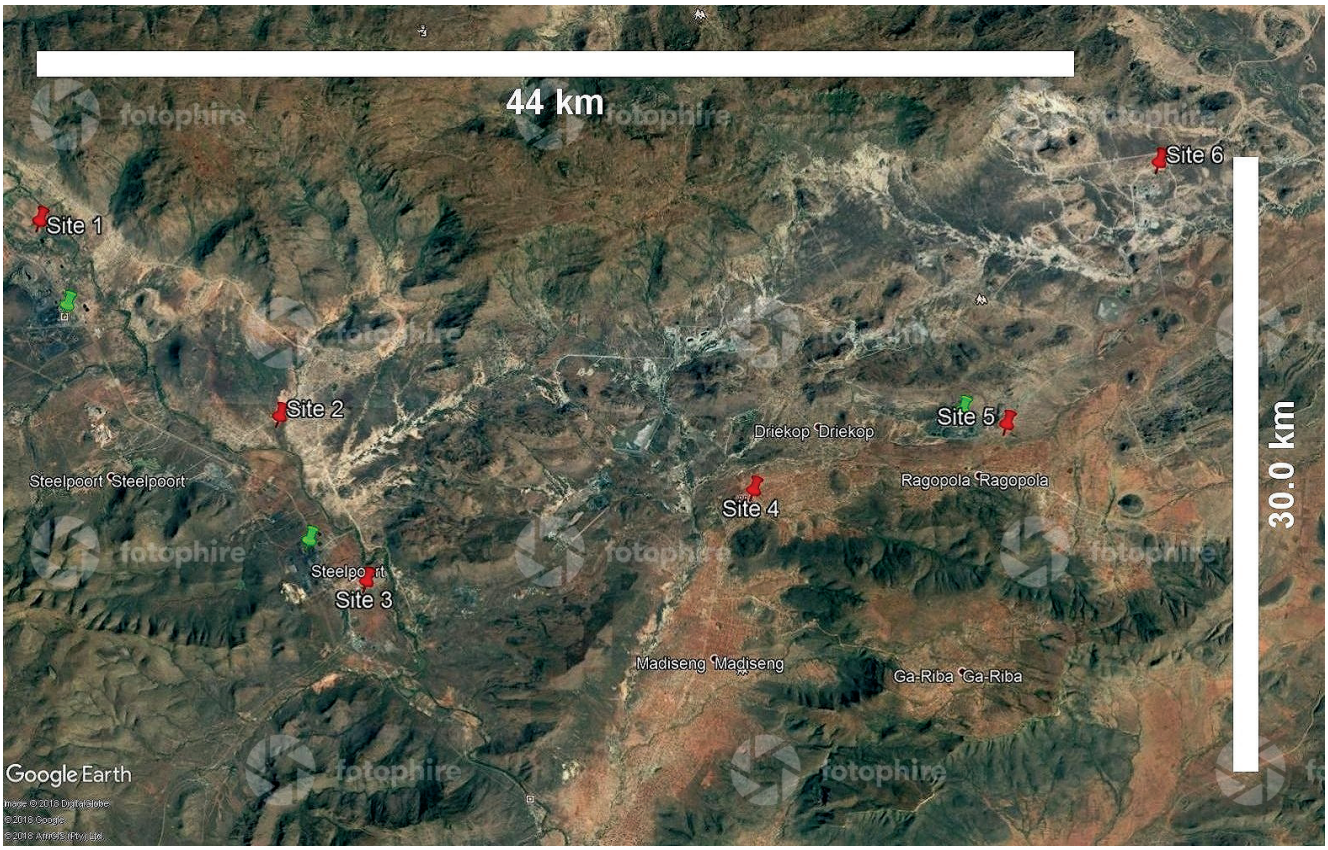

Figure A1: *Map of the study area showing passive sampler locations (red place-marks on Google map), with smelters shown as green place-marks.*

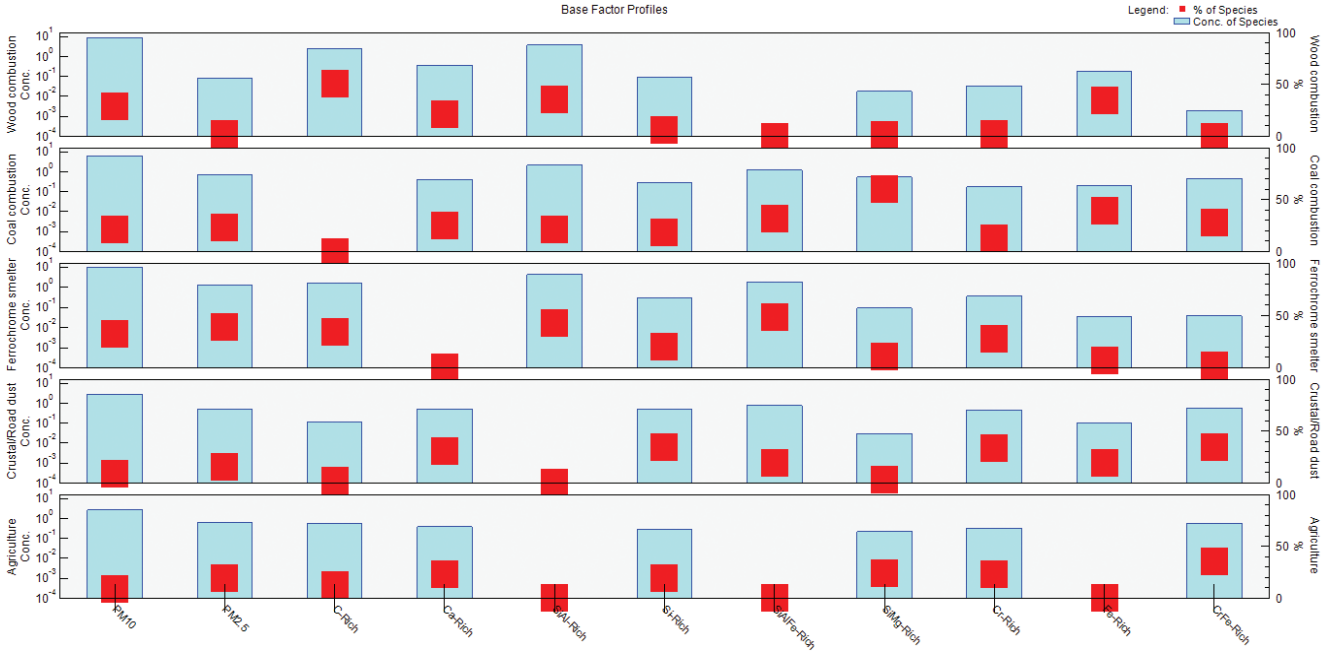

Figure A2.1: *Factor fingerprints for species at site 1.*

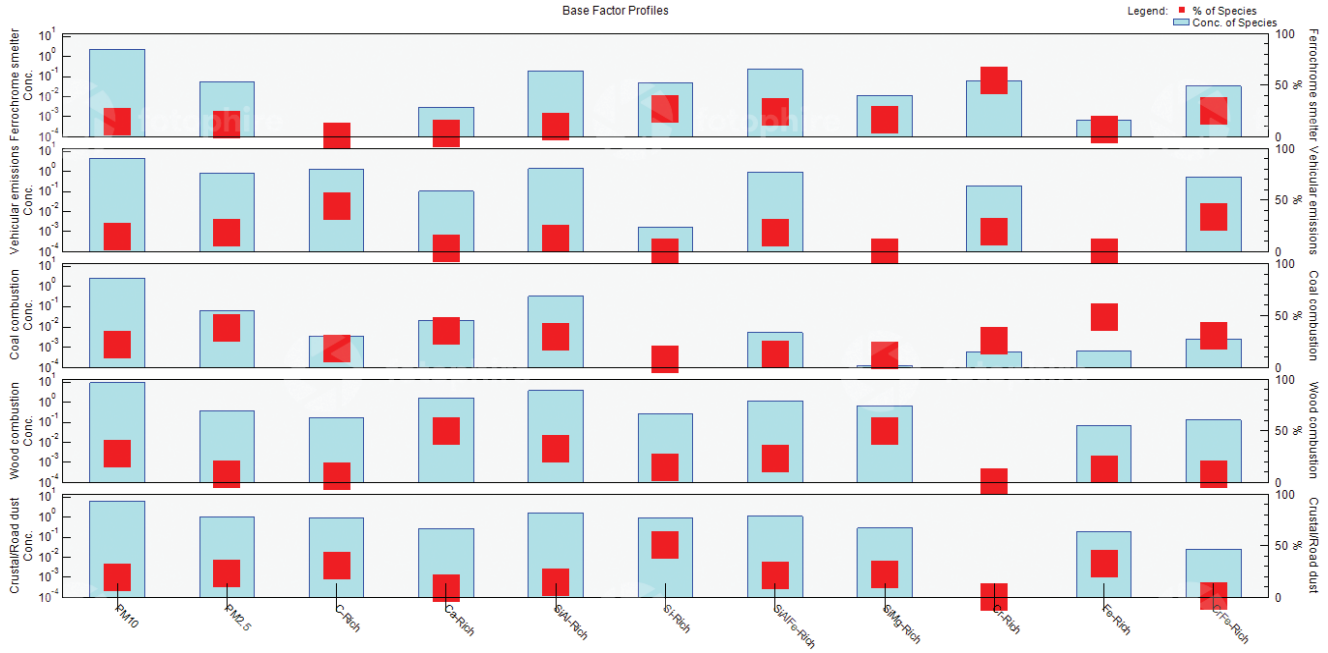

Figure A2.2: *Factor fingerprints for species at site 2.*

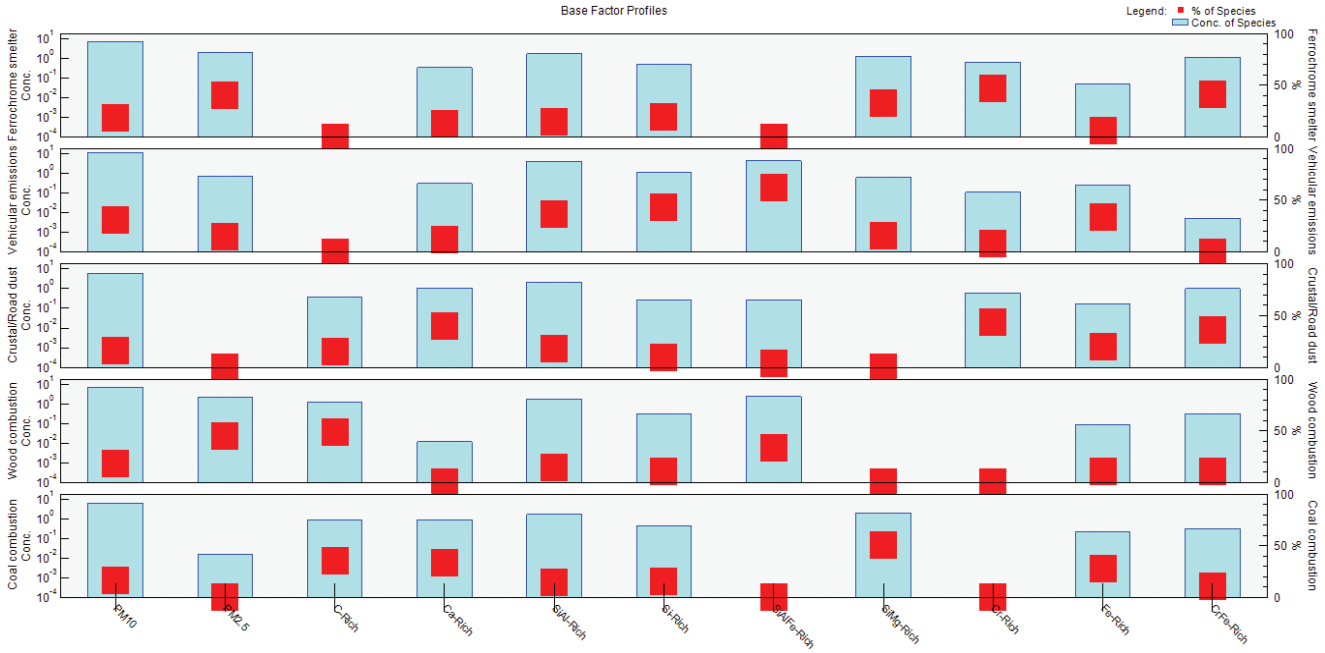

Figure A2.3: *Factor fingerprints for species at site 3.*

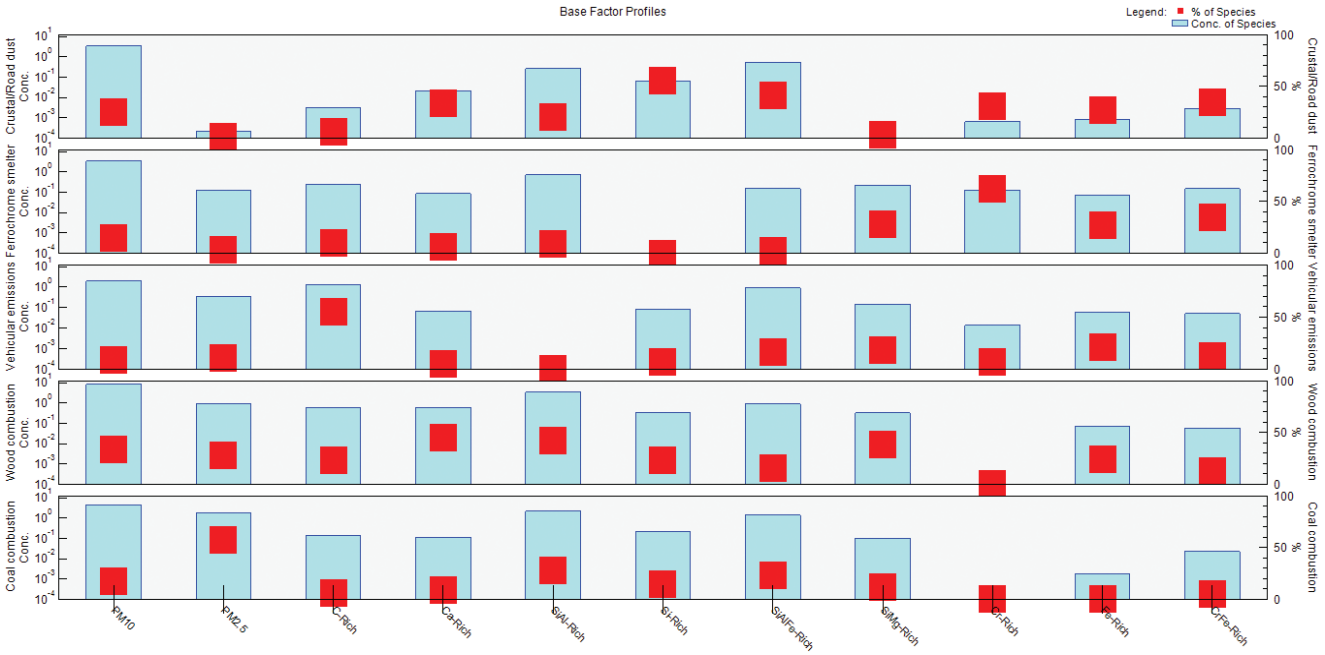

Figure A2.4: *Factor fingerprints for species at site 4.*

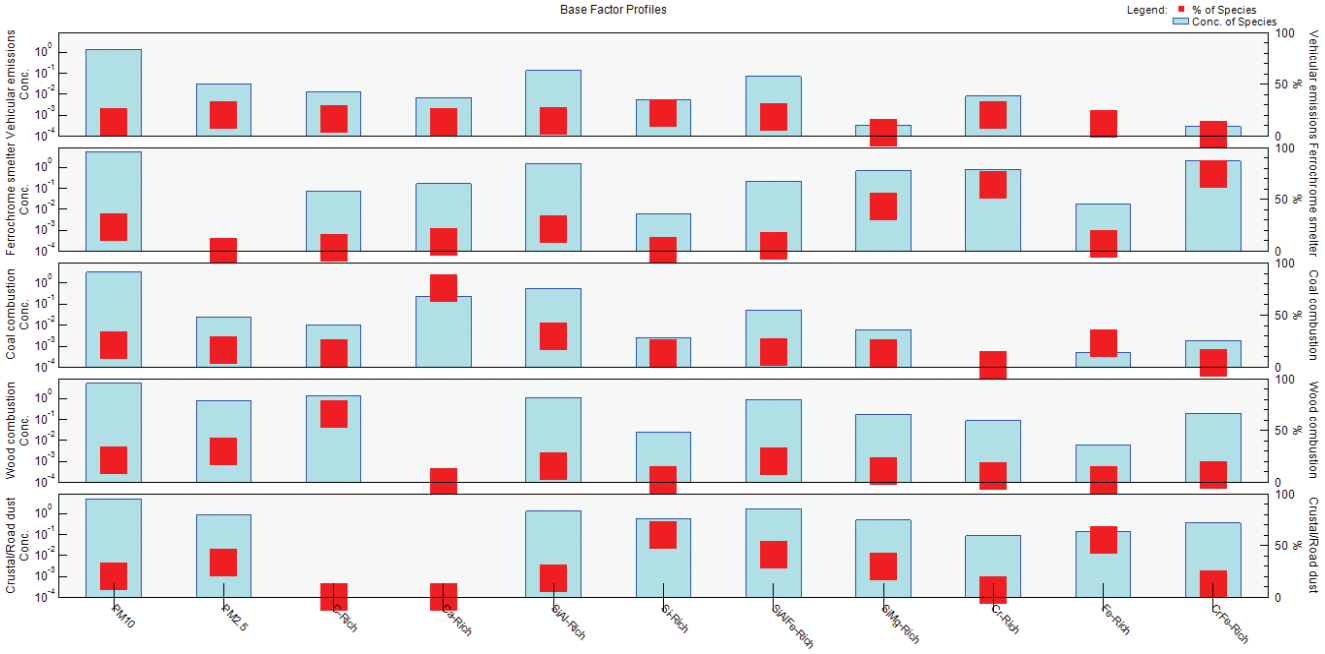

Figure A2.5: *Factor fingerprints for species at site 5.*

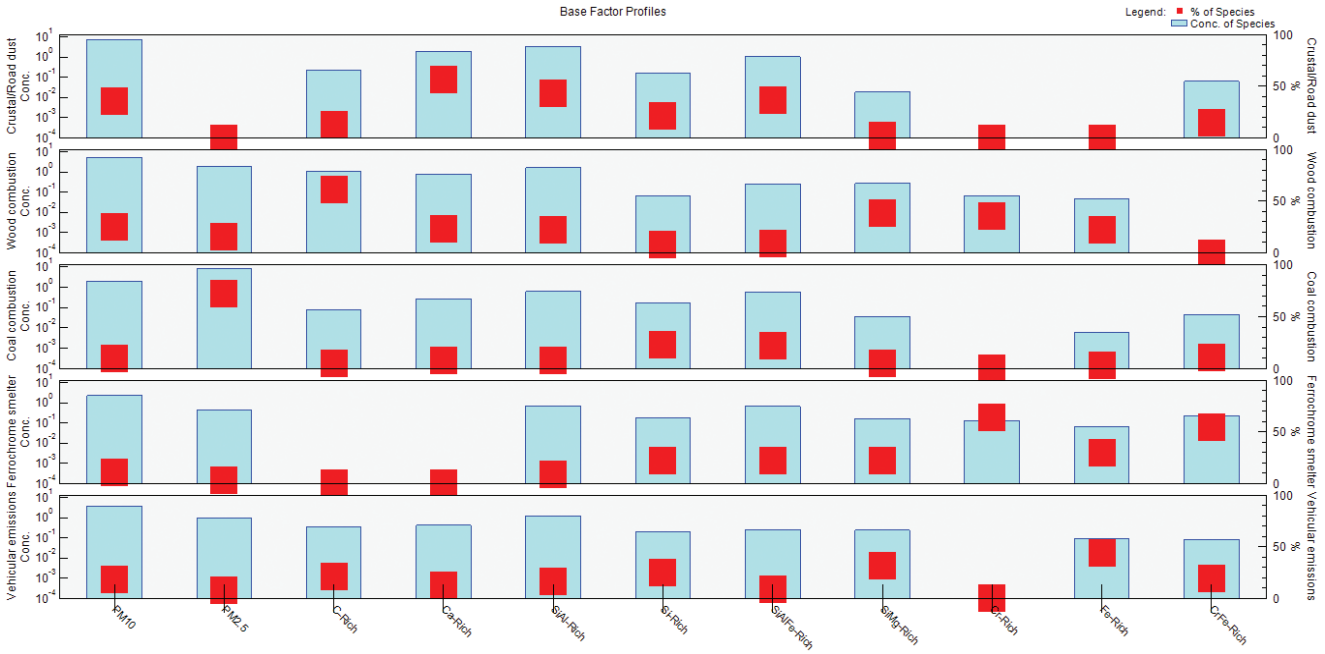

Figure A2.6: *Factor fingerprints for species at site 6.*

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Research article

A critical review of health risk assessments of exposure to emissions from coal-fired power stations in South Africa

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Abstract

Emissions from coal-fired power stations increase the incidence of respiratory, cardiovascular and cardiopulmonary diseases and contribute to premature deaths. Wildly varying estimates of the magnitude of these health impacts have been published, however. This paper investigates the reasons for the large discrepancies calculated in five comprehensive health risk assessments of South African coal-fired power station emissions. We review the approaches and input data used by the studies. We also evaluate the exposure-response functions (which relate the pollution concentration to which the population is exposed, to the increase in health risk) used by each study and pay specific attention to whether the exposure-response functions are relevant to the South African context. Health risks are under-estimated in studies that only consider impacts in industrialised areas, or assume high counterfactual concentrations. Health risks are probably over-estimated, however, in other studies which use linear or exponential exposure-response functions that are not applicable in areas where exposure levels are much higher (such as solid fuel-using communities). 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. A fair assessment of the health risk of South Africa's older coal-fired power stations should also weigh the health costs of the emissions against the health benefits of electricity use in lower income households.

Keywords

health impact, coal-fired power stations, exposure-response function, electrification

Introduction

The World Health Organisation (WHO 2016) estimates that there were 3 million premature deaths worldwide in 2012 due to ambient particulate matter pollution. A *premature death* is a death that occurs before the average age of death in a certain population (National Cancer Institute 2015). The health risk is spread disproportionately across the globe, with highest risks in low- and middle-income countries in the Western Pacific (where there were on average 65 age-standardised deaths per 100 000 capita attributable to ambient air pollution in 2012). There is a much lower risk due to ambient air pollution exposure in North and South America (only 7 age-standardised premature deaths per 100 000 capita in high-income countries, and 18 age-standardised premature deaths per 100 000 capita in low- and middle-income countries in 2012; WHO 2016). In South Africa, 14 356 premature deaths were estimated to have occurred in 2012 due to acute lower respiratory illness (ALRI), chronic obstructive pulmonary disease (COPD), lung cancer, ischemic heart disease (IHD) and strokes. These account for 3.0% of the 480 476 deaths that occurred in South Africa in 2012 (StatsSA 2014).

The South African Comparative Risk Assessment (Norman et al. 2007) estimated premature mortality from urban air pollution to be 4 637 (0.9% of all deaths) in 2000. These figures were calculated considering monitored particulate matter with aerodynamic diameter of less than or equal to 2.5 micrometres ($PM_{2.5}$) and particulate matter with aerodynamic diameter of less than or equal to 10 micrometres ((PM_{10}) concentrations, in the large metropolitan areas of Cape Town, eThekwini, Johannesburg and surrounds, Ekurhuleni, the Vaal Triangle and Nelson Mandela Metro, and so largely exclude the impact of power station emissions and other industrial activities on the Mpumalanga Highveld.

Fine particulate matter is derived from a large number of combustion sources, including power stations, industries, mines, vehicles, vegetation fires and domestic burning. There have been many attempts to apportion particulate matter to the sources from which it is derived, based on chemical signatures of particulate samples (for example, Annegarn et al. 1999) and chemical transport or dispersion modelling (for example, the Highveld and Vaal Triangle Priority Area Baseline Assessments). Apportionment of the particulate matter is no easy task because the bulk of fine particulate matter is formed in the atmosphere (Maenhaut et al. 1996; Piketh et al. 1999). A source apportionment is, however, necessary in order to apportion the harmful health effects of particulate pollution.

The health impacts of emissions from power stations have received particular attention, with Myllyvirta (2014) and Holland (2017) calculating that around 2 239 deaths per year in South Africa are due to particulates from coal-fired power stations. These figures are much higher than those previously calculated by van Horen (1996) – 174 premature deaths per year; Scorgie et al. (2004; henceforth called the FRIDGE (Fund for Research into Industrial Development Growth and Equity) study) – 10 deaths per year; Scorgie and Thomas (2006; henceforth called the Airshed and Infotox study) – 17 deaths per year; and Grobler (2016) – 57 deaths per year. The reasons for the disparities in the estimates of the health effects of South Africa's coal-fired power station emissions are explored in this paper. Coal-fired power generation is but one source of many that contribute to ambient particulate matter levels in South Africa. Nevertheless, emissions from coal-fired power stations are the sole focus of this paper because of the magnitude of the emissions, their extensive area of impact, and the large investments required for emission reduction that are the subject of much debate in South Africa at the moment.

Health effects of coal-fired power station emissions

Impurities in coal are released during combustion in the boiler of a coal-fired power station. More than 99% of ash is removed by fabric filter plants or electrostatic precipitators in South African power stations, but most of the other pollutants, like sulphur dioxide (SO_2), oxides of nitrogen (NO_x) and trace substances like mercury are released, unabated, to the atmosphere (Pretorius et al. 2015). These pollutants are released through tall stacks (ranging between 152 m and 300 m for Eskom's power stations), are diluted and undergo chemical transformations, and finally come to the surface where they may be inhaled or affect the physical environment. As the plume ages, most of the SO_2 and NO_x is converted to secondary fine particulate matter in the atmosphere. Highest ground-level concentrations of SO_2 and NO_x typically occur during the day when turbulent eddies bring less diluted plumes to the ground, but plumes may also be advected for long distances (many tens of kilometres) before they are brought to the ground.

When pollutants from coal combustion (and other types of combustion) are inhaled, they may have a range of harmful effects on health during the entire human lifespan. This review only considers health risk assessments that focus on exposure to ambient particulate matter, SO_2, NO_2 and ozone (O_3) (where included in the studies), but a more thorough review of the effects of power station emissions on health is given here to provide a comprehensive picture.

Exposure to air pollution affects early childhood development. Heavy metal and ultrafine particulates are able to cross the placental barrier and have the potential to harm the foetus and its developing organs (Wick et al. 2010). There is strong evidence that ozone and SO_2 are associated with premature birth, with weaker evidence for particulates (Ha et al. 2014). Exposure to particulates, and perhaps also to ozone, NO_2 and carbon monoxide (CO) during pregnancy may affect foetal growth and increases the risk of low birth-weight (Glinianaia et al. 2004; WHO 2013). The concern with premature birth and low birth-weight is that they have an impact on the developing organs.

Heavy metals, like lead and mercury, have been associated with neurodevelopmental harm, leading to reduced cognitive function, lower intelligence quotient (IQ), attention deficit hyperactivity disorder and possibly autism spectrum disorder during childhood (Canfield et al. 2003; Liu and Lewis 2014.

Young children are particularly vulnerable to the effects of air pollution. After birth, the organs are still maturing and infants have a relatively high metabolic rate so they breathe a greater volume of air than adults, relative to their size (RCP 2016). Early-life exposure to air pollution is also thought to cause epigenetic modification through changes in DNA methylation (Janssen et al. 2013; Jiang et al. 2014).

Long-term exposure to air pollution (particularly particulates, black carbon and NO_2) suppresses the development of lung function (Chen et al. 2015; Hwang et al. 2015; Kulkarni et al. 2006), and may speed up the decline of lung function into older age (Rice et al. 2015; Adam et al. 2015). Even relatively small disturbances in the normal development or functioning of organs can significantly change the number of individuals in a population who develop diseases as a consequence. For example, a small change in lung function can shift the normal distribution of lung function in a population downwards, increasing the number of people in the lower tail who have low enough lung function to cause disease (RCP 2016). Acute exposure to high levels of air pollution results in an increased incidence of respiratory symptoms in children.

Outdoor air pollution causes instances of lung cancer (IARC 2013; Raaschou-Nielsen 2013). Long-term exposure to air pollution (NO_2 and particulates) has been linked to the development of asthma (McConnell et al. 2010; Gasana et al. 2012; Anderson et al. 2013; Chen et al. 2015), and there is evidence that air pollution can make asthma worse in people who already have it. There is a strong link between air pollution and cardiovascular disease (myocardial infarction, heart disease and stroke) (Brook et al. 2010; WHO 2013; Newby et al. 2015).

Calculating health effects of air pollution

Ostro's (1994) impact pathway approach to calculate the health impact of outdoor particulate air pollution has been adopted by the majority of health risk assessments conducted to date. The method for calculating the health impact of elevated particulate matter concentrations is as follows, and is similar to the methods used for SO_2, NO_2 and O_3:

i. Assess ambient exposure of the population to particulate matter based either on fixed-site measurements from ambient air quality monitoring stations or on model-based estimates.
ii. Identify a counterfactual exposure (also called a 'target' or 'background' PM concentration), below which it is assumed there is no harmful health effect. This level is used for comparison, to determine the potential benefit (in terms of disease reduction) of reducing the risk factor.
iii. Determine the size of the population group exposed to particulate matter.
iv. Determine the incidence of the health effect being estimated, e.g. the underlying mortality rate of the population, in deaths per hundred thousand.
v. Use exposure-response functions (also called dose-response functions or concentration-response functions) that relate the concentrations of $PM_{2.5}$ or PM_{10} to selected health effects
vi. Calculate the attributable fraction for each health outcome based on the relative risk of the exposed population
vii. Calculate the attributable health burden by multiplying the population-attributable fraction by the health outcome.

Health outcomes

A rather bewildering array of health effects are reported by different studies. Premature mortality and morbidity (such as number of hospital admissions or incidences of an illness) outcomes are often calculated. Disability-adjusted life years (DALYs) are a measure of the total health impact (quality of life and longevity) and are calculated by adding the years of life lived with disability and the years of life lost. The effects of pollution on the development of foetuses and young children are usually not directly accounted for.

Health effects are typically divided into categories, based on the length of exposure (short-term or long-term), the type of health effect and the age of the population:

i. all-cause mortality, due to short-term exposure to PM_{10}. This is not used in DALY calculations and should not be added to any other health estimate;
ii. respiratory mortality, due to short-term exposure to PM_{10} for children <5 years;
iii. cardiopulmonary mortality due to long-term exposure to $PM_{2.5}$ for adults >25 years; and
iv. lung cancer mortality, due to long-term exposure to $PM_{2.5}$ for adults >25 years (Ostro 2004; WHO 2016).

Exposure-response relationship

The relationship between exposure to a particular concentration of pollutants and the health effect is expressed either as an exposure-response function, or as a relative risk. The relative risk of exposure is determined by two main methods. In the first method, the relationship between short-term exposure (one- or two-day averages) and daily health impact (e.g. daily mortality) is determined through time series studies where the associations between changes in health outcomes and changes in exposure indicators are evaluated. The outcome is a count (i.e. the Poisson distribution) and the model form is usually log-linear. The Harvard Six Cities time-series analysis is an example of such a study. In the second method, the health effects of chronic, long-term exposure to (annual average) ambient concentrations are determined by cross-sectional 'ecologic' studies which compare exposure and responses at the community level and, more usefully, by prospective cohort studies which use data from a sample of individuals observed over time (for example the Harvard Six Cities study (Dockery et al. 1993) and the American Cancer Society (ACS) study (Pope et al. 1995)). The outcome of these studies is continuous (person-time per exposure) and the relationship is (somewhat) linear (USEPA 2004). The types of exposure-response functions used in the five studies reviewed in this paper are listed in Table 2.

Künzli et al. (2001) (and the USEPA (2004)) conclude that "time-series analyses underestimate causes of death attributable to air pollution and that assessment of the impact of air pollution on mortality should be based on cohort studies." Time series approaches only capture the deaths of already frail persons, induced by exposure to air pollution shortly before death. Cohort studies capture all cases of deaths related to pollution exposure, including the risk of underlying diseases leading to frailty.

Considering the example of PM_{10} concentrations, an exposure-response relationship is multiplied by the increase in PM_{10} concentrations and the number of exposed people to calculate the health impact I:

$$I = ERF * pop\ size * \Delta X \quad ...Equation\ 1$$

where
ERF is the exposure-response function;
$pop\ size$ is the size of the exposed population; and
ΔX is the change in PM_{10} concentration ($\mu g/m^3$) due to the source of concern

As an illustration, considering the central estimate for PM^{10} mortality for people <65 years old of $0.23*10^{-8}$ in the van Horen (1996) study (Table 2), one person in $0.23*10^8$ (23 million) will die for each 1 $\mu g/m^3$ increase in PM_{10} concentration.

Health impact I, expressed as the number of cases that can be attributed to the exposure, can also be calculated from the relative risk as:

$$I = AF \times incid \times pop\ size \quad ...Equation\ 2$$

where
AF is the attributable fraction of deaths due to exposure to PM_{10}; and
incid is the incidence of death in the population (non-accidental mortality).

The attributable fraction *AF* is given by:

$$AF = RR - 1/RR \quad ...Equation\ 3$$

where *RR* is the relative risk of death due to exposure to PM_{10} and is given by:

$$RR = exp\ [\beta(X-X_0)] \quad ...Equation\ 4$$

where
β is the fractional increase in mortality/morbidity incidence per 1 µg/m³ increase in pollution concentration;
X is the current annual mean concentration of the pollutant of concern; and
X_0 is the threshold/baseline concentration of the pollutant.
($X-X_0$ corresponds to ΔX in equation 1.)

For example, if there is an increase of 0.8% in the mortality rate due to a 10 µg/m³ increase in PM_{10} concentration, the value of β would be 0.0008.

The exposure-response function and the relative risk function calculated for an incremental increase in pollution of X-X_0 are related as follows:

$$ERF = \frac{AF \times incid}{X-X_0} \quad ...Equation\ 5$$

In many studies, a linear relationship is assumed between amount of pollution inhaled and severity of the health response (e.g. van Horen 1996). However, the development of integrated exposure-response functions which combine exposure-health relationships for ambient air pollution, household air pollution, second-hand tobacco smoke and active smoking has shown that there is a levelling-off of the health impact at higher exposure levels (Pope et al. 2009; Pope et al. 2011; Smith et al. 2014; Burnett et al. 2016).

Counterfactual exposure

The counterfactual exposure, also called the background or threshold concentration, is the baseline concentration for comparison of the health risks. The World Health Organisation (2013) argues that there is no safe level of exposure to particulate matter below which there are no negative health effects. Several studies select zero µg/m³ as the counterfactual concentration (for example, van Horen (1996) and in all likelihood, Myllyvirta (2014)). However, others argue that zero exposure is not a practical counterfactual level because this is impossible to achieve, even in pristine environments (Brauer et al. 2012). Lim et al. (2012) define the theoretical-minimum-risk exposure distribution based on the minimum concentration in the studies used to estimate risk (below which there is clearly no evidence of an association between exposure and health).

Studies on health impacts of South African coal-fired power stations

To date, five comprehensive assessments have been performed on the impact of South African coal-fired power station emissions on human health. Van Horen's (1996) PhD thesis is a comprehensive assessment of the environmental externalities in South Africa's energy sector. He considers the health effects of particulate matter and ozone that form as a result of power station emissions. The FRIDGE study by Scorgie et al. (2004) was sponsored by the National Economic, Development and Labour Council (NEDLAC), and considers the health costs of air pollution from a comprehensive range of sources. It also considers the cost-benefit ratio of interventions to improve air quality. The Airshed and Infotox study (Scorgie and Thomas 2006) was commissioned by Eskom and also considers most major sources of air pollution. The Myllyvirta (2014) study was commissioned by Greenpeace, and is the basis for the Holland (2017) study. It considers emissions in excess of the 2020 limits for new plants in the *Listed Activities and Associated Minimum Emission Standards published in terms of Section 21 of the National Environmental Management: Air Quality Act (Act No 39 of 2004)*. Steyn and Kornelius (2018) calculate the health benefits (expressed in monetary terms) of reducing SO_2 emissions from power stations, from current levels to compliance with the new plant SO_2 emission standard of 500 mg/Nm³. A comparison between the inputs used for these five studies is given in Table 1.

All studies use dispersion or chemical transport models that calculate the secondary formation of particulates from SO_2 and NO_x emissions. The van Horen (1996) and Myllyvirta (2014) studies use models that are not customised to the South African environment. Steyn and Kornelius (2018) only consider ambient SO_2 and secondary particulate (sulphate) levels for SO_2 emissions in excess of 500 mg/Nm³.

The FRIDGE (2004) study assumes daily counterfactual concentrations of 25 µg/m³ for PM_{10} and 15 µg/m³ for $PM_{2.5}$, based on the recommendations of the CEPA/FPAC Working Group (1998). The Airshed and Infotox (2006) study uses annual counterfactual concentrations of 15 µg/m³ for PM_{10} (as per Cohen et al. 2004), 25 µg/m³ for SO_2 and 20 µg/m³ for NO_2 (these levels are 50% of the annual average air quality limits recommended by the WHO (2000)). Van Horen (1996) and Steyn and Kornelius (2018) use a counterfactual exposure of zero. The counterfactual concentration used by Myllyvirta (2014) is not clear. The Krewski et al. (2009) study, from which Myllyvirta's relative risk factors are drawn, uses a counterfactual concentration with uniform distribution between 5.8 and 8.8 µg/m³, but Myllyvirta (2014) applies the relative risk factors to exposure concentrations

Table 1: *Inputs used for health risk assessments of South African coal-fired power station emissions*

	van Horen (1996)	FRIDGE (2004)	Airshed & Infotox (2006)	Myllyvirta (2014)	Steyn & Kornelius (2018)
Ambient air pollution concentrations	EXMOD model's embedded air quality dispersion models	CALPUFF dispersion modelling suite	CALPUFF dispersion modelling suite	Regression models derived from single-source CTM (CAMx and CALPUFF) model runs	CALPUFF dispersion modelling suite
Counterfactual concentration	Zero	Daily PM_{10}: 25 µg/m^3 Daily $PM_{2.5}$: 15 µg/m^3 (CEPA/FPAC Working Group, 1998)	PM_{10}: 15 µg/m^3 (Cohen et al., 2004) SO_2: 25 µg/m^3; NO_2: 20 µg/m^3	Not clear (zero?)	Zero (for the difference between baseline and compliance ambient concentrations)
Power stations considered	Arnot, Duvha, Hendrina, Kendal, Kriel, Lethabo, Matimba, Matla, Tutuka	Arnot, Duvha, Hendrina, Kendal, Kriel, Lethabo, Majuba, Matla, Tutuka	Arnot, Duvha, Hendrina, Kendal, Kriel, Lethabo, Majuba, Matla, Tutuka	Arnot, Camden, Duvha, Grootvlei, Hendrina, Kendal, Komati, Kriel, Lethabo, Majuba, Matimba, Matla, Medupi, Tutuka	Arnot, Camden, Duvha, Grootvlei, Hendrina, Kendal, Komati, Kriel, Majuba, Matla, Tutuka and Sasol Synfuels steam plants
Power station emissions considered	PM, SO_2, NO_x – 1994 annual emissions	PM, SO_2, NO_x, CO, N_2O, benzene, lead, CH_4, TNMOC, CO_2 – 2002 annual emissions	PM, SO_2, NO_x – 2003 annual emissions	PM, SO_2, NO_x – 2012/13 annual emissions in excess of Minimum Emission Standards	SO_2 only – difference between 2012/13 annual emissions for Eskom power stations and 2014 operations for Sasol Steam Stations, and compliance with new plant SO_2 emission standard
Other polluting sources considered	None	Industries, services, agriculture, transport, domestic fuel burning	Industries, open cast mines, ash dumps, household fuel burning, vehicle exhausts	None	None
Population data	1991 census data, per magisterial district	2001 census data	2001 census data	GPWv3# projections for 2010	2011 census
Geographical domain	South Africa	Mpumalanga Highveld & Vaal Triangle	Industrialised Highveld	South Africa	Highveld Priority Area
Exposed population size	36.2 million	18.7 million	10.83 million	~50 million	Not stated

#GPWv3 is the Gridded Population of the World, Version 3 (GPWv3): Population Count Grid, Future Estimates. Edition: 3.00

Table 2: *Exposure-response risk functions used in the health risk assessments of South African coal-fired power station emissions*

	van Horen (1996) (after Rowe et al. 1994)	FRIDGE (2004)	Airshed & Info-tox (2006)	Myllyvirta (2014)	Steyn & Kornelius (2018)
	Exposure-response functions per 1 µg/m³ change in PM_{10} concentration or 1 ppb change in O_3 concentration (number in brackets denotes the probability)		**Percent increased risk per 50 µg/m³ concentration increase**	**Relative risks for a 10 µg/m³ $PM_{2.5}$ concentration increase**	**Relative risks for concentration increase stipulated below**
PM_{10} mortality	Daily exposure: ≥65 years: L# $10.1*10^{-8}$ (P 33%) C# $16.9*10^{-8}$ (P 34%) H# $25.4*10^{-8}$ (P 33%) <65 years: L $0.14*10^{-8}$ (P 33%) C $0.23*10^{-8}$ (P 34%) H $0.35*10^{-8}$ (P 33%)	Daily exposure: ≥65 years: $4.42*10^{-7}$ (EXMOD) <65 years: $2.35*10^{-8}$ (EXMOD)	Annual exposure 6.1% (Krewski et al. 2000 and ACS study, USEPA 2004)	Annual $PM_{2.5}$ exposure Lung cancer: 1.14 IHD: 1.26 COPD: 1.05 Stroke: 1.12 ALRI (<5 yrs): 1.12	Annual sulphate exposure All-cause mortality: 1.07 for a 5 µg/m³ concentration increase (Krewski et al. 2009)
SO_2 mortality		Daily exposures ≥65 years: $1.01*10^{-8}$ <65 years: $1.38*10^{-9}$ (Watkiss and Holland for EC DG Environ)	Annual exposure 10%		Annual exposure All-cause mortality: 1.02 for a 14 µg/m³ concentration increase (Krewski et al. 2009) Daily exposure Infant mortality <5 years: 1.06 for a 9.2 µg/m³ concentration increase (Lin et al. 2004)
NO_2 mortality			Annual exposure 1.3%		
PM_{10} respiratory hospital admission	Daily exposure: L $1.8*10^{-8}$ (P 25%) C $3.3*10^{-8}$ (P 50%) H $4.8*10^{-8}$ (P 25%)	Daily exposure: $1.2*10^{-5}$ (Ostro, 1994)	Daily exposure 7.3% (Mean of COMEAP 1998; McGowan et al. 2002 and USEPA 2004)		Daily exposure for sulphate 0.14% change per 1 µg/m³ concentration increase (Atkinson et al. 2014)
SO_2 respiratory hospital admission		Daily exposure: $2.01*10^{-6}$ (Maddison, 1997)	Daily exposure 2.5%		Daily exposure 0.05% change per 1 µg/m³ concentration increase (COMEAP as cited by Stedman et al. 1999)
NO_2 respiratory hospital admission		Daily exposure: $1.65*10^{-6}$ (Maddison, 1997)	Daily exposure 2.5%		
PM_{10} cardio-vascular hospital admission		Daily exposure: $1.01*10^{-7}$ (Dockery et al., 1989)			Daily exposure for sulphate 0.12% change per 1 µg/m³ concentration increase (Atkinson et al. 2014)

#L, C and H denote the low, central and high estimates, respectively

of less than 3.4 µg/m^3, so it is assumed that a counterfactual concentration of zero was in fact used.

The scope of the studies also differs. Van Horen (1996) and Myllyvirta (2014) consider the impact over the whole of South Africa, while the FRIDGE (2004) and Airshed and Infotox (2006) studies only consider the industrialised Highveld. Steyn and Kornelius (2018) consider the Highveld Priority Area (the industrialised Mpumalanga Highveld and eastern Gauteng), and include emissions from the Sasol Synfuels Steam Stations. Myllyvirta (2014) also considers the three return-to-service power stations (Camden, Grootvlei and Komati) and is the only study to consider Medupi Power Station.

The exposure-response functions used to relate exposure to the pollutant and health outcome are given in Table 2. The van Horen (1996) and FRIDGE (2004) studies use functions for acute (daily) exposures derived from time series studies for mortality estimates, while the Airshed and Infotox (2006), Myllyvirta (2014), and Steyn and Kornelius (2018) studies use functions for chronic (annual average) exposure derived from cohort studies for the mortality estimates. The van Horen (1996) and FRIDGE (2004) calculations of premature mortality are probably an underestimate of the actual impact (Künzli et al. 2001; USEPA 2004). All hospital admissions are calculated from acute (daily) exposure functions. Myllyvirta's (2014) relative risk functions are sourced from the American Cancer Society study, with the reference given as Krewski et al. (2009). However, the relative risks appear rather to come from an original analysis of the American Cancer Society Study by Burnett et al. (2012 – supplemental material).

It is difficult to compare the dose-response functions with the relative risk functions as they are presented in Table 2, so a few of the relative risk functions used by Myllyvirta (2014), Airshed and Infotox (2006) and Steyn and Kornelius (2018) have been converted to dose-response functions using Equation 5 (Table 3). The PM_{10} daily mortality exposure-response functions used by van Horen (1996) and FRIDGE (2004) differ by an order of magnitude, which is surprising since they are both apparently the factor used in the EXMOD model used by van Horen (1996). There may be an error in van Horen's table. The dose-response function used by Steyn and Kornelius (2018) for mortality due to chronic exposure to sulphates is slightly lower than the function used by Myllyvirta (2014) for mortality due to chronic exposure to $PM_{2.5}$, which is consistent since sulphates are just one of many components of $PM_{2.5}$. The exposure-response factor used by van Horen (1996) for PM_{10} respiratory hospital admissions is three orders of magnitude lower than that used by FRIDGE (2004) and Airshed/Infotox (2006), while the factor used by Steyn and Kornelius (2018) is one order of magnitude lower.

There are also significant differences in the incidence of death or disease in the general population, as used in the health risk studies (Table 4). The rates of respiratory hospital admissions used in FRIDGE (2004) and by Steyn and Kornelius (2018) are much higher than those used in Airshed and Infotox (2006). The total mortality rates used in FRIDGE (2004), Airshed and Infotox (2006) and Steyn and Kornelius (2018) are similar, while the cardiovascular mortality rate used in Airshed and Infotox (2006) is higher than that used in the FRIDGE (2004) and Myllyvirta (2014) studies.

The FRIDGE study predicts by far the highest number of hospital admissions due to exposure to air pollution – 5 456 per year. Steyn and Kornelius (2018) calculate the lowest number of hospital admissions (98 in total), since they are only considering the impact of SO_2 emissions in excess of the Minimum Emission Standards in the Highveld Priority Area.

Table 5 is not an exhaustive list of the health impacts considered in the health risk assessments. Rather, a number of health outcomes covered by most studies have been selected for comparison. Other types of health outcomes considered include chronic bronchitis, restricted activity days (van Horen 1996 and FRIDGE 2004) and asthma attacks (van Horen 1996 and Steyn and Kornelius 2018).

Relevance of exposure-response functions

It is worth reflecting on the shape of the exposure-response functions used in the different studies, and the exposure levels over which they are applied. The exposure-response functions by van Horen (1996) and FRIDGE (2004) are linear, while those used by Airshed and Infotox (2006), Myllyvirta (2014) and Steyn and Kornelius (2018) are exponential (as per Ostro 2004). The exposure-response relationships given by these two types of functions are similar at low concentrations, but differ significantly at higher concentrations.

Table 3: *Comparison of exposure-response functions used in the health risk assessments of South African coal-fired power station emissions*

	van Horen (1996)	FRIDGE (2004)	Airshed and Infotox (2006)	Myllyvirta (2014)	Steyn & Kornelius (2018)
PM_{10} mortality – daily exposures < 65 years	$2.30*10^{-9}$	$2.35*10^{-8}$			
PM_{10} mortality – daily exposures ≥ 65 years	$1.69*10^{-7}$	$4.42*10^{-7}$			
PM_{10} / $PM_{2.5}$ mortality – annual exposures			$2.60*10^{-5}$ for PM_{10}	$3.35*10^{-5}$ for $PM_{2.5}$	$8.52*10^{-6}$ for sulphates
PM_{10} respiratory hospital admissions	$3.30*10^{-8}$	$1.20*10^{-5}$	$1.39*10^{-5}$		$1.28*10^{-6}$ for sulphates

Table 4: *Population incidence of death/disease (per 100 000 people) used in the health risk assessments of South African coal-fired power station emissions*

	Incidence of death/ health outcome	Source
Van Horen (1996)	Not published	
FRIDGE (2004)	Total mortality: 1 235.0 Cardiovascular mortality: 43 Respiratory mortality: 141 Respiratory hospital admissions: 3 100	Bradshaw et al. 2003 StatsSA 2002 StatsSA 2002 Joburg 2000
Airshed & Infotox (2006)	Total non-accidental mortality: 1 065.0 Cardiovascular mortality: 204.9 Respiratory mortality: 93.7 Respiratory hospital admissions: 477.6	Bradshaw et al. 2004 (sum of provincial data) KZNDOH 2004
Myllyvirta (2014)	Lung cancer mortality: 9.1 IHD mortality: 34.6 COPD mortality: 11.8 Stroke mortality: 48.6 Lower respiratory infection (<5 years old): 12.2	Global Burden of Disease 2010
Steyn & Kornelius (2018)	Total mortality: 1 110 Child mortality <5 years: 85.5 Respiratory hospital admissions: 5 420 Cardiac hospital admissions: 1 500	StatsSA 2014 StatsSA 2014 Da Costa 2009 Da Costa 2009

Table 5: *Selected mortality and morbidity estimates from the health risk assessments of South African coal-fired power station emissions*

	van Horen (1996)	FRIDGE (2004)	Airshed and Infotox (2006)	Myllyvirta (2014)	Steyn & Kornelius (2018)
Premature mortality: PM_{10} and O_3	174 (56-266)				
Premature mortality: PM		5 (PM_{10})	0 (PM_{10})	2 238 ($PM_{2.5}$)	32 (sulphates)
Premature mortality: SO_2		5.2	16.6		25
Premature mortality: NO_2			0		
Respiratory hospital admissions: PM_{10} and O_3	672 (360-962)				
Respiratory hospital admissions: PM_{10}		1 981	0.0		47 (sulphates)
Respiratory hospital admissions: SO_2		1 500	661.2		40
Respiratory hospital admissions: NO_x		1 958	0.01		
Cardiovascular hospital admissions		16.7			11

Several recent studies have developed integrated exposure-response (IER) functions that model disease risks across three orders of magnitude in the range of $PM_{2.5}$ exposure associated with three main sources of combustion pollution: ambient air pollution, second-hand tobacco smoke and active tobacco smoking (Pope et al. 2009; Pope et al. 2011; Smith et al. 2014). Burnett et al. (2016) also included household air pollution due to domestic solid fuel burning. The WHO's *Ambient air pollution: Global assessment of exposure and burden of disease* (2016) uses IER functions developed by Burnett et al. (2016) for the Global Burden of Disease (GBD) 2010 study, and updated by the GBD 2013 study. The IER functions show a greater increase in relative risk of mortality for an incremental increase in $PM_{2.5}$ concentration at lower concentrations, but a much smaller increase in relative risk at higher concentrations. It is preferable to use the IER functions when exposure levels of the population in question exceed the ambient concentrations in the North American or European studies from which the exposure-response functions used in the health risk assessments reviewed here were derived.

The relative risk factors derived from the American Cancer Society study used by Myllyvirta (2014) were derived at an annual average $PM_{2.5}$ concentration of 14.2 µg/m^3, with $PM_{2.5}$ concentrations ranging between 5.8 and 22.2 µg/m^3. Measured annual average PM_{10} concentrations in South Africa ranged between 10 µg/m^3 in Brackenham, Umhlathuze and 98 µg/m^3 in Sharpville, Gauteng, in 2016. $PM_{2.5}$ concentrations are not measured as widely, but in the Highveld Priority Area the annual average $PM_{2.5}$ concentrations ranged between 11 µg/m^3 in Middelburg and 43 µg/m^3 in Secunda in 2016. In the Vaal Triangle Priority Area, annual average $PM_{2.5}$ concentrations in 2016 ranged between 24 µg/m^3 in Diepkloof and 42 µg/m^3 in Kliprivier (Khumalo, 2017).

Table 6: *Indoor PM_4 concentration and personal PM_4 exposure levels in two low-income areas on the Mpumalanga Highveld (concentrations in µg/m³; Wernecke 2018)*

Kwadela	Winter (2013, 2014)	Summer (2014, 2015)	Calculated annual average
Indoor	166	99	116
Personal	70	38	46
KwaZamokuhle	**Summer (2016)**	**Winter (2016)**	**Calculated annual average**
Indoor	65	193	97
Personal	53	161	80

Table 7: *Relative risk factors for an increase of 50 µg/m³ from Burnett et al.'s (2016) integrated-exposure response function and the relative risk factors used by Myllyvirta (2014) from the American Cancer Society study*

	Burnett et al., 2014	Myllyvirta, 2014
IHD	1.3	3.2
Stroke	1.55	1.8
COPD	1.3	1.3
Lung cancer	1.3	1.9

In more remote areas, the exposure-response function used in the Myllyvirta (2014) study is certainly applicable, but in the industrialised areas it may well not be. Two out of the five monitoring stations in the Highveld Priority Area, and all six monitoring stations in the Vaal Triangle Airshed Priority Area, have annual average concentrations which exceed the upper level of concentrations measured in the American Cancer Society study from which the relative risk functions were derived.

In each of the health risk assessments, the predicted ambient air pollutant concentration was taken to be the exposure; i.e. it was assumed that the pollutant concentrations predicted for a certain location were inhaled by the people living in that location. This is not the case in areas where people burn solid and liquid fuels in their homes for cooking or space heating, as shown by measurements collected by Wernecke (2018) in the low-income areas of Kwadela and KwaZamokuhle on the Mpumalanga Highveld (Table 6). Coal is burnt for heating in around 70% of households in Kwadela, and 60% of households in KwaZamokuhle. Personal exposure to PM_4 ranges between an average of 38 µg/m³ in summer and 161 µg/m³ in winter. A rough calculation, assuming that summer conditions prevail for nine months of the year and winter conditions for three months, suggests that annual average personal exposure to PM_4 in communities where domestic burning is prevalent ranges between 46 µg/m³ and 80 µg/m³. (While the proportion of PM_4 which is $PM_{2.5}$ is not known, measurements by Nimra et al. (2015) show that the $PM_{2.5}$:PM_4 ratio is at least 0.95).

The Myllyvirta relative risk factor for IHD of 1.26 for an increase of 10 µg/m³ in annual average $PM_{2.5}$ concentrations is similar to the IER function at lower concentrations, but they diverge sharply for a $PM_{2.5}$ concentration increment of 25 µg/m³ or more. As shown in Table 7, if the Myllyvirta relative risk factors were to be used to account for the health impacts of all sources contributing to $PM_{2.5}$ exposure levels, the health risks would be significantly over-estimated. A much more accurate estimate of health effects would be obtained by applying an IER function to quantify health risks at actual exposure levels, and then apportioning the health effects relative to the contribution that each source makes to total exposure levels.

Health benefits of coal-fired power stations: electrification

A discussion of the negative health effects of coal-fired power stations is not complete without a consideration of the health benefits brought about by electrification, mainly through decreasing the inhalation of smoke from indoor fires. Spalding-Fecher and Matibe (2003) compare the costs of the impact of air pollution on health with the health costs avoided through electrification, based on van Horen's (1996) analysis. They consider total avoided health costs for low income households electrified by 1999 (relative to power stations operating in 1999), based on reduced mortality and morbidity from air pollution from indoor coal and wood fires; reduced mortality and morbidity from accidental paraffin poisoning; reduced mortality and morbidity from fires and burns caused by paraffin and candles; and the reduced social costs of fuel wood scarcity. They calculate that the health costs avoided by electrification were R958 million in 1999 (range: R173 million – R2 324 million), while the health costs arising from air pollution from power stations was only slightly higher at R1 177 million (range: R852 million – R1 450 million).

The FRIDGE (2004) study estimates that electrification would reduce annual respiratory hospital admissions (due to exposure to PM_{10}, SO_2 and NO_2 exposure) by 7 946, and reduce mortality by 22. These benefits outweigh the health costs of coal-fired power stations calculated by this study (5 439 respiratory hospital admissions and 10 deaths a year). While the costs and health impact figures calculated in these studies are highly uncertain and likely to be underestimated, the comparison in each study is valuable because similar assumptions were used to calculate the impact of both electrification and exposure to coal-fired power station emissions.

Conclusions

A careful analysis of the assumptions and inputs used in the four health risk assessments of emissions from South African coal-fired power stations conducted to date has shown reasons for the disparities in the calculated health effects. The health impacts calculated by the FRIDGE (2004) and Airshed and Infotox (2006) health risk assessments are in all likelihood too low because of the high counterfactual concentrations used (daily PM_{10} concentration of 25 µg/m³ and daily $PM_{2.5}$ concentration of 15 µg/m³ in the FRIDGE (2004) study, and annual PM_{10} concentration of 15 µg/m³ in the Airshed and Infotox (2006) study). Health risks at lower PM concentrations are not accounted for. In the

Airshed and Infotox (2006) study, modelled PM_{10} annual average concentrations are below the counterfactual concentrations everywhere in the modelling domain, so no health effects from PM exposures are considered at all. Steyn and Kornelius (2018) restrict themselves to an assessment of SO_2 emissions and impacts in the Highveld Priority Area. The FRIDGE (2004) and Airshed and Infotox (2006) studies also only cover a limited geographical area, while the van Horen (1996) and Myllyvirta (2014) studies consider exposure throughout South Africa.

It is difficult to comment on the van Horen (1996) results, because it seems that there was an error in the transcription of the exposure-response functions.

The Myllyvitra (2014) health risk assessment of exposure to power station emissions appears to be a reasonable quantification of the health risk in remote areas, but is probably a large over-estimation of the health risk in more polluted areas, and especially in communities where personal exposure is elevated due to people inhaling smoke from domestic burning.

A fair health risk assessment of coal-fired power station emissions should offset the health costs of emissions from power stations with the health benefits from converting low-income households from solid/liquid fuels to electricity. This requires a quantification of the relationship between the electricity price and the rate of domestic solid/liquid fuel use. At a minimum, the increase in domestic electricity use, relative to domestic fuel use, achieved by keeping the electricity price low by continuing to use old coal-fired power stations (rather than replacing them with cleaner generation options) should be weighed against the health costs of coal-fired power stations.

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Research article

Spatial and Temporal Trends of PM_{10} and SO_2 in the Richards Bay Area

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Abstract

Air pollution is a public health emergency (WHO, 2016a). It is the biggest environmental risk to health with a global responsibility for about 1 in 9 deaths annually. In 2013, Statistics SA indicated that 10 % of all deaths in South Africa were attributed to respiratory diseases. Areas with increased industrial growth such as Richards Bay are particularly vulnerable. The paper analyses the spatial and temporal concentration trends of PM_{10} and SO_2 in Richards Bay over the last two decades; specifically, since the promulgation of national ambient air quality standards (NAAQS) and minimum emissions standards (MES). Mann-Kendell trend tests was applied to monitoring data from the Richards Bay Clean Air Association (RBCAA) to test for an improving or worsening trend and the significance thereof. The pollution concentration data was also compared to NAAQS and the WHO annual guidelines. Overall, results indicate that although there has been a downward trend in the concentration of PM_{10} and SO_2 emissions in the study area, the trends are not statistically significant. However, there was indication of statistically significant reducing trends in ambient concentrations at some stations. The concentrations at all stations were below NAAQS.

Keywords

air quality, trends, status, Richards Bay

Introduction

Air pollution is a public health emergency with the biggest environmental risk and carries a global responsibility for about 1 in 9 deaths annually (WHO, 2016a). In 2015, close to 10 % of all deaths in South Africa were attributed to respiratory diseases (Stats SA, 2016a).

It is known that industrial growth often results in increased emissions and with this a concern for people's health (Cohen et al., 2004; Scorgie, 2012). Research has shown that air pollution exacerbates and increases the probability of infections such as acute lower respiratory infections (ALRI) which include acute bronchitis and bronchiolitis, influenza and pneumonia. In addition, air pollution is associated with stroke, chronic obstructive pulmonary disease (COPD) including chronic bronchitis and emphysema as well as ischemic heart disease (IHD) and cancers of the trachea, bronchus and lung (Chiu, 2013; World Health Organization, 2016).

Whereas accelerated industrial growth is an important aspect to unlock poverty and improve people's livelihoods, environmental considerations are important to moderate potential adverse effects. Pollution trends in growing areas such as Richards Bay need to be monitored to provide a basis for advice and planning for future development and most importantly to protect people and the environment.

To understand the potential for burden of disease due to air pollution, and to ascertain whether policy intervention or other drivers have led to an improvement in in air quality, it is important to understand the spatial and temporal trends of air pollution. This paper focuses on analyses of two pollutants of concern PM_{10}, as well as SO_2, and their concentration over the last two decades and specifically since the promulgation of national minimum emissions standards.

Study area

Richards Bay is a growing metropolitan with a number of industries in relatively close proximity to residential areas (Okello and Allan, 2015). The city is an economic centre and one of the country's strategic economic hubs designated as an 'Area of National Economic Significance' (uMhlathuze Municipality, 2016). The city hosts the largest coal export terminal in the

world, which is the second largest port in South Africa as well as the largest aluminium and iron smelters in Africa. In addition, the City also hosts several commercial, light and heavy industrial activities such as paper, fertilizer and sugar production, which collectively contribute to most of the air quality concerns in the region. Sugarcane and forestry burning, pesticide usage and dust associated with agricultural processes are also common.

The City has a population of 364, 473 (Stats SA, 2016b) and an area of 1233.3 km^2 (Cogta, 2017). The geographical areas covered include suburbs such as Esikhaleni, Arboretum, Felixton, Mtunzini, Veld n Vlei, as well as the Central Business District (CBD) St. Lucia, Brackenham and Harbour West. These areas are representative of the entire Richards Bay and surroundings.

Air quality review

Air pollution definition and its effects

The National Environmental Management Air Quality Act (NEM: AQA, 39 of 2004) defines "air pollution" as any change in the composition of the air caused by smoke, soot, dust (including fly ash), cinders, solid particles of any kind, gases, fumes, aerosols and odorous substances. This definition is not far from the WHO, which defines air pollution as a contamination of the indoor or outdoor environment by any chemical, physical or biological agent that modifies the natural characteristics of the atmosphere (World Health Organization, 2016). Consequently, air pollution increases the risk of respiratory and heart disease in the population. Both short and long-term exposure to air pollutants has been associated to health impacts. More severe impacts affect the already ill, children, the elderly and poor people who are more susceptible (Chiu, 2013); (Craig et al., 2007); (WHO, 2013, 2016b). This is also the case in South Africa and in Richards Bay (Jaggernath, 2013).

Pollutants of major public health concern include particulate matter (PM), carbon monoxide (CO), ozone (O_3), nitrogen dioxide (NO_2) and sulphur dioxide (SO_2). While risk is depended on the exposure dose and time, the South African Department of Environmental Affairs has declared these pollutants as criteria pollutants (DEA, 2011). The health risks associated with particulate matter of less than 10 and 2.5 microns in diameter (PM_{10} and $PM_{2.5}$ respectively) is well documented (Cohen et al., 2004; Craig et al., 2008). $PM_{2.5}$ is capable of penetrating deep into lung passageways and entering the bloodstream causing cardiovascular, cerebrovascular and respiratory impacts (Rai, 2015). There is good evidence of the effects of short-term exposure to PM_{10} on respiratory health particularly irritation and acute respiratory disorders, but for mortality, and especially as a consequence of long-term exposure, $PM_{2.5}$ is a stronger indicator (WHO, 2016c).

Additionally, PM from aerospora is linked to the onset of influenza and allergic cases especially during summer and spring (Renaut and Bamford, 2004). SO_2 is known to affect the respiratory system and the functions of the lungs and causes irritation of the eyes. Consequently, the inflammation of the respiratory tract causes coughing, mucus secretion, aggravation of asthma and chronic bronchitis and makes people more prone to infections of the respiratory tract (World Health Organization, 2016). From an ecological perspective, when SO_2 combines with water, it forms sulphuric acid; this is the main component of acid rain.

In 2007, an estimated 3.7 % of national cardiopulmonary disease related deaths and 5.1 % of trachea, bronchus and lung cancers in adults older than 30 years were attributed to ambient air pollution (CSIR, 2015). In 2013, Statistics SA indicated that 10 % of all deaths were attributed to respiratory diseases.

Legislation and policy instruments for air emissions reduction

From a policy perspective, WHO global emission standards are useful to gauge pollution (WHO, 2005). South Africa too has an enabling legal framework that sets out the minimum emission standards for point source pollution as well as ambient air quality standards (DEA, 2010). The Atmospheric Pollution Prevention Act (APPA) was enacted in 1965 and was repealed on 1st April 2010 with the National Environmental Management: Air Quality Act (NEM: AQA) which was partially enacted on 9th September 2005 and fully enacted on 1 April 2010. In order to protect human health and the integrity of the environment, the national ambient air quality standards covering priority pollutants including PM_{10} and SO_2 were introduced on 24th December 2009. Similarly, industrial emission standards were developed and formally introduced on 22nd November 2012 (DEA, 2010). These were revised in 2013 (DEA, 2012).

Table 1: *Ambient PM_{10} average concentration (µg/m^3) NAAQS standard and WHO guideline adopted from (RBCAA, 2018)*

Source	Daily Average	Annual average
Initial NAAQS[a,b]	120 µg/m^3[d]	50 µg/m^3[e]
Current NAAQS [a,c]	75 µg/m^3[d]	40 µg/m^3[e]
WHO[f]	50 µg/m	20 µg/m

Notes:
[a] SA Government Gazette 32816 (24th December 2009) in terms of the National Environmental Management: Air Quality Act 39 of 2004, [b] Effective from 2010 to 2014
[c] Effective from 2015
[d] Not to be exceeded more than 4 times in one year.
[e] Not to be exceeded.
[f] World Health Organisation guideline

Until now, the influence of these policy instruments has not been adequately reviewed in the context of a reduction of the health risks that are attributable to air pollution in Richards Bay. The key question of whether these policy instruments have been adequate, and their implementation is producing the desired effect is an important aspect that needs to be investigated.

Table 2: *Ambient SO_2 average concentration (µg/m³) NAAQS standard and WHO guideline adopted from (RBCAA, 2018)*

Source	10 min Average	1 hour Average	Daily Average	Annual average
NAAQS[a]	500 µg/m³[b]	350 µg/m³[c]	125 µg/m³[d]	50 µg/m³[e]
	191 ppb[b]	134 ppb[c]	48 ppb[d]	19 ppb[e]
WHO[e]	20 µg/m³		20 µg/m³	

Notes:
[a] SA Government Gazette 32816 (published 24th December 2009) in terms of the National Environmental Management: Air Quality Act 39 of 2004 effective 2010
[b] Not to be exceeded more than 526 times in one year.
[c] Not to be exceeded more than 88 times in one year.
[d] Not to be exceeded more than 4 times a year
[e] Not to be exceeded
[e] World Health Organisation guideline

Table 1 and Table 2 represent the National Ambient Air Quality Standards (NAAQS) for PM_{10} and SO_2 respectively as the focus pollutants in this paper.

In this study, only the annual average limit of the national ambient standards was considered consistent with other national and international air quality trend studies and long-term exposure (Mohan and Kandya, 2007; Colette et al., 2011; Guerreiro et al., 2014; Butt et al., 2017). Data is however aggregated from the daily averages and thus diurnal fluctuations have been considered.

Global and local trends in air emission concentration

On a global scale, a study conducted by Butt et al.(2017), estimates that global population-weighted particulates concentrations increased by 37.5 % over the period 1960 to 2009. Increases in China and India were dominant, attributed to economic expansion, and growth in emissions. The study also concluded that in contrast, air quality regulation and emission controls in the European Union (EU), and United States (US) have reduced population-weighted $PM_{2.5}$ concentrations over the same period.

WHO compared 795 cities in 67 countries for levels of small and fine particulate matter (PM_{10} and $PM_{2.5}$) during the five-year period, 2008-2013. In the study, global urban air pollution levels increased by 8 %, despite improvements in some regions.

Despite very sparse pollution data in the Africa region, the study revealed that PM levels remained above the world median (WHO, 2016a).

In South Africa, the air quality in most urban centres is considered relatively good. However, there are a number of "hotspots" around the country where severe air quality problems are experienced (Gwaze, 2017). High ambient SO_2, NO_2 and PM_{10} concentrations are common in these areas and are primarily a result of fossil fuel burning in the residential areas as well as from industrial and power generation sources (Scorgie, 2012). The State of Air Report for South Africa indicates that PM is still the greatest national cause for concern in terms of air quality due to numerous pollution sources but climatic conditions are also a major factor (Khumalo, 2016).

Areas considered as priority include Highveld Priority Area, Vaal Triangle Priority Air-Shed, and Waterberg Bojanala Priority Area (DEA, 2018). These areas are highly polluted mainly due to anthropogenic influence. They are declared as priority areas due to heavy industrial pollution that often exceeds the NAAQS (DEA, 2018).

Despite the implementation of air pollution interventions, the air quality is said to remain poor and noncompliant with ambient standards. One could argue that the Richards Bay area may have similar characteristics to these priority areas given the number and scale of industrial operations, pollutant sources and history of emission concentration (Renaut and Bamford, 2004; Scorgie, 2012). Indeed, a study conducted in 2011-2013, focusing on perceptions of air pollution in different suburbs based on economic class concluded that air quality is a priority for the residents of Richards Bay in general and poorer areas are mostly affected by probably as a result of higher solid fuel burning compared the fuel sources in the more affluent sections of the area (Jaggernath, 2013).

Methodology

Data collection

The ambient air quality data used in these analyses was obtained from the Richards Bay Clean Air Association (RBCAA). Since 1997, the RBCAA has grown and established a real time monitoring network able to characterise the local ambient air quality by monitoring ground level pollutant concentrations (Okello and Allan, 2015). The monitoring network, operated, maintained and managed by an external service provider, comprises 11 continuous monitoring stations, recording meteorological data, SO_2, PM_{10} and Total Reduced Sulphide (TRS) concentrations.

Table 3 show the RBCAA stations and surrounding land use areas while Figure 1 shows the spatial locations of the stations.

The monitoring and data collection network is robust and well maintained, with a system in place that satisfies the South Africa National Accreditation System (SANAS) requirements (RBCAA, 2018). Furthermore, the appointment of an external service provider allows the RBCAA to remain objective and independent from the results (Okello & Allan, 2015). The RBCAA submit data to the Department of Environmental Affairs' South African Air Quality Information System (SAAQIS). This data is also submitted to the WHO Global Ambient Air Quality Database from which WHO derives the global ambient air quality concentrations.

Table 3: *RBCAA stations and surround area land use*

Codes	Station	Latitude	Longitude	Predominant Land use surrounding station
Stn1	Arboretum	-28.752704	32.066172	Residential
Stn2	Brakenham	-28.731269	32.039014	Residential
Stn3	CBD	-28.744667	32.055011	Industrial, commercial and partly residential
Stn4	Esikhaleni	-28.865253	31.911651	Residential
Stn5	Felixton	-28.831189	31.895903	Mostly agricultural, partly industrial
Stn6	Habour West	-28.787292	32.027125	Industrial
Stn7	Mtunzini	-28.956439	31.750979	Mostly residential, partly agricultural/ forestry
Stn8	Scorpio	-28.769697	32.034283	Industrial
Stn9	St Lucia	-28.377076	32.414732	Mostly residential, partly agricultural/ forestry

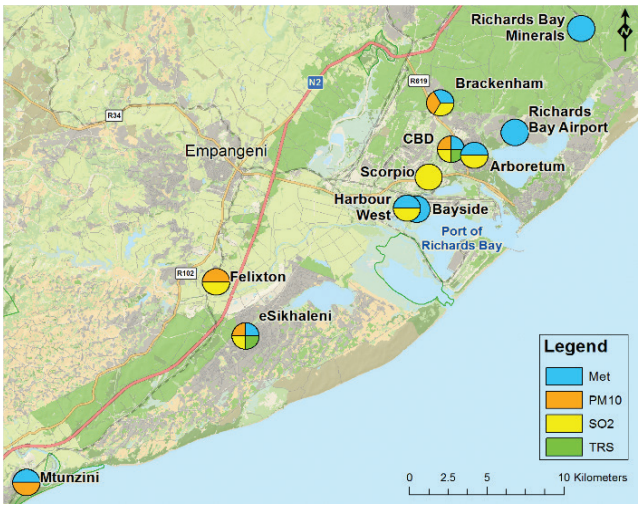

Figure 1: *RBCAA Monitoring Network (Source: RBCAA)*

Daily average data per station from 1997 to 2017 was aggregated into annual averages for the nine stations. Seven of these stations are in Richards Bay within the City of uMhlathuze municipality while two are in neighbouring municipalities (St. Lucia station in Mtubatuba and Mtunzini in uMlalazi Municipalities respectively). The St. Lucia and Mtunzini stations were considered as reference sites for PM_{10} given the predominant North Easterly and South Westerly wind directions in the area. Seven of the nine stations had PM_{10} data while six had SO_2 data.

Data availability per station was considered. Data used was filtered to include capture/ availability of over 80% as much as possible where enough data was available.

Trend analyses

Data was analysed using Mann Kendall test (MK-tests) which has been used widely to statistically analyse for trends in climatologic and in hydrologic time series (Guerreiro, Foltescu and deLeeuw, 2014; Koudahe et al., 2017). This test is used to analyse data collected over time for consistently upward or downward trends ("monotonic trends"). The advantage of the MK-tests is that it is a non-parametric test. This means it works for all distributions including for data that is not normally distributed. Also the test can be used to find trends for as few as four samples (Guerreiro, Foltescu and deLeeuw, 2014). Nonetheless, the more data points analysed the more the likelihood to find a true trend (as opposed to chance). The minimum number of recommended measurements is therefore at least 8 to 10 (Colette et al., 2011; Karmeshu, 2015; Pohlert, 2018).

According to this test, the null hypothesis H_0 assumes that there is no trend (the data is independent and randomly ordered) and this is tested against the alternative hypothesis H_1, which assumes that there is a trend. Data values are evaluated as an ordered time series. Each data value is compared with all subsequent data values considered. If a data value from a later period is higher than a data value from an earlier period, the statistic S is incremented by one. On the other hand, if the data value from a later period is lower than a data value sampled earlier, S is decremented by 1. The net result of all such increments and decrements yields the final value of S (Shahid, 2011). The MK-test statistic S is calculated using the formula below (Gilbert, 1987).

$$sgn\left(x_j - x_k\right) = \begin{cases} +1 \; if \; x_j - x_k > 0 \\ 0 \; if \quad x_j - x_k = 0 \\ -1 \; if \; x_j - x_k > 0 \end{cases} \tag{1}$$

where x_j and x_k are the annual values in years j and k, j > k, respectively.

$$S = \sum_{k=1}^{n-1} \sum_{j=k+1}^{n} sgn\left(x_j - x_k\right) \tag{2}$$

The S statistic is calculated by summing over all pairs of values:

S will be negative for a negative trend, zero for no trend, and positive for an upward trend.

For n≤10, the p value is taken from a table of exact values (Gilbert, 1987). For n>10, a normal approximation is used, that is by determining the total number of groups of ties g and the number of tied values t_j within each group, in the sorted sequence. Then the standard deviation of S is estimated using the formula:

$$SD = \sqrt{\frac{1}{18}\left[n(n-1)(2n+5) - \sum_{j-1}^{g} t_j\left(t_j - 1\right)\left(t_j + 5\right)\right]} \tag{3}$$

The z statistic is then:

$$Z = \frac{|S|-1}{SD} \quad (4)$$

The Z value is then used to calculate p from the cumulative normal distribution. If the p value is less than the significance level α (alpha) = 0.05, H_0 is rejected. Rejecting H_0 indicates that there is a trend in the time series, while accepting H_0 indicates no trend could be detected. On rejecting the null hypothesis, the result is said to be statistically significant. (Helsel and Hirsch, 2002; Mohan and Kandya, 2007).

Results and findings

Overall since 1997, for the Richards Bay area, the PM_{10} and SO_2 trends examined for significance indicated a reducing trend at 95 % significance level. However, the trends were not deemed statistically significant using the Mann-Kendall test. Figure 2 shows a spatial distribution of the stations and relative PM_{10} and SO_2 concentrations against the annual average of the NAAQS.

Particulate matter (PM_{10})

The data analysed indicates that there was a statistically downward trend in ambient PM_{10} concentrations over last 14 years. Caution was however taken due to the lower number of stations used for sampling prior to 2009. When the same data was taken from 2009 to 2017, a downward trend was observed but it was not statistically significant. In Table 4, the results indicate that null hypothesis H_0 was accepted for 5 out of 6 stations where PM_{10} data was available. This means no significant trends. A downward trend was observed in four stations (Brakenham, CBD, Esikhaleni and Felixton) (Table 4). Only the decrease at Esikhaleni was determined to be statistically significant although sampling had only been a short period of time (2014 to 2017). Mtunzini and St. Lucia, the reference site, had upward trends but was not statistically significant. The CBD and Brakenham had higher PM_{10} values compared to the other stations, although all measurements were within the stipulated NAAQS annual average limit of 50 µg/m^3.

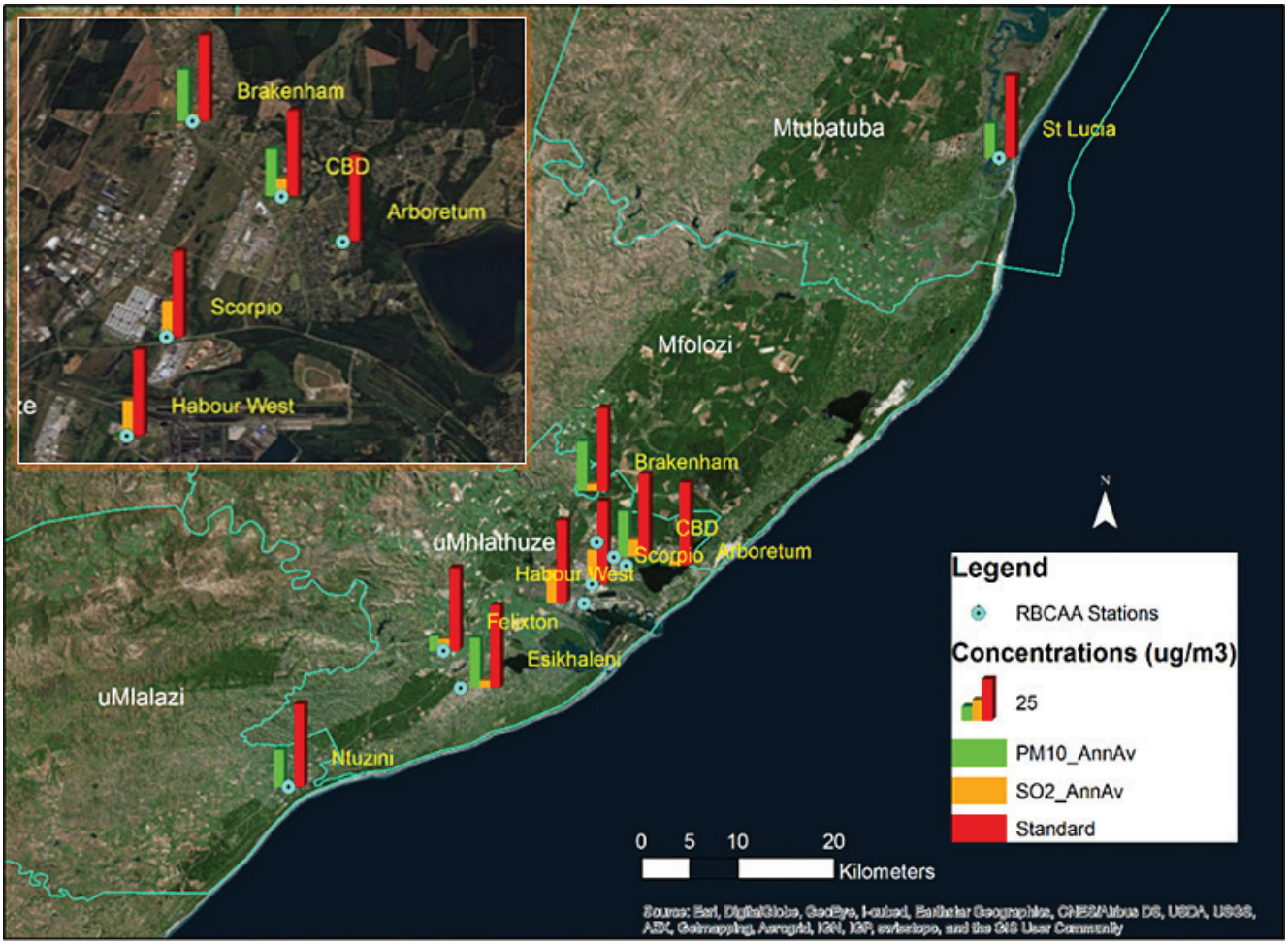

Figure 2: *Spatial distribution PM_{10} & SO_2 annual concentrations in relation to NAAQIS 50µg/m^3 annual average.*

Table 4: *Trends of annual mean concentration of PM_{10} ($\mu g/m^3$) per station in Richards Bay and in reference locations (2004-2017)*

Station/ Year	2004	2005	2006	2007	2008	2009	2010	2011	2012	2013	2014	2015	2016	2017	MK Trend Test			
															MK Statistic	Trend	Std. dev	Significance
Brakenham					32	34.4	29.9	27.0	30.7	26.6		31.8	28.5	32.2	S=-4; p=0.38	↓	2.6	NS
CBD	25	30	25	42.2	34.6	28.5	30.9	23.4	26.6	26.9	29.4	30.5	24.6	26.2	S=-12; p=0.55	↓	4.9	NS
Esikhaleni											36.3	33.0	27.8	22.1	**S=-6; p=0.042**	↓	6.2	S
Felixton									9.1	9.2	10.2	9.4	8.2	8.6	S=-3; p=0.36	↓	0.7	NS
Mtunzini							14.5	20.7	24.6	22.3	25.6	20.9	24.9	24.7	S=14; p=0.054	↑	3.7	NS
St. Lucia							17.9	20.0	22.5	21.2	22.5				S=6 ; p=0.12	↑	1.9	NS
WHO Guidelines	20	20	20	20	20	20	20	20	20	20	20	20	20	20				
NAAQS Annual Average							50	50	50	50	50	40	40	40				

KEY

Good quality	↓ Decreasing trend	S= Significant
Moderate quality	↔ Neither increasing nor decreasing	
NAAQS Annual Limit	↑ Increasing trend	NS=Not significant

Note: There were no PM_{10} measurements in Arboretum, Harbour West and Scorpio during the measurement period.

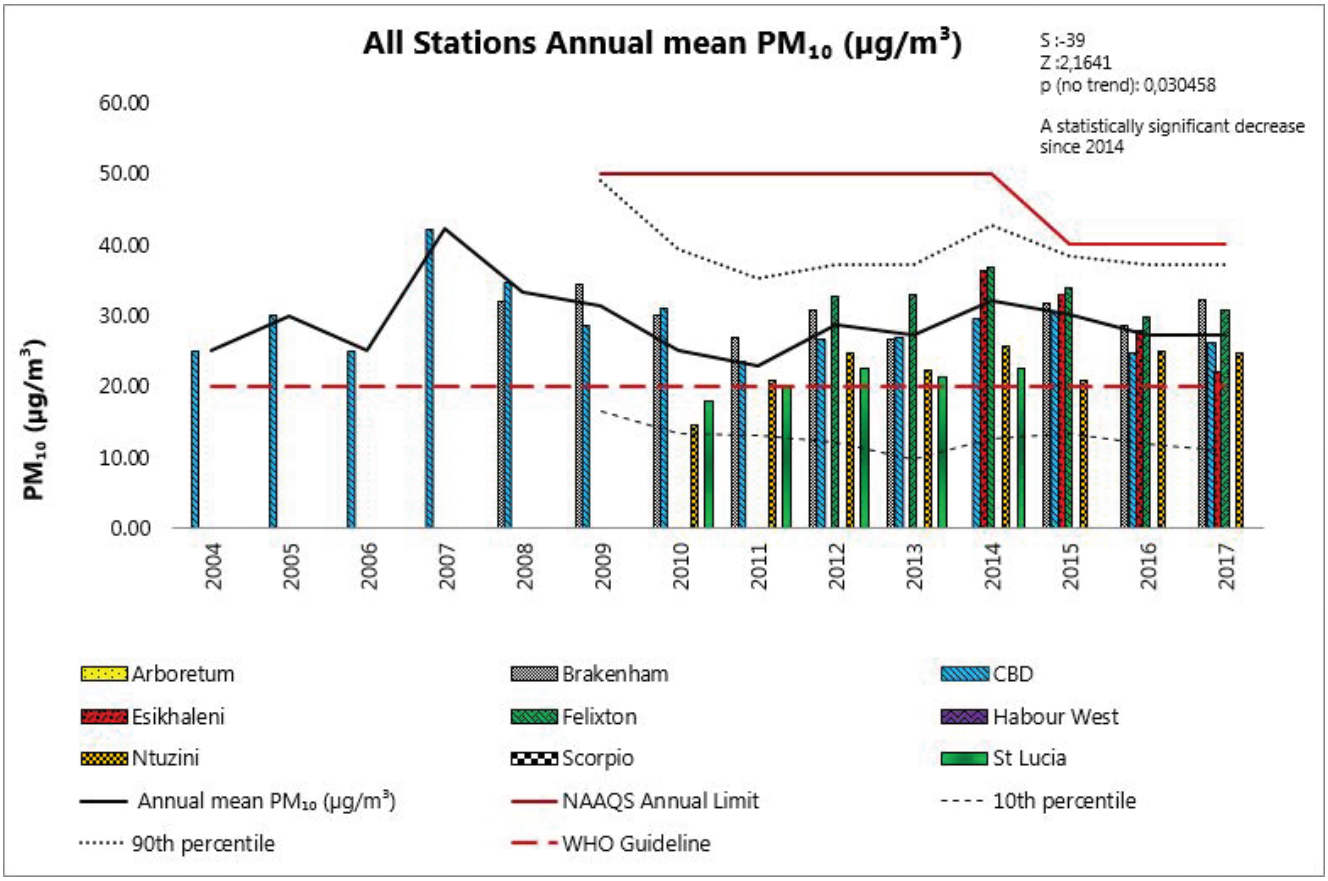

Figure 3: *Richards Bay PM_{10} status and trends (2004-2017)*

Table 5: *Trends of annual mean concentration of SO_2 ($\mu g/m^3$) per station in Richards Bay and in reference locations (1997 - 2017)*

Station/ Year	1997	1998	1999	2000	2001	2002	2003	2004	2005	2006	2007	2008	2009	2010	2011	2012	2013	2014	2015	2016	2017	MK Trend Test			
																						MK Statistic	Trend	Std. dev	Significance
Arboretum	5.0	10.5	7.9	9.2	9.7	9.2	9.5	12.1	10.3	10.5	6.8	5.5	6.5	6.7	7.8	8.4	6.6	5.5	3.6	2.4	3.2	**S=-93; p=0.005**	↓	**2.6**	S
Brakenham							8.2	10.5	11.6	8.9	5.3	3.7	2.9	3.6	6.9	3.9	2.4		6.9	3.1	2.4	**S=-49; p=0.009**	↓	**3.1**	S
CBD					13.2	15.0	13.9	17.9	15.5	15.8	13.9	11.6	11.0	10.8	10.7	8.7	9.7	14.0	11.5	6.4	11.1	**S=-67; p=0.007**	↓	**2.9**	S
Esikhaleni																		2.7	5.8	3.6	5.3	S=2; p=0.375	↑	**1.5**	NS
Felixton																9.0	6.8	8.0	7.3	4.6	4.4	**S=-11; p=0.028**	↓	**1.8**	S
Harbour west										21.1	21.3	10.3	21.6	22.6	21.3	21.4	24.7	22.1	14.9	18.5	18.0	S=0; p=0.945	↔	**3.9**	NS
Scorpio							18.2	27.9	28.4	23.9	20.0	15.0	15.5	18.9	19.6	23.4	31.4	22.9	18.9	18.5	19.4	S=-11; p=0.621	↓	**4.8**	NS
NAAQS Annual Av.														50	50	50	50	50	50	50	50				

KEY

Good quality	↓ Decreasing trend	S= Significant
Moderate quality	↔ Neither increasing nor decreasing	
NAAQS Annual Limit	↑ Increasing trend	NS=Not significant

Note: There were no SO_2 measurements in St. Lucia and Mtunzini during the measurement period.

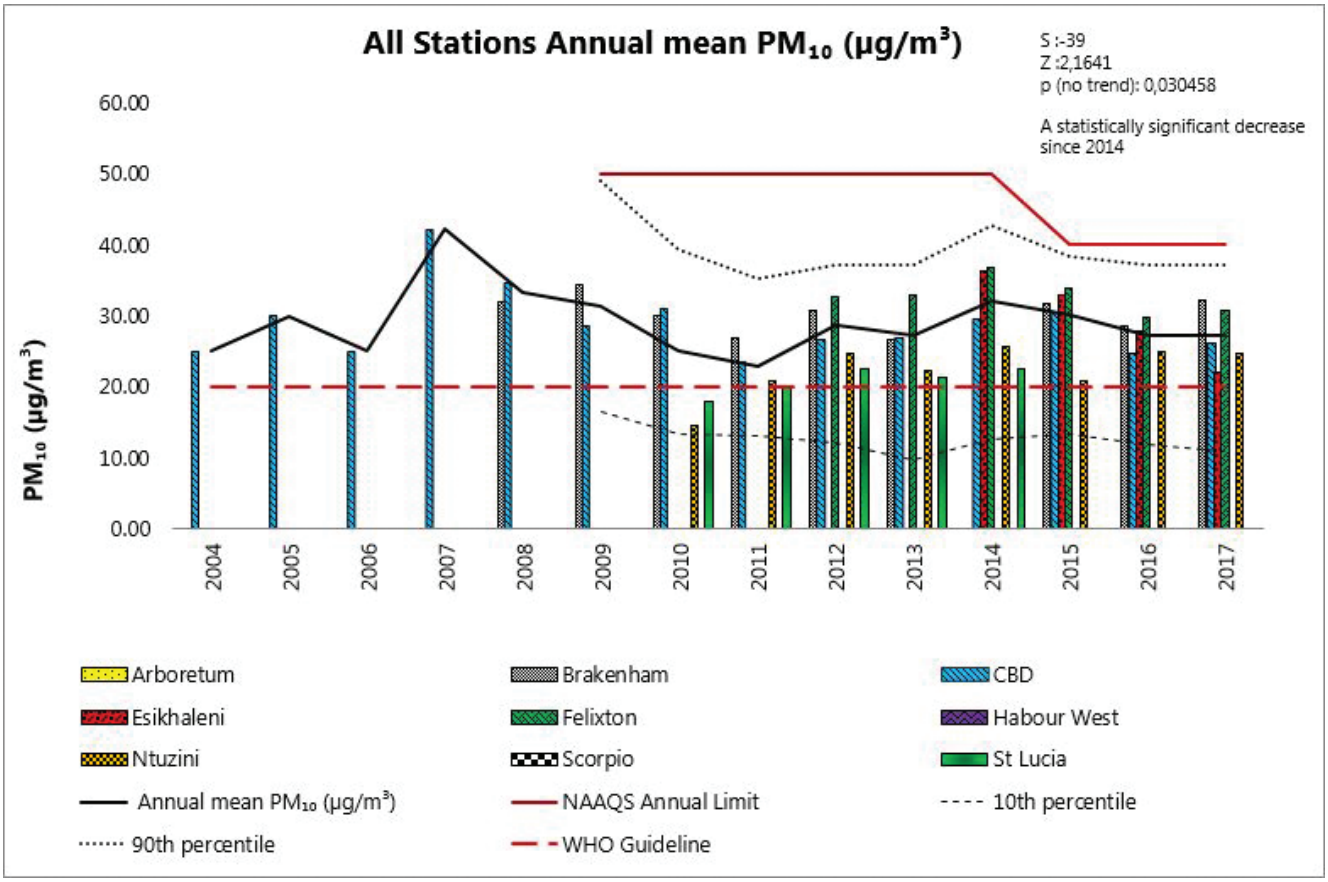

Figure 4: *Richards Bay SO_2 status and trends (1997-2017)*

Most importantly, the annual NAAQS was not exceeded in any of the years where measurements were taken. Although considering the area average, the CBD, Brakenham and Esikhaleni were consistently above the average with data predominantly within the 90th percentile (Figure 3). Esikhaleni is a highly populated area with mostly low-income households and fewer industries compared to areas around the CBD. In this area PM_{10} was likely a result of solid fuel burning. St. Lucia and Mtunzini were the reference sites with PM_{10} levels averaging at 20.8 µg/m^3 and 22.3 µg/m^3 respectively (Figure 3). This is probably a good indication of the background PM_{10} concentration of the whole study area as both sites were relatively unaffected by the main sources of PM_{10}. The background in both cases was above the WHO guideline indicating the potential contribution of other sources such as pollen and sea salts due to proximity to the sea (Renaut and Bamford, 2004).

An upward trend of PM_{10} is observed at Mtunzini although not statistically significant. This could be attributed to PM transport due to wind from pollution sources but could also be because of sea salts, forest fires and vehicles emissions etc.

Sulphur Dioxide (SO_2)

Like the PM_{10} measurements, SO_2 concentrations in all seven stations where data was available were within the stipulated annual limit of 50 µg/m^3 (Table 5). Statistically significant downward trends were observed in Arboretum, Brakenham, CBD and Felixton. Harbour west neither had an upward nor a downward trend. Esikhaleni showed an upward trend although with ambient concentrations well below the annual limit value. Scorpio had the least favourable SO_2 trends attributable to their close vicinity to industry while Arboretum, Brakenham, Esikhaleni and Felixton looked better in terms of the SO_2 profile (Table 5).

Data taken over the long term (1997 to 2017) for SO_2 indicate an upward trend although not statistically significant. From 2013 to 2017 however, a statistically significant downward trend is observed (Figure 4). The Scorpio and Harbour West Stations have consistently been above the 20-year average with annual average ambient concentration within then the 90th percentile. This can be attributed mostly to emissions from the surrounding industries. The CBD had SO_2 annual average ambient concentration just below the 20-year regional annual average. Measurement from residential areas such as Arboretum, Mtunzini and Esikhaleni showed low concentrations of SO_2. Importantly, the annual NAAQS for SO_2 of 50 µg/m^3 was not exceeded in all stations.

Practical managerial implications and recommendations

The results obtained indicate a generally downward trend of ambient PM_{10} and SO_2 concentrations in Richards Bay over the last two decades and particularly in the recent 5 years. This trend could be a result of the introduction of NAAQS in 2009 as is seen with the step changes in trends when these standards were issued. It could also be a result of the implementation of minimum emissions standards for listed activities introduced in 2010 with achievement milestones stipulated for 2015 and 2020. The King Cetshwayo District Municipality Air Quality Management Unit and the local uMhlathuze Municipality have incorporated these standards into the Air Emission Licenses (AEL) and trade permits of industries. It can be argued that these regulatory changes have pushed emission sources particularly industry to invest more on abatement equipment.

Another possible factor contributing to the reduction in emission concentration could be the closure of some big industries particularly, Bayside Aluminium, Tata Steel and recently Lafarge that were near the Central Business District. It is worth noting that increase in SO_2 and PM_{10} concentration levels around 2008, 2009 coincide with introduction of industries for example Tata Steel. Also, the Bayside Aluminium and Tata steel closures coincide with the sharp decrease in SO_2 post 2014.

More so, an important contributor to the reducing trend could be pressure on industrial emission sources from NGOs such as the RBCAA that has handled numerous complaints from the public and tried to apportion emission to different sources through their stations and case studies. The RBCAA operates on a polluter pays principle with industry involvement who form part of the management committee (Okello and Allan, 2015). The RBCAA reviews Environmental Impact Assessments for new developments in the area that are likely to alter emission concentration in the air-shed and advises on potential mitigation options. In the words of Jim Phelps, formerly of the Zululand Environmental Alliance (ZEAL) “the RBCAA has done its best to work with polluting industries while raising serious concerns about air pollution in Richards Bay for the sake of the community and environmental health”. Its role thus cannot be under estimated.

The downward trend in ambient concentrations is also mirrored in the number of air quality related complaints received through the RBCAA and the City of uMhlathuze’s air quality unit which peaked in 2004 and have since showed a downward trend with 2017 having the least number of complaints (RBCAA, 2018).

Caution should be taken however, as Richards Bay is rapidly urbanising with new industrial development underway through the Richards Bay Industrial Development Zones (RBIDZ). The population is also rapidly increasing and with that, more vehicular and other activities that are likely to add to the PM_{10} and SO_2 concentration in the ambient air. Monitoring efforts should thus be continued and implemented in a more collaborative and efficient manner to ensure that emission sources are reduced for the benefit of all living in the area.

Conclusion

Based on the results analysed, the ambient trends for PM_{10} and SO_2 over the last two decades in Richards Bay and surrounding areas are generally decreasing. This is a positive aspect for

the region and could be an indication that efforts to reduce emissions by imposing of the national minimum emission standards especially on surrounding industries is achieving the desired effect. However, the trends were not deemed statistically significant using the Mann-Kendall test. Although the overall decrease is not statistically significant, significant downward SO_2 trends were observed in Arboretum, Brakenham, and CBD. For PM_{10}, downward trends were observed in Brakenham, CBD and Felixton with Esikhaleni having the only statistically significant downward trend.

This paper thus concludes that efforts to reduce air emissions, particularly particulate matter in Richards Bay and surrounding areas are bearing fruit. Industrial abatement and collaborative work through involvement of different stakeholders in forums such as the RBCAA needs to continue. Government intervention through air emission licensing and trade permits and complaints handling is also critical to ensure that pollution reduction trends are sustained to a significant level.

It is worth noting that the measured background PM10 in the reference sites of St. Lucia and Mtunzini (20.8 µg/m^3 and 22.3 µg/m^3 respectively) are in the range of the WHO guideline. These sites were generally not affected by industrial sources and thus could be an indication of other pollution sources mainly sea salts and probably sugar cane burning.

Recommendation

It is recommended that the current monitoring of pollution concentrations continue to further improve the database of air quality in the area. Additionally, current efforts by the RBCAA to monitor Total Reduced Sulphur compounds (TRS) should be continued and supported. Measurements should also include other priority pollutants such as nitrogen dioxide, ozone, benzene, and carbon monoxide where necessary to ascertain baseline and need for continuous monitoring. These additional measurements would present a better picture of air quality pollution in the area. Incoming industries with these priority pollutants as emissions should be encouraged to be in involved in pollution monitoring. More so, recent government involvement in ambient air monitoring should be encouraged and perhaps even better consider synergy with existing monitoring networks such as the RBCAA to ensure better utilisation of resources and consistent monitoring.

Although the focus of this paper was on the trends of air quality pollutants, these results are important and could be applied in studying health impacts associated with long term air quality pollution and in interpreting health risk data. The results are also an important indicator of policy efficacy and a reminder that pollution reduction is possible if policy implantation is effective.

Acknowledgments

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