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

Gregor Feig1,2,4*, Rebecca M. Garland1,2,3, Seneca Naidoo1, Amukelani Maluleke1, Marna van der Merwe1
1Council for Scientific and Industrial Research, Pretoria, South Africa
2Department of Geography, Geoinformatics and Meteorology, University of Pretoria, South Africa
3Unit for Environmental Sciences and Management, North West University, South Africa
4South African Environmental Observation Network (SAEON), Pretoria, South Africa
*gregor@saeon.ac.za

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

Abstract
Ambient air pollution has important impacts on a variety of environmental issues, particularly on human health and ecosystem processes. A key tool for understanding the impacts of atmospheric pollution is through the long-term measurement of the ambient concentrations of criteria atmospheric pollutants. Monitoring of ambient pollution concentrations has been conducted in two of the National Air Quality Priority Areas since 2007. During this time period, significant changes in the management of air pollution have occurred, including the adoption of the ambient air quality standards, and the implementation of Section 21 emission standards. This paper examines the long-term evolution of ambient concentrations for PM and SO2 in the Vaal Triangle Airshed Priority Area and Highveld Priority Area. These trends will be evaluated against the implementation of management interventions and the variation in the measured concentrations and emerging areas of concern are highlighted.

Keywords
Particulate matter, SO2, Priority areas, air quality trends, Vaal Triangle Priority Area, Highveld Priority Area, Theil-Sen.

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

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

While this legislation has been enacted, certain areas have been identified as being of particular concern and have been declared as air quality priority areas, these include: the Vaal Triangle Airshed Priority Area (VTAPA; DEAT, 2006), the Highveld Priority Area (HPA;DEAT, 2007) and the Waterberg-Bejanala Priority Area (DEA, 2012a). Within these areas, particular concern has been placed in the development and review of management plans, including the VTAPA in 2009, the HPA in 2012 and the Waterberg in 2013 (DEA, 2015c).

Beyond the scope of changing legislation, there have been changes in the emissions profiles within the priority areas that have had significant impacts on emission profiles in the priority areas (Pretorius et al., 2015). These include:
The increase in the number of vehicles in the country has contributed to changes in the emissions profiles, including an increase in vehicle emissions. Between January 2009 and June 2018, South Africa has seen an increase in motor vehicles, rising from ~9 to ~12 million. Notable increases have been seen in the Gauteng and Mpumalanga provinces (where VTAPA and HPA are located), with increases from ~3.5 to ~4.7 million in Gauteng, and ~600,000 to ~900,000 motor vehicles in Mpumalanga have been recorded (ENATIS, 2018).

The closure of Highveld Steel in February 2015 (Goldswain, 2010), which resulted in a decrease in industrial emissions in the HPA.

The large scale domestic electrification programmes in the priority areas which resulted in the rapid electrification, across South Africa more than 5 million households received access to electricity between 1990 and 2007 in South Africa (Bekker et al., 2008) which resulted in a reduction in household emissions but an increase in electricity consumption and associated emissions at the power generation plants.

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

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

Methods and Materials

Data for the 11 monitoring stations in the VTAPA and HPA priority area ambient air quality monitoring networks were requested from SAAQIS at an hourly temporal resolution (Table 1). The data were provided in a quality controlled form, but a subsequent quality control of the data was conducted to remove a limited number of negative concentration values and the removal of a significant number of values below the detection limit of the instruments (typically zero values) and other anomalous measurements. The data were analysed using the R language for statistical computing (R Core Team, 2013), specifically using the Open Air Package (Carslaw and Ropkins, 2012). The trends in the concentrations of PM$_{10}$, PM$_{2.5}$, and SO$_2$ were calculated for each station for the period of available data using the Theil-Sen trend analysis following a deseasonalisation step as recommended, which uses the Loess method. The Theil-Sen Estimator approach uses monthly averages, for which an 80% data availability threshold was set. A comparison of the trends in the continuously monitored SO$_2$ concentrations can be made with a dataset of measurements conducted by the CSIR extending from 1959-1968 (Kemeny and Halliday, 1972; Kemeny, 1980; Kemeny and Vleggaar, 1983; Walker, Ellerbeck and Kemeny, 1986; Walker, Galpin and Pienaar, 1987). The historical CSIR measurements were made using the Hydrogen Peroxide Method, which consisted of passing the air volume through a dilute solution of hydrogen peroxide and measuring the change in the pH of the solution through titration with a sodium borate solution (Kemeny, 1980). Comparison with these historical results serves as a valuable benchmark as to how the ambient concentrations of SO$_2$ have changed in the last 60 years.

Results

PM$_{10}$

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

The monthly PM$_{10}$ concentrations have shown a general decrease at all the sites in the VTAPA and HPA (Figure 2), except for Sharpeville (which shows a large increase in 2016), Kliprivier and Three Rivers. This decrease has been significant at the p < 0.001 confidence interval at Diepkoof (-1.56 µg/m$^3$/year), Sebokeng (-1.37 µg/m$^3$/year), Zamdela (-4.68 µg/m$^3$/year), Hendrina (-1.97 µg/m$^3$/year), Middelburg (-3.54 µg/m$^3$/year) and Secunda (-4.35 µg/m$^3$/year). At the p <0.01 confidence level Ermelo shows a trend of -1.22 µg/m$^3$/year and at the p<0.05 level, Sharpeville shows a decreasing trend of -1.16 µg/m$^3$/year (the strength of the trend is negatively influenced by the 2016 value). The Witbank site shows a decrease but this is not significant, while the Kliprivier site shows a non-significant increase. The Three Rivers site shows an increase of 0.99 µg/m$^3$/year at a p<0.05 confidence interval.
Table 1: Monitoring site Location and dominant emission sources

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

Figure 1: Annual average PM$_{10}$ concentrations for the VTAPA and HPA (the black dot represents the median, the red horizontal line represents the historical (2009-2014) annual standard of 50µg/m$^3$, the orange line represents the current (2015-present) annual standard of 40µg/m$^3$)
Using the trend in the change in PM$_{10}$ concentrations over the measurement period, a simplistic assessment was made of how long it would take for each of the monitoring stations to reach compliance with the national PM$_{10}$ standard based on the annual average concentration for the last full year with data (Table 2), it is acknowledged that this is an overly simplistic approach as it is not expected that the ambient concentrations of PM$_{10}$ will follow a continuous linear trend. However, this approach is illustrative as to how long it may take to get to compliance following the long term current trend. Currently three stations are in compliance with the PM$_{10}$ annual standard for the remainder it is (simplistically) expected that compliance will be reached between 2018 for Secunda and 2065 for Sharpeville. In the Sharpeville case, the annual concentration for 2016 (57.2 µg/m³) was considerably higher than for 2015 (22.78 µg/m³) in this case if the 2015 annual value is used with a continuation of the historical trend, compliance is expected by 2035.

PM$_{2.5}$

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

Monthly PM$_{2.5}$ concentrations show a decreasing trend for all the sites, except for Zamdela (a non-significant increase in PM$_{2.5}$) (Figure 4). A decreasing concentration at a confidence interval $p \leq 0.001$ was found for: Diepkloof (-2.33 µg/m³/year), Middelburg

![Figure 2: Thiel-Sen Trend analysis for monthly PM$_{10}$ at the VTAPA and HPA air quality monitoring sites. *** indicates significance at the $p=0.001$ confidence level, ** indicates significance at the $p=0.01$ confidence level and * indicates significance at the $p=0.05$ confidence level.](image-url)
## Table 2: Summary of trends in PM$_{10}$ concentrations in the VTAPA and HPA (NS = not significant)

| Site         | Priority Area | Average annual PM$_{10}$ Concentration (µg/m$^3$), most recent full year | Year of last measurement | Dif erence from NAAQS | Rate µg/m$^3$/year | Significance | Years to compliance with current standard | Year of compliance with current standard |
|--------------|---------------|-------------------------------------------------------------------------|--------------------------|-----------------------|--------------------|--------------|------------------------------------------|-------------------------------------------|
| Diepkloof    | VTAPA         | 31.99                                                                   | 2016                     | -8.01                 | -1.56              | 0.999        | 0                                         | In compliance                             |
| Ermelo       | HPA           | 51.73                                                                   | 2015                     | 11.73                 | -1.22              | 0.99         | 9.6                                      | 2026                                      |
| Hendrina     | HPA           | 24.43                                                                   | 2016                     | -15.57                | -1.97              | 0.999        | 0                                         | In compliance                             |
| Kliprivier   | VTAPA         | 51.88                                                                   | 2014                     | 11.88                 | 0.1                | NS           | No Trend                                  |                                           |
| Middelburg   | HPA           | 21.90                                                                   | 2016                     | -18.1                 | -3.54              | 0.999        | 0                                         | In compliance                             |
| Sebokeng     | VTPA          | 43.97                                                                   | 2016                     | 3.97                  | -1.37              | 0.999        | 2.9                                      | 2019                                      |
| Secunda      | HPA           | 53.82                                                                   | 2014                     | 13.82                 | -4.35              | 0.999        | 3.2                                      | 2018                                      |
| Sharpewille  | VTAPA         | 97.2                                                                    | 2016                     | 57.2                  | -1.16              | 0.95         | 49                                       | 2065                                      |
| Three Rivers | VTAPA         | 62.78                                                                   | 2015                     | 22.78                 | -1.16              | 0.95         | 19.6                                    | 2035                                      |
| Witbank      | HPA           | 53.35                                                                   | 2016                     | 13.35                 | -0.06              | NS           | No Trend                                  |                                           |
| Zamdela      | VTAPA         | 65.72                                                                   | 2016                     | 25.72                 | -4.68              | 0.999        | 5.5                                      | 2022                                      |

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

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

## Table 4: Summary of trends in SO$_2$ concentration in the VTAPA and HPA (NS = not significant)

| Site         | Average annual SO$_2$ Concentration (ppb) | Year of last measurement | Dif erence from NAAQS | Rate ppb/year | Significance | State of compliance with current standard |
|--------------|------------------------------------------|--------------------------|-----------------------|---------------|--------------|------------------------------------------|
| Diepkloof    | 4.18                                     | 2016                     | -14.82                | -0.3          | 0.999        | In compliance                            |
| Ermelo       | 10.76                                    | 2016                     | -8.42                 | -0.52         | 0.999        | In compliance                            |
| Hendrina     | 9.5                                      | 2016                     | -8.5                  | -0.73         | 0.999        | In compliance                            |
| Kliprivier   | 5.17                                     | 2014                     | -13.83                | 0.01          | NS           | In compliance, no trend                   |
| Middelburg   | 5.77                                     | 2015                     | -13.23                | -0.52         | 0.999        | In compliance                            |
| Sebokeng     | 4.96                                     | 2015                     | -14.04                | 0.08          | NS           | In compliance, no trend                   |
| Secunda      | 6.83                                     | 2016                     | -12.17                | -0.23         | NS           | In compliance, no trend                   |
| Sharpewille  | 5.79                                     | 2016                     | -13.21                | -0.1          | NS           | In compliance, no trend                   |
| Three Rivers | 5.93                                     | 2016                     | -13.07                | +0.08         | 0.95         | In compliance                            |
| Witbank      | 19.29                                    | 2014                     | +0.29                 | -0.21         | NS           | Out of compliance, no trend               |
| Zamdela      | 8.47                                     | 2016                     | -10.53                | +0.03         | NS           | In compliance, no trend                   |
(-1.88 µg/m³/year), Secunda (-1.84 µg/m³/year), Ermelo (-1.75 µg/m³/year), Sebokeng (-1.61 µg/m³/year), Witbank (-1.12 µg/m³/year), and Hendrina (-1 µg/m³/year). A decreasing trend with a confidence interval of p <0.01 was found for Three Rivers (-0.36 µg/m³/year), while Kliprivier showed a decreasing trend (-0.72 µg/m³/year) at the p<0.05 level. Sharpeville and Zamdela did not show a statistically significant trend in the PM$_{2.5}$ concentrations.

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

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

(-1.88 µg/m³/year), Secunda (-1.84 µg/m³/year), Ermelo (-1.75 µg/m³/year), Sebokeng (-1.61 µg/m³/year), Witbank (-1.12 µg/m³/year), and Hendrina (-1 µg/m³/year). A decreasing trend with a confidence interval of p <0.01 was found for Three Rivers (-0.36 µg/m³/year), while Kliprivier showed a decreasing trend (-0.72 µg/m³/year) at the p<0.05 level. Sharpeville and Zamdela did not show a statistically significant trend in the PM$_{2.5}$ concentrations.
Using a 75% data capture requirement, it is only the Witbank site that exceeds the annual \( \text{SO}_2 \) standard within the priority areas (Figure 5). \( \text{SO}_2 \) has not proven to be out of compliance with the NAAQS.

In comparison to the PM measurement where there was a significant decreasing trend in almost all the monitoring sites, the trends in \( \text{SO}_2 \) concentrations were only significant at five of the 11 sites (Figure 6). A significant decrease in \( \text{SO}_2 \) concentrations at the \( p<0.001 \) level was found at Hendrina (-0.73 ppb/year), Ermelo (-0.52 ppb/year), Middelburg (-0.52 ppb/year), and Diepkloof (-0.3 ppb/year). At the Three Rivers site an increasing trend of 0.08 ppb/year in the \( \text{SO}_2 \) concentration was observed at the \( p<0.05 \) confidence level. No significant trends were observed at the other sites (Table 4).

### Historical \( \text{SO}_2 \) Concentrations

Long term historical data are not available for many of the sites under consideration in this study, however the measured \( \text{SO}_2 \) concentrations reported for the major metros in South Africa during the 1960s show that historically the ambient concentrations of \( \text{SO}_2 \) were considerably higher than is currently recorded. Within Johannesburg, Durban and East London annual average \( \text{SO}_2 \) concentrations above 20 ppb were common (Figure 7).
Discussion

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

As stated by the National Air Quality Officer, in previous State of Air Reports, the compliance with the PM$_{10}$ and PM$_{2.5}$ standards is a significant problem over the Vaal and Highveld regions with almost all of the monitoring sites being non-compliant with the current NAAQS this is well known and has been previously extensively documented (Venter et al., 2012; Hersey et al., 2015; Wernecke et al., 2015; Feig, et al, 2016; Garland et al., 2017). However, annual SO$_2$ is generally (with the exception of Witbank) considerably lower than the NAAQS. These results are similar to those reported by Feig et al, (2016) for the Waterberg priority area and Venter et al. (2012), for the Western Bushveld Igneous complex, which identified the major sources of SO$_2$ as high stack emissions and PM$_{10}$ from domestic combustion emissions. Across the Highveld annual average concentrations of SO$_2$ were reported as being below the NAAQS (Lourens et al.,...
Figure 6: Thiel-Sen trend in $SO_2$ concentration (ppb) over the VTAPA and HPA. *** indicates significance at the p<0.001 confidence level, ** indicates significance at the p<0.01 confidence level and * indicates significance at the p<0.05 confidence level.

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

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

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

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

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

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

In contrast to the decreasing trend in particulate matter, there is little to no trend in the concentrations of SO$_2$ over the VTAPA and HPA monitoring stations, where a significant increasing trend was found in five of the 11 sites. Four of the sites showed a negative trend in SO$_2$ (Diepkloof, Ermelo, Hendrina and Middelburg) which ranged between -7.3 ppb/year for Hendrina to -0.3ppb/year for Diepkloof. Supporting the previous findings, the ambient concentrations of SO$_2$ at the annual averaging period are in compliance with the NAAQS (Lourens et al., 2011; Venter et al., 2012; Wernecke et al., 2015).

Long term historical measurements at some of the major South African cities in 1960s to 1980s show a decreasing trend in SO$_2$ concentration during the 1960s and 1970s however it levelled out during the 1980s at the end of the measurement time series (Kemeny and Halliday, 1972; Kemeny, 1980; Kemeny and Vleggaar, 1983). During the time period between the observations reported in the Kemeny papers and the beginning of the observations in the VTAPA and HPA reported here there has been a considerable decrease in the ambient
concentrations, once again highlighting the importance of continuous observation record.

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

Conclusion
Despite the existence of the current air quality management regime for a period of 15 years (since the promulgation of the NEM:AQA in 2004) the air quality in the VTAPA and HPA is still considered to be poor and these areas are out of compliance with the PM$_{2.5}$ and PM$_{10}$ NAAQS. However, in most instances over the monitoring time period, the ambient concentrations of particulate matter are improving and in some cases are improving fairly rapidly. These trends are not as evident for SO$_2$ concentrations, however, in contrast to PM, there is only one station where ambient SO$_2$ concentrations exceeded annual NAAQS.

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

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

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

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

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

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

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

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

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

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

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

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

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

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

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

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

DEA (2015a) Amendments to National List of Activities which result in atmospheric emission which have or may have a significant detrimental effect on the environment, including health, social conditions, economic conditions ecological conditions or cultural heritage.
DEA (2015b) Declaration of small-scale char and small-scale charcoal plants as controlled emitters and establishment of emission standards.

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

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

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

DEAT (2007) Declaration of the Highveld as a priority area in terms of section 18 of the National Environmental Management: Air Quality Act (3/2004).

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

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

Feig, G. T. et al. (2015) ‘Measurement of atmospheric black carbon in the Vaal Triangle and highveld priority areas’, Clean Air Journal - Tydskrif vir Skoon Lug, 25(1), pp. 46-50. Available at: http://reference.sabinet.co.za/webx/access/electronic_journals/cleanair/cleanair_v25_n1_a14.pdf?%5Cnhttp://dx.doi.org/10.17159/2410-972X/2015/v25n1a4.

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

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

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

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

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

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

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

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

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

Pretorius, I. et al. (2015) ‘A perspective on South African coal fired power station emissions’, Journal of Energy in Southern Africa, 26(3), pp. 27-40. Available at: http://www.scielo.org.za/scielo.php?script=sci_arttext&pid=S1021-447X2015000300046

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

Sangeetha, S. K. et al. (2017) ‘SO2 seasonal variation and assessment of Ozone Monitoring Instrument (OMI) measurements at Sharpeville (27.86°E; 26.68°S) a South African ground-based station’, International Journal of Remote Sensing, Taylor & Francis, 38(23), pp. 6680-6696. doi: 10.1080/01431161.2017.1363433.

Sangeetha, S. K. and Sivakumar, V. (2019) ‘Long-term temporal and spatial analysis of SO2 over Gauteng and Mpumalanga monitoring sites of South Africa’, Journal of Atmospheric and Solar-Terrestrial Physics. Elsevier Ltd, 191(March), p. 105044. doi: 10.1016/j.jastp.2019.05.008.

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

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

Tshehla, C. and Wright, C. Y. (2019) ‘15 Years after the National Environmental Management Air Quality Act: Is legislation failing to reduce air pollution in South Africa?’, South African Journal of Science, 115(9), pp. 2-5. https://doi.org/10.17159/sajs.2019/6100
Research article: Changes in criteria pollutants in the Vaal and Highveld priority areas

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

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

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

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