Research article

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

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Abstract

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

Keywords

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

Introduction

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

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

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

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

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

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

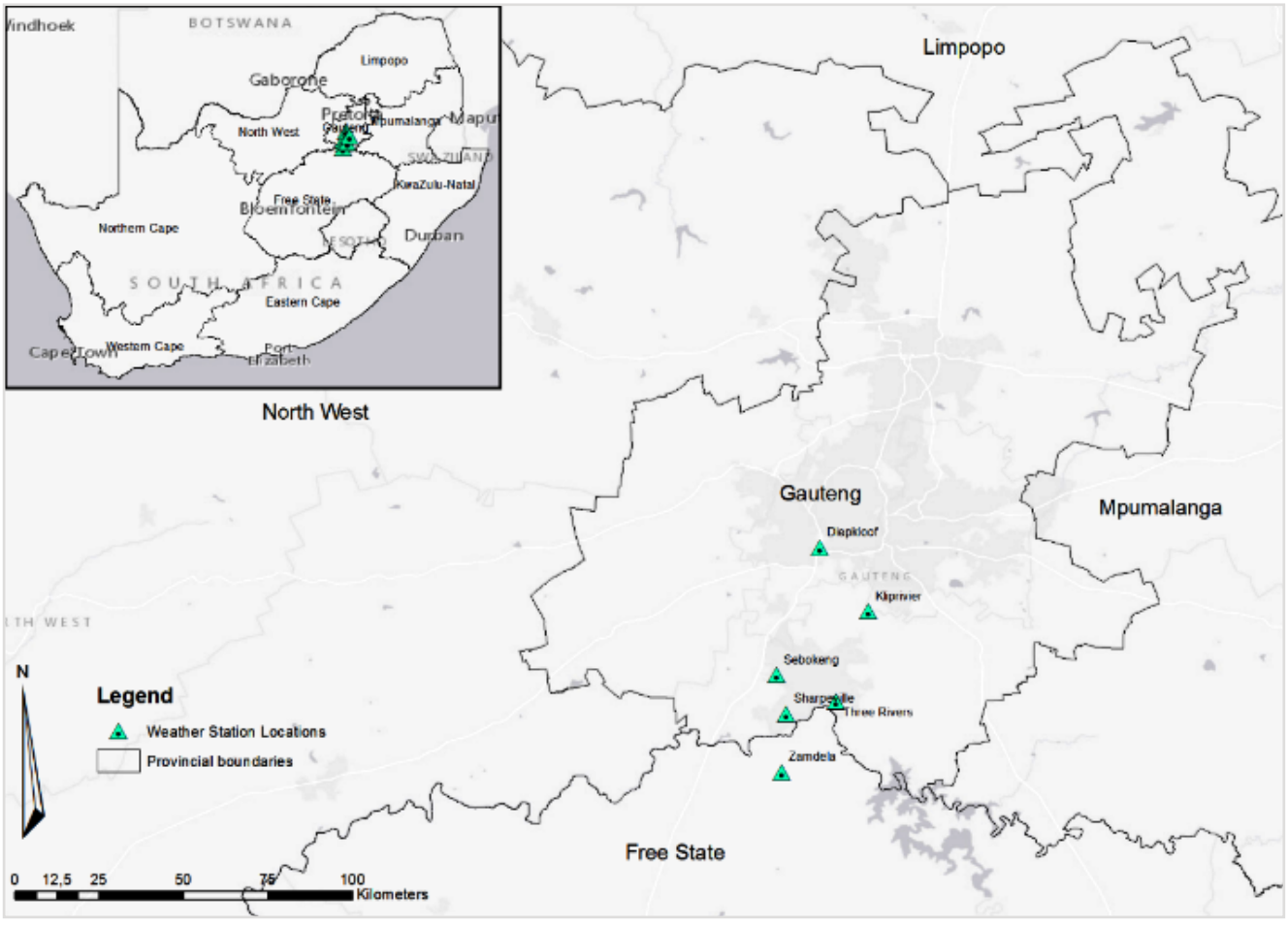

Figure 1: *Geographical map of the selected six air quality monitoring stations in the Vaal Triangle Airshed Priority Area of South Africa.*

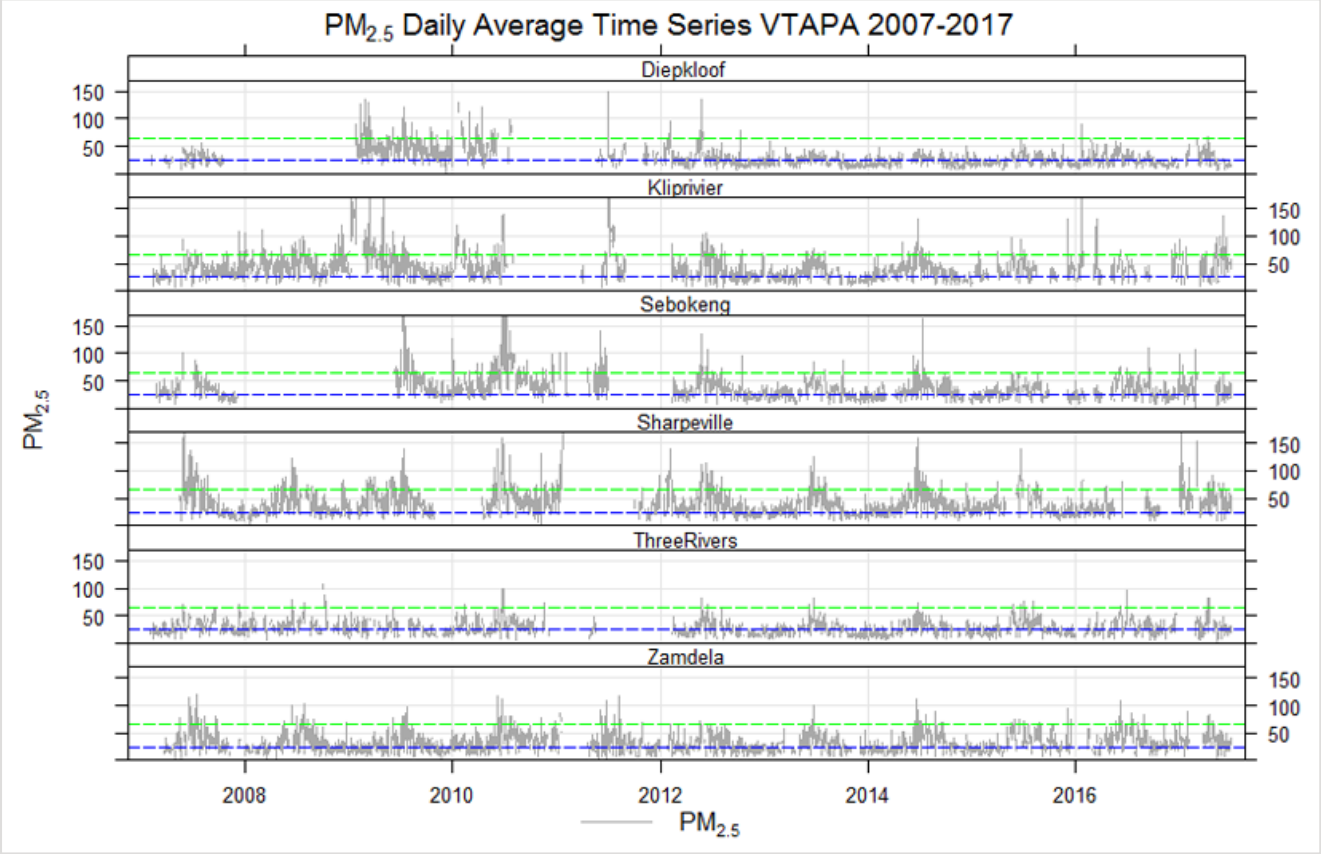

Figure 2: *Daily Average of $PM_{2.5}$ concentration of hourly data for February 2007 to June 2017*

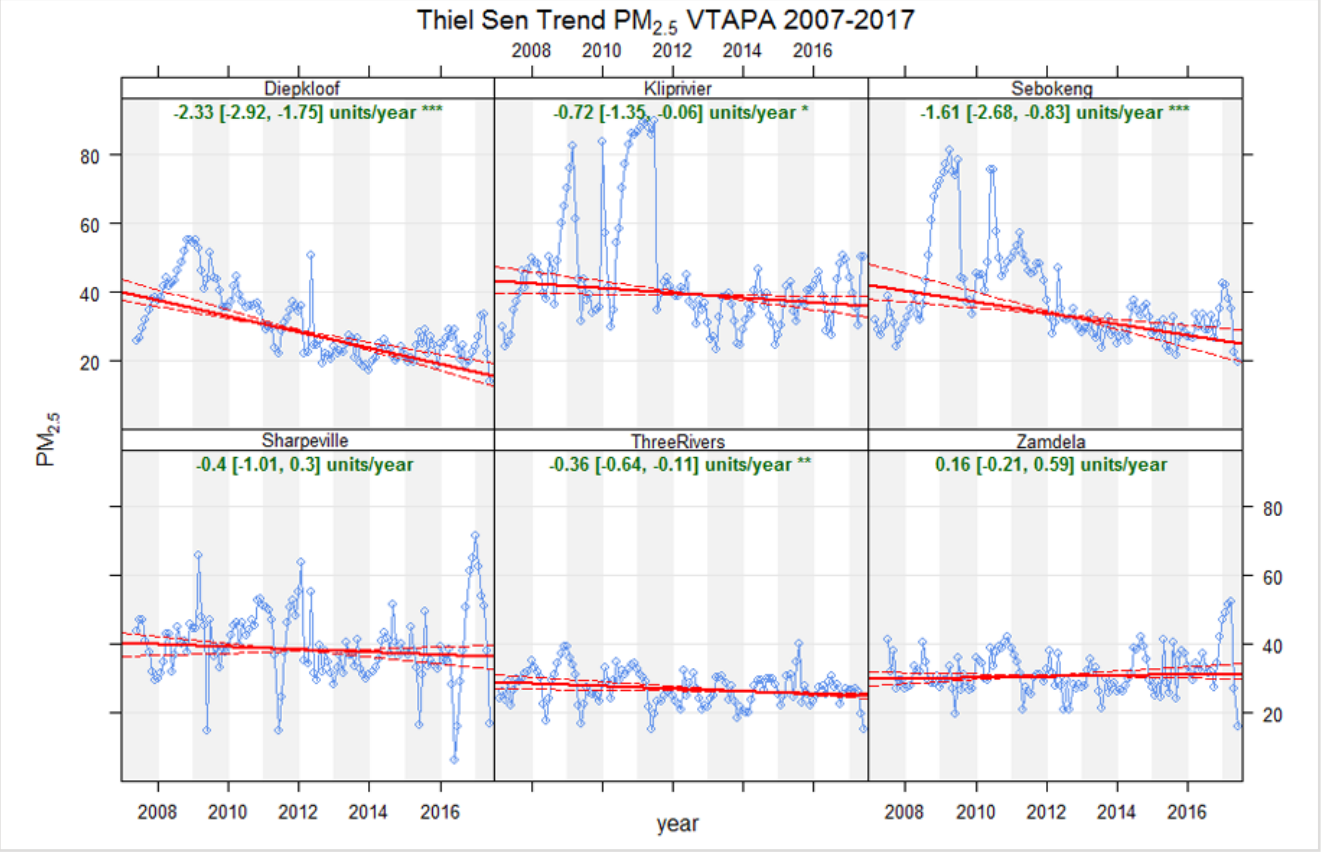

Figure 3: *Mann Kendall and Theil-Sen trend analysis for $PM_{2.5}$ concentrations for 2007-2017*

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

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

Study site

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

Table 1: *Air quality monitoring stations' location*

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

Data and Methodology

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

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

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

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

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

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

Results and discussion

Particulate Matter ($PM_{2.5}$)

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

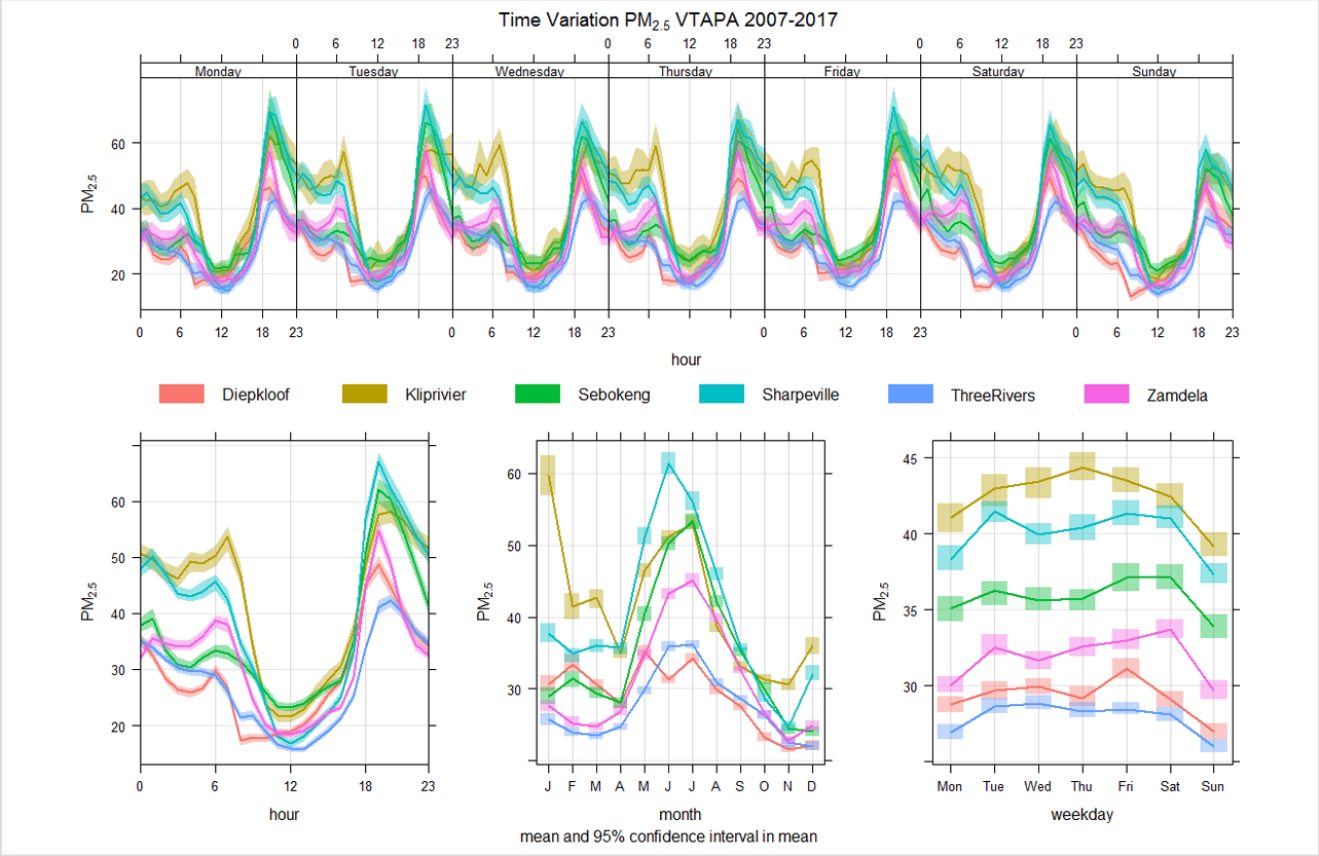

Figure 4: *Time variation plot for daily average of $PM_{2.5}$ concentration of hourly data for February 2007 to June 2017*

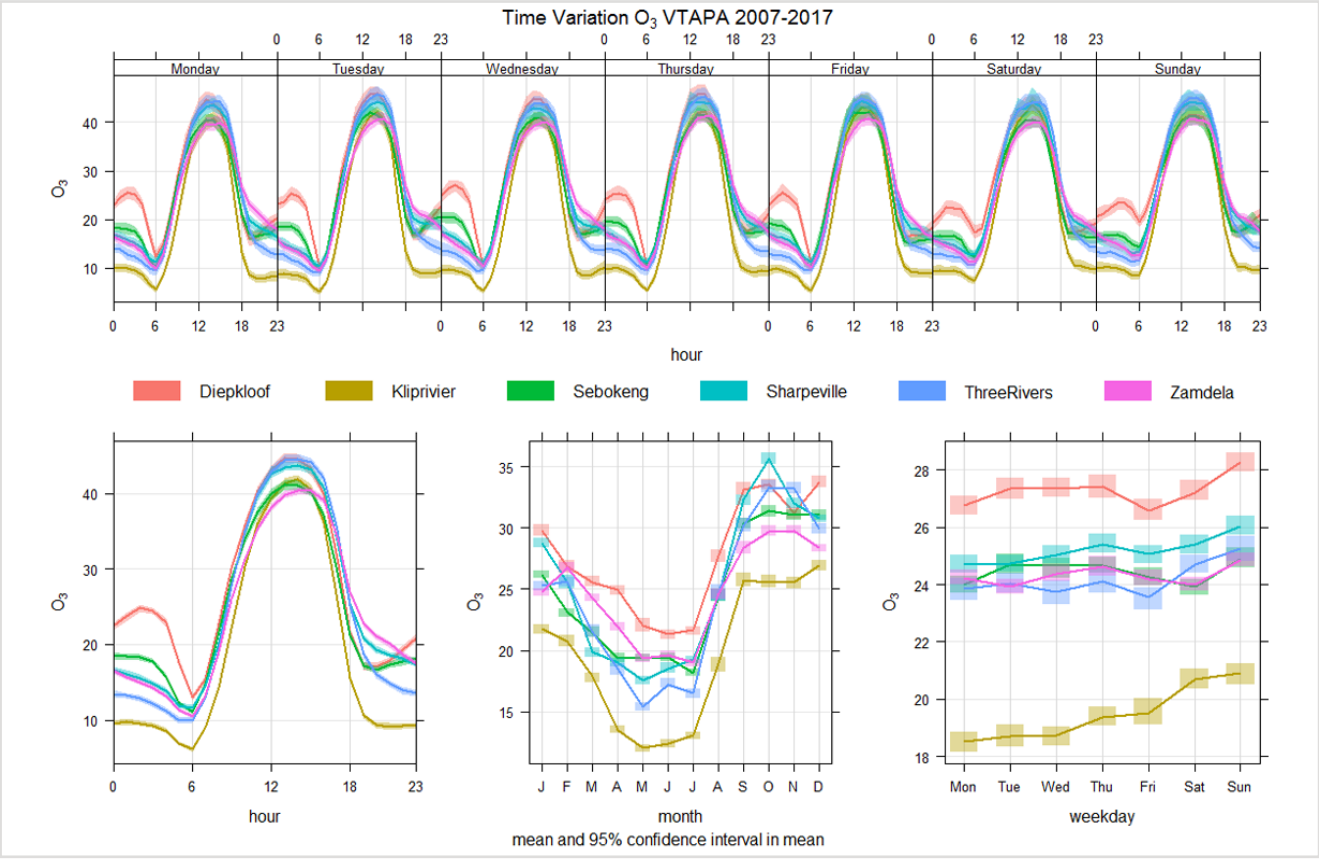

Figure 5: *O_3 time variation plot for daily average of concentration of hourly data for February 2007 to June 2017*

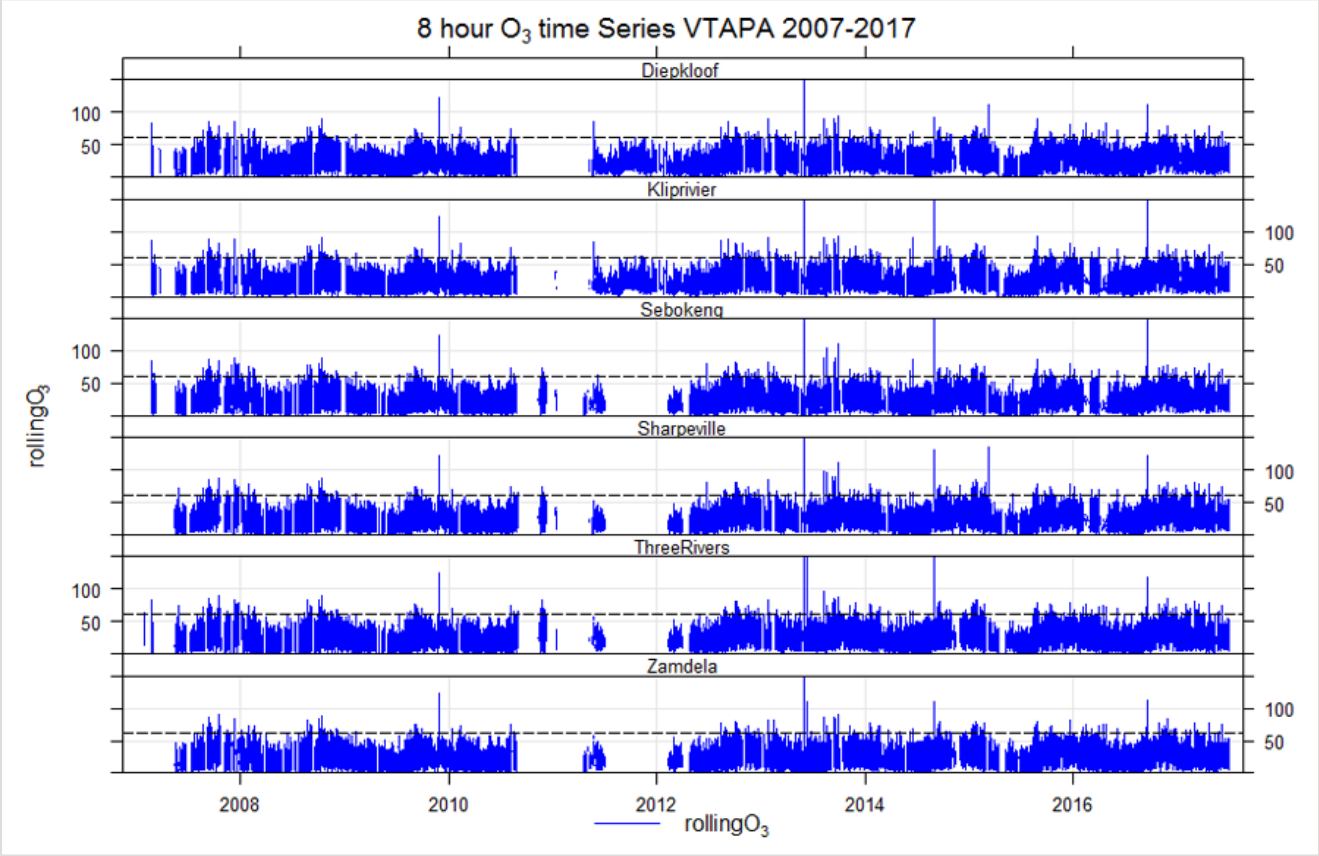

Figure 6: *Time Series Plot illustrating the 8-hourly O_3 concentration for the period February 2007 to June 2017*

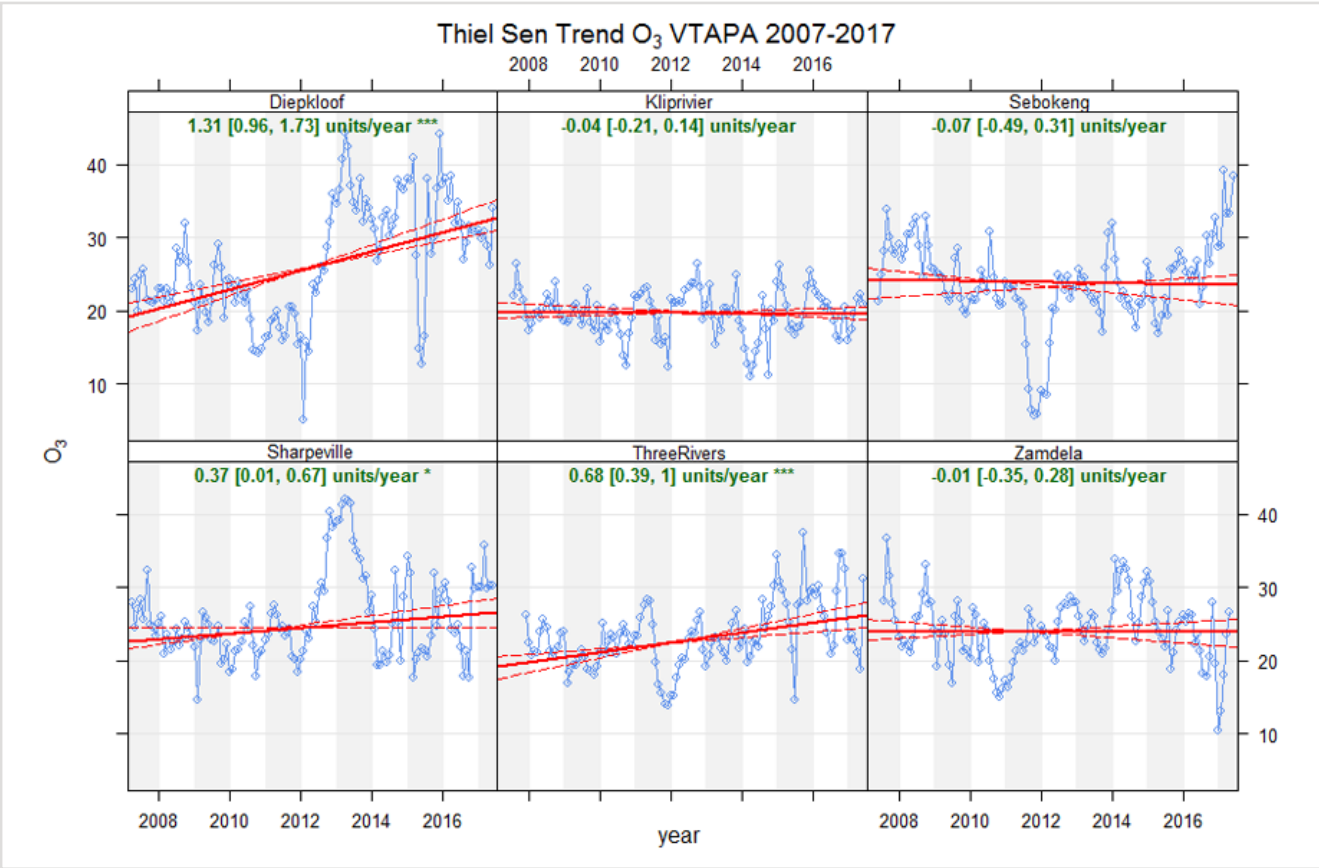

Figure 7: *Mann Kendall and Thielsen for O_3 concentrations for 2007-2017*

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

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

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

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

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

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

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

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

Ozone (O_3)

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

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

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

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

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

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

Discussion

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

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

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

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