Research article

Decreasing trend in SO_2 concentrations over Durban: 2004 - 2014

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Abstract

The climatology of ambient SO_2 air pollution was investigated in Durban, South Africa using data collected by seven air quality monitoring stations (Ferndale, Grosvenor, Jacobs, Wentworth, Settlers School, Southern Works and Prospecton) operated by eThekwini municipality (2004–2014, 2018-2019). These sites constitute a mix of urban and industrial locations.

Yearly average trends indicated that no site exceeded the yearly average national guideline (19ppbv) 2004-2014, 2018-2019. Southern Works, Wentworth and Jacobs, recorded highest yearly averages, a consequence of situation within Durban South Industrial Basin (DSIB) while Ferndale recorded lowest yearly averages, a reflection of location in an urban environment. Results of linear fitting to yearly average data (2004-2014) indicated negative trends (all sites). The largest trend was recorded at Jacobs (-0.48ppbv yr-1) and smallest trends observed at Prospecton (-0.12ppbv yr^{-1}), Ferndale (-0.084ppbv yr^{-1}) and Grosvenor (-0.024ppbv yr^{-1}). Using these linear trends, projected SO_2 levels were calculated and compared to actual data where it existed (2019 - Wentworth, Settlers, Prospecton), (2018, 2019 Southern Works). Comparison of actual with projected data indicated that except for Prospecton, projected yearly averages are lower than actual yearly averages for these sites. The largest difference between projected and actual data occurred for Southern Works in 2019 (4.10 ppbv). Monthly averages displayed periodic behaviour with maxima recorded in winter and minima in summer. Highest monthly averages were consistently recorded at Wentworth, Jacobs or Southern Works. Maximum monthly average (2004-2019) was reported at Jacobs, June 2009 (21.71± 2.82ppbv) while minimum monthly average (2004–2019) was reported at Ferndale, December 2012 (0.32±0.04ppbv).

Jacobs recorded maximum seasonal values June (11.54±6.37ppbv) to November (8.32±3.59ppbv), February (7.18±5.33ppbv) and April (9.02±6.38ppbv) while Southern Works recorded maximum seasonal average January (7.37±5.09ppbv), March (9.03±5.81ppbv). During May, Southern Works, Jacobs, Wentworth reported closely matched values (9.04±4.50ppbv, 9.17±6.34ppbv, 9.49± 5.22ppbv). Minimum seasonal levels were recorded at Ferndale (1.06±0.58ppbv – 2.29±0.88ppbv). Ferndale, Grosvenor, Wentworth and Settlers School reported secondary maxima in September/October potentially indicating the influence of biomass burning at these locations.

Seasonal averages illustrated that cooler conditions favoured higher SO_2 levels and warmer conditions, lower concentrations. Comparison of average difference between seasonal maximum and seasonal minimum (average width of seasonal envelope) indicated that Jacobs had the largest seasonal envelope and Ferndale the smallest.

Previous Durban SO_2 studies are considerably older or of shorter duration than the analysis presented here and are limited as they principally focus on yearly average trends. The determination of SO_2 levels over additional averaging periods (monthly average, seasonal variation, seasonal average) coupled with a larger data set and the comparison across sites (industrial vs urban) provides a more detailed exposure profile as experienced by individuals living and working in the eThekwini municipality and thereby expands on previous investigations.

Keywords

Ambient SO_2 pollution, Durban, yearly average, monthly average, seasonal variation, seasonal average.

Introduction

Air pollution is a major problem of the new millennium, and it has become increasingly clear that human activities are playing an important role in the cycling of trace gases in the atmosphere (Carslaw and Carslaw 2001). Poor air quality can affect health and the wider environment, particularly in urban areas where the majority of people live and work. Due to its abundance and substantial health impacts, SO_2 was chosen as the target pollutant in this investigation.

Several factors influence air quality in urban areas. The levels of pollutants released into the atmosphere are directly related to the number of emission sources, distribution of emission sources and volume of pollutants released by each source. These sources may be in the form of stationary sources e.g. emissions from a combustion furnace flue stack or mobile sources such as exhaust emissions from cars / aeroplanes etc. and accompanying transport of pollutants (Masiol et al. 2014).

Air quality is also affected by the rate at which pollutants disperse. Dispersion is dependent on both wind direction and strength. Strong winds result in rapid dispersal of emissions whereas little or no wind results in the accumulation, and in some cases, high concentration of air pollutants. Local factors such as topography and proximity to coast, building height and time of year all affect local wind conditions and can play a role in increasing air pollution levels (Diab et al. 2002; Thambiran and Diab 2010).

As in the case of NO_2, SO_2 has multiple emission sources both natural and anthropogenic in origin (Masiol et al. 2014). Natural emission sources of SO_2 include volcanoes, grassland, and forest fires. Coal and petroleum often contain sulphur compounds and their combustion generates SO_2. Anthropogenic SO_2 emission sources therefore include combustion of fossil fuels / crude oil and coal transformation processes. It is also produced as a by-product of metal smelting (of sulphur containing ores) and other industrial processes (Masiol et al. 2014). Approximately 90% of sulphur present in fossil fuels enters the gas phase in the form of SO_2 during combustion unless it is deliberately removed from fuel gas (Hewitt 2001). As a result of these combined industrial activities, approximately 99% of the SO_2 present in ambient air is of anthropogenic origin (Hewitt 2001). The main sinks of SO_2 are the oxidation by OH radicals and wet deposition, namely the solution in cloud droplets where it is converted to sulphurous (H_2SO_3) and sulphuric acid (H_2SO_4) (Pham et al. 1995).

The main route of SO_2 exposure is inhalation and the target organs comprise the respiratory system. Once inhaled, SO_2 is absorbed through the respiratory tract and distributed to all parts of the body, including the brain and bone marrow. Exposure to SO_2 is linked to numerous adverse effects on the respiratory system including broncho-constriction, wheezing and increased asthmatic symptoms (Matooane and Diab 2003; Mentz et al. 2018). It has also been found that SO_2 is toxic to plants (Lee et al. 2017).

South Africa has been identified as a source of industrial pollution, significant on a global scale (Josipovic et al. 2010). Furthermore, the problem of air pollution in Durban has a long history, particularly in relation to the Durban South Industrial Basin (DSIB). The DSIB is an approximately 4 km wide area on the eastern seaboard of South Africa, extending south from the Durban Central Business District (CBD) for 24 km to Umbogintwini. This area includes the CBD and Port of Durban, which is the busiest port in Africa. Poor historical land-use planning has resulted in juxtaposition of residential and industrial areas in South Durban. This has led to a long-term ongoing conflict between local communities and industry, particularly regarding concerns about possible effects of ambient air pollution on the health of residents (South Durban basin multi-point plan case study report 2007; Matooane and Diab 2003).

Industrial development in the area started in the mid 1950's. At present, some 600 industries are reportedly located in South Durban Industrial Basin. Emission sources include Mondi (paper manufacturing), sugar refinery, sewage treatment works, a cluster of chemicals industries, major petrochemical and chemical storage facilities, textile manufacturing, metal smelting, breweries, factories relating to the paint and motor industries. During the study period (2004-2014, 2018-2019) the Engen refinery and SAPREF were also significant sources of air pollutants. The fact that several of the major facilities have relatively low stack height (50 – 100 m) further facilitates the increase in pollutants close to the ground (South Durban Basin multi-point plan case study report 2007). The DSIB is also the focal point of many of the city's major transport routes and this adds a further large contribution to emissions from vehicular traffic and shipping (South Durban Basin multi-point plan case study report 2007).

Other factors apart from emission sources affect air pollution levels in DSIB. The local climate has a direct influence on the fate of particulates and gases released into the atmosphere. Concentration of pollutants varies on a daily / hourly basis in response to changes in atmospheric stability, resultant mixing depth and atmospheric circulation patterns. High concentration of air pollutants is associated with poor dispersion conditions. Furthermore, South Durban has a basin-like topography that is conducive to the accumulation of pollution (South Durban Basin multi-point plan case study report 2007), (Air quality scoping report: Royal Haskoning 2014).

Winds in Durban basin blow predominantly from the south-south-west to south-west and north-north-east to north-east in approximately equal proportions. Winds from north-north-east / north-east are associated with high atmospheric pressure and fine weather however winds from south-south-west / south-west are associated with the passage of coastal low-pressure systems and cold fronts and hence accompany unfavourable weather. The direction of predominant winds parallel to the coast together with the DSIB topography results in the channelling of pollutants within the basin (South Durban Basin multi-point plan case study report 2007).

The effect of temperature inversion conditions on pollutant levels should also be considered. A temperature inversion occurs when the air temperature increases with height as opposed to the usual decrease in temperature with increasing altitude. Inversions are common overnight during periods of calm weather and are generally strongest in the early morning hours. This inversion phenomenon acts like the ‘lid’ on a containment vessel that traps pollutants close to the ground and prevents upward air movement. Temperature inversions coupled with low wind speed can result in high levels of pollutants near the surface. A brown haze is a common feature of air quality in Durban during winter and can be attributed to the photochemical action of NO_x, O_3, SO_2, PM and VOCs. Once air flow is restored, circulation of pollutants can occur. Finally, air quality may be negatively impacted through the transport of pollutants from inland areas down to the coast by north-westerly land breezes at night, particularly during winter.

The effect of rainfall on air pollution is also significant. Annual rainfall in Durban is 1009 mm with most of this rain falling in summer. This period of high rainfall is associated with periods of improved air quality as the overall effect of rainfall is to remove dust and pollutant gases from the atmosphere. During summer humidity is high (sometimes approaching 100%) while winter is characterised by low humidity (as low as 20%). This high humidity in summer implies that chemical reactions that require water vapour are accomplished more efficiently, hence airborne pollutants are removed more effectively and rapidly than during the dry winter conditions (South Durban Basin multi-point plan case study report 2007), (Naidoo et al. 2007). Overall, dispersion conditions in summer improve due to less stable air conditions, higher wind speed and the effect of rainfall. As a result of the contributing factors discussed above, DSIB one of the most heavily polluted industrial areas in South Africa.

Figure 1: *Location of seven air pollution monitoring sites employed in this investigation – Ferndale, Grosvenor, Jacobs, Wentworth, Settlers School, Southern Works and Prospecton.*

As SO_2 is a principal by-product of many industrial processes it is often used as an 'indicator pollutant'. The National Ambient Air Quality Standards (NAAQS) for SO_2 are 191ppbv (10-minute average), 134ppbv (hourly average), 48ppbv (daily average) and 19ppbv (yearly average) (Government Gazette RSA 2009). Air quality monitoring in Durban was initiated at Wentworth in 1958 (Diab and Motha 2007) and as a result SO_2 is therefore the pollutant with the longest record of near continuous monitoring. The present investigation aims to characterize the climatology and seasonal variation of SO_2 emissions in order to compare and expand on previous studies at this location (Diab et al. 2002; Guastella and Mjoli 2005; Gounden 2006; Diab and Motha 2007; Guastella 2008; Mdluli 2015; Khumalo 2020).

It should be noted that previous SO_2 trend studies are considerably older and for significantly shorter study periods than the present investigation, as in the case of Diab et al. 2002; Guastella and Mjoli 2005; Gounden 2006; Diab and Motha 2007; Guastella 2008. Furthermore, these studies largely focus on yearly trends. There are very limited studies on monthly averages, seasonal variation or seasonal averages. Analysis of monthly average trends are presented in Guastella and Mjoli (2005), however this work is based on data collected for the period 1996 to 2003. More recent studies (Mdluli 2015 and Khumalo 2020) solely focus on analysis of SO_2 yearly average trends. The motivation for this investigation was therefore to complete a detailed long term trend study of SO_2 climatology in Durban, including analysis of not only yearly averages but additionally monthly averages, seasonal variation and seasonal averages as well as an analysis of data recorded in both industrial and urban environments.

Data collection, processing and instrumentation

eThekwini Municipality commissioned the continuous monitoring network in December 2003 as one of the major elements of its Air Quality Management System. The network is composed of instrumentation owned and operated by the eThekwini municipality. The network consists of monitoring stations situated at a range of sites representing heavy industry, urban, residential and rural locations. The network instruments continuously measure priority pollutants (measurement techniques) CO (infrared spectrometry), NO_x (chemiluminescence), SO_2 (UV fluorescence), O_3 (UV photometry), PM_{10} (TEOM - tapered element oscillating microbalance) as well as other species at selected sites.

In this investigation, seven sites were chosen to examine the long-term trends on SO_2 ambient air pollution in South Africa, these are: Ferndale, Grosvenor, Jacobs, Wentworth, Settlers School, Southern Works and Prospection. Figure 1 indicates the locations of the seven monitoring stations.

The seven selected stations can be grouped in the following way: One northern station – Ferndale, which is located at the Ferndale Primary School, is situated some distance from roads and industry. The closest industries are found approximately 2 km to the SE in River Horse Valley Industrial Park and Briardene Industrial Park. This site is representative of an urban environment, and it is the only monitoring station situated in the north of Durban. The remaining six stations - Jacobs, Grosvenor, Wentworth, Settlers School, Southern Works, Prospection are all located in the heart of the DSIB with the exception of Prospecton. It is the most southerly site of the selected locations and serves as an industrial background station measuring pollutant levels entering the DSIB from the Prospecton/Umbogintwini area.

The quality of data collected at the above monitoring stations is assured in a number of ways as a robust server-based data acquisition sytem called Envista Air Rosources Manager is employed. This system is globally used and has built-in data flags which are in accordance with ISO/IEC 17025:2017. Data validation is accomplished by adhering to the National Norms & Standards for Ambient Air Quality Monitoring developed by the Department of Forestry, Fisheries and Environment (DFFE)

Table 1: *Monitoring station (and GPS coordinates), SO_2 monitoring instrumentation, location characteristics and years of available data.*

Station	GPS Coordinates	SO_2 monitoring instrumentation	Location characteristics	Years of available data
Ferndale	-29.77806 30.97805	Monitor labs ML, 9850B	Urban	2004 - 2013
Grosvenor	-29.92089 31.00436	Thermo Scientific, Model 43iQ series	Industrial	2004 - 2012
Jacobs	-29.92831 30.97937	Thermo Scientific, Model 43iQ series	Industrial	2004 - 2014
Wentworth	-29.93306 30.98774	Teledyne API, Model T100	Industrial	2004 – 2014, 2019
Settlers School	-29.95875 30.97905	Teledyne API, Model T100	Industrial	2004 – 2014, 2019
Southern Works	-29.95984 30.97395	Thermo Scientific Model 43iQ series	Industrial	2004 – 2014, 2018, 2019
Prospecton	-30.00311 30.92960	Environment SA, Model AF22M	Industrial background	2004 – 2014, 2019

(National). Data outliers are flagged such as duplications, and erroneous data such as spikes and dips often due to abrupt shutdowns (loadshedding). Data validation checks generally take place every 15 days.

In terms of quality control, the practice of ISO 17025, SANAS TR0703 and the National Department (DFFE) Draft Norms and Standards for Ambient Air Quality Monitoring are followed. Station checks are done daily. In addition to data collection, the Envista system assists with data management and the operation of the monitoring stations. Each station has a datalogger installed with Envida software. Envidas is a package/component of Envista. All instruments are configured to transmit data from instrument to the datalogger, Envidas captures and saves this data on the logger PC. Envidas is configured to the remote server running the Envista DMS, data is transmitted from station level to server level via network connection and these applications. Data for each station and instrument is remotely checked each day on Envista by the data team and the technicians and technicians respond to stations that display no online status for stations or instruments, and/or any data that appears spurious.

Bi-weekly calibration is done in-house and is a one-point check on the gas instruments, using low concentration (ppb) certified and accredited gas. Multi-point calibration is done quarterly, in-house - three times per year. Here a dilution calibrator, zero air generator and a high concentration (ppm) certified and accredited gas is employed. Calibration at zero and high span is undertaken, and gas is diluted to multiple points in order to achieve a linear relationship. Finally, one external calibration is done by a SANAS accredited lab yearly.

Instrument maintenance is done in accordance with manufacturer specifications and standards including preventative maintenance such as planned services. In addition, to bi-weekly calibrations instrument checks are conducted and instrument check sheets completed. In these checks, monitoring of the instrument response not only to gas but also to electronic signals that have a defined operating range/threshold value e.g., instrument flows, temperature, pressure, voltage of certain components: lamps, PMT, etc. takes place (eThekwini air quality monitoring network – Annual Report 2009).

For each monitoring station, Table 1 gives details of GPS coordinates, SO_2 monitoring instrumentation and total years of available data during the period 2004 – 2019. All instrumentation is based on the UV fluorescence technique for SO_2 monitoring.

All original data was obtained in hourly intervals in excel format and processed with the use of MATLAB. In order to remove any spurious values from that dataset (such data gaps, zero and negative values), a 70% data completeness criterion was applied thereby removing any average data point that was calculated from less than 70% available data in that particular dataset. This rule was applied throughout the analysis presented in this work.

All raw hourly data sets were manipulated in order to calculate SO_2 yearly averages, monthly averages, seasonal variation and seasonal averages. It should be noted that not all sites have complete data sets 2004 -2019. For sites where data was available 2018-2019, this was included in order to optimize the data set and consequent findings. For yearly averages, monthly averages and seasonal averages, graphs are plotted for the entire time period in order to facilitate comparison between sites.

Results and discussion

Annual average trends

As a first step in investigating trends in the Durban air pollution SO_2 dataset, yearly averages were calculated for each site and the results presented below. Figure 2 shows a composite of seven plots, each illustrating the yearly average 2004 – 2019 for the monitoring sites Ferndale, Grosvenor, Jacobs, Wentworth, Settlers School, Southern Works and Prospecton.

Figure 2 yields a number of important observations. Of the chosen monitoring sites, Southern Works, Wentworth and Jacobs recorded the highest yearly averages over the study period. In 2004, the maximum values recorded were reported

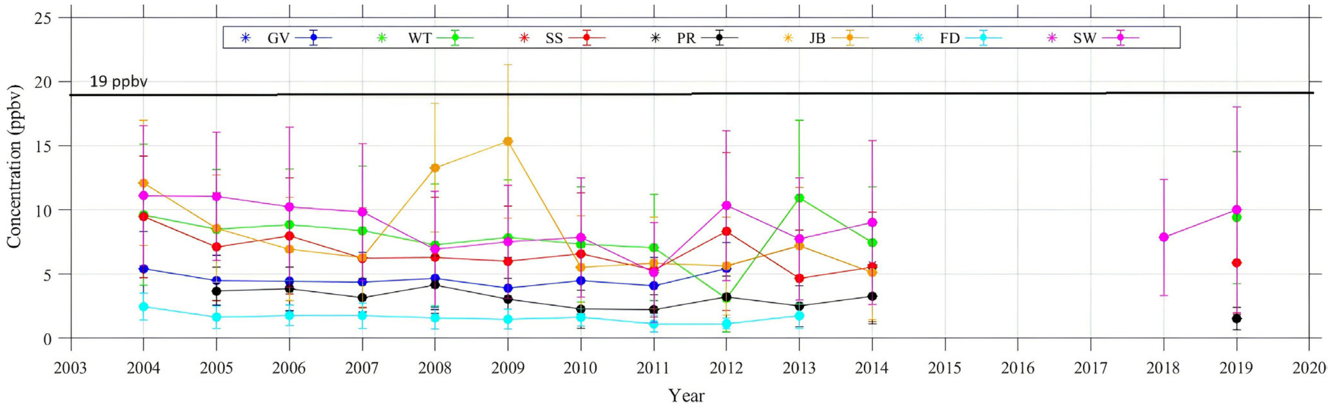

Figure 2: *Comparison of yearly average SO_2 concentration (ppbv) for the study period 2004 – 2019 at monitoring sites Ferndale (FD), Grosvenor (GV), Jacobs (JB), Wentworth (WT), Settlers School (SS), Southern Works (SW), Prospecton (PR). The permissible NAAQS early average exposure limit of 19ppbv is indicated by the solid black line for reference.*

at Jacobs (12.09 ± 4.87ppbv) and Southern Works (11.12 ± 5.46ppbv). Between 2005 – 2007 Southern Works recorded maximum yearly averages (for the dataset) in the range 11.04 ±4.99ppbv – 9.83 ±5.36ppbv, respectively. During 2008 and 2009, Jacobs reported maximum values of 13.27 ± 5.02ppbv and 15.34 ± 5.97ppbv. These levels significantly exceed those recorded at the remaining 6 sites. Yearly average SO_2 concentration recorded at Jacobs in 2009 is the highest value recorded for all seven sites (2004 – 2019). In 2011 and 2013, Wentworth recorded maximum values for the dataset of 7.06 ± 4.14ppbv and 10.92 ± 6.03ppbv respectively. During the remaining years 2010, 2012, 2014, 2018, 2019 Southern Works reported maximum values for the dataset, and these were in the range 7.85 ± 4.62ppbv to 10.34 ± 5.83ppbv.

From 2011 to 2014, Southern Works, Settlers school and Prospecton show a correlation in SO_2 levels. It is possible that Settlers School and Southern Works may have been influenced by the same emission sources which resulted in this behaviour, however, Prospecton is the most southerly site of the 7 locations, and it is therefore uncertain why this would show a similar pattern of SO_2 observations during this period (2011 – 2014).

The Prospecton and Ferndale monitoring sites are characterized by consistently low SO_2 concentrations with Ferndale recording the lowest yearly average for the study period, during 2011 (1.11 ± 0.64ppbv). Since the Ferndale monitoring site is situated north of Durban and is also some distance from main roads and heavy industry, these factors must therefore be the main contributing factor to the low SO_2 levels observed at this site. Finally, the guideline for permissible yearly average SO_2 exposure is 19ppbv and for the period of study, all yearly average values for the seven monitoring stations dataset are below this limit.

For each location, maximum and minimum yearly average (2004 – 2019) were determined together with difference between maximum and minimum values. In each case, a linear trend was fitted to the data and the gradient and norm of residuals of the trend calculated. Because of the substantial data gap, 2015 – 2018 (Southern Works), 2015 - 2019 (remaining sites), the linear trend was fitted for the period 2004 - 2014 and projected data for 2018, 2019 (Southern Works), 2019 (all remaining sites), compared with actual data. It should be noted that actual data only exists for 2018, 2019 (Southern Works), 2019 (Wentworth, Settlers School, Prospecton). For comparison, Table 2 shows maximum value and year, minimum value and year, difference between maximum and minimum values, linear trend fitted to yearly average data (2004-2014) and projected yearly average data (2019) for seven monitoring sites. Actual data recorded at Wentworth, Settlers School, Prospecton (2019), Southern Works (2018, 2019) is also presented. Maximum, minimum yearly average values and actual data (2018, 2019) are expressed with standard deviation in parenthesis.

Consideration of Table 2 indicates that Jacobs shows the largest maximum value for the dataset, namely 15.34 ± 5.97ppbv recorded in 2009. Southern Works, Wentworth and Settlers School all record maximum values in the range 9.47 – 11.12ppbv. The locations Grosvenor, Prospecton and Ferndale all record significantly lower maximum values for their datasets with these values in the range 2.47 - 5.45ppbv. Furthermore, three sites record dataset maximum values in 2004 and these are Ferndale, Settlers School and Southern Works. There is a second grouping of dataset maximum values in 2008-2009 and these are recorded at Prospecton and Jacobs respectively. There is a third grouping of dataset maximum values in 2012-2013 recorded at Grosvenor and Wentworth respectively. The observation that maximum values appear to fall into three groupings seems to indicate that the three groupings reflect similar causal factors at specific locations at similar times. Both Settlers School and Southern Works are located within close proximity to each

Table 2: *Maximum value and year, minimum value and year, difference between maximum and minimum values, linear trend fitted to yearly average data (2004-2014) and projected yearly average data (2018, 2019) for seven monitoring sites. For comparison, actual data recorded at Wentworth, Settlers School, Prospecton (2019), Southern Works (2018, 2019) is also presented. Maximum and minimum yearly average values are expressed with standard deviation in parenthesis.*

Monitoring site	Maximum of data set (ppbv) and year	Minimum of data set (ppbv) and year	ΔSO_2 (ppbv)	Gradient of linear trend (ppbv yr^{-1})	Norm of residuals	Projected yearly average 2019 (ppbv)	Actual yearly average 2019 (ppbv)
Ferndale	2.47 (1.03) 2004	1.11 (0.64) 2011	1.36	-0.084	0.84	0.75	No data
Grosvenor	5.45 (2.01) 2012	3.91 (2.37) 2009	1.54	-0.024	1.47	4.34	No data
Jacobs	15.34 (5.97) 2009	5.13 (3.65) 2014	10.21	-0.48	10.07	3.55	No data
Wentworth	10.92 (6.03) 2013	3.13 (2.63) 2012	7.80	-0.19	5.83	5.97	9.40 (5.14)
Settlers School	9.47 (4.73) 2004	4.68 (3.76) 2013	4.79	-0.27	3.51	3.94	5.88 (4.01)
Southern Works	11.12 (5.46) 2004	5.12 (3.88) 2011	6.00	-0.29	5.22	2018: 6.19 2019: 5.92	2018: 7.89 (4.53) 2019: 10.02 (8.00)
Prospecton	4.17 (2.24) 2008	1.53 (0.86) 2019	2.64	-0.12	1.66	2.02	1.53 (0.86)

***Table 3:** Results obtained from Gustella and Mjoli (2005) for SO_2 five-year mean (1998-2002) and SO_2 mean for 2003 with data from this investigation (2004).*

Location	Five-year mean (1998 – 2002) (ppbv) from Gustella and Mjoli (2005)	Mean for 2003 (ppbv) from Gustella and Mjoli (2005)	Mean for 2004 (ppbv) from present study
Wentworth	18	11	9.61
Southern Works	26	21	11.12
Settlers School	15	14	9.46

other, and Wentworth and Grosvenor form a further pair of locations presumably similarly impacted by the same factors at approximately the same time. Minimum values recorded for the dataset seem to follow a less well-defined pattern. For all sites excluding Prospecton (minimum recorded in 2019), minima for the remaining six sites are recorded in the range 2009 – 2014.

Table 2 also illustrates that the greatest difference in maximum and minimum values is seen in the data recorded at the Jacobs site (10.21ppbv). This is followed by Wentworth (7.80ppbv), Southern Works (6.00ppbv) and Settlers School (4.79ppbv). These sites are also located closest to the Durban South Industrial Basin (DSIB) and are presumably more directly impacted by variability in the emissions from the heavy industry in this area. Sites such as Ferndale and Prospecton show smaller differences between the maximum and minimum values for the period of study. These sites are located further from the heart of the DSIB – taking Jacobs as a central location, Ferndale and Prospecton are located 16.72 km and 9.59 km respectively from the Jacobs monitoring site. These smaller differences between maximum and minimum values are presumably related to the fact that emissions have had the chance to disperse substantially before impacting at these sites.

Inspection of Table 2 shows that fitted trends for all sites are negative (2004-2014). The site with the largest negative trend is Jacobs (-0.48ppbv yr^{-1}). This is followed by Southern Works with a value of -0.29 ppbv yr^{-1}. The remaining sites fall within the range -0.024 to -0.27ppbv yr^{-1} in decreasing order: Settlers School (-0.27ppbv yr^{-1}), Wentworth (-0.19ppbv yr^{-1}), Prospecton (-0.12ppbv yr^{-1}), Ferndale (-0.084ppbv yr^{-1}) and Grosvenor (-0.024ppbv yr^{-1}). It is important to note that although the gradients of the linear fits in Table 2 are small, they do indicate that SO_2 emissions at all monitoring sites have decreased over the study period, which is a significant observation.

Comparison of actual data 2018, 2019 (Southern Works), 2019 (Wentworth, Settlers School, Prospecton) with projected data obtained from the fitted linear trend, indicates that except for Prospecton, projected yearly averages are lower than actual yearly averages for these sites. The largest difference between projected and actual data occurs at Southern Works (2019) with a difference of 4.10pppbv. For the remaining sites this difference in descending order is Wentworth (3.43ppbv), Settlers School (1.94ppbv) and Southern Works for 2018 (1.70ppbv). These sites are all located in the DSIB and it is concerning to note that significant differences between actual and projected data are reported for these locations. The case of Prospecton is interesting in that it records a smaller than projected value for 2019, indicating that SO_2 remediation measures in this location are resulting in a positive impact on ambient SO_2 levels. It should also be noted that this site is located in the extreme south of the chosen study area and is not characterized by the high degree of heavy industry as seen in the DSIB. Considering the above, it is important to note, that although three sites report increases in actual data relative to projected data (which is a concern), the projected data falls withing the standard deviation associated with the actual data in all cases (including Prospecton). This illustrates that although the presence of the data gap within the dataset is not optimal, its effect is not as significant as might be initially expected and the comparison of actual and projected data (2018, 2019) is a useful tool for validating the dataset.

As an aside, a detailed study of the major SO_2 emitters in close proximity to these sites would be useful in determining what has changed 2004-2014 relative to 2018, 2019 to produce the observed increases. This may be related to changes in the dominant types of industry in these areas, but also to possible modification in manufacturing processes that may result in increased SO_2 production. A thorough investigation in this regard could form the basis of a future investigation.

Using the linear fit determined from yearly average data for the seven sites (2004-2014) and projected data (2019), reduction in SO_2 levels (2004-2019) corresponded to the following sequence: Jacobs (67%), Ferndale (63%), Settlers School (50%), Southern Works (47%), Prospecton (47%), Wentworth (32%) and Grosvenor (8%). It is interesting to note that the two largest reductions are seen at an industrial site followed by an urban one. However as discussed previously, this finding should be placed in the contexts of the increases in actual data compared to projected data seen for Wentworth, Settlers School, Southern Works and Prospecton.

In Diab and Motha (2007), data were continuously monitored at Wentworth (1958 – 2005) and results from a long term SO_2 trend study presented. Data (µg m^{-3}) were derived from SO_2 bubbler records based on the hydrogen peroxide method of collection. Significant changes in ambient air quality were noted, namely extended periods of increasing trends in SO_2 interspersed with shorter periods of decreasing trends. During the 1980's and early 1990's an increasing trend in SO_2 levels was reported with levels exceeding 80 µg m^{-3} (30ppbv) in 1989 and 1991 and approximating maximum levels recorded in 1962. After 1991, SO_2 concentrations declined, and levels stabilized between 50 – 60 µg m^{-3} (19 – 23ppbv) over the following few years. Since

1998 they reported a further decline to a mean annual value of approximately 40 µg m^{-3} (15 ppbv) by the end of the dataset (2005). This should be compared with the values obtained in this work, the maximum value recorded at Wentworth is 10.92ppbv (2013) with maximum yearly average and minimum yearly average bound in the range 7.80ppbv. Furthermore, the continuation of the decline in yearly average SO_2 since 1998, as reported by Diab and Motha (2007), is confirmed by this work through the negative linear trend fitted to Wentworth yearly average data presented here. It is unfortunate that Diab and Motha (2007) only provided a descriptive analysis of their dataset, and a linear trend was not calculated (1958 – 2005), hence no direct comparison of a linear trend is possible with the present investigation. Comparison is further limited as Diab and Motha (2007) only contained data recorded at Wentworth.

In Guastella and Mjoli (2005), data from permanent SO_2 monitoring stations at Wentworth, Southern Works, AECI and Settlers School were presented. Measurements of SO_2 were undertaken by means of continuous ultraviolet fluorescence analysers with data logged at five-minute intervals. Data were recorded for the period 1996 – 2003 at Wentworth, Southern Works and AECI while data acquisition only started at Settlers School from 2000 resulting in a comparably shorter dataset (2000 – 2002). The authors reported that SO_2 concentrations were highest at Southern Works where the National Guideline of 19ppbv was exceeded for all years, except 2002. Data recorded at Wentworth showed that this guideline was also exceeded for the years 1996 – 1999. They emphasized that for the early part of their dataset, concentrations at Southern Works and Wentworth were comparable however in the later part of their dataset, levels at Wentworth showed a sustained decrease. Although yearly average data were presented for the above-mentioned sites, no linear trends were fitted to it, other than to comment on the obvious average decline in levels as seen in each of the relevant plots. They did however present mean SO_2 values (1998 – 2002) in comparison with levels recorded in 2003 for Southern Works, Wentworth and Settlers School (the mean for Settlers School was only applicable for the period June 2000 – December 2002). Their findings included the following: At Southern Works, there was an increase in annual average for 2003 relative to 2002, however levels in 2003 were still below those of the 5-year mean, thereby indicating an improvement in air quality. At Wentworth they noted a substantial reduction in annual average for 2003 relative to the 5-year mean while the annual average 2003 at Settlers School was consistent with the mean of the previous two years. A comparison of results obtained by Guastellla and Mjoli (2005), with those presented in this investigation is given in Table 3.

Consideration of Table 3 indicates that historically (prior to the dataset presented here) SO_2 levels at Southern Works were substantially higher than those recorded at other monitoring stations and also exceeded in both instances (five year mean and mean for 2003) the yearly average exceedance of 19 ppbv but that by 2004, concentrations have decreased to below this recommended level. For all three sites, it is also seen that the 2004 yearly average value recorded in the present study is lower than the mean (1996 – 2002) and 2003 level as presented in Guastella and Mjoli (2005). This further supports the evidence that, on average, air quality has improved at these three locations for the period 1996 – 2004.

Monthly average trends and seasonal variation

In order to investigate month to month variation in SO_2 levels at each monitoring site, monthly averages were calculated for the entire dataset and for each location. Figure 3 shows a composite

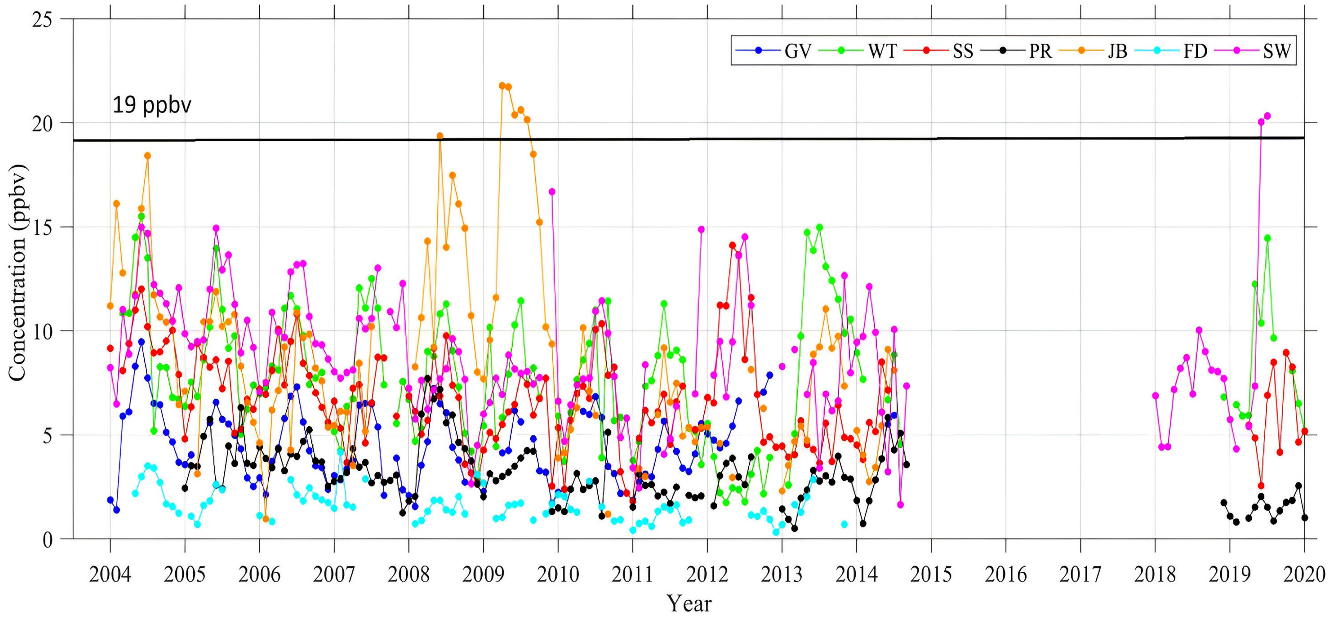

Figure 3: *Comparison of monthly average SO_2 concentrations (2004 – 2019) for the monitoring stations Grovenor (GR), Wentorth (WT), Settlers School (SS), Prospecton (PR) Jacobs (JB), Ferndale (FD) and Southern Works (SW). The permissible NAAQS yearly average exposure limit of 19ppbv is indicated by the solid black line for reference.*

of seven plots, each illustrating the monthly average 2004 – 2019 for the monitoring sites Ferndale, Grosvenor, Jacobs, Wentworth, Settlers School, Southern Works and Prospecton. For clarity, error bars have been removed in this figure. This determination of SO_2 monthly averages, allows the dataset to be investigated on a shorter time scale in order to determine more accurately, individual exposures experienced by those living in close proximity to the study sites.

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Figure 3 indicates that on average, all data sets exhibit the well documented pattern as observed by Diab et al. (2002) and Guastella and Mjoli (2005), namely high SO_2 concentrations in winter and low concentrations in summer. When datasets for multiple years are displayed together, a characteristic periodic behaviour is seen. This observation is a direct consequence of poor dispersion conditions experienced in Durban during winter. Temperature inversions trap a layer of air close to the ground causing the build-up of pollutants at this low level. This effect is further amplified by the fact that the DSIB is located in a basin type structure which favours the formation of temperature inversions. These factors all contribute to produce elevated SO_2 levels in the cooler months. The observations of these previous studies are therefore reaffirmed by the present investigation.

Comparison of the monthly data sets reveals that the highest monthly averages (2004 - 2019) are consistently observed at one of the three sites: Wentworth, Jacobs or Southern Works. The exception to this trend is Settlers School which records values comparable to those at Southern Works during Winter 2010 and Winter 2012. The highest monthly averages for the whole dataset were recorded at Jacobs during 2008 and 2009 with a maximum (21.71 ±2.82ppbv) measured during June 2009. June 2008 also recorded elevated SO_2 levels (19.36 ± 3.23ppbv) at this site relative to the rest of the datasets. It is interesting to note that the years 2008 – 2010 saw unusually high concentrations of SO_2 at this location. This is presumably attributable to locally increased emissions from industry in the area such as change in operational procedures with resulting impact on emission profiles.

The lowest monthly averages for the whole dataset are consistently recorded at Prospecton and Ferndale and these fall within the approximate range 1 – 7ppbv. Monthly average data for all monitoring stations appears to follow an approximately downward trend as would be expected given the negative linear trend as determined from yearly average SO_2 data.

Maximum and minimum monthly averages for each monitoring site (2004 - 2019) were determined and are presented in Table 4. Anomalous readings that do not conform to the well documented pattern of maximum SO_2 levels in winter and minimum SO_2 levels in Summer are indicated in Table 4 by (*).

It is noted from Table 4 that in most instances all sites conform to the expected periodic pattern of maximum and minimum levels. However, there are several instances that do not adhere to this pattern namely: Ferndale – maximum monthly average recorded in February 2007 and minimum monthly average measured in May 2012 at Wentworth. For the anomalous maximum reading, it possible that this corresponds to non-standard emissions from industry during this time and in close proximity to this site such that this effect dominated over the increased dispersion conditions typically experienced in summer. This would be unusual given that the Ferndale site is situated some distance from industrial sources.

For the anomalous minimum value recorded at Wentworth, observed high wind-speed from a south westerly direction may have also favoured the dispersion of pollutants. The case of Prospecton is puzzling as both minimum and maximum monthly average do not adhere to the established seasonal trend. The reason for this behaviour is unknown. The monitoring sites,

Table 4: *Maximum and minimum monthly averages for each monitoring site (2004 - 2019). Anomalous readings that do not conform to the well documented pattern of maximum SO_2 levels in Winter and minimum SO_2 levels in Summer are indicated by (*). Maximum and minimum values expressed with standard deviation in parenthesis.*

Monitoring site	Maximum monthly average (2004 - 2019) (ppbv) and date		Minimum monthly average (2004 - 2019) (ppbv) and date	
Ferndale	4.16 (0.35)	February 2007 (*)	0.32 (0.04)	December 2012
Grosvenor	9.47 (2.50)	June 2004	1.56 (1.14)	February 2008
Jacobs	21.71 (2.82)	June 2009	0.96 (0.61)	February 2006
Wentworth	15.50 (3.80)	June 2004	1.75 (1.35)	May 2012 (*)
Settlers School	14.40 (6.68)	June 2012	1.81 (0.54)	January 2011
Southern Works	20.32 (7.26)	July 2019	2.46 (1.37)	February 2011
Prospecton	7.71 (1.91)	October 2008 (*)	0.51 (0.19)	March 2013 (*)

Southern Works, Settlers School, Jacobs and Grosvenor do not show any anomalous behavior in terms of the typical seasonal trend.

In Guastella and Mjoli (2005), the following long-term trends in monthly averages were noted: For Wentworth, distinct seasonal fluctuations in SO_2 levels were recorded for the period 1996 – 2003 with monthly average concentrations measured in winter higher than those recorded in the summer months. This dataset indicated a general decrease in SO_2 concentrations from 1996 and the authors report that this trend is related to a decrease in SO_2 in winter as averages in summer were approximately constant. The authors propose that this reduction was related to reduction in emissions from refineries and coal-burning by smaller industries. At Southern Works, data collection for this location began in May 1998. Monthly averages (1998 - 2003) were seen to be fairly variable and the authors stated that this was clearly related to emission scenarios. This dataset showed a general decreasing trend over the study period with a slight increase in 2003. At Settlers School, data collection began from June 2000. Although this dataset is variable, the long-term trend in SO_2 showed an approximate decrease in levels. Monthly average SO_2 levels dropped to a minimum for the study period in December 2002. The results presented in the present investigation clearly confirm these previous observations in particular, maximum monthly averages recorded in cooler conditions with the converse also confirmed. The continued decline in SO_2 monthly levels reported in this work is clearly supported and expands upon this previous study.

In order to investigate seasonal variation in SO_2 levels at each monitoring site, average months were calculated for the entire dataset (2004 – 2019) and for each location. Figure 4 shows a composite of seven plots, illustrating the seasonal variation for the monitoring sites Ferndale, Grosvenor, Jacobs, Wentworth, Settlers School, Southern Works and Prospecton.

Figure 4 indicates that all monitoring stations record a maximum in the cooler months and minimum in warmer months, which is in agreement with the previous finding of Diab et al. (2002); Guastella and Mjoli (2005). In Durban, wind speeds are higher in spring (September – November) while low wind speeds are typically recorded in autumn / winter (April – June). High levels of pollutants are generally connected to low wind speed conditions in winter and at night / early morning. This is because of poor vertical mixing and low horizontal transport out of the source area (South Durban Basin multi-point plan case study report 2007), (Air quality scoping report, Royal Haskoning DHV, 2014). The observations presented in this investigation are clearly confirmed by the above findings.

From Figure 4 it is seen that Jacobs records the highest average seasonal values for the months June to November as well as February and April while Southern Works records the highest seasonal average for the months January and March. For the month of May, three sites are approximately equal, namely, Wentworth, Jacobs and Southern Works. The locations Ferndale and Prospecton consistently record the lowest seasonal SO_2 levels.

For all sites, the seasonal maximum is recorded in Winter (June – August) except for Prospecton which records a maximum in September. All sites record a minimum in Summer (December, January, February) except for Ferndale which records a minimum in early autumn (March). A possible explanation for this observation may be due to increased dispersion conditions at this site prevailing later in the year than at other locations.

The influence of biomass burning on ambient air pollution is well documented (Sinha et al. 2003; Behera and Balasubramanian 2014; Agbo et al. 2021). Savanna fires in Africa, constitute approximately two thirds of the savanna burned world-wide and chemical species released as emissions from these fires, include CO, NO_x, SO_2, hydrocarbons, halocarbons together with particulates. It has been reported in numerous sources (Crutzen and Andreae 1990; Blake et al. 1996; Yokelson et al. 1996 and 2003; Andreae and Merlet 2001, Sinha et al. 2003a) that the number of Southern African savanna fires peaks in the dry season (April – October). The effects of these fires on ambient air pollution levels are further amplified by meteorological

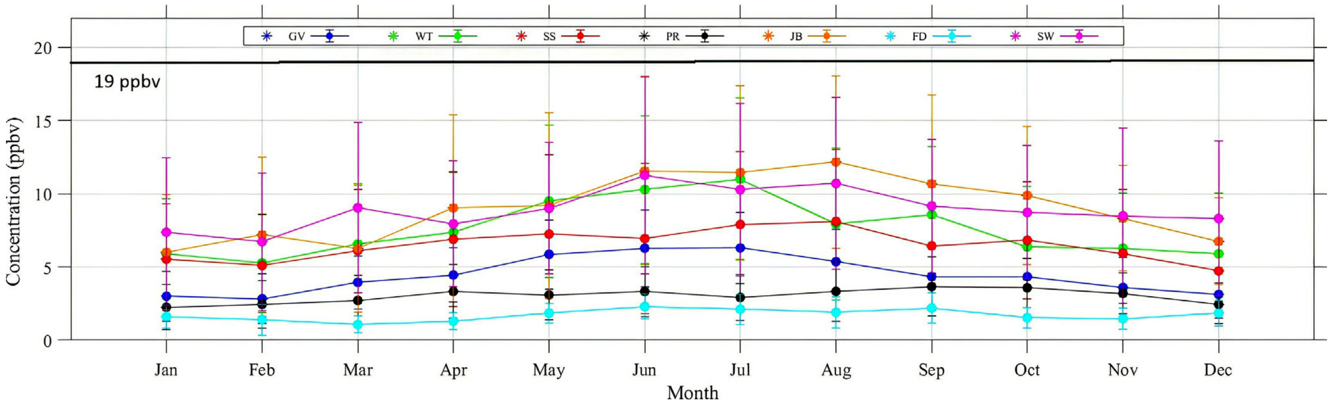

Figure 4: *Comparison of SO_2 seasonal variation (2004 – 2019) for the monitoring stations Grovenor (GV), Wentorth (WT), Settlers School (SS), Prospecton (PR) Jacobs (JB), Ferndale (FD) and Southern Works (SW). The permissible NAAQS yearly average exposure limit of 19ppbv is indicated by the solid black line for reference.*

conditions during this dry season such as stable air masses, southeasterly trade winds and subtropical high pressure over South Africa (McGregor and Nieuwolt 1998). Furthermore, the presence of stable layers reduces vertical mixing resulting in pollution build-up close to the surface (Cosijn and Tyson 1996; Hobbs 2002 and 2003).

Four sites, Ferndale, Grosvenor, Wentworth and Settlers School show a secondary maximum in September - October which (given the previous work mentioned above) may be attributed to the influence of biomass burning at these locations. Prospection is characterized by a single maximum in September, and it is possible that for this site, the impact of biomass burning dominates all other factors influencing the behaviour of SO_2 levels at this site.

Table 5 shows a comparison of time and level of maximum, minimum and percentage change in seasonal SO_2 levels over the year (between maximum and minimum), for the seven monitoring sites. The seasonal percentage change in SO_2 levels appears to fall into two separate groups. Ferndale, Grosvenor, Jacobs and Wentworth are characterized by seasonal percentage change in the range 122 – 103% while Settlers School, Southern Works and Prospecton are characterized by seasonal percentage change in the range 62 - 72%. Assuming that rainfall is approximately constant for all sites (and that emission profiles from industry are approximately the same throughout the year), it is possible that this observation is more closely related to other dispersion factors such as wind speed. High percentage changes indicate a greater variability in SO_2 levels over the year which must be related to conditions where more efficient stagnation mechanisms result in pollution build up in winter. The implication is that winds speeds are therefore lower in Winter at the sites Ferndale, Grosvenor, Jacobs and Wentworth, thereby resulting in higher levels of SO_2 and hence larger percentage changes between winter and summer levels. The converse is therefore that Prospecton, Settlers School and Southern Works experience higher wind speeds in winter, resulting in less build up pollutants near the surface and hence a smaller percentage change between winter and summer values at these locations. Prospecton is an interesting case in that the seasonal profile at this location is different from the remaining sites and this is probably the result of other factors impacting on the seasonal behaviour at this site.

Seasonal averages per year

In order to investigate the variation from season to season for each year of the dataset (7 sites), a seasonal average was calculated in the following way: For each site and each year of the dataset, an average for Spring (September, October, November), Summer (December, January, February), Autumn (March, April, May) and Winter (June, July, August) was calculated. The results are presented in Figure 5 which is a composite of seven plots, each illustrating the seasonal averages per year for the monitoring sites Ferndale, Grosvenor, Jacobs, Wentworth, Settlers School, Southern Works and Prospecton. The results are discussed individually for each site below. For clarity, error bars have been removed from the data points in the individual graphs.

For each monitoring site (and each year), difference between maximum and minimum seasonal value (or width of seasonal envelope) $\Delta_{Max,\ Min}$, was calculated together with associated percentage change. This difference between maximum and minimum values is clearly a combination of many factors such as changes in emission profile at the location but is possibly more representative of differences in meteorological factors (such as rainfall, wind speed) this is because the effect due to seasonal changes is generally expected to dominate over the effect of changes in emission profile over the time period of 3 months (1 season). For comparison purposes, the difference between minimum and maximum seasonal values is only significant for years where the average for all four seasons is available. Furthermore, for each monitoring site, the number of anomalous observations was calculated as a percentage of the total number of observations (where this is taken as the reduced data set using only years where data for all seasons is available). An anomalous observation is defined as not conforming to the well documented pattern of maximum in winter and minimum in summer. For each monitoring site, Table 6 shows $\Delta_{Max,\ Min}$ and

***Table 5:** Maximum and minimum SO_2 seasonal values and corresponding percentage change over the year for seven monitoring sites. Sites with a secondary maximum are indicated by (*). Maximum and minimum values expressed with standard deviation in parenthesis.*

Monitoring site	Maximum of seasonal variation (ppbv) and month		Minimum of seasonal variation (ppbv) and month		Percentage change between maximum and minimum over year
Ferndale	2.29 (0.88) 2.19 (*) (1.04)	June September	1.06 (0.58)	March	116
Grosvenor	6.28 (2.62) 4.33 (*) (2.13)	June - July October	2.83 (1.70)	February	122
Jacobs	12.15 (5.91)	August	6.00 (3.93)	January	103
Wentworth	11.00 (5.55) 8.57 (*) (4.64)	July September	5.27 (3.34)	February	109
Settlers School	8.11 (4.92) 6.81 (*) (4.02)	August October	4.72 (3.57)	December	72
Southern Works	11.26 (6.73)	June	6.71 (4.70)	February	68
Prospecton	3.65 (2.02)	September	2.25 (1.53)	January	62

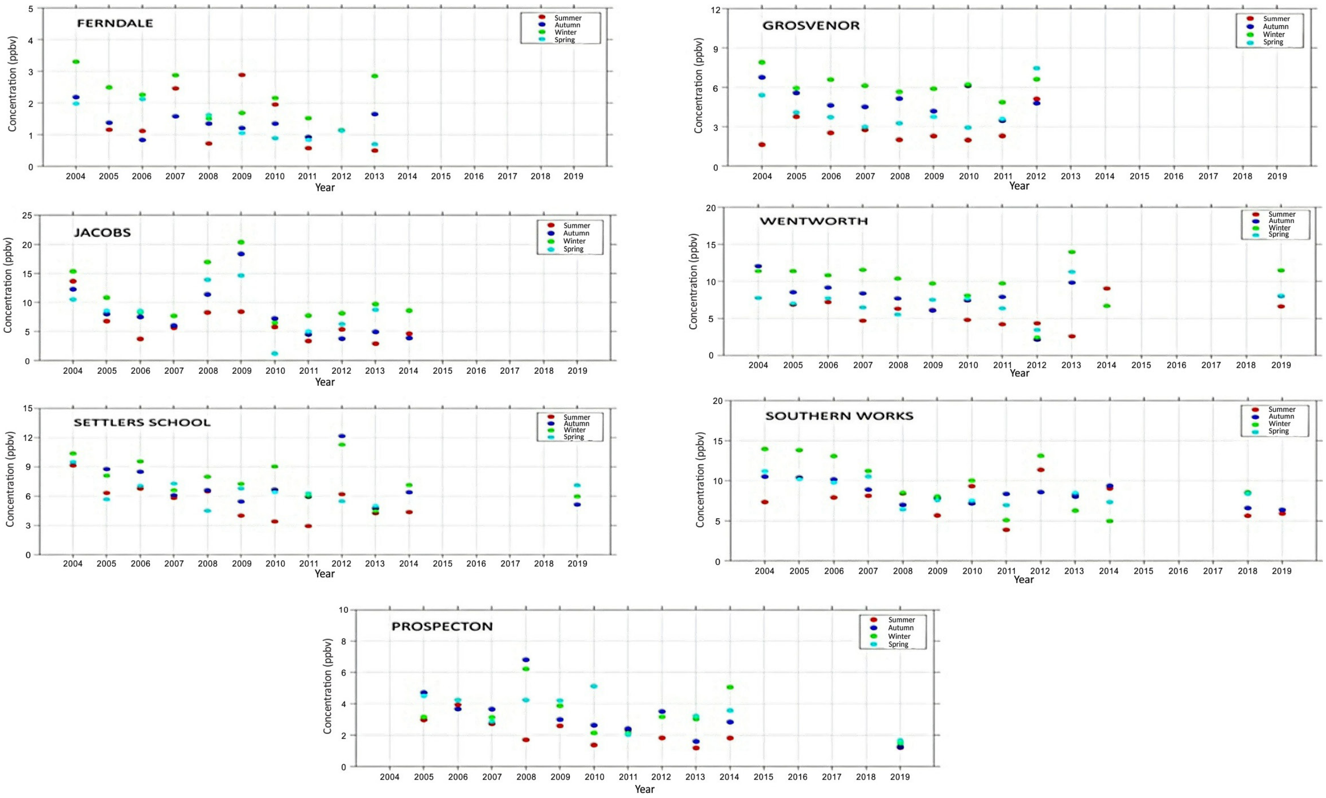

Figure 5: *Seasonal averages per year (ppbv) for the study period 2004 – 2019 at monitoring sites Ferndale, Grosvenor, Jacobs, Wentworth, Settlers School, Southern Works, Prospecton.*

corresponding percentage, average $\Delta_{\text{Max, Min}}$ (or average width of seasonal envelope) and percentage of anomalous observations in each data set. Values in red indicate years where data for all four seasons are available. Maximum and minimum $\Delta_{\text{Max, Min}}$ are indicated for each monitoring site.

Table 6 indicates that the largest $\Delta_{\text{Max, Min}}$ is recorded at Jacobs (11.95ppbv) in 2009 while the smallest $\Delta_{\text{Max, Min}}$ is recorded at Prospecton (0.38ppbv) in 2011. Furthermore, during 2004 Grosvenor and Southern Works both record maximum $\Delta_{\text{Max, Min}}$ while in 2013, Ferndale and Wentworth record maximum $\Delta_{\text{Max, Min}}$. In 2005, Grosvenor and Jacobs record minimum $\Delta_{\text{Max, Min}}$. In the case of 2004 and 2005, it is possible that this reflects real meteorological impacts on SO_2 levels as the sites Grosvenor and Southern Works (2004) and Grosvenor and Jacobs (2005) are in close proximity however the link between Ferndale and Wentworth (2013) may be more coincidental as these locations are further apart allowing for more significant differences in meteorological factors.

Comparison of average difference between seasonal maximum and seasonal minimum (or average width of seasonal envelope), for years with all seasonal data available, and for each location, is in descending order: Jacobs - 6.21ppbv, Wentworth - 5.05ppbv, Southern Works - 3.64ppbv, Grosvenor - 3.62ppbv, Settlers School - 3.35ppbv, Prospecton - 2.35ppbv and Ferndale - 1.43ppbv.

From the above it appears that Jacobs and Wentworth (5.05 – 6.21ppbv) form a separate grouping from Southern Works, Grosvenor and Settlers School (3.35 – 3.64ppbv). Prospecton and Ferndale have the lowest average difference between seasonal maximum and minimum 1.43 – 2.35ppbv). The average difference or width of envelope for seasonal average is clearly a combination of factors and it seems that at a minimum, the groupings outlined above constitute locations where the same sets of factors dominate in terms of SO_2 fluctuations. This is evidenced by consideration of the map of monitoring sites in that Jacobs and Wentworth are closely associated while (with the exception of Grosvenor) the same is true for the proximity of Southern Works and Settlers School. Finally, it is reasonable to assume that since Ferndale and Prospection are the furthest from the central DSIB, that they would be impacted by lower industrial emissions and a different set of meterological factors than those experienced in the DSIB.

In order to understand the above findings, an investigation of the location of emission sources in close proximity to each monitoring site and their individual contributions would prove difficult due to the very varied nature of industry at these sites, the complicated issue of quantifying the contribution from individual sources (e.g. factories/manufacturing plants, etc.) but also the temporal complexity of obtaining such data (if it existed at all) for the historical period 2004-2014. For these reasons, the authors have not pursued this line of enquiry in this work. There is however, limited meteorological information in the form wind direction and wind speed data for three of the sites, namely Grosvenor, Wentworth and Southern Works and this was used to generate wind roses for these locations. This analysis showed that during summer, the dominant wind direction for Grosvenor, Wentworth, Southern Works was NNE, NNE and NNW respectively while in winter, dominant wind direction was

WSW/SW, NNE and SW respectively. On average, wind speed at Wentworth was higher for all seasons compared to that at Grosvenor and Southern Works. Furthermore, assuming that $4ms^{-1}$ is a wind speed capable of efficient pollutant dispersal, the frequency of occurrence (%) of winds in excess of $4m^{-1}$, over all directions, was calculated for the three sites, for summer and winter. This analysis showed that for Grosvenor, frequency of occurrence (%) of winds with speed > 4 ms^{-1} was 12.3 (summer), 15.5 (winter), for Wentworth the corresponding values were 49.0 (summer), 48.5 (winter) and for Southern Works 7.7 (summer), 12.2 (winter). From these results, a counter-intuitive pattern of behaviour is seen. It would be expected that since summer is characterized by lower levels of SO_2 relative to winter, it could be assumed that the frequency of occurrence for winds in excess of $4ms^{-1}$ should be higher during this season, however the opposite pattern is seen for Grosvenor and Southern Works, with both seasons of approximately equal frequency of occurrence for Wentworth. It is possible that this anomaly is related to wind direction. The phenomenon of higher wind speed increasing surface SO_2 levels has been highlighted in Diab et al. 2002. They reported that high SO_2 values of up to 40ppbv at Southern Works were recorded under low wind speed conditions. This relationship was observed until a critical speed of 3.5 ms^{-1} was reached, at this point a sustained rise in SO_2 corresponded to an increase in wind speed. An analogous observation was reported at Wentworth, where the critical wind speed was slightly higher, namely 4.5 ms^{-1}. They proposed that such behaviour was related to the location of monitoring stations in relation to principal SO_2 sources and could result from stack down-drafting in strong winds, resulting in high SO_2 concentrations close to the ground. They further suggested that this was indicative of SO_2 emissions originating from elevated sources such as industrial stacks. The above results from the present work appear to broadly support the findings of Diab et al. (2002) and show that similar sets of

Table 6: *For each monitoring site, difference between seasonal maximum and minimum* $\Delta_{Max, Min}$ *(ppbv) and corresponding percentage change, average* $\Delta_{Max, Min}$ *and percentage of anomalous observations in each data set. Values in red indicate years where data for all four seasons are available. Maximum and minimum* $\Delta_{Max, Min}$ *are indicated for each monitoring site.*

Year	Ferndale $\Delta_{Max, Min}$ %	Grosvenor $\Delta_{Max, Min}$ %	Jacobs $\Delta_{Max, Min}$ %	Wentworth $\Delta_{Max, Min}$ %	Settlers School $\Delta_{Max, Min}$ %	Southern Works $\Delta_{Max, Min}$ %	Prospecton $\Delta_{Max, Min}$
2004	1.32 66.4	6.27 ^{MAX} 383.8	4.81 45.7	4.28 55.2	1.22 13.3	6.60 ^{MAX} 89.7	_
2005	1.33 115.1	2.18 ^{MIN} 57.9	4.06 ^{MIN} 59.9	4.37 62.4	3.09 54.4	3.59 35.1	1.74 58.4
2006	1.29 154.0	4.06 160.7	4.81 129.1	3.62 50.2	2.51 35.6	5.15 65.1	0.56 15.1
2007	1.29 81.8	3.36 122.0	2.04 36.1	6.85 145.3	1.45 24.8	3.09 37.9	0.92 33.9
2008	0.90 ^{MIN} 123.9	3.65 182.2	8.68 105.0	4.84 87.7	3.48 77.1	2.05 31.9	5.11 ^{MAX} 299.1
2009	1.82 173.3	3.60 157.62	11.95 ^{MAX} 141.9	3.65 60.2	3.25 80.7	1.91 ^{MIN} 33.7	1.61 61.9
2010	1.26 115.9	4.23 213.8	6.04 507.8	3.28 68.3	5.63 164.5	2.86 39.8	3.75 273.1
2011	0.95 163.8	2.57 111.8	4.36 129.2	5.51 130.5	3.34 351.6	4.47 114.9	0.38 ^{MIN} 18.4
2012	-	2.66 55.6	4.36 115.7	1.92 ^{MIN} 79.7	6.68 ^{MAX} 121.7	4.52 52.5	1.68 92.2
2013	2.35 ^{MAX} 467.4	-	6.79 233.1	11.37 ^{MAX} 437.5	0.74 ^{MIN} 17.4	2.20 35.0	2.03 170.6
2014	-	-	4.72 121.9	2.36 35.3	2.76 63.1	4.40 88.4	3.24 178.1
2018	-	-	-	-	-	2.73 48.4	-
2019	-	-	-	4.86 73.2	1.96 38.1	0.45 45.4	0.38 30.7
Average $\Delta_{Max, Min}$ (ppbv)	1.43	3.62	6.21	5.05	3.35	3.64	2.35
Anomalous observations %	33.3	11.1	22.2	16.7	38.9	33.3	50.0

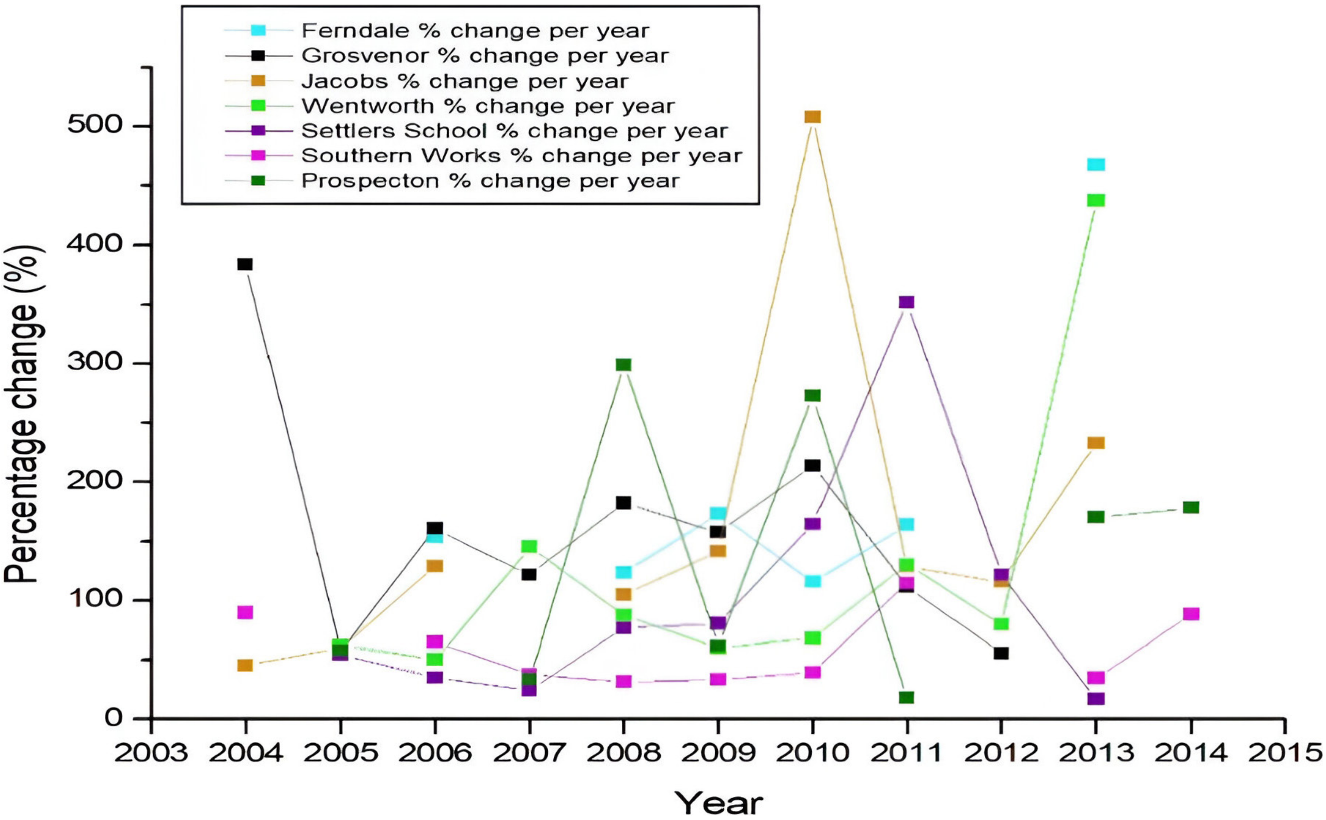

Figure 6: *Percentage change between SO_2 maximum seasonal value and SO_2 minimum seasonal value, for each location and years that have all seasonal data points.*

conditions are operative at Grosvenor and Southern works (as evidenced in this study by similar seasonal envelopes). The case of Wentworth clearly requires a more detailed study to fully determine relevant contributing factors.

Grouping the sites in terms of total number of anomalous seasons expressed as a percentage of the total (for the entire dataset) in descending order: Prospecton - 50.0%, Settlers School - 38.9%, Southern Works - 33.3%, Ferndale - 33.3%, Jacobs - 22.2%, Wentworth - 16.7%, Grosvenor - 11.1%. Excluding Prospecton, the percentage of anomalous seasons falls in a more closely bound range 22.2 – 38.9% for sites Jacobs, Ferndale, Settlers School and Southern Works. It is possible that this percentage change is related to the impact of dispersion factors at these sites, increased emissions or both. Wentworth and Grosvenor appear to form a further grouping. Finally, it is difficult to draw any further definite conclusions due to the small sizes of the data sets.

In order to compare seasonal variations between sites, percentage changes between SO_2 maximum seasonal value and minimum seasonal value were plotted for each year and for each location and are shown in Figure 6. This is only done for years for which four seasonal data points are available. It is difficult to draw any firm conclusions from this figure. From 2009-2011, there appears to be a correlation between Wentworth and Southern Works together with a correlation between Prospecton and Jacobs for the same time interval. A number of very large percentage changes occur in 2004 (Grosvenor), 2008 and 2010 (Prospecton), 2010 (Jacobs), 2011 (Settlers School) and 2013 (Wentworth). The cause of this behaviour is unknown.

Summary and conclusion

Trends in yearly average SO_2 levels at the seven sites indicated that no location exceeded the yearly average national guideline of 19ppbv for the study period (2004 – 2019).

Southern Works, Wentworth or Jacobs consistently recorded maximum yearly averages (2004-2019) and this is clearly a direct result of the location of these sites in the DSIB. In 2004, maximum was reported at Jacobs (12.09±4.87ppbv), and 2005–2007, Southern Works recorded maximum yearly averages ranging 11.04±4.99ppbv to 9.83±5.36ppbv, respectively. During 2008, 2009, maxima of 13.27±5.02ppbv and 15.34±5.97ppbv were observed at Jacobs. In 2011, 2013, Wentworth recorded maxima of 7.06±4.14ppbv and 10.92±6.03ppbv respectively. During 2010, 2012, 2014, 2018, 2019 Southern Works reported maximum values ranging 7.85±4.62ppbv to 10.34± 5.83ppbv. Ferndale consistently reported low SO_2 concentrations with minimum for the entire dataset measured in 2011 (1.11±0.64ppbv). These observations are clearly a consequence of its location in an urban environment.

Results of linear fitting to yearly average SO_2 data (2004-2014) indicated that SO_2 emissions show a negative downward trend over the study period for all monitoring sites. The largest negative linear trend was recorded at Jacobs (-0.48ppbv yr^{-1}) while the smallest trends were observed at Ferndale (-0.084ppbv yr^{-1}) and Grosvenor (-0.024ppbv yr^{-1}). It is important to note that although the gradients of the linear fits are small, they do indicate that SO_2 emissions at all monitoring sites have decreased over the study period, which is a significant observation.

Comparison of actual data for Southern Works (2018, 2019) and Wentworth, Settlers School, Prospection (2019) with projected values determined from the fitted trends indicate that although there are increases in actual data relative to projected data (Southern Works, Wentworth, Settlers School) and a decrease (Prospection), the projected data values for these years still fall within the standard deviation of the actual data recorded. This illustrates that the effect of the data gap (2015-2018/2019) is not as significant as might have been expected and this comparison provides a useful tool for data validation.

Using the linear fit determined from yearly average data for the seven sites (2004-2014) and projected data (2019), reduction in SO_2 levels (2004-2019) corresponded to the following sequence: Jacobs (67%), Ferndale (63%), Settlers School (50%), Southern Works (47%), Prospecton (47%), Wentworth (32%) and Grosvenor (8%). It is interesting to note that the two largest reductions are seen at an industrial site followed by an urban one.

Monthly average SO_2 levels at all sites, displayed the well-documented periodic behaviour of higher concentrations recorded in winter compared to lower concentrations in summer. This is in agreement with previous studies (Diab et al. 2002, who reported trends for Wentworth and Southern Works). The highest monthly average values were consistently recorded at one of the three sites: Wentworth, Jacobs or Southern Works. The highest value of the dataset was 21.71 ± 2.82 ppbv recorded at Jacobs in June 2009. The lowest monthly averages are consistently recorded at Prospecton and Ferndale with the lowest value of this data set (0.32 ± 0.04 ppbv) recorded at Ferndale in December 2012.

Seasonal variation in SO_2 concentration were investigated and determined that Jacobs recorded the highest average seasonal values for the months June to November as well as February and April while Southern Works recorded the highest seasonal average for the months January and March. For the month of May, three sites were approximately equal, namely, Wentworth, Jacobs and Southern Works. The locations Ferndale and Prospecton consistently reported the lowest seasonal SO_2 levels.

For all sites, the seasonal maximum was recorded in Winter (June – August) except for Prospecton which recorded a maximum in September. All sites recorded a minimum in Summer (December, January, February) except for Ferndale which recorded a minimum in early autumn (March). Four sites, Ferndale, Grosvenor, Wentworth and Settlers School reported a secondary maximum in September – October. This may indicate the impact of biomass burning at these locations as the effect of biomass burning on surface SO_2 levels is well documented (Agbo et al. 2021). Prospection is characterized by a single maximum in September, and it is possible that for this site, the influence of biomass burning dominates all other factors influencing the behaviour of SO_2 levels at this site.

It should be noted that few investigations of seasonal averages in Durban exist which emphasizes the importance of the present work. Two short studies (both less than two years in duration) reported seasonal behaviour at monitoring sites including some of those employed in this study. In Naidoo et al. (2007), SO_2 was monitored at 16 sites using ultraviolet fluorescence spectrometry methods (January 2004 – October 2005). In a more recent investigation, Tularam et al., 2020 employed Ogawa passive samplers to measure SO_2 levels at 40 monitoring sites, including 23 sites located in the south of Durban and 17 sites in the north (July 2015 to June 2016). Both of these studies reported strong seasonality with on average, high levels were recorded in winter (June – August) and the lowest levels in summer (December – February).

For the first time, this investigation presented analysis of seasonal averages for each site using a data set longer than 2 years. These seasonal averages illustrated that on average, cooler conditions generally favoured higher SO_2 levels and warmer conditions were characterized by lower SO_2 concentrations – which is in alignment with previous studies. However, there are exceptions to this rule with Prospection showing the greatest number of anomalous incidences to this general rule and Grosvenor the least number of exceptions.

Comparison of average difference between seasonal maximum and seasonal minimum (or average width of seasonal envelope), for years with all seasonal data available, and for each location, indicated that Jacobs and Wentworth formed a separate grouping from Southern Works, Grosvenor and Settlers School. Prospecton and Ferndale had the lowest average difference between seasonal maximum and minimum. The average difference or width of envelope for seasonal average is clearly a combination of factors and it seems that at a minimum, the groupings outlined above constitute locations where the same sets of factors dominate in terms of SO_2 fluctuations. The finding of this grouping is significant as it highlights the range of seasonal variation for industrial sites vs urban locations, with associated implications for personal exposure. It should be noted that the data sets for the calculated seasonal averages at each site are relatively small and that a larger, more complete data set would allow for a more accurate determination of the above grouping. This is clearly an area where further research is required.

Finally, through the use of a substantially larger dataset coupled with a range of averaging periods (yearly average, monthly average, seasonal variation, seasonal average) differences in exposure scenarios between industrial and urban locations have been determined together with the characterization of a more detailed exposure profile for individuals living and working in the greater Ethekwini municipality (2004-2019).

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Author contributions

B.L. Duigan: Conceptualization and drafting of manuscript, S.K. Sangeeetha: Contribution towards method and data analysis and V. Sivakumar: Supervision and advice on research. All authors contributed towards results and discussion and finalizing the manuscript for journal submission.

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