Measurement of atmospheric black carbon in the Vaal Triangle and Highveld Priority Areas

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Abstract
Atmospheric black carbon is an important atmospheric pollutant; it has impacts on human health and a strong climate impact. Black carbon particles are functionally defined by their optical properties (viz. characteristics in light absorption). As a result, black carbon particles are derived from a wide range of sources, but are largely the result of incomplete combustion processes. In order to quantify the atmospheric load of black carbon particles, multi angle absorption photometer (MAAP) instruments have been installed in 8 of the ambient air quality monitoring stations in the Vaal Triangle and Highveld Priority areas. Three of the instruments have been in operation since 2012 and the other 5 were installed in August 2013. This paper presents an analysis of the initial black carbon monitoring data. The impacts of seasonality and meteorological conditions as well as the relationship of the black carbon concentration to PM_{10} and $PM_{2.5}$ concentrations are discussed.

Keywords
Vaal Triangle Priority Area, Highveld priority area, black carbon

Introduction
Black carbon (BC) is a component of the atmospheric aerosol that is highly absorbent of visible light and is resistant to chemical transformation (Petzold et al. 2013). Black carbon is formally defined through its optical properties as "ideally light absorbing substances comprised of carbon", this definition does not take into account the formation processes. Black carbon is predominantly formed through the incomplete combustion of organic materials; however pyrolysis and dehydrogenation of wood and sugars under anaerobic conditions may also result in its formation (Petzold et al. 2013). The fact that BC is largely formed through combustion processes makes it a useful indicator for combustion sources of particulate matter.

Black carbon makes up an important component of the particulate matter less than 2.5µm in aerodynamic diameter ($PM_{2.5}$) fraction and therefore is implicated in the health impacts of $PM_{2.5}$. It has been suggested that the BC concentration is a better indicator of the risk to human health from PM than the PM_{10} or $PM_{2.5}$ mass concentration (Janssen et al. 2012). Further evidence suggests that $PM_{2.5}$ mixtures with a large BC component have a more adverse health effect than other mixtures (Anenberg et al. 2011).

The absorption of solar radiation by BC significantly enhances the heating of the atmosphere. The influence of BC can cause changes in cloud cover and surface albedo, therefore affecting the Earth's radiative budget both directly and indirectly. These aerosol particles not only influence atmospheric temperature but cause considerable changes in atmospheric chemistry. Black carbon particles have a relatively short lifespan within the atmosphere and for this reason tend to have more localised effects, impacting the regions closest to the source (Bauer et al. 2010).

Since atmospheric BC has such strong human health and climatic impacts, and is an indicator of combustion sources of particulate matter, it was deemed necessary to monitor the concentrations of BC at locations in the Vaal Triangle and Highveld Priority areas. This study examines a 12 month period (September 2013 to August 2014) for the measurement of BC in the Vaal Triangle and Highveld Priority Areas with the aim of characterising the ambient BC concentrations in terms of the seasonal, diurnal and air flow patterns.

Methods
During 2012, Multi Angle Absorption Photometer (MAAP; Thermo Scientific) instruments Model 5012 for the measurement of atmospheric BC were installed in the Witbank, Secunda and Zamdela monitoring stations. During August 2013, further instruments were installed at the monitoring stations in Diepkloof, Sharpeville, Sebokeng, Three Rivers and Kliprivier in the Vaal Triangle Network. The specific instrumentation used for this study is presented in Table 1. The instruments report to the South African Air Quality Information System (SAAQIS)

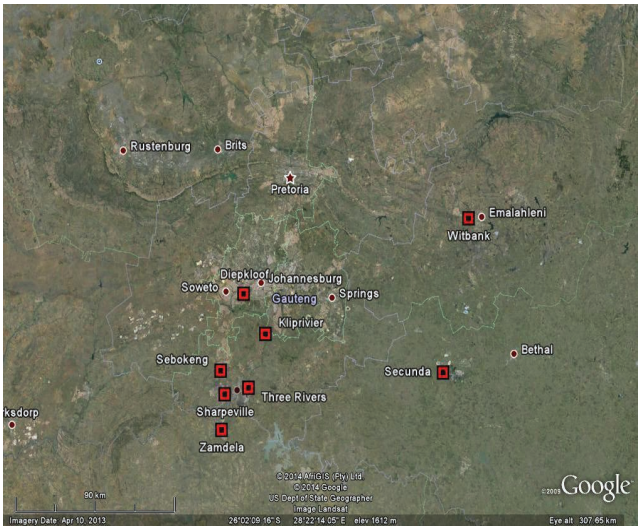

Figure 1: *Google Earth image showing locations of the monitoring stations*

(www.saaqis.org.za) at a 5 minute temporal resolution. In addition to the measurement of BC, all stations in the Vaal Triangle and Highveld Priority Area networks are instrumented for the measurement of PM_{10}, $PM_{2.5}$, SO_2, NO_x, CO, O_3, Benzene, Toluene and Xylene (BTX), and the meteorological parameters wind speed, wind direction, rainfall, temperature, pressure, humidity and solar radiation. All the data from the Vaal Triangle and Highveld networks are available from the SAAQIS, and has been validated to remove calibration periods, instrument drifts and spikes, all validation processes are detailed in the monthly network reports for the Vaal Triangle and Highveld Priority Area networks. These reports are available online on the SAAQIS website.

For this study, BC, PM_{10}, $PM_{2.5}$ mass concentration data and meteorological data for the period 1 September 2013-31 August 2014 was downloaded from the SAAQIS at an hourly temporal resolution for the Diepkloof, Sharpeville, Sebokeng, Zamdela, Three Rivers, Kliprivier Witbank and Secunda stations (Figure 1). The site and instrument specifications are presented in Table 1. The data was analysed using Excel and the "openair" package of R (Uria-Tellaetxe and Carslaw 2014).

It has been reported that there is an artefact in the MAAP instrument at high BC concentration (Hyvärinen et al. 2013). The correction suggested by Hyvärinen et al. (2013) was not applied for this study as not all of the parameters required for implementing the correction were logged, and the manufacturers did not recommend the implementation of the correction when inquiry was made. At high concentrations the instrument may under-report the BC concentration.

Table 1*: Site and instrument specifications*

Site	BC instrument	PM instrument	Site characteristics
Diepkloof	Thermo Model 5012 MAAP	Thermo FH62C14	Location in school in middle to low income residential with impacts from traffic light industry and domestic combustion
Sebokeng	Thermo Model 5012 MAAP	Thermo FH62C14	Location in community centre in low income residential with impact from metallurgical industry, and domestic combustion
Sharpeville	Thermo Model 5012 MAAP	Thermo FH62C14	Location in school in low income residential area with impact from metallurgical industry, and domestic combustion
Sharpeville	Thermo Model 5012 MAAP	Thermo FH62C14	Location in school in low income residential area with impact from chemical and petrochemical industry, and domestic combustion
Three Rivers	Thermo Model 5012 MAAP	Thermo FH62C14	Location in school in middle income residential area with impact from a coal power plant
Kliprivier	Thermo Model 5012 MAAP	Thermo FH62C14	Location in Police station in low income residential area with impact from traffic and domestic combustion
Witbank	Thermo Model 5012 MAAP	GRIMM EDM180	Location in school in low income residential area with impact from metallurgical industry, coal power generation and domestic combustion
Secunda	Thermo Model 5012 MAAP	GRIMM EDM180	Location in sports centre in low income residential area with impact from chemical and petrochemical industry, and domestic combustion

Results

The results for this study are divided according to the seasonal and diurnal effects; the impact of the wind direction and speed, and the relationship between the mass concentrations of BC and the other PM classes measured at the sites.

Average Concentrations

The average concentration of atmospheric BC is between 2.5 and 4.5 µg/m^3 for all stations, however hourly values of up to 20 µg/m^3 occurred at all sites except Three Rivers (Figure 2).

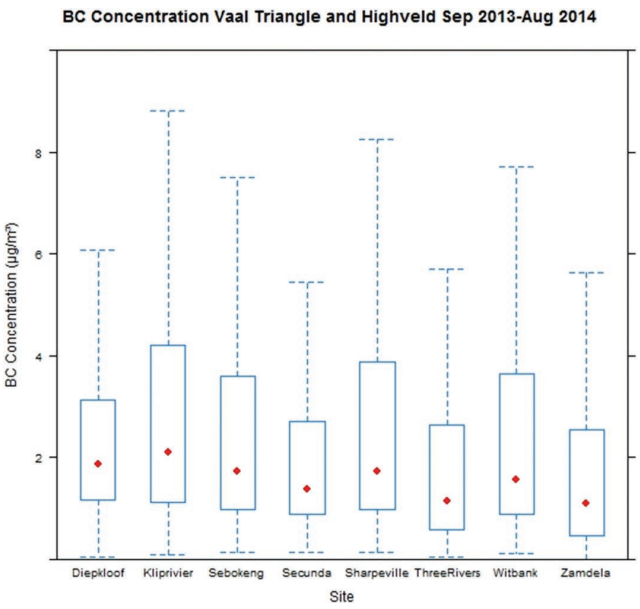

***Figure 2**: Average hourly BC concentration (µg/m^3) for all the sites for the period September 2013-August 2014*

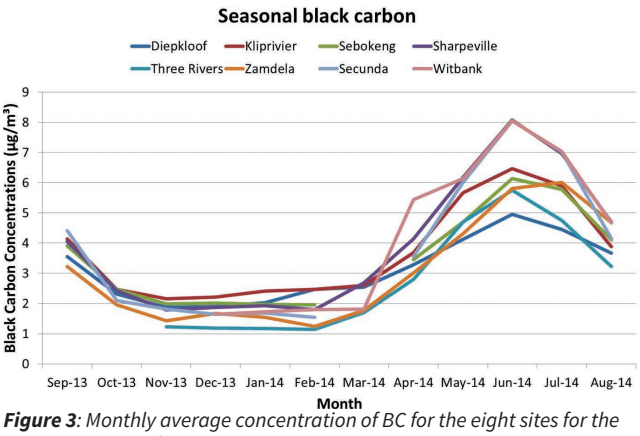

***Figure 3**: Monthly average concentration of BC for the eight sites for the monitoring period*

Seasonal and diurnal effects

A strong seasonal trend in the ambient concentration of atmospheric BC was observed (Figure 3). The concentration of BC increased significantly with the onset of winter. Increases in the ambient concentrations were observed during the months of April, May and June 2014, with reductions occurring from August. During the warmer months (October – March), the monthly average concentration remained fairly constant and similar between the stations. During the cooler months (April – September), stronger differences are observed between the sites, with higher concentrations being observed in the Witbank, Secunda and Sharpeville stations.

The "openair" calendar plot function for the Zamdela station during 2013 (Figure 4) illustrates that the BC mass concentration peaks during the cooler period (May – July as well as September) and remains fairly low during the warmer months. During the June and July period the daily average concentrations of BC vary with days of relatively low concentration following

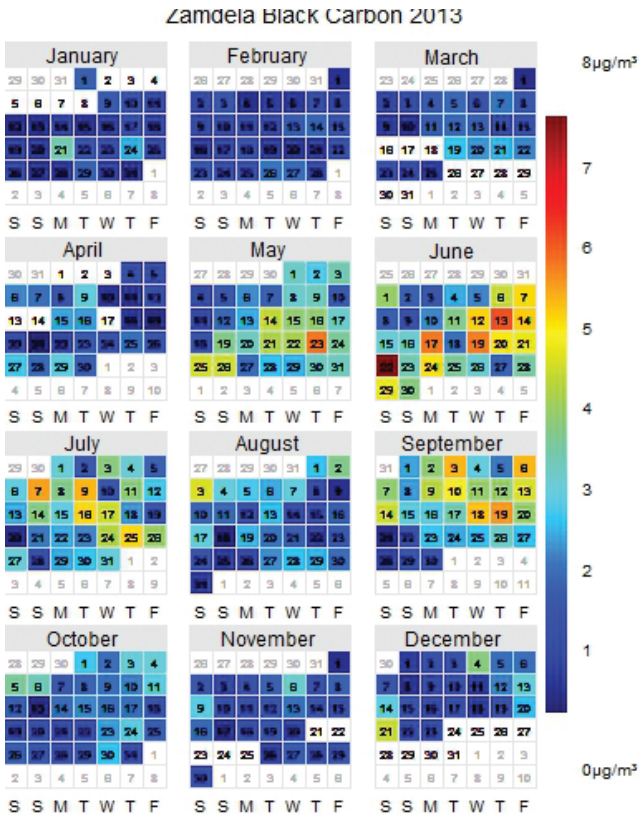

***Figure 4**: Calendar plot of the daily BC concentrations recorded at Zamdela for 2013*

periods of high concentration, many of these periods of low BC concentration are associated with the passing of a cold front over the interior of the country, particularly on the 2 June 2013, 8 June 2013, 3 July 2013 and 28 July 2013. During September 2013 there is an increase in the BC concentrations; this is potentially due to long range transport of biomass burning emissions.

The hourly profile of the ambient BC concentrations (for the entire time period) recorded at all the stations show a strong bimodal distribution (Figure 5) with peaks occurring in the mornings (5-8 am) and in the evenings (6-8 pm). The concentrations remain elevated during the night and then reduce during the day time. This pattern of increased concentrations is fairly typical of domestic burning emissions. This can be seen in more detail in Figure 6 which is a time variation plot of BC in Zamdela for the entire time period. The diurnal pattern of high BC concentration remains consistent across the days of the week, but the peak concentrations are reduced over the weekends and the morning peaks are spread out over a longer period on Saturday and Sunday, presumably due to people starting their activities later in the morning.

Impact of air flow

The "Polar Plot" function from "openair" plots the concentration of BC (in colour) in relation to the wind speed and wind direction (Figure 7). All the stations considered in this study show that local sources are important and high concentrations occur when there is a fairly low wind speed. Five of the stations in the

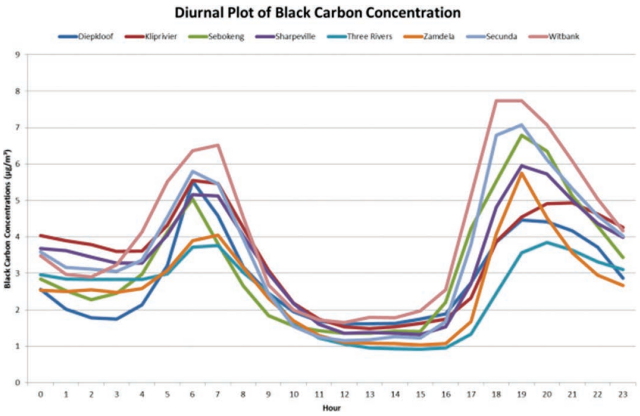

***Figure 5**: Diurnal characteristics of the ambient BC concentration*

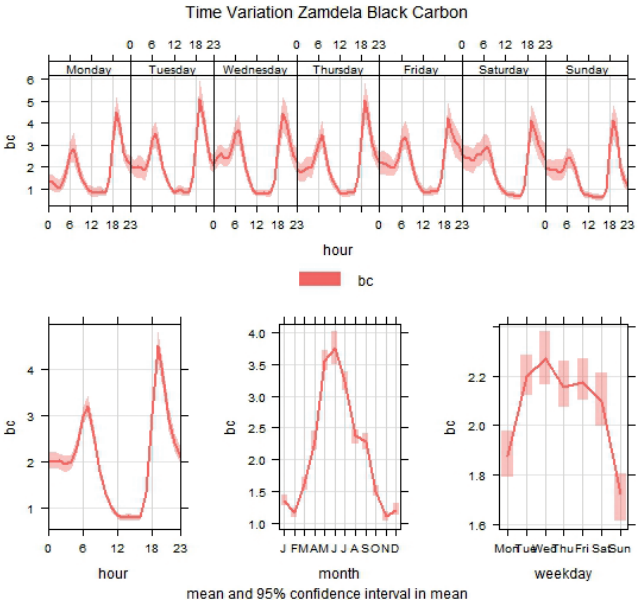

***Figure 6:** Time Variation Plot Zamdela BC. In this plot the top panel shows the diurnal pattern for each day of the week, while the lower panel shows the diurnal pattern for the year, the monthly average and the day of week concentrations. The mean for each time period is indicated by the red line with the 95% confidence interval in the mean shown in the shaded area.*

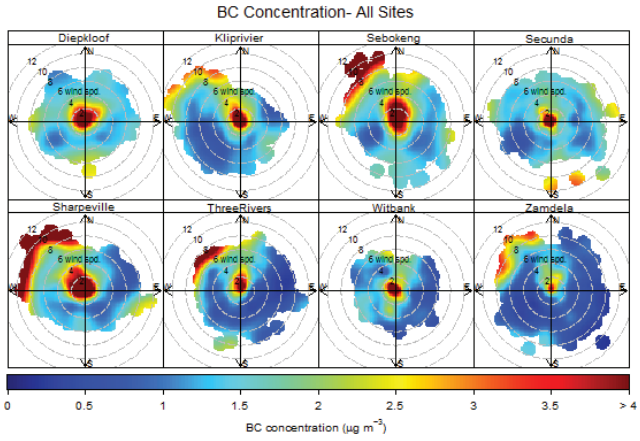

***Figure 7**: Polar plot of site specific BC concentration and meteorological conditions*

Vaal Triangle (Kliprivier, Sebokeng, Three Rivers, Sharpeville and Zamdela) also show high concentrations of BC associated with strong winds from the north-westerly directions sources to the north west of these stations may include the gold fields of Randfontein/ Carletonville and the Bojanala platinum belt, further analysis is required to identify potential sources. Zamdela shows very low BC concentrations associated with winds from the south and easterly sectors where there is very little industrial activity.

Relationship between BC, PM_{10} and $PM_{2.5}$

There is a strong relationship between the 1-hr mass concentrations of BC, and the concentrations of PM_{10} and $PM_{2.5}$ as shown in Figure 8. The mass concentrations of $PM_{2.5}$ and PM_{10} are plotted against each other while the BC concentration is represented by the colour of the point. It can be seen that as the concentrations of PM_{10} and $PM_{2.5}$ increase so do the concentrations of BC, however the BC concentration tracks more closely with the $PM_{2.5}$ values.

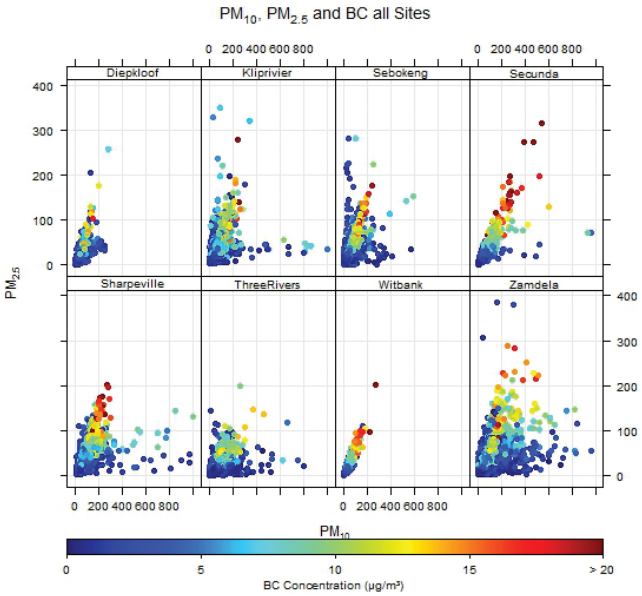

***Figure 8**: Scatter Plot 1-hr averaged mass concentrations of PM_{10}, $PM_{2.5}$ and BC*

Using the full time period of measurements (July 2012-June 2014) at Zamdela the monthly linear relationship between BC concentration and the $PM_{2.5}$ concentration was plotted using the linear relation function from "openair" (Figure 9). The "linear relation" function looks at the relation between two pollutants over differing time periods (in this case monthly) the error bars represent the 95% confidence interval while the red trend line represents the long term trend in the relationship. The ratio between BC and $PM_{2.5}$ changes seasonally with increases in the BC component during the winter months. The value of the plotted linear relationship is the ratio of BC:$PM_{2.5}$. While in general there is fairly good correlation between the concentrations of $PM_{2.5}$ and BC, the BC makes up a small portion of the total $PM_{2.5}$ concentration, typically less than 10%.

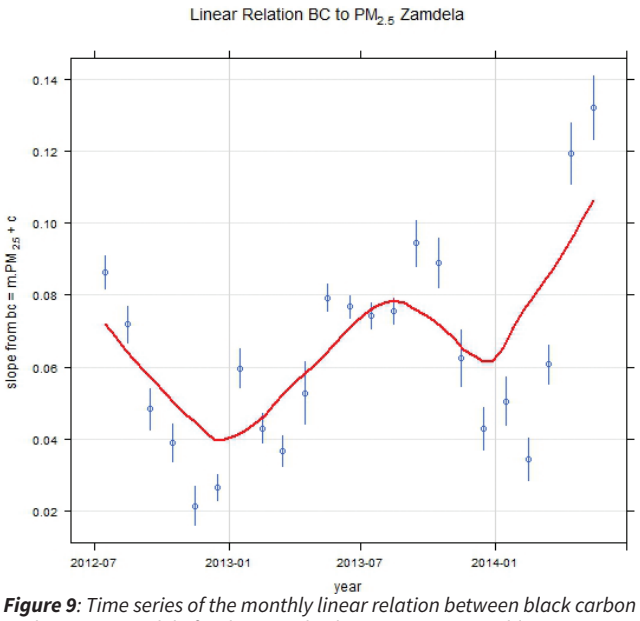

***Figure 9**: Time series of the monthly linear relation between black carbon and $PM_{2.5}$ at Zamdela for the period July 2012 to May 2014 blue points and error bars represent the monthly linear relationship between the parameters while the red line represents the trend in the monthly linear relationship observed*

Discussion

The ambient concentrations of BC are monitored at 8 ambient air quality monitoring stations in the Vaal Triangle and Highveld Priority Areas. The concentrations of BC show a distinct seasonal pattern, with higher concentrations occurring in the cooler months. This is similar to what has been reported previously in the North West Province (Venter et al. 2012). This observed seasonal trend is likely linked to greater emissions of BC from domestic burning and biomass burning sources, and the presence of highly stable atmospheric conditions, which reduce mixing. The diurnal profile shows a strong bimodal distribution with peaks in the early morning and evening. Such concentration profiles are indicative of either domestic combustion and/or traffic sources. Since the maximum BC concentrations occur between 5:00 and 7:00 in the morning and between 18:00 and 20:00 in the evening the predominant source may be domestic combustion as the maximum traffic flows are expected to occur later in the morning and finish earlier in the evening. This is confirmed when looking at the weekday diurnal profiles as the morning peak in BC concentration is spread over a greater time period as people tend to start their daily activities later on Sundays.

The analysis of the BC concentration in relation to the air flow indicates that for most of the stations local sources are important, however, the stations in the Vaal Triangle show high BC concentrations associated with strong winds from the north-west. In a previous study of ozone concentrations in the Vaal Triangle, high ozone concentrations were associated with biomass burning events and the approach of a cold front, drawing in air masses from the north west (Feig et al. 2014).

Black carbon accounts for approximately 6%-12% of the mass concentration of $PM_{2.5}$. The proportion of BC in the $PM_{2.5}$ fraction is impacted by season with a higher BC contribution occurring in the winter months, presumably due to the greater emissions from domestic and biomass burning sources.

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