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ATMOSPHERIC METHANE CHARACTERISATION OVER THE SOUTH AFRICAN INTERIOR

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The concentrations of atmospheric methane have an important impact on the global climate system and are important in the production of tropospheric ozone as it acts as an ozone precursor. The ambient concentrations of methane have increased more than 150% since the beginning of the industrial revolution and are currently greater than levels at any point within the last 800 000 years. The sources of atmospheric methane include natural sources such as wetlands, termites and oceans as well as anthropogenically influenced processes such as energy production, ruminant husbandry, waste disposal and treatment, and combustion processes. Methane is a long lived pollutant with an atmospheric lifetime of approximately 8.5 years, and a global warming potential 23 times that of CO_2. Due to the long atmospheric lifetime, ambient methane concentrations are influenced by both local and global processes. The CSIR and NWU have been operating a network of Cavity Ring Down Spectrometry (CRDS) instruments at Kwazamokuhle, Elandsfontein and Lephalale within the Highveld and Waterberg priority areas in the interior of South Africa, measuring at both regional background and low income residential sites. Results from these measurements are characterised in relation to potential local sources and temporal changes in the ambient concentrations that are measured.

Keywords: Methane, Highveld Priority Area, Waterberg Priority Area, Cavity Ring Down Spectroscopy.

1. Introduction

Atmospheric methane (CH_4) is the second most important greenhouse gas, it is believed that methane has been responsible for approximately 20%-30% of the global warming signal resulting from anthropogenic production of long lived greenhouse gases (Kirschke et al. 2013; Miller et al. 2013). The ambient concentrations of methane have increased more than 150% since the beginning of the industrial revolution and are currently greater than levels at any point within the last 800 000 years (Kirschke et al. 2013). Methane has an atmospheric residence time of approximately 8-10 years (Anenberg et al. 2012). Ambient concentrations of CH_4 are typically in the range of 1.8 ppm and have risen significantly since the start of the industrial revolution at which time the concentrations were 0.68-0.715 ppm (Miller et al. 2013). It had been thought that the rate of increase in the global CH_4 concentrations had stabilised and there was a period fairly constant CH_4 concentrations between 1992 and 2006, however since 2007 the global CH_4 concentrations have been increasing. This is especially evident in the southern tropics where the rise in CH_4 concentration has been above the global trend. Since 2007 the average CH_4 concentration at Ascension Island increased by >10 ppb per year (Kirschke et al. 2013).

Methane is produced through a number of natural and anthropogenic processes, including microbial metabolism in soil under anaerobic conditions (Conrad 1996; Krüger et al. 2013), enteric fermentation in ruminants and their manure (Moeletsi & Tongwane 2015), industrial processes (Thambiran & Diab 2011), waste water treatment (Miller et al. 2013) and the exploitation of fossil fuels such as coal and shale gas (Altieri & Stone 2016; Cohen & Winkler 2014).

While CH_4 is an important greenhouse gas, it has an important impact on atmospheric chemistry. By reacting with hydroxyl radicals, CH_4 reduces the oxidizing capacity of the atmosphere and generates ozone in the troposphere (Kirschke et al. 2013; Crutzen & Lelieveld 2001) hence it is a known ozone precursor and has the potential to impact the production of secondary organic aerosols, particularly in the smaller sub PM2.5 size classes. It has been estimated that the control of CH_4 production would have significant benefits in terms of improving both air quality and climate impacts (Anenberg et al. 2012; Thambiran & Diab 2011).

Due to the impact that CH_4 has on both air quality and climate it is important that an understanding of the emission sources is developed. It has been found that top down approaches to the estimation of CH_4 emissions can lead to a significant under estimation. In the USA Miller et al. (2013) used a combination of atmospheric data, available emissions inventories and inverse modelling techniques to derive spatially resolved source based CH_4 emissions, they found that the EDGAR and EPA emissions inventories had underestimated the emissions by between 40% and 70%.

In order to use techniques such as inverse modelling it is necessary to have long term and high quality measurements of atmospheric CH_4. However there is a paucity of CH_4 measurements in South Africa with the only continuous measurement site being Cape Point and a series of CH_4 flux measurements conducted in the Drakensberg (Krüger et al. 2013). To date, and to the best of our knowledge, there have been no published measurements of atmospheric CH_4 over the interior of South Africa. This study presents the preliminary results of ongoing measurements of CH_4 in the interior of South Africa that are being conducted by the CSIR and the Climate Research Group at NW University.

2. Methods and Materials

The CSIR and NWU have been operating a network of Picarro Cavity Ring Down Spectrometer instruments at Kwadela, Kwazomokuhle, Elandsfontein and Lephalale in the Highveld and Waterberg Air Quality Priority Areas, measuring at both regional background and low income residential sites. The instruments were placed at established air quality monitoring sites or deployed during comprehensive field campaigns. Campaign sites include Kwadela and Kwazamokuhle and permanent sites are at Eskom Air Quality monitoring stations at Lephalale and Elandsfontein (Table 1). In addition, measurements from Cape Point have been included for comparison to a well-established marine-influenced site that forms part of the Global Atmospheric Watch Program (GAW). A map depicting the locations is presented in Figure 1.

Figure 1 Location of methane measurement used

Measurements at the Kwadela site occurred during both the winter campaign of July-September 2014 and the summer campaign of February to April 2015. The Kwadela study focused on the impact of domestic combustion on air quality within a low income residential area located within the Highveld Priority area. The Kwadela site is situated approximately half way between Bethal and Ermelo and removed (~40-50 km) from any major point sources of pollution. Meteorological data was measured at site.

The Kwazamokuhle site is in a low income residential area outside of Hendrina in the Mpumalanga Highveld. The site has been operational since June 2015 and is intended to quantify the impact of emission offsetting activities on ambient air quality. Meteorological information was obtained from the South African Weather Service ambient air quality monitoring station in Hendrina located ~3 km to the south west.

The Elandsfontein site is an Eskom managed regional background site. The proximal land use is crop and livestock agriculture although there are numerous power plants and coal mines surrounding the station at a distance of greater than 20 km. During the period January – June 2016 the wind sensor was not functional and therefore the wind data was obtained from the South African Weather Service ambient air quality monitoring station in Hendrina located ~25 km to the north east.

The Lephalale site is located approximately 15 km to the SW of the town of Lephalale and south of the Matimba and Medupi power stations.

The Picarro instrument utilises the Cavity Ring Down Spectroscopy technique which is highly

accurate and highly stable. The nameplate precision at a 5 second averaging interval is <1 ppb for CH_4. The nameplate drift is less than 3 ppb per month.

Table 1 Location, atmospheric parameters measured, periods of operation and characteristics of each of the CH_4 measurement sites

Site	Co-Ordinates	Parameters Measured	Period installed	Site characteristics
Kwadela	26°27'47.84"S 29°39'48.34"E Alt: 1721m	CO_2,CH_4, SO_2, NO_x, CO, O_3, PM10, PM2.5, meteorology	16/07/2014 → 18/08/2015 11/02/2015 → 14/04/2015	Low income residential area, strong influence of domestic combustion, regional industry and power generation
Kwazomokuhle	26° 8'12.87"S 29°43'46.87"E Alt: 1652m	CO_2, CH_4, SO_2, NO_x, CO, O_3, PM10, PM2.5, meteorology	22/06/2015 → 12/05/2016	Low income residential area, strong influence of domestic combustion, regional industry and power generation
Elandsfontein	26°14'44.91"S 29°25'4.45"E Alt:1657m	CO_2, CH_4, SO_2, NO_x, CO, PM10, PM2.5, meteorology	14/01/2016 → 29/06/2016	Regional background station local influence of agricultural activities and regional industrial and power generation sources
Lephalale	23°44'21.05"S 27°32'22.85"E Alt: 901	CO_2, CH_4, SO_2, NO_x, CO, PM10, PM2.5, meteorology	14/04/2016 → 08/06/2016	Regional background station local influence of agricultural activities and regional industrial and power generation sources
Cape Point	34°21'12.06"S 18°29'23.02"E Alt: 140m	CO_2, CH_4, N_2O, SF_6, CO, ^{222}Rn, halocarbons, O_3, meteorology	20/03/2014 → 31/12/2015	Global Atmosphere Watch Station, measurement of atmospheric chemical constituents

3. Results and Discussion

The observations of CH_4 at the measurement sites have been in operation since early 2014. Figure 2 shows the periods where measurements have taken place and the overlap of observations up until the most recent data availability.

The observed CH_4 concentrations are fairly constant between sites and range from 1.75 ppm to 2.2 ppm (Figure 3). The lowest CO_4 concentrations are observed at the Cape Point site which primarily measures air masses of marine origin and represents a southern hemisphere background concentration. The concentrations measured at the inland sites represent the background concentrations with additional input from local terrestrial sources. Using a one way ANOVA significant differences ($p \leq 0.05$) in the CH_4 concentration exist between all the sites.

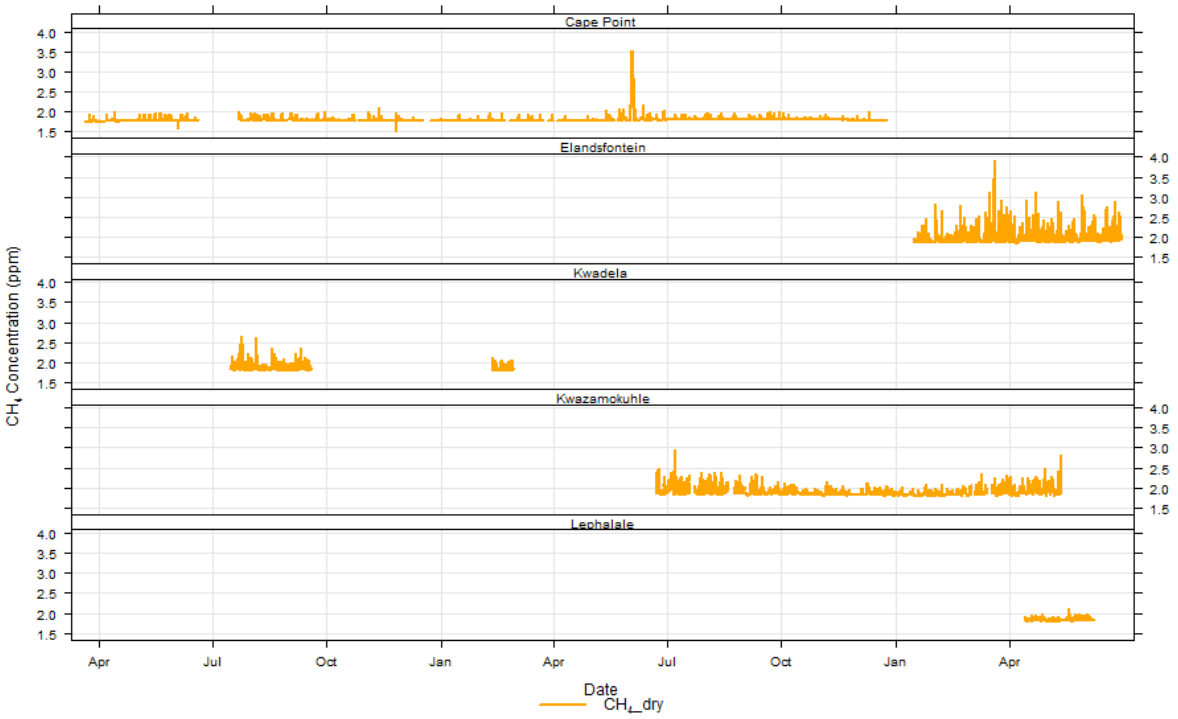

Figure 2. Time series of CH_4 measurements at the 5 sites from March 2014 – July 2016

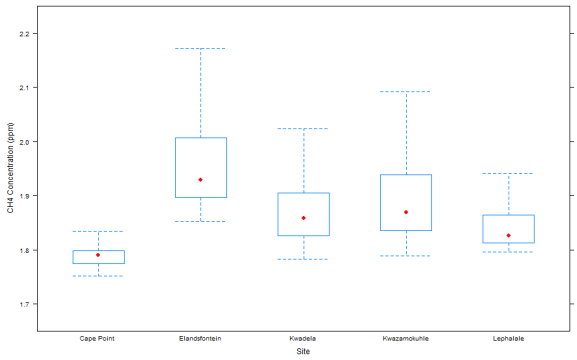

Figure 3. Box and Whisker plot of the observed CH_4 concentrations at the 5 monitoring sites. The red point represents the mean value; the solid lines represent the 25 and 75 percentile values and the dashed line the 5 and 95 percentile values.

There is an observable diurnal variation in the measured CH_4 concentrations that is consistent across all the inland sites (Figure 4). The CH_4 concentration is lowest during the day and increases at night. It is presumed that this is a function of the atmospheric mixing, so that with the development of stable night time conditions the CH_4 concentrations are able to increase as a result of ground level sources.

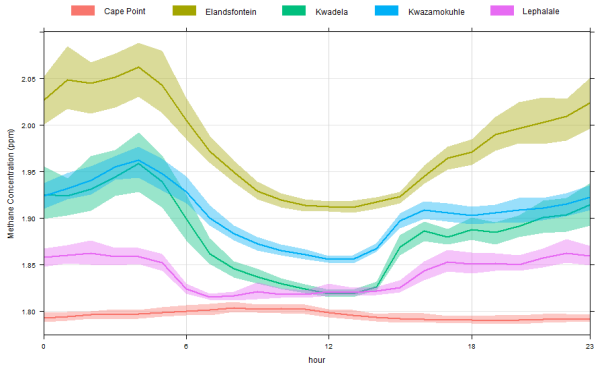

Figure 4. Diurnal variation of CH_4 concentration across all sites. The dark line represents the mean value with the colour bands representing the 95% confidence interval around the mean

The seasonal variation of the CH_4 concentration is presented in Figure 5. While a clear cycle cannot be observed at all the sites due to an insufficient monitoring period, a clear seasonal pattern is observable at Kwazamokuhle and Cape Point which each have at least a full year's measurement. The trend at Elandsfontein, Lephalale and Kwadela, where there are measurements for part of the year appear to follow the trends at Kwazamokuhle and Cape Point. An interesting feature is the spike in CH_4 concentrations observed in June, which is evident at all the sites. The cause of this is currently under further investigation.

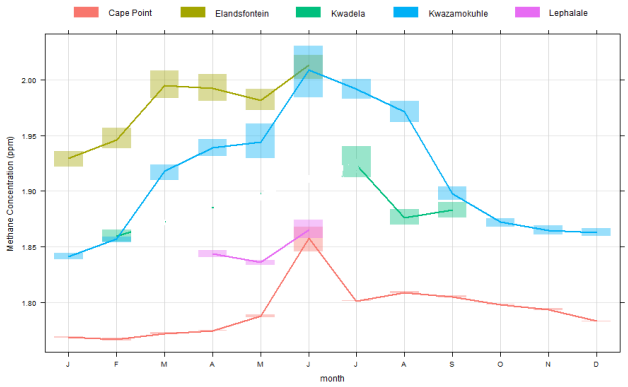

Figure 5. Methane seasonal cycle

The incorporation of wind data with the CH_4 concentration data allows one to identify potential sources of CH_4 near the monitoring site (Carslaw 2014; Uria-Tellaetxe & Carslaw 2014). This is shown for the inland sites for both the day-time (6:00-18:00) and night-time conditions (18:00-6:00) (Figure 6).

At the Elandsfontein and Kwadela sites higher CH_4 concentrations are associated with air movement from the north-westerly areas; the increase in concentration is most prominent at night. At Elandsfontein this accounts for an increase in the CH_4 concentrations of approximately 150 ppb. A potential source of these CH_4 emissions is the coal mining operations in the Ogies and Emalahleni area.

There is an additional local source of CH_4 at Kwadela associated with calm night time conditions to the NW of the town. This corresponds with the location of a lake and reed beds. No such clear CH_4 sources are observed at the Lephalale site.

For the Cape Point site there is a strong nocturnal signal from the NNE, which corresponds to the Cape Flats Waste Water Treatment Works Figure 7.

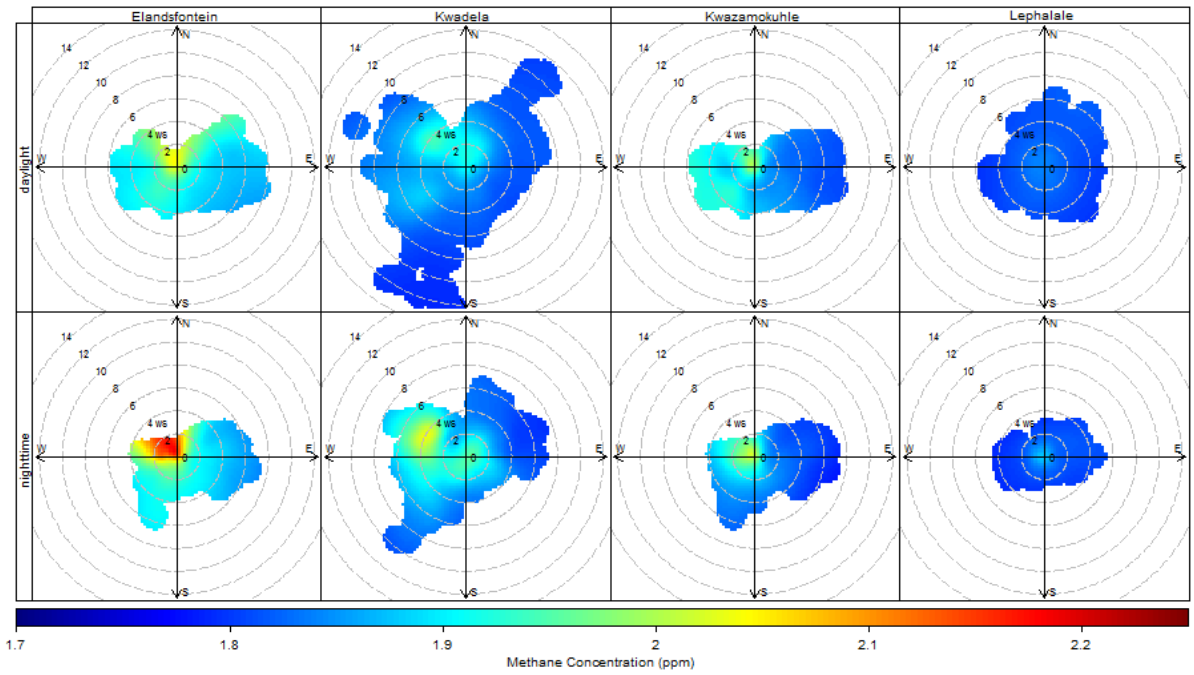

Figure 6 Polar plot of CH_4 concentration Elandsfontein, Kwadela, Kwazamokuhle and Lephalale, top row day-time and bottom row night-time, rings indicate the measured wind speed and concentration is indicated by the colour

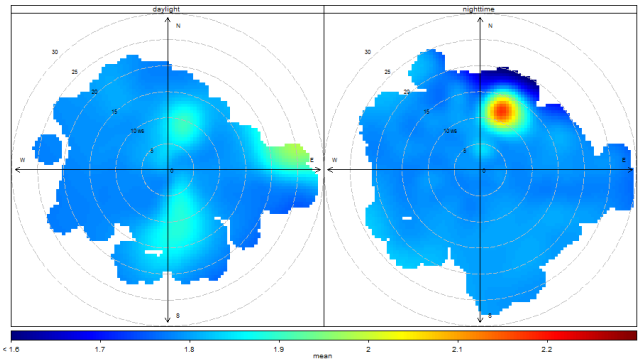

Figure 7 Polar plot of CH_4 concentration at Cape Point left day-time and right night-time, rings indicate the measured wind speed and concentration is indicated by the colour

4. Conclusion

This study shows preliminary results from a network of CH_4 measurements conducted in the Highveld and Waterberg air quality priority areas. Average values across the sites are fairly similar and range between 1.75 and 2.2 ppm. A clear diurnal pattern in the concentrations is observed at all the sites, with an increase in the CH_4 concentrations observed during the night time.

It appears that there is a seasonal cycle with the CH_4 concentrations, however the length of measurements is not sufficient to elucidate the seasonal cycles fully.

These measurements provide the basis for quantifying changes in the ambient CH_4 concentrations as a result of mining, agriculture and waste treatment and provide a tool for identifying local CH_4 sources and providing an independent estimate of CH_4 emissions.

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6. References

Altieri, K.E. & Stone, A., 2016. Air pollutant emissions inventory for NOx from the development of unconventional natural gas wells in the Karoo basin, South Africa. *Atmospheric Environment*, 129, pp.34–42. Available at: http://dx.doi.org/10.1016/j.atmosenv.2016.01.021.

Anenberg, S.C. et al., 2012. Global air quality and health co-benefits of mitigating near-term climate change through methane and black carbon emission controls. *Environmental Health Perspectives*, 120(6), pp.831–839.

Carslaw, D.C., 2014. Editorial Modern tools for air quality data analysis – openair. *Clean Air Journal*, 24(2), pp.4–5.

Cohen, B. & Winkler, H., 2014. Greenhouse gas emissions from shale gas and coal for electricity generation in South Africa. *South African Journal of Science*, 110(3), pp.2–6.

Conrad, R., 1996. Soil microorganisms as controllers of atmospheric trace gases (H2, CO, CH4, OCS, N2O, and NO). *Microbiological reviews*, 60(4), pp.609–40. Available at: http://www.pubmedcentral.nih.gov/articlerender.fcgi?artid=239458&tool=pmcentrez&rendertype=abstract.

Crutzen, P.J. & Lelieveld, J., 2001. Human impacts on atmospheric chemistry. *Annual Review of Earth and Planetary Sciences*, 29(1), pp.17–45. Available at: http://www.annualreviews.org/doi/abs/10.1146/annurev.earth.29.1.17 [Accessed July 14, 2011].

Kirschke, S. et al., 2013. Three decades of global methane sources and sinks. *Nature Geoscience*, 6, pp.813–823.

Krüger, J.P. et al., 2013. Greenhouse gas emission peaks following natural rewetting of two wetlands in the southern Ukhahlamba-Drakensberg Park, South Africa. *South African Geographical Journal*, 96(2), pp.113–118. Available at: http://dx.doi.org/10.1080/03736245.2013.847798.

Miller, S.M. et al., 2013. Anthropogenic emissions of methane in the United States. *Proceedings of the National Academy of Sciences of the United States of America*, 110(50), pp.20018–22. Available at: http://www.pubmedcentral.nih.gov/articlerender.fcgi?artid=3864315&tool=pmcentrez&rende

rtype=abstract.

Moeletsi, M.E. & Tongwane, M.I., 2015. 2004 methane and nitrous oxide emissions from manure management in South Africa. *Animals*, 5(2), pp.193–205.

Thambiran, T. & Diab, R.D., 2011. Air quality and climate change co-benefits for the industrial sector in Durban, South Africa. *Energy Policy*, 39(10), pp.6658–6666. Available at: http://dx.doi.org/10.1016/j.enpol.2011.08.027.

Uria-Tellaetxe, I. & Carslaw, D.C., 2014. Conditional bivariate probability function for source identification. *Environmental Modelling and Software*, 59, pp.1–9. Available at: http://dx.doi.org/10.1016/j.envsoft.2014.05.002.