

Source region plume characterisation of the interior of South Africa as observed at Welgegund

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1. Introduction

Before 2005, air quality measurements in South Africa were mainly performed by industries for compliance monitoring. These data sets were not generally available in the public domain and in most cases not scrutinised by peer reviewing. Other atmospheric studies have mainly been limited to short-term intensive campaigns such as SAFARI-92 and SAFARI 2000, or long-term campaigns with less comprehensive sets of instrumentation. The Cape Point Global Atmospheric Watch station is the most comprehensive long-term station in South Africa. However, the Cape Point GAW station is not regionally representative, since it is mostly influenced by marine background.

In order to generate comprehensive long-term data sets that are regionally representative, the University of Helsinki (Finland), the North-West University (South Africa) and the Finnish Meteorological Institute (Finland) deployed a mobile measurement trailer in 2006 (e.g. Vakkari et al., 2013; Venter et al., 2012). A two year measurement campaign (2009 to 2010) was also initiated as part of an European Union project EUCAARI (e.g. Laakso et al., 2012). In mid 2010, the afore-mentioned mobile measurement station was moved to a permanent location approximately 30 km north of Potchefstroom. Additional instruments were also added. This resulted in the establishment of a very comprehensively equipped permanent atmospheric measurement station.

In this paper air masses that have passed over the major source regions of the interior of South Africa, before arriving at the Welgegund station are characterised in term of trace gases.

2. Experimental

2.1 Methods

Currently installed, continuously operating equipment at Welgegund includes trace gases (SO₂, NO/NO_x, O₃, CO), PM₁₀ aerosol mass, aerosol light absorption (for black carbon), aerosol light scattering, aerosol number size distribution (12-840nm), ion number size distribution (0.4-40nm), direct and reflected solar radiation, flux measurements (H₂O,

CO₂ and sensible heat), soil measurements (temperature and moisture at different depths) and meteorological parameters (temperature, relative humidity, wind speed and direction, precipitation and vertical temperature gradient). Additional campaign based measurements have also been conducted, e.g. a one year PM₁ Aerosol Chemical Speciation Monitor (ACSM) campaign, a six months vertical column measurement campaign of NO₂, O₃, CH₂O and H₂O, a one year VOC measurement campaign and a one year GCXGC-TOFMS analysis campaign of organic components in aerosols. One year atmospheric trace metal and Cr(VI) studies are currently in progress. Vegetation surveys are also conducted on various transects around the site four times per year and ad hoc soil chemical analyses have also been conducted to enhance interdisciplinary cooperation. Recently (September 2012) a ceilometer was installed to help resolve issues related to the layered structure of the atmosphere above the South African Highveld.

2.2 Air mass history

The air mass history was determined by calculating back trajectories with the Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model (version 4.8). All back trajectories were calculated for 96 hours, arriving every hour at a height of 100m throughout the entire measurement period.

2.3 Spatial classification of source regions

Several source regions, as observed from Welgegund, were defined, i.e. the Regional Background (Back), western Bushveld Igneous Complex (WBIC), the Jhb-Pta megacity (MegaC), the Vaal Triangle (VaalT), the Mpumalanga Highveld (MpHV) and the combined Anticyclonic Recirculation-Eastern Bushveld Igneous Complex (Anti-EBIC). These source regions are indicated in Figure 1. More detail on how these source regions were defined will be included in a future paper.

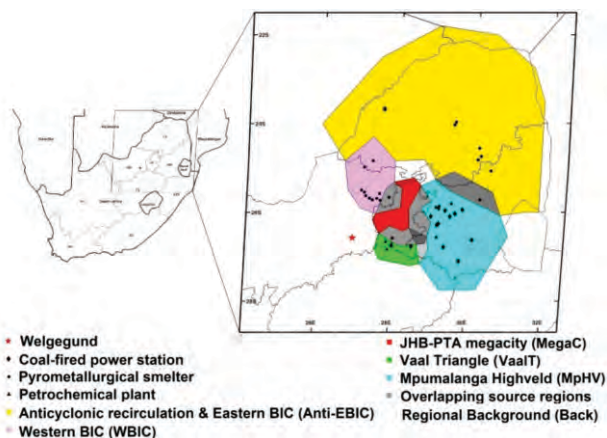


Figure 1. Regional map indicating large point sources in the industrial hub of South Africa and source regions impacting on the Welgegund measurement station as defined in this paper. Areas in grey indicate overlapping areas.

2.4 Linking air mass history with measurements at Welgegund

In order to establish a link between air masses arriving at Welgegund after passing over the defined source regions and the measurements conducted, the time of arrival of calculated back trajectories were correlated to the time periods of the measurements. Since back trajectories were calculated to arrive at Welgegund on the hour, the data collected during the half hour before and half hour after the arrival time of the back trajectory was allocated to that specific trajectory arrival time.

3. Results

3.1 Allocation of air masses

For the period considered (06/2010-04/2012), 16 416 back trajectories were calculated and classified. 75.7 % of all trajectories could be classified as passing over one of the six source regions defined. The remaining 24.3% of trajectories passed over multiple source regions. Data collected during such periods were not included in further data analysis.

A substantial fraction of the air masses arriving at Welgegund passed over the Back (27.59 %), the WBIC (23.97 %), the Anti-EBIC (16.22 %) and the MegaC (5.58 %) source regions. However, only 1.87 % (307 trajectories) and 0.51% (83 trajectories) of the hourly arriving air mass back trajectories passed over the VaalT and the MpHV regions, respectively.

3.2 Trace gas source region plume characterisation

The NO and NO₂ characterisations of the air mass source region histories are presented in Figure 2. Similar figures for the other trace gases measured at Welgegund, i.e. O₃, SO₂, CO, will be presented in a future paper.

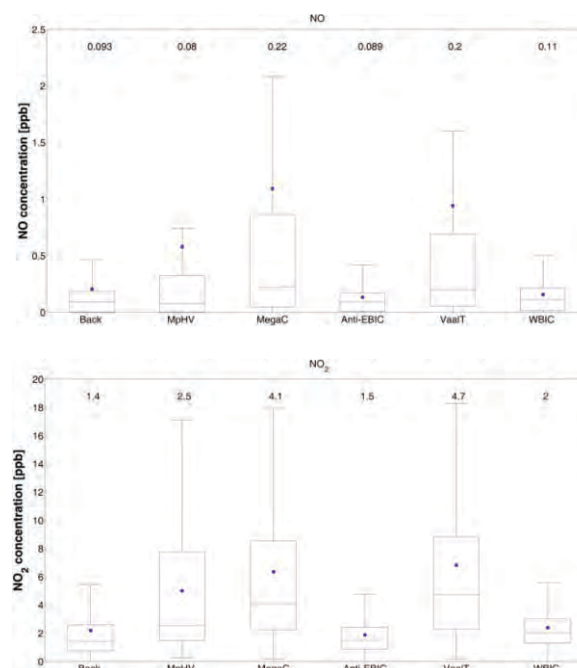


Figure 2. NO and NO₂ source region plume characterisation. The red line of each box is the median (value displayed below the top edge), the top and bottom edges of the box the 25th and 75th percentiles and the whiskers $\pm 2.7\sigma$.

Median NO levels were the highest in air masses that had passed over the MegaC and VaalT source regions, i.e. 0.22 and 0.2 ppb, respectively. However, these source region air masses also had the largest spread in NO levels. Median NO levels in air masses that had passed over the other source regions were substantially lower than the afore-mentioned two source regions. NO levels measured within the large anthropogenic source regions in South Africa are substantially higher than that reported here, e.g. Collett et al. (2010) reported monthly average NO values between 1 and 7 ppb at Elandsfontein, which is situated within the MpHV source region. However, lower values can be expected at Welgegund, since oxidation of NO during transport will reduce levels substantially. Biomass burning (veld fires) occurring upwind of Welgegund resulted in high NO levels. However, specific analyses of such events fall beyond the scope of this paper.

Air masses that had passed over the MpHV, the MegaC and the VaalT source regions had the highest median NO₂ concentrations, i.e. 2.5, 4.1 and 4.7 ppb, respectively. As expected, these NO₂ levels were lower than levels measured within these source regions, due to oxidation during transport to Welgegund, e.g. Lourens et al. (2011) reported annual average NO₂ levels of up to 8.6 ppb in the MpHV source region. Median NO₂ concentrations were the lowest in air masses that had passed over the Back source region, i.e. 1.4 ppb. This was expected, since the Back source region contained no large combustion point sources apart from biomass combustion that is endemic to southern Africa.

However, this NO₂ level (1.4 ppb) is still elevated above the 0.34 ppb 10-year average NO₂ level reported for a remote background station at the Estosha pan in Namibia (Martins et al., 2007). This indicates that the Back source region is not totally without anthropogenic emissions. Venter et al. (2012) recently indicated that household combustion for space heating and cooking in informal settlements could contribute to NO₂ pollution. Additionally, biomass burning events are also sources of NO and NO₂. These events occur across the entire southern Africa, hence pollutant levels in the Back source regions will also be influenced. Air masses that had passed over the Anti-EBIC and WBIC source regions only had slightly higher median NO₂ concentrations than the Back source region. This confirms that the Anti-EBIC source region have a much less dense occurrence of large point sources, but might also additionally indicate that NO and NO₂ had substantially been oxidised to nitrate (NO₃) during the transport of air masses via the anticyclonic recirculation from the other source regions. This will be further explored when the non-refractive PM₁ chemical data is discussed in a future paper.

The differences in the NO and NO₂ levels measured in air masses that had passed over the various anthropogenic source regions can possibly also partially be explained by the nature of the sources within each of these regions. The large point sources in the WBIC almost exclusively consist of pyrometallurgical smelters that produce metals from ores via reducing processes. This is in contrast with large point sources in the MpHV and the VaalT source regions, where oxidative combustion point sources dominate. The air mass history from MegaC is also strongly influenced by the MpHV with its combustion type point sources. Additionally, large traffic volumes and household combustion contribute to NO and NO₂ pollution from the MegaC (Lourens et al., 2012). Of the three dominant NO and NO₂ source regions, i.e. the MpHV, the MegaC and the VaalT, the MpHV source region had the lowest NO/NO₂ ratio. This was expected since this source region's back trajectories on average had to travel further before reaching Welgedund, leading to increased oxidation from NO to NO₂.

O₃ levels were relatively high irrespective of the source region air mass history. Plumes from all the source regions had O₃ median values between 30 and 40 ppb, with the 75th percentile of all the source regions, with the exception of the Back source region, exceeding the 40 ppb threshold often used as an indicative value for O₃ impacts on crops (Zunckel et al., 2006). Even the WBIC and the Anti-EBIC source regions that have less NO₂ point sources had quite high O₃ levels. This is likely due to the aging of the O₃ precursor rich air masses via the dominant anticyclonic recirculation pattern, leading to high O₃ levels, even from source regions that had lower NO₂ levels. Additionally, the release of CO that is also a known O₃ precursor specie, is not restricted to the

anthropogenic source regions, since biomass combustion occurs on a regional scale. The O₃ results presented here further supports the suggestion by Venter et al. (2012) that O₃ pollution is currently the most wide-spread air quality problem in South Africa.

SO₂ levels in the air masses arriving from the MpHV, the MegaC and the VaalT were substantially higher than that of the other source regions. The dominance of the afore-mentioned three anthropogenic source regions can be explained, since the MpHV and the VaalT have large combustion point source that utilise low grade coal, resulting in substantial SO₂ pollution (Lourens et al., 2011; Collett et al., 2010). Additionally, the MegaC is strongly influenced by the MpHV and household combustion of low grade coal for space heating is also a common occurrence in the low income settlement areas of the megacity (Lourens et al., 2012). Air masses that had passed over the WBIC had relatively low SO₂ levels, notwithstanding that especially the PGM smelters in this source region could have substantial SO₂ emissions. However, most of these PGM smelters have implemented de-SOx technologies over the last decade resulting in lower SO₂ pollution. This is in contrast to the large point sources in the MpHV and the VaalT, which have not yet implemented de-SOx technologies. SO₂ levels in air masses arriving from the Back source region were the lowest, i.e. 0.23 ppb. This concentration compared well with the SO₂ levels reported for a remote background station at the Estosha pan in Namibia (Martins et al., 2007).

CO levels from the various source regions did not differ as substantially as the above-discussed NO, NO₂ and SO₂ levels. CO originates primarily from biomass combustion that is a regional phenomenon impacting on all source regions, even the Back source region. However, most biomass combustion (veld fires) occurs in the sector between north and south in the region east of Welgedund, due to the differences in vegetation types. This could possibly contribute to the lower levels of CO measured in air masses that had passed over the Back source region. Additionally, the more anthropogenically influenced source regions also have industrial activities that release CO, e.g. combustion point sources. As with most of the other trace gasses discussed, the MpHV, MegaC and the VaalT source regions again had the highest levels. CO levels should not only be considered in isolation, since CO is a well-known O₃ precursor specie.

3.3 Additional source region plume characterisations

In addition to the trace gas source region plume characterisation discussed in the previous section, source region plume characterisations of PM₁₀, sub-micron aerosol particle number concentration, black carbon, aerosol optical properties, as well as non-refractive PM₁ chemical composition will be discussed in a full paper that will be published soon.

4. Conclusions

The strategic positioning of the Welgegund measurement site, i.e. positioned to be able to sample regionally representative background, as well as air masses passing over all the major anthropogenic source regions of the industrial hub of South Africa, was demonstrated. The comprehensiveness and integrated nature (including chemical, physical, botany, entomology) of measurements being conducted at the Welgegund measurement site is unprecedented in South Africa. However, the true value of such measurements will only be realised if this measurement effort can be sustained on a permanent basis in future.

5. Acknowledgments

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6. References

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