



# Re-evaluating the NO<sub>2</sub> hotspot over the South African Highveld

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Globally, numerous pollution hotspots have been identified using satellite-based instruments. One of these hotspots is the prominent NO<sub>2</sub> hotspot over the South African Highveld. The tropospheric NO<sub>2</sub> column density of this area is comparable to that observed for central and northern Europe, eastern North America and south-east Asia. The most well-known pollution source in this area is a large array of coal-fired power stations. Upon closer inspection, long-term means of satellite observations also show a smaller area, approximately 100 km west of the Highveld hotspot, with a seemingly less substantial NO<sub>2</sub> column density. This area correlates with the geographical location of the Johannesburg–Pretoria conurbation or megacity, one of the 40 largest metropolitan areas in the world. Ground-based measurements indicate that NO<sub>2</sub> concentrations in the megacity have diurnal peaks in the early morning and late afternoon, which coincide with peak traffic hours and domestic combustion. During these times, NO<sub>2</sub> concentrations in the megacity are higher than those in the Highveld hotspot. These diurnal NO<sub>2</sub> peaks in the megacity have generally been overlooked by satellite observations because the satellites have fixed local overpass times that do not coincide with these peak periods. Consequently, the importance of NO<sub>2</sub> over the megacity has been underestimated. We examined the diurnal cycles of NO<sub>2</sub> ground-based measurements for the two areas – the megacity and the Highveld hotspot – and compared them with the satellite-based NO<sub>2</sub> observations. Results show that the Highveld hotspot is accompanied by a second hotspot over the megacity, which is of significance for the more than 10 million people living in this megacity.

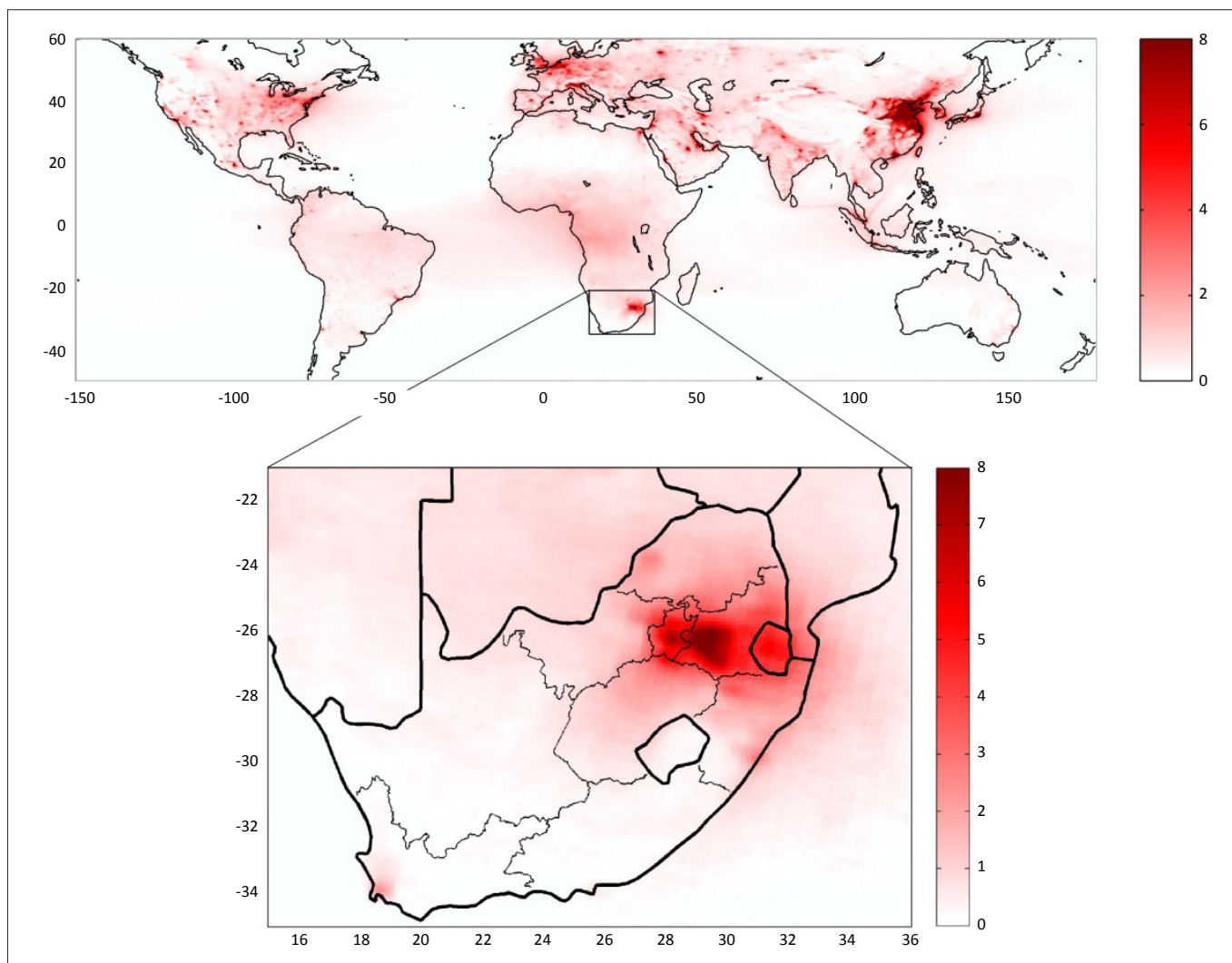
## Introduction

Satellite-based instruments such as the Global Ozone Monitoring Experiment (GOME 1 & 2),<sup>1</sup> the Scanning Imaging Absorption spectroMeter for Atmospheric CHartographY (SCIAMACHY)<sup>2</sup> and the Ozone Monitoring Instrument (OMI)<sup>3</sup> have been used to quantify trends of various atmospheric trace gas concentrations on a global scale.<sup>4</sup> Of interest here are the observations of tropospheric nitrogen dioxide (NO<sub>2</sub>) column densities<sup>5,6,7,8</sup> and the identification of global NO<sub>2</sub> pollution hotspots.<sup>9,10</sup> NO<sub>2</sub> plays a key role as a precursor for tropospheric ozone (O<sub>3</sub>), aerosol nitrate (NO<sub>3</sub><sup>-</sup>), and the hydroxyl radical (OH<sup>\*</sup>).

One prominent NO<sub>2</sub> hotspot seen in global maps from satellite retrievals is over the South African Highveld (referred to hereafter as the Highveld hotspot). The tropospheric NO<sub>2</sub> column density of this area (shown in Figure 1) is comparable to that observed for central and northern Europe, eastern North America and south-east Asia.<sup>9</sup> In addition to the well-known source of NO<sub>2</sub> – 11 coal-fired power stations – there are also other widespread area sources, such as road transport, biomass burning and human settlement, as well as other large point sources including metallurgical, mining and petrochemical operations.<sup>11</sup> A total of 90% of the industrial NO<sub>x</sub> emissions in South Africa originate from the Highveld hotspot.<sup>12</sup> In a recent assessment of global emissions, South African emissions amounted to 420 kt/year of nitrogen, which constituted 1.6% of global anthropogenic NO<sub>2</sub> emissions.<sup>13</sup>

Located 100 km west from the Highveld hotspot is the largest conurbation in southern Africa, which developed around Johannesburg and Pretoria, with a population of over 10 million.<sup>14</sup> Conurbations of this magnitude have been defined as Megacities.<sup>15</sup> The Johannesburg–Pretoria conurbation or megacity is the central hub of economic activity and growth in southern Africa. The main pollution sources in this conurbation are traffic emissions, biomass and domestic combustion (space heating and cooking), and industrial activities.<sup>16</sup>

In Figure 1, an area smaller than the Highveld hotspot, with a less substantial NO<sub>2</sub> column density, is seen to correspond with the geographical location of the megacity. Ground-based measurements conducted in this study indicate that satellite observations may underestimate the importance of this conurbation to local NO<sub>x</sub> pollution as a result of specific diurnal cycles of



Measurements were obtained for 2003-2009 from the Scanning Imaging Absorption Spectrometer for Atmospheric Chartography (SCIAMACHY).

**FIGURE 1:** Satellite image of the tropospheric vertical  $\text{NO}_2$  column (in  $10^{15}$  molecules/ $\text{cm}^2$ ) over the South African highveld.

emissions. However, ground-based measurements cannot be compared directly to satellite-based column measurements because satellite measurements reflect on the total column, whereas ground-based measurements do not indicate the total column density. Nonetheless, the aim of this study was to evaluate the potential contribution of the megacity to the  $\text{NO}_2$  Highveld hotspot. The diurnal cycles of ground-based measurements of  $\text{NO}_x$  were evaluated for the megacity and the Highveld hotspot, and compared to satellite-based  $\text{NO}_2$  observations.

## Satellite- and ground-based observations

### Satellite retrievals

Multi-year  $\text{NO}_2$  retrievals obtained from (1) SCIAMACHY (Max Planck Institute retrieval,<sup>8</sup> for the years 2003–2009) and (2) OMI (Dutch OMI  $\text{NO}_2$  product DOMINO (v1.02)<sup>7</sup>, available from the Tropospheric Emission Monitoring Internet Service for the years 2005–2009) were used. Both instruments orbit earth in a polar, sun-synchronous orbit, with a local South African observation time of 10:00 (SCIAMACHY) or

13:30 (OMI). Thus, the comparison of  $\text{NO}_2$  measurements from SCIAMACHY and OMI holds limited information on diurnal cycles.<sup>17</sup>

From the spectral measurements of the satellite instruments, slant column densities (SCDs), that is, integrated concentrations along the effective light path, of  $\text{NO}_2$  were derived. The stratospheric slant column is subtracted from the total slant column, yielding the tropospheric slant column density (TSCD).<sup>7,8</sup> For translation of the TSCD into a tropospheric vertical column density, tropospheric air mass factors (AMF) are generally needed. This translation requires prior assumptions on the ground albedo, as well as the vertical profiles of aerosols, clouds and  $\text{NO}_2$ , which makes it the most uncertain step within satellite retrievals.

To compare SCIAMACHY and OMI data sets meaningfully (including differences in viewing geometry, local time, ground pixel size and the AMF calculations), we (1) focused on TSCDs (which are far less uncertain than TVCDs) and (2) investigated relative patterns (i.e. ratios of TSCDs for the megacity versus the Highveld). This approach mostly



eliminated potential systematic biases caused by the use of different satellite instruments and retrievals.

## Ground-based observations

Hourly near-surface concentrations of  $\text{NO}_2$  were obtained from four air-quality monitoring stations within the megacity and at five stations in the Highveld hotspot for a period of 3 months in autumn 2009 (March to May). These stations were specifically chosen as their locations are regarded to be representative of the ambient concentrations of these areas.

Figure 2 indicates the locations of these measurement stations within the megacity and the Highveld hotspot. These stations are operated in the megacity and the Highveld hotspot by the town council of Johannesburg and the industries, respectively. Apart from their strategic positioning, these stations also complied with the South African Air Quality Information System quality-control requirements. Data received were also manually checked to identify possible errors and outliers.

Ground-based  $\text{NO}_2$  measurements were made with chemiluminescence instruments using molybdenum oxide converters.  $\text{NO}_2$  is catalytically converted to  $\text{NO}$  on a heated molybdenum surface and then measured as  $\text{NO}$  by means of chemiluminescence in the reaction with ozone. This method not only converts the  $\text{NO}_2$  to  $\text{NO}$ , but also partially converts other oxidised nitrogen compounds and can therefore cause overestimations of  $\text{NO}_2$  by between 17% and 30%.<sup>18</sup> However, this technique is still widely applied and measurements with these instruments currently provide the only available near-surface data for the regions under discussion.

## Results

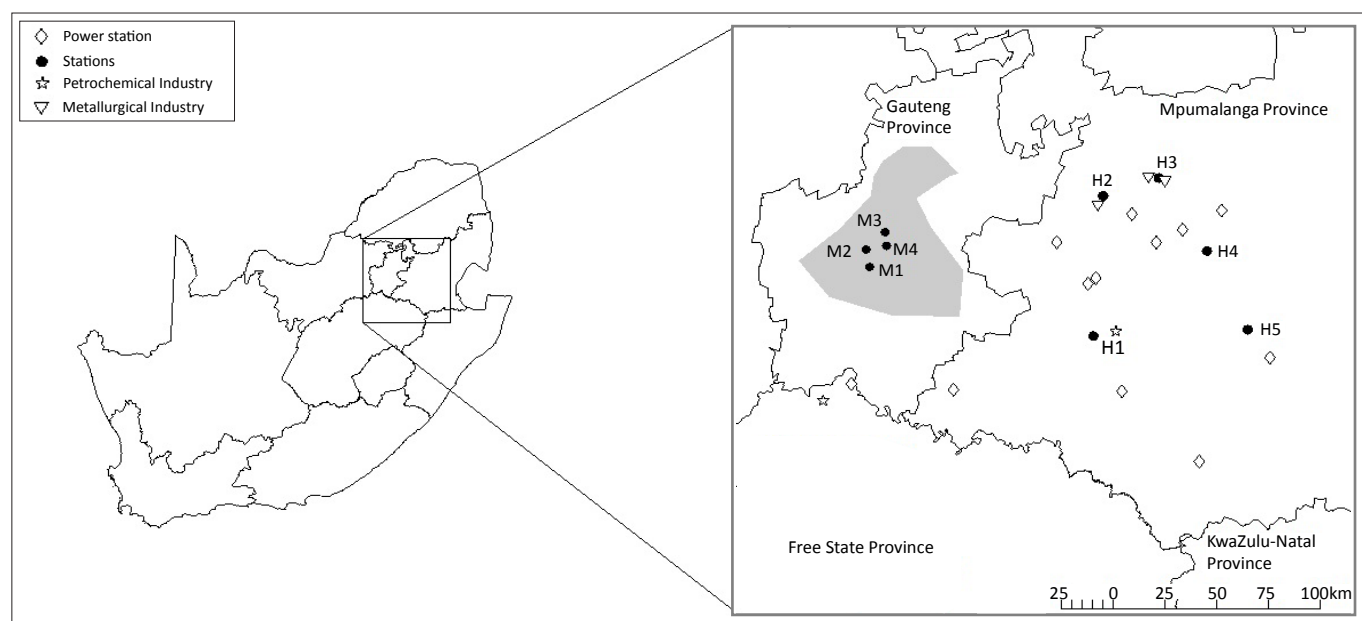
Figure 3 shows the  $\text{NO}_2$  diurnal cycles for each of the ground-based stations in the megacity and the Highveld hotspot for the 3-month period.

It is evident from Figure 3 that the  $\text{NO}_2$  mixing ratios of the megacity and the Highveld hotspot were mostly of the same order of magnitude. The megacity diurnal cycles were characteristic of urban areas dominated by traffic emissions, with maximum atmospheric  $\text{NO}_2$  levels during peak traffic periods in the early morning and late afternoon (Figure 3). In contrast, the  $\text{NO}_2$  diurnal cycles measured in the Highveld hotspot were less pronounced. This difference can be ascribed to the fact that the industrial activities that are mainly responsible for  $\text{NO}_2$  emissions operate 24 h per day. Measurement station H2, however, was an exception, as it showed a distinct diurnal cycle, presumably because it is relatively close to a city (Witbank) and is influenced by traffic emissions.

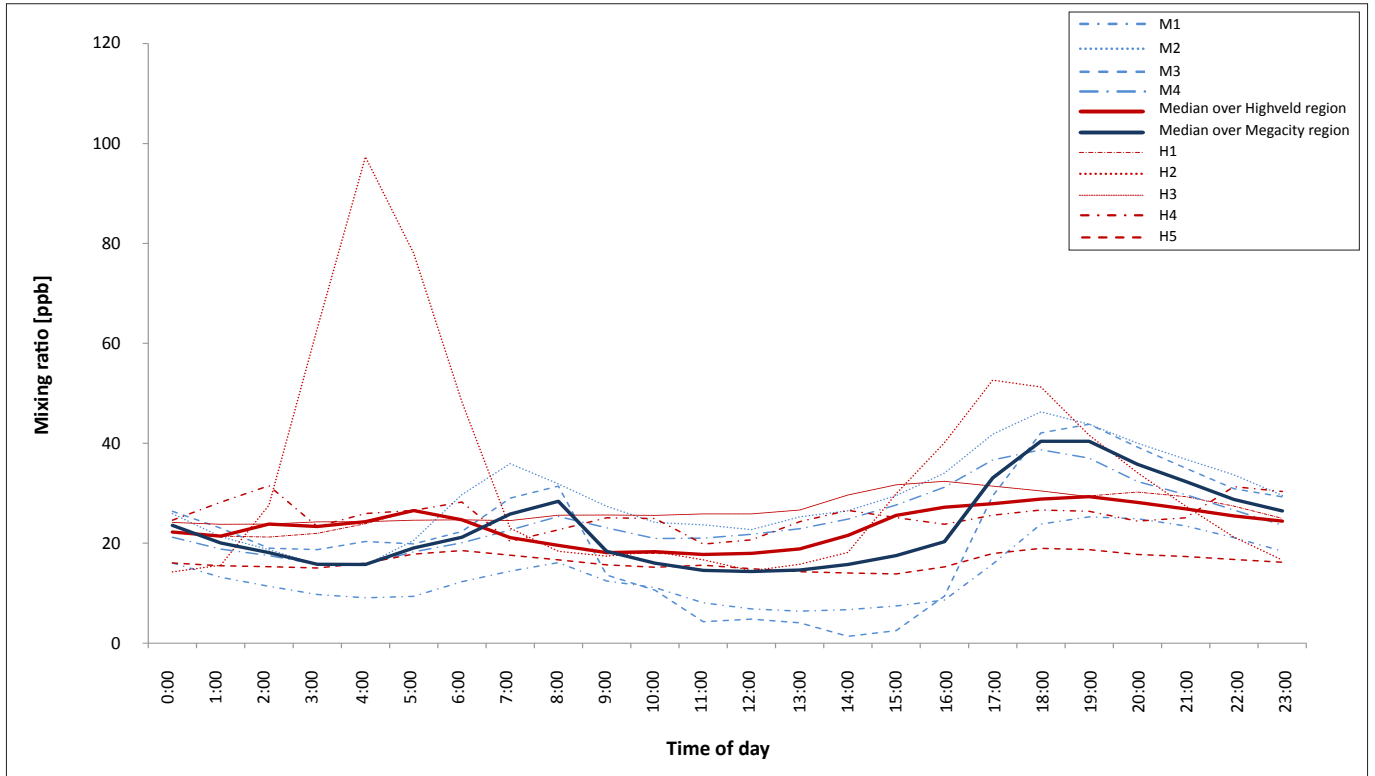
In order to facilitate the comparison of  $\text{NO}_2$  levels of the two areas, the ratio of the median concentration of the four megacity stations versus the median concentration of the five Highveld stations was computed, and is plotted in Figure 4 as a function of the time of day. Ratios greater than 1 indicate higher  $\text{NO}_2$  concentrations over the megacity than the Highveld, while ratios less than 1 signify higher  $\text{NO}_2$  concentrations over the Highveld than the megacity.

From this average diurnal cycle ratio, it is apparent that there are two distinct periods: 06:00–09:00 and 17:00–21:00, when near-surface megacity  $\text{NO}_2$  mixing ratios are higher than those of the Highveld hotspot. These time periods coincide with peak traffic hours in the megacity.

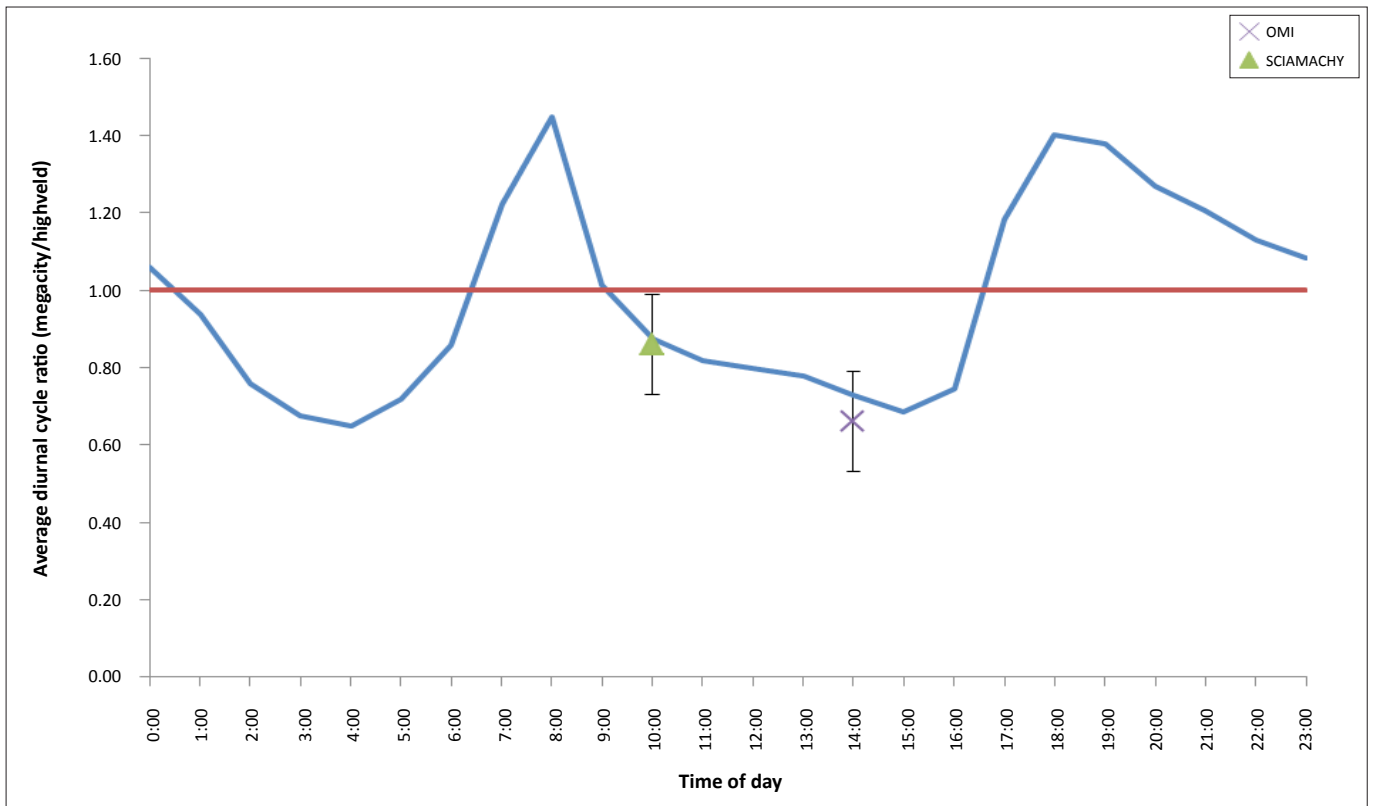
The ratios of the TSCD for the megacity divided by the TSCD for the Highveld hotspot are also shown in Figure 4, for measurements obtained from SCIAMACHY (2003–2009) and from OMI (2005–2009). The ratios were calculated from mean maps on a  $0.35^\circ \times 0.35^\circ$  grid. The actual numbers depend on the choice of the areas used for averaging. For the Highveld, an area of  $3 \times 3$  pixels (i.e.  $\approx 1^\circ \approx 1^\circ$ ) was selected that contained



**FIGURE 2:** A map of the sampling sites showing the major point sources, with 'M' denoting the Johannesburg-Pretoria megacity and 'H' the Highveld hotspot. The area shaded in grey indicates the Johannesburg-Pretoria megacity.



**FIGURE 3:** Median diurnal NO<sub>2</sub> mixing ratios for each of the ground-based stations for the period from March to May 2009 and the medians over each region. ‘M’ denotes a megacity station and ‘H’ a Highveld hotspot station, with numbers corresponding to the locations indicated in Figure 2.



The upper ends of the error bars were calculated using a megacity area of 1 pixel whereas the lower ends were calculated using a megacity area of 3x3 pixels (the same area as the Highveld). **FIGURE 4:** Average diurnal cycle ratio, defined as the averaged NO<sub>2</sub> diurnal mixing ratio of the four megacity measurement stations divided by the averaged NO<sub>2</sub> diurnal mixing ratio of the five monitoring stations in the Highveld hotspot, for a 3-month period (March–May 2009). The triangle and cross are the average ratios of the tropospheric slant column density (TSCD) for the megacity divided by the TSCD for the Highveld hotspot, which were obtained from the Scanning Imaging Absorption Spectrometer for Atmospheric Chartography (SCIAMACHY) and the Ozone Monitoring Instrument (OMI), respectively.



all H stations. For the megacity, two different areas were considered: (1) 1 pixel (containing all M stations) and (2)  $3 \times 3$  pixels (the same area as the Highveld). The upper ends of the error bars reflect the first choice, the lower end the second choice and the cross and triangle the averages of each ratio.

The ratios of the TSCDs could be biased if the AMFs for the Highveld and megacity were different. However, terrain height, aerosol load (MODIS aerosol optical density) and mean cloud fraction (from OMI) were similar for both regions.

The satellite-observed  $\text{NO}_2$  TSCDs over the Highveld hotspot were generally higher than those over the megacity. The 24-h average ratio of the ground-based observations is close to one (0.90), indicating that, at ground level, the overall  $\text{NO}_2$  pollution levels in the megacity were comparable to those of the well-known Highveld hotspot. Although the geographical area covered by the Highveld hotspot is larger than the area of pollution in the megacity, this area of the megacity deserves more attention than it has received, because of the large urban population that is affected by the pollution.

## Discussion and conclusions

We have shown that the Highveld hotspot – well known in the international science community because of its prominence in satellite images – is accompanied by a second South African hotspot over the Johannesburg–Pretoria megacity, with peak  $\text{NO}_2$  levels exceeding the maximum daily Highveld values during the morning and evening traffic rush hours. This finding is of importance for the more than 10 million people living in this megacity. Although satellite instruments have been valuable in pointing out hotspots worldwide, we have shown here a limitation as a result of their specific overpass times. Satellite overpass times that coincide with the peak traffic hours of the megacity would be more ideal. However, early morning overpasses would not necessarily be more suitable for satellite detection, as a result of the trapping of pollutants near the surface where satellite measurements do not provide a suitable response. Early evening overpasses might be more feasible to detect gaseous concentrations because peak traffic emissions would be better mixed in the boundary layer by evening. Further progress on this issue will require more ground-based and aircraft profile measurements of the region. Direct observation of tropospheric  $\text{NO}_2$  vertical column densities with ground-based Multi AXis Differential Optical Absorption Spectroscopy (MAX-DOAS) measurements<sup>19</sup> or airborne instruments may be particularly valuable. Airborne DOAS measurements can be used for direct observation of tropospheric  $\text{NO}_2$ .<sup>20</sup> A comprehensive study of the Highveld hotspot and the megacity is required in order to develop a better understanding of the true extent of the  $\text{NO}_2$  hotspot over the interior of South Africa. An improved understanding, as well as greater public and political awareness of this pollution hotspot will be valuable, especially in light of

eventually determining the consequences of this additional hotspot for  $\text{O}_3$  production, impacts on health and agriculture, and effective mitigation measures.

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## Competing interests

We declare that we have no financial or personal relationships which may have inappropriately influenced us in writing this paper.

## Authors' contributions

A.S.M.L. conducted this work as part of her PhD study and was therefore the main investigator; T.M.B., J.P.B. and P.G.v.Z. were study leaders; S.B. assisted with satellite data interpretation; M.G.L., G.D.F., J.J.P., T.W. and K.-P.H. made conceptual contributions.

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