

***Interactive comment on* “Basic characteristics of atmospheric particles, trace gases and meteorology in a relatively clean Southern African Savannah environment” by L. Laakso et al.**

Anonymous Referee #2

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1. General Comments.

In this article measurements of particle number size distribution, PM_x mass concentrations, trace gases concentrations and meteorology, performed in Savannah environment in South Africa are presented. The paper is written in a fluid way and is friendly to read. The study is of high interest because air pollution has not been deeply investigated in this type of environment. Moreover data have been collected with suitable methods and ensure a high quality.

This study is of interest and fit with the objectives of ACP. However, I think that a deeper analysis of the data is required. I guess that the idea of the authors was to prepare a

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general paper presenting the data and the general behaviour of the particles and gases at this site, and not prepare a paper focused on any specific topic of aerosol or gases (e.g. new particle formation or dust transport from distant arid regions). However, when reading the paper it seem that the results are presented into unconnected boxes or analysis, each focused on CO, NO_x, SO₂, PM_x, nucleation particles and growth rates. My question is, what is the relationship among these parameters?. Three examples:

1. It is argued that biomass burning may contribute to CO levels, however and that industrial emission contributes to SO₂ levels. These two sources may also contribute to NO_x levels, together with soil emissions and transport from urban areas. For this it would be of interest to quantify what is the relationship among CO, SO₂ and NO_x.

2. In the manuscript it is described that there are biogenic emissions of monoterpenes and isoprenes and industrial emissions of SO₂. These can contribute to new particle formation events, however it is not discussed in the paper. It would be of interest to quantify what is the relationship between SO₂ and nucleation particles (see details below in point 8).

3. It is discussed that both long range transport of dust from arid regions and it is suggested that the biomass burning realising CO may contribute to PM_x concentrations. Are the high PM_x events associated with high CO, NO_x or SO₂ concentrations?.

In my opinion, a deeper analysis on the relationship between the traces gases and particles is necessary. I suggest using any statistical tool for doing this (correlation coefficients, factor or cluster analysis). This additional analysis will provide more details on the nature of the pollution in this region. Some of the questions I asked my self when reading the paper are: are the new particle formation events associated with SO₂ events?, are the SO₂ episodes associated with NO_x events (industrial emissions)?, are the CO episodes associated with NO_x events (biomass burning)?. I encourage to the authors to do this when preparing the revised version. The data base is really very interesting and I think that this small additional analysis may provide a more general

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view that will enrich the paper.

2. Specific Comments

1. Section-3. PM_x concentrations were monitored with a modified TEOM sensor. The system used allows obtaining (not simultaneous) data on PM₁₀, PM_{2.5} and PM₁. Was the PM_x aerosol dried before measure it?. How was it done?, The standard TEOM1400a sensor heats the sample to 50degrees Celsius and it is very well known that this may result in a negative artefact due to evaporation of semi-volatile compounds (e.g. ammonium-nitrate and organic matter). Have the authors any quantitative estimation on this potential negative artefact or any estimation on what is the contribution of ammonium-nitrate and organic matter to PM₁₀ concentrations (e.g. by filter analysis)?

2. Section 4.1. In the general description performed for each trace gas (i.e. NO_x and CO) the seasonal evolution is described, except for SO₂. In Figure 6a it can clearly be observed that SO₂ experience a seasonal evolution with a maximum in local winter (July), however this is not described in this section. I suggest including this in the text.

3. Section 4.1. The seasonal evolution of O₃ is neither described nor discussed. In Figure 6d it can be observed how ozone exhibits a seasonal evolution with a maximum in local spring (Sept-Nov). However, this is not described in the text. The seasonal evolution with the maximum in spring is a feature of O₃ in islands of the North Atlantic Ocean located at the same latitude than the region studied in this article. However, this is not cited in the article. See for example Rodriguez and Guerra (Atmospheric Environment 2001, 35, 1829 - 1841), Rodriguez et al. (Atmospheric Environment 2004, 38, 4733 - 4747) and references therein.

4. The processes contributing to the seasonal evolution of NO_x, CO and SO₂ are discussed in a scattered way long the manuscript. I suggest to perform this discussion into 1 single section and not along several sections. Moreover, for the case of ozone is not discussed. More details: 4.1. CO, page 6322, line 18-21 (section 4.1): The carbon monoxide concentration had a clear seasonal trend with elevated values observed

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during the late winter and spring (Fig. 6c). The most probable reason for 20 the elevated CO concentrations (by the way, please add s) during the driest months were the regional bush fires and possibly more distant biomass burning sources. This is OK for me. 4.2. NO_x, page 6322, line 16-17 (section 4.1): The highest values were measured during the local autumn (March - April). The cause of this seasonal evolution is cited or discussed here as done for CO. 4.3. SO₂, page 6325, line 4-6 (section 4.2): The high values during the winter compared 5 with other seasons can be attributed to less effective mixing, which affects the dilution of pollution originating from the Rustenburg mining area. 4.4. Ozone. The origin of the seasonal evolution of O₃ is not discussed in the text. As pointed above, O₃ exhibited higher values in spring. In Islands of the North Atlantic Ocean these high values of ozone in spring are associated with high O₃ concentrations both during daylight and at night due to downward transport from the free troposphere (e.g. Parrish et al., 1998, Journal of Geophysical Research 103 (13), 357-376, Oltmans and Levy, 1992, Nature 358, 392 - 394, Rodriguez et al. Atmospheric Environment 2004, 38, 4733 - 4747). How does the high spring O₃ events occurs at this site?, are they associated with an enhancement in the regular daily evolution (i.e. much higher values during daylight than at night), or are they associated with high O₃ levels both during daylight and at night?. This type of analysis may provide useful information on the potential role of ozone production into the boundary layer (probably linked to biomass burning) or downward transport from the free troposphere. For details on the O₃ spring maximum I also recommend the paper of Crutzen et al. 1999 (Tellus 51A - B, 123 - 146).

5. Section 4.1. When discussing the variations of PM₁₀ concentrations with wind direction (Figure 9), it is stated that the fact that high PM₁₀ concentrations are recorded when wind blows from NW are probably due to transport of mineral dust from Kalahari Desert. Have author studied these potential dust transport events with satellite observations (e.g. OMI or MSG), model simulations (e.g. NAAPs: <http://www.nrlmry.navy.mil/aerosol/#currentaerosolmodeling>), or back-trajectories?. It would be useful to cite in the text what is the distance between the measurement site

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and the Kalahari Desert.

6. Section 4.1, page 6324, line 9-10, it is stated that there is no clear seasonal evolution in the particle number concentration and a reference to Figure 10 is performed. However, in the Abstract (lines 7-8) it is stated that gases and particles had a clear seasonal evolution. This contradiction on the seasonal evolution of particles should be corrected.

7. Section 4.2, page 6325, lines 3-8. The daily evolution of SO₂ is described. It is argued that concentrations of SO₂ are higher during daylight than at night owing to SO₂ deposition at night. In my opinion other processes are also involved on this SO₂ daily evolution. Studies performed in rural areas (several tens of kilometres) around power stations with high stacks (>200 meters high) have observed that SO₂ concentrations are higher during daylight than at night. It has been widely documented that this is mainly due to downward transport of the SO₂ plume (located above the boundary layer due to the SO₂ emission take place in a high stack) owing to vertical mixing processes activated by the thermal convective activity. See for example the studies performed in Eastern Spain by Querol et al. (Atmospheric Environment 1998, 32, 1963 - 1978), Querol et al. (Environmental Pollution 1999, 105, 397 - 407), Alastuey et al. (Atmospheric Environment 2004, 38, 4979 - 4992). I suggest to the authors to investigate how the SO₂ emissions take place in the mines located at the East of the measuring site. Probably these SO₂ emissions take place in a medium to relatively high stack. In my opinion, the downward transport of the SO₂ plume due to thermal convective activity during daylight is the most important process contributing to the daily evolution observed in this study.

8. In my opinion more details should be provided on the features of the new particle formation events. Two examples: 8.1. Does nucleation events seem to be linked to nucleation of sulphuric acid, or is there any involvement of the biogenic emissions (monoterpenes and isoprenes) in the new particle formation?. For performing this discussion other relevant information may be provided: are SO₂ levels higher during

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nucleation events than during non-nucleation events?, does SO₂ correlates with the number concentration during NPF events?, 8.2. it seem that the daily evolution of O₃ and N is significantly correlated. Are O₃ levels higher during nucleation events than during non-nucleation events?. This has been observed in Europe (e.g. Hamed et al., Atmospheric Chemistry Physics 2007 7, 355 - 376 and references therein).

9. The time variations of the PM_{2.5}/PM₁₀ can not be studied in details because PM₁, PM_{2.5} and PM₁₀ concentrations were not simultaneously performed. However, I think it worth to highlights that the mean PM_{2.5}/PM₁₀ ratio is close to that observed in semi-arid regions of Southern Europe (Rodrigues et al., 2007, Environ Chem Lett 2007 5, 1 - 7) and much lower than that found in urban polluted sites of Central and Northern Europe (Atmospheric Environment 38 (2004) 2561 - 2577).

Interactive comment on Atmos. Chem. Phys. Discuss., 8, 6313, 2008.

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