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Atmospheric
Chemistry
and Physics
Discussions

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

L. Laakso et al.

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Anonymous Referee 2

1. General Comments.

In this article measurements of particle number size distribution, PMx 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.

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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 general paper presenting the data and the general behavior 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).

*Correct, the idea of this paper is to show basic characteristics and act as a reference paper for our more detailed case studies so that in stead of describing the site, basic meteorology, emission sources, instrument etc. we can just refer to this paper and another paper now in progress which describes technical solutions and calibrations in detail.

Second reason to publish only part of the first data set (as soon as possible) was to provide reference for other scientists; we have already provided our data to global NPF modelers.*

However, when reading the paper it seem that the results are presented into unconnected boxes or analysis, each focused on CO, NOx, SO2, PMx, nucleation particles and growth rates.

The structure of the article is now changed

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 NOx 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 NOx.

*Our original idea was to keep this first article short, and easily readable. As also discussed above, we plan to do thorough study on gas phase chemistry based on the whole data set, not only the first one year. However, we calculated correlations for different times of the day, and between all variables. The tables are given at the end of

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this letter. However, to keep the article easily readable, we do not include the tables to the actual manuscript.*

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).

*We added some material related to this issue to the manuscript. It looks like the SO2 is not typically the limiting factor in NPF, but the process is rather limited by pre-existing particle surface. Please see also comment above. *

3. It is discussed that both long range transport of dust from arid regions and it is suggested that the biomass burning realizing CO may contribute to PMx concentrations.

Are the high PMx events associated with high CO, NOx 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 NOx events (industrial emissions)?, are the CO episodes associated with NOx 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 view that will enrich the paper.

*These are important - and extensive- questions. We have answered these questions partly in section 4.4 where we discuss the correlations between different variables. In addition, one of our students is currently preparing his MSc-thesis on this topic. So we ask the referee to be patient and wait until early 2009. *

2. Specific Comments

1. Section-3. PMx concentrations were monitored with a modified TEOM sensor. The system used allows obtaining (not simultaneous) data on PM10, PM2.5 and PM1. Was the PMx 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)?

Typically, RH is rather low, so we did not dry the PM sample. As our monitoring station is part of provincial measurement network in Rustenburg from February 2008 until mid-2009, we decided to keep the standard setting, even we know that in doing that, we very probable loose the volatile compound from our samples. This was a choice we needed to do. We are also collecting samples by impactors, but due to long storing times in less-well-defined conditions, and unknown contribution from vapor phase organics, we do not have the courage to use the data to correct our mass concentrations.

2. Section 4.1. In the general description performed for each trace gas (i.e. NOx 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.

The seasonal variability of SO2 is now discussed in paragraph 7 in section 4.2 (see below).

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.

Discussion about the seasonal variability of O3 has been added to the paper (paragraph 8 in section 4.2). The valuable comments by the referee are taken into account in the text with appropriate references. We also appreciate this comment from future point of view, as we plan to prepare a separate manuscript on gas phase chemistry.

4. The processes contributing to the seasonal evolution of NOx, CO and SO2 are discussed in a scattered way long the manuscript. I suggest performing this discussion into 1 single section and not along several sections. Moreover, for the case of ozone is not discussed. More details:

We agree on this. The seasonal variability of NOx, CO and SO2 is now discussed in a single paragraph (paragraph N in section 4.2). This is followed by a corresponding paragraph for O3.

4.1. CO, page 6322, line 18-21 (section 4.1): The carbon monoxide concentration had a clear seasonal trend with elevated values observed 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.

*Moved to paragraph 7 in section 4.2 with slight modification. *

4.2. NOx, 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.

Moved to paragraph 7 in section 4.2 with slight modification.

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

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dilution of pollution originating from the Rustenburg mining area.

Moved to paragraph 7 in section 4.2 with slight modification.

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).

The seasonal variation of O3 concentration is now discussed briefly in the text (paragraph 8 in section 4.2). A comparison to observations at other sites in included as well. A detailed analysis of the origin of the seasonal O3 cycle at our measurement site has, however, left out of the paper. We feel that such an analysis would be a work on its own and, anyway, it would occupy too much space of this paper.

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

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would be useful to cite in the text what is the distance between the measurement site and the Kalahari Desert.

*We have planned to conduct an analysis on this topic as part the M. Sc. thesis of one of our students in the near future. Unfortunately, at this point we are not able to back up our statement with more detailed information. Defining the edge of Kalahari is somewhat difficult, but we can safely say that there are dry areas able to produce significant amount of dust approximately 500 km away from Botsalano. *

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.

Abstract 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.

We agree. That's actually what we tried to say, but with poor success. We clarified this in the text. A reference was added into the text.

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 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).

*Based on correlations calculated, there was no correlation between O3 and nucleation mode particles. To our surprise, SO2 seem not to be the limiting factor in nucleation, but rather the concentration of pre-existing surface. We will discuss this more in detail in our oncoming paper on new particle formation.

We can not say much on connection between the biogenic VOC emissions and new particle formation, since our data on VOC is very limited. However, as we have some VOC-samples from October 2006, January 2007, May 2007, October 2007 and January 2008, we may return to this topic, with help of BL-chemistry models (e.g. Boy et al, 2006) later on, or potentially in our on-coming gas phase chemistry paper.*

9. The time variations of the PM2.5/PM10 can not be studied in details because PM1,

PM2.5 and PM10 concentrations were not simultaneously performed. However, I think it worth to highlights that the mean PM2.5/PM10 ratio is close to that observed in semiarid 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

This point was added to the text with relevant references.

*References:

M. Boy, O. Hellmuth, H. Korhonen, E. D. Nilsson, D. ReVelle, A. Turnipseed, F. Arnold, and M. Kulmala, MALTE-model to predict new aerosol formation in the lower troposphere; Atmos. Chem. Phys., 6, 4499-4517, 2006*

Appendix to the referee comments: correlation coefficients (we apologize for slighly inconvenient format due to latex-input)

Whole day

	O3	SO2	Nox	CO	Nucl	Ait	Acc	Tot	PM1	PM2.5	PM10
O3	1.00										
SO2	0.11	1.00									
NOx	-0.26	0.35	1.00								
CO	0.31	0.12	0.02	1.00							
Nucl	0.04	0.21	0.03	-0.09	1.00						
Ait	0.17	0.40	0.08	0.10	0.20	1.00					
Acc	0.20	0.19	0.03	0.45	-0.06	0.11	1.00				
Tot	0.17	0.42	0.07	0.02	0.81	0.71	0.16	1.00			
PM1	0.28	0.31	0.07	0.38	-0.13	0.14	0.23	0.07	1.00		
PM2.5	0.18	0.28	0.04	0.32	-0.11	0.14	0.21	0.08	0.76	1.00	
PM10	0.14	0.31	0.02	0.20	-0.05	0.18	0.16	0.12	0.58	0.57	1.00

Morning correlations (06-12)

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O3 SO2 Nox CO Nucl Ait Acc Tot PM1 PM2.5 PM10 O3 1.00
SO2 0.05 1.00
Nox -0.2 0.54 1.00
CO 0.38 0.09 0.01 1.00
Nucl 0.08 0.29 0.08 0.00 1.00
Ait 0.13 0.31 0.09 0.09 0.32 1.00
Acc 0.42 0.39 0.14 0.47 -0.04 0.09 1.00
Tot 0.14 0.38 0.11 0.06 0.92 0.66 0.07 1.00
PM1 0.16 0.26 0.14 0.36 -0.12 0.04 0.69 -0.04 1.00
PM2.5 0.13 0.29 0.10 0.34 -0.09 0.05 0.63 -0.02 0.75 1.00
PM10 0.14 0.33 0.06 0.24 -0.02 0.08 0.61 0.06 0.64 0.55 1.00

Afternoon (12-18)

O3 SO2 Nox CO Nucl Ait Acc Tot PM1 PM2.5 PM10
O3 1.00
SO2 0.13 1.00
Nox -0.04 0.67 1.00
CO 0.30 0.20 0.00 1.00
Nucl -0.20 0.19 0.18 -0.19 1.00
Ait 0.13 0.48 0.24 0.11 0.13 1.00
Acc 0.14 0.11 0.05 0.41 -0.07 0.08 1.00
Tot 0.03 0.52 0.31 -0.01 0.67 0.80 0.15 1.00
PM1 0.45 0.30 0.14 0.39 -0.16 0.13 0.10 0.09 1.00
PM2.5 0.34 0.37 0.20 0.33 -0.11 0.23 0.14 0.18 0.67 1.00
PM10 0.24 0.37 0.19 0.23 -0.10 0.20 0.12 0.15 0.62 0.62 1.00

Night (21-03)

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	O3	SO2	Nox	CO	Nucl	Ait	Acc	Tot	PM1	PM2.5	PM10
O3	1.00										
SO2	0.18	1.00									
Nox	-0.31	0.05	1.00								
CO	0.33	0.14	0.03	1.00							
Nucl	-0.33	-0.25	-0.13	-0.46	1.00						
Ait	0.10	0.50	-0.03	0.08	-0.06	1.00					
Acc	0.47	0.51	-0.04	0.45	-0.51	0.27	1.00				
Tot	0.13	0.52	-0.09	-0.04	0.13	0.94	0.38	1.00			
PM1	0.36	0.37	-0.05	0.36	-0.45	0.17	0.77	0.24	1.00		
PM2.5	0.27	0.32	-0.06	0.31	-0.40	0.16	0.70	0.22	0.78	1.00	
PM10	0.26	0.36	-0.14	0.07	-0.19	0.22	0.57	0.31	0.68	0.70	1.00

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

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