Response to Anonymous Referee #1

This work presents four year data of submicron aerosol particle number size distributions for two different locations in southern African Savvanah. Similar observations in the southern hemisphere are scarce and of great significance for the atmospheric community. The manuscript is well written and presents data illustrating the effect of anthropogenic activities, incomplete burning and nucleation processes to the aerosol size distributions in the area. However the presentation of the results can be made more effectively. Therefore, it should be accepted after the authors have made some minor changes

We would like to thank the Anonymous Referee 1 for his/her comments, which have improved the quality of the manuscript.

General comments

The two sites under study are presented in a way, so that at some points the reader gets the feeling that the authors refer to a single location representative for the whole southern Africa. However, it should be made even more pronounced in the manuscript that the aerosol populations in the two locations are significantly different. Nevertheless, these differences are of great importance since the comparison between the two sites can give a more detailed picture of heavily polluted and semi- clean conditions in S. Africa, and thus valuable information for modelers about the parameterization of aerosols in the region. It is recommended that the comparisons between all the different sites refered in the manuscript to be summarized and given in two separate paragraphs, one for polluted and one for semi-clean conditions.

We have collected the comparisons with other sites to a new chapter, which is divided into semi-clean and polluted sections.

Mineral dust originating from the Kalahari region can be present and play some role in the area, however it is not included in the analysis. The Kalahari sector in Figure 11 (N100 panel), the Kalahari distribution in Fig.12 perhaps indicate the presence of large particles. The mobilization of the Kalahari region, is maybe not very significant on a global scale, however can be regionally important. It would be a great improvement to include some analysis on the dust contribution, using some satellite or AERONET data (available for the region) if no other means are available.

We would like to thank Anonymous Referees 1 and 2 for bringing up the significance of desert dust from the Kalahari, it definitely should be discussed with the Kalahari source region. Unfortunately the coarse mode size distribution was not measured at Botsalano, and therefore the presence of desert dust cannot be quantified for the Kalahari source region as defined here. Nevertheless, it would be important to quantify in the future also the coarse mode size distribution over the area covered in this study.

To address these questions better we have added a figure on satellite retrieved aerosol optical depth (AOD) over southern Africa and modified the discussion in chapter 3.4:

"The Kalahari is the only source region that does not exhibit regional new particle formation in the median diurnal variation. This is probably due to smaller concentrations of both

biogenic and anthropogenic precursors if compared to the re-circulation and industrial hub source regions, because the condensation sink (CS) in Kalahari ($2.5\ 10^{-3}$ s^{-1}) is lower than that of the eastern source regions (Vakkari et al., 2011). In the Karoo source region the combination of an even lower condensation sink ($1.4\ 10^{-3}$ s^{-1}) than in the Kalahari source region to lower growth rates (Vakkari et al., 2011) results in the nucleation mode being continuously present.

Despite lower accumulation mode concentration than the re-circulation and industrial hub source regions, the Kalahari source region has a higher AOD as seen in Fig. R1. Most likely the increase in AOD over the Kalahari source region originates in desert dust in the coarse mode size range. However, the PM2.5 and PM10 mass concentrations observed at Botsalano are not elevated for the Kalahari source region (Fig. R1), which implies that the desert dust from the Kalahari is not transported to Botsalano. As the condensable vapours have short lifetime (approximately CS^{-1}) it seems that the Kalahari desert dust cannot explain the lack of new particle formation at Botsalano for the Kalahari source region, although dust storms have been shown to scavenge effectively sub-100 nm aerosol particles (Jayaratne et al., 2011)

Now if the condensable vapour source rate and aerosol formation rate at 2 nm are assumed equal during new particle formation for Karoo and Kalahari source regions, even the difference in the submicron CS is enough to suppress new particle formation events for the Kalahari source region. The Kerminen and Kulmala (2002) formulation connects the ratio of observed formation rate to nucleation rate at a smaller diameter with CS and growth rate, and from the assumptions above and observations for Karoo (Vakkari et al., 2011) it follows that the J10 for Kalahari source region would be lower than the J10 from Karoo by a factor of 10^{13}, i.e. the nucleated particles will be lost by coagulation before they reach the 10 nm detection limit."
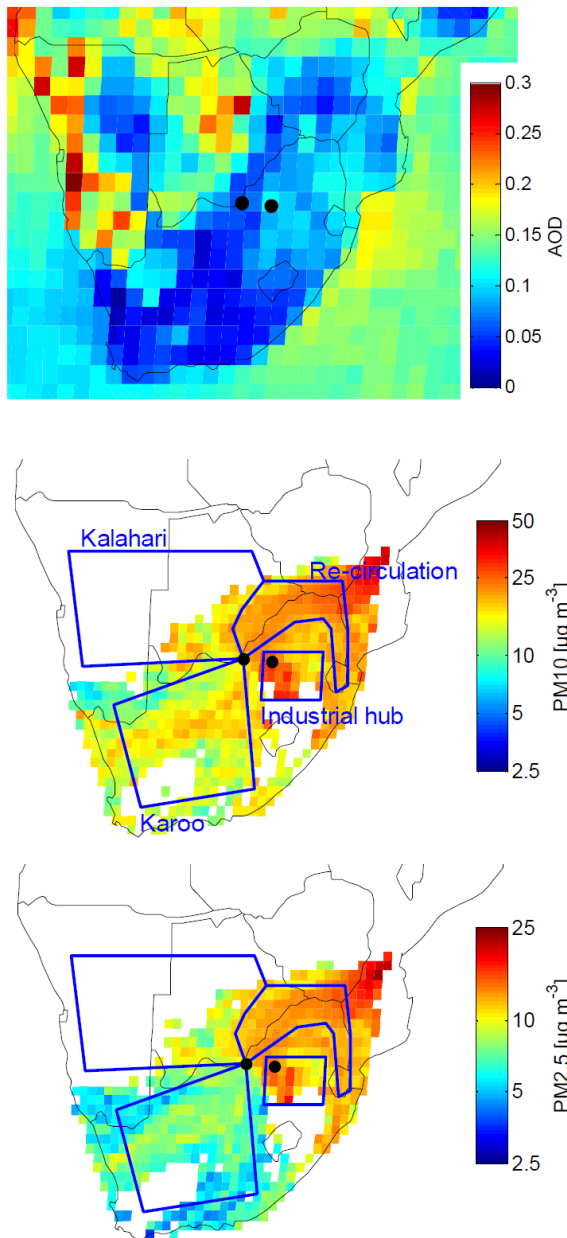
Figure R1. Top: median AOD over southern Africa from July 2006 to January 2008 from MODIS aerosol product at 550 nm (Remer et al., 2005). In the middle the mean PM10 mass concentration and at the bottom the mean PM2.5 mass concentration for the four defined source regions at Botsalano. Black dots indicate Botsalano (on the left) and Marikana (on the right).

Specific Comments

Figure 2: Please use different colour or/and marker for the different sites

The Fig. 2 has been redrawn as a bar plot and the different sites are now indicated with different colour.
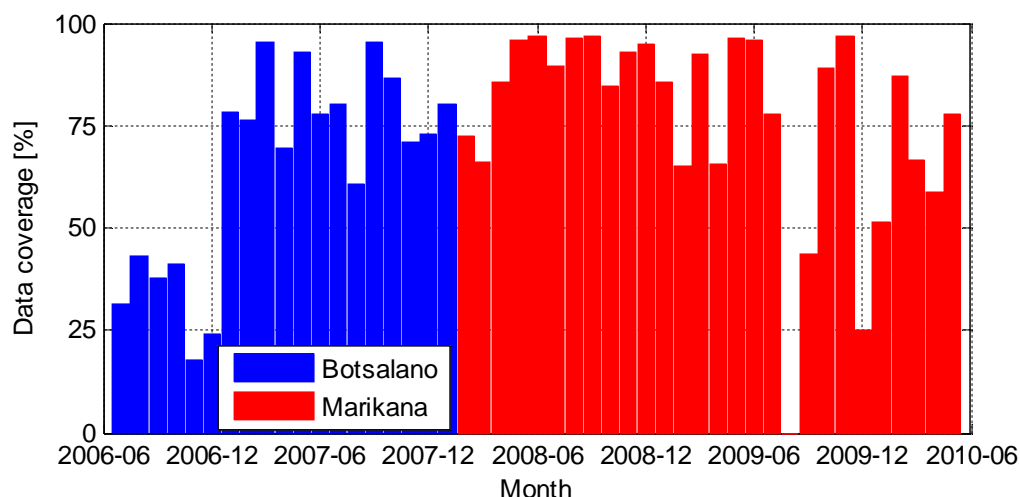
Figure R2. New Fig. 2 for the manuscript.

Paragraph 3.1, Line 5: The numbers given in the table are 1856 and 7805 respectively.

We have corrected the text to match the table.

Figure 4: The contour plots are dominated by the nucleation events, suppressing the rest of the regional characteristics. Same plot separated in event and non-event days will be more enlightening for both the nucleation processes and the size distributions diurnal cycle the rest of the days.

We agree with Anonymous Referee 1 that the nucleation events may suppress other regional phenomena in Fig. 4. However, there are so few non-event days at both Botsalano and Marikana that the non-event surface plots would not be representative. We have added the following discussion in chapter 3.2:

"The regional new particle formation in Fig. 4 dominates the diurnal variation to a degree where other daytime regional phenomena may be suppressed by it. However, as new particle formation frequency is so high in Botsalano and Marikana, the diurnal variation of only non-event days would not be representative: 6% of days at Botsalano and only 0.3% (i.e. two days) at Marikana were classified as non-event."

Figure 5, 7. There is a conflict here with the determination of what a mode is. To my opinion the evolution of a mode with time is dynamic, modes interact to each other. Therefore the growth of particles to larger diameters does not reflect just the growth of the mode mean diameter but the contribution of the growth processes to the number of the next mode. For instance, in Fig. 5 mode mean diameter panel, Mode 3 stops to exist at 18:00, but the mode diameter of Mode 2 can be attributed after 18:00 to Mode 3.

Anonymous Referee 1 is certainly correct in that mode 3 does not vanish at 18:00, but merges with previous mode 2. In the manuscript Figs. 5 and 7 and related discussion is to some extent unclear with how this is presented. The modal fittings are done independently for each time step in Figs. 5 and 7 and therefore the fitting routine does not provide the labels for the

modes, but they are grouped afterwards to help the eye catch the diurnal patterns and connect the correct modal mean diameter, particle number and standard deviation together.

We modified the rows 20-27 on page 24056 in chapter 3.2 as follows:

"Considering the median diurnal distribution, the accumulation mode concentration increased at the onset of the new particle formation event: the increase in N100 in Fig. 4 is concurrent with the appearance of the nucleation mode. Even in the one hour median size distribution parameters in Table 3 the N100 increased from 6:00 to 12:00 LT. This is due to the growth of the pre-existing Aitken mode particles, as is seen in Fig. 5 modal fitting parameters. The mode 2 mean diameter in Fig. 5 starts to increase rapidly from 70 nm already at 12:00 LT, which is clearly before the mode 1 particles could have reached this size as the median growth rate in new particle formation at Botsalano was 8.9 nm h^{-1} (Vakkari et al., 2011). At 18:00 LT the mode 2 has grown out from the Aitken mode size range and merges with the previous accumulation mode (mode 3). The growth of the pre-existing Aitken mode therefore seems to be an important process producing CCN-sized particles in a semi-clean savannah environment such as Botsalano.

Note also that the modal fittings in Fig. 5 are calculated independently for each size distribution letting the modal fitting algorithm decide the number of modes from one to three. Also the diameters of the modes are let to vary freely (Vartiainen et al., 2007), which may lead to more than one mode in e.g. Aitken mode size range in some cases. The division into three (or four for Marikana) modes in Fig. 5 is then done independently of the modal fittings to better illustrate the diurnal patterns in the size distribution."

Paragraph 3.3 Information about the frequency of occurrence on an annual base of each particle mode described earlier in a figure or in plain text would be very interesting.

We have calculated modal fittings to the monthly median diurnal variation shown in Figs. A1 and A2 and have added the following figure and discussion in chapter 3.3:

"The seasonality of log-normal modal parameters was studied by fits to the monthly median distributions presented in Appendix A. Interestingly from the modal fitting point of view the seasonality in N100 for both semi-clean and polluted savannah (Fig. 8) originates in an increase in the Aitken mode (mode 2) number concentration rather than in the accumulation mode (mode 3) concentration. The frequency of occurrence of the mode 4 at Marikana has a clear seasonality with maximum during the colder months as seen in Fig. R3. Also the frequency of occurrence of mode 3 at Botsalano seems to have a seasonality with a minimum from July to August, Fig R3, which is when the number concentration of mode 2 is elevated. The monthly median modal fitting parameters are included in the supplement.
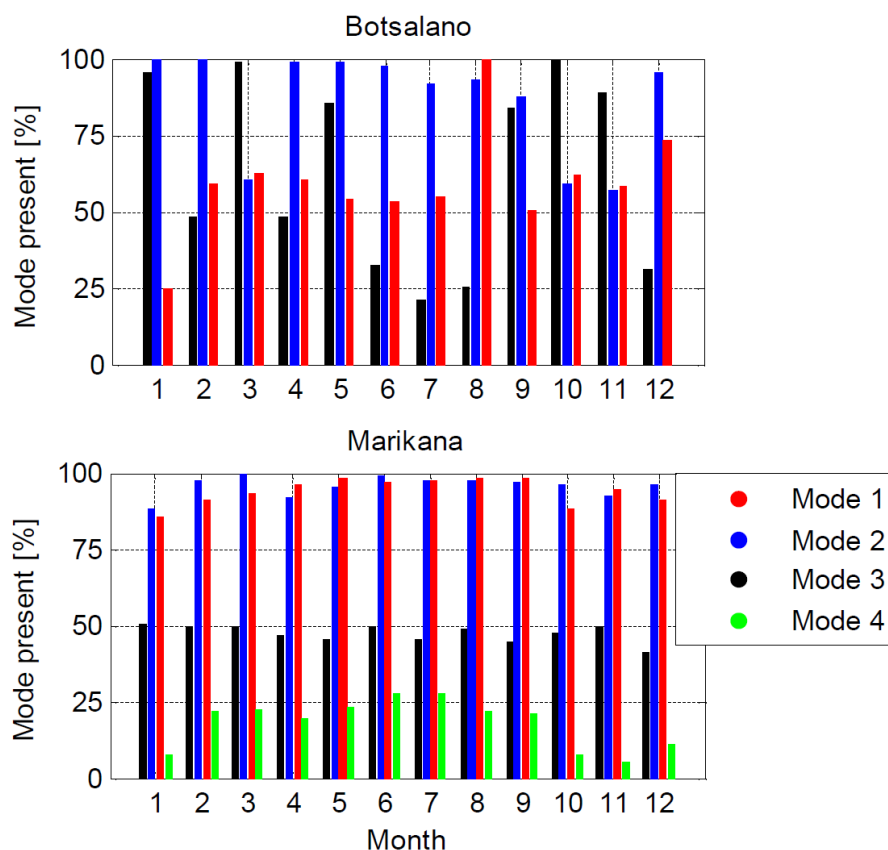
Figure R3. Monthly frequency of occurrence of each mode in Botsalano (upper panel) and Marikana (lower panel). The frequency is based on modal fits to the monthly median diurnal plots in Appendix A."

Figure 9: The secondary maximum of CO at Marikana probably attributed to the maximum of fire observations in the area as shown in Fig 10 is not represented in the N100 annual variation. Please explain.

We would like to thank the referee for bringing this up and have added the following discussion in chapter 3.3 to address this question:

"The monthly median CO concentration, Fig. 9, has two peaks at Marikana: one in July, the coldest month, and a secondary one in September, which is the wild fire peak month, Fig. 10. The N100, however, does not peak in September but only in July (Fig. 8). In contrast in Botsalano September is the peak month for both CO and N100, which is approximately 560 particles cm^{-3} higher than during summer (cf. Table 4).

In Marikana the N100 in September is approximately 470 particles cm^{-3} higher than during summer (Table 4), not far from the increase at Botsalano, but still 1350 particles cm^{-3} lower than in July. The intensity of the evening burning peak in September is, however, close to what it is in February and March, Fig. A2, which is reasonable as the mean temperature is 19.4°C and there is no or very little need for heating in the evenings. Therefore the elevated N100 concentration at Marikana in September is due to regional wild fires; only the concentration appears low compared to the July peak from domestic heating and cooking.

Why does the CO then peak in September at Marikana? The reason is that CO has lifetime of 30–90 days in troposphere, while aerosol particle lifetime varies from a few days to a few weeks (Seinfeld and Pandis, 2006). Therefore CO accumulates in the atmosphere over a longer period than aerosol particles, which leads to an increase in ratio of CO to N100 towards the end of the dry season and especially in the early wet season, when increased wet removal decreases aerosol particle concentration but not CO concentration."

References

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