

## Response to Anonymous Referee #2

*The paper by Vakkari et al. presents an interesting data set of aerosol size distributions from South African savannahs, a region where there were only few reported measurements. As such this represents a valuable set of measurements that would be very useful for the global aerosol community. The data analysis and discussions are of a very high quality and overall the manuscript is very well written. I would recommend that it is published after some minor revisions.*

We would like to thank Anonymous Referee 2 for the positive criticism, which has improved the quality of the manuscript.

*Introduction: In the introduction part where the authors give an overview of the measurements in the southern hemisphere I think that they are overlooking some of the work done by the group at QUT in Brisbane. Maybe it is worth looking at some of the papers by Mejía, J. F. and Morawska, L. from ACP or at some recent work from the same group by Cheung et al 2011 (also published in ACP).*

We would like to thank the Anonymous Referee 2 for bringing up these references. We have updated the introduction as follows:

“However, for the southern hemisphere there is a very limited number of long-term data sets of sub-micron aerosol particle size distributions in the continental boundary layer (Laakso et al., 2006; Laakso et al., 2008; Hirsikko et al., 2012; Laakso et al., 2012). Even from the Amazon basin, where several intensive measurement campaigns have been carried out during recent years, no size distribution measurements that cover a full year have been published (see e.g. the review by Martin et al. (2010) and references therein). From Australia the majority of observations are from coastal or urban locations (e.g. Gras, 1995; Mejia and Morawska, 2009; Cheung et al., 2011).”

*Figure 6. At the Marikana site there are 2 maxima for the CO concentration during the day and one maxima for SO<sub>2</sub>. The SO<sub>2</sub> one is attributed to the change in the boundary layer height. What are the 2 CO maxima attributed to?*

The CO maxima are attributed to domestic heating and cooking in the nearby residential areas. We have modified lines 4-7 on page 24057 to bring this up better:

“The morning and evening peaks originate from domestic space heating and cooking in the surrounding informal and semi-formal settlements (Venter et al., 2012; Hirsikko et al., 2012), which is seen also as two peaks in the CO concentration during corresponding time periods in Fig. 6.”

*p. 24057, l.27: “: : :The night-time nucleation mode originates from the evening household combustion peak, although most of these emissions are in Aitken mode.” I do not agree with this statement. The household combustion will produce much larger particles than the ones in the nucleation mode. Maybe there is some other source of the nucleation mode particles in this region. Is there any extensive gas (CNG, LPG or LNG) burning in the region?*

We do not know of any extensive gas burning in the region – the sources around the station have been characterised in more detail by Venter et al. (2012), which is now published.

Nights at Marikana are very calm and inversion is often strong, so that transport from sources further away is not feasible. However, also residential wood combustion has been observed to produce small amounts of nucleation mode particles (e.g. Tissari et al., 2008). The nucleation mode increase in Marikana is approximately 10% of the total concentration increase in the evening peak, which is in the same order of magnitude as the wood combustion experiments (Tissari et al., 2008).

We have modified this sentence as follows:

“The night-time nucleation mode appears simultaneously with the evening household combustion peak, as seen in Fig. 7. However, the increase in the nucleation mode particle concentration is approximately only 10% of the total particle concentration increase, which is comparable to previous measurements on residential wood combustion (e.g. Tissari et al., 2008).”

*p. 24059, l 1: “The diurnal variation in Rondonia, Brazil has a resemblance to the Marikana diurnal variation in that the highest concentrations from biomass burning are had during evening and night-time, however, there is no or little new particle formation during daytime in Rondonia (Rissler et al., 2006).” Rewrite the sentence as it does not make sense.*

This has been rewritten as:

“In Rondonia, Brazil, the highest concentrations from biomass burning occur during evening and night-time, which is similar to Marikana. However, there is no or little new particle formation during daytime in Rondonia (Rissler et al., 2006).”

*Figure 10. y-axis labels are not clear in this figure. Explain them in more detail in the caption. Also define in the caption what is on the top and what is on the bottom 2 figures.*

Fig. 10 caption has been rewritten as:

“On the top are presented the monthly correlation coefficient for CO and N100 concentrations for Botsalano and Marikana. The correlation coefficient has been calculated for hourly median CO and N100. On the bottom are presented the monthly median temperature and the number of MODIS burned area fire observations within 500 km radius of Botsalano (left panel) and Marikana (right panel).”

#### *Section 3.4 Spatial variation of the size distribution*

*I find the findings in this section very interesting, unfortunately I have had some problems in understanding the methodology of determining the size distribution within the source regions. What do the authors mean by: “The calculated times were then linearly interpolated to the DMPS time stamps, : : :” (p. 24062, l.11). Can the whole procedure be explained in more detail.*

*You use 10 minutes tie intervals. Does the algorithm for calculating back-trajectories have a resolution of 10 minutes? If not maybe it would have been better to calculate the average of the size distributions over the time period that the back-trajectories could have been meaningfully calculated.*

The procedure is easiest to illustrate with an example: let's assume that the back-trajectory arriving at 14:00 stays 80 hours over the re-circulation source region and 0 h over any other source region so that it will classify as re-circulation according to Table 5. Now if the back-trajectory arriving at 15:00 stays still 80 hours over the re-circulation source region but also some time over the industrial hub source region, it is discarded as unclassified.

Now there are two options: if we use hourly averaged size distributions, all size distributions from 13:30 to 14:30 are classified as re-circulation and all size distributions from 14:30 to 15:30 are unclassified. However, if the time over source region is interpolated between 14:00 and 15:00, the rate of change in the back-trajectories comes into play. If the back-trajectory at 15:00 visited the industrial hub source region for only 1 hour, all size distributions from 14:00 to 14:30 are classified as re-circulation. However, if the back-trajectory at 15:00 stayed 3 hours over the industrial hub source region, only size distributions from 14:00 to 14:10 are classified as re-circulation and the rest are unclassified.

The paragraph on page 24062, lines 8-18 has been modified to make it clearer, but we omitted longer explanations to keep the paper more compact:

“In order to obtain a more detailed picture of the size distribution within the source regions, hourly back-trajectories were used to select a subset of the measurements best representing each source region. For the selection the time spent over each source region in Fig. 11 was first calculated for each back-trajectory. The calculated time-over-source-region was then linearly interpolated to the DMPS time stamps, thus attributing to each size distribution a time the air mass had spend over each of the source regions. In this manner, each 10 minute size distribution could be classified according to the criteria listed in Table 5, which resulted in a total of 17 000 10 minute size distributions with well-defined source region origins. The criteria in Table 5 were set to select the air masses best representing each source region, while simultaneously minimizing the contribution from other source regions.”

*p.24064, 2nd paragraph. One of the reasons that you did not see any nucleation modes from the Kalahari region and in general a very low concentration of particles smaller than 100nm could be due to the scavenging of the small particles by dust particles. Kalahari is a desert region with a high concentrations of dust (sand). A similar effect has been observed in regions under the influence of dust storms (see Jayaratne, Rohan, Johnson, Graham R., McGarry, Peter D., Cheung, Hing Cho, & Morawska, Lidia (2011) Characteristics of airborne ultrafine and coarse particles during the Australian dust storm of 23 September 2009. Atmospheric Environment, 45(24), pp. 3996-4001.). This would be worth discussing in the paper.*

This has been included in the discussion. Please see also the reply Anonymous Referee 1.

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