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Interactive comment on “Boundary layer nucleation as a source of new CCN in savannah environment” by L. Laakso et al.

Anonymous Referee #2

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General comments:

In this work, the authors investigated particle formation and growth in the South African savannah region environment. They found that CCN-sized particles during the dry winter season are from biomass burning, while CCN-sized particles during the wet summer season are mainly from boundary layer new particle formation and growth. They suggested that the higher growth rates during the wet season are attributed to vapors of biogenic origin. Global model results were presented here to interpret the observations at the Botsalano game reserve site.

This is a very interesting and important work, one that has a good potential to deepen the understanding on biogenic emissions and the factors controlling nucleation and growth in such savannah environment. I would ultimately like to see the results of this

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effort published. However, the lacks of a critical analysis of the results and the large uncertainties of model simulation at the Botsalano game reserve site made the current version of this manuscript could not be accepted for publication in ACP. Based on the assessment above, I recommend major revision of the manuscript in its present form.

Most of the detailed discussions about the observed new particle formation, growth, and the contribution due to sulfuric acid at the Botsalano game reserve site during the same period have already been published in Vakkari et al. (2011). The potential highlight of this work is the discussion of the contributions of biogenic organic vapors to new particle formation and growth in such savannah environment. However, there was no attempt to look at the detailed measurements at this site to provide a convincing idea about the role of biogenic organic vapors in the formation of CCN-sized particles. It is no surprise that biogenic organic vapors are important in savannah environment. But the more interesting part is how biogenic organic vapors affect new particle formation and growth in such environment.

The simulated new particle formation and growth are only approximately 10-20% of the observations. It means amount of unknown sources and physical processes are missed in this simulation. The model does not provide acceptable results for boundary layer nucleation at this site, which is closely related to the major conclusion made in this study. When we use the model simulation to interpret the observations, one of the basic requirements is that model simulation should show good/reasonable agreement with the measurements. Otherwise, this kind of interpretation could mislead the readers' understanding.

Special comments:

1. There are a number of citation errors of references. For example, the manuscript of Laakso et al. (2008) appeared several times in this manuscript is not included in references list. Please check the citations carefully in the manuscript.

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2. P8505, L4-6. In addition to particle number concentration, aerosol composition and mixing state are also very important for cloud properties.
3. P8507, L17-18. How about the frequency of electricity breaks at the observation site? How to consider the missing data in the statistical analysis? Will the frequent electricity breaks bring any trends to the statistical analysis?
4. P8509, L26-28. Carbonaceous aerosol emission from large scale biomass burning is important for the simulations of primary particle number concentration and condensation sink, especially in this region. van der Werf et al. (2010) pointed out that the Global Fire Emissions Database version 3 (GFEDv3) monthly inventory data were highly variable. Therefore, the GFEDv3 data based on satellite data averaged over the years 1997–2002 is not suitable here. I suggest the authors to use the GFED3 emission monthly inventory for individual years (2006-2008) to do the simulation. Carbonaceous aerosol emission from large scale biomass for the individual years can be found in <http://www.falw.vu/gwerf/GFED/GFED3/partitioning/>.
5. P8510, L13-14. Biogenic monoterpenes are major sources of the condensable secondary organic vapors which are suggested to have significant contribution to particle growth here. Did the modeled monoterpene emissions taken from the GEIA database have seasonal variation? What are the major differences between GEIA biogenic monoterpene emissions and those from MEGAN?
6. P8513, L19-21. As shown in Fig.3 (a) and (b), the simulated particle formation rate is only approximately 10% of the observations. It means 90% of the observed particle formation rate could not be explained by model simulation. And it is clear that the model shows an opposite cycle in particle formation rate. The large differences between the observations and model simulations indicate that the simulation of new particle formation at this site may miss some important underlying physical processes which dominate the performance of nucleation here. Therefore, the following discussions and conclusions which are based on the modeling study are not strongly supported.

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7. P8513, L21-23. What is the role of vapors of organic origin in particle formation? Can it cover the rest 90% of the observed particle formation rate which is not captured by the model simulation in this study?

8. P8514, L13-15. The simulated particle growth rate in this study is only approximately 10-20% of the observations. I do not think model simulation presented here is suitable to interpret the observations at the Botsalano game reserve.

9. P8515, L1-2. Figure 4 (a) suggests that approximated fractions of particle growth due to sulfuric acid are less than 20% during the whole year. But model simulation shows that approximated fractions of particle growth due to sulfuric acid from May to October are high up to 40%. In my opinion, the observation and the simulation provide opposite conclusions. The observation suggests that sulfuric acid is not an important role in particle growth at this site. In contrast, model simulation suggests that sulfuric acid play a comparable role in particle growth at this site during dry winter. In additional, if organic vapors are important for seasonal cycle of particle growth rates at this site, the model employed in this study, which has large uncertainties of organic vapors simulation, is not suitable to interpret the observations.

10. P8515, L6-7. The model significantly underestimates particle formation rate and growth rate. Why can the model still capture the absolute number of different CCN-sized particles?

11. P8516, L1-9. The discussion of observed and simulated CCN indicates that model simulation significantly underestimates the contribution of new particle formation to CCN number concentration in wet season. It indicates that the model cannot capture the major characteristics of new particle formation at the site. Therefore the discussion of the contributions of primary particles, upper tropospheric nucleation, and boundary layer nucleation, which is based on model simulation here, is not convincing.

12. P8526, Figure 2. Latitude and longitude should be marked on the map. The legend for the colored trajectories should be added here.

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