Response to anonymous referee 1

We would like to thank the referee for the constructive comments on the manuscript. We present our answers to the comments below. The referee comments have been pasted below *in italic*.

General:
This paper investigates atmospheric new particle formation in a Savannah environment in Southern Africa. The analysis is based on a 18-month measurement data set using state-of-the-art instrumentation. While similar kinds of analyses have been made elsewhere, the unique location of the site increases the value of this paper. The paper is relatively well-written and, after some revisions, it can be considered scientifically sound. Before publication in ACP, there are a number of (mostly minor) issues that should be addressed in more detail.

Scientific comments:

Section 1

The authors mention the importance of new particle formation on the global aerosol budget. Please define whether you mean the budget of aerosol number, CCN or mass concentration and add a couple of more references to back up your statement. My impression is that new particle formation is very important for aerosol number, of some importance to CCN, but not at all for aerosol mass.
Author response: We have made the following changes:
"The importance of new particle formation to the global aerosol number and cloud condensation nuclei budgets has also been confirmed by global models (Spracklen et al., 2008; Merikanto et al., 2009)."
In addition, we are currently preparing a paper to quantify this effect in Southern Africa.

The authors mention long-term measurement conducted in North America and Europe. A few more examples of published measurement data sets should be given here.
Author response:
We have added more references:
Iida et al., 2008
Kulmala and Kerminen, 2008 and references therein,
Venzac et al., 2009

Section 3.3

The authors assumed that the total concentration of 2-3 nm particles was a hundred time the concentrations of similar-size charged particles. The assumption is justified with only one set of measurements. I am pretty sure in reality that this factor is not equation to 100 but varies quite a lot with atmospheric conditions. The authors should discuss a bit how sensitive their results are for this fact and what implications this variability might have on their

interpretations. The natural place for this discussion is page 30788 (lines 13-16) where the authors touch this subject but do not really discuss the resulting consequences.
Author response:
We have calculated the J_2(ions) also for factor of 50 and 200 for the ratio of ion and neutral clusters to estimate the sensitivity of J_2(ions) to this assumption.

Also the section on page 30788 has been changed:
"The fifth term in Eq. (2), which carries the assumption of the neutral cluster concentration being hundred times the measured total ion cluster concentration, is on average 25% (maximum 90%) of the sum of the terms 1–4 in Eq. (2). As a sensitivity study, we calculated the 2 nm ion formation rates for neutral cluster concentrations of 50 and 200 times the ion cluster concentration. For the factor of 50, the total ion formation rate, J_2(ions), is on average (median) 15% higher than for the factor of 100. For the factor of 200 the J_2(ions) is on average (median) 31% lower, respectively.

It seems that the authors use fundamentally different approaches when analyzing the particle growth rates from AIS and DMPS measurement? Why not to use the same method for these two data sets? Or why not use both methods for both data and then compare all these combinations? Please provide some reasoning.
Author response:
We have used two fitting techniques to determine the growth rate from the size distribution. In the log-normal technique a log-normal size distribution is fitted to the size spectrum at every time step. In the normal distribution technique a normal distribution is fitted to the concentration in each size channel. When the aerosol particle mode is smaller than approximately 5 nm, the mode is not log-normal and therefore the growth rate analysis cannot be based on log-normal fitting for the smallest size ranges of 1.5 – 3 nm and 3 – 7 nm for the AIS measurements.

On the other hand, the size resolution of both instruments is logarithmically evenly spaced and therefore the size channels are wider for larger particles. Naturally the growing particles stay longer in wider channels, which leads to broadening of the peak in time in larger sizes. Consequently above 20 nm the peak concentration in the size channel (in time) cannot any longer be defined with enough accuracy and we have to use for instance the log-normal technique to calculate the 10–30 nm growth rate from the DMPS measurements. Therefore, and also for consistency with earlier studies, the growth rates have been defined with different methods for AIS and DMPS measurements.

A recent modelling study comparing the different methods for determining the growth rate indicates that on the size regime from 5 to 20 nm both methods used here give similar results (Leppä et al., 2011). A comparison on these methods for this data set has been carried out by Vakkari (2009), but as we wanted to keep this article compact and the focus is on atmospheric measurements, not on method development, it has been left out.

Section 4.1
Page 30786, line 24: please explain what is meant by non-growing ion-bursts, i.e. how they are related to the different events categorized in Table 1.

Author response:
The non-growing ion bursts refer here to the cases when ions appear in the 2–20 nm regime in the AIS spectrum, but they do not present growth. In Table 1, this means classes apple, hump, rain and mixed-type. We have added this clarification to the manuscript.

As far as I understand, the reported aerosol formation (J10) and growth (GR) rates, as well as the frequency of the new-particle formation events, are at the high end of values observed in different environments. This is an important result and should be highlighted in the paper.
Author response:
We certainly agree with the referee. We state this result now more clearly in the results and conclusions and also in the abstract.

Related to the previous comments, at least a brief comparison of observed values of J10, GR, CS and event frequency to other locations should be made. How these values compare with other continental locations of different character (heavily polluted, urban, rural, remote).
Author response:
We have added a short comparison of J_{10}, GR and CS observations in continental locations in the discussion for J_{10}, GR and CS in Sect. 4.1 based on Kulmala et al. (2004 and 2005), Hamed et al. (2007), Kulmala and Kerminen (2008) and Shen et al. (2011).

Page 30787, lines 18-23: I am not fully convinced about the analysis here. I principle, the formation rate of 10 nm particles depends on 3 things: the nucleation rate, particle growth rate and coagulation sink. The latter two are in addition coupled with each other, such that the "survival probability" of growing nuclei depend on the ratio between the particle growth rate and their coagulation sink. The authors should make this clear with relevant references. Since authors have not measured the nucleation rate, they cannot really say for sure what causes seasonal variation in J10 (could be either the nucleation rate or the growth rate).
Author response:
We would like to thank the referee for pointing this out. This analysis is better fitted on page 30789, after the seasonal variation of ion formation and growth rates and CS have been discussed. It seems that the changes in the CS cannot explain the observed seasonal cycle of J_{10}. Although the J_2(ions) does not have seasonal cycle, it can be that the neutral particle nucleation rate has a seasonal cycle. However, the growth rate, also below 10 nm, has an identical seasonal cycle to the J_{10} seasonal cycle. We have added discussion on the possibility of the real nucleation rate seasonal cycle in the manuscript.

Page 30788: It is stated that CS is clearly elevated during the dry season. In my opinion, the seasonal variation of CS is quite moderate.
Author response:
The seasonal variation of the CS is not as clear as for e.g. the J_{10}, but the CS is continuously at least 30% higher during the dry season (from May to October) than during the wet season, which in our opinion is already quite clear difference. We would like to point this out for the reader by saying that the CS is clearly elevated during the dry season.

Page 30789, lines 1-5 and lines 14-19: The statements about correlations and relations should be supported by statistical measures, such as correlation coefficients and limits of confidence.
Author response:
We have added the correlation coefficients to the discussion. Correlation coefficient between J_{10} and CS is 0.26 and between GR and CS 0.15, which indicate that there is no correlation and all references to relations and correlations are removed.

For the fittings with the York bivariate fitting method we have given the standard errors, which can be used to calculate confidence limits at the confidence level required. However, we have decided to leave the comparison between AIS and DMPS out from the manuscript.

We have also left Fig. 7 and 8 out to reduce the number of figures in the manuscript.

End of section 4.1: What is really compared when comparing J10 from AIS and DMPS measurements with each other? The authors give a very hand-waving explanation that is really hard to follow. Please explain what this comparisons really means, or remove it from the paper.
Author response:
This was included in the manuscript mainly to show that the instruments are likely working as they give linear relationship for GR and J_{10}. A more thorough way of carrying out the comparison would be to calculate the total particle size distribution from the AIS spectrum assuming that above 10 nm the size distribution is in charge equilibrium, as measurements indicate (e.g. Laakso et al., 2007), and then calculating the J_{10}. However, as this topic is outside the scope of this article, we will leave the comparison out.

Section 4.2
Since sulfuric acid is calculated based on proxies, its concentrations probably have larger uncertainties as those based on direct sulfuric acid measurements. What are the expected uncertainty bars and how these uncertainties affect the interpretation of results in this section? I suppose that the results from Figure 9 are solid but how about Figure 10? Is proxy information accurate enough to state there is a clear seasonal cycle in sulfuric acid concentration?
Author response:
In Hyytiälä the measured sulphuric acid concentrations correlated well with all of the proxies and the proxy median matches exactly the measured median (Petäjä et al., 2009). There is noise, but also the relationship between proxy and measured concentration is linear, so there is no bias and therefore averages over longer periods will agree well with the actual concentrations.

However, the environment where the proxy was developed is very different from the environment in Botsalano. Therefore the rate constants, which have a small temperature and pressure dependency, and also the sinks and sources of the precursor oxidants could be different in Botsalano than in boreal forest. To assess the effect of these differences direct measurements of sulphuric acid are required; before that a quantified estimate of error cannot be given.

The estimated sulphuric acid concentrations in Botsalano are reasonable as compared to observations from Europe considering the oxidising potential of the atmosphere: the highest concentrations in Botsalano are $1 \cdot 10^8$ molecules cm^{-3}, whereas the measured sulphuric acid concentrations in boreal forest are $1 \cdot 10^6$ molecules cm^{-3} in midday and in Hohenpaissenberg, rural Southern Germany the daily highest concentrations reach $1 \cdot 10^7$ molecules cm^{-3} (Petäjä et al., 2009).

The authors report of bigger contribution of growth by sulfuric acid for smaller particles. Has similar been observed before in any other studies?
Author response:
This kind of behaviour has been reported by Fiedler et al. (2005). We have added this reference in the discussion.

Section 4.3
It is stated that 2-nm ion and 10-nm particle formation rates have similar patterns, as do also CS and GR source areas. What is the scientific interpretation of these findings?
Author response:
This pattern reflects the general pattern of the anthropogenic and natural emissions in southern Africa. The Karoo region has very little anthropogenic and biologic activity and therefore has low sources, which in turn result in low GR, CS and J. We have added reference to vegetation mapping (Friedl et al., 2002) and will point this out in the discussion.

Technical issues:

When providing equations like (3) and (4), the units of variables used in these equations should be given as well. Otherwise it is very difficult to apply these equations by others.
Author response:
Units have been added.

Page 30790, lines 19-24: The dependence of the growth rate on sulfuric acid concentration in different size regimes is more complicated as assumed here. Have a look at the recent paper by Nieminen et al. (2010, ACP 9777-9779) for a more detailed analysis of this issue.
Author response:
We appreciate the referee pointing out this issue.
Considering that we have estimated the sulphuric acid concentration with a proxy, the uncertainties in the growth by sulphuric acid are minor. However, we have re-calculated the sulphuric acid contribution based on Nieminen et al. (2010) as it is more accurate. Assuming 50% relative humidity gives following relations: for particles between 1.5 and 3 nm $1.15 \cdot 10^7$ molecules of H_2SO_4 cm^{-3} corresponds to 1 nm h^{-1} growth rate, for particles between 3 and 7 nm $1.5 \cdot 10^7$ molecules of H_2SO_4 cm^{-3} corresponds to 1 nm h^{-1} growth rate, for particles between 7 and 20 nm $1.7 \cdot 10^7$ molecules of H_2SO_4 cm^{-3} corresponds to 1 nm h^{-1} growth rate, and for particles between 10 and 30 nm $1.8 \cdot 10^7$ molecules of H_2SO_4 corresponds to 1 nm h^{-1} growth rate. These concentrations are approximately 50% higher than the previous estimates, which will reduce the estimated fraction of growth explained by sulphuric acid.

We also found that the parameters for the scaling factor k_3 (Eq. 4 in the discussion paper) were in wrong order in the text in Petäjä et al. (2009). The correct equation 4 is

$$k_3 = 1.4 \cdot 10^{-7} \cdot Glob^{-0.70} .$$

The correct scaling factor k_3 increases the estimated sulphuric acid concentration by approximately 50% from the previous estimate. However, the shape of the seasonal variation or the source area of sulphuric acid does not change (Fig. 10 and Fig. 18).

The increase in the estimated sulphuric acid concentration and the better estimate of contribution of sulphuric acid to the aerosol particle growth nearly cancel each other, so that the changes in the results (Fig. 11 and Fig. 19) are small.

I do not see the extra value of Figure 5 as compared with Figure 4. It is sufficient to say in the text that both polarities give very similar results. The statement "The differences between the polarities are negligible" in the figure caption is not correct: there are clear differences in J2 during months 9-12.
Author response:
Differences between the polarities indicate differences in the chemistry of the growing particles. Comparing the different polarities, we see for example that J_2 for positive ions has different annual cycle than the one for negative. The differences in growth rates of the smallest size fraction indicate differences in condensing vapours.

Although the paper is clearly written and easy to read, there are grammatical problems and clumsy sentences here and there. The language of the paper should be carefully checked out when preparing the final version of the paper. Below a few examples:
Author response:
We will check it carefully. The examples below as well as the comments by anonymous referee 2 have been corrected.

Page 30779, line 10: "..budget, measurements. . ."
Corrected.

Page 30779, lines 16-17: The sentence, "The concentrations of climatically important aerosol particles are due to their sources and sinks", is structured in a bit strange way.
Corrected.

Page 30782: Data. . .were, not was
Corrected.

Page 30783: "No a one to three modal log-normal. . ."
Corrected.

After equation 2: ". . .refer to. . ., to..2-3 nm particles, . . .to the coagulation sink. . .to the growth rate. . .". I do not think that the list can be structured like this.
Reformulated.

References

Fiedler, V., Dal Maso, M., Boy, M., Aufmhoff, H., Hoffmann, J., Schuck, T., Birmili, W., Hanke, M., Uecker, J., Arnold, F., and Kulmala, M.: The contribution of sulphuric acid to atmospheric particle formation and growth: a comparison between boundary layers in Northern and Central Europe, Atmos. Chem. Phys., 5, 1773-1785, doi:10.5194/acp-5-1773-2005, 2005.

Friedl, M. A., McIver, D. K., Hodges, J. C. F., Zhang, X. Y., Muchoney, D., Strahler, A. H., Woodcock, C. E., Gopal, S., Schneider, A., Cooper, A., Baccini, A., Gao, F. and Schaaf, C.: Global land cover mapping from MODIS: algorithms and early results, Remote Sens. Environ., 83, 287-302, doi:10.1016/S0034-4257(02)00078-0, 2002.

Hamed, A., Joutsensaari, J., Mikkonen, S., Sogacheva, L., Dal Maso, M., Kulmala, M., Cavalli, F., Fuzzi, S., Facchini, M. C., Decesari, S., Mircea, M., Lehtinen, K. E. J. and Laaksonen, A.: Nucleation and growth of new particles in Po Valley, Italy, Atmos. Chem. Phys., 7, 355-376, doi:10.5194/acp-7-355-2007, 2007.

Iida, K., M. R. Stolzenburg, P. H. McMurry, and J. N. Smith (2008), Estimating nanoparticle growth rates from size-dependent charged fractions: Analysis of new particle formation events in Mexico City, J. Geophys. Res., 113, D05207, doi:10.1029/2007JD009260.

Kulmala, M. and Kerminen, V.-M.: On the formation and growth of atmospheric nanoparticles, Atmos. Res., 90, 132–150, 2008.

Kulmala, M., Petäjä, T., Mönkkönen, P., Koponen, I. K., Dal Maso, M., Aalto, P. P., Lehtinen, K. E. J., and Kerminen, V.-M.: On the growth of nucleation mode particles: source rates of condensable vapor in polluted and clean environments, Atmos. Chem. Phys., 5, 409-416, doi:10.5194/acp-5-409-2005, 2005.

Kulmala, M., Vehkamäki, H., Petäjä, T., Dal Maso, M., Lauri, A., Kerminen, V.-M., Birmili, W. and McMurry, P. H.: Formation and growth rates of ultrafine atmospheric particles: A review of observations, J. Aerosol Sci., 35, 143–176, 2004.

Laakso, L., Gagne, S., Petäjä, T., Hirsikko, A., Aalto, P.P., Kulmala, M. and Kerminen, V.-M.: Detecting charging state of ultra-fine particles: instrumental development and ambient measurements, Atmos. Chem. Phys., 7, 1333–1345, 2007.

Leppä, J., Anttila, T., Kerminen, V.-M., Kulmala, M., and Lehtinen, K. E. J.: Atmospheric new particle formation: real and apparent growth of neutral and charged particles, Atmos. Chem. Phys. Discuss., 11, 2077-2122, doi:10.5194/acpd-11-2077-2011, 2011.

Merikanto, J., Spracklen, D. V., Mann, G. W., Pickering, S. J., and Carslaw, K. S.: Impact of nucleation on global CCN, Atmos. Chem. Phys., 9, 8601-8616, doi:10.5194/acp-9-8601-2009, 2009.

Nieminen, T., Lehtinen, K. E. J. and Kulmala, M.: Sub-10 nm particle growth by vapor condensation – effects of vapor molecule size and particle thermal speed, Atmos. Chem. Phys., 10, 9773-9779, doi:10.5194/acp-10-9773-2010, 2010.

Petäjä, T., Mauldin, III, R. L., Kosciuch, E., McGrath, J., Nieminen, T., Paasonen, P., Boy, M., Adamov, A., Kotiaho, T. and Kulmala, M.: Sulfuric acid and OH concentrations in a boreal forest site, Atmos. Chem. Phys., 9, 7435–7448, 2009.

Shen, X. J., Sun, J. Y., Zhang, Y. M., Wehner, B., Nowak, A., Tuch, T., Zhang, X. C., Wang, T. T., Zhou, H. G., Zhang, X. L., Dong, F., Birmili, W. and Wiedensohler, A.: First long-term study of particle number size distributions and new particle formation events of regional aerosol in the North China Plain, Atmos. Chem. Phys., 11, 1565-1580, doi:10.5194/acp-11-1565-2011, 2011.

Spracklen, D. V., Carslaw, K. S., Kulmala, M., Kerminen, V.-M., Sihto, S.-L., Riipinen, I., Merikanto, J., Mann, G. W., Chipperfield, M. P., Wiedensohler, A., Birmili, W. and Lihavainen, H.: Contribution of particle formation to global cloud condensation nuclei concentrations, Geophys. Res. Lett., 35, L06808, doi:10.1029/2007GL033038, 2008.

Vakkari, V.: Aerosol particles in a semi-clean savannah environment, Master's thesis, Helsinki University of Technology, Espoo, 2009.

Venzac, H., Sellegri, K., Villani, P., Picard, D., and Laj, P.: Seasonal variation of aerosol size distributions in the free troposphere and residual layer at the puy de Dôme station, France, Atmos. Chem. Phys., 9, 1465-1478, doi:10.5194/acp-9-1465-2009, 2009.