

Response to anonymous referee 2

We would like to thank the referee for the constructive and detailed comments on the manuscript, which certainly helped to improve the manuscript. We present our answers to the comments below. The referee comments have been pasted below *in italic*.

General comments

I recommend the paper for publication upon minor revisions. If the other comments by other reviewers are not too extensive, the paper needs no further review from my own side.

The current paper presents statistical results and characteristics of new particle formation events in Southern Africa on the African continent, where there is previously a limited amount of data. Formation events have a potential to significantly affect the global radiation balance of the earth-atmosphere system, since these events are a source of cloud condensation nuclei. Regional and global models however, have a difficulty in predicting where and when new particle formation takes place and how strong these formation events are since the mechanism behind it and what is required to observe them is still not known. Therefore, every new measurement in a new environment, such as in this study is extremely valuable to validate modeling attempts to predict the events and how they can affect the aerosol population, and hence the climate.

The overall quality of the paper is excellent. There are not too many messages, which makes the paper rather short and to the point. The paper is also extremely well structured, very easy to understand and the line of argument is very good. I have only some minor language remarks, and a few other points that need to be answered as outlined below. Finally, I think that there are too many figures in the paper.

Specific comments

Page 30778, lines 12-14. Voluntary correction: Consider removing this sentence to make abstract shorter and to the point.

Author response: Sentence removed.

Page 30778, lines 14-25. Voluntary correction: Consider rephrasing to make abstract shorter and avoiding repetition. It is mentioned twice that VOC emissions are important for growth rates.

Author response: We have rephrased is as:

“The growth rates had a clear seasonal dependency with minimum during winter and maxima in spring and late summer. The relative contribution of estimated sulphuric acid to the growth rate was decreasing with increasing particle size and could explain more than 20% of the observed growth rate only for the 1.5–3 nm size range. Also the air mass history analysis indicated the highest formation and growth rates to be associated with the area of highest VOC (Volatile Organic Compounds) emissions following from biological activity rather than the areas where the highest estimated sulphuric acid concentrations originated from.”

Page 30780, lines 9-17. *“In this study, supporting the EUCAARI project South African component (Kulmala et al., 2009), we give a more detailed analysis on the new particle formation observed on background savannah (Laakso et al., 2008).” Should read: “In this study, supporting the EUCAARI project South African component (Kulmala et al., 2009), we provide a more detailed analysis and an extension on the new particle formation observed at Botsalano (Laakso et al., 2008).” This helps the reader to understand that you are talking about the same measurement site in the second last and last paragraph respectively in section 1.*

Author response: Modified as suggested.

Page 30781, line 9. *Please include in the same sentence where the Karoo region is situated.*

Author response:

“The region west and south of Botsalano, on the Botswana side and the Karoo region in South Africa, has little industrial or other anthropogenic activities and forms the clean sector.”

Was replaced with

“The region west and south of Botsalano on the Botswana side and the Karoo region in western and central South Africa has little industrial or other anthropogenic activities and forms the clean sector. In this sector also biological activity is lower as the landscape is mainly semi-arid shrublands (Friedl et al., 2002).”

Page 30783, line 2. *Please remove “and the aerosol particle growth rate”, since the current paragraph and formula (1) do not include the growth rate explanation.*

Author response: Modified as suggested.

Page 30783, lines 17-18. *“The actual growth rate is obtained by tracing the maximum of the log-normal fit to the nucleation mode as it grows from 10 to 30 nm and making a linear fit to the obtained data points”. Consider replacing with for example: “The actual growth rate is obtained by tracing the increase of the geometric mean diameter of the log-normally fitted nucleation mode as it grows from 10 to 30 nm diameter and making a linear fit to the obtained data points”.*

Author response: Modified as suggested.

Page 30784, last paragraph. *I don’t understand this paragraph. Is it possible to explain it better without having to read Manninen et al. (2009b), or is this reference necessary for understanding? If the reference is not crucial for the understanding, then please rephrase.*

Author response: We would like to thank the referee for pointing this out. We have rephrased this as:

“The production of neutral 2–3 nm particles from recombination of opposite polarity ions, i.e. ion-ion recombination rate, was calculated similarly as in Manninen et al. (2009b). The recombination rate was calculated for the complete measurement period. For the new particle formation events the ion-ion recombination rate is given as median value during the linear growth of the 2–3 nm ion concentration, i.e. the period when the first term in Eq. (2) was defined. The upper limit of ion induced nucleation was estimated as a sum of recombination rate and the total ion formation rate $J_2(\text{ions})$.”

See also the reply to anonymous referee 1.

Page 30786, last paragraph of chapter 3.5. I don't understand this paragraph. Please rephrase.

Author response: This was rephrased as:

“For the air mass history study a $0.5^\circ \times 0.5^\circ$ grid was defined over the southern Africa. Each grid cell was then assigned with the mean of the observed concentrations at Botsalano for the trajectories passing through that cell. In other words, the value for each grid cell represents the mean concentration observed at Botsalano for all the trajectories passing through this grid cell. A minimum of five trajectories per cell was required for the statistical reliability.”

Page 30788, line 28. Please consider replacing “was” with “is”. You use the present tense in all other places.

Author response: Modified as suggested.

Page 30789, lines 10-12. Do you mean that if DMPS/SMPS data is missing, then the only way to calculate J10 and GR10-30 nm is to use AIS data? However, to be able to use AIS data for this, you first need to know if the AIS data method works? Is this what you are trying to say? If so, then please explain it more clearly.

Author response:

We agree with the referee that this issue was not discussed in enough length and thank the referee for pointing this out.

This would be one way to calculate J_{10} and GR_{10-30} if the DMPS is unavailable. However, this was not used here. A more direct way to calculate J_{10} from AIS would be to calculate the total aerosol particle size distribution from the ion size distribution assuming charge equilibrium. See also the response to anonymous referee 1 for more discussion.

We have decided to remove this comparison from the manuscript as it is outside the scope of this study.

Page 30789, lines 27-28. Do we expect based on the size dependent charging efficiency of the ambient aerosol that the ratio between DMPS and AIS J10 should be close to 6.6? If so, please add a one or two sentences explanation regarding this and the value of 6.6.

Author response:

We decided to remove this from the manuscript.

Pages 30791-30792, lines 25-28, and 1-9 respectively. If I understand it correctly, the SO2 concentration values are calculated using the SO2 data at the Botsalano site and by examining where the back trajectories came from when these SO2 values were measured? In any case, it is hard to understand this. Please explain more thoroughly. It is also hard to understand the part with the Highveld blue polygon area. Please explain also this more thoroughly.

Author response:

In order to clarify the method we modified also the description in sect. 3.5. This paragraph was modified as:

“The high SO₂ emissions from the industry in the Highveld, see discussion in Sect. 2.1 and Fig 1, show clearly on the SO₂ source area map based on the air mass history, Fig 13. The emissions are so large that for trajectories passing over the highest sources the concentration measured at Botsalano does not represent the concentration in the air mass before Highveld. In fact, if this is not accounted for, there appear artificial sources due south of Botsalano, Fig. 13 left side.

Therefore all back-trajectories passing over the highest sources in the Highveld – the area limited by the blue polygon in Fig. 13 right-hand map – were truncated so that the history of the back trajectory before it passed over Highveld was left out from the analysis. This removes the artificial sources in the left-hand map in Fig. 13 and results in the right-hand map. This map compares well with the SO₂ emission inventory, Fig. 1. Sources of SO₂ on the south western coast over 1000 km away from Botsalano (in Western Cape) are not visible, but otherwise the SO₂ source areas follow the shape of the emission inventory. The same cut was applied to air mass history studies of aerosol particles and other variables as well.”

Page 30795, lines 2-4. I don't fully agree. To me it looks like the hot spots of the high growth rates of all size ranges in Figure 19 look more like the hot spots of SO₂ and H₂SO₄ rather than like the hot spots of VOC emissions.

Author response:

The Fig. 19 presents the fraction of growth rate explained by estimated H₂SO₄, which naturally peaks at the highest H₂SO₄ sources. The conclusion on lines 2-4 is based on Fig. 14 and Fig. 16, which present the source areas of growth rate.

Page 30809, figure caption of Figure 7. Please try to incorporate the text “The GR10–30 increase slightly with increasing condensation sink, but the observations are too scattered for a linear fit” in the first paragraph of page 30789 and remove it from the figure caption. We want to avoid discussion of results in the figure caption.

Author response: This figure was removed.

Pages 30816-30820, Figures 14-18. Please avoid writing about the results in the figure captions. Please also consider removing as many figures as possible, since there are too many already. Especially the results from figures 15-18 can be based on figures 13 and 14, where the minimal differences between the figures can be explained by words rather than figures in the manuscript.

Author response:

The differences in the source areas of different polarity ion formation rate and 1.5–3 nm growth rate indicate changes in the chemical composition of the vapours contributing to the formation and growth. For instance, in Fig. 16 is visible how sulphuric acid contributes more to the growth of negative ions than positive. See also the reply to referee 1.

The CS and H₂SO₄ can affect the particle growth rates. Therefore Fig. 17 and 18 are important for the discussion related to the effect of these two variables to the observed high growth rates in the northern South Africa.

Fig. 19 is kept to show in a more quantitative way how small the area where sulphuric acid is relevant for the observed growth is.

Below are listed captions from Fig. 14-18 and the corrections made.

Was: “Figure 14. The source areas of the observed 10 nm formation rate and 10–30 nm growth rate from the DMPS measurements. The black dots represent (from left to right) Botsalano, Rustenburg and Johannesburg. The highest values originate in the north-eastern direction from Botsalano.”

New:

“Figure 14. The source areas of the observed 10 nm formation rate and 10–30 nm growth rate from the DMPS measurements. The black dots represent (from left to right) Botsalano, Rustenburg and Johannesburg.”

Was: “Figure 15. The source areas of the 2 nm ion formation rates for positive and negative ions separately and the total 2 nm ion formation rate. The black dots represent (from left to right) Botsalano, Rustenburg and Johannesburg. The highest values originate in the north-eastern direction from Botsalano.”

New:

“Figure 15. The source areas of the 2 nm ion formation rates for positive and negative ions separately and the total 2 nm ion formation rate. The black dots represent (from left to right) Botsalano, Rustenburg and Johannesburg.”

Was:

“Figure 16. The source areas of the ion growth rates for size ranges 1.5–3nm, 3–7nm and 7–20nm from the AIS measurements. The black dots on the map represent (from left to right) Botsalano, Rustenburg and Johannesburg. The highest values originate in the north-eastern direction from Botsalano, except for the 1.5–3nm growth rate, which exhibits higher values also for the Johannesburg-Highveld area.”

New:

“Figure 16. The source areas of the ion growth rate for size ranges 1.5–3 nm, 3–7 nm and 7–20 nm from the AIS measurements. The black dots on the map represent (from left to right) Botsalano, Rustenburg and Johannesburg.”

Was:

“Figure 17. Condensation sink. The CS originating in the Karoo south west from Botsalano are very low, whereas the CS from north easterly directions are slightly above the average of $4 \cdot 10^{-3} \text{ s}^{-1}$. The highest CS originates in the Highveld.”

New:

“Figure 17. The source areas of the condensation sink. The black dots on the map represent (from left to right) Botsalano, Rustenburg and Johannesburg.”

Not modified: “Figure 18. The source areas of the estimated H_2SO_4 concentration. The black dots represent (from left to right) Botsalano, Rustenburg and Johannesburg.”

Technical corrections

Page 30779, lines 16-17. “The concentrations of climatically important aerosol particles are due to their different sources and sinks.” Suggested correction: “The concentrations of climatically important aerosol particles depend on their different sources and sinks.”

Author response: Modified as suggested.

Page 30779, lines 26-28. “On the other hand, the number of long-term continuous measurements of aerosol particle properties in Europe and North America, observations are increasing (e.g., Manninen et al., 2010).” Suggested correction: “Conversely, the number of long-term continuous measurements of aerosol particle properties in Europe and North America are increasing (e.g., Manninen et al., 2010).”

Author response: Modified as suggested.

Page 30779, line 28. “For instance, the only reported . . .”. Please delete “For instance”, and place the text in a new paragraph. Otherwise it feels as if the Botsalano measurements have got something to do with the European and American measurements.

Author response: Modified as suggested.

Page 30780, line 26. “north of”. Should read: “north of the”.

Author response: Modified as suggested.

Page 30781, line 7. “Highveld”. Should read: “Highveld area”.

Author response: Modified as suggested.

Page 30783, line 10. “was taken to equal”. Should read: “was set to zero”.

Author response: Modified as suggested.

Page 30784, line 9. “polarity 1.5-3 nm”. Should read: “polarity for 1.5-3 nm”.

Author response:

“GR[±] to the growth rate of respective polarity 1.5–3 nm ions”

was modified as

“GR[±] to the growth rate of 1.5–3 nm ions of respective polarity”

Page 30784, equation (2). No explanation is made regarding the difference between $N_{(<3nm)^{\pm}}$ and N_{\pm}

Author response: Subscript <3nm was removed. It is not needed and is misleading.

Page 30784, line 24. “the period when the first term in Eq. (2) was defined.” Should read: “during the period when the first term in Eq. (2) was calculated.”

Author response: Modified as suggested.

Page 30784, line 25. “a sum of recombination”. Should read: “the sum of the recombination”.

Author response: Modified as suggested.

Page 30785, lines 13-14. “to the Eq. (4) by Petäjä et al. (2009)”. Should read: “to Eq. (4) in Petäjä et al. (2009)”.

Author response: Modified as suggested.

Page 30786, line 15. “to a 0.5”. Should read: “in a 0.5”.

Author response: Modified as suggested.

Page 30787, line 1. “continental”. Should read “the continental”.

Author response: Modified as suggested.

Page 30788, line 17. *“with mean”*. Should read: *“with a mean”*.

Author response: Modified as suggested.

Page 30788, line 25. *“for period 09:30–10:30 LT, which is the hour following the median observed new particle formation onset.”*. Correction suggestion: *“for the period 09:30–10:30 LT, which is one hour later compared to the median hour of the onset of new particle formation events.”*.

Author response: Modified as suggested.

Page 30789, line 3. Please consider replacing *“defined”* with *“calculated”*.

Author response: Modified as suggested.

Page 30789, lines 19 and 24. *Standard errors of what?*

Author response: These were removed from the discussion.

Page 30791, line 5. Please consider to remove *“However”* in the sentence, since it gives the impression that the results are not consistent with what you discuss in lines 3-4.

Author response: Modified as suggested.

Page 30791, lines 10-12. Please rephrase sentence, since it is hard to understand.

Author response: The first two paragraphs on page 30791 (lines 1-15) were modified:

“The estimated sulphuric acid calculated according to Petäjä et al. (2009) has a clear seasonal cycle in Botsalano with maximum during the dry season from May to July, Fig. 10. This cycle is nearly the opposite of the cycle of the 10–30 nm aerosol particle growth rates. However, the growth rate calculated from sulphuric acid does not have seasonal cycle, Fig. 11. For this comparison the estimated sulphuric acid values are medians for the period when the growth rates are determined.

The fraction of growth rate explained by the estimated sulphuric acid is decreasing with increasing particle size, which is in line with previous observations from northern hemisphere (Fiedler et al., 2005). On average, the estimated sulphuric acid concentration can explain 25% of the 1.5–3 nm growth rate, 16% of the 3–7 nm growth rate, 14% of the 7–20 nm growth rate and 10% of the 10–30 nm growth rate.”

Page 30791, line 23. Please consider reversing *“air”* and *“polluted”*.

Author response: Modified as suggested.

Page 30792, line 18. *“show a same kind”*. Should read: *“show the same kind”*.

Author response: Modified as suggested.

Page 30793, line 24. *“in semi-“*. Should read: *“in a semi-“*.

Author response: Modified as suggested.

Page 30794, line 15. *“did not have seasonal variation”*. Should read: *“did not have a seasonal variation”*.

Author response: Modified as suggested.

Page 30795, lines 7-8. Consider replacing “The seasonal variation and the source areas suggest the growth to originate in VOC emissions following from biological activity” with “The seasonal variation and the source areas suggest that occasions with high growth rates are connected to areas with high VOC emissions following from biological activity”.

Author response:

This was rephrased as:

“The seasonal variation in the growth rate suggests biological activity to contribute significantly to the growth; furthermore, the air mass history study connects highest growth rates to the areas of highest biogenic VOC emissions emphasizing the importance of biogenic VOC’s in the growth.”

Page 30802, Table 2. Please reverse the order of the GR (1.5-3nm) and GR (7-20nm) for better readability.

Author response: Modified as suggested.

Page 30805, Figure 2 caption. Please reverse the order of “formation” and “growth”.

Author response: Assuming that the referee means Fig. 3 caption, this was modified as suggested.

Page 30806, Figure 4 caption. Please delete “The formation rate does not have a seasonal dependence.” to avoid discussion of results in the figure caption.

Author response: “The seasonal variation is strongest in the 3–7nm size range.” and “The formation rate does not have a seasonal dependence.” were removed from the caption.

Page 30807, Figure 5 caption. Please delete “The differences between the polarities are negligible.” to avoid discussion of results in the figure caption. Please also explain averaging period and the numbers in the left panel.

Author response: The caption was modified as:

“Monthly averages of positive and negative ion growth and formation rates presented separately for both polarities. The number of events analysed for each month and size range are also shown.”

Page 30808, Figure 6 caption. Please change the caption text to something like: “The monthly averaged condensation sink calculated during 09:30 and 10:30 LT for all days, i.e. including event and non-event days”.

Author response: Modified as suggested.

Page 30809, Figure 7. Please consider to remove the figure. There are too many already, and the relation is anyway not linear. It is enough to describe the findings using written words in the manuscript.

Author response: This figure will be removed from final manuscript.

Page 30810, Figure 8 caption. “is mean”. Should read: “is the mean”.

Author response: Modified as suggested.

Page 30810, Figure 8 caption. Please reverse the order of “formation” and “growth”.

Author response: Modified as suggested.

Page 30810, Figure 8 caption. Please add a text explaining the fitting depicted with the red curve and the equation of the fit.

Author response: This figure will be removed from final manuscript.

Page 30812, Figure 10 caption. Please add that it is monthly averages

Author response: The caption was modified as:

“The monthly averaged estimated H₂SO₄ concentration calculated during 08:00 and 14:00 LT.”

Page 30813, Figure 11. Please consider to replace figure with a table. There are too many figures, and there are so many values in the figure, that a table might be more suitable.

Author response:

We agree that there were too many numbers in this figure and we will remove the percentages indicating the fraction of growth rate explained by sulphuric acid. However, in our opinion this figure is important to show that the sulphuric acid cannot explain the observed seasonal variation in the growth rates even granting the high uncertainty in the sulphuric acid proxy.

Page 30816, Figure 14. Please reverse the order between the left and the right panel in the graph.

Author response:

To be consistent with figures 3-5 GR is kept on left panel. However, in the caption the order of formation and growth was reversed: “The source areas of the observed 10–30 nm growth rate and 10 nm formation rate from the DMPS measurements. The black dots represent (from left to right) Botsalano, Rustenburg and Johannesburg.”