Reviewer#2:

For the present study the authors measured particle chemical composition in a subtropical urban environment by means of an Aerosol Mass Spectrometer (c-TOF-AMS). During the measurements five new particle formation (NPF) events were observed. Regardless of the instrument's lower detection limit of 50 nm, the authors asses the data with the clear aim to evaluate particle chemical composition during NPF and the consecutive growth.
One major result of the study is that all measured particles are predominantly composed of organic molecules, both when NPF takes place and also on non-nucleation days. From calculating the mass fractions f43 and f44 the authors assign the particle chemical composition to different classes of organic species. By plotting f43 vs. f44 it is shown that the chemical composition of particles on NPF event days differs from composition during non-event days. According to the authors the f43 vs. f44 plotting method can be used to distinguish the particle source, whether they are pure traffic generated particles or origin from NPF.
The study is giving valuable information on particle composition in a subtropical urban environment. The allocation of the particle composition to different sources, by means of the f43 vs. f44 plot, is a straight forward approach and is a considerable contribution to the scientific community. The manuscript is written in a comprehensible way and is well structured.
Therefore, the manuscript is suited for publication in ACP. However, several issues need be assessed before publication.

General comments:

1. A major deficiency of the study is the fact that the lower detection limit of the c-TOF-AMS is in the size range between 50 and 100 nm. This size range is not appropriate to analyze the chemistry driving NPF. However, in many parts of the manuscript the authors state that the data gives insight into the chemistry of NPF. In my opinion, this is not the case. The data obtained from the c-TOF-AMS can only be attributed to the growth of particles several hours after the nucleation process. This fact should be made much more clear throughout the whole manuscript.

   Response:

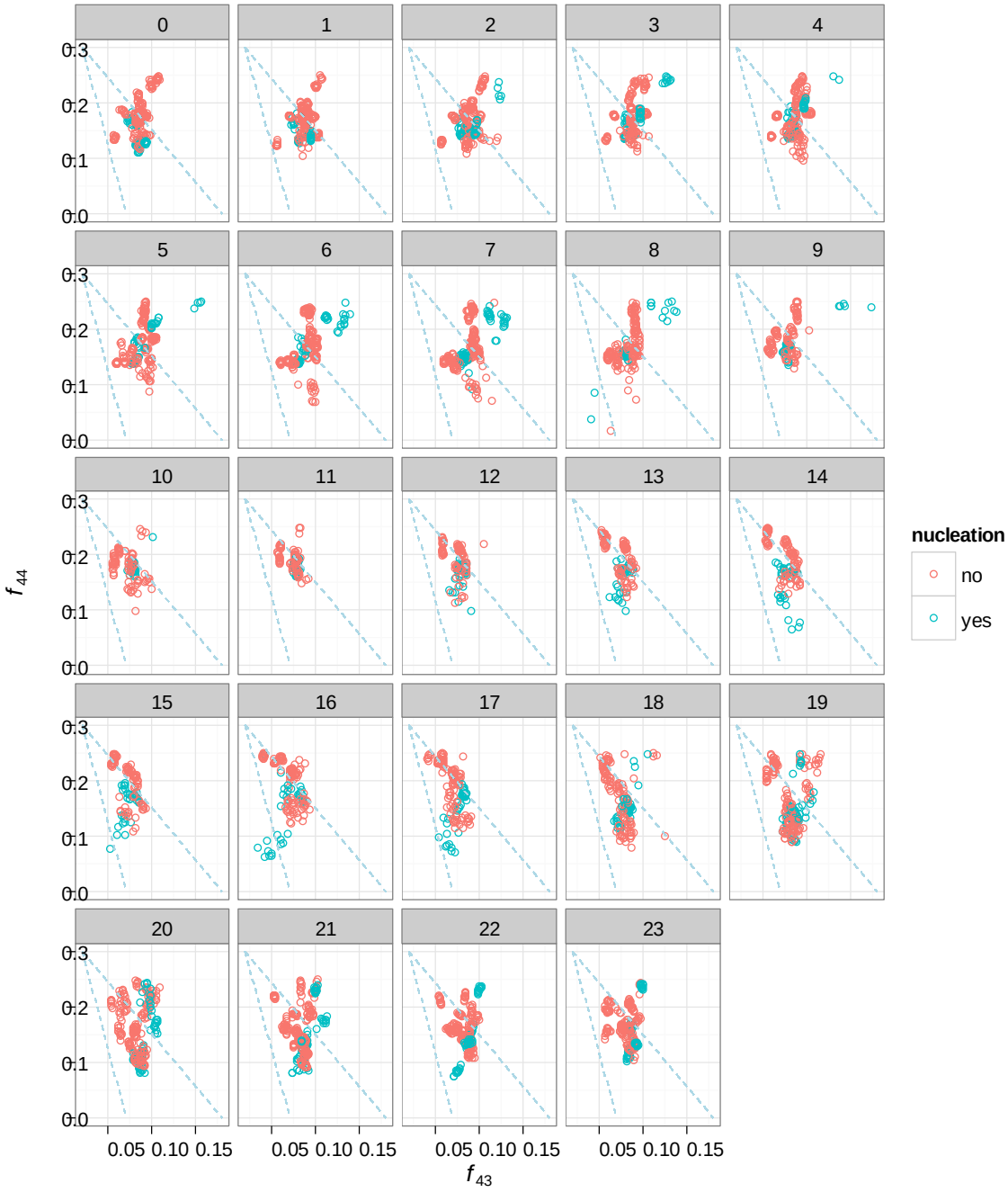
   We have added the following sentence to the manuscript to address this issue (page7, line 23):

   "The analysis and interpretation of the chemical composition of the newly formed particles focusses mostly on the particles in the growth phase due to the lowest detection limit of the C-TOF AMS which does not permit to see them in the formation phase."

2. A possible distinction of traffic related aerosols from particles originating from NPF, by means of the f43 vs. f44 plot, is the principal finding of the present study. However, these conclusions are only based on three measured nucleation events. In my opinion some more evidence should be supplied. According to the authors two more NPF events were recorded. Therefore, at least these two events should also be analyzed in respect of the f43 vs. f44 plot.

   Response:

   The additional two NPF events have been analysed for f_{43} vs f_{44}. To make the comparison between events and non-event days easier, event and non-event days have been plotted on the same figure. Therefore, figure 6 and 7 have been replaced with the updated figure6 which is shown below:



3. Further, the c-TOF-AMS is giving the possibility to analyze chemical composition for different particle size ranges. In my opinion, this is a clear advantage of the instrument. Unfortunately the measurements were not analyzed in this respect to a satisfying degree. Especially in section 3.3, where the chemical composition is assessed, the size information should be considered. I think it would be beneficial for the study to compare the chemical composition of particles in different size ranges. Therefore, some insight into the growth process could be obtained.

Response:
Please see the response to the reviewer3's comment number 10.

4. Additionally, the captions of virtually all figures should contain more information. Detailed descriptions give the reader the possibility to understand the figures in a minimum of time.

Response:
More detailed descriptions have been added to the captions as follows:

- *Figure 1: Average diurnal pattern of solar radiation (SR), humidity, temperature and condensation sink (CS) as well as the measured data on nucleation and non-nucleation day at s12 and s25. Shaded areas represent the 95% confidence interval in the mean. Data includes 5 and 51 nucleation and non-nucleation days.*

- *Figure 2: Average diurnal pattern of organics, nitrate, sulphate and ammonium concentrations as well as the measured data on nucleation and non-nucleation days at s12 and s25. Shaded areas represent the 95% confidence interval in the mean. Data includes 5 and 51 nucleation and non-nucleation days.*

- *Figure 3: Time series of the particle number/volume size distributions (top two graphs) and mass concentration of particle species and their fraction to the total (bottom two graphs) at s12. Solid lines are calculated using LOESS smoothing technique and the shaded areas represent the 95% CI.*

- *Figure 4: Time series of the particle number/volume size distributions (top two graphs) and mass concentration of particle species and their fraction to the total (bottom two graphs) at s25. Solid lines are calculated using LOESS smoothing technique and the shaded areas represent the 95% CI.*

- *Figure 5: f_{44}vs f_{43} at each hour of the day for all data measured during nucleation and non-nucleation days. The triangle from Ng et al. (2010) is drawn as a visual aid. Data includes 5 and 51 nucleation and non-nucleation days.*

- *Figure 6: SSE versus the number of clusters. The sharp decrease in SSE stops at 5 clusters forming an elbow.*

- *Figure 7: The f44 vs f43 plot for nucleation days and non-nucleation days with the five clusters and their 95% confidence epsilon (a). The contributing proportion of each cluster to the total for nucleation and non-nucleation days (b). The triangle from Ng et al. (2010) is drawn as a visual aid.*

- *Figure 1: Diurnal variation of the f57 during the nucleation and non-nucleation days. Solid lines have been calculated using the LOESS smoothing method and the shaded areas represent the 95% confidence interval. Data includes 5 and 51 nucleation and non-nucleation days. The data includes weekdays and weekend as no exclusions were applied to increase the statistical power.*

- *Table 1: Duration of each nucleation event as well as their average growth rates. Growth rates were calculated by a linear fit from 9nm to 20nm.*

Specific comments:

Abstract

5. Page 27946, line 6: Please state in which diameter range the chemical composition and number size distribution was measured.

Response:
The relevant sentence has been changed to (page 1, line 18):
"An Aerodyne compact Time-of-Flight Aerosol Mass Spectrometer (c-TOF-AMS) and a TSI Scanning Mobility Particle Sizer (SMPS) measured aerosol chemical composition (particles above 50nm in vacuum aerodynamic diameter) and PNSD (particles within 9-414 nm in mobility diameter),…"

6. Page 27946, line 11: Please state whether you relate to absolute- or relative humidity.

Response:
The relative humidity was measured, therefore, "humidity" has been replaced with "relative humidity" throughout the manuscript.

Introduction

7. Page 27947, lines 7-8: Please add some references for the statement:" … *these events are one of the main sources of ultrafine particles (UFPs; particles smaller than 100 nm), in addition to combustion emitted particles.*"

   Response:
   The following paper has been cited (page2, line 15):

   "Morawska, L., Wang, H., Ristovski, Z., Jayaratne, E. R., Johnson, G., Cheung, H. C., Ling, X., and He, C.: JEM Spotlight: Environmental monitoring of airborne nanoparticles, Journal of Environmental Monitoring, 11, 1758-1773, 10.1039/B912589M, 2009."

8. Page 27948, lines 4-6: Please state clearly that particle chemical composition was not measured during NPF but during the following growth process.

   Response:
   To address the reviewer's comment, the relevant sentence has been modified as follows (page3, line 7):
   "To address this gap in knowledge, the main aim of this study was to determine the role of chemical species in NPF events growth process in a subtropical urban environment."

Materials and methods

9. Page 27949, line 11: Please state which meteorological parameters were measured.

   Response:
   The following sentence has been added (page3, line 19):
   "Meteorological parameters including wind speed/direction, relative humidity and solar radiation were measured using a 'Monitor Sensor' weather station."

10. Page 27950, line 7: Which particle diameter definition does Dva stand for?

    Response:
    To address the reviewer's comment, the relevant sentence has been modified as follows (page5, line 12):
    "Ratio of mobility diameter (D_m), measured by the SMPS, to vacuum aerodynamic diameter (D_{va}) is a function of size, composition, shape and relative humidity for ambient particles."

11. Page 27951, line 6: Generalised Additive Model (GAM): Why was a modeling approach necessary to analyze the diurnal patterns? Some more explanation would be appropriate at this point. A figure comparing the measured data and the model results would be helpful.

    Response:
    GAM modelling was initially selected to help us better understand the data pattern and trend, however, we have found LOESS method a simpler approach which suits our aims and well represent our data. Therefore, GAM model has been replaced with the LOESS model throughout the paper. Consequently, the following modifications have been applied throughout the paper (page 6, line 8):

    "LOESS Smoothing: In order to better understand the trends and patterns of the data, LOESS technique as a locally weighted polynomial regression smoothing method was selected and applied to the data. The resulting smooth functions, and their 95% confidence intervals, indicate the trends of the relevant data throughout the paper.(Cleveland and Devlin, 1988)"

    In addition, all the related graphs have been replotted and they now include the actual measured the data as well.

Results and discussion

12. Page 27951, lines 18-20: Please state how many non-event days were considered at each site.

Response:
The following sentence has been added (page6, line 23):

"PNSD data was available for 6, 18, 11, 13, and 8 days in s1, s4, s11, s12, and s25 respectively."

13. Page 27952, lines 3-4: What could be the reason for a lower CS on nucleation days? Perhaps on NPF event days there was less traffic or another prevailing wind direction. Did the authors observe a typical wind direction during NPF?

Response:
The following sentence has been added (page 7, line 4):
"The wind speed/direction have not been plotted as they did not show a typical trend during the nucleation and non-nucleation days"

14. Page 27952, lines 24-26: The authors state that "*… the role of precursors on the NPF events can still be investigated as the condensable vapours responsible for NPF events do condense on pre-existing particles which are detectable by the instrument.*" I am not convinced by this statement. It was shown by Winkler et al. (2012) that different organic species condense on particles of different diameters. In their study Winkler et al. (2012) found that particle chemical composition differed significantly when comparing particles with 10 nm to particles with 40 nm diameter. As the lower detection limit of the c-TOF-AMS is somewhere between 50 nm and 100 nm no conclusion on the composition of freshly formed particles can be drawn from this data.

Response:
We agree with the reviewer's comment and have removed the related sentence

15. Page 27953, lines 10-13: In line 10 the authors state that "*…no distinctive trend in sulphate mass concentration was observed during the first event.* ". However, contradicting this statement the authors state in line 13: "*Sulphate followed similar trend to this during the events observed at S25.*" The paragraph clearly needs to be rephrased.

Response:
Section 3.2 (evolution of chemical composition of newly formed particle) has been re-written as follows (page 7, line 23):

"The analysis and interpretation of the chemical composition of the newly formed particles focusses mostly on the particles in the growth phase due to the lowest detection limit of the C-TOF AMS which does not permit to see them in the formation phase. Particle volume distribution (PVD) was calculated from the PNSD data as it is a better measure for comparison with the mass.

It has been previously determined in the literature that the main contributing species to NPF events and subsequent particulate growth are sulphate, nitrate, organics and ammonium (Zhang et al., 2004). Therefore, the percent fractions of each of these chemical species were calculated by dividing the mass concentration of each chemical species by the total (sulphate + nitrate + ammonium + organics).

Figure 3 illustrates the evolution of PNSD, PVD, and mass concentration of chemical species (organics, nitrate, sulphate and ammonium) during the three consecutive NPF events at s12. In general, mass concentration of the aerosol species followed the evolution of particle volume distribution as expected. Ammonium, sulphate and nitrate mass fractions peaked just before the particles volume increase due to the growth of newly formed particles. However, organics mass fraction followed the opposite trend, with a significant rise after the increase in particles volume due to the growth of new particles. In other words, the fraction of organics increased and the fraction of ammonium, sulphate and nitrate decreased when the newly formed particles grew enough to dominate the particles volume. This shows the import contribution of organics to the growth of newly formed particles.

Time series of PNSD and mass concentration of particle species during two NPF events happening on two consecutive days at s25 are illustrated in Figure 4. The mass concentrations of the chemical species and their fractions followed similar trends as the ones at s12. At s12, the magnitude of mass fractions changed from almost 50, 30, 10, and 10% before the nucleation to 70, 20, 6, and 4% after the event for organics, sulphate, ammonium and nitrate and respectively. At s25, the changes in mass fractions were more dramatic as they changed from 40, 25, 25, and 10 % before the event to 85, 5, 5, and 5% after the nucleation occurred."

16. Page 27953, lines 22-23: Referring to Fig. 5 the authors state: "*Ammonium, sulphate and nitrate mass fractions peaked around the start of nucleation and subsequently decreased after the event.*" With a little good will I can see this behavior for site S12 in Fig. 5. But at site S25 I cannot see this behavior. Especially sulphate does not behave as the authors stated. I suggest the authors focus on the temporal behavior of the individual mass fractions after the particles reached the detection limit of the c-TOF-AMS. This is reasonable, as it is not clear where the signal comes from before the particles reached the threshold diameter.

Response:
This has been addressed, please see the response to the previous comment (15).

17. Page 27954, lines 10-11: I miss information on particle size in Figs. 6 and 7. Which particle diameters are plotted? I think it would be most interesting to add different particle diameters to the figures. This could be done by differently coloring the single particle diameter ranges. . *Response:*

Response:
Total chemical mass data was used, the relevant sentence has been modified to clarify it (page 8, line 23):
"Firstly, f_{43} and f_{44} were calculated for the total chemical mass data on nucleation days and non-nucleation days"

18. Page 27954, lines 18-20: Please rephrase or delete the sentence, as its content seems to be already stated in the sentence above.

Response:
The relevant sentence has been deleted.

19. Page 27954, lines 21-27: At the beginning of the paragraph the authors state: "… *the aerosol components reached the bottom left side of the triangular region*". Contradicting this statement, the authors write at the end of the paragraph: "… *they clustered at the middle right hand side of the triangle.*" I suggest a revision of the paragraph.

Response:
"middle right" has been replaced with "middle left" (page 9, line 4)

Summary and conclusions

20. Page 27955, lines 23-26: Please state that the nucleation events were only observed at two of the five measurement sites.

Response:
The relevant sentence has been rephrased to (page 9, line 30):
"Five NPF events, with growth rates ranging from 3.3-4.6 nm.hr^{-1}, were observed at two of the five sites, and the NPF events happened on days with relatively lower relative humidity, and higher solar radiation and temperature than non-event days."

21. Please state whether you relate to absolute- or relative humidity.

Response:

The relative humidity was measured, therefore, "humidity" has been replaced with "relative humidity" throughout the manuscript

Table and figures

General comment: Please add more details to the figure captions.

Response:
Please see the response to the 4th comment.

22. Page 27962, Table 1: Please state for which particle size range the growth rate was determined.

Response:
The following sentence has been added to the caption of Table 1:
Growth rates were calculated by a linear fit from 9nm to 20nm.

23. Page 27963, Figure 1: What do the shaded areas denote?

Response:
The following sentence has been added to the figure's caption for clarification:
"Shaded areas represent the 95% confidence interval in the mean"

24. Please replace humidity by relative humidity.

Response:
The relative humidity was measured, therefore, "humidity" has been replaced with "relative humidity" throughout the manuscript.

25. How many nucleation and non-nucleation days were averaged for the figure?

Response:
The following sentence has been added to the caption of Figures 1 and 2:
"Data includes 5 and 51 nucleation and nucleation days respectively."

26. Comparing Fig. 1 to Figs. 3 and 4, I cannot consent that all nucleation events started at 10 AM. Is the point in time an average value? The same fact applies to Fig. 2.

Response:
It was an average value but we decided to remove the vertical lines representing the start of nucleation as it can be misleading.

27. Page 27965 & 27966, Figures 3 & 4: The figures would be much easier to read with the colorbars moved to the right hand side.

Those figures have been replotted as follows:

Figure 3

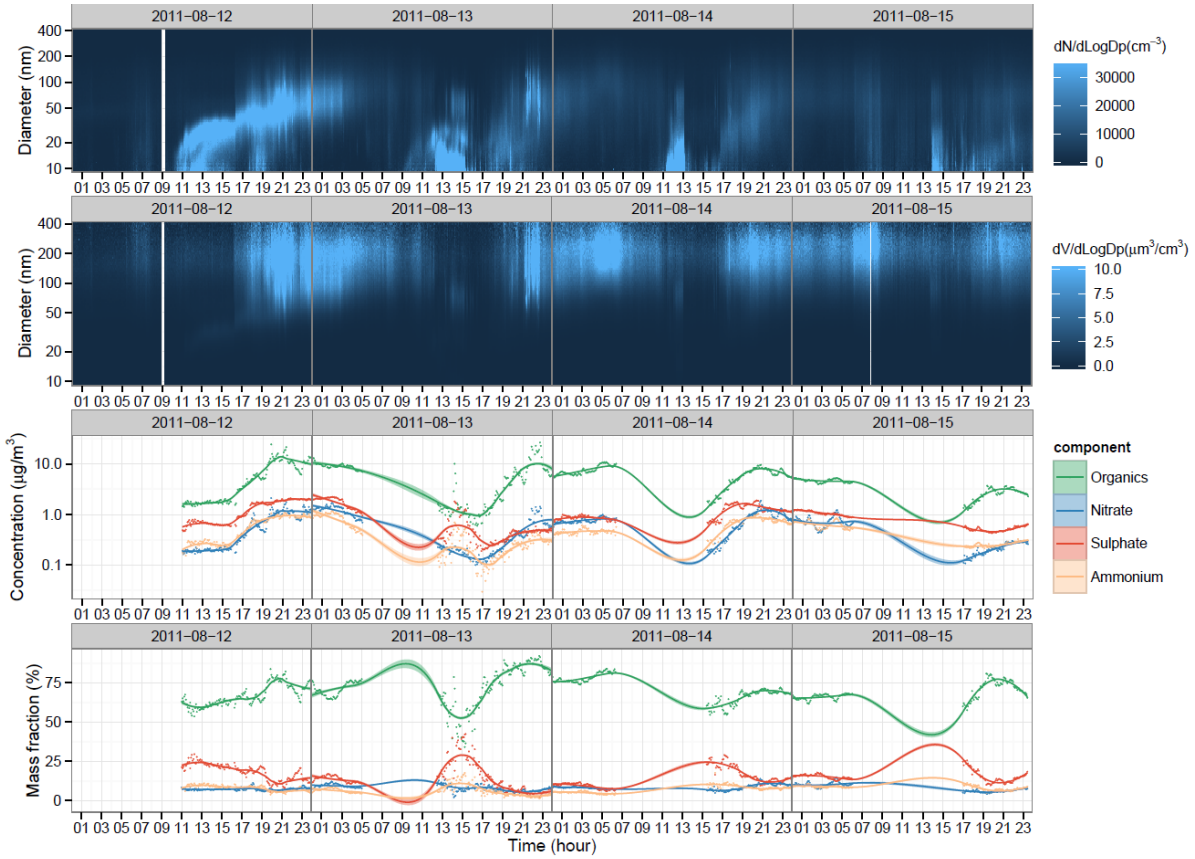
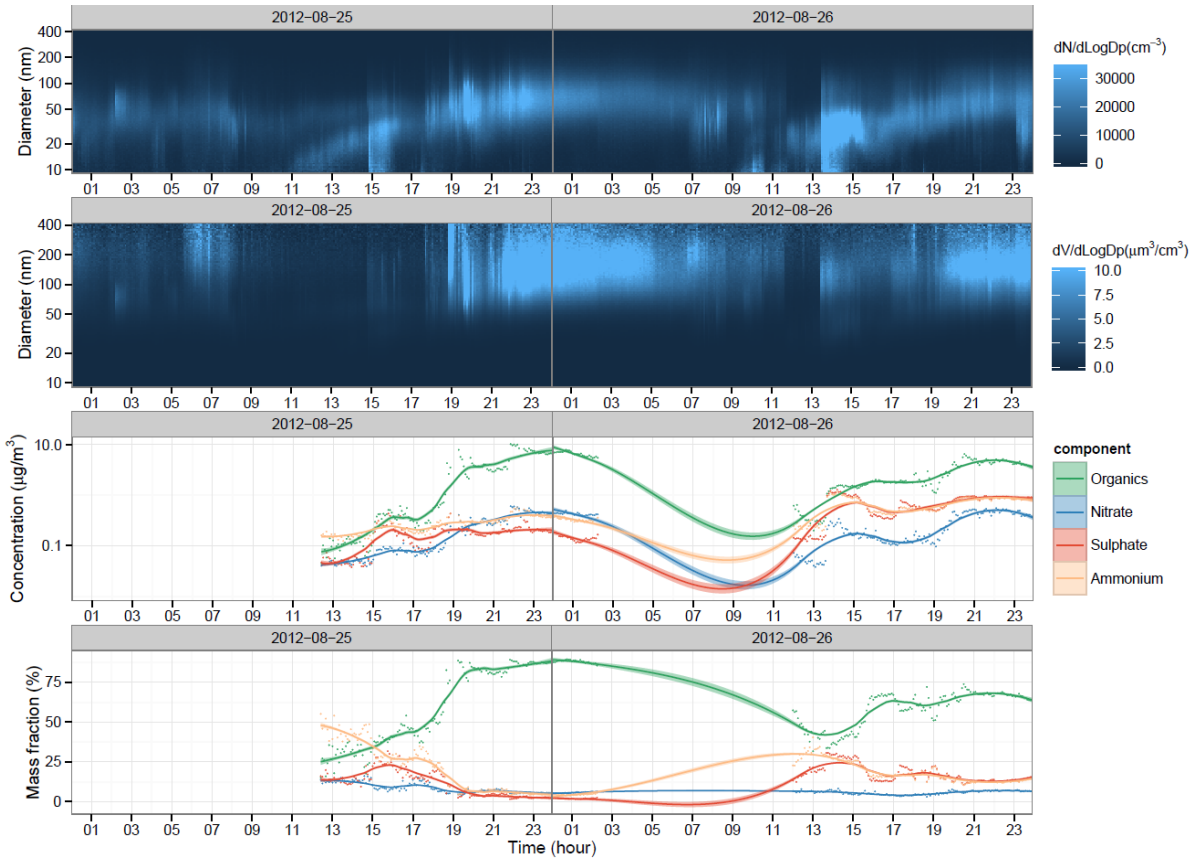


Figure 4



28. The time scale of the figures should be revised, with focus on an even hourly spacing of the ticks. Please denote what the blue vertical lines indicate.

Response:
Those figures have been replotted, please see the response to the previous comment.

29. Page 27967, Figure 5: Please denote the particle size range in which the chemical species were determined. I'm aware that it is already mentioned in the text but it is still helpful to have this information also in the figure caption.

Response:
That figure has been replaced with Figure3&4.

30. The time scale of the figures should be revised, with focus on an even hourly spacing of the ticks. In the moment a period of 15 hours is denoted by 5 and 10 ticks, respectively.

Response:
Please see the response to the comment 24.

31. Page 27968, Figure 6: Which particle size range is considered in this figure? How many non-event days were analyzed for this plot?

Response:
Figures 6&7 have been replaced with Figure5 and it includes the total chemical mass data. Please see the response to the 17th comment.
The following sentence has been added to the caption of figure5:
"Data includes 5 and 51 nucleation and non-nucleation days"

32. Page 27969, Figure 7: It would be interesting to denote at what times the growing particles reached the detection limit of the AMS.

Response:
We have added the following sentence to the Figure's (now Figure 5) caption:
"Within the period of 15-17, the newly formed particles grew enough to have significant contribution to total signal"

33. Page 27970, Figure 8: How can the authors be sure the red arrow represents the typical composition of traffic aerosol, did other studies find similar patterns?

Response:
Further details regarding the clustering of f_{44}vs f_{43} data has been added, please see the response to reviewer3's comment number 12.

34. How many NPF event and non-event days were analyzed for this plot?

Response:
The following sentence has been added to the caption of Figure 8:
"Data includes 5 and 51 nucleation and non-nucleation days."

35. Did all considered days show comparable meteorological conditions (e.g. wind direction & solar irradiance)?

Response:
The following sentence has been added (page 9, line 25):
"All non-nucleation days did not have exactly the same meteorological conditions but the effects of their variation on f_{57} were minimal."

36. Are only workdays considered, with typical rush hours, or does the figure also include weekends?

Response:
The following sentence has been added to the caption of Figure 8:

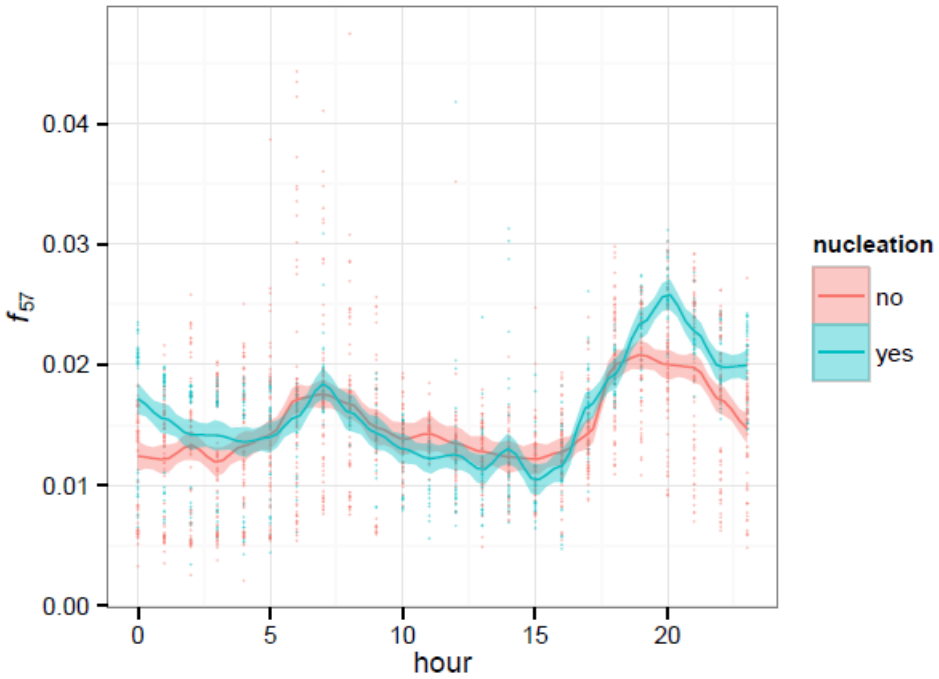
"The data includes weekdays and weekend as no exclusions were applied to increase the statistical power."

37. Did the authors apply the f44 vs. f43 plot also for measurement site S25? Are the patterns comparable at S25 and S12?

Response:
Please see the response to the comment number 2.

38. Page 27971, Figure 9: Why is the unit on the y-axis denoted in hours? According to my understanding f57 should have no unit as it is the ratio of m/z 57 divided by the total c-TOF-AMS signal. For better comparison, both y-axis in the figure should be scaled equally.

Response:
Figure 9 (which is now Figure 7) has been replotted as follows:



Technical comments

39. Page 27946, line 26: Please add "e.g." in front of the references.

Response:
It has been added.

40. Page 27947, line 13: Please delete "been".

Response:
Done.

41. Page 27947, line 17: Please replace "process" with "NPF".

Response:
Done.

42. Page 27948, line 10: "UPTECH" is mentioned twice in the same line, please delete one.

Response:
Done.

43. Page 27949, line 8: Please remove the brackets of "(Crilley et al., 2013)".

Response:
Done.

44. Page 27953, line 26: Please replace "reaching" with "reached"

Response:
Done.

References

Winkler, P. M., Ortega, J., Karl, T., Cappellin, L., Friedli, H. R., Barsanti, K., McMurry, P. H., and Smith, J. N.: Identification of biogenic compounds responsible for size-dependent nanoparticle growth, Geophys. Res. Lett., 39, L20815, 2012.

Reviewer#3:

This manuscript presents measurements of particle size distributions and chemical composition in Brisbane, Australia, with a special focus on new particle formation (NPF) events. The authors conclude that the organic markers f43 and f44 behave differently when organics are produced from NPF and when it is traffic-produced. The paper is mostly well-written, and does address an important scientific question. However, I do find several major shortcomings that need to be addressed before this manuscript can be considered for publication in ACP.

Major comments

Statistics:

1. One of the main problems with this manuscript is the statistics. In part because the analysis is based on only 5 NPF days, but even more so because at no point do the authors acknowledge this limitation of the analysis. An effect that is found based on the analysis of three specific days is taken as a general truth without further discussion, and stated in the abstract that "this finding can be used as a tool for source apportionment".

Response:
We agree with the reviewer and have added the following sentence (page 10, line 15):

"However, it should be noted that the analysis were based on only five NPF events and the results may not be a general rule"

We have also replaced "this finding can be used as a tool for source apportionment" with "this finding can be potentially used as a tool for source apportionment" (page 2, line 3)

2. More discussion is needed on the applicability of the findings in this manuscript. Related to the comment of the use for source apportionment based on f43 and f44, to arrive to this conclusion the authors have already used f57 as an accepted marker for traffic-produced aerosol. What is then the use, or additional value, of the f43 and f44 markers?

Response:
Further analysis and discussion has been applied on the f43 and f44 data. Please see the response to comment number 12 for details.

3. How many non-NPF days are included in the analysis from S12 and S25?

The following sentence has been added (page 6, line 23):

"PNSD data was available for 6, 18, 11, 13, and 8 days in s1, s4, s11, s12, and s25 respectively."

4. What are the shaded areas in Figures 1 and 2? Certainly there has to have been more variability over these periods. This needs to be shown in the figures.

Response:
The following sentence has been added to the captions of Figures 1 and 2: "Shaded areas represent the 95% confidence interval in the mean."

The mean and 95% confidence intervals in the mean were calculated using the bootstrap simulations through "timeVariation" function in "openair" package. However, further investigation showed that the LOESS smoothing approach better represent the data. Therefore, Figures 1 and 2 have been updated and they now also include all the data, as well as the smoothed trend illustrating the overall trend as well as its variability (Please see below).

Figure 1:

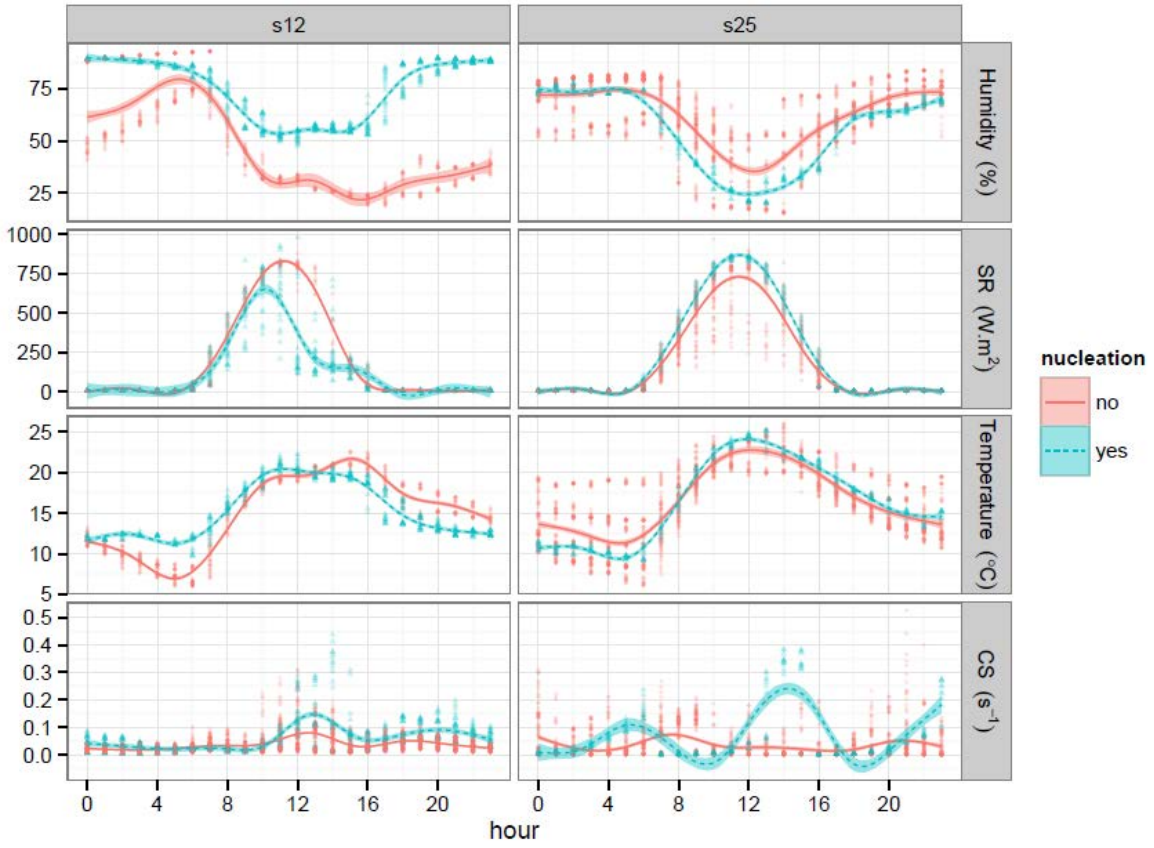
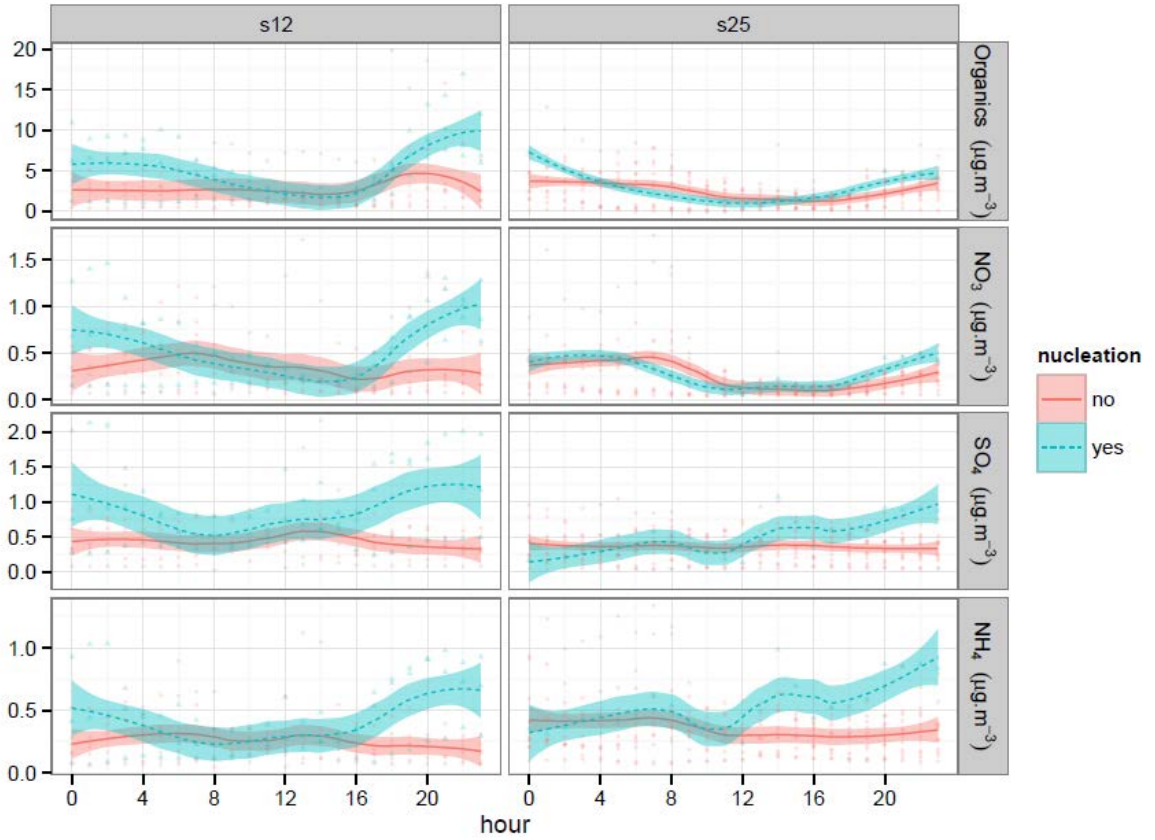


Figure 2:



5. If Cheung et al (2011) found 65 NPF events per year in Brisbane, why did the authors now only detect 5 events over the course of 10-15 weeks (five 2-3 week deployments, as I understood from the text, although it was a bit unclear in the formulation)? Such variability needs to be discussed.

Response:
The measurement campaign was performed for about 11 weeks but the data did not cover the whole period due to instrument malfunction.

The following sentence has been added (page 6, line23):

"PNSD data was available for 6, 18, 11, 13, and 8 days in s1, s4, s11, s12, and s25 respectively."

The frequency of observed nucleation events are still less than the ones found in Cheung et al (2011) which can be due to the temporal/spatial trend of these events as this project covered different location and time of the year.

6. What triggers NPF? Certainly there has to be more meteorological data available that would help in understanding chemistry of NPF than what is presented here. These might have been discussed in Cheung et al, but also closely relate to the results in this manuscript, and therefore need to be discussed here as well.

Response:
The meteorological conditions inducing the NPF events were not the focus of this study and therefore were not covered in the manuscript.

Condensation sink:

7. The way the condensation sink (CS) is discussed and used in this paper, raises the question whether the authors analyzed and interpreted their data correctly. P27949, L23-24: "The surface area of aerosol particles that is available for condensation can be measured using condensation sink (CS)." This is a very awkward statement, and certainly the surface area is not "measured using CS". CS can be calculated from the measured size distribution.

Response:
We agree with the reviewer's comment and replaced that sentence with (page 4, line 18):
"The surface area of aerosol particles available for condensation can be estimated by the condensation sink (CS) parameter which is calculated from the measured PNSD."

8. If the formula for the calculation is given explicitly, the variables also have to be defined in the text. In order to show that the equation has been used correctly, some discussion should be included e.g. on the value of the mass accommodation coefficient used.

Response:
The following discussion related to the calculation of CS has been added to pages 4 and 5:

"where D is the diffusion coefficient, d_p is the particle diameter, N_i is the concentration of particles and β_M can be expressed as (Soo et al., 1971):

$$1) \quad \beta_M = \frac{Kn+1}{0.377Kn+1+\frac{4}{3}\alpha^{-1}Kn^2+\frac{4}{3}\alpha^{-1}Kn}$$

where α is the sticking coefficient and is assumed to be 1 (Clement et al., 1996) and Kn is the Knudsen number, which is equal to $\frac{2\lambda_v}{d_p}$. The mean free path (λ_v) is a function of pressure and temperature and can be calculated using the following formula (Willeke, 1976):

$$2) \quad \lambda_v = \lambda_o\left(\frac{101}{p}\right)\left(\frac{T}{296.2}\right)\left(\frac{\left(1+\frac{110}{296.2}\right)}{\left(1+\frac{110}{T}\right)}\right)$$

where P is in kPa and T in K. λ_o = 0.039 µm, which is the mean free path of H_2SO_4 at standard conditions (Bae et al., 2010). CS's were calculated using the above mentioned procedure on the PNSD data."

9. Fig. 1: The absolute values of the CS in these plots are extremely high. While I am not aware of the specific aerosol sources at this location, I still expect that these numbers are erroneous as they are presumably calculated from the PSD of particles smaller than 400nm and get values orders of magnitude higher than I have ever seen. This also raises the question about the huge variability in the values, especially at S25. While I believe the numbers are erroneous, the interpretation based on the CS plots, that it is lower a few hours before NPF onset in accordance with previous work is also highly questionable. The CS is all over the place, and is on average higher during most of the NPF days, which certainly is not in agreement with typical findings. These data need to be thoroughly checked, and if the authors still seem to find such high CS and CS variability, the calculations (and reasons) need to presented in great detail to be convincing.

Response:
Thanks for noticing the issue related to the CS's high values. The calculations were double checked and an error was found in the calculation of the condensation sink. The error was related to the normalisation of the SMPS data and only affected the absolute values and did not affect the trends, therefore, interpretation which were based on the trends of the CS's remain unchanged.

The CS's were found to be lower a few hours before the start of NPF events and increased rapidly after the start of NPF events which is expected and aligned with the previous findings.

AMS data:

10. For anyone familiar with AMS data, it is obvious from Figures 3-5 that the data has not been analyzed correctly with regard to ammonium. Ammonium is typically present as ammonium sulfate or ammonium nitrate, and in both salts the ammonium makes up less than a third of the total mass.
    In Fig. 5 the authors propose that ammonium makes up 2-3 times more mass than nitrate+sulfate in the 50-100 nm range, which would be a shocking finding in itself. However, when looking at the size distribution (or in fact lack thereof) in Fig. 3-4, it becomes evident that this must be an error in data processing.
    The authors need to improve their knowledge on how to correct the ammonium signal in the AMS and replot and then reinterpret all these figures.
    In addition, if the ammonium was so badly off, it does raise the question how well for example the organics have been evaluated, as there exists many pitfalls there as well. Perhaps this only relates to the size dist. data of the AMS, as the NH4 values in Fig. 2 seem more reasonable.

Response:
Thanks for noticing the issue with the analysis of the size distribution data of the AMS. We investigated the issue and found that the AMS particle size data were not reliable, therefore, we replaced our analysis with the total chemical mass data which we are confident in. The related Figures (3&4) have been replotted and Figure 5 has been included in Figures 3&4. Updated Please see response to the reviewer2's 27th comment for Figure 3& 4:

11. The PSD should also be converted to volume in order to be able to compare properly to the AMS data size distributions. I recommend the authors compare to Fig. 3 in Zhang et al (2004), and use this as a goal for their own plots.

Response:
Particle volume distribution has been calculated and plotted in Figures 3 and 7. We also tried to follow the Figure recommended by the reviewer: please see the updated Figures in the response to the reviewer2's 27th comment.

Other comments

12. P27955, L1-5: With such a short description, it is very hard to understand what was clustered how, and what the real outcome was. The authors need to expand on this, possibly including a figure.

Response:
We have modified the clustering method we used and have also discussed it in further details. The updated methodology, discussion and figures are as follows:

(Page 6, line 12)
"K-means clustering: This clustering algorithm minimises the within-cluster sum of squares in order to divide M observations with N dimensions into K clusters (Hartigan and Wong, 1979). A common method of choosing the number of clusters is to compare visually a measure of error, such as sum of squared error (SSE), with sequential number of clusters by plotting the measure of error on y axis and the number of clusters on x axis (Everitt et al., 2010). Optimum number of clusters is the point at which the measure of error flattens and form an elbow."

(Page 9, line 7)
"In order to assess the possible clustering of f44 vs. f43 for nucleation days compared to non-nucleation days, K-means clustering technique was applied on the f43 and f44 data. A period between 3-5 pm was selected for this purpose as this was the initial stage where the newly formed particles grew enough to have significant contribution to the total signal. In order to find the optimum number of clusters, SSE was plotted against the sequential number of clusters and five number of clusters was found to be appropriate as it was located at the elbow in the plot (Figure6).

The five identified clusters as well as their 95% confidence ellipse are illustrated in Figure 7.a, 93% of the data measured in nucleation days were in clusters 3-5 (54%,15%, and 24% in clusters3,4,and 5 respectively) while clusters1 and 2 contained 77% of the data measured in non-nucleation days (Figure 7). In addition, cluster 1 and 5 contained less than 1% of nucleation and non-nucleation days respectively. These show a distinct clustering on f44 vs. f43 for nucleation days compared to non-nucleation days and indicating a potential application of f44 vs. f43 plot for identification of the newly formed particles."

Figure 7:

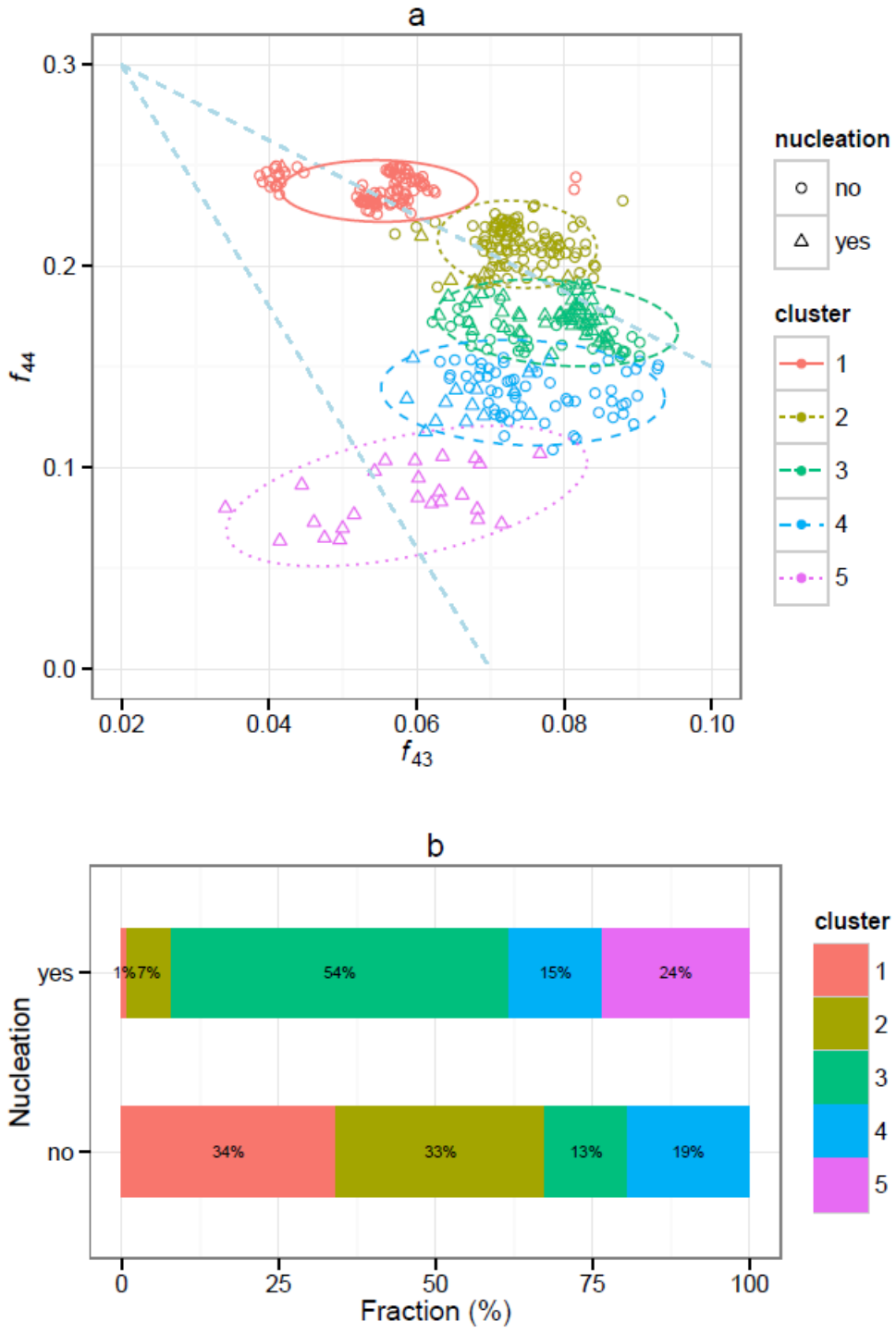
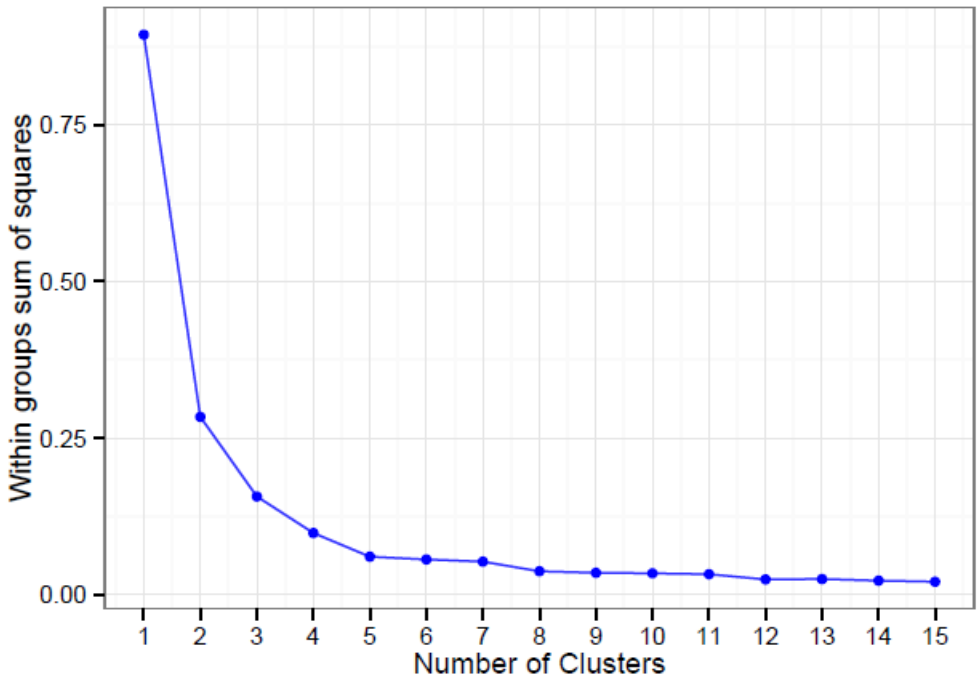

a
0.3
0.2
0.1
0.0
f_{44}
0.02
0.04
0.06
0.08
0.10
f_{43}
nucleation
no
yes
cluster
1
2
3
4
5
b
yes
no
Nucleation
1%
7%
54%
15%
24%
34%
33%
13%
19%
0
25
50
75
100
Fraction (%)
cluster
1
2
3
4
5

Figure 6:


Within groups sum of squares
0.00
0.25
0.50
0.75
1
2
3
4
5
6
7
8
9
10
11
12
13
14
15
Number of Clusters

13. P27955, L18-20: While the behavior of f57 convincingly shows that traffic related aerosol seemed to stay constant between NPF and non-NPF days, it certainly cannot "explain the pattern observed in f44 vs f43".

Response:
We agree with the reviewer and have removed that sentence

Specific comments

14. Look over use of parentheses in references. At least P27948 L23, P27949 L9 and L22.

Response:
Fixed

15. P27950, L23: What does the term "component mass spectrum" mean? Why not just say total organic signal? Also, I would not say that f44 and f43 "can characterize" the degree of oxidation. Please reformulate.

Response:
"component mass spectrum" and "can characterise" have been replaced with "total organic signal" and "indicate" respectively.

16. P27951, L6-11: I would like to see a statement on why the authors used this model. Both in general, why fitting was needed, and why specifically this model.

Response:
Please see the response to reviewer2's 10th comment.

17. P27953, L13-14. What does "similar" refer to here?

Response:
Please see the response to reviewer2's comment number 15.

18. P27953, L19-20. "percent"

Response:
Fixed.

Additional modifications:

1. *We have changed the title slightly to:*
   "Investigation into the chemistry of new particle formation and growth in a subtropical urban environment"

2. *The introduction has been modified to include a recent study (page 3, line 2):*

"Leigh et al (2014) found ammonium and sulphate as the dominant species on particle formation days in a short term study based on measurements at a fixed site. More studies need to be carried out, particularly in less investogated areas, using direct measurements techniques, and including comprehensive and advanced analysis, in order to determine the species involved in NPF events and the nature of their contribution."

3. *For consistency, the names of the sites (s1, s4, s11, s12, and s25) have been written in lower case throughout the paper.*

4. *The following sentence in the section 2.2 (Instrumentation) has been removed as the issue with the AMS particle data still exist.*
   "The issue related to particle size mentioned in Crilley et al. (2013) has been fixed for this paper as it was a calculation error."
5. *The "Abstract" and "Summary and Conclusion" have been modified aligned with the changes throughout the manuscript:*

Abstract:

"The role of different chemical compounds, particularly organics, involved in the new particle formation (NPF) and its consequent growth are not fully understood. Therefore, this study was conducted to investigate the chemistry of aerosol particles during NPF events in an urban subtropical environment. Aerosol chemical composition was measured along with particle number size distribution (PNSD) and several other air quality parameters at five sites across an urban subtropical environment. An Aerodyne compact Time-of-Flight Aerosol Mass Spectrometer (c-TOF-AMS) and a TSI Scanning Mobility Particle Sizer (SMPS) measured aerosol chemical composition (particles above 50nm in vacuum aerodynamic diameter) and PNSD (particles within 9-414 nm in mobility diameter), respectively. Five NPF events, with growth rates in the range 3.3-4.6 nm, were detected at two of the sites. The NPF events happened on relatively warmer days with lower condensation sink (CS). Temporal percent fractions of organics increased after the particles grew enough to have a significant contribution to particles volume, while the mass fraction of ammonium and sulphate decreased. This uncovered the important role of organics in the growth of newly formed particles. Three organic markers, factors f43, f44 and f57, were calculated and the f44 vs f43 trends were compared between nucleation and non-nucleation days. K-means cluster analysis was performed on f44 vs f43 data and it was found that they follow different patterns on nucleation days compared to non-nucleation days, whereby f43 decreased for vehicle emission generated particles, while both f44 and f43 decreased for NPF generated particles. It was found for the first time that vehicle generated and newly formed particles cluster in different locations on f44 vs f43 plot and this finding can be potentially used as a tool for source apportionment of measured particles."

Summary and Conclusions

"In summary, PNSD, chemical composition and meteorological parameters were measured at five sites across the Brisbane Metropolitan Area. Five NPF events, with growth rates ranging from 3.3-4.6 nm.hr-1, were observed at two of the five sites, and the NPF events happened on days with lower CS and higher temperature than non-event days. Higher sulphate, nitrate, ammonium and organics were observed on nucleation days compared with days when no nucleation was observed. Percent fractions of nitrate, sulphate, ammonium and organic chemical species were calculated and their diurnal trends were modelled using the LOESS. Ammonium, sulphate and nitrate mass fractions increased before the newly formed particles grew enough to have a significant contribution to the particles volume, peaked around that time and decreased after that. Conversely, the organics percent fraction increased significantly after the contribution of new particles to total volume, indicating the important role of organics in the growth phase of NPF events. f_{44} and f_{43} were analysed to investigate the role of organics more in depth, as the f_{44} vs f_{43} would reveal information regarding the level of oxidation and volatility of OOA. f_{43} and f_{44} both decreased after the start of nucleation, while f_{44} decreased only in case of particles generated from vehicle emission. K-means clustering analysis revealed that the aerosol particles generated by vehicle emissions and NPF events clustered in different locations on the f_{44} vs f_{43} plot. This determined the potential application of the f44 vs f43 plot for identification of the source/s and transformation of the OOA components as they clearly followed different patterns. However, it should be noted that the analysis were based on only five NPF events and the results may not be a general rule."

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