Anonymous Referee #2
Received and published: 15 October 2015
General comments The manuscript by Budisulistiorini et al. presents new and interesting data from two years of measurements using ACSM at an urban and a rural site in south-eastern USA. This is a very large dataset and the analysis presents important and new findings. Unfortunately the manuscript needs considerable editing, before submission should be considered. There are numerous spelling errors and grammatical errors, which must be corrected. I have identified some, but it is not the task of the reviewers to correct spelling and standard grammatical errors. My only other major point is that the authors should remember the limitations of the study, namely that the measurements were performed during two different years. This should be written much more clearly in the discussion of the data. Generally the figures look very good, but they are too small and should be divided into more separate figures to enhance readability. Please describe and use a uniform description of the r2 values (high/low/moderate/low).

Specific comments.
Page 22383 line 4: Please include a reference to the statements of sources to sulfate, nitrate and OA.

We removed the statement and re-phrased the paragraph.

Page 22383 line 9-10: Please add a reference to the discussion of POA/SOA variation during the day.

We revised the statement and add reference.
"Contribution of hydrocarbon-like OA (HOA) associated with POA to urban OA mass may be significant during morning traffic, while oxygenated OA (OOA) associated with SOA exceeds POA at midday or in the afternoon (Zhang et al., 2005)."

Page 22383 lines 13-16: Please add a reference here.

We removed the statement and re-phrased the paragraph.

Page 22383 line 29: Anthropogenic sources might be low in rural areas, but they are not "absent".

We revised it to "low".

Page 22384 Lines 7-8: Please clarify this sentence.
What is the distance between the sites?

The current and previous studies were located at the same site. We revised the sentence as follows.
"Moreover, OC at LRK is the primary component of SOA in summer, while POA from wood burning can contribute significantly during fall [1]."

Page 22384 Line 16: collocated -> co-located.

Both words are appropriate according to Oxford dictionary. We will keep "collocated".

Page 22385 Line 23: Please state how often the calibration procedure was done.

We added the number of calibrations performed. The sentence has been revised as follow:

"The ACSM was tuned for ionizer and electronic offset and calibrated for ionization efficiency on site (5 – 7 times) throughout each year of sampling at each site."

Page 22387: The results and discussion should start with a somewhat broader presentation to introduce the data, including some average concentrations.

We separated the results and discussion into different section. We re-organized the paragraphs so that there is a general introduction to the results section.

Page 22388 line 11: Please correct language here. How close are the coal-fired power plants? Given the time-scale for sulfate formation, would you expect them to contribute to sulfate at your sites?

The closest coal-fired power plant is ~7 km from JST and $\geq$100 km from LRK. These power plants contributed to regional sulfate variability in the southeast region, and thus, sulfate concentration measured at the site.

We have revised the sentence and paragraphs as follow:
"Average OA contributions to NR-PM_1 were higher in spring and summer at JST and LRK, suggesting that biogenic SOA plays significant role during these periods. OA characterization is further discussed in section 3.2."

"Average sulfate concentrations were highest in summer for LRK (2.1 μg m^{-3}) and fall for JST (~2 μg m^{-3}) as illustrated in Fig. 1. The enhanced sulfate concentrations coincided with increased OA concentration, which is consistent with prior studies that sulfate may contribute to enhanced SOA formation (Lin et al., 2013a, Xu et al., 2015, Budisulistiorini et al., 2015). Changes of sulfate concentrations at LRK are likely affected by changes in SO_2 emissions from electrical generating units in the region (Tanner et al., 2015). At JST, SO_2 emissions from coal-fired power plants nearby Atlanta contributed to the spatial variability of sulfate concentrations (Peltier et al., 2007). …"

Page 22389: in accord with -> in accordance with? The word "correlation" should only be used if you actually calculated the correlation of the data. did you do that? Which technique did Guo et al. use – please state.

"in accord with" and "in accordance with" are both appropriate English phrase based on Oxford dictionary. We keep "in accord with".

We did calculate correlation between aerosol pH and organic. The sentence has been revised to include correlation value:

"No direct correlation (r^2 <0.1) was observed between aerosol pH and OA at both sites."

Guo et al. (2015) used CCN measurements to calculate organic hygroscopic parameter, which was then used to calculate organic water. The sentence has been revised as follows:

"It should be noted that the possible LWC contributions from OA are not included because cloud condensation nuclei (CCN) measurements, such as that recently used by Guo et al. (2015) to

measure this quantity, are lacking at our sites."

Page 22390
Line 14: Specify "during fall and winter". Is this only referring to JST site?

The original text referred to winter season only and comparing trend at night and day.
We clarified the text into:

"The BBOA factor concentration increased during the night and decreased during the day at both sites (Fig. 8),…"

Line 20-21: r^2 = 0.2-0.5 is not moderate, but low.

It has been revised.
"The time series of BBOA showed low to moderate correlation (r^2 0.4–0.5 at JST and r^2 0.2 – 0.4 at LRK; Tables S2–S3) with BC, suggesting that it is likely influenced by some local sources (e.g., fires)."

Page 22391
line 19: "most abundant component of OA"

It has been revised as follows:
"SV-OOA, which was observed only in urban OA, is the most abundant component of OA."

Lines 25-: Please correct and clarify this sentence.

We rephrased the sentence as follows:

"Since isoprene emission is expected to be negligible during winter season, SV-OOA might not relate to IEPOX-derived SOA."

Page 22393 lines 10-12: This sentence needs further editing to be clear.

We rephrased the sentences as follow:

"The average IEPOX-OA concentration was slightly higher at LRK than at JST, which is expected due to abundant emissions of isoprene at the forested site."

Page 22394 line 2: r^2 of 0.2 is very weak, approaching non-existent.

It has been revised as follows:

"The time series of LV-OOA was weakly correlated with sulfate (r^2 ~0.2) at JST, but more strongly correlated with sulfate at LRK (r^2 = 0.6–0.7) (Table S3)."

Page 22394 lines 20-21: What do you mean here?

We rephrased the sentences as follow:

"Average concentration of IEPOX-OA at JST and LRK increased during summer. At LRK, the average concentration of IEPOX-OA reached a maximum in summer, but its relative contribution

to total OA mass was lower due to the increasing concentration of 91Fac."

Page 22395 line 4: Please clarify/use correct language.

We rephrased the sentence as follow:

"At JST, PMF analysis of fall OA resulted in a four-factor solution (i.e., HOA, BBOA, SV-OOA, and LV-OOA), while at LRK a three-factor solution was resolved (i.e., LV-OOA, 91Fac, and IEPOX-OA)."

Page 22396: Avoid repeating the results presented in the very long section above.
Furthermore the discussion should reflect the fact that the data were collected in two different years.

We re-organized the results and discussion and added information about measurement period for the two sites.

Page 22397
line 13: The argument about "loss of foliage as a major source of isoprene emissions" is unclear.

We rephrased the argument as follows:

"Decrease of IEPOX-OA concentration in fall season is consistent with loss of tree foliage as a major source of isoprene emission. Additionally, IEPOX-OA was not observed during winter suggesting that isoprene emission was negligible."

Line 23: What are "sources datasets"?

The "sources datasets" refers to OA mass spectra datasets from field and experiment measurements. We revised the sentence as follows:

"The IEPOX-OA component has been observed in chamber experiments and field OA (Hu et al., 2015)."

Page 22398
lines 18- . Please remove and replace "we offer" with eg. "the study presents".

It has been revised.

"Although parameterizations of IEPOX-OA factor based on its markers (Hu et al., 2015) was not done in this study, this study presents some insights of role of the *m/z* 82 fragment ion over different seasons in southeastern U.S. "

line 22: identification -> detection.

It has been revised.

"Observation of the *m/z* 75 fragment ion variation over different seasons indicates its potential as an additional marker ion for IEPOX-OA detection."

Page 22399 lines 9-14: This is very speculative, given the quite similar correlations.

We rephrased the argument as follow:
"SOA tracers of isoprene ozonolysis were moderately correlated (r^2 = 0.2–0.5) with both IEPOX-OA and 91Fac, adding to fair correlation between 91Fac component and monoterpene SOA tracers reported in Budisulistiorini et al. (2015) . "

Figure 6 does not provide enough information to include it in the main manuscript. Please move to SI or leave out.

We moved Fig. 6 to SI.