Response to Referee #1

We thank the referee for comments that have improved this paper. We believe that we have addressed all the concerns.

Comment:
I do not agree with some of the discussions on air mass processing related to lake breeze circulation. (OOA)/deltaCO and SO4/(SO2+SO4) ratios were used as indicators of the extend of air mass chemical processing. The authors observed higher (OOA)/deltaCO and SO4/(SO2+SO4) ratios in air masses sampled during flights compared to the estimated ratios in regional background and subsequently suggested that lake-breeze circulations are an important dynamic in the formation of SO4 and SOA. However, the differences in these ratios may simply reflect differences in SO2 and VOC concentrations between air masses.

Response:
We agree that precursor concentrations may be different in the different air masses which are reflected in the ratio differences. However, we focus on the connection of airmasses across the LSC lake-breeze front (CBL (flight 4) and AF (flight 5) air masses) where we have data to show evidence of processing-related enhancements. Based on our detailed analyses (observations, modelling, trajectories), we interpret these differences to be due to oxidation processes.

Comment:
In this manuscript the formation rate of SO4 is reported in the unit of % per hour, defined as the increase rate of SO4 normalized by total sulfur (= SO4 + SO2). Assuming no depositional loss of sulfur, the total sulfur content in an air mass should conserve and the formation rate of SO4 should be proportional to SO2 concentration. As SO2 reacts away in more aged air mass, the production rate of SO4 should decrease. So the low SO4 production rate estimated in regional background may simply be due to low SO2 concentrations in such air masses.

Response:
We are reporting the SO_4^{-2} production as a percentage (so it is relative to the total amount of sulphur), and since the percentage is very low, we are effectively comparing the relative instantaneous formation rate of SO_4^{2-}. Under this situation the absolute amount of SO_2 is not a factor. Consider the following hypothetical reaction

SO_2 + oxidant → SO_4^{2-} k

We have

$$\frac{d[SO_4]}{dt} = k[oxidant][SO_2]$$

The relative instantaneous SO_4^{2-} formation rate would be

$$\frac{1}{[SO_2]}\frac{d[SO_4]}{dt} = k[oxidant]$$

And this is dependent on the oxidant level.

Comment:
Similarly, the lower OOA production rate in regional background air mass may be because lower concentrations of VOC precursors in regional background than in polluted air masses.

Response:
A similar argument can be made for the OA formation as for SO4 formation. Since, in this case we do not have the precursor VOC concentration to compare to, we use CO as the tracer for the VOCs.

Comment:
The unit for OOA production rates should be explained. I could not figure out how the OOA production rates (200 ug m-3 ppmv-1 %-1 for flight 5 and 80 ug m-3 ppmv-1 %-1 for flight 4) were calculated. Were they determined based on the slopes in Fig. 12 and % indicates changes in SO4/(SO2+SO4)? If so, shouldn't the slope for the Flight 4 data points be ~1 ug m-3 ppmv-1 %-1?

Response:
Agreed that the units are confusing and we believe this is because in Fig. 12 the x-axis should have been expressed as a percentage (as described in the text) rather than a fraction. The units of the slope are then in ug m^{-3} $ppmv^{-1}$ $\%^{-1}$. This change also addresses the same comment by referee #2. In the text, Section 3.7, lines 576-579, the description of Fig 12 has been modified to be more clear, statement now reads: "Figure 12 shows that the relationship between OA/ΔCO and $SO_4^{2-}/(SO_2+SO_4^{2-})$ for Flight 5 is confined to a narrower range of OA production (~2.25 µg m^{-3} $ppmv^{-1}$ $\%^{-1}$), whereas in Flight 4, a range of OA production (relative to SO_4^{2-} production) extending from 1.2-1.75 µg m^{-3} $ppmv^{-1}$ $\%^{-1}$ is observed with a range of OA production (relative to SO_4^{2-} production) extending from 1.2-1.75 µg m^{-3} $ppmv^{-1}$ $\%^{-1}$." The numbers have changed because we changed OOA to OA (based upon a later comment made by referee #1) and a conversion to percentage from fraction on the x-axis.

Comment:
Nevertheless, it is not clear to me the physical bases of assuming that (OOA)/deltaCO should change linearly with SO4/(SO2+SO4) ratios?

Response:
We are not assuming that a linear relationship must exist between these two ratios. We hypothesized that there could be a relationship between these two ratios based on the fact that they are both closely coupled to oxidant chemistry in air. Fig. 12 for flight 4 data shows us that there is a relationship between these two terms and we fit a linear function to it. We then use this relationship to back calculate what the OA/ΔCO would be.

Comment:
It is also a question of determining the OOA formation rate for regional background air by multiplying the slope in Fig. 12 (Flight 4, m= 71.39 ugm-3 ppmv-1 fraction) by the regional background SO4 formation rates taken from Fig. 13 (1–2% h-1). For the least, didn't such slopes vary between air masses, as show in Fig. 14?

Response:
In Fig. 12 there appears to be a linear relationship between the two ratios including a number of different air mass types in flight 4 and a linear regression is therefore applied. This is the slope that is taken to represent a number of different air mass types. If by slope the referee is actually referring to the OA/ΔCO ratios, the ratios did vary between air masses. We focus on a comparison of ratios in the CBL (flight 4) and AF (flight 5) by presenting a linkage between air masses via the LSC circulation.

Comment:
In addition, it was said in the texts that the studies were all carried out under clear sky conditions, so what are the bases for speculating that cloud processes are responsible for enhancement of OOA and sulfate production? Overall, the linkage of the enhancements of OOA and sulfate formation rates to the presence of cumulus clouds associated with the lake-breeze fronts seems weak.

Response:
Agreed, this sentence referring to 'clear sky conditions' (Section 2.1) is misleading. The aircraft flew under visual flight rules and thus would adjust the flight path so as avoid flying through thick cloud. Clouds associated with lake-breeze fronts were present on most days during the study. The statement in Section 2.1, lines 91-92, referring to clear air measurements was removed and replaced with "Slight adjustments were made to the flight paths to avoid flying through thick cloud."

Additional description linking enhancement of OA and sulphate formation rates to clouds is provided in Section 3.7, lines 647-660.

Comment:
Abstract, it is said that the formation rate for OOA was found to be 2.5-6.2 ug m-3 ppmv-1 hr-1. This unit is confusing and should be clarified.

Response:
We realize this is a long unit expression, so the sentence, lines 30-31, has been rewritten to try to make it more clear as: "The formation rate for OA (relative to excess CO in ppmv) was found to be 2.2-9.1 ug m^{-3} $ppmv^{-1}$ hr^{-1} and the SO_4^{2-} formation rate was 1.0-4.6 % hr^{-1}."

Comment:
Page 11502, line 14- 15, how do the D.L. determined in this study compare to the values reported from other AMS studies?

Response:
Added the following sentence Section 2.3, lines 127-128 "These detection limits are within the range of AMS detection limits reported (e.g. DeCarlo et al., 2006)."

Comment:
Page 11503, line 5-7, give citation(s) for the discussions on various causes for AMS CE not being 1.

Response:
Added references into the following sentence, Section 2.3, lines 149-153 “The CE is a function of the particle transmission through the aerodynamic lens (Jayne et al., 2000, Liu et al., 2007), the efficiency of particles being focussed by the lens and directed onto the vaporizer (Quinn et al., 2006; Salcedo et al., 2007; Huffman et al., 2005; Jayne et al., 2000), and the extent to which particles bounce off the vaporizer (Matthew et al, 2008; Huffman et al., 2005).”

Comment:
In the 2nd paragraph of “3 Results and Discussion”, in addition to referencing to Sills et al., 2011, it seems useful to reiterate some details on the identification of lake breeze fronts and give some details on the spatial and vertical changes in pollutant concentrations.

Response:
Added two sentences to provide more details on the lake breeze identification. Section 3, lines 205-207, “The identification and classification of lake breezes in this study was based on meteorological data collected from several different sources including surface measurements, satellite observations of clouds, and radar data.”

Section 3, lines 222-224, “The light gradient flow on this day was conducive to the formation of lake-breeze fronts around LSC and along the edge of LE, and thus, classified as a LD (or a ‘classic’) circulation.”

It is unclear as to what is being requested with regard to providing details on spatial and vertical changes in pollutant concentrations – these details are presented in Section 3.3 and discussed in the following sections. The first two paragraphs just provided a general lead-in prior to discussing the details on pollutant variability on the day of interest. No changes made.

Comment:
Page 11509, line 11, extra “in”?

Response:
This was a typo, changed ‘in’ to ‘an’.

Comment:
Page 11514, line 20, extra “being”?

Response:
Removed ‘being’ page 11514, line 23.

Comment:
Section 3.7, it seems that OOA was used as a proxy for SOA and HOA for POA, but the associations were not directly stated or referenced in this work.

Response:
OA is now used as a proxy for SOA and is stated in the first paragraph of section 3.7. The beginning of the first paragraph in Section 3.7 has been rearranged so that it begins with an explanation of the use of OA/ΔCO (and $SO_4^{2-}/(SO_2+SO_4^{2-})$) and then presents

Fig. 12. The appropriate references that demonstrate the use of OA as a proxy for SOA are listed in first sentence. We also added two references for the use of SO_4^{2-}/($SO2+SO_4^{2-}$) to indicate extent of SO_2 oxidation.

Comment:
Page11518, line 26 – 29, all three references uses increase in the OA/deltaCO ratio, not OOA/deltaCO ratio to indicate SOA production.

Response:
We decided to use OA instead of OOA to use as a proxy for SOA. We believe this may be a better proxy than OOA so as not to have a preconceived notion of the composition of OOA (OOA is determined based on the ion intensity at m/z 44). Also, this allows a more direct comparison with recent literature that uses OA. Fig. 12 and 14 were modified to show OA instead of OOA. The text in section 3.7 has been changed from OOA to OA and the sentence describing and referencing the deconvolution of OA into OOA and HOA has been removed. Rechecked the references and made the following changes to reflect OA/ΔCO work. Added the DeCarlo et al., 2010 reference. Removed de Gouw et al., 2005 reference. Changed Kleinman et al., 2009 reference to Kleinman et al., 2008 and removed Kleinman et al., 2009 reference from Reference list.

Comment:
Figure 7, please show latitude and longitude scales.

Response:
Selected latitudes/longitudes are overlaid on Figure 7.

Comment:
Figure 10, please explain all the lines in the figure.

Response:
The lines are all now described in the figure caption of Fig. 10.

Comment:
Figure 12, what's b, what's m, what are the error bars corresponding to?

Response:
On Fig. 12 slope and intercept are written out explicitly. In the Figure caption, the following text is added: "The error bars are the standard deviations for each point."

Comment:
Page 11519 line 20 and Figure 12, there are 4 significant figures in the b and m values. Are the third and fourth digits of these values really statistically significant?

Response:
In Fig. 12, and in the text, the slope and intercept values are adjusted to only display statistically significant values.

Comment:
It is said in the text that the extent to which lake-breeze circulations were deformed by the synoptic wind is classified as low, moderate and high deformations. It will be useful to give a bit explanation on the significance of this classification.

Response:
Section 3, lines 209-215, added the following text to provide a bit more description/significance of the lake-breeze classifications: “LD lake-breeze circulations are most similar to the classic textbook depiction, with the light synoptic wind regime allowing lake-breeze fronts to develop around the perimeter of the lake and air parcels to move along quasi-closed circuits. On the opposite end of the spectrum are HD lake-breeze circulations in strong synoptic wind regimes where lake-breeze fronts form only along segments of shoreline quasi-parallel to the synoptic wind and the influence of the lake-breeze circulation on the movement of air parcels is masked.”