

## Response to Referee #2

We thank the referee for insightful and useful comments and we believe that we have addressed all of the concerns.

### Comment:

p. 11514, lines 1-4: The key hypothesis of the paper is that "pollutants in the CBL were lofted upward at the LSC lake-breeze front, transported northward in the synoptic flow, transported in the downdraft on the north side [of] the front, and then confined by the LSC onshore flow along the south shore of LSC." This is possible, but the more likely explanation should be considered first: that pollutants behind the LSC lake-breeze front were lofted upward at the front, transported northward, transported in the downdraft, and confined along the south shore of the LSC. This is a known circulation pattern, while the trajectory described in the hypothesis has not previously been documented anywhere. Further, the paper does not provide convincing evidence of this. The trajectory analyses show recirculation, but the recirculation seems to be consistent with my "more likely explanation". Nowhere in the trajectory analysis is it shown that air initially landward of the lake breeze front actually becomes ingested in the lake breeze air. The two air masses will not be completely separate, because mixing does take place along and behind the lake breeze front. Note that the air behind the lake breeze front can be dirty because polluted air can be transported over LSC during the early morning and post-dawn hours prior to the development of the lake breeze, and concentrations will be relatively high because the air will not be mixed within a CBL while the air resides over LSC during the morning. In the absence of stronger evidence, this portion of the analysis (including a large part of section 3.7) should be removed.

### Response:

As it is very difficult to measure and track these types of complex flows, the aircraft measurements only suggest that that CBL air could be entrained into the LSC circulation – convective motion in the CBL, northward flow of air and a strong downdraft behind the LSC front (Fig. 5c). The model vertical cross sections (Fig. 8) then provide a further indication that the circulation presented in the paper is possible. Following on the referee's comments and suggestions, forward trajectories were computed at a number of different locations (with an emphasis on starting in the CBL), altitudes and starting times to provide stronger evidence that the trajectory pathway proposed in the paper is a possibility. In Fig. 9, a forward air parcel trajectory is shown overlaid on the 285 m agl model wind fields at a) 12:00 LT and b) 13:00 LT. Although, the model predicts the LE and LSC fronts merging earlier than shown by the observations, the trajectory does show that air in the CBL at 12:00 LT is connected to that behind the LSC front 4 hours later (as the LE and LSC fronts begin to merge). The air parcel drops close to the surface in the downdraft behind the LSC front and moves onshore in the LSC lake breeze in a helical pattern. The circulation pattern proposed by the referee which is consistent with the trajectories shown in the old Fig. 9 (as stated later by the referee), are possible, but the evidence with the new Fig. 9 forward trajectory, now shows that the circulation proposed by the authors is also possible. The text in section 3.5, lines 509-520 has been changed to describe the new Fig. 9.

Added on lines 419-423, "Another possible circulation pattern could be that pollutants already present behind the LSC lake-breeze front were lofted upward at the front, transported northward, caught in the downdraft and confined along the south shore of

LSC. Pollutants from the CBL also appear to have been transported northward and observed during the later flight 5 time period discussed below.”

Comment:

p. 11506, line 3: Show Sarnia on Fig. 1 or refer to what's already on Fig. 1.

Response:

Sarnia is now included on Fig. 1 as point #12.

Comment:

p. 11509, lines 5-12: Make it clear that you are looking for evidence that the LSC front is present beneath the flight level of the aircraft, rather than supposing that the front itself extends up to 800m.

Response:

These sentences were re-written so that it was clearer that we were looking for evidence of the lake-breeze front at 800 magl rather than making the apriori assumption that the front actually extended to 800 magl. The new sentences, lines 315-323, “Guided by the location of the surface lake-breeze front, finding evidence of the presence of the south shore LSC lake-breeze front beneath the 800 m a.g.l. flight level was more difficult based on meteorology alone. Since CO is a relatively long-lived species in the troposphere, these measurements were examined for perturbations related to lake-breeze boundaries. A brief, sharp change in CO of 24 ppbv (green solid arrow at 12:01 LT), was observed (Fig. 3b2) suggesting the influence of the lake-breeze front.”

Comment:

p. 11509, lines 13-18: Since the LSC front is expected to be shallower than 800 m, no dew point change is expected at flight level. Since it's not relevant to the rest of the paper anyway, I suggest not attempting to infer the LSC front location north of LSC. This idea is further supported by the lack of evidence of a front at 300 m.

Response:

The text referring to the front north of LSC has been removed and replaced with ‘A LSC front north of LSC was not detectable in the aircraft data and there were no discernible features suggesting the presence of a surface-based front.’ The green dashed arrow depicting this front has been removed from Figure 3.

Comment:

Fig. 4: Are the time labels misplaced? For example, the text (p. 11510, lines 17-18) talks about a feature at 16:36 at 800 m, but according to Fig. 4's x axis, 16:36 occurs to the left of the start of the flight-level data.

Response:

The data on both Figs 3 and 4 are plotted so that LE is always on the left side and Lambton is always on the right side of the x-axis regardless of the direction of flight (and thus time). The following text is added at the beginning of Section 3.3. “Note that in order to compare one transect to the next, the data are plotted so that geographically LE is always on the left and Lambton is always on the right of the x-axis; this means then

that the time will either go from left to right or right to left depending on the direction of the aircraft transect.” The following text is also added in the Fig. 3 caption “Data are plotted so that LE is on the left and Lambton is on the right of the x-axis and the direction of the aircraft transect is depicted as a grey arrow” and for Fig. 4 caption “Data are plotted as in Fig. 3.”

Comment:

p. 11511, line 10: The relevant assumption is of slow processes, not stable processes.

Response:

Replaced the word stable with ‘slow’, line 368.

Comment:

Figs. 5 and 6: Single locations are given for the frontal positions, yet the fronts moved between aircraft passes at different altitudes. Did you correct for this motion by applying a horizontal adjustment to the flight-level data? If not, indicate the frontal location on each pass and note in the text that the vertical sections don’t actually represent the vertical structure of the atmosphere at any given time.

Response:

A horizontal adjustment was not made to the flight data. In Fig. 5, arrow heads denoting the LE front are marked for both the 800 m and 300 m passes that indicate the movement in this front. There was little movement in the LSC front. The dashed green arrow north of LSC was removed as there was not strong evidence for a front in this location.

The assumption is stated in the text that ‘the processes are sufficiently slow (changed from stable) that we can build these cross sections’. Although there is some movement in the LE front, we believe that these plots provide useful information about the vertical chemical and physical structure. At the beginning of Section 3.4, line 368-370, a statement is added to indicate that the LE front moved during the flight 4 time period. “Since there was some northward movement of the LE front during Flight 4, the front locations are marked on each transect.” The start and end times of the flights are also now labelled on the figure.

Comment:

p. 11513, lines 10-13: The TIBL growth does not limit vertical mixing. Any pollutant will take time to be lofted upward, and when the air mass is especially unstable (lake breeze air moving over hot land), the lofting should be especially rapid. The mixing limit is provided by the lake breeze inversion.

Response:

Agreed. This sentence, lines 425-430, is changed to: “Fig. 3b2, c2 shows that CO and SO<sub>2</sub> behind the LSC front are confined to lower layers and not mixed through the same depth as the CBL. This is hypothesized to be because the development of a thermal internal boundary layer (TIBL) behind the lake-breeze front growing vertically with inland distance would not get as high as the fully developed CBL in the short distance between the lake shore and the front. This would result in pollutants within the TIBL being mixed over a shallower layer, and thus less diluted, compared with the CBL.”

Comment:

p. 11513, lines 20-22: A simpler (or alternative) explanation is that there are fewer industrial pollutant sources on the north side of Detroit, where the air came from according to the trajectories.

Response:

Agreed. Lines 434-437, modified sentence to read "There was no evidence of any significant pollutant buildup north of LSC, which could be because there was less impact from sources along the back trajectory path (Fig. 2) and/or the weaker offshore lake-breeze circulation compared to the south and east sides of the lake."

Comment:

p. 11516, line 15: What is "LSC inflow"? How are you diagnosing along-shore transport from the vertical sections?

Response:

Changed 'inflow' to 'lake-breeze'. Along-shore transport is difficult to diagnose with the vertical cross sections that are presented – the text has been changed to eliminate such interpretation from this figure. The relevant sentence, lines 494-497, now reads "The model results suggest a circulation where air just south of the LSC lake-breeze front moves upwards and northward over LSC, gets caught in the downdraft behind the front, and moves ashore in the LSC lake breeze; the black arrow in Fig. 8a depicts this motion in the plane of the flight path."

Comment:

p. 11516, lines 20-23: Fig. 9a doesn't show a complete circuit. At most half a circuit is shown, and it's relevant to my main point above whether the circuit ever includes air ahead of the LSC breeze front. With half a circuit taking 2 h, a complete circuit would be 4 h, consistent with Fig. 9b.

Response:

A new Fig. 9 has been produced that shows a forward trajectory overlaid on the 285 m agl wind fields at 12:00 LT and 13:00 LT. The forward trajectory at 12:00 LT is within the convective boundary layer (CBL) at ~1000 magl and the model shows that as the LE and LSC fronts push together, the air parcel in the CBL drops in altitude, then pops back up over the merging fronts, drifts a little ways over LSC, gets caught in the downdraft and then moves in a helical trajectory back along the south shore of LSC in the lake breeze. Consistent with the referee's comment, the trajectory completes the circuit in about 4 hours.

Comment:

p. 11517, lines 17-19: The dashed grey arrow is incorrect. The text correctly diagnosed the light wind regime as the return flow aloft, where the air is "returning" only in a front-relative sense. Thus the light wind area corresponds to the top part of the solid gray arrow, where it's pointing slightly to the south. But then, as you showed, there's nowhere that the air is actually moving toward the south. The key to this paradox is the fact that the front is moving to the north. Streamlines and trajectories cannot be parallel to the front, or else it would not move. All along the front, the northward or upward flow must be sufficient to propel the front forward, so there must be flow toward the front (horizontally or vertically) within the lake air adjacent to the front.

Response:

We think that this comment is about the interpretation of the word 'circulation' and the frame of reference in which the figures are presented. Rather than present the figures as a conceptual model of a lake-breeze circulation, we changed the frame of reference to be a conceptual model of flow in the vicinity of a lake-breeze circulation. We are trying to synthesize all the pieces of information (measurements and model) in and around the lake-breeze circulation and account for synoptic and lake-breeze circulation with the winds being additive. The dashed grey arrow represents the resulting flow due to the interaction of the lake-breeze circulation with the synoptic wind. Thus, in this context, we also straightened the darker grey arrow so that it was not slightly pointing south. We changed Fig. 10 caption to read "Conceptual models of the flow in the vicinity of a lake-breeze circulation for..." and similarly, the text referring to this figure.

Comment:

p. 11519, lines 6-10: I'm having difficulty reconciling the units and description in the text with Fig. 12.

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  $\mu\text{g m}^{-3} \text{ppmv}^{-1} \%^{-1}$ . This change also addresses the same comment by referee #1. 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  $\text{OA}/\Delta\text{CO}$  and  $\text{SO}_4^{2-}/(\text{SO}_2+\text{SO}_4^{2-})$  for Flight 5 is confined to a narrower range of OA production ( $\sim 2.25 \mu\text{g m}^{-3} \text{ppmv}^{-1} \%^{-1}$ ), whereas in Flight 4, a linear relationship is observed with a range of OA production (relative to  $\text{SO}_4^{2-}$  production) extending from 1.2-1.75  $\mu\text{g m}^{-3} \text{ppmv}^{-1} \%^{-1}$ ."

Comment:

p. 11521, lines 13-14: If there's subsidence between 4.2 km and 12 km, it's not at all discernable in Fig. 5. Use a color palette that clearly depicts the downdraft. Up to this point, I had thought the downdraft referred to in the text was the one just behind the LSC front, since that's the only one I could see.

Response:

The measured vertical wind data in Fig. 5c show downdrafts behind the LSC front at  $\sim 4.2$  km from the front; note that the LSC front remains relatively stationary during the flight 4 time period (11:00-13:30 LT, see Fig. 1a,b). The aircraft vertical wind data however did not detect subsidence over LSC that is expected to be present given the lake-breeze situation around this lake. This is likely because the wind measuring system is more sensitive to updrafts/downdrafts over smaller horizontal distances and the subsidence over LSC may be over a broader horizontal scale. We do not make use of the time estimates from this figure anymore.

Comment:

p. 11521, lines 17-18: As I've argued above, I do not believe that the air has made a circuit from ahead of the LSC front to behind it. Thus the calculation of aerosol generation rates is dubious. But if the air did make such a circuit, the estimate of the time required for the circuit is off because it neglects the time air spends within the

downdraft over the lake at levels where the horizontal wind is very weak.

Response:

As Fig. 9 now shows, an air parcel trajectory is possible that connects the air ahead of the LSC front to behind it. The estimate of the time required for the circuit is taken from the forward trajectory (Fig. 9) as  $4\pm 1$  hours. The text describing circulation time estimates in section 3.7 from measurements and model output has been removed and modified to account for the trajectory estimate.

Comment:

p. 11521, lines 18-20: The difference in our interpretations could be settled with a model-output trajectory that originates ahead of the LSC front and ends up at 300 m behind it. In the absence of such a trajectory, there's no point in estimating the circuit time of a hypothetical trajectory with the model wind speeds.

Response:

A new Fig. 9 has been produced that demonstrates a model-output trajectory that originates ahead of the LSC front and ends up at 300 m behind it (more details in response to first comment). A complete circuit takes  $4\pm 1$  hours.

Section 3.5, lines 509-520 - modified to describe the new trajectory in Fig. 9.

Modified Fig. 10b to reflect the trajectory data over LSC (down arrow for downdraft)

Removed the whole paragraph on circulation time estimates in section 3.7 and modified the text using the circulation time derived from the forward trajectory analyses (Fig. 9) of  $4\pm 1$  hours.

Comment:

p. 11521, lines 27ff: The calculation relies on the unstated (and dubious) assumption that the air in the CBL is completely unchanged for 1-2 (or, by my calculation, 4) hours. Discuss this.

Response:

We agree that this is a very important point and have done some further analyses to address this. Since the new forward trajectory analyses (Fig. 9) shows a helical return pattern about the LSC circulation as the LE and LSC fronts are merging on a time scale of  $\sim 4$  hours, a comparison of data from flight 4 and 5 can then be made. The CBL in flight 4 is compared to AF air in flight 5 (AF was selected just north of the merged LE/LSC front to represent air that has been processed via LSC lake-breeze circulation on a time scale of  $\sim 4$  hours).

Section 3.3, lines 351-355, small modification (shown here in italics) to the following sentence: "Pollutant concentrations in this later afternoon flight were generally less variable compared to Flight 4 *in the region between LE and LSC*, but elevated pollutant *levels* were observed north of LSC, possibly from the CBL between LE and LSC, or pollutants from over LSC earlier that were advected northward and then mixed upward."

Comment:

p. 11522, lines 11-13: The important issue is not the enhancement of reaction rates

above the regional background, but rather the enhancement of reaction rates above the undisturbed CBL.

Response:

We agree with this comment, and this is what we have attempted to do by selecting a  $\text{SO}_4^{2-}$  production rate in the CBL in a non-plume and non-lake-breeze influenced area (Fig. 13, northeast side of LSC) to represent the undisturbed CBL.

Section 3.7, lines 585-586 added “(non-plume influenced and non-lake-breeze influenced)” to be more explicit that we are attempting to extract a value that is representative of a CBL that has not been influenced by lake-breeze circulations.

Comment:

p. 11522, lines 14-19: This explanation works the wrong way. Air within the CBL will spend much of its time within the clouds topping the CBL, while air within the LSC circulation will only experience clouds near the LSC front.

Response:

We agree with this. However, in light of our current analyses, we examined the cloud situation on June 25 in more detail. At 11am LT there were no clouds present and then an hour later, there was enhanced cloud growth at the LE front, some enhanced cloud growth at the LSC front, and shallow cumulus cloud in the CBL. This was the time, ~12pm, when the data in the CBL (flight 4) were taken. Thus, clouds started to form about the time the measurements were made (or just prior) indicating that the measurements reflect only limited cloud processing. Between 1pm-2pm, there was enhanced cloud growth at LE and LSC fronts with a small area of shallow cumulus cloud in the CBL, and there was significantly enhanced cloud growth as the LE and LSC fronts merged. By 3pm, there were no clouds in this particular area. At ~5pm when the measurements were made (flight 5), the forward trajectory indicates that the air in the CBL, at least partially, would be behind the LSC front. So given the air parcel trajectory and timeline, we believe that it is possible that cloud processing contributed to the enhancements in  $\text{SO}_4^{2-}$  and OA formation rates.

Additional explanation was added Section 3.7, lines 647-660.

Comment;

Fig. 8: Enlarge these panels to at least the size and aspect ratio of Figs. 5 and 6 to which they are to be compared.

Response:

Fig. 8 panels have been enlarged to the size and aspect ratio of Figs 5 and 6.