Author's response to reviewers: *Exploring the Nature of Air Quality over Southwestern Ontario: Main Findings from the Border Air Quality and Meteorology Study*

First Reviewer:
I must add that while the results summarized in this paper are clearly at the leading edge of our understanding of regional scale tropospheric pollution photochemistry, there are no analogous locations worldwide, so the results have mostly regional importance.

Response:

We agree that the physical, climatological and meteorological characteristics of the BAQS-Met Study area, combined with the quantity and distribution of emissions, are unique. Consequently, the specific features of the complex meteorology occurring over the study region, and the BAQS-Met results probing specifically how they impact air quality, are clearly most relevant to the southern Great Lakes and the large population effected by such conditions (i.e., from Chicago to Toronto and many large communities nearby). However, this uniqueness does not limit the relevance of the results towards other regions; the results are relevant to any region with coastlines co-existing with population centres and/or similar sources of pollutants.

Our overarching message that local circulation in this form of environment induces highly heterogeneous pollutant patterns, enhanced local secondary pollutant formation, and high concentrations of secondary pollutants, is relevant to many other regions. Such patterns are important to appreciate and understand in order to effectively avoid or reduce their adverse effects on human and ecosystem health. BAQS-Met's process-studies and modelling results are also of broader relevance demonstrating current capabilities of state-of-the-art models in challenging domains and areas requiring improvement (from emissions to representation of processes, to approaches for model evaluation). We appreciate this reviewer's perspective, though we expect that readers of this paper and those in the special ACP issue will find results that offer insights regarding other study areas worldwide.

Specific comments:
1) Line 83: The term "model observations" has no useful meaning. Elsewhere the authors use "model output", which does.
2) Line 88: "highly" is awkward. "strongly" would be better.
3) Line 155: "Great Lakes region" is misleading, since the study really only covers southwestern Ontario.
4) Line 252: "AOD" must be defined. It finally is on line 546.
5) Line 524: "Large differences" between what and what?
6) Line 530: OM/ΔCO must be defined.
7) Lines 548-549: The time zone must be specified. In the present case, since all chemical and physical processes under consideration are local, it should be local solar time, though local standard time would suffice.

8) Line 578: The “downward” in “downward subsidence” is redundant.
9) Line 639: See specific comment 7).
10) Line 689: It is not clear which the “first flight” is.
11) Line 742: Zhang et al 2012 is not in the list of references.

12) Line 742: Gordon et al 2012 is not in the list of references.
13) Line 835: “Similar to similar” is awkward.
14) Figure 5: “AWS” in the vertical axis label should be defined in the caption.

Response:

The revised manuscript addresses all of the specific and largely editorial comments above. We thank the reviewer for bringing these issues to our attention.

Reviewer #3

1) I suggest adding a plot showing a time series of air temperature and other meteorological variables at a surface station (similar to Figure 5)

Response:

This is a good suggestion and will clearly help readers gain a better understanding of the conditions during the study. The figure below has been generated to show both a time series of relevant meteorological parameters and the origin of the airmass(es) affecting the region during the two more significant O_3 and $PM_{2.5}$ episodes. The text has been modified to highlight this new figure and some of its key features and how they relate to the concentrations shown in Figure 5.

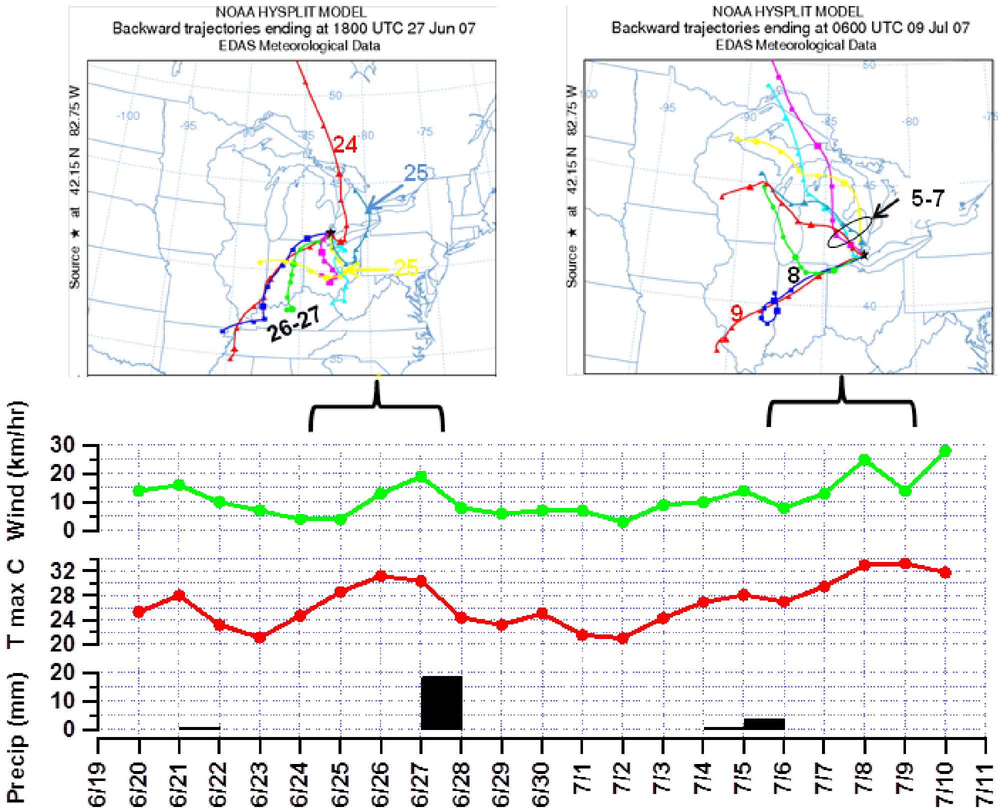

Figure 6 Time series of mean 850 hPa wind speed, daily maximum temperature and daily precipitation accumulation at Harrow. Three-day back-trajectories arriving every six hours corresponding to the two episode periods are shown in the top two panels. Trajectories are labelled with dates in June (24-27) and July (5-9) of arrival at Harrow.

2) Can you provide a short analysis/discussion of a possible weekday versus weekend effect observed in the pollutant concentrations (O_3, OM etc.)?

Response:

The duration of the intensive period was too short to be able to convincingly isolate how emissions modulations related to weekday vs. weekend cycles affected our observations given the dominant effect of regional and local scale meteorology. There were only three weekends (June 23-24, June 30-July 1, July 7-8) and as the new figure above shows, the subtle weekend/weekday effect is secondary in importance relative to the large meteorological effects, which were our primary focus. Furthermore, for the intensive period there were also likely to have been different local emissions arising due to the Canadian and U.S. holidays on

July 1st and July 4th, respectively. Even for the longer 'Mesonet" period, disentangling meteorological and emissions effects on O3 or OM would require an in-depth analysis to attempt to tease that signal out of the dominating meteorological effects. As uncovering weekend and/or holiday effects on pollutant levels was not an objective of the BAQS-Met, the papers published in the special issue did not focus on this issue. This paper is intended to be an overview of that already published work and not a source for significant new analyses. However, while including an in-depth analysis/discussion is beyond the scope of this paper, we now have highlighted the issue of weekend and holiday effects on the observations in the context of presenting Figure 5 and the new figure (6) showing the meteorological time series.

3) If possible, can you add two vertical cross-sections from the model output to display the impact of the lake and land breezes on ozone transport across the region? This phenomenon is widely discussed in the paper, therefore I think a nice illustration of the process would be very valuable.

Response:

This is a good suggestion and we agree that cross-sections help demonstrate the phenomena discussed. Several cross-sections demonstrating the circulation and ozone impacts were already shown in the individual study papers. We do not feel it is necessary to reproduce them in the summary paper as readers can refer to the earlier work, but we highlight these figures again here in this response so that the reviewer, and future readers, will be able more quickly access such information.

From Makar et al, 2010(b) (all references appear in the original text): Figure 12(a) and 14 (below) show respectively the modelled surface ozone concentration/wind field for a frontal convergence line between lake breeze fronts from Lake Huron and Lake Erie and a cross-section from Lake Huron to Lake Erie on the same day. This demonstrates the impact of the convergence line on the local ozone production: a very significant and local increase in ozone production. Figure 14 shows the narrow cross-sectional extent of the convergence line's ozone, and the extent to which this ozone carried aloft (to a height of 1.5km). The helical nature of flow along the convergence lines may be inferred from the circular wind patterns in the cross-section of Figure 14, coupled with the horizontal flow in the surface concentration map; the flow follows a helix moving downwind along the frontal convergence line.

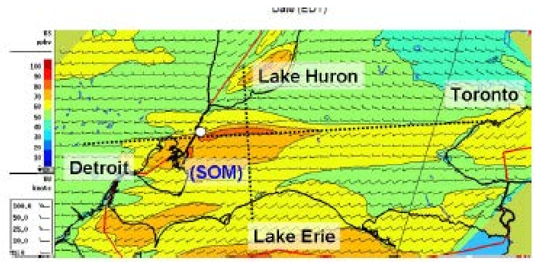

Fig. 12. (a) Model-predicted ozone versus observations, 8 July, Sombra station. (b) Model-predicted surface ozone and surface winds at 17:00 UT (01:00 p.m. local time).

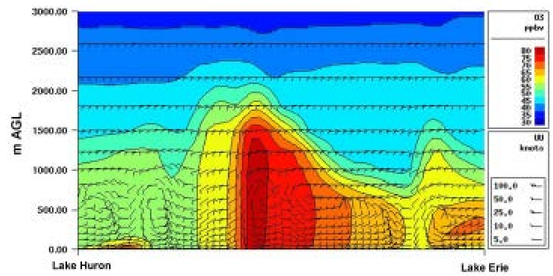

Fig. 14. Model-predicted ozone and wind fields, 8 July, 17:00 UT, Lake Huron to Lake Erie cross-section.

Also from Makar et al, 2010(b), the figures below show the modelled average afternoon surface wind field and ozone concentrations for the study area during the measurement intensive. The purple lines indicate, on average, where convergence and pollutant confinement can be expected to occur. A key feature of the second figure is the high concentrations of ozone over Lake Erie. These are driven by subsidence over Lake Erie and outflow at the surface, seen in Figure 17.

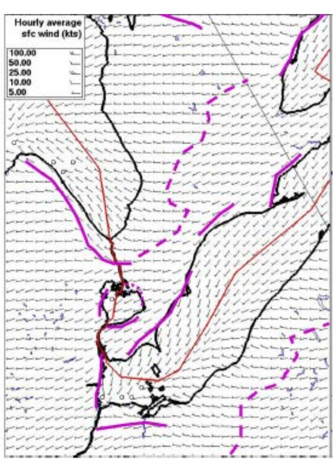

Fig. 17. 23-day average wind fields at **(a)** 12:00 UT, **(b)** 16:00 UT, **(c)** 20:00 UT, and **(d)** 00:00 UT; 08:00 a.m., 12:00 p.m., 04:00 p.m., and 08:00 p.m. local time, respectively. Convergence regions marked as solid mauve lines, boundary of divergence outflow regions marked with dashed mauve lines.

Figure 19 from Makar et al 2010(b) shows the modelled averaged surface ozone concentrations(a) as well as the average concentrations in a cross-section extending from Lake St. Clair to Lake Erie (b) showing both a “dome” of ozone over the smaller Lake St. Clair due to the coupled downdraft over the lake and updraft over the city of Detroit, and a much shallower lake-breeze-subsidence-induced “pancake” of ozone over Lake Erie. Figures 19(c) and (d) show regions of chemical production of ozone resulting from this circulation, and total change in ozone due to transport, respectively.

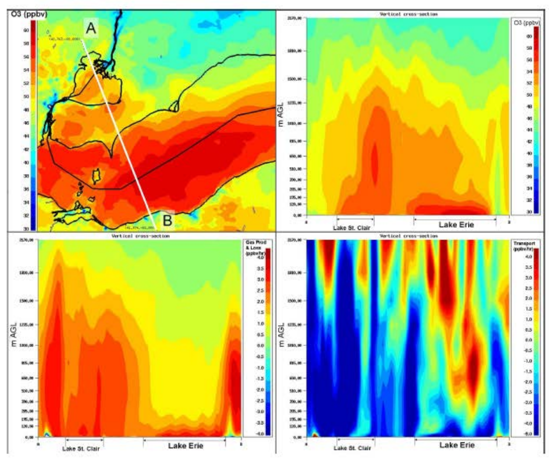

Fig. 19. 23-day average values at 20:00 UT (04:00 p.m. local time): **(a)** surface ozone over Lake St. Clair and Lake Erie, showing location of cross-section used for **(b)** ozone concentration, **(c)** ozone gas-phase production and loss, **(d)** ozone total transport.

Figure 20 from Makar et al shows modelled average wind fields in the afternoon over the same Lake St. Clair to Lake Erie cross-section, with the wind barbs overlaid with dotted lines showing the circulation patterns. The average winds suggest a pair of linked vorticies, reaching the surface for Lake St. Clair at the left, and remaining aloft above the north shore of Lake Erie. The strong afternoon subsidence over Lake Erie is also shown.

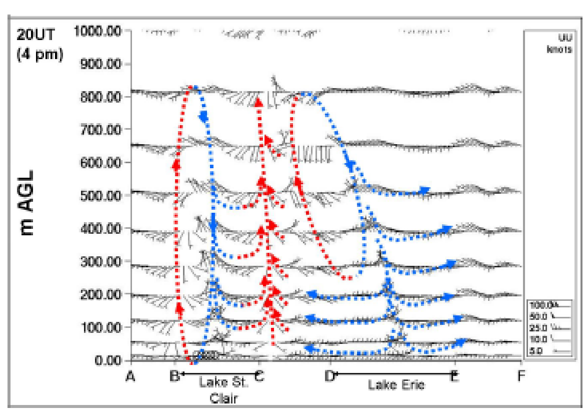

Fig. 20. Cross-section of 3-D wind fields across Lakes St. Clair and Erie at **(a)** 12:00 UT (08:00 a.m.), **(b)** 16:00 UT (12:00 noon), and **(c)** 20:00 UT (04:00 p.m.). Net upward motion streamlines are sketched in red, downward motion in blue. Inset: location of the cross-section, superimposed on 20:00 UT surface winds.

From Levy et al.2011, Figure 4 below, shows the modelled average surface ozone concentration (top row), and cross-sections between Lakes St Clair and Erie for ozone (middle row), and mean vertical velocity (bottom row), for additional times. These demonstrate how the circulation varies (bottom row of figures), with the up and downdraft pairs at 4pm corresponding to the highest local ozone concentrations. The figure also shows significant updrafts over the city of Cleveland at night, resulting from local heat island effects – heat island circulation coupled with lake subsidence also drives ozone production over Lake St. Clair, in the figures above. Figure 4 also demonstrates the strong diurnal nature of the average circulation, and regions of ozone formation and chemical destruction and/or transport removal.

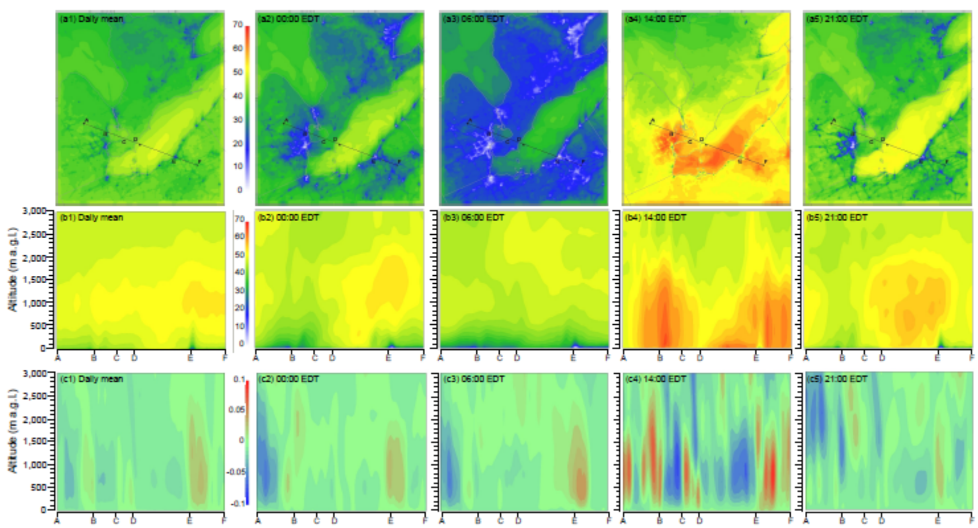

Fig. 4. Mean AURAMS model output showing **(a)** mean ground-level ozone, **(b)** mean ozone cross sections (ppbv), and **(c)** mean vertical velocity ($m\,s^{-1}$) for all 24 h (a1, b1 and c1) and selected times (00:00, 06:00, 14:00 and 21:00 EDT, a2-5, b2-5 and c2-5, respectively), calculated for the entire 23 days of the intensive campaign (18 June–10 July 2007). Cross sections are along transect A-F marked in (a), with contour intervals of 2 ppbv for ozone and 0.01 $m\,s^{-1}$ for vertical velocity.

Hayden et al. (2011) also included a number of cross-section diagrams, both from model output and schematics. The cross-section example shown in Figure 8 of their work shows the model representation of lake breeze winds, including the return flow aloft and subsidence over the lake. In this case, there was a local enhancement in sulphate, relative to SO2, trapped near the surface (below 500 M AGL) of Lake St. Clair and the adjacent on shore area. When available the study's measurement data have tended to corroborate the modeled concentration patterns described above, although the model clearly provides the complete picture while measurements were obtained at discrete locations and times.

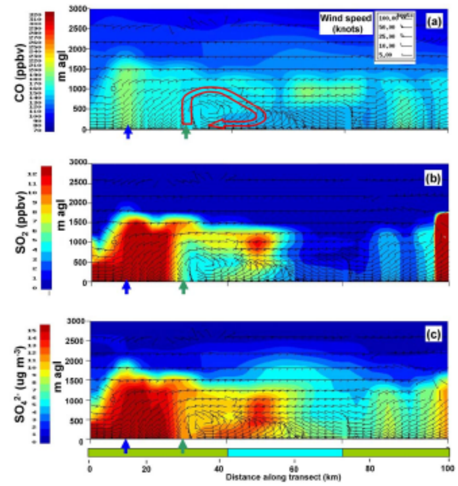

Fig. 8. Vertical cross section for model-predicted **(a)** CO and **(b)** SO_2 and **(c)** SO_4^{2-} at 12:00 LT along the axis of the aircraft transect (dashed red line in Fig. 7). Wind direction overlaid is the 3-D wind field in the plane of the cross-section and, thus can be used to indicate horizontal and vertical motion in the plane of the cross-section. Red arrow in panel **(a)** represents the LSC lake-breeze motions. Land = green bar; lake = blue bar. Coloured arrows indicate lake breeze fronts (blue = LE, green = LSC).

Key BAQS-Met papers with vertical cross-sections:

Hayden, K. L., Sills, D. M. L., Brook, J. R., Li, S.-M., Makar, P. A., Markovic, M. Z., Liu, P., Anlauf, K. G., O'Brien, J. M., Li, Q., and McLaren, R.: Aircraft study of the impact of lake-breeze circulations on trace gases and particles during BAQS-Met 2007, Atmos. Chem. Phys., 11, 10173-10192, doi:10.5194/acp-11-10173-2011, 2011.

Levy, I., Makar, P. A., Sills, D., Zhang, J., Hayden, K. L., Mihele, C., Narayan, J., Moran, M. D., Sjostedt, S., and Brook, J.: Unraveling the complex local-scale flows influencing ozone patterns in the southern Great Lakes of North America, Atmos. Chem. Phys., 10, 10895-10915, doi:10.5194/acp-10-10895-2010, 2010.

Makar, P. A., Zhang, J., Gong, W., Stroud, C., Sills, D., Hayden, K. L., Brook, J., Levy, I., Mihele, C., Moran, M. D., Tarasick, D. W., He, H., and Plummer, D.: Mass tracking for chemical analysis: the causes of ozone formation in southern Ontario during BAQS-Met 2007, Atmos. Chem. Phys., 10, 11151-11173, doi:10.5194/acp-10-11151-2010, 2010b.