Atmos. Chem. Phys. Discuss., 11, C5184–C5187, 2011
www.atmos-chem-phys-discuss.net/11/C5184/2011/

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Aircraft study of the impact of lake-breeze circulations on trace gases and particles during BAQS-Met 2007" *by* K. L. Hayden et al.

Anonymous Referee #2

Received and published: 20 June 2011

This manuscript reports an airborne study on the influence of lake-breeze circulations on particle chemistry in the southern Great Lake region of Canada. Lake-breeze circulation events were identified and interpreted using chemical and meteorological observations in conjunction with air quality model simulations. The complexity of lake-breeze circulation patterns and their interactions with the synoptic flow are discussed. The authors also attempted to estimate the production rates of secondary aerosol species. The topic of this work is a good fit for ACP. However, I have some comments on data analysis and interpretations that I hope could be addressed prior to publication.

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. 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. 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.

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 $\sim$ 1 ug m-3 ppmv-1 %-1? Nevertheless, it is not clear to me the physical bases of assuming that (OOA)/deltaCO should change linearly with SO4/(SO2+SO4) ratios? 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 μgm-3 ppmv-1âĂć 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? 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.

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.

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

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

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.

Page 11509, line 11, extra "in"?

Page 11514, line 20, extra "being"?

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.

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

Figure 7, please show latitude and longitude scales.

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

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

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?

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.

Interactive comment on Atmos. Chem. Phys. Discuss., 11, 11497, 2011.