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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Ozone distributions over southern Lake Michigan: comparisons between ferry-based observations, shoreline-based DOAS observations and air quality forecast models" *by* P. A. Cleary et al.

Anonymous Referee #3

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This study compares O3 measurements made by Differential Optical Absorption Spectrometry (DOAS) on the shoreline of Lake Michigan, Thermo-based O3 measurements over Lake Michigan on a commercial ferry and O3 predictions by the National Air Quality Forecast System. The measurements show largest differences during the evening and night, with levels higher over the lake, and with little dependence on ferry position or temperature. On-shore DOAS measurements of NO2, SO2 and HCHO demonstrate differences between onshore and offshore air masses which are attributed to the diurnal lake breeze cycle. Comparisons of the ferry O3 measurements with the National

Weather System's National Air Quality Forecast System indicate mean model biases of 12 ppb with non-uniformity in model biases for ferry location, time of day or month.

The paper is well written and the data are logically presented.

Over-water O3 measurements are sparse and in combination with onshore measurements make this dataset unique and would be of interest to ACP readers.

I have three major concerns with the paper as outlined below.

1. It is not clear to me what new information is being presented in this paper that is not already in the literature. The authors describe previous work in the Lake Michigan area and indicate that there has been O3 reduction in the past 20 years, but are there still problems with non-attainment? How significant are the current problems? What was the impetus for this particular study? The need for routine offshore comparisons with onshore measurements has been highlighted, but the reasons why are not described. This study may be the first to present regular O3 measurements over Lake Michigan and compare with national air quality forecasts (p. 23207 L. 11), but what new information does this study provide in that is any different from previous studies? This needs to be highlighted. As an aside, why do the authors not make use of other land-based air quality measurements that exist in this region to help characterize the onshore air quality, sources, and the behaviour of the lake breeze? This would also provide context for the measurements presented in this paper.

2. Uncertainties and statistical comparisons are not presented and discussed. Particularly because comparisons (ie. differences) between DOAS and TECO O3 data and TECO O3 and model output are being presented, it is important to quantify uncertainties and statistically compare the data in order to ascertain whether differences are significant or not. What is the accuracy of the DOAS? Were the DOAS and O3 systems run side by side to intercompare and substantiate whether there is a bias in either measurement? This is a critical starting point to ensure the measurements are comparable. In addition, the uncertainties in digitizing the model images needs to be

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quantified.

3. The model comparison with the ferry O3 measurements in Section 3.3 is very limited. Interpretation of measurements versus model requires an in depth understanding of the model itself. Little information is provided about the model used in this paper. What emissions inventory is being used? How often is the meteorology updated? What chemistry is in the model? What is the spatial resolution i.e. can lake breeze circulations be captured and how does this impact the comparisons? What can the model tell us about the vertical O3 structure over the lake? What are the most significant model limitations in the context of these comparisons? Also, it is odd to me why the authors needed to digitize model images – why would the data not be available to presumably obtain much better quality comparisons with presumable much better accuracy than using images. No information is provided about the uncertainty associated with digitizing these images. If this paper is to include a large section on model comparisons as it does, then a much more comprehensive comparison needs to be done, most preferably using direct model output. Otherwise interpretation is left completely to speculation with an all-encompassing list of possible causes of discrepancies - meteorology, emissions, and chemistry. The authors are referred to Makar et al. (2010), reference provided below, for work that is relevant to this paper.

Minor comments

In order to better understand the O3 measurements presented in this paper, it would be helpful to have further information and interpretation of emission sources and possible impacts. The following comments (1-5) are related to this:

1. p. 23206, L.24- p. 23207 L. 5 How does the combination of DOAS species provide a 'unique breadth of information about air masses'? and What is meant by this 'measurement strategy'?

2. p. 23204 L. 21 What is unique about the distribution of o3 precursor emissions and why not state more explicitly what and where they are? A figure depicting emission

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sources might be helpful.

3. p. 23210 L 1-2, Not a very comprehensive picture for the reader to know where the major sources are located and what they are.

4. p. 23210 L9-11 'may implicate influence of power plant emissions over longer distances'. . .where are the sources implicated? Any attempt at connecting the measurements to these sources through e.g. back trajectories, calculating time scales etc.

5. p. 23207, L. 5-7 I don't see in this study how the DOAS measurements used to determine changes in chemical composition to describe emissions, processing and transport of plumes over Lake Michigan.

6. p 23211, L 16-18 so why not discuss the different sources that impact your measurements How were the meteorological measurements made? What are their uncertainties? Was a minimum wind speed criteria used for the wind direction measurement to avoid stagnant conditions which may be unreliable (Fig 2)? The O3 inlet seems like it would be a fairly lengthy line – what are the details on this e.g. residence time, subsampled off faster flow? Was an inlet filter used and how often was it changed? Was there some kind of 'rain cover'? How often were the O3 zeros done?

Should indicate in the Figure 1 caption that the ferry path is shown in the inset.

p. 23204 L. 20 Please add references for forecast models predicting high lake O3

p. 23206, L. 12, typo synoptic

Figure 2, SO2 also looks like it is elevated from the south (land) in addition to over water

Change concentrations to mixing ratios throughout manuscript.

p. 23209 L 15-26 description here doesn't seem to lead to any interpretation of the measurements and thus why is this even discussed?

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p. 23210 L 3-5 what about night time losses of NO2?

Figures 11, 12 and 14, Differences between west and east: how does the 2.5 hour time period for a trip factor into the data interpretation? I would expect changes in O3 chemistry over this time period. How much uncertainty is estimated from these changes?

p. 23214, L 10-13, sentence doesn't make sense

Figure 13 – need to clarify what 'number' represents (ie number of days) on both the yaxis and the caption

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

Interactive comment on Atmos. Chem. Phys. Discuss., 14, 23201, 2014.