Atmospheric Environment 109 (2015) 52–60

Contents lists available at ScienceDirect

Atmospheric Environment

journal homepage: www.elsevier.com/locate/atmosenv

Impact of lake breezes on ozone and nitrogen oxides in the Greater Toronto Area

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HIGHLIGHTS

- Impacts of lake breezes on air quality is investigated in the Greater Toronto Area.
- Lake Ontario breezes form on 74% of summer (May–September) days.
- O_3 is 42–49% (13–15 ppb) higher when a lake breeze is present.
- Only sites within the circulation exhibit enhanced O_3.

ARTICLE INFO

Article history:
Received 24 November 2014
Received in revised form
27 February 2015
Accepted 3 March 2015
Available online 5 March 2015

Keywords:
Lake breeze
Ozone
Greater Toronto Area
Nitrogen oxides

ABSTRACT

Meteorological and air quality datasets from summertime (May to September, 2010–2012) were analysed in order to assess the influence of lake-breeze circulations on pollutant levels in the Greater Toronto Area (GTA). While previous estimates of the frequency of summer days experiencing lake breezes range between 25 and 32 % for the GTA, a simple algorithm using surface meteorological observations suggested Lake Ontario breezes occurred on 56% of summer days, whereas a more reliable multiplatform approach yielded a frequency of 74%. Data from five air quality stations across the GTA were used to compare air quality on days during which a lake-breeze circulation formed ("lake breeze days") versus days when one did not ("non-lake breeze days"). Average daytime O_3 maxima were 13.6–14.8 ppb higher on lake breeze days relative to non-lake breeze days. Furthermore, the Ontario Ambient Air Quality Criteria (AAQC) for 1-h average O_3 (80 ppb) and 8-h average O_3 (65 ppb) were exceeded only on lake breeze days and occurred on a total of 30 and 54 days throughout the study period, respectively. A causal link between lake-breeze circulations and enhanced O_3 was identified by examining several days in which only some of the air quality sites were inside the lake-breeze circulation. O_3 mixing ratios at sites located within the circulation were at least 30 ppb higher than sites outside the circulation, despite similar temperatures, cloud conditions and synoptic regimes across the region. Rapid O_3 increases were concurrent with the arrival of the lake-breeze front, suggesting O_3-rich air from over the lake is being advected inland throughout the day. Lake-breeze circulations were found to have less impact on nitrogen oxide (NO_x) levels. Morning NO_x was greater on lake breeze days, probably due to the stagnant conditions favourable for lake breeze formation. During the late afternoon, only inland sites experience increased NO_x on lake breeze days, likely as a result of being downwind from near-shore city centres.
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1. Introduction

1.1. Air quality and the Greater Toronto Area (GTA)

Highly populated urban areas often experience poor air quality and photochemical smog as a result of large emissions of pollutants from industrial activities and transport. Tropospheric ozone (O_3) can be a major constituent of smog and has adverse health effects for both humans and vegetation (Seinfeld and Pandis, 2006). Ozone is produced in the troposphere through a series of complex, non-linear reactions that depend on two main precursors: volatile organic compounds (VOCs) and nitrogen oxides (NO_x). Meteorology also exerts a strong control on O_3 levels, with clear skies, high temperatures and stagnant conditions generally favouring higher concentrations. The behaviour of these pollutants and their

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http://dx.doi.org/10.1016/j.atmosenv.2015.03.002
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precursors is heavily influenced by meteorology and local topography. For instance, Lu and Turco (1995) found sea breezes and mountain-induced flows in the Los Angeles Basin to be a key contributing factor to poor air quality in Los Angeles.

In this context, we explore air quality in Toronto, Canada (43°40′N, 79°23′E), which is situated along the north-western shore of Lake Ontario. The Greater Toronto Area (GTA) has a population of roughly 5.5 million and encompasses 7000 km^2. It is one of the most populated and industrialized regions in Canada and is frequently afflicted with poor air quality. A report released by the Ontario Medical Association (OMA) estimated that the annual economic burden of air pollution in Ontario in 2015 will be CAD $9.8 billion (OMA, 2005). Establishing the major determinants of air quality in the GTA will help in the development of more accurate air quality forecasts (including smog alerts) and of more effective mitigation strategies. Previous studies have looked at the impact of meteorology and/or reduction of VOC and NO_x emissions on O_3 (Geddes et al., 2009; Pugliese et al., 2014) in the GTA. However, the influence of lake-breeze circulations on these pollutants has not been well characterized in the GTA.

1.2. Lake-breeze circulations

Lake (and sea) breezes are mesoscale meteorological phenomena that result from a pressure difference triggered by preferential heating of land relative to water, and typically develop a few hours after sunrise and persist until sunset (Physick, 1980; Crosman and Horel, 2010). The positive land-lake temperature difference is more pronounced in summer; hence most lake-breeze circulations develop from May to September in the Northern hemisphere. Fig. 1 shows a vertical cross-section of an idealized lake-breeze circulation under light synoptic winds. As the sun rises the land will heat up faster than the water, which results in the air over land becoming warmer than the air over water. This thermal contrast results in a pressure differential that causes cooler, higher pressure air from over the lake to flow inland. As the air mass penetrates inland it warms up and the shallow thermal internal boundary layer (TIBL) deepens. A narrow updraft region, called the lake-breeze front, forms and progresses inland throughout the day. Subsiding air in the return flow completes the lake-breeze circulation and creates a very shallow capping inversion over the lake. The capping inversion helps to suppress cloud formation behind the lake-breeze front, whereas the updraft region can promote a narrow band of clouds along the front (Segal et al., 1997). As a result, defining features of a lake-breeze circulation are high solar insolation, shallow inflow boundary layer, and the entrainment of pollutants in a circulating pattern. Some lake-breeze fronts penetrate as far inland as 200 km; however, most stay within tens of kilometres of the shoreline (Sills et al., 2011). Land breezes are analogous phenomena that occur at night-time due to a negative land-lake air temperature difference and result in an offshore flow. Extensive reviews of the lake breeze phenomenon are prevalent in the literature (e.g. Simpson, 1994; Crosman and Horel, 2010).

Numerous studies have explored the effect of lake (and sea) breezes on air quality in coastal environments, and several have focused on impacts to O_3 in the Great Lakes region of North America (e.g. Biggs and Graves, 1962; Sills, 1998; Hastie et al., 1999; Lennartson and Schwartz, 2002; Hayden et al., 2011). These studies conclude that O_3 concentrations at inland locations are generally higher during the presence of a lake-breeze circulation, which may be a result of: (1) increased solar insolation, (2) decreased mixing height, and/or (3) recirculation of pollutants. However, the conditions most favourable for lake breeze formation (clear skies, light winds, warm air temperature) are also conducive to rapid O_3 production (Seinfeld and Pandis, 2006). Hence, it is difficult to establish whether a cause-and-effect relationship exists between lake-breeze circulations and enhanced O_3, or whether the relationship is merely a correlation. Other investigations have examined the behaviour of other pollutants in lake (and sea) breeze circulations, such as $PM_{2.5}$ (Harris and Kotamarthi, 2005; Hayden et al., 2011), PM_{10} (Papanastasiou and Melas, 2009), and NO_x (Reid et al., 1996; Hastie et al., 1999). However, these studies are less prevalent and the role(s) lake breezes play in modifying the levels of these pollutants is much less clear. This present study investigates the impact lake-breeze circulations have on air quality in the GTA using a representative (season-long), multi-year approach. To our knowledge, this is the first attempt to ascertain the impact of Lake Ontario breezes on O_3 and NO_x across the GTA using complete, multiyear datasets. The specific goals of this paper are to:

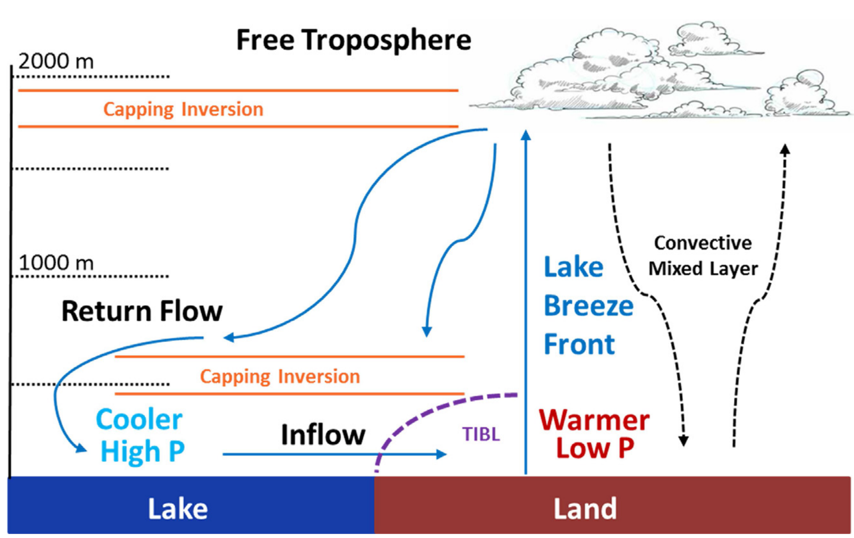

Fig. 1. Vertical cross-section of a typical lake breeze under light synoptic winds. Arrows depict motions of air masses both inside and outside the lake-breeze circulation. A thermal internal boundary layer (TIBL) is shown in purple and grows in height with distance inland until it reaches the height of the convective mixed layer. The lake-breeze front is denoted by the vertical blue arrow and the orange lines represent capping inversions. This figure is adapted from Sills et al. (2011). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

Fig. 2. Location of air quality (yellow pins) and meteorological (purple pins) stations. The red and green pins indicate the Lake Ontario buoy used to record lake surface temperature and meteorology station used for solar insolation data, respectively. Map data: Google, NOAA. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

- Determine the frequency of lake breezes impacting the GTA during the warm season (May to September) for 2010, 2011 and 2012 (section 3.1)
- Assess the applicability of different methodologies used to identify lake breezes (see sections 2.2 and 2.3, results in section 3.1)
- Compare meteorology, O_3, and NO_x on days when a lake breeze forms versus days without a lake breeze (sections 3.2 and 3.4)
- Determine if a causal link exists between high O_3 concentrations and lake-breeze circulations (section 3.3)

2. Methods and materials

2.1. Air quality and meteorological observations

Data from five air quality monitoring stations across the GTA were chosen to explore the pollutant concentrations on lake breeze versus non-lake breeze days. These stations are operated by the Ontario Ministry of the Environment and Climate Change (OME) and report hourly averages of NO_x, NO_2, and O_3 mixing ratios. Measurements of NO_x/NO_2 and O_3 were performed using automated continuous chemiluminescence and UV absorption spectroscopy, respectively. This historical data is accessible online and data for the months May to September (inclusive) were obtained for the years 2010, 2011 and 2012 (Ontario Ministry of the Environment and Climate Change, 2014). Only the months May to September are considered because the frequency of lake-breeze circulations is low outside these months. Corresponding meteorological observations were acquired during the same period from 5 sites across the GTA which are in close proximity to the OME air quality sites. These meteorological stations were managed by Environment Canada (EC) and record standard atmospheric parameters (e.g. temperature, RH, wind speed, wind direction) (EC, 2014). Hourly lake surface temperature measurements were retrieved from a buoy operated by Environment Canada in western Lake Ontario. The locations of these stations are shown in Fig. 2 with additional information regarding location, identification code, and distance from Lake Ontario in Table 1. Solar radiation was measured using both a net radiometer (CNR1, Campbell Scientific Corp.) and a pyranometer (CMP 11, Kipp and Zonen B.V.) run by the University of Toronto Mississauga Department of Geography's Meteorological Station (UTM).

2.2. Lake breeze identification: algorithm method

To determine the influence of lake-breeze circulations on pollutant levels it is critical to accurately identify the existence of a lake-breeze circulation. One common method that has been employed is to detect the passage of a lake (or sea) breeze front by analysing surface station data for a rapid change in temperature and/or dew point, as well as an abrupt wind shift from offshore to onshore flow (e.g. Hastie et al., 1999; Zumpfe and Horel, 2007; Atkins and Wakimoto, 1997). Early efforts by Biggs and Graves (1962) and Lyons (1972) created lake-breeze indices that were used to parameterize the occurrence of lake breezes with data from surface meteorological stations. Laird et al. (2001) attempted to improve upon these indices by using criteria to ensure other atmospheric phenomena (i.e. synoptic-scale fronts) do not yield false positives. The four conditions from Laird et al. (2001) that must be met in order to positively identify a lake-breeze circulation are:

1. Change in average wind direction from offshore or calm conditions (<5.5 m s^{-1}) in the morning (5:00–7:00 LST) to onshore during the afternoon (16:00–18:00 LST).
2. Positive temperature difference between the daily maximum at the inland station and the lake surface measured at the same hour.
3. Average air temperatures in the morning (5:00–7:00 LST) lower than during the afternoon (16:00–18:00 LST).

Table 1
Location of the observation stations.

Station name	Station code	Site type	Latitude (°N)	Longitude (°W)	Distance from Lake Ontario (km)
Burlington Piers	WWB	Met	43°18′00	79°48′00	0.2
Billy Bishop Airport	YTZ	Met	43°37′49	79°23′46	0.2
Pearson Airport	YYZ	Met	43°40′38	79°37′50	12.6
Buttonville Airport	YKZ	Met	43°51′44	79°22′12	19.7
Oshawa Airport	YOO	Met	43°55′22	78°53′00	8.0
Burlington	BUR	Air Quality	43°18′54	79°48′90	0.1
Toronto Downtown	TDT	Air Quality	43°39′46	79°23′17	2.9
Toronto North	TON	Air Quality	43°46′53	79°25′30	16.3
Newmarket	NEW	Air Quality	44° 20′39	79°28′59	42.6
Oshawa	OSH	Air Quality	43°56′45	78°53′41	10.8
Lake Ontario Buoy	WLO	Lake Temp	43°15′50	79°32′27	–
U of T Mississauga	UTM	Solar Data	43°32′58	79°39′43	6.5

4. An average wind speed in the morning (5:00–7:00 LST) less than 5.5 m s^{-1}.

Laird et al. (2001) constructed the four criteria based on 15 years of observations and then validated it by comparing its ability to detect lake breezes against 2 years of detailed observations (similar to the methodology outlined in section 2.3). The authors found that the four criteria listed above could reasonably identify Lake Michigan lake breeze events. Lennartson and Schwartz (2002) used this same methodology in conjunction with O_3 measurements to determine that 82% of 1-h O_3 exceedances off the western shores of Lake Michigan occur in association with lake-breeze circulations. Therefore, we adopt this same algorithm as our first approach to identify lake breeze events off of Lake Ontario. Onshore winds in this paper are defined as 45–135° for WWB, 70–225° for YTZ, 90–200° for YYZ and YKZ, and 135–225° for YOO. If any one of these stations meets the criteria then that day is classified as a "lake breeze day". It is important to note that many other analogous algorithms have also been invoked to identify a lake (or sea) breeze day (e.g. Papanastasiou and Melas, 2009; Furberg et al., 2002; Steyn and Faulkner, 1986).

2.3. Lake breeze identification: manual method

These algorithms are useful because of their objective criteria, which only require standard meteorological measurements and can be easily applied to multi-year datasets. However, conditions 1 and 4 frequently prevent identification of lake breezes that form in a moderate to strong onshore synoptic regime, leading to false negatives. Furthermore, there are several scenarios that can result in the algorithm detecting a false positive; for example, sudden wind shifts from storm outflows or the passage of other mesoscale features. Therefore, it can be advantageous to use a manual, multi-platform approach rather than relying solely on surface station data. This manual method incorporates visible channel satellite imagery and radar, and has been successfully applied for mesoscale boundary analysis (e.g. Roebber and Gehring, 2000; Wilson and Roberts, 2006; Sills et al., 2011). The manual identification method used for this study follows that developed by Sills et al. (2011). Images showing surface weather station observations and low-level radar reflectivity data superimposed on GOES-13 visible-channel satellite imagery were generated for each hour in the study period using the Aurora workstation (Greaves et al., 2001). An example is provided in Fig. 3.

For each day,[1] the images were inspected visually at hourly intervals and via animations to identify clear evidence of a Lake Ontario lake-breeze circulation. Criteria used to manually identify lake-breeze fronts, listed in Sills et al., 2011 (Table 1), include a rapid shift in wind direction to an onshore wind, and the presence of a line of cumulus clouds and/or radar fine line quasi-parallel to the shore that moves gradually inland (and is not associated with a convective gust front or synoptic-scale front). A day with at least one hour of lake breeze conditions affecting onshore locations was classified as a "lake breeze day".

Fig. 3. Example of an analysis image used for manual lake breeze identification with surface weather station observations and radar data superimposed on visible satellite imagery (valid 2:00 PM LST on 16 Aug 2011). The magenta lines indicate the analysed positions of lake-breeze fronts. Weather station data shown are temperature (upper left) and dew point temperature (lower left) in degrees Celsius, and wind direction and speed (wind direction indicator points in the direction from which the wind is blowing, short barb = 2.5 m s^{-1}, long barb = 5 m s^{-1}). Low-reflectivity radar returns (<25 dBZ with green lowest, blue highest) are due mainly to the presence of airborne insects. The locations of the BUF (Buffalo, NY) and WKR (King City, ON) radar stations are indicated. Clouds are shown in greyscale with white being the highest albedo. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

3. Results and discussion

3.1. Lake breeze frequency

The frequency of lake breeze days for each year and both methods is shown in Table 2. The year-to-year variability of lake breeze frequency is low and lake-breeze circulations are established in the GTA on 56% and 74% of the study days as computed using the algorithm and manual methods, respectively. Previous estimates for lake breeze occurrence off of Lake Ontario during summer days are 32% (Guski and Miller, 1980), 25% (Chermack, 1986) and 30% (Comer and McKendry, 1993). The lake breeze frequencies from this study are much higher, which may be partly due to improvements in both the quality and spatial coverage of observation stations. The discrepancy could also be attributed to the criteria used for classifying lake breezes, as these previous studies employed more rudimentary algorithms; Comer and McKendry (1993) explicitly state that their value is a conservative estimate. The results from this work are similar to values obtained for other Great Lakes; Lyons (1972) and Laird et al. (2001) used an algorithm method and reported lake breezes occurring on 46% and 62% of summer days off of Lake Michigan. Sills et al. (2011) found

Table 2
Frequency of lake breeze events in the GTA from May to September.

Year	Algorithm method		Manual method	
	Number	Percentage	Number	Percentage
2010	80	52%	110	72%
2011	88	58%	114	75%
2012	89	58%	116	76%
Total	*257*	*56 %*	*340*	*74 %*

[1] Analysis images were missing for several days in 2012. For these days, surface station observations, radar data and/or satellite imagery were obtained from other sources and carefully examined for clear evidence of lake breeze occurrence.

lake breeze frequency to be approximately 75% for each of Lake Huron, Lake Erie, and Lake St. Clair. The results from this study conclude that summertime lake-breeze circulations encompassing the GTA are much more prevalent than previously reported.

It is unsurprising that the manual method identified more lake breeze events than the algorithm developed by Laird et al. (2001). As discussed in section 2.3, the algorithm method will not identify lake breeze events that developed in a moderate to strong synoptic regime (wind speeds > 5.5 m s^{-1}), whereas the more comprehensive manual approach can distinguish lake breezes under all synoptic regimes. Therefore we can interpret the lower occurrence of lake breeze events identified by the algorithm as a result of false negatives in the algorithm method. Nonetheless, there were also several days which were identified as lake breeze days using the algorithm but not the manual method (i.e. false positives). Therefore, it is advantageous (albeit more time-consuming) to use a manual approach to identify lake breeze events given that the algorithm method is an underestimate.

3.2. Ozone concentrations

Fig. 4 compares the average hourly O_3 levels on lake breeze days (dashed lines) versus non-lake breeze days (solid lines) as a function of time of day and is coloured by air quality station. The daily maxima of O_3 in the mid-afternoon are consistent with higher temperatures and increased solar insolation which are favourable for high levels of O_3 (Jacob et al., 1993). The afternoon O_3 mixing ratios are significantly higher on lake breeze days than non-lake breeze days, with this trend being more pronounced when the manual method is used to identify lake breeze events. This likely stems from false positives where the algorithm method classified the passage of rain showers and thunderstorms as lake breezes. Also, some lake-breeze circulations are not detected with the algorithm method which would further dampen the differences between lake breeze and non-lake breeze days. This reinforces the importance of using a robust (manual) method when studying the influence of lake breezes on air quality.

The average maximum O_3 mixing ratio on lake breeze days versus non-lake breeze days for both methods, as well as the percent increase in maximum O_3 on lake breeze days, is shown in Table 3. Average maximum O_3 increases by 22–28% and 42–49% on lake breeze days for the algorithm and manual methods, respectively. Furthermore, "extreme" O_3 episodes only occur on lake breeze days: not once over this 3-year study period was the Ontario Ambient Air Quality Criteria O_3 standard (1-h average of 80 ppb) exceeded on a non-lake breeze day. The standard was only exceeded during lake-breeze circulations and occurred on 30 different days throughout the study period. The ubiquitous O_3 increase across all sites on lake breeze days suggests a strong correlation between lake-breeze circulations and high O_3, consistent with numerous reports in the literature (e.g. Hastie et al., 1999; Lennartson and Schwartz, 2002; Hayden et al., 2011). These studies identified several mechanisms that could enhance O_3 mixing ratios as a result of lake-breeze circulations: (1) increased solar insolation due to suppressed cloud formation, (2) decreased mixing height, (3) entrainment of pollutants in a recirculating pattern, and (4) transport of O_3 rich-air from over the lake. To examine these mechanisms, the meteorology on lake breeze days must be compared to meteorology on non-lake breeze days. For conciseness, from this point on we only consider results obtained using the manual method since it is a more comprehensive method for identifying lake breezes as discussed previously in this section.

Table 3
Average hourly maximum O_3 mixing ratios (ppb) on lake breeze and non-lake breeze days, as well as the percent increase on lake breeze days relative to non-lake breeze days.

Site	Algorithm method			Manual method		
	LB days	Non-LB	% Increase	LB days	Non-LB	% Increase
BUR	47.6	38.5	24%	47.2	33.0	43%
TDT	46.6	38.3	22%	46.6	32.1	45%
TON	45.3	36.9	23%	45.3	30.5	49%
NEW	48.6	38.4	27%	47.4	33.3	42%
OSH	46.6	36.5	28%	45.5	31.9	43%
Average	*46.9*	*37.7*	*24%*	*46.4*	*32.2*	*44%*

Fig. 5 shows the time of day averages for wind speed, solar insolation, and air temperature on lake breeze days (dashed lines) and non-lake breeze days (solid lines). Average wind speed is about 30% lower on lake breeze days. These relatively stagnant conditions favour the formation of lake-breeze circulations but can also increase pollutant levels in urban areas due to reduced ventilation. Lake breeze days have both higher average mid-day solar insolation (by ~50%) and air temperatures (up to 6 °C). These factors create a larger land-lake temperature difference and hence promote the formation of lake breezes while simultaneously contributing to faster O_3 production.

3.3. Evidence for a causal link between O_3 and lake breezes

Clearly the relationship between meteorology, lake breezes and air quality is complicated, and it is difficult to identify the causes of higher O_3 observed during lake breeze events. Nonetheless, Reid

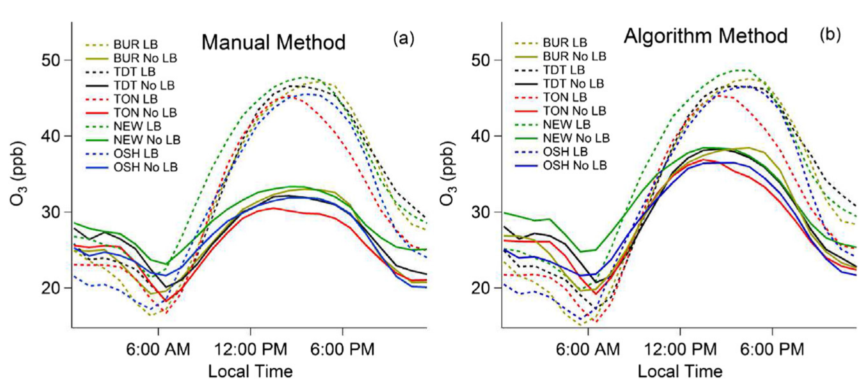

Fig. 4. Time of day plots for average O_3 mixing ratio on lake breeze days (dashed lines) and non-lake breeze days (solid lines). Colours represent different air quality stations explained in Table 1. Lake breeze days were identified using both the (a) manual method and (b) algorithm method. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

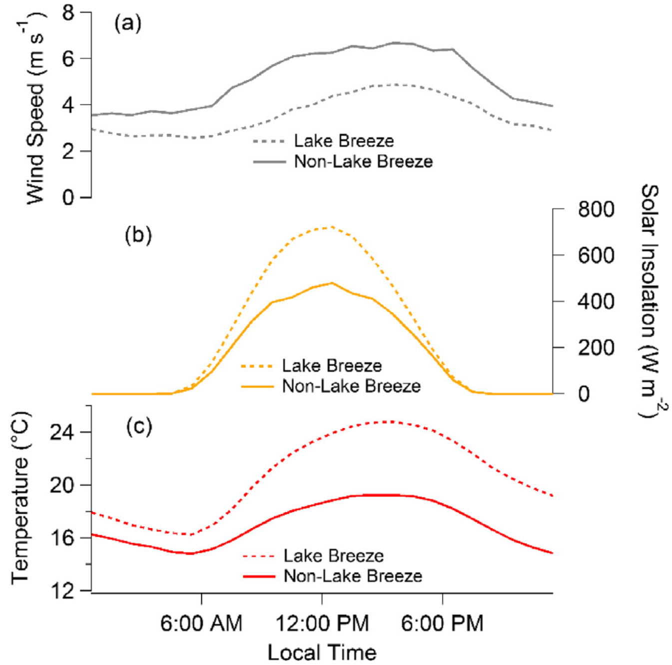

Fig. 5. Time of day plots for average (a) wind speed, (b) solar insolation and (c) air temperature on lake breeze (dashed lines) and non-lake breeze (solid lines) days as determined via the manual method. Wind speed and air temperature data are from the YYZ meteorology station, and solar insolation is from the UTM site.

et al. (1996) and Hastie et al. (1999) focused on several case studies (days) where the unambiguous arrival of a lake-breeze front at specific locations near Lake Ontario is associated with a sharp increase of O_3 (up to 30 ppb in an hour). This is strong evidence for the advection of O_3-rich air at the lake-breeze front leading to observations of high O_3 at near-shore monitoring sites. This paper aims to further explore this link by identifying days over the 3-year study period where some air quality sites are definitively *inside* the lake-breeze circulation and comparing their O_3 mixing ratios to sites definitively *outside* the circulation. The proximity of the air quality sites means they will be subject to the same synoptic scale features that dictate long-range transport and meteorology (temperature and solar radiation). Therefore, any differences in O_3 mixing ratios between sites inside and outside the lake-breeze circulation are most likely attributable to the presence of the lake-breeze circulation.

For the purposes of this analysis, the unambiguous arrival of a lake-breeze front was determined by a sudden (within an hour) wind shift from offshore to onshore during the daytime. Hence, sites that exhibit this wind shift are within the circulation (i.e. within the solid blue arrows in Fig. 1.), whereas those that maintain an offshore flow throughout a lake breeze day are outside the circulation. Examples of five days where only some sites are unambiguously within a lake-breeze circulation are shown in Fig. 6. On 7 July 2010 (Fig. 6a) there is a clear lake-breeze front passage at WWB (10:00), YTZ (11:00), YYZ (11:00) and YOO (11:00) as judged from sudden shifts in offshore to onshore wind direction. However, a lack of wind shift at YKZ implies that NEW remains outside the lake-breeze circulation since NEW is further inland than YKZ. NEW is the only air quality site that does not experience O_3 above 60 ppb during the day, contrary to the other four sites (within the circulation). In addition, the wind direction shifts back to offshore at WWB later in the day (16:00) indicating the lake-breeze circulation no longer encompasses BUR which coincides with a 29 ppb drop in ozone. At around the same time thunderstorms were initiated

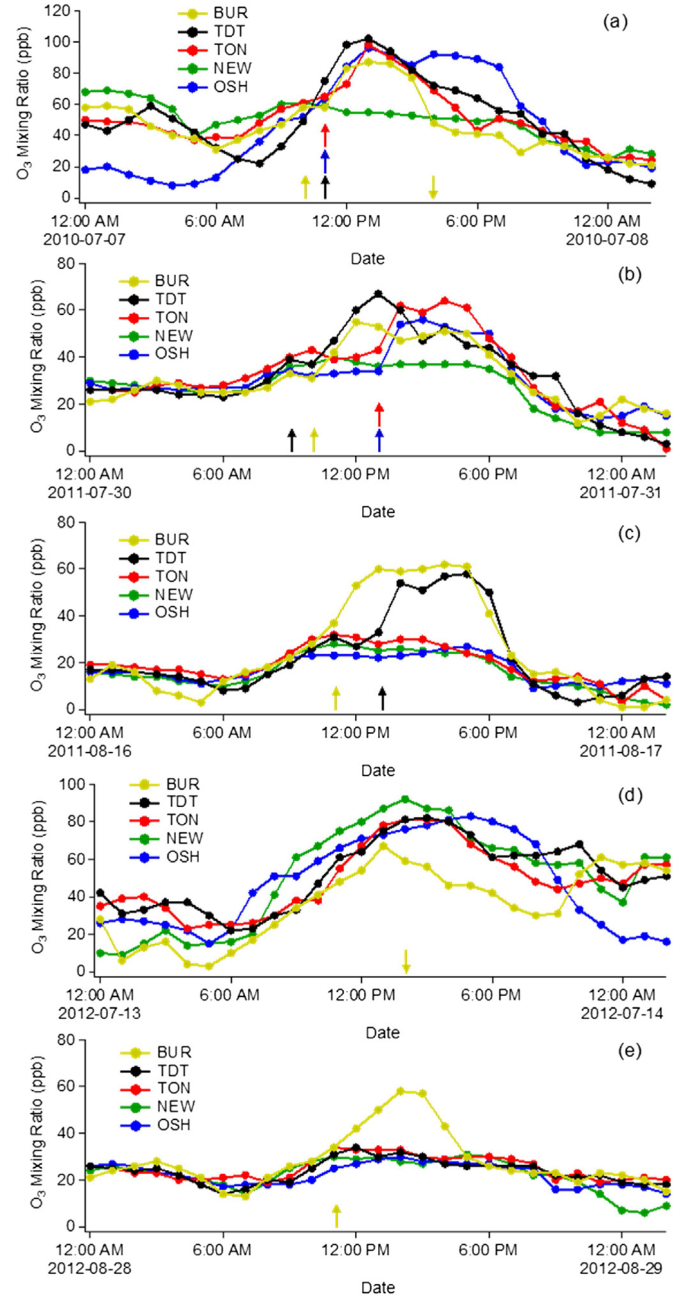

Fig. 6. O_3 mixing ratios measured at each air quality station represented by a different colour on (a) 7 July 2010, (b) 30 July 2011, (c) 16 August 2011, (d) 13 July 2012 and (e) 28 August 2012. Upward and downward arrows (coloured by station) indicate the timing of on-shore and offshore wind shifts, respectively. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

along the lake-breeze front which likely affected pollutant concentrations further.

Similar O_3 increases upon arrival of a lake-breeze front are seen on 30 July 2011 (Fig. 6b). Onshore wind shifts occur at YTZ (9:00), WWB (10:00), YYZ (13:00) and YOO (13:00) but not at YKZ. Again NEW is outside of the lake-breeze circulation and is spared from high O_3 levels. In contrast to 7 July 2010 (Fig. 6a), the front on 30 July 2011 moves more slowly and substantial O_3 increases are not seen at OSH and TON until the front passes, despite meteorology that is favourable to O_3 formation. A stronger northerly (offshore) synoptic flow on 16 August 2011 (Fig. 6c) causes the lake-breeze front to be slower yet and only reach two sites (BUR and TDT). Onshore wind shifts occur at WWB (11:00) and YTZ (13:00) but not at sites further inland. In this case, TON, OSH and NEW are all outside the lake-breeze circulation (confirmed by the analysis in Fig. 3) and O_3 mixing ratios remain below 30 ppb despite equivalent temperatures and cloud conditions to WWB and TDT.

On 13 July 2012 (Fig. 6d) on-shore wind shifts have occurred at all sites by 10:00 suggesting the lake-breeze front quickly penetrates inland. The synoptic regime in the morning is a light westerly flow which intensifies throughout the day and causes an offshore wind shift at WWB (14:00). The lake-breeze circulation dissipates at WWB and a sudden decrease in O_3 is observed at BUR. On the other hand, a moderate north-westerly synoptic flow on 28 August 2012 (Fig. 6e) prevents a lake-breeze circulation on the north-western shore of Lake Ontario and winds are offshore for YTZ, YYZ, YKZ and YOO throughout the entire day. However, an onshore wind shift at WWB (11:00) indicates a lake-breeze circulation forms near Burlington, where the shoreline orientation is north-south. As a result, the only air quality site afflicted with O_3 higher than 34 ppb is BUR which reaches 58 ppb.

The five days depicted in Fig. 6 show that sites within a lake-breeze circulation have higher O_3 than nearby sites outside the lake-breeze circulation which suggests a causal link between elevated O_3 measured at inland sites and the presence of lake-breeze circulations. While this provides strong evidence that air masses advected from over the lake have high ozone, it does not identify what factors are contributing to the higher ozone (e.g. different precursor concentrations, changing rates of oxidation, reduced deposition). We do not have access to any datasets that allow us to evaluate the influence of lake-breeze circulations on mixing heights over the GTA. The role of reduced ventilation cannot be assessed either. Similar cloud conditions at sites within and outside the lake-breeze circulations on days in Fig. 6 suggest that, in these instances, increased insolation behind the front is not the primary cause of high O_3 inside the circulation.

There is evidence of advection of O_3-rich air from over Lake Ontario since BUR and TDT experience sharp O_3 increases and are situated so close to the shore. Indeed, early studies proposed enhanced O_3 over Lake Michigan as a result of a very shallow inflow layer as well as reduced dry deposition due to the relative insolubility of O_3 (Sillman et al., 1993; Dye et al., 1994). More recently, high O_3 has been both observed and modelled over Lake Michigan (Foley et al., 2011), Lakes Erie and St. Clair (Makar et al., 2010), and Chesapeake Bay (Goldberg et al., 2014; Loughner et al., 2014). These studies also highlight the importance of land breezes that can persist in the early morning, which advect rush hour emissions of O_3 precursors (NO_x and VOCs) over the lake. As solar insolation increases during the morning these precursors begin to rapidly form O_3 over the lake in a very shallow boundary layer with reduced deposition. If a lake breeze forms, this O_3-rich air is then advected back onshore which causes a sharp increase in O_3. It is also possible that O_3-rich air from aloft (above the nocturnal boundary layer) subsides in the return flow and further increases O_3 mixing ratios over the lake (Sills, 1998).

3.4. NO_x concentrations

Time of day plots for average NO_x (Fig. 7) on lake breeze and non-lake breeze days reveal a distinct diurnal pattern. NO_x has a lifetime on the order of hours and is primarily emitted from combustion processes which are abundant across the GTA. A large morning peak around 7:00 is a result of morning rush hour traffic. NO_x mixing ratios during morning rush hour are higher on lake breeze days, but since the lake breeze circulation pattern is not present this early in the day, it is likely because of the stagnant

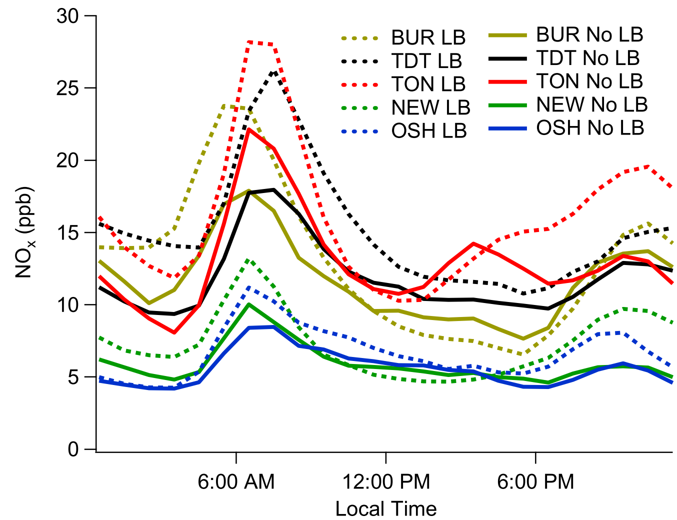

Fig. 7. Time of day plots for average NO_x mixing ratios on lake breeze days (dashed lines) and non-lake breeze days (solid lines). Each air quality site is denoted by a different colour. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

conditions that favour lake breeze formation. Late morning and mid-afternoon mixing ratios are similar on lake breeze and non-lake breeze days suggesting that lake-breeze circulations have little effect on inland NO_x levels during these periods. In the late afternoon and early evening, sites further inland (TON, OSH and NEW) experience significantly more NO_x on lake breeze days – this is likely a consequence of being downwind from heavily populated near-shore urban centres on lake breeze days. The lack of difference in NO_x on lake breeze days at near-shore sites (BUR and TDT) implies there is not a significant enhancement of NO_x over the lake. When examining the NO_x data from the case study days from Fig. 6, there are occasions where NOx increases during lake breeze front passages, but typically the signal is not noticeable, especially compared to the strong signals in ozone. However, Levy et al. (2010) did observe a sharp increase in NO_x during the passage of a lake-breeze front in the Lake Erie region and attributed this rise to an accumulation of NO_x over the lake during the previous night.

4. Conclusions

This study reports, for the first time, the impact of summertime lake-breeze circulations on pollutant (O_3 and NO_x) levels in GTA using a multiyear dataset. Observations from surface meteorological stations were employed to identify days impacted by lake breezes from Lake Ontario using an algorithm from Laird et al. (2001). A manual method from Sills et al. (2011) using data from multiple platforms (surface stations, satellite and radar) was also employed to detect lake breezes. The former method computed lake breezes on 56% of the summer (May to September) days from 2010 to 2012, whereas the manual method determined the frequency to be 74%. These values are much higher than previous estimates for summertime Lake Ontario, which range from 25 to 32%. The algorithm method is less reliable since it underestimates lake breeze occurrences in moderate-to-strong onshore synoptic regimes and is subject to false positives caused by other atmospheric phenomena (i.e. frontal passages, wind shifts due to showers or thunderstorms).

Hourly averaged concentrations of O_3 and NO_x from five sites across the GTA were used to compare pollutant levels on lake breeze days versus non-lake breeze days. Using the manual method, the average daily 1-h maximum of O_3 was found to be 42–49% higher on lake breeze days at all five air quality sites. Corresponding meteorological measurements show that conditions favourable for high O_3 (lower wind speeds, higher temperatures, and increased insolation) are present on lake breeze days, since they are also conducive for lake breeze formation. A detailed analysis revealed several lake breeze days where only some sites were located within the circulation. Sites within the circulation experienced higher O_3 (up to 30 ppb) than sites outside the circulation suggesting that the high levels of ozone can be attributed to the presence of lake-breeze circulations. There is likely O_3-enriched air over Lake Ontario that is being advected onshore via the lake breeze since near-shore sites (BUR and TDT) experience sharp increases in O_3 with the arrival of a lake-breeze front. Mechanisms responsible for high O_3 over Lake Ontario could be: (1) reduced mixing height, (2) reduced deposition, (3) night-time land-breeze circulation that fumigates the lake with O_3 precursors before sunrise, and/or (4) subsidence of O_3-rich from aloft (over both land and lake) in the return flow.

NO_x mixing ratios on lake breeze mornings are enhanced, likely due to the stagnant conditions favourable for lake breeze formation. However, afternoon mixing ratios are comparable on lake breeze and non-lake breeze days at each site. Late afternoon levels on lake breeze days are enhanced relative to non-lake breeze days at inland sites only (TON, NEW and OSH). This is caused by onshore winds from lake-breeze circulations which transport NO_x emissions from near-shore city centres further inland.

Lake breezes are common summertime phenomena in the GTA and contribute to increased O_3 mixing ratios, but exhibit a much smaller influence on NO_x. In the context of the GTA, the link between lake-breeze circulations and air quality (specifically O_3 levels) has been underappreciated in the literature. Future studies should focus on determining the exact mechanisms which lead to high O_3 over the lake. Measurements of ozone and its chemical precursors over the lake would be beneficial for this type of analysis. Furthermore, this analysis suggests that chemical transport models need to accurately represent lake-breeze circulations, including the details of the lake-breeze front, to improve air quality predictions in the region.

Acknowledgements

The authors are grateful to Environment Canada, the Ontario Ministry of the Environment and the Geography Department at University of Toronto Mississauga for providing the meteorology and air quality data sets. Assistance with the Aurora workstation was provided by Emma Hung at Environment Canada. G.R.W. acknowledges funding from the NSERC Integrating Atmospheric Chemistry and Physics from Earth to Space (IACPES) program. The authors wish to thank D. J. Donaldson and J. P. D. Abbatt for useful discussions and feedback.

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