Statistical quantification of COVID-19 lockdown effect on air quality from ground-based measurements in Ontario, Canada

Hind A. Al-Abadleh,[ξ]* Martin Lysy,[‡]* Lucas Neil,[§] Priyesh Patel,[ξ] Wisam Mohammed,[ξ] and Yara Khalaf[ξ]

[ξ] Department of Chemistry and Biochemistry, Wilfrid Laurier University, Waterloo, Ontario N2L 3C5, Canada. [‡] Department of Statistics and Actuarial Science, University of Waterloo, Waterloo, Ontario N2L 3G1, Canada. [§] Hemmera Envirochem Inc., Oakville, Ontario L6J 7W5, Canada.

ABSTRACT

Preliminary analysis of satellite measurements from around the world showed drops in nitrogen dioxide (NO_2) with lockdowns due to the COVID-19 pandemic. A number of studies have found these drops to be correlated with local decreases in transportation and/or industry. None of these studies, however, has rigorously quantified the statistical significance of these drops relative to natural meteorological variability and other factors that influence pollutant levels during similar time periods in previous years. Here, we develop a novel statistical testing framework that accounts for seasonal variability, transboundary influences, and new factors such as COVID-19 restrictions in explaining trends in several pollutant levels at 16 ground-based measurement sites in Southern Ontario, Canada. We find statistically significant and temporary drops in NO_2 (11 out 16 sites) and CO (all 4 sites) in April-June 2020, with pollutant levels 20% lower than in the previous three years. Much fewer sites (2-3 out of 16) experienced statistically significant drops in O_3 and PM2.5. The statistical testing framework developed here is the first of its kind applied to air quality data, and highlights the need for rigorous assessment of statistical significance, should analyses of pollutant level changes post COVID-19 lockdowns be used to inform policy decisions in Ontario, Canada.

INTRODUCTION

The province of Ontario in Canada declared a state of emergency on March 17, 2020 in an effort to limit the spread of COVID-19, which caused the first related death in mid-March 2020. As a result, lockdown restrictions affected the majority of workplaces, which shifted to working from home, including schools and universities, and the closure of recreational and shopping facilities that gather large numbers of people. Table S1 lists the timeline of restrictions in Ontario, the state of Michigan in the U.S., which borders the southwestern part of the province, and Ohio, which can influence pollution levels in Ontario via transboundary movement of pollutants. The imposition of the lockdown measures drastically reduced traffic, aviation and industrial activity in the province as reported from satellite analysis.[1] Satellite data for nitrogen dioxide (NO_2) column using the Tropospheric Monitoring Instrument (TROPOMI) operated by NASA and European Space Agency were analyzed for the Greater Toronto area, home to Ontario's capital and Canada's most populous urban region.[1] The analysis showed drastic reduction in NO_2 levels by roughly 40% relative to pre-lockdown. This reduction is similar in magnitude to those reported in cities in China, Europe and the United States during their respective lockdowns and/or states of emergency.[2] Comparisons of data in 2020 were made to the same period in 2019 to quantify the drop in NO_2 levels since weather and seasonal changes also affect the levels of these pollutants.[3] Griffin *et al.*[1] estimated a 20% reduction in satellite-measured NO_2 attributed to meteorology in Toronto. Analysis of satellite and ground-based (i.e., surface) measurements of pollutant levels pre- and post-COVID-19 closures was also reported for different cities from around the globe (see updated list of papers in reference [4]). In the Supporting Information, we highlight a few examples from Bejing, Wuhan, and Northern China[5,6], the City of Pittsburgh, Pennsylvania in the U.S.[7], and over 10,000 air quality stations in 34 countries.[8] However, none of these studies has undertaken a rigorous quantification of the statistical significance associated with these findings, in that such quantification is either absent or heavily reliant upon modeling assumptions which cannot be verified.[8] This raises serious concerns over causality conclusions about the potential lockdown effect, and highlights the considerable challenge in disentangling the contribution of short-term seasonal effects and natural variability in atmospheric chemistry from the observed reduction in pollutant levels when comparing pre- and post- lockdown data.[9]

Indicator pollutants of air quality in Ontario are monitored by a network of 39 stations across the province maintained by the Ministry of the Environment, Conservation and Parks (MECP).[10] These pollutants include nitrogen oxides (NO_x), CO, O_3 and PM2.5. Sources of NO_x are closely associated with combustion. In 2016, 69% of NO_x originated from road vehicles and other transportation in Ontario.[10] Seasonal variations of NO_2 levels are observed with maximum levels occurring in the winter and minimum levels observed in the summer. The seasonal NO_2 signature can be attributed to seasonal fluctuations in the boundary layer height. In general, wind speeds increase in spring and summer as the height of the boundary layers increases, which enhances dispersion and lowers concentrations.[11] Also, NO_2 is a photoactive molecule that dissociates to NO and O and hence, contributes to ground-level O_3 formation. Another by-product of incomplete combustion of fossil fuels is CO. Similar to NO_x, the transportation sector accounts for 71% of all CO emissions in Ontario[10], and as much as 95% of all CO emissions in metropolitan areas in the US.[12] Seasonal variations of CO levels mimic those of NO_2, with maximum levels occurring during late winter and minimum levels observed during late summer.[12] This seasonal trend is the result of inversion conditions being more frequent during winter months than summer months. The major source of ground-level O_3 is secondary processes from the photochemical reaction of NO_x and volatile organic compounds (VOCs). Transportation and general solvent use account for 43% of

VOCs emissions in Ontario.[10] As a result of its formation chemistry, concentrations of ground level O_3 are highly variable on an hourly, daily, seasonally, and yearly basis. The scavenging effect of NO reduces local O_3 levels in urban centres in Ontario, especially during summer months. Over the 10-year period from 2007 to 2016, progressive reduction in NO_x emissions in Ontario and the US resulted in a decrease in the summer means of local O_3. Still, ground level O_3 in the summer continues to exceed the Ontario Ambient Air Quality Criteria (AAQC) of 80 ppb (1-hr), particularly in Southern and Eastern Ontario. As for PM2.5, residential sources account for 56% of all sources by sector from fuel wood combustion in fireplaces and wood stoves, followed by industrial (21%) and transportation sectors (12%).[10] Together, O_3 and PM2.5 drive smog episodes in May-September in Ontario, which are affected by local and regional weather patterns and long-range transboundary influences from industrial and urbanized US states.

The objective of this investigation is to rigorously quantify the statistical significance of changes to air quality indicators from ground-based measurements in Southern Ontario as a result of the COVID-19 restrictions. To this end, we develop a novel statistical testing framework, which accounts for seasonal variability, transboundary influences, and new factors such as COVID-19 restrictions in explaining trends in the levels of NO_2, CO, O_3 and PM2.5. Importantly, our quantification of statistical significance makes minimal modeling assumptions about the data. We expand on the relevance of this framework to the analysis of a one-time event such as the lockdown in the Methods section and Supporting Information, and on implications for policy-making in the Discussion.

METHODS

Data acquisition

For the selected sites in this paper, ground-based hourly data of pollutant concentrations were downloaded from the MECP website (http://www.airqualityontario.com). More details on data quality are available in the Supporting Information. Hourly and daily meteorological data collected by the Meteorological Service of Canada network of stations were obtained from the National Climate Archives website (https://climate.weather.gc.ca/historical_data/search_historic_data_e.html). Solar irradiance monitoring data were obtained by contacting the surface weather observation network maintained by Environment and Climate Change Canada (ECCC).

Statistical information.

Our novel statistical protocol hinges on the assessment of statistical significance via the following randomization test.[13,14] Suppose that N_{post} daily pollutant concentrations are recorded post-lockdown and N_{pre} daily pollutant concentrations are recorded pre-lockdown under similar conditions. Specifically, in our analysis, N_{post} corresponds to the weekdays of a given month – say April 2020 – whereas N_{pre} corresponds to the weekdays of the same month in the three reference years April 2017-2019. By comparing the same month across years, we account for natural variation in seasonal meteorology. We eliminate weekends from our analysis since COVID-19 restrictions affect traffic activity on weekdays and weekends quite differently.

Suppose that each of the N_{pre} and N_{post} daily pollutant concentrations come from an independent and identically distributed (iid) sample. Under the null hypothesis H_0 that there is no pre/post-lockdown difference, every permutation of the $N_{\mathrm{pre}} + N_{\mathrm{post}}$ observations into groups of size N_{pre} and N_{post} is equally likely. Moreover, a random permutation should produce a difference in medians Δ_{rand} which is not too far from Δ_{obs}, the difference in medians recorded from the actual data. Thus, the p-value against H_0 is the probability of Δ_{rand} being greater than Δ_{obs} with all random permutations being equally likely. This probability can be estimated to arbitrarily high precision by Monte Carlo simulation, i.e., by reporting the fraction of times Δ_{rand} exceeds Δ_{obs} on a large number M of random permutations (all of our p-values are calculated with $M = 10000$, thus having a Monte Carlo standard error of no more than 0.005).

The randomization test described above is nonparametric, making no modeling assumptions other than the lockdown data and the reference year data both originating from iid samples. Moreover, the resulting p-value calculation is exact, in contrast to most statistical tests for which the p-value is only valid asymptotically for large pre- and post-lockdown samples.

To the best of our knowledge, this is the first application of such a randomization test to air quality data. It is also worth noting that randomization tests such as ours do not necessarily rely on assumptions about iid sampling or other elements of a statistical model, which can be especially advantageous for the analysis of one-time events such as COVID-19. Additional explanation for both points is provided in the Supporting Information.

Data availability. The data that support the findings of this study are available from the corresponding authors upon reasonable request.

Computer code. A self-contained library written in the R programming language documenting all p-value and boxplot calculations are available from the corresponding authors upon request.

RESULTS

Assessing variation in temperature and solar irradiance in 2017-2020. The overlap of seasonal variations in the concentrations of NO_2, CO, O_3 and PM2.5 with measures enforced by the Ontario government to limit the spread of COVID-19 complicated the assessment of reductions associated with reduced traffic, aviation, and industry emissions. Figure 1 shows the locations of the air quality stations, meteorology and solar irradiance stations that collect hourly data on pollutant levels, temperature and radiative forcing, respectively. To assess meteorological changes in 2020 relative to reference years, 2017-2019, Figure S1 shows box plots of daily mean temperature for three locations selected based on their type (rural versus urban) from January until June. Below each box in the plot is the p-value value calculated from the randomization test described in the Methods section. The set of p-values on the right test whether there is a statistically significant difference between the median monthly temperature in 2020 compared to 2017-2019.

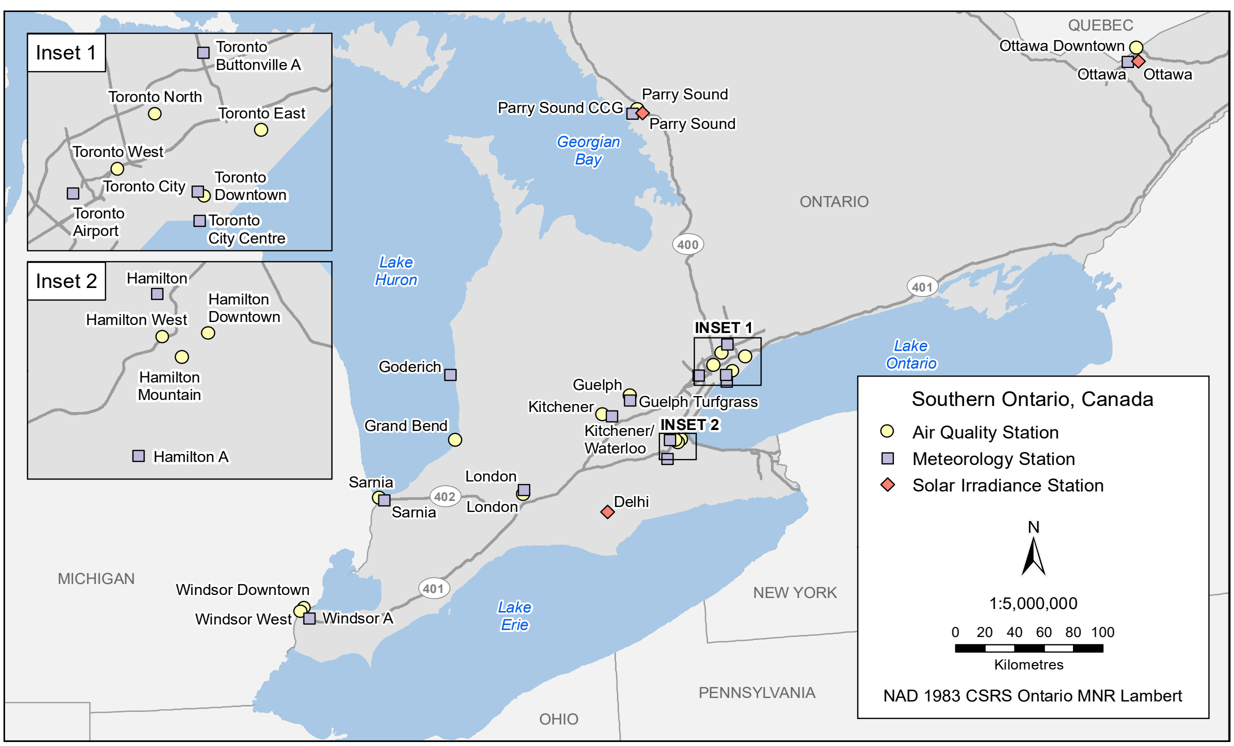


Figure 1: Map of Southern Ontario showing locations of air quality stations maintained by MECP, national meteorological and irradiance stations maintained by ECCC.

For March 2020, the p-value is less than 0.05 for all sites, suggesting that a potential lockdown effect on air pollutants might be masked by unusually high temperatures relative to the reference years. On the other hand, $p > 0.05$ for April, May, and some sites in June 2020. These data suggest that temperature was not significantly different in 2020 compared to reference years, and therefore does not confound pollutant concentration months when the lockdown is both in full and waning force. The other set of p-values below the boxes to the left side tests the difference between medians in the reference years (a generalization of the randomization test above to more than two samples is provided in the Supporting Information). The p-values for February, March, and June are all greater than 0.05, indicating that 2017-2019 median temperatures were not statistically different during those months. In contrast, the corresponding p-values for January, April, and May 2017-2019 are well below 0.05. Since the months within these reference years did not experience the lockdown effect, the low p-value indicates that there is considerable natural variation in seasonal meteorology during these months, making it difficult to detect the specific impact of COVID-19 in 2020.

As shown in Figure 1, the two stations in Southern Ontario for measuring solar irradiance are in Ottawa and Delhi. Figure S2 shows the daily solar global horizontal irradiance (GHI) at these locations from January till June between 2017-2020. GHI values were obtained from the measured radiation field. In this case, the p-values are all generally greater than 0.05, indicating that there is little difference in solar irradiance between these years.

Assessing variation in pollutant levels. Figure 1 shows the locations of selected air quality stations in Southern Ontario that collect hourly data on pollutant levels analyzed here. Each air quality station measured hourly levels of NO_2, O_3 and PM2.5. Only four out of the sixteen stations reported CO measurements: Hamilton Downtown, Ottawa Downtown, Toronto West, and Windsor

Downtown. The MECP's rationale behind choosing these sites for CO measurements is that Hamilton and Windsor are in the top five of the most polluted cities in Ontario. Toronto West station is near the busiest highway in North America, Hwy 401.[15] Ottawa was likely chosen because it is the nation's capital city and is technically located in Eastern Ontario, further away from the US border with little industrial activity. The next few sections describe the variation in pollutant levels at different resolutions: hourly, daily, weekly and monthly in order to show how data resolution affects the type of conclusions that can be made. As detailed above, the statistical approach we developed here aims at quantifying the significance in the difference between median pollutant levels of weekdays (no weekends) per month in 2020 and the previous three years, 2017-2019, used as reference.

Variation in NO_2 levels. Figure S3 shows the diurnal average levels of NO_2 in April over 2017-2019 and 2020 for Grand bend (rural), Kitchener (urban), and Toronto West (urban). These data are superimposed with solar irradiance and average hourly temperature for each location. April was chosen because it followed two weeks of COVID-19 lockdown measures in Ontario. As a photoactive molecule, the data show a reduction in the NO_2 levels with increasing solar irradiance, which peaks around 12:00-13:00. Overall, the concentrations of NO_2 in Grand Bend range from 1.5 – 3 ppb, Kitchener from 2.5 – 11 ppb, and from 5 – 22 ppb for Toronto West, which peak around 06:00 during the morning rush hour. While the average data in Figure S3 show lower diurnal NO_2 levels in 2020 compared with the average data in 2017 – 2019, standard deviation calculations ($\pm 1 \sigma$) revealed extensive overlap between the two cases (see shaded areas). Based on this data, we could not conclude that the reduction observed in April 2020 is statistically significant relative to 2017-2019.

We then calculated the daily NO_2 levels for each station from January until June in 2020 for comparison with the daily average of each month from 2017-2019. Figure S4-S6A show selected data for the same locations in Figure S3. The values of the standard deviation were removed for clarity. The trends in the daily NO_2 concentrations over a five-month period shows a great degree of overlap between the 2020 and average 2017 – 2019 data. Also, these data show the seasonal reduction in NO_2 in the spring months compared to winter. The start of the COVID-19 lockdown in March 2020 is marked in these Figures. There is no clear evidence that additional reductions in daily NO_2 levels were observed in the daily values in 2020 compared with the average daily values in reference years in any of the stations we analyzed. We then looked at median values of NO_2 levels for weekdays only (no weekends) for all weeks from January until the end of June, per year in 2017-2020. Figures S4-S6B show selected data from this type of analysis for the same stations in Figure S3. The median of weekdays analysis did not reveal clear reduction in NO_2 levels in the weeks after the COVID-19 lockdown either.

Following the hourly, daily and weekly analyses described above, the weekdays distribution in NO_2 levels in a given month in 2020 and in reference years was graphically analyzed using box and whisker plots. Figure S7 shows representative plots for the three air quality stations shown in Figure S3. The p-values for March 2020 are all statistically insignificant, perhaps linked to unusually high temperatures during this month. On the other hand, many stations recorded drops in NO_2 concentrations below the 0.05 significance level in April-June. Of particular note is Toronto West in April 2020, for which a significant drop was reported despite the statistically significant differences between the reference years. In other words, the difference between April 2020 and the reference years is large, even compared to the considerable seasonal variability of pollutant levels which naturally occurs during the month of April. As presented in the following sections, the weekdays median values for NO_2 were used to calculate the percentage difference in 2020 relative to the reference years, 2017-2019, and also to calculate the p-values used to quantify the statistical significance of the percent difference.

Variation in CO levels. Figure S8 shows box and whisker plots for the weekday distribution in CO levels in four air quality stations, Hamilton Downtown, Ottawa Downtown, Toronto West and Windsor Downtown. These are all urban stations, each of them having a significantly lower median CO value in April 2020 than in the reference years. There is also some evidence that the lockdown is easing, with many p-values above 0.05 in May-June 2020. Similar to NO_2, the weekdays median values for CO were used to calculate the percentage difference in 2020 relative to the reference years, 2017-2019.

Variation in O_3 and PM2.5 levels. Figure S9 shows box and whisker plots for the weekday distribution in the concentrations of O_3 and PM2.5 over 2017-2019 and 2020 for the sites that experienced statistically significant drops in each pollutant per Table 1. For O_3, the sites shown in the figure are Sarnia and Windsor West, with Toronto West added for comparison given its proximity to Hwy 401. For PM2.5, the sites shown in the figure are Hamilton West, Ottawa Downtown and Windsor Downtown. The raw data show the seasonal changes in O_3 levels for these selected sites that increase in spring and summer months. The apparent trend in PM2.5 levels is a narrower distribution of data points in May and June compared to earlier months for all years, and in 2020 in general, compared to reference years, 2017-2019. Similar to NO_2 and CO, the weekday median values for O_3 and PM2.5 were used to calculate the percentage difference in 2020 relative to the reference years, 2017-2019.

Table 1: Summary of the percentage *decrease* in pollutant levels in 2020 relative to the same period in 2017-2019. The statistically significant values are highlighted in p-values below 0.05 listed in parentheses.

AQ station name [a] R = Rural U = Urban	Pollutant											
	NO_2 (ppb)			CO (ppm)			O_3 (ppb)			PM2.5 (µg m^{-3})		
	COVID-19 related decrease (%) [b]											
	Apr	**May**	**Jun**	**Apr**	**May**	**Jun**	**Apr**	**May**	**Jun**	**Apr**	**May**	**Jun**
Grand Bend (R)	39 (0.01)	n.o.	n.o.	No measurements			n.o. (0)	0.1	n.o.	n.o.	2	n.o.
Guelph (U)	22 (0.05)	22	n.o.	No measurements			n.o.	2	n.o. (0.05)	n.o.	27	n.o. (0.02)
Hamilton Downtown (U)	27	15	38 (0.05)	20 (0.01)	4	10	n.o.	n.o.	n.o. (0.02)	6	15	n.o.
Hamilton Mountain (U)	27	5	28	No measurements			n.o.	3	n.o. (0.04)	19	27	n.o.
Hamilton West (U)	22	21	17	No measurements			n.o.	n.o.	n.o. (0.02)	11	32 (0.03)	n.o.
Kitchener (U)	24	29 (0.03)	39 (0)	No measurements			2	2	n.o.	10	25	n.o.
London (U)	29 (0.01)	20 (0.02)	18 (0.02	No measurements			n.o.	4	n.o.	32	15	n.o.
Ottawa Downtown (U)	16	15	17	18 (0)	14 (0.01)	6	n.o.	n.o.	n.o. (0.02)	n.o.	24 (0.04)	n.o.
Parry Sound (U)	7	14	n.o.	No measurements			n.o.	n.o.	n.o.	15	n.o.	n.o.
Sarnia (U)	42 (0.02)	30	13	No measurements			14 (0)	18 (0)	1	n.o.	2	n.o.
Toronto Downtown (U)	n.o.	n.o. (0.01)	n.o.	No measurements			5	21	n.o.	n.o.	12	n.o. (0.01)
Toronto East (U)	30 (0.01)	22	21	No measurements			n.o. (0.02)	2	n.o. (0.01)	9	18	n.o.
Toronto North (U)	30 (0.03)	7	2	No measurements			n.o. (0.01)	1	n.o. (0.03)	0.4	29	n.o.
Toronto West (U)	27 (0)	21 (0.01)	13	18 (0)	10	2	n.o. (0.01)	n.o.	n.o. (0.03)	n.o.	21	1
Windsor Downtown (U)	17	40 (0)	19 (0.03)	11 (0.03)	17 (0.01)	11	4	n.o.	n.o.	n.o.	26 (0.04)	0
Windsor West (U)	7	40 (0)	11	No measurements			12 (0)	8	n.o.	n.o. (0.03)	n.o.	n.o.

Notes: [a] See Table S2 for station type and Figure S1 for location. [b] % decrease in 2020 in a given month = (median in 2020 – median in 2017-2019)*100% / (median in 2017-2019). See Figures 1,2, 4a-c for examples. 'n.o.' = no decrease observed, on the other hand, an increase was observed in pollutant level in 2020 relative to 2017-2019.

Assessing the variability of pollutant levels within the reference years 2017-2019. Table S2 lists the p-values calculated for the concentration distribution of each pollutant within the three year period 2017-2019. The main assumption is that seasonal factors are the major contributors to the concentration distribution in each year, which are similar over a three year period. The statistical significance test used here resulted in $p < 0.05$ for a number of sites in a given month. Tables S3-S18 list the median values for each pollutant in April – June over 2017-2019. Median values for 2020 are also listed. The median values provide an accurate indication of the similarity between years reflected in the calculations of the p-values. For example, the p-value for May in 2017-2019 for NO_2 levels at Grand Bend station is 0. The median values for NO_2 listed in Table S3 are 2.8, 4.0, and 1.6 for 2017, 2018 and 2019, respectively. Hence, the p-value of 0 indicates that there is considerable natural variation in NO_2 concentration levels from year to year between 2017-2019. Other examples of statistically significant differences over the reference years are highlighted in Table S2 with underlined p-values. When the p-value for 2020 is less than 0.05, this indicates that there is a significant difference between 2020 and the past three years that could be attributed to new factors such as the COVID-19 lockdown restrictions. In the case when p-values for the reference years are also less than 0.05, this indicates that 2020 stands out despite considerable variability among the reference years. This suggests the presence of new unique factors in 2020 that are separate from those causing the difference in pollutant levels among the reference years. This result is different from the scenario where p-values are less than 0.05 for the reference years, but greater than 0.05 for 2020. This result would suggest that seasonal meteorology can account for large differences between years, compared to which the lockdown effect is insignificant.

Assessing the variability of pollutant levels in 2020 relative to the reference years 2017-2019. As detailed in the Methods section, the calculated p-values reflect the degree of similarity in the distribution of daily pollutant levels in 2020 and the reference years: p-values < 0.05 indicate statistically significant difference between the 2020 median weekday levels and those in reference years. The calculated percentage difference, which could indicate increase, decrease, or no change in pollutant levels, can be attributed to new factors other than temperature and solar irradiance because it was calculated for the same monthly period. These factors would include the effect of COVID-19 measures on reducing traffic, aviation and industrial activities. It could also include new residential sources, which increased in contribution due to ‘go home and stay at home’ public health advisories starting in March 2020. Another important factor that has been known to influence air quality in Ontario is transboundary air pollution from the United States that increases the concentration of pollutants studied here. The US did not enforce COVID-19 lockdown measures during the same time period as Ontario. As highlighted in Table S1, ‘stay at home orders’ in Michigan and Ohio were implemented after Ontario and were beginning to be lifted well before Ontario lifted its ‘stay at home’ order. This difference in lockdown enforcement was expected to have a big impact on NO_x and CO levels for stations in the Windsor and Sarnia area, along the US border, which are heavily impacted by transboundary transport, in so much that any impact from COVID-19 lockdown measures would be difficult to disentangle from US sources impacting these sites.

The weekday median values for the pollutants analyzed here were used to calculate the percentage difference in two ways to highlight two cases: In case 1, percentage difference values were calculated for each month in 2020 relative to the corresponding month in the reference years, 2017-2019 (Table 1). This type of calculation assumes that seasonal variability is similar for each

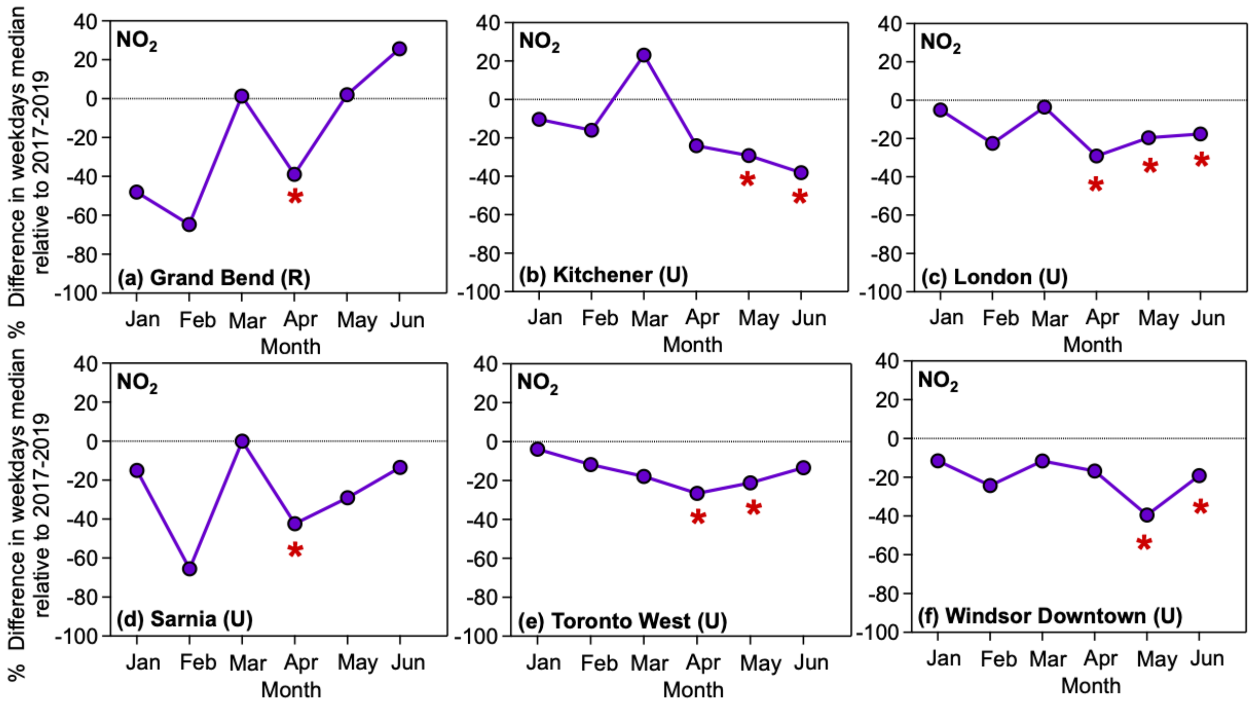


Figure 2: Percentage difference in weekday median levels of NO_2 in 2020 relative to the same period in reference years, 2017-2019 for selected sites. The data for April – June are listed in Table 1 for these sites. The '*' highlight the statistically significant decreases based on the p-values.

month, and hence any statistically significant difference in pollutant levels is due to new factors such as transboundary influences or COVID-19 restrictions. Figure 2 shows graphical representation of the percentage decrease in pollutant levels for selected sites. In case 2, percentage difference values were calculated relative to January in 2020 and in the reference years 2017-2019. Then, if an extra decrease was observed for a given pollutant in 2020 relative to reference years, the difference in the percentages was calculated to quantify that extra decrease as reported in Table S19. This type of calculation shows the magnitude of seasonal changes in each pollutant for 2020 and reference years 2017-2019 relative to their highest levels in January. The assumption here was that new factors that might influence pollutant levels in 2020 beyond seasonal changes will be manifested as either increases or decreases in percentage. Figure S10 shows graphical representation of this extra decrease in NO_2 concentrations for selected sites. Therefore, the calculated p-values were used to quantify the statistical significance of these percentages in cases 1 and 2. For Tables 1 and S19, the data are only shown for April until June since COVID-19 lockdown measures started March 17, 2020 in Ontario. The statistically significant percentages are highlighted in shaded areas based on the p-values listed in parentheses. These p-values are the same as those listed in the monthly 2020 columns in Table S2. Calculated percentages that indicate no change or an increase in pollutant levels were assigned 'n.o.' in an effort to highlight *decreases* attributed to the impact of COVID-19 lockdown measures or other new factors.

Figures 2 and 3 show selected data from Table 1 for selected sites to graphically demonstrate differences in NO_2 and CO changes among different sites over the months in 2020 before and after the COVID-19 measures came into effect. Percentages were calculated in these figures according to case 1 described above. Figures S10 and 4 show the extra decreases observed in 2020 for NO_2 and CO, respectively, for selected sites from percentages calculated according to case 2 described above, which are also listed in Table S19. Transportation sources contribute 69% and 87% of NO_2 and CO emissions in Ontario, respectively.[10] The statistically significant decreases in NO_2 levels occurred in April and ranged from 22-42% depending on the location of the station (Table 1). For example, Figure 2a shows that the rural station, Grand Bend, experienced a 39% reduction in NO_2 levels in April, no change in May, and a 20% increase in June. The p-value associated with the latter percentage is 0.2 (Table S2), and hence the calculated increase in NO_2 June 2020 levels is considered statistically insignificant (i.e., June weekday median levels in 2020 are within the distribution of the corresponding values in June 2017-2019). Urban sites that experienced a statistically significant reduction in NO_2 levels in April 2020 include Guelph (22%), London (29%), Sarnia (42%), Toronto East, North, and West (~30%). Other urban sites experienced a statistically significant reduction in NO_2 levels in May 2020, which include Kitchener (29%), London (20%), Windsor Downtown

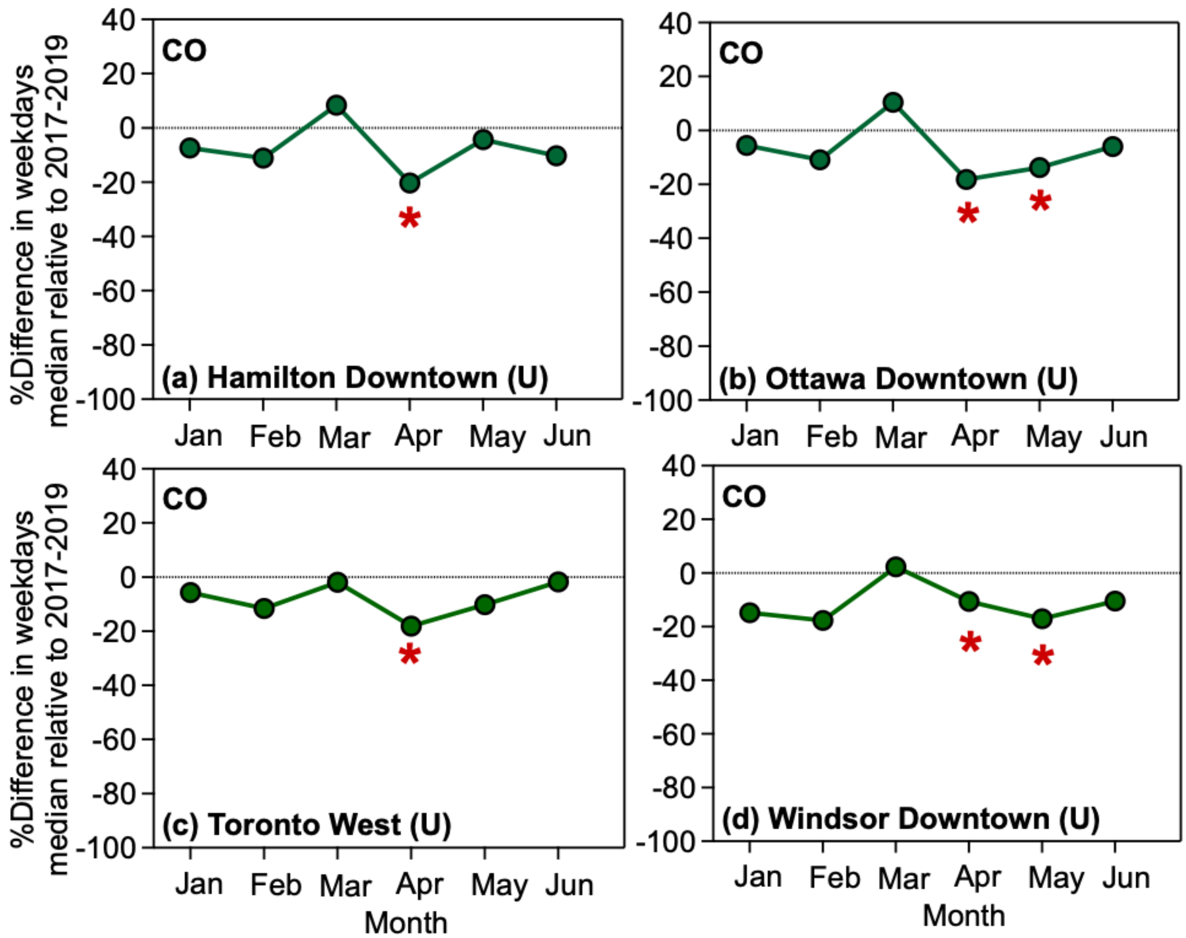


Figure 3: Percentage difference in weekday median levels of CO in 2020 relative to the same period in reference years, 2017-2019 for selected sites. The data for April – June are listed in Table 1 for these sites. The '*' highlight the statistically significant decreases based on the p-values.

(40%), Windsor West (40%), and Toronto West (21%). A few urban sites experienced a statistically significant reduction in NO_2 levels in June 2020, and those include Hamilton Downtown (38%), Kitchener (39%), London (18%), and Windsor Downtown (19%). Our findings underscore the major contributors to the reduction in NO_2 observed by satellite measurements in Southern Ontario.[1]

Moreover, data in Table S19 show that the majority of sites in Southern Ontario experienced a statistically significant 5-28% extra decrease in NO_2 levels in 2020 beyond seasonal variability observed in the same months in 2017-2019. This trend in the data agrees with that shown in Table 1, with the exception of Grand Bend, where the statistically significant drop shown in Figure 2a in

April does not align with that in Figure S10a. The box plot for the NO_2 data in Grand Bend is shown in Figure S7a where there is a clear fluctuation in the median 2020 data over February – June relative to January compared to a progressive decrease in the corresponding data for 2017-2019. Given the location of this site on the Canadian shore of Lake Huron, it is very likely that these fluctuations are due to transboundary influences from Michigan, USA.

The data in Figure 3 for CO levels in different urban sites show a *ca.* 20% statistically significant reduction in April 2020 for Hamilton Downtown, Ottawa Downtown, and Toronto West. Windsor Downtown experienced 11% reduction in April 2020. The statistically significant reduction in CO levels continued in May 2020 for Ottawa Downtown (14%) and Windsor Downtown (17%). All of these urban sites experienced a statistically insignificant reduction in CO in June 2020, which coincided with the second phase of lifting restrictions in Ontario (see Table S1). Moreover, data in Figure 4 show that these sites experienced a statistically significant 2-16% extra decrease in CO levels in 2020 beyond seasonal variability observed in the same months in 2017-2019. This trend in the data agrees with that shown in Figure 3.

The data in Figure 5 for O_3 levels in different urban sites show statistically significant reductions in March - May 2020 for Sarnia and Windsor West, both of which are border cities with Michigan, USA with extensive industrial activity. The reduction observed in March 2020 for these sites of nearly 40%, is higher than that observed for the Toronto West site at 24% (Figure 5e), which is near Hwy 401. For the latter site, the reductions observed in April and May were not statistically significant, suggesting dominance of seasonal factors or other factors that affects the chemistry of ozone production in these sites.[9]

Figure 6 a-c show the variability in PM2.5 levels for the urban sites that experienced a 35-40% statistically significant reduction in May 2020, which are Hamilton West, Ottawa Downtown

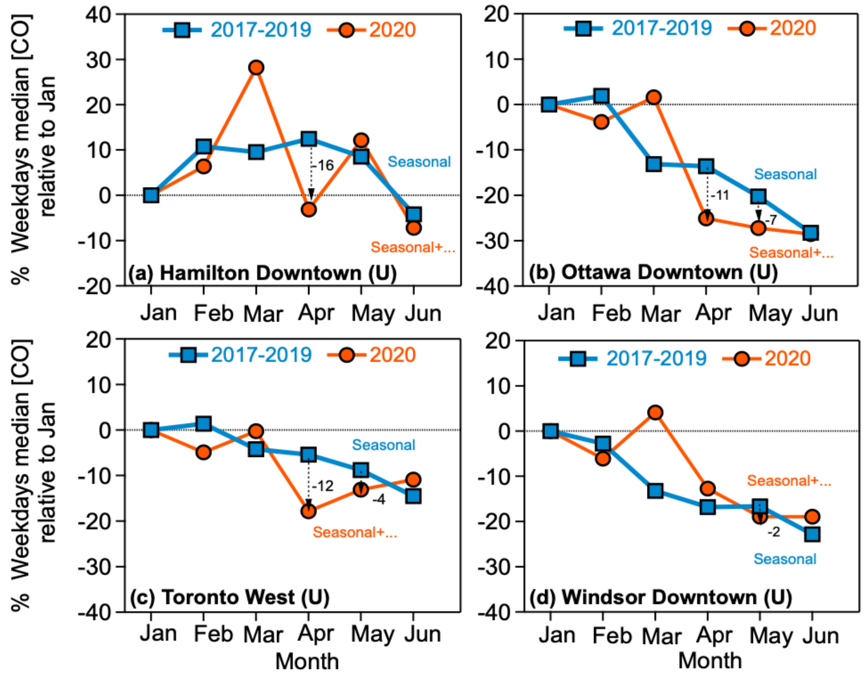


Figure 4: Percentage difference in weekday median levels of CO in 2020 and 2017-2019 relative to January of the same year(s). The vertical lines highlight the statistically significant percentage decreases based on the p-values listed in Table 1 for April – June.

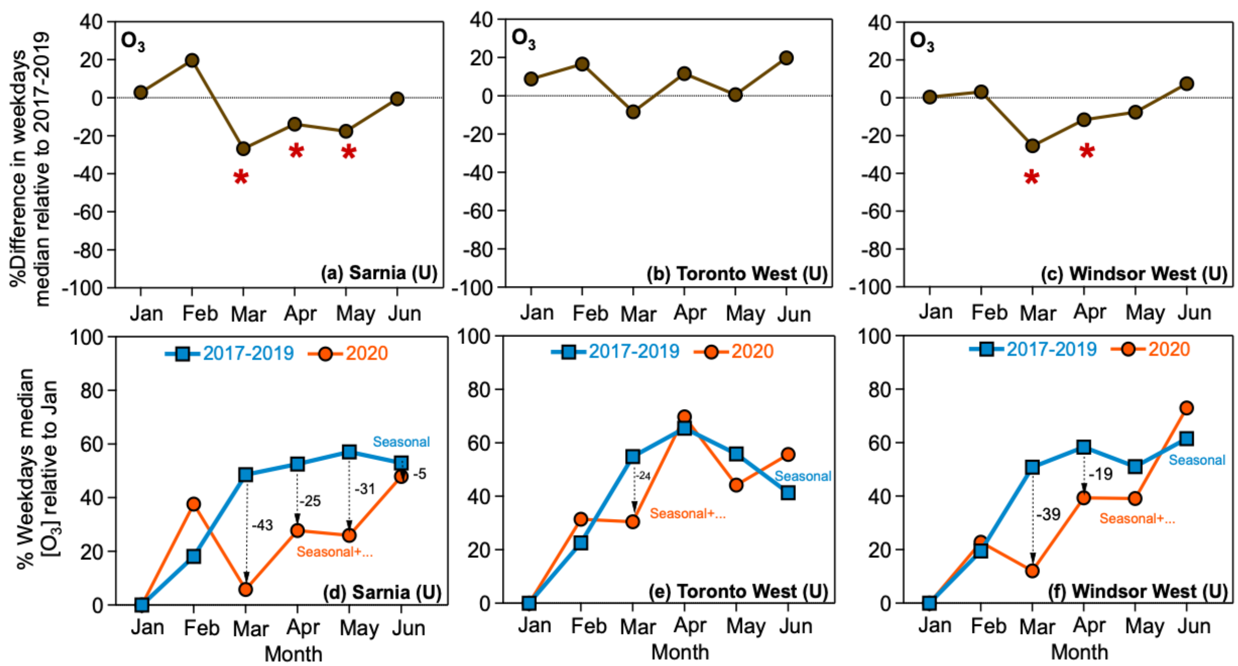


Figure 5: (a-c) Percentage difference in weekday median levels of O_3 in 2020 relative to the same period in reference years, 2017-2019. The data for April – June are listed in Table 1 for these sites. (d-f) The percentage in O_3 median values in 2020 and 2017-2019 relative to January of the same year(s). The vertical lines highlight the statistically significant percentage decreases based on the p-values listed in Table 1 for Apr – June.

and Windsor Downtown. The relatively large reduction in Hamilton West site was also observed when the percentage was calculated relative to January of the same year(s) (Figure 6d). This result suggests that the Ontario lockdown on the industrial activity in Hamilton had a significant impact on the levels of PM2.5, which was not observed in the other sites. The Ottawa Downtown site experienced 11% reduction in PM2.5 (Figure 6e), which likely reflects the effect of the City's lockdown on transportation. The Windsor Downtown site experienced only 7% reduction in PM2.5 (Figure 6f), which was likely influenced by the industrial activity in Michigan, USA. Interestingly, levels of PM2.5 were higher in June 2020 compared to previous years in all of the sites analyzed.

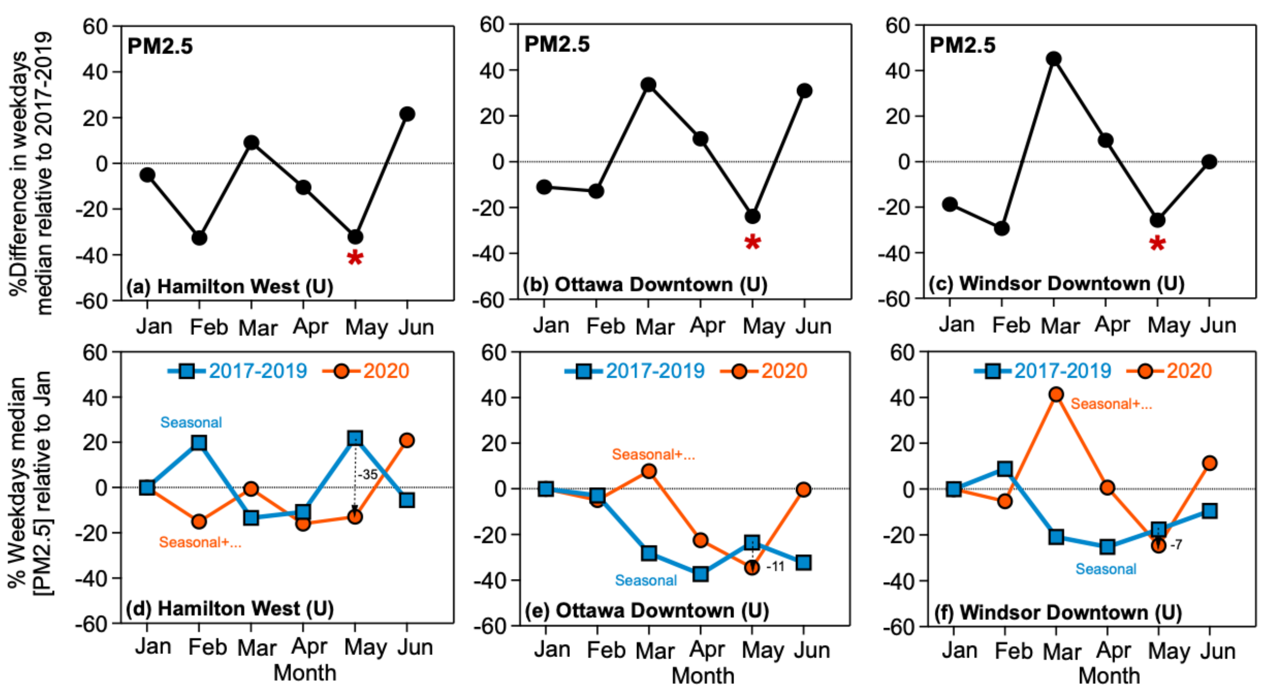


Figure 6: (a-c) Percentage difference in weekday median levels of PM2.5 in 2020 relative to the same period in reference years, 2017-2019. The data for April – June are listed in Table 1 for these sites. (d-f) The percentage in PM2.5 median values in 2020 and 2017-2019 relative to January of the same year(s). The vertical lines highlight the statistically significant percentage decreases based on the p-values for Apr – June.

DISCUSSION

Sites within the City of Hamilton were expected to see little impact from decreased transportation and industrial activity, as many of the city's industry were likely classified as "essential services" during the lockdown that started in mid-March. As a result, only one site in the city had a statistically significant drop in NO_2 (Hamilton Downtown – June). The statistically insignificant drops in NO_2 at all other sites in Hamilton could be due to slowed production or could be due to annual variability driven by atmospheric chemistry.[9] This result matches the observations of Shi and Brasseur[5], who found substantial variability in NO_2 levels, as well as other pollutants, in Beijing, which had less severe lockdown measures than Wuhan.

In the Toronto region, NO_2 levels were expected to be significantly impacted by local sources, such as transportation and industry given their relatively large distance from significant U.S. sources of the Ohio Valley. All Toronto sites saw large drops in NO_2 levels in April 2020 relative to 2017-2019, except Toronto Downtown. The drop in NO_2 levels observed here are similar to reported by Griffin *et al.*[1] after accounting for seasonality estimated in their analysis. The Toronto West site also saw a drop in measured CO levels. While not all decreases in pollutant levels were significant compared to previous years, the trend suggests that decreased movement of the population and industry played a considerable part in the observed drops. These results corroborates the findings of Griffin *et al.*[1] who found that reductions in NO_2 in the Toronto region are not entirely due to COVID-19 related emissions reductions. The mix of significant and insignificant decreases from previous years could be due to the fact that a number of industries within the Toronto region were likely still operating during the lockdown, given their "essential services" status. These findings could also be highlighting the importance of other factors such as meteorology and atmospheric chemistry.[9]

Medium sized cities in Southwestern Ontario were also expected to have little impact from transboundary sources for NO_2 with transportation making a larger impact on NO_2 and CO sources than industry. While some of these cities have manufacturing facilities, they are not expected to be on the scale of Toronto or Hamilton. As seen in Table 1, Kitchener and London had statistically significant drops in NO_2 in all but one month, with the remaining month still showing a large drop in NO_2. Guelph had a statistically significant drop in April and a large (albeit insignificant) drop in May. This data suggest that the drop in NO_2 could be directly linked with decreased traffic in these cities. This is corroborated by transportation data from Kitchener that saw a 55% decrease in traffic in early May, and a 47% decrease in late May – early June in 2020 compared to previous traffic counts within the city (Table S20). Furthermore, Ottawa experienced statistically insignificant drops in NO_2 in all three months, as well as statistically significant drops in CO in April and May. This likely reflects the effect of the City's lockdown on transportation and is reinforced by the findings for PM2.5 levels in the city.

Sarnia, Windsor Downtown and Windsor West were expected to have a large transboundary influence from both Michigan and Ohio, but also a potentially significant influence from transportation. Given the wide range of dates of closures and re-openings across the two US states and Ontario, it was expected that little to no difference would be seen in 2020 compared to previous years. Any difference was expected to be seen in April since all three jurisdictions were closed in this month. However, both Windsor sites saw significant decreases in NO_2 in May, with Windsor Downtown also seeing a significant drop in NO_2 in June and a significant decrease in CO in May. Only Sarnia saw a significant decrease of NO_2 in April. This may suggest that Sarnia is more impacted by local transportation, including cross border traffic, as opposed to Windsor, which is impacted by local industry immediately across the border in and around Detroit. It is not clear why the Windsor sites did not see a significant drop in April but did in May and June, which requires further analysis. Again, this finding is similar to that of Griffin *et al.*[1] who found that NO_2 levels in this region of the province were difficult to recreate as a result of difficulties in estimating changes in US-based emissions due to reduced activity during COVID lockdowns.

The statistically significant drops in CO at sites across the province, especially in April, highlight the drop in transportation during the pandemic. The drop in CO concentrations continued into May, with half of the sites recording a statistically significant drop. As the province relaxed quarantine measures and the population re-emerged during May and June, the drops in CO were generally smaller than April, and not always statistically significant.

In conclusion, the government measures to limit the spread of COVID-19 in Southern Ontario resulted in statistically significant reduction in pollutant levels emitted from the transportation and industrial sectors in the majority of the sites analyzed. These reductions were beyond the seasonal variability observed within the last three years. Other sites were influenced by transboundary and/or other local influences (i.e., industry) that countered local reductions in human activity. Results presented here are highly significant because (1) they highlight the need to carry out rigorous statistical analysis that accurately quantifies the significance of short term events on pollutant levels, (2) our analysis provides numerical evidence to the magnitude that large scale lockdowns have on air quality in Southern Ontario since worsening air quality is one of the impacts of climate change[16], and (3) policy makers would be better informed when planning for mitigation and adaptation for long-term and lasting positive effects of reducing air pollution.[17] That being said, meaningful change with respect to air pollution and air quality can only be solved with meaningful local change in select circumstances. Our results highlight the impact that transboundary pollution and local industrial sources can have, limiting the effect that changing local transportation modes, as an example, can have on local air quality. Furthermore, our results also suggest that seasonal meteorology can account for large differences between years, compared to which the lockdown effect is insignificant. This highlights the importance of considering all factors that influence air pollution and that policies critical to one jurisdiction may not have a significant impact in another jurisdiction. As the province continuously monitors and reports the effect of air quality regulations for different sites, future data collection should also focus on specific chemical compounds or classes that affect local O_3 and PM2.5 formation[9] to disentangle local versus transboundary sources.

In light of recent research that correlates long term exposure to NO_2,[18] PM2.5 and PM10 in polluted cities[19] with fatalities caused by COVID-19, future analysis should also focus on analyzing the relationship between pollution levels, number of confirmed COVID-19 cases and deaths in the sites analyzed here. Since airborne transmission is identified as the dominant route for the spread of COVID-19,[20,21] research that correlates PM levels in Southern Ontario and the rates of infections and deaths are worth investigating. It is important to account for population density, age, race, socioeconomic status and establish a clear baseline from previous years on major causes of respiratory diseases and fatality.

AUTHOR INFORMATION

Corresponding Authors
* H.A.A.: Phone: (519)884-0710, ext.2873; e-mail: halabadleh@wlu.ca.
* M.L.: Phone: (519) 888-4567, ext.35503; e-mail: mlysy@uwaterloo.ca .

Notes
The authors declare no competing financial interest.

Content in this paper was previously submitted to *ChemRxiv*, a preprint server: Hind A. Al-Abadleh, Martin Lysy, Lucas Neil, Priyesh Patel, Wisam Mohammed, and Yara Khalaf. Statistical

quantification of COVID-19 lockdown on air quality from ground-based measurements in Southern Ontario, Canada. 2020.

ASSOCIATED CONTENT

Supporting Information Available

Examples of earlier studies, Tables S1-S20, Figures S1-S10, additional details on methods. This material is available free of charge on the ACS Publications website at DOI: XXX

Acknowledgements. The authors acknowledge funding from the Natural Science and Engineering Council of Canada (NSERC). The authors thank the City of Kitchener for providing the traffic count data.

References

(1) Griffin, D.; McLinden, C.; Racine, J.; Moran, M.; Fioletov, V.; Pavlovic, R.; Eskes, H., Assessing the impact of Corona-Virus-19 on nitrogen dioxide levels over southern ontario, canada. *Geophys. Res. Lett.* **2020,** *Pre-print*, Earth and Space Science Open Archive. https://doi.org/10.1002/essoar.10503538.2.

(2) Bauwens, M.; Compernolle, S.; Stavrakou, T.; Muller, J.-F.; van Gent, J.; Eskes, H.; Levelt, P. F.; van der A, R.; Veefkind, J. P.; Vlietinck, J.; Yu, H.; Zehner, C., Impact of coronavirus outbreak on NO_2 pollution assessed using TROPOMI and OMI observations. *Geophys. Res. Lett.* **2020,** *47*, e2020GL087978. https://doi.org/10.1029/2020GL087978.

(3) Schiermeier, Q., Why pollution is falling in some cities — but not others. *Nature* **2020,** *580*, 313.

(4) Covid-19 and air pollution (accessed august 27, 2020). https://docs.google.com/document/d/1UTQvW_OytC37IatMNR5qJK7qKfSylNpI2fT3pdteVZA/edit#heading=h.8lu90y3nmm7o

(5) Shi, X.; Brasseur, G. P., The response in air quality to the reduction of chinese economic activities during the COVID-19 outbreak. *Geophys. Res. Lett.* **2020,** *47*.

(6) Le, T.; Wang, Y.; Liu, L.; Wang, J.; Yung, Y. L.; Li, G.; Seinfeld, J. H., Unexpected air pollution with marked emission reductions during the COVID-19 outbreak in China. *Science* **2020,** *369*, 702-706.

(7) Tanzer-Gruener, R.; Li, J.; Eilenberg, S. R.; Robinson, A. L.; Presto, A. A., Impacts of modifiable factors on ambient air pollution: A case study of COVID-19 shutdowns. *Environ. Sci. Technol. Lett.* **2020,** *7*, 554-559.

(8) Venter, Z. S.; Aunan, K.; Chowdhury, S.; Lelieveld, J., COVID-19 lockdowns cause global air pollution declines. *PNAS* **2020,** *117*, 18984-18990.

(9) Kroll, J. H.; Heald, C. L.; Cappa, C. D.; Farmer, D. K.; Fry, J. L.; Murphy, J. G.; Steiner, A. L., The complex chemical effects of COVID-19 shutdowns on air quality. *Nature Chem.* **2020,** *12*, 777-779.

(10) *Air quality in ontario*; Ministry of the Environment and Climate Change (currently Ministry of the Environment, Conservation and Parks): Toronto, ON, 2016.

(11) Atkins, D. H. F.; Lee, D. S., Spatial and temporal variation of rural nitrogen dioxide concentrations across the united kingdom. *Atmos. Environ.* **1995,** *29*, 223-239.

(12) *Toxicological profile for carbon monoxide*; Agency for Toxic Substances and Disease Registry (ATSDR), U.S. Department of Health and Human Services Atlanta, USA, 2012.

(13) Fisher, R. A., *The design of experiments*. Oliver and Boyd: Edinburgh, 1935.

(14) Manly, B., *Randomization, bootstrap and monte carlo methods in biology*. Chapman & Hall/CRC: Boca Raton, FL, 2006; Vol. 70.

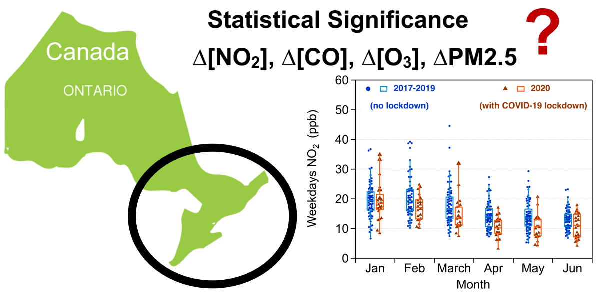
(15) Dabek-Zlotorzynska, E.; Celo, V.; Ding, L.; Herod, D.; Jeong, C.-H.; Evans, G.; Hilker, N., Characteristics and sources of PM2.5 and reactive gases near roadways in two metropolitan areas in Canada. *Atmos. Environ.* **2019,** *218*, 116980 (1-13).

(16) von Schneidemesser, E.; Monks, P. S.; Allan, J. D.; Bruhwiler, L.; Forster, P.; Fowler, D.; Lauer, A.; Morgan, W. T.; Paasonen, P.; Righi, M.; Sindelarova, K.; Sutton, M. A., Chemistry and the linkages between air quality and climate change. *Chem. Rev.* **2015,** *115*, 3856-3897.

(17) Ching, J.; Kajino, M., Rethinking air quality and climate change after COVID-19. *Int. J. Environ. Res. Pub. Health* **2020,** *17*, 5167 (1-11).

(18) Ogen, Y., Assessing nitrogen dioxide (NO_2) levels as a contributing factor to coronavirus (COVID-19) fatality. *Sci. Total Environ.* **2020,** *726*, 138605 (1-5).
(19) Zoran, M. A.; Savastru, R. S.; Savastru, D. M.; Tautan, M. N., Assessing the relationship between surface levels of PM2.5 and PM10 particulate matter impact on COVID-19 in Milan, Italy. *Sci. Total Environ.* **2020,** *738*, 139825 (1-12).
(20) Zhang, R.; Li, Y.; Zhang, A. L.; Wang, Y.; Molina, M. J., Identifying airborne transmission as the dominant route for the spread of COVID-19. *PNAS* **2020,** *117*, 14857-14863.
(21) Asadi, S.; Bouvier, N.; Wexler, A. S.; Ristenpart, W. D., The coronavirus pandemic and aerosols: Does COVID-19 transmit via expiratory particles? *Aerosol. Sci. Technol.* **2020,** *54*, 635-638.

TOC Graphic



Statistical Significance
Δ[NO2], Δ[CO], Δ[O3], ΔPM2.5
Canada
ONTARIO
2017-2019
(no lockdown)
2020
(with COVID-19 lockdown)
Weekdays NO2 (ppb)
0
10
20
30
40
50
60
Jan
Feb
March
Apr
May
Jun
Month