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Impact of the COVID-19 lockdown on the chemical composition and sources of urban PM$_{2.5}$

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ABSTRACT

The lockdown measures caused by the COVID-19 pandemic substantially affected air quality in many cities through reduced emissions from a variety of sources, including traffic. The change in PM$_{2.5}$ and its chemical composition in downtown Toronto, Canada, including organic/inorganic composition and trace metals, were examined by comparing with a pre-lockdown period and respective periods in the three previous years. During the COVID-19 lockdown, the average traffic volume reduced by 58%, whereas PM$_{2.5}$ only decreased by 4% relative to the baselines. Major chemical components of PM$_{2.5}$, such as organic aerosol and ammonium nitrate, showed significant seasonal changes between pre- and lockdown periods. The changes in local and regional PM$_{2.5}$ sources were assessed using hourly chemical composition measurements of PM$_{2.5}$. Major regional and secondary PM$_{2.5}$ sources exhibited no clear reductions during the lockdown period compared to pre-lockdown and the previous years. However, cooking emissions substantially dropped by approximately 61% due to the restrictions imposed on local businesses (i.e., restaurants) during the lockdown, and then gradually increased throughout the recovery periods. The reduction in non-tailpipe emissions, characterized by road dust and brake/tire dust, ranged from 37% to 61%, consistent with the changes in traffic volume and meteorology across seasons in 2020. Tailpipe emissions dropped by approximately 54% and exhibited even larger reductions during morning rush hours. The reduction of tailpipe emissions was statistically associated with the reduced number of trucks, highlighting that a small fraction of trucks contributes disproportionally to tailpipe emissions. This study provides insight into the potential for local benefits to arise from traffic intervention in traffic-dominated urban areas and supports the development of targeted strategies and regulations to effectively reduce local air pollution.

1. Introduction

More than 150 million cases and 3 million deaths from the coronavirus disease (COVID-19) have been reported globally since the SARS-CoV-2 virus was first reported, affecting all countries and territories around the world (Johns Hopkins University, April 2021). Canada reported its first case on January 25, 2020, in Toronto, Ontario. On March 17, the provincial government of Ontario declared a state of emergency and implemented a provincial-wide lockdown in response to COVID-19 (Ontario, 2020). Similar to other countries, strict restrictions were imposed to limit transportation, schools, commerce, and cultural activities that resulted in abrupt and significant changes in vehicle traffic in urban areas.

The COVID-19 lockdown provided an unprecedented natural experiment to explore the potential benefits of air quality by limiting different sources. In particular, local and regional changes in traffic emissions during the lockdown influenced air quality. Recent studies from across the globe during the COVID-19 pandemic have reported a wide range of reductions in concentrations of commonly monitored air pollutants, such as nitrogen dioxides (NO$_2$) and carbon monoxide (CO) (e.g., Kumari and Toshniwal, 2020). A comparison of how PM$_{2.5}$ (particulate matter with an aerodynamic diameter smaller than 2.5 μm) changed at 21 sites in the USA showed a mix of increases and decreases such as 44% in Seattle, 46% in Indianapolis, versus −41% in Los Angeles, and −45% in San Jose (Chen et al., 2020). A wide range of reductions (i.e., no change to −50%) in PM$_{2.5}$ were also observed in other countries (Chauhan and Singh, 2020). This is not surprising given that PM$_{2.5}$ is a complex mixture of primary and secondary pollutants originating from various local/regional scale anthropogenic/biogenic sources, thus requiring a more comprehensive analysis to understand the

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changes in PM$_{2.5}$. Few studies have explored the influence of the COVID-19 lockdown on the changes in the chemical components of PM$_{2.5}$, such as black carbon (BC), trace metals, inorganics species, and organic aerosol (OA). In urban areas across Europe, BC declined by approximately 33–55% (Greece), 41% (Spain), and 58–71% (Italy) during the COVID-19 lockdown compared to the pre-lockdown period (Grivas et al., 2020; Collivignarelli et al., 2020; Tobias et al., 2020). Some traffic-related trace elements in PM$_{2.5}$ were reduced by 43–55% during the partial lockdown in an urban area of Vietnam (Nguyen et al., 2021). A source apportionment study of OA exhibited that secondary OA decreased by 86% during the lockdown in Delhi, India as compared to the pre-lockdown period, whereas primary OA related to traffic emissions decreased by 18% during the lockdown in India (Manchanda et al., 2021). The mixed results reflect the complex contributions of primary and secondary ambient pollutants from local and regional sources that may vary by location. Thus, a comprehensive analysis is necessary to properly assess and understand the impact of COVID-19 lockdowns on PM$_{2.5}$. The objective of this study was to assess the influence of the COVID-19 lockdown on the sources contributing to PM$_{2.5}$, with an emphasis on traffic-related contributions by comparing measurements in the 2020 lockdown period to a pre-lockdown period and baselines established in 2017–2019. The influence of the changes in traffic volume and fleet composition on PM$_{2.5}$ were examined to elucidate their relationship with local traffic emissions. Specifically, changes in the chemical composition of PM$_{2.5}$, including inorganic/organic species and trace metals, were assessed to identify traffic-related PM$_{2.5}$ and understand the impact of the COVID-19 lockdown on the level of traffic-related PM$_{2.5}$ sources.

2. Methods

2.1. Monitoring location and instrumentation

The concentration of PM$_{2.5}$ and its components were continuously measured from February 2017 to July 2020 at a near-road monitoring station at the Southern Ontario Centre for Atmospheric Aerosol Research (SOCAAR), located in downtown Toronto, Ontario, Canada (at 43°39′32″ N, 79°23′43″ W). This downtown site is surrounded by multi-story buildings and commercial market/restaurants districts. The sampling inlet of the downtown site was situated 3 m above the ground, and 15 m from the 4-lane arterial road. Detailed information on the near-road monitoring station and instrumentation can be found elsewhere (Evans et al., 2019). The mass concentrations of PM$_{2.5}$ were measured by a 5030 Synchronized Hybrid Ambient Real-time Particulate Monitor (SHARP, Thermo Scientific). Black carbon was measured every minute by a 7-wavelength Aethalometer (AE33, Magee Scientific). Hourly averaged Aethalometer data measured at a wavelength of 880 nm were referred to as total BC. The absorption characteristics of BC aerosol from fossil fuel combustion (BCf) and biomass burning (BCbb) were obtained by an optical method (Healy et al., 2017).

Thirty-minute averages of organic aerosol (OA) and inorganic aerosol (sulphate, nitrate, and ammonium) were measured by a quadrupole mass spectrometer (Aerosol Chemical Speciation Monitor, ACSM, Aerodyne), while hourly concentrations of trace metals (i.e., potassium (K), sodium (Na), calcium (Ca), titanium (Ti), manganese (Mn), iron (Fe), copper (Cu), zinc (Zn), selenium (Se), strontium (Sr), barium (Ba), and lead (Pb)) were determined by a continuous X-ray fluorescence monitor (625 Xact, Cooper Environ). The description and calibration of the ACSM and Xact metal monitor are described in the supplementary material.

As auxiliary measurements, nitrogen oxides (NO$_x$, NO + NO$_2$), CO, ozone (O$_3$), and carbon dioxide (CO$_2$) were measured by a chemiluminescence (42i Thermo Scientific), infrared photometric (48i Thermo Scientific), UV photometric (49i Thermo Scientific), and non-dispersive infrared analyzer (840 A, LI-COR Biosciences, Precision: ±1.5% of reading), respectively. The number concentrations of ultrafine particles (UFP, >8 nm) were measured by a Condensation Particle Counter (CPC, 651 API). In addition to the measurements of air pollutants, vehicle counts, speeds, and lengths (i.e., C2: 1.0–7.6 m, C3: 7.6–15 m, C4: 15–36.5 m) were measured by a radar detector (SmartSensor HD, Waveotronix). Routine calibration and maintenance including reference and background checks of the gas and particle monitors were conducted as discussed elsewhere (e.g., Hilker et al., 2019; Evans et al., 2019). Meteorological parameters including ambient temperature, relative humidity (RH), wind speed (WS), and wind direction (WD) were measured using a weather sensor (Vaisala WXT520) at the SOCAAR site.

2.2. Source identification of PM$_{2.5}$

To resolve the sources contributing to PM$_{2.5}$ and to quantify the impact of the reduction of traffic on PM$_{2.5}$, a multivariate factor analysis, Positive Matrix Factorization (PMF), was applied to hourly concentrations of organic and inorganic speciation data, trace elements, and BC at the downtown site using EPA PMF 5 (Norris, 2014). A two-step PMF approach was used to resolve OA factors more efficiently by reducing the number of variables of the large dataset in the PMF analysis (Jeong et al., 2019). A detailed description of the source apportionment analysis is shown in the supplementary material.

2.3. Relative change in concentrations

In this study, data analyses were conducted for the 4-weeks between March 22 and April 25, 2020, as the first-phase lockdown period (Lockdown I) and the following 4-weeks between April 26 and May 23, 2020, as the second-phase lockdown period (Lockdown II). As relaxed restrictions were implemented at the end of May, the 4-week periods, May 24-June 27 and June 28-July 25, 2020, were referred to as Recovery I and Recovery II, respectively. Data for a transition period (March 15 to March 21, 2020) were excluded from the analysis. To estimate the relative change of PM$_{2.5}$ composition during the strict lockdown and relaxed lockdown periods, the mean values were compared with the 4-week Pre-Lockdown period (February 16 to March 14, 2020) and the corresponding baseline periods of the 3 previous years. The former helped account for any year-to-year variability in emissions, while the latter was used to evaluate the effect of restricted emissions under similar seasonal conditions. The seasonality is accounted for by comparing measurements during the lockdown period to measurements during the corresponding periods in 2017–2019 as follows:

\[
\text{Relative Change, } RC = \frac{|\text{Lockdown}_{2020} | - |\text{Lockdown}_{2017-19} |}{|\text{Lockdown}_{2017-19} |}
\]

(1)

where \([\text{Lockdown}_{2020}]\) and \([\text{Lockdown}_{2017-19}]\) were the mean concentrations during the Lockdown I period in 2020 and the corresponding period of Lockdown I in 2017–2019, respectively. The implicit assumption was that in the absence of the lockdown, the average concentration during the lockdown period would have been equal to the average concentration of the same period across 2017–2019. Admittedly, this method also has limitations, as year-to-year differences in seasonality often occur at this time of year. Non-parametric Spearman correlations and Mann-Whitney U tests were used to examine the correlations and the significance of differences between two datasets, respectively.

3. Results and discussion

3.1. Change in traffic volume and meteorology

During the measurement period from 2017 to 2020 (including the COVID-19 lockdown period), traffic counts of passenger cars (C2), trucks and buses (C3), and heavy-duty trucks (C4) were continuously measured at the downtown site (Fig. 1 and Table 1). On average, the volume of total vehicles in 2017–2019 was 930 vehicles hour$^{-1}$ with
approximately 88% being passenger cars (C2) and 8% being trucks and buses (C3). The number of heavy-duty trucks was very low on this arterial road and many of the C4 vehicles were in fact 15–16 m-long streetcars. In 2020, the average traffic volume reduced from approximately 910 vehicles per hour (Pre-Lockdown) to 390 vehicles per hour (Lockdown I), a reduction of about 57%. The comparable vehicle number during Pre-Lockdown compared to the previous three years indicates interannual stability in traffic volumes at the nearest road. The speed of vehicles also increased by about 27% due to the reduced traffic volume. Similar reductions of C2 and C3 were observed, indicating no change in the fleet composition between Pre-Lockdown and Lockdown I with the truck fraction of approximately 6%. The relative change was approximately a decrease of 58% and 64% for both C2 and C3, respectively (Mann-Whitney U test, p < 0.01). Thus, the lockdown due to the COVID-19 pandemic provided an interesting natural experiment that allowed us to examine how air quality changed when more than half of all vehicles were taken off the road.

The average temperature varied from 2 °C to 26 °C during the measurement periods (Table 1). As compared to the three previous years, there was no statistical difference in the average temperature during Lockdown I in 2020 and the corresponding baseline period (p = 0.81). Relative humidity and wind speed were relatively constant across the Pre-Lockdown and Lockdown I periods during 2017–2020. A wind rose comparison shows the same prevailing wind directions (southwest and northwest) between Lockdown I and the corresponding baseline period (Fig. 52). These findings indicate that changes in temperature, RH, and wind parameters during Lockdown I were comparable to those in the corresponding period in 2017–2019. In this study, no normalization in the meteorological conditions ambient during the lockdown year with respect to previous years was applied. A further study is recommended to isolate the impact of the COVID-19 lockdown from other sources of variability such as temporal trends of air pollutants over time and meteorological variables.

3.2. Change in black carbon and PM$_{2.5}$

Table 1 summarizes the monthly averages and standard deviations of BC and PM$_{2.5}$. Fig. S1 illustrates the weekly average concentrations of BC and PM$_{2.5}$ from February 16 to July 26 in 2017–2020. A clear decrease of BC was observed between Lockdown I and the corresponding baseline period in the previous years, whereas PM$_{2.5}$ showed no statistically significant differences (RC = 4% for PM$_{2.5}$ with p = 0.63 vs. 64% for BC with p < 0.01). The BC data in the three previous years showed relatively constant levels ranging from 500 ng m$^{-3}$ to 700 ng m$^{-3}$ between February and April in 2017–2019, whereas BC substantially dropped in

![Fig. 1. Weekly average counts of passenger cars (C2) and trucks and buses (C3) from Feb. 16 to Jul. 26 in 2017–2019 (light blue) and 2020 (dark blue). Error bars represent the 95% confidence intervals. Data from June 14 to July 26 in 2017–2019 are not available.](image-url)
2020, presumably due to reduced anthropogenic emissions. It should be noted that the estimated reduction in BC was potentially influenced by a construction site located 30 m from the monitoring station that was active from 2017 to 2019. The BC baseline may be affected by emissions from diesel construction machinery and trucks involved in construction. The ratio of BCf to total BC during Pre-Lockdown was lower than that of the corresponding baseline period (0.64 vs. 0.81, Table 1 and Fig. S1), supporting the presence of additional fossil fuel sources such as emissions from the construction activities from 2017 to 2019.

The diurnal pattern of BC (and BCf) was dominated by the strong peak during morning rush hours (7:00 a.m. to 9:00 a.m.), whereas no clear trend was observed for PM$_{2.5}$ (Fig. 2 and Fig. S3). The morning peak of BC during Lockdown I dropped by 66% compared to the corresponding baseline period (0.64 vs. 0.81, Table 1 and Fig. S1). A clear trend was observed for PM$_{2.5}$ during the morning peak of BC during Lockdown I, whereas no peak during the corresponding baseline period (0.64 vs. 0.81, Table 1 and Fig. S1). The increase in BC during Recovery in June and July might also reflect the typical summertime rise in BC concentrations as found in a previous BC apportionment study at nine urban sites in Ontario (Healy et al., 2017).

It is also interesting to note that the evening peak between 6:00 p.m. and 8:00 p.m. (local time) observed for BC, BC$_{10}$, NO$_x$, and UFP during Pre-Lockdown was not observed during Lockdown I and Recovery II. Detailed analysis of the evening peak is discussed in the supplementary material. In brief, the nighttime increase of BC, NO$_x$, and UFP prior to Lockdown was not solely due to meteorological conditions, but also potentially due to contributions of additional sources that changed during Lockdown. Since this downtown site was situated amongst commercial districts, cooking and heating in local restaurants and commercial/residential buildings may have contributed to additional local emissions in the evening. Further investigation was undertaken which revealed a reduction of cooking emissions during Lockdown, as discussed later in the section.

3.3. Change in chemical composition of PM$_{2.5}$

The relative change to PM$_{2.5}$ during Lockdown I was about 4%, which indicated a weak correlation to observed traffic reductions. To better understand the impact of the lockdown on PM$_{2.5}$, the changes in the chemical composition of PM$_{2.5}$ were examined (Fig. 3 and Fig. S5, Tables S2 and S3). The ACSM measured non-refractory PM up to approximately 1 μm in aerodynamic diameter (NR-PM$_1$). The sum of OA, sulphate, nitrate, and ammonium by the ACSM was comparable to the PM$_{2.5}$ mass measured by a collocated SHARP PM$_{2.5}$ monitor ($r^2 = 0.69$, AC SM PM$_1 = 0.79 \times$ SHARP PM$_{2.5}$). Overall, OA, nitrate, sulphate, and ammonium accounted for approximately 56%, 11%, 7%, and 6% of PM$_{2.5}$, respectively. As the largest contributor, the temporal trend of OA was similar to that of PM$_{2.5}$, showing the lowest concentration during Lockdown (Spring) and the highest during Recovery (Summer). In contrast, ammonium nitrate, the second-largest contributor, exhibited a strong winter-high pattern, while sulphate showed a summer-high pattern. The formation of the major PM$_{2.5}$ chemical components predominantly depends on secondary reactions of gas and particle-phase pollutants originating from regional-scale sources. For example, nitrate, sulphate, and ammonium are the main secondary inorganic aerosol components in PM$_{2.5}$ mainly occurring as ammonium nitrate and ammonium sulphate that is formed by the neutralization of nitric acid (HNO$_3$) and sulphuric acid (H$_2$SO$_4$) with ammonia (NH$_3$), respectively (Seinfeld and Pandis, 2016). The strong seasonality of the chemical components during the measurement period spanning from February to July makes it difficult to properly estimate the change in PM$_{2.5}$ during the COVID-19 lockdown. This may explain the wide range of PM$_{2.5}$ changes attributed to lockdowns, reported across the world (Kumari and Toshniwal, 2020). In addition to the major chemical species, hourly concentrations of trace elements were measured over the last 4 years at the downtown site. Diurnal variations of eleven trace elements on weekdays and weekends indicated clear weekday-high patterns for Ca, Ti, Mn, Fe, Cu, Zn, and Ba (p < 0.01, Fig. S4). Elements Ca and Fe mostly come from soil and road dust, whereas Cu, Ba, Zn, and Mn are marker elements for abrasion of brake pads and tire wear (Councell et al., 2004; Grigoratos and Giorgio, 2014; Jeong et al., 2020). In Toronto, Zn is also associated with local iron/steel making processes (Jeong et al., 2019). Elevated concentrations of these elements were typically observed during the corresponding baseline period due to favorable meteorological conditions and drier road surfaces without snow cover and dust accumulated over the winter when no street sweeping occurs. The average concentrations of the selected elements, Ca, Fe, Mn, Cu, Zn, and Ba are compared in Fig. S5. The highest concentrations of these elements were found during the corresponding baseline period in 2017–2019, except for Zn. During Lockdown I, the strongest reduction was observed for Ba with a relative change of 76%, followed by Cu (66%) and Zn (66%), indicating substantial reductions as compared to the baseline period (Table S3).

3.4. Changes in sources of PM$_{2.5}$

In the OA source apportionment analysis, the most reliable solution was explained by four factors, including two primary OA factors, hydrocarbon-like OA (HOA) related to fresh traffic emissions and cooking OA (COA) dominated by a tracer ion for cooking emissions, and two oxygenated OA factors, more-oxygenated oxygenated OA (MOOxOA) and less-oxygenated oxygenated OA (LOOOA) differentiated by their oxidation states. Distinct features of the mass spectral pattern and the diurnal variations of each factor were used to characterize these OA factors as described in the supplementary material (Figs. S6 and S7). The factor profiles of the OA factors show similarity to the reference mass spectra presented elsewhere (e.g., Sun et al., 2012). The comparison of
the average concentrations of the OA factors during Lockdown is displayed in Fig. 3. Detailed descriptions of the four OA factors are discussed in the supplementary material.

To better identify the changes in the sources of PM$_{2.5}$ during Lockdown, the OA factors were used as variables in the second PMF analysis including three inorganic species (sulphate, nitrate, ammonium), eleven trace elements, and BC data. In this comprehensive PMF analysis, a nine-factor solution was found to represent the most reliable sources of PM$_{2.5}$; aged organic aerosol (Aged OA), secondary organic aerosol (Sec OA), secondary nitrate (Nitrate), coal combustion related to industrial use (Coal), cooking emissions (Cooking), tailpipe emissions (TE), non-tailpipe emissions related to road dust (NTE_RD), non-tailpipe emissions related to brake dust (NTE_BD), and non-tailpipe emissions related to tire dust and metal industry (NTE/Metal). The chemical profiles and contributions of the nine sources are depicted in Figs. S8 and S9, respectively. The evaluation of the PMF solution is discussed in the supplementary material. The Aged OA, Sec OA, and Nitrate factors accounted for approximately 25% (1.9 ± 1.6 μg m$^{-3}$, Mean ± Standard deviation), 20% (1.5 ± 1.5 μg m$^{-3}$), and 24% (1.8 ± 2.8 μg m$^{-3}$) of the total PM$_{2.5}$ mass in 2017–2019. As major contributors, the Aged OA and Sec OA factors were associated with the MOOOA and LOOOA factors identified based on the degree of oxidation (Fig. S8). Aged OA (or oxygenated OA in general) has been observed in many OA source apportionment studies using ACSM and aerosol mass spectrometer (AMS) studies at many other sites around the world (Jimenez et al., 2009; Mohr et al., 2012; Xu et al., 2015; Gu et al., 2018). These secondary OA factors were found to be associated with regional-scale sources in the urban area (Jeong et al., 2019). The seasonality for the secondary OA factors showed a similar summer-high pattern with a strong correlation with ambient temperature (Fig. S10 and Table S4). No weekend/weekday differences were observed for the secondary OA factors, supporting that these factors are unlikely to be primary traffic emissions (Fig. S11). The presence of some trace elements in the source profile of Aged OA probably indicates that aged biomass burning (K) and transboundary industrial emissions (Se and Pb) are possible sources for Aged OA (Xu et al., 2015; Sofowote et al., 2019). Nitrate characterized by ammonium nitrate exhibited distinct seasonality with strong correlations with NO$_2$ and temperature ($p < 0.01$). These results support that the formation of particulate nitrate (Nitrate) is thermodynamically favored by cold winter temperatures to form HNO$_3$ from NO$_2$ and react with NH$_3$ (Seinfeld and Pandis, 2016; Wang et al., 2020).

The large PM$_{2.5}$ sources exhibited no clear reductions during Lockdown I compared to Pre-Lockdown and the baseline years, but strong seasonal variations (Fig. 4 and Fig. S10). Due to the formation of secondary organic aerosols (SOA) via photochemical reactions in summer and ammonium nitrate favored by low temperature in winter (Hayes et al., 2013; von der Weiden-Reinmüller et al., 2014), the contributions of Sec OA and Nitrate to PM$_{2.5}$ increased to 35% in summer and 44% in winter, respectively, in the previous years. The absence of any clear reduction in the Nitrate factor during Lockdown was surprising, given that emission of the precursor compounds decreased to some extent. Ambient temperature during Lockdown I in 2020 was comparable to that during the corresponding period in 2019 (6.2 ± 4.9 °C in 2020 vs. 6.4 ± 4.4 °C in 2019). Given the similar meteorology condition between the periods in 2019 and 2020, the Nitrate factor dropped by 23% in 2020 compared to the period in 2019 which was comparable to the relative change in NO$_2$ (25%). From Lockdown II to Recovery II, the Nitrate factor decreased by 40% in 2020 compared to the corresponding baseline periods, indicating the overall reduction of nitrate was due to the reduced traffic volume.

The coal combustion factor was characterized by ammonium sulphate and Se, accounting for 10% (0.8 ± 0.9 μg m$^{-3}$) of total PM$_{2.5}$ mass. The highest secondary sulphate concentrations were observed during mid-day (noon to 3:00 p.m., Fig. S11), consistent with the diurnal pattern of sulphate observed in the eastern US and Canada (e.g., Sun et al., 2011; Jeong et al., 2019). The presence of Se was also associated with a trace metal from coal-fired power plants in the eastern US (Squizzato et al., 2018). Previous studies in Toronto reported that the sulphate-rich factor can be attributed to the long-range transport of coal-fired power plant emissions from the US including the Ohio River Valley region (Jeong et al., 2020; Sofowote et al., 2015). The negligible change during Lockdown I was also found for the coal combustion factor. However, the concentration of the Coal factor in the summer (Recovery II) dropped by 36% in 2020 compared to the corresponding baseline period (Fig. S4, Table S4).
Table S6). The presence of the reduction seems credible given that overall sulphur dioxide emissions from coal-based electricity production in the US decreased by 34% in 2020 compared to 2017–2019 (US EPA, 2021). Historically, the contribution of the coal combustion factor in Toronto has dropped significantly which is mainly due to the complete phase-out of coal-fired power plants in the province of Ontario and the continuous reduction of coal burning in the US (Jeong et al., 2020).

Cooking, with a high loading of cooking OA, dominated by COA accounted for 9% of PM$_{2.5}$ ($0.7 \pm 0.9 \ \mu\text{g m}^{-3}$). Higher correlations between this factor and CO ($r = 0.41, p < 0.01$) and UFP ($r = 0.38, p < 0.01$) were observed compared to other sources. The cooking source was distinguished by apparent peaks at lunch and dinner times (Fig. S11), consistent with the diurnal patterns of COA in previous studies (Zhang et al., 2011; Shah et al., 2018) There was no statistically significant difference between weekends and weekdays, even though the contribution of the cooking source on weekend evenings was slightly higher than the level on weekdays. Unlike these major PM$_{2.5}$ sources (i.e., Aged OA, Sec OA, Nitrate, and Coal) affected by the secondary formation of regional-scale sources, a local PM$_{2.5}$ source related to primary cooking emissions dropped by 61% compared to the baseline averages. The sharp difference between Pre-Lockdown and Lockdown I/II in the cooking emissions factor and the gradual increase over the Recovery periods corresponds to the timeline of government interventions on restaurant activity (e.g., lowering capacity) to curb the first surge of COVID-19 cases. The Pre-Lockdown diurnal trend of the cooking factor shown in Fig. 5 presents pronounced peaks at noon and 8:00 p.m., consistent with lunch and dinner mealtimes (Zhang et al., 2011; Shah et al., 2018). The distinct drop from 0.62 $\mu$g m$^{-3}$ to 0.25 $\mu$g m$^{-3}$ during Lockdown I compared to the corresponding baseline period indicates that cooking emissions mainly originate from commercial businesses such as restaurants in the downtown area rather than cooking activities in residential sectors. Robinson et al. (2018) found elevated OA concentrations in the vicinity of many restaurants in an eastern US city and a majority of acute exposure to OA was from restaurant cooking emissions. In the presence of nighttime peaks of BC, NO$_x$, and UFP, cooking emissions from local restaurants can be an additional local PM$_{2.5}$ source in the downtown area. This also highlights how the lockdown policies have impacted emissions from local businesses in the urban community. Traffic-related PM$_{2.5}$ sources, which include tailpipe emissions (TE) and non-tailpipe emissions (NTE_RD, NTE_BD, and NTE/Metal), accounted for 5% (0.4 ± 0.7 $\mu$g m$^{-3}$), 3% (0.2 ± 0.2 $\mu$g m$^{-3}$), 2% (0.2 ± 0.2 $\mu$g m$^{-3}$), and 1% (0.1 ± 0.1 $\mu$g m$^{-3}$) of PM$_{2.5}$ on average, respectively, in the urban area from 2017 to 2020. The traffic-related PM$_{2.5}$ sources were distinguished by the prominent peak during the morning rush hour with strong weekday/weekend differences ($p < 0.01$, Fig. S11). The TE factor was represented by emissions from internal combustion engines including HOA and BC (Fig. S6). Higher correlations with traffic-related air pollutants (i.e., NO$_x$, UFP) and vehicle counts were found for TE, compared to the NTE factors. During Lockdown I, TE dropped by 54% in 2020 compared to the previous baseline years, in agreement with the reduction in the traffic volume (57–59%). The TE factor characterized by HOA was highly correlated with tracers for primary emissions, NO$_x$, CO, and UFP (Table S4). The diurnal pattern of TE was also consistent with the emissions from traffic exhaust characterized by traffic rush hour in the morning. The morning peak at 8:00 a.m. sharply decreased by 59% during Lockdown I, compared to the baseline period, which is consistent with the reduced traffic volume. This also implies that the TE factor can be used as a surrogate for the reduced traffic volume during Lockdown. In terms of the change in the fleet composition of motor vehicles during Lockdown and Recovery, the number of passenger cars (mostly gasoline vehicles) were still lower by half during Recovery, whereas truck counts gradually increased over time and were almost back to the...
Pre-Lockdown level by July (Recovery II). This resulted in an increased truck fraction as shown in Fig. 1. The concentrations of TE generally followed the trend of C3 truck counts during Recovery (Fig. 4). The TE factor was more correlated with the number of C3 trucks \( (r = 0.33, p < 0.001) \) than C2 passenger cars \( (r = 0.21, p < 0.001) \). This suggests that the reduction of TE was driven by the reduced number of trucks in the urban environment. These findings also highlight that a small number of vehicles are heavy emitters and emit disproportionately compared to the rest of the fleet \( (Dallmann et al., 2012; Hudda et al., 2013) \). Wang et al. (2015) reported that a small fraction of vehicles are heavy emitters and emit 25\% more than half to the total fleet emissions at this site in Toronto.

The factor of non-tailpipe emissions related to road dust (NTE_RD) was characterized by high loadings of crustal elements, Ca, Ti, Mn, and Fe, while Cu and Ba were key markers of brake wear dust (NTE_RD) \( (Grigoratos and Giorgio, 2014; Jeong et al., 2019) \). The NTE factor, characterized by a high loading of Zn, was believed to in part be associated with tire wear \( (Harrison et al., 2012) \). However, it was also found that the Zn-rich source (NTE/Metal) was potentially associated with local metals processing (Metallurgy) in previous source apportionment studies in the urban area \( (Jeong et al., 2019; Sofowote et al., 2014) \). The diurnal trends of the NTE factors were similar to that of the TE factor, showing prominent morning rush hour peaks and clear differences between Lockdown in 2020 and those baselines in 2017–2019 (Fig. 5).

Furthermore, these NTE factors were also characterized by weekday-high patterns \( \text{weekday/weekend} = 1.81 \) for NTE_RD, 1.17 for NTE_BD, 1.58 for NTE/Metal, \( p < 0.01 \) having moderately good correlations with the tracers of primary traffic emissions \( (Table S4) \). During Lockdown I, relative changes of up to 37\%, 60\%, and 61\% for NTE_RD, NTE_BD, and NTE/Metal, respectively, with respect to the corresponding baseline period were also found as shown in Fig. 4. Compared to Pre-Lockdown, the reductions varied ranging from \( -14\% \) for NTE_BD and 45\% for NTE/Metal. Typically, non-tailpipe emissions can be influenced not only by traffic volume and motions, but also by meteorology. \( Table S4 \) shows a statically strong negative correlation between RH and the road dust factor, NTE_RD, implying that the resuspension of road dust is enhanced by dry road conditions. The lower reduction for NTE_RD may imply that soil dust that is less influenced by traffic activity might be included in the NTE_RD factor due to the similarity in their chemical profiles dominated by crustal elements. The mixed reductions for the NTE factors can be explained by the meteorology-induced seasonality of the NTE in March and April. The upward trend of the NTE sources during the Recovery periods is also consistent with the higher fraction of trucks observed during this time as discussed for the TE factor. Thus, these results underscore the importance of targeted strategies to reduce heavy emitters on roadways to mitigate the levels of the tailpipe and non-tailpipe emissions more effectively. This has important implications for future traffic-restricting intervention. Policies implemented to remove the small fraction of heavy emitting vehicles from populated areas could yield large benefits.

The mass reconstruction of the PM_{2.5} sources during Lockdown I was compared to that in the corresponding baseline period \( (Fig. 6) \). While the total PM_{2.5} mass dropped by approximately 4\% between Lockdown I in 2020 and the corresponding baseline period without a significant difference \( (p = 0.63) \), local primary PM_{2.5} sources (Local PM_{2.5}: Cooking, TE, NTE_RD, NTE_BD, NTE/Metal) dropped by 54\%. No difference was observed for the sum of the major secondary PM_{2.5} sources (Regional PM_{2.5}: Aged OA, Sec OA, Nitrate, Coal) between the periods.
Substantial changes were found in local primary sources of PM during the winter and spring months. More mass in the urban area was influenced by counterbalanced regional and local sources, the chemical composition of PM$_{2.5}$ (including both tailpipe and non-tailpipe emissions) was more likely caused by the reduced number of trucks. This implies that removing and/or replacing the small fraction of heavy emitting vehicles can yield large benefits to more effectively mitigate tailpipe and non-tailpipe emissions. These findings support the development of more effective policies and regulations that aim to reduce local air pollution. In terms of the risks of air pollutants on human health, trace elements related to non-tailpipe traffic emissions may lead to reactive oxygen species generation (Yang et al., 2014; Weichenthal et al., 2016). The unprecedented reductions to traffic may create potential health benefits as it led to a reduction of traffic-related PM$_{2.5}$ emissions. Future work on the infrastructure of ambient air monitoring stations and data analysis strategies are required to adequately measure these benefits of the interventions.

### 4. Conclusions

Primary and secondary air pollutant concentrations along with traffic data before, during, and after the first government-imposed lockdown in 2020 were compared to those during reference periods (a pre-lockdown period in 2020 and a baseline from 2017 to 2019). Whereas the volume of traffic decreased by approximately 60% during Lockdown I in 2020, the contribution of the traffic-related PM$_{2.5}$ sources dropped from 24% in 2017–2019 to 13% in 2020. The contributions of cooking emissions to PM$_{2.5}$ also dropped from 21% in 2017–2019 to 11% in 2020 during the evening hours of 6:00 p.m. to 8:00 p.m. due to the lockdown.

### Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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### Appendix A. Supplementary data

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