Black Carbon Particles Physicochemical Real-Time Data Set in a Cold City: Trends of Fall-Winter BC Accumulation and COVID-19

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Abstract Black carbon (BC) plays an important role in climate and health sciences. Using the combination of a year real-time BC observation (photoacoustic extintciometer) and data for PM2.5 and selected co-pollutants, we herein show that annual BC Mass concentration has a bi-modal distribution, in a cold-climate city of Montreal. In addition to the summer peak, a winter BC peak was observed (up to 0.433 μg/m3), lasting over 3 months. A comparative study between two air pollution hotspots, downtown and Montreal international airport indicated that airborne average BC Mass concentration in downtown was 0.344 μg/m3, whereas in the residential areas around Montreal airport BC Mass values were over 400% higher (1.487 μg/m3). During the numerous snowfall events, airborne BC Mass concentration decreased. High-resolution scanning/transmission electron microscopy with energy dispersive X-ray spectroscopy showed BC or carbon nanomaterials, at snow-air interface analysis of the snow samples provided evidence that airborne BC particles or carbon nanomaterials were indeed transferred from polluted air to snow. During the COVID-19 lockdown, the BC concentration and selected co-pollutants, decreased up to 72%, confirming the predominance of anthropogenic activities in BC emission. This first cold-climate BC data set can be essential for more accurate air quality and climate modeling. About one-third of the Earth's land surface receive snow annually, the impact of this study on air quality, health and climate change is discussed.

1. Introduction

Aerosols or airborne particles have drawn great scientific interests in the past decades due to their essential roles in the nucleation of ice, clouds, formation, type and intensity of precipitation, radiation budget, and climate change (Bond et al., 2013; de Oliveira Alves et al., 2015; Fuzzi et al., 2015; IPCC, 2013; Knopf et al., 2018; McCluskey et al., 2014; Munoz-Alpizar et al., 2017; Murray et al., 2012; Nazarenko et al., 2017; Petters et al., 2009; Qian et al., 2014; Rangel-Alvarado et al., 2019; Washenfelder et al., 2015). The Intergovernmental Panel for the Climate Change (IPCC) has thereby identified aerosols and aerosol-climate interactions as the major uncertainty in climate change (IPCC, 2013). The World Health Organization (WHO) has shown that aerosols have critical impacts on human health, contributing to over 8 million premature deaths per year, including 4.2 million deaths attributed to ambient air pollution and 3.8 million deaths caused by indoor air pollution (WHO, 2021a).

Aerosol composition is diverse, originating from natural and anthropogenic sources such as organic, inorganic (including metallic), and biological particles (Bond et al., 2013; Hall et al., 2020; IPCC, 2013; Rahim et al., 2019; Rangel-Alvarado et al., 2015; Rivas et al., 2020). Airborne black carbon (BC) is a type of aerosol that formed as by-product of incomplete combustion of fossil fuel, accounting for 5%–15% of the annual urban particulate matter concentration (Xu et al., 2020; Yang et al., 2011).

Like other aerosols, BC can impact climate directly by absorbing and scattering solar radiation (Bahadur et al., 2012; Bond & Bergstrom, 2007; Bond et al., 2013; Qian et al., 2014; Xu et al., 2019), and indirectly by serving as cloud condensation nuclei and ice nuclei (Brooks et al., 2014; Fan et al., 2016; Kulkarni et al., 2016; McCluskey et al., 2014; Parent et al., 2016; Qian et al., 2014; Vergara-Temprado et al., 2018). BC is a significant component in climate change (Bond et al., 2013) and is estimated to be +1.1 W/m2 with 90% uncertainty bounds from +0.17 to +2.1 W/m2, only second to CO2 (Bond et al., 2013). Yet, the question of how BC affects climate change is still debated (Dou & Xiao, 2016; Grandey et al., 2018; IPCC, 2014; Malavelle et al., 2019) due to lack of knowledge of the physicochemical properties of BC.
The lack of understanding of the BC’s physicochemical characteristics such as morphology, size and mixing state is the cause of the existing large uncertainty for an accurate evaluation of its radiative forcing (Bond & Bergstrom, 2007; Chakrabarty et al., 2014; Lack et al., 2014; Long et al., 2013). For instance, organic aerosols (OAs) are commonly co-emitted with BC from biomass burning. The internal and external mixing of OA and BC have been suggested to strongly alter the optical properties of BC, increasing the absorption ability in a specific wavelength, causing a stronger climate warming effect (Cappa et al., 2012; Nakayama et al., 2014). Whereas the heterogeneous ice nucleation of BC under diverse environmental conditions shows contrasting results (Brooks et al., 2014; Fan et al., 2016; Kulkarni et al., 2016; McCluskey et al., 2014; Petters et al., 2009; Qian et al., 2014; Vergara-Temprado et al., 2018).

BC is indeed linked to adverse health effects (Bove et al., 2019; Brown, 2013; Jia et al., 2020). For instance, toxic compounds such as polycyclic aromatic compounds (PAHs) have been shown to be bound with BC. A large proportion of anthropogenic BC emissions occur in urban regions, home to most of the human population. The urban BC sources include industrial production, transportation, electrical production, and heating systems (IPCC, 2013). Air traffic is a major transportation venue in the world (ICAO, 2016). The contribution of BC emission from airplanes has been rising because of the expansion of aviation in the last decades (ICAO, 2016). There are several studies on the air pollution characterizations at airports (Rahim et al., 2019; Rivas et al., 2020) and experimental research of BC emission from airplane engines (Abegglen et al., 2016; Chen et al., 2019; Elser et al., 2019; Parent et al., 2016), revealing BC emission is even larger when airplanes are near or on the ground, in comparison with climbing and flying in the sky (ICAO, 2016). To our knowledge, there have been limited studies on in situ real-time BC emission at the airport that are needed for a comprehensive assessment of the link between airplanes emitted BC and the health hazards of people living close to the airports.

The importance of snow on atmospheric chemistry and physics of the urban regions has been reported (Ariya et al., 2018). Aerosols are known to promote the formation of ice nuclei that can ultimately lead to the precipitation of snow in the atmosphere (Rangel-Alvarado et al., 2015). During the snowfall, aerosols like BC can be scavenged by snowflakes (Ariya et al., 2018; Pal et al., 2020; Rangel-Alvarado et al., 2019; Wang et al., 2021). Furthermore, as snowpack is porous, it can adsorb combustion emitted gas and particles like BC and PAHs (Nazarenko et al., 2016, 2017). BC has been suggested to change snow albedo, and affect the snow melting process (Bond et al., 2013). Snow organic compounds have been shown to undergo physicochemical processes including re-emission to air, upon melting (Nazarenko et al., 2016, 2017). Halogenated salts are commonly added as a de-icing agent in snowy cities, affecting the oxidation potential of the lower atmosphere (Hall et al., 2020; Pal et al., 2020; Rahim et al., 2019; Rangel-Alvarado et al., 2019).

In this study, we aimed to develop real-time data set on selected physicochemical characteristics of BC, in a model cold-climate city of Montreal, Canada (Nazarenko et al., 2017; Pal et al., 2020; Rangel-Alvarado et al., 2015). Concurrently, selected air co-pollutants (such as particulate matters [PM], carbon monoxide [CO], and nitrogen dioxide and monoxide [NO$_x$]) were analyzed. We herein evaluated BC’s distributions and accumulation in the boundary layer. We also explored whether the BC concentration close to urban hotspots in residential areas is drastically elevated in the cold climate to cause potential adverse health. Lastly, as there are limited data on the impact of COVID-19 pandemic lockdowns on pollutant emissions in cold-climate cities, we explored a suite of air pollutants prior to, during the lockdown and after partial re-openings, pointing to the significance of anthropogenic activities in air quality.

2. Methodology

2.1. Black Carbon Analysis

A photoacoustic extinctiometer (PAX, Droplet Measurement Technologies, Inc.) with 870 nm laser module is used to in situ measure absorption coefficient ($B_{abs}$) of BC from ambient air. PAX samples air with a 1 L/min flow rate. When air is drawn into the sampling chamber of PAX, the 870 nm laser will heat up BC particles in the air since few aerosols, except BC, can absorb light at 870 nm wavelength. Subsequently, the heated BC particles transfer heat to the surrounding air, generating pressure waves which can be detected by a sensitive microphone inside the chamber to get $B_{abs} (\text{Mm}^{-1})$ of the sample in 1-s resolution:
\[
\text{BC Mass}\left(\mu g/m^3\right) = B_{\text{abs}}\left(Mm^{-1}\right)/\text{BC MAC}\left(m^2/g\right)
\] (1)

The calibration of PAX was performed by using manufacturer-recommended material, glassy carbon spheres (GCS). GCS were nebulized by an aerosol generator and then passed through PAX to generate \(B_{\text{abs}}\) values which were greater than 5,000 Mm\(^{-1}\) for 30–60 s. BC Mass concentration was calculated directly by using \(B_{\text{abs}}\) and literature and manufacturer’s recommended mass absorption cross-section (MAC) (4.74 m\(^2/g\) at 870 nm) at ambient temperature and pressure (Bond & Bergstrom, 2007; Selimovic et al., 2019), as is shown in Equation 1. It should be noted that BC MAC can increase depending on the BC mixing state.

PAX also provides a scattering coefficient (\(B_{\text{scat}}\)). \(B_{\text{scat}}\) is measured by a nephelometer inside the chamber, and respond to all kind of particles regardless the size, morphology, and chemical compositions. (NH\(_4\))\(_2\)SO\(_4\) was used to calibrate \(B_{\text{scat}}\) as it can scatter light, yet it can hardly absorb it. (NH\(_4\))\(_2\)SO\(_4\) solution was aerosolized to pass through PAX to generate \(B_{\text{scat}}\) values which were greater than 5,000 Mm\(^{-1}\) for 30–60 s.

\[
B_{\text{ext}} = \frac{1}{0.354}\ln \frac{I}{I_0} \times 10^6 \left[Mm^{-1}\right]
\] (2)

The calculation of the calibration process is shown in Equation 2, where \(B_{\text{ext}}\) is the extinction coefficient which can also be calculated by summing up \(B_{\text{abs}}\) and \(B_{\text{scat}}\) \((B_{\text{ext}} = B_{\text{scat}} + B_{\text{abs}})\). \(I_0\) is the average laser power before and after calibration, and \(I\) is the laser power during calibration. To calculate \(B_{\text{scat}}\) \(B_{\text{ext}}\) must be calculated based on Equation 2, then plot calculated \(B_{\text{ext}}\) with measured \(B_{\text{scat}}\) to gain the correction factor (the slope of the linear regression) since \(B_{\text{abs}}\) of (NH\(_4\))\(_2\)SO\(_4\) can be negligible. Yet to calibrate \(B_{\text{abs}}\) (\(B_{\text{ext}}-B_{\text{scat}}\) should be plotted against \(B_{\text{abs}}\) because the scattering of GSC cannot be neglected. It should be noted that, the calibration process assumes a linear response of the instrument up to the calibration absorption values.

The systematic error of PAX on BC Mass mainly comes from the value of BC MAC. The value of BC MAC used in this study is 4.74 m\(^2/g\). This value is calculated based on two reasons: (a) Bond and Bergstrom (2007) recommended the value of BC MAC at 550 nm is 7.5 ± 1.2 m\(^2/g\); and (b) BC MAC is inversely related to wavelength. Yet, Bond and Bergstrom (2007) noted that, BC MAC can increase with the mixing state. For thickly coated particles, the absorption enhancement can be up to 50% for BC aggregates (Bond & Bergstrom, 2007). BC MAC varies from 4.74 to 7.11 m\(^2/g\). Thus, BC Mass reported in this work can be overestimated if BC particles are more coated than the value of 4.74 m\(^2/g\).

Both BC Mass and \(B_{\text{scat}}\) are reported in the format of average values adding or subtracting standard errors.

### 2.2. High-Resolution Electron Microscopy

The details of high-resolution scanning/transmission electron microscopy (S/TEM) with energy dispersive X-ray spectroscopy (HR-S/TEM-EDS) were described elsewhere (Pal et al., 2020; Rahim et al., 2019; Rangel-Alvarado et al., 2015). The TEM SiO\(_2\)-membrane grids were analyzed using a high-resolution FEI. Tecnai G2F20 S/TEM microscope with a field emission gun. Images were acquired using an Advanced Microscopy Technique, Corp. (AMT) XR80C CCD Camera System, which was previously used for snow samples (Rangel-Alvarado et al., 2015). The AMT was adapted for collecting aerosols directly on the grid to further analyze the size, morphology, and chemical compositions of aerosol samples with high resolution.

### 2.3. Sampling Sites, Observation, and Snow Collection

Air sampling is continuously conducted in downtown Montreal. The instruments are placed inside the laboratory of Otto Maass Building (45°30’N 73°34’W), with direct ambient air inlets, in the heart of downtown Montreal at McGill University (Pal et al., 2020; Rahim et al., 2019). The laboratory maintains a constant temperature, pressure, and relative humidity to ensure the instruments work optimally. Whereas particle observations near the Montreal-Pierre-Elliott-Trudeau International Airport (Quebec, Canada) were performed at a public part (Westwood Park; 45°27’N 73°45’W) which is only 200 m away from the airport runway and in the residential areas. The BC observations were conducted on the weekends prior to the COVID-19 shutdowns, during the total closure and after partial reopening in Montreal.
2.4. Aerosol and Snow Collection

A Micro Orifice Uniform Deposit Impactor (MOUDI, model 100-R, MSP Corp., Shoreview, MN, USA) was used to collect size-fractionated aerosol samples. MOUDI was operated on the roof with an inlet flow rate of 30 L/min and set up for 12-hr runs starting from 8 a.m. Teflon quartz substrate with TEM grids attached was used to make sure the samples can be used in TEM based on the method our group developed (Hudson & Ariya, 2007).

Snow samples were collected at Westwood Park by following the procedures described in detail elsewhere (Rangel-Alvarado et al., 2015). Generally, snow samples were collected during the snow precipitation events from the top 2 cm of the surface, and the aged snow taken at least 5 cm beneath the snowpack surface with sterile equipment and clean suits. All samples were kept in pre-sterilized amber chambers, frozen immediately after collection at −10 ± 2 °C, like ambient temperature during sampling. Small amounts of collected samples were transferred to a centrifuge tube with sterilized spoon under clean conditions. The centrifuge tube was kept frozen for HR-S/TEM-EDS analysis.

2.5. Other Air Co-Pollutant Data Analysis

The hourly averaged concentrations of fine PM$_{2.5}$, CO, NO$_x$, and ozone (O$_3$) are obtained from The Air Quality Monitoring Network (RSQA), conducted by Environmental and Climate Change Canada (ECCC) and City of Montreal, which provides data from the monitoring stations on the Island of Montreal including stations both in downtown and near the Montréal-Pierre-Elliott-Trudeau International Airport (YUL Airport) (NAPS, 2021). The PM$_{2.5}$ continuous measurements are done by a synchronized hybrid ambient real-time particulate monitor (SHARP 5030). A CO Analyzer (Thermo 48i) is used, which utilizes the gas filtration correlation technology. A chemiluminescence NO-NO$_2$-NO$_x$ Analyzer (Thermo 42i) is used for the quantification of NO$_x$, O$_3$ is measured by a UV-photometric Ozone Analyzer (Thermo 49i) (Hall et al., 2020). All the data are provided in hourly averages with 24 data points per day. The unit of CO, O$_3$ and NO$_x$ is ppb, while the unit of PM$_{2.5}$ is μg/m$^3$, as is shown in Tables 2–5. All the concentrations are reported in the format of average values adding or subtracting standard errors.

The back trajectory analysis was performed on internet based HYSPLIT model provided by Air Resources Laboratory (Rolph et al., 2017; Stein et al., 2015).

3. Results and Discussions

3.1. BC and Selected Co-Pollutants Emission in a Cold-Climate City

Montreal is a typical cold-climate city (Järvi et al., 2014) where winter lasts for 5 months (spanning from December 1 to April 20, according to ECCC), receiving about 209 cm of snow per year. The long winter and low temperature lead to the need for additional heating, such as the heating system of buildings in the whole city and car idling, increasing the usage of fuels and, thereby, emission of BC (Bond et al., 2013).

Previous observations have shown that the highest number density of airborne nanoparticles in City of Montreal occurs during the winter (Pal et al., 2020). Hall et al. (2020) have shown the high oxidation potential from anthropogenic photolabile chlorine in the photochemical process is due to de-icing salts during winter. In this study, we explored whether the mass concentration of BC showed a similar trend to what was previously observed for nanoparticles (Pal et al., 2020) and chlorine (Hall et al., 2020), that is, a maximum peak for BC Mass during the long winter in this cold-climate city and whether the existence of the snow affects the BC distribution and snow physicochemical properties.

To test this hypothesis, BC Mass from July 2019 to June 2020 (except October 2019) is shown in Figure 1a. The real-time $S_{abs}$ data is also presented in the supplementary information (Figure S5 in Supporting Information S1). Although data of October were absent because of repairing the instrument, BC Mass in downtown Montreal showed a bimodal variation. BC emissions reached a peak before or in July (0.365 ± 0.007 μg/m$^3$), yet decreased through summer and autumn, and showed another peak during the winter (0.433 ± 0.013 μg/m$^3$), then decrease again until next spring. The long duration of sunshine in summer has been suggested to promote the photochemical reactions of organic carbon internally or externally mixed
with BC, increasing the hygroscopicity of BC, and accelerating the wet deposition of BC in the atmosphere (Bond et al., 2013; Brooks et al., 2014; Zhang et al., 2019). Summer rainfalls may facilitate frequent wet deposition of BC. However, the BC fate in winter in the cold climate is indeed complex. More snowfalls in long winter can decrease BC Mass by ice nucleation and wet deposition (Hadley et al., 2010; Liu, He, et al., 2020), yet the application of the heating system and increasing consumption of fuels because of low temperature contribute to an extra BC emission.

Cold air has a higher density than warm air, the height of the boundary layer is thereby lower and thus more compact in the winter in contrast to the summer. Figure S6 in Supporting Information S1 shows HY-SPLIT modeling results on two typical days in summer and winter, January 20, 2020 (winter) and June 28, 2020 (summer). It has already been shown that the height of the atmospheric boundary layer is higher in

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**Figure 1.** Monthly mean values of black carbon (BC) mass (a), day-night comparison of BC Mass in each month (b), comparison of BC with $O_3$ (c), PM$_{2.5}$ (d), CO (e), and NO$_2$ (f). The error bars in panels (a, b) indicates the standard deviations.
summer than in winter (Chan & Wood, 2013). Figure S6 in Supporting Information S1 demonstrates that BC entrapment is more important, and air dilution is less pronounced in cold seasons in comparison with warmer seasons, which can also partly explain the decreasing tendency from July 2019 to September 2019. Since snow is a ubiquitous feature of cold-climate, airborne BC particles can also be deposited on snowpack, following multiple cycles of melt and precipitations and release to the atmosphere, hydrosphere, and lithosphere (Ariya et al., 2018; Nazarenko et al., 2016, 2017). Such processes may explain the peaks observed between December 2019 to February 2020.

Due to the COVID-19 Pandemic in winter 2020, a lockdown was implemented from March 16 to May 25 in Montreal, which restricted most the non-essential human activities, and led to a significant reduction of BC emission from March to May (Figure 1 and Figure S1 in Supporting Information S1). The sharp increase in June 2020 can be attributed to the partial reopening of the city. After a long term of lockdown, human activities gradually increased, which led to the increment of BC emission in the end.

The impact of photochemistry can be reflected in Figure 1b and Figure S1 in Supporting Information S1. Daytime is set from 9 a.m. to 6 p.m., whereas Nighttime is from 10 p.m. to 5 a.m. In this way, the variation of sunlight hours induced by seasons can be excluded. BC Mass at night is more than, or close to BC Mass during daytime. BC Mass during daytime decrease is due to photochemistry. Nighttime BC emission in the atmosphere sustained for a relatively long time due to the lack of sunlight.

Figure S1 in Supporting Information S1 shows the diurnal variation of BC Mass based on all-year data, indicating the net effects of emissions due to anthropogenic activities. Like other aerosols (Pal et al., 2020; Rahim et al., 2019), BC has two peaks during the traffic rush hours. BC emission reached the maximum at 7–8 a.m. when citizens begin to commute to their workplaces on workdays. Around noon, it has been shown that BC Mass gradually decreases likely due to the variation of the atmospheric boundary layer (Begam et al., 2016). The height of the atmospheric boundary layer gradually increases and then peaks value at noon, causing the dilution of BC in the atmosphere (Begam et al., 2016). Later in the afternoon, BC emission increases again due to vehicle traffic and a decrease in the height of the boundary layer. Whereas at night, with less traffic and industrial production, the BC concentration variation remains steady unless strong wind or precipitation takes place. In brief, the diurnal variation, besides natural processes, also reveals the important role of human activities such as commute and transportation in BC emissions in urban areas (Bond et al., 2013; Jia et al., 2020; Liakakou et al., 2020).

The comparisons of BC Mass with concentrations of O₃, PM₁₀, CO, and NO₂ are shown in Figures 1c–1f. BC is one of the anthropogenic pollutants in this urban area. PM₁₀, CO, and NO₂ in city are also mainly originated from human activities. Pearson correlation analysis was performed between BC Mass and concentrations of O₃, PM₁₀, CO, NO₂, NO, and SO₂ respectively for each month (Table S5 in Supporting Information S1).

The results indicate a positive correlation between BC Mass with PM₁₀, CO, NO₂ and a negative correlation between BC Mass with O₃. BC consists of particles of various sizes. BC particles whose sizes are smaller than 2.5 μm accounts for part of PM₁₀, which can confirm the positive correlation between BC Mass and concentrations of PM₁₀. The positive correlation between BC with CO and NO₂ can be explained by anthropogenic activities including transport sector emissions. PM₁₅, CO, and NO₂ are common anthropogenic pollutants. The positive correlation coefficients with these air pollutants confirms that BC emission in an urban area is mainly from anthropogenic sources. Whereas O₃ plays an essential role in photochemical processes, which oxidize organic carbon internally or externally mixed with BC in the atmosphere, facilitating the wet deposition of BC (Bond et al., 2013; Brooks et al., 2014; Zhang et al., 2019), and decreasing the atmospheric BC Mass, likely indicating a negative correlation between O₃ and BC.

### 3.1.1. Meteorological Winter in a Cold Climate: Up to 5 Months

Although the variation of BC Mass shows a peak during winter, it should be noticed that BC Mass in warm-climate cities, like Nanjing (Tan et al., 2020), Athens (Liakakou et al., 2020), and Czech (Mbengue et al., 2020), is greatest during winter as well, which indicates large BC Mass during winter is prevalent. Yet, winter in the cold-climate sites lasts longer, comparing with warm-climate sites where winter usually ends in February. The 5-month winter in Montreal, a cold-climate city, makes it possible that the duration of large BC Mass is longer than warm-climate sites. Although, this speculation cannot be fully examined...
directly because of the lack of BC Mass data from January 2019 to May 2019, the increase of CO in fall and winter can be used as BC proxy, indirectly (Rahim et al., 2019). Furthermore, according to the results illustrated above, CO shows a positive correlation with BC, with Pearson correlation coefficients varying from 0.2909 to 0.9194 (Table S5 in Supporting Information S1). Consequently, CO can be used to estimate the variation of other anthropogenic pollutants like BC.

Table S2 in Supporting Information S1 shows the variations of CO from January to June in the last four years. Table S3 in Supporting Information S1 illustrates the results of t-test between January with the other five months respectively. The values in the bracket of March 2020 are obtained by using data of the first 15 days of March 2020 (before lockdown). There is no significant difference between January with February and March (except for March 2018) in the past four years. It accounted for high concentration of CO maintaining until April, that is, still meteorological winter in Montreal. As such, it is logical to speculate that the duration of large BC Mass can also maintain from December to the end of March, showing the uniqueness of cold-climate sites. The longer duration of large BC Mass in cold-climate sites may cause more health hazards to human health.

3.2. COVID-19 Pandemic and Impacts on the Urban Environment

SARS-CoV-2 causing COVID-19 disease was first reported in Wuhan, China in December 2019. Since then, the WHO declared it to be a worldwide pandemic, infecting about 150 million people around the world, causing over 3 million deaths all around the world, and it is still counting (WHO, 2021b). During the COVID-19 lockdowns, several researchers have shown a decrease of air pollution around the city worldwide (Bao & Zhang, 2020; Briz-Redón et al., 2021; Gautam, 2020; Kanniah et al., 2020; Lee et al., 2020; Lian et al., 2020; Liu, Sha et al., 2020; Liu et al., 2021; Mahato et al., 2020; Mandal & Pal, 2020; Nakada & Urban, 2020; Park et al., 2021; Putaud et al., 2021; Zhang et al., 2020).

As the lockdown policy obviously constrain the spread of COVID-19 among citizens, many countries complied with it to protect their people. COVID-19 not only changes the world profoundly but also provides an opportunity for scientists to observe and evaluate anthropogenic impacts on the environment during the lockdown period when almost all unnecessary economical activities are stagnant. Earlier in China and Italy, the sharp decreases in CO, NOx, and particulate matter (PM2.5 and PM10) emissions were immediately reported (Bao & Zhang, 2020; Gautam, 2020; Liu, Sha et al., 2020; Putaud et al., 2021; Zhang et al., 2020). Similar phenomena were found in India, Brazil, and Southeast Asia (Kanniah et al., 2020; Mandal & Pal, 2020; Nakada & Urban, 2020). The decrease of air pollutants including CO, NOx, PM2.5, and the increase of O3 has been widely reported (Table 1).

It is noteworthy that in urban air quality both meteorological variability and anthropogenic emission changes impact urban air pollutant concentrations and should be thus considered together. In this study, we consider bi-modal distribution of BC Mass based on two points: (a) decreasing trend from July 2019 to September 2019; and (b) peaking trend during wintertime (January 2020). BC emission during these two periods was not impacted by the COVID-19 pandemic. Furthermore, BC Mass peaking in January has been widely reported in cities around the world (Liakakou et al., 2020; Mbengue et al., 2020; Tan et al., 2020). Accordingly, we believe the conclusion on bi-modal distribution is likely reliable in this study.

The first lockdown in Montreal started on March 16, 2020 and lasted for more than 2 months. The city partially reopened starting from May 25, 2020. During the lockdown, most the unnecessary activities were restricted. Transportation and industrial production decreased dramatically. With restricted human activities, emissions of BC and other air pollutants from anthropogenic sources thereby decreased.

BC Mass variation before the lockdown, during the lockdown and partial reopening in downtown Montreal, is shown in Figure 2A and Figure S4a in Supporting Information S1. The average BC Mass were 0.340 ± 0.014 μg/m3, 0.268 ± 0.004 and 0.551 ± 0.012 μg/m3, respectively. The statistical analysis showed that, there was a significant difference when comparing BC Mass during the lockdown with BC Mass before the lockdown or partial Reopen (Table 3). Since the lockdown spanned from mid-March to the end of May, the seasonal change effect on BC Mass distribution was considered. To exclude the impacts from seasonal changes, the statistical analysis was conducted for BC Mass from March 1 to 15 (before lockdown) with BC Mass from March 16 to March 31 (during lockdown) and BC Mass from May 1 to May 24 (during lockdown)
### Table 1

*Selected Variations of Air Pollutants During Lockdown All Over the World*

| Location          | Pollutant | Variations (%) | Comments                                      | References                  |
|-------------------|-----------|----------------|-----------------------------------------------|-----------------------------|
| California, USA   | NO<sub>2</sub> | −38%           | March 19–May 7, 2020, compared with January 26–March 18, 2020 | Liu, Sha et al. (2020) |
|                   | CO        | −49%           |                                               |                             |
|                   | PM<sub>2.5</sub> | −31%         |                                               |                             |
|                   | O<sub>3</sub> | +14%          |                                               |                             |
|                   | PM<sub>10</sub> | +10%          |                                               |                             |
| Seoul, Korea      | NO<sub>2</sub> | −39.91%        | March 2020 compared with March 2019          | Park et al. (2021)          |
|                   | CO        | −15.25%        |                                               |                             |
| Barcelona, Spain  | CO        | < −35%         | April 2020, compared with April 2019         | Briz-Redón et al. (2021)   |
|                   | NO<sub>2</sub> | < −35%         |                                               |                             |
|                   | NO<sub>3</sub> | +15%–25%      |                                               |                             |
|                   | PM<sub>10</sub> | −35% ~ −25%   |                                               |                             |
|                   | SO<sub>2</sub> | < −35%         |                                               |                             |
| UK                | NO<sub>2</sub> | −42%           | March 23–May 31, 2020, compared with the same period 2015–2019 | Lee et al. (2020)          |
|                   | O<sub>3</sub> | +11%           |                                               |                             |
| Wuhan, China      | PM<sub>2.5</sub> | −36.9%        | January 24–February 23, 2020, compared with December 24, 2019–January 23, 2020 | Lian et al. (2020)          |
|                   | NO<sub>2</sub> | −53.3%         |                                               |                             |
|                   | O<sub>3</sub> | +116.6%        |                                               |                             |
|                   | PM<sub>10</sub> | −40.2%        |                                               |                             |
|                   | CO        | −22.7%         |                                               |                             |
|                   | SO<sub>2</sub> | −3.9%          |                                               |                             |
| NCT Delhi, India  | PM<sub>10</sub> | −51.58%       | March 25–April 14, 2020, compared with March 2–March 21, 2020 | Mahato et al. (2020) |
|                   | PM<sub>2.5</sub> | −53.11%       |                                               |                             |
|                   | SO<sub>2</sub> | −17.97%        |                                               |                             |
|                   | NO<sub>2</sub> | −52.68%        |                                               |                             |
|                   | CO        | −30.35%        |                                               |                             |
|                   | O<sub>3</sub> | +0.78%         |                                               |                             |
|                   | NH<sub>3</sub> | −12.33%        |                                               |                             |
| São Paulo, Brazil | CO        | −29.8%         | March 24–April 20, 2020, compared with February 25–March 23, 2020 | Nakada and Urban (2020)  |
|                   | PM<sub>10</sub> | +7.7%         |                                               |                             |
|                   | PM<sub>2.5</sub> | −0.3%         |                                               |                             |
|                   | NO        | −40.4%         |                                               |                             |
|                   | NO<sub>2</sub> | −21.5%         |                                               |                             |
|                   | O<sub>3</sub> | +10.8%         |                                               |                             |
| Northern China    | SO<sub>2</sub> | −6.76%         | January 28–March 21, 2020, compared with January 1–27, 2020 | Bao and Zhang (2020)       |
|                   | PM<sub>2.5</sub> | −5.93%        |                                               |                             |
|                   | PM<sub>10</sub> | −13.66%        |                                               |                             |
|                   | NO<sub>2</sub> | −24.67%        |                                               |                             |
|                   | CO        | −4.58%         |                                               |                             |
| Malaysia          | PM<sub>10</sub> | −26% ~ −31%   | March 18–April 30, 2020, compared with the same period in 2018 and 2019 | Kanniah et al. (2020) |
|                   | PM<sub>2.5</sub> | −23% ~ −32%   |                                               |                             |
|                   | NO<sub>2</sub> | −63% ~ −64%    |                                               |                             |
|                   | SO<sub>2</sub> | −9% ~ −20%     |                                               |                             |
|                   | CO        | −25% ~ −31%    |                                               |                             |
with BC Mass during May 25 to March 30 (partial reopening). As shown in Table S1 in Supporting Information S1, significant differences between these time periods were observed. The results were not surprising as BC is generated by anthropogenic sources in urban areas. When the lockdown policy restrained the BC emissions, BC Mass certainly decreased too.

The impact of COVID-19 on other air pollutants, rather than only BC were illustrated in Table 3 and Figure 2. The concentrations of PM$_{2.5}$, CO, NO, and NO$_2$ decreased from ~26% and up to 72%. Yet, the concentration of O$_3$ increased by about 30% likely due to the declining emission of other photochemical-related pollutants including CO and NO$_2$. Fan et al. (2021), Huang et al. (2021) and Putaud et al. (2021) attributed the increment of O$_3$ to the decline of NO$_x$ emission during the lockdown. As NO emission decreased, the number of NO$_2$ converted by NO decreased. Additionally, since there was less NO titrating O$_3$, the concentration of O$_3$ increased correspondingly. Yet, Fan et al. (2021) and Huang et al. (2021) observed enhanced secondary pollution in China, which was not observed by Putaud et al. (2021) in northern Italy and this work in Montreal, Canada, which may be due to a lack of data on volatile OAs and PM$_{10}$ in Montreal, or because of regional disparity. Although BC itself is photochemical inert, organic carbon internal or external mixed with BC can be oxidized by O$_3$. Thus, the decreasing emission of BC also contributes a part to the increment of O$_3$. However, the situations in the period of partial Reopen are more complex. The partial reopening was a gradual process during which the restrictions were gradually and selectively lifted. Not all urban activities were fully recovered. Consequently, the recovery of O$_3$, CO, NO, and NO$_2$ emissions was not as clear as during the complete lockdown (Figure S4 in Supporting Information S1). The recovery of BC and PM$_{2.5}$ emissions was statistically clearer, as shown in Figure S4 in Supporting Information S1. With the restrictions removed gradually, a sharp increase of mass concentrations of BC and PM$_{2.5}$, notwithstanding the impact of seasonal changes should be also considered.

As depicted in Figure 1a, BC Mass decreased since July 2019, indicating the existence of another peak around July 2019. During the partial reopening period from May 25 to June 30, 2020, the BC trend was in accordance with BC values in 2019. The recovery of PM$_{2.5}$ was less pronounced than BC. It can be explained partly by the fact that PM$_{2.5}$ contains not only part of BC but also other non-BC particulate matter emitted by anthropogenic sources (Rahim et al., 2019). The observation of low concentrations of CO, NO, and NO$_2$ are in accordance with transportation and industries which gradually recovered since June 2020.

The size of BC particles spans from nano-size to micro-size (Chakrabarty et al., 2014; Long et al., 2013). As such, BC particles greater than 2.5 μm are not included in PM$_{2.5}$ values. The variations of BC and PM$_{2.5}$ are not expected to be identical, as herein confirmed. It is to note that $B_{\text{scat}}$ values may reflect the variation of particulate matters in wider size distribution, as the scattering chamber used in this study, responds to all particles, regardless of their size, morphology, and chemical composition.

As is shown in Table 3 and Figure S2 in Supporting Information S1. A $B_{\text{scat}}$ decreased from $11.30 \pm 0.39$ to $5.60 \pm 0.08$ Mm$^{-1}$ because of COVID-19 but increased back to $12.12 \pm 0.52$ Mm$^{-1}$ during the partial Reopen period, almost at the same levels to $B_{\text{scat}}$ before lockdown. Yet, seasonal changes are likely play a role. As shown in Figure S3 in Supporting Information S1, an increase of $B_{\text{scat}}$ in June 2019 was observed, coinciding with solar radiation hinting to the importance of seasonal changes. Yet, in addition to photochemistry, the recovery of $B_{\text{scat}}$ can also be in part, due to the reopening period in June 2020.

Table 2 and Table S4 in Supporting Information S1 provide detailed information for a suite of air pollutants. The impact of COVID-19 lockdown is statistically clear. Moreover, the variations of BC Mass, PM$_{2.5}$, CO,
Table 2
The Averages, Standard Deviations, Median Values, 99th Percentile Values, 1st Percentile Values, 25th Percentile Values, and 75th Percentile Values for BC Mass, O$_3$, PM$_{2.5}$, CO, NO, NO$_2$, and B$_{scat}$ From January 2020 to June 2020

|                      | Average | Std   | Median | 99%  | 1%   | 25%  | 75%  |
|-----------------------|---------|-------|--------|------|------|------|------|
| **BC Mass (μg/m$^3$)**|         |       |        |      |      |      |      |
| January 2020          | 0.433   | 0.252 | 0.418  | 1.09 | 0.060| 0.248| 0.569|
| February 2020         | 0.336   | 0.226 | 0.320  | 0.991| 0     | 0.173| 0.447|
| March 2020            | 0.310   | 0.269 | 0.280  | 1.18 | 0     | 0.152| 0.401|
| April 2020            | 0.245   | 0.170 | 0.244  | 0.743| 0     | 0.127| 0.334|
| May 2020              | 0.343   | 0.251 | 0.301  | 1.12 | 0     | 0.186| 0.466|
| June 2020             | 0.563   | 0.275 | 0.525  | 1.42 | 0.102| 0.367| 0.714|
| **O$_3$ (ppb)**       |         |       |        |      |      |      |      |
| January 2020          | 18.6    | 8.6   | 19.3   | 34.4 | 1.1  | 12.4 | 25.4 |
| February 2020         | 24.2    | 9.4   | 26.5   | 42.5 | 0.8  | 19.2 | 30.1 |
| March 2020            | 30.0    | 7.9   | 31.3   | 43.3 | 3.7  | 25.8 | 35.4 |
| April 2020            | 33.4    | 7.4   | 33.9   | 48.6 | 10.8 | 29.3 | 38.7 |
| May 2020              | 33.4    | 9.8   | 32.5   | 59.8 | 10.7 | 26.9 | 39.3 |
| June 2020             | 31.3    | 12.4  | 30.6   | 59.7 | 6.8  | 21.4 | 39.4 |
| **PM$_{2.5}$ (μg/m$^3$)**|       |       |        |      |      |      |      |
| January 2020          | 8.6     | 7.2   | 7.3    | 37.2 | 0.7  | 3.8  | 10.5 |
| February 2020         | 10.0    | 8.8   | 7.1    | 41.9 | 1.1  | 4.4  | 12.7 |
| March 2020            | 6.5     | 5.0   | 5.2    | 25.3 | 0.7  | 3.3  | 8.0  |
| April 2020            | 5.2     | 2.7   | 4.5    | 13.5 | 0.8  | 3.2  | 6.7  |
| May 2020              | 5.3     | 3.0   | 4.7    | 13.9 | 0.4  | 3.2  | 6.8  |
| June 2020             | 7.7     | 7.5   | 6.4    | 31.9 | 1.1  | 4.2  | 9.2  |
| **CO (ppb)**          |         |       |        |      |      |      |      |
| January 2020          | 240.3   | 75.1  | 225.8  | 545.6| 134.5| 191.8| 262.5|
| February 2020         | 243.5   | 91.2  | 215.4  | 583.5| 139.2| 188.1| 258.2|
| March 2020            | 203.5   | 54.2  | 193.8  | 397.1| 146.2| 172.6| 219.2|
| April 2020            | 166.8   | 23.1  | 163.6  | 239.6| 130.9| 150.4| 180.3|
| May 2020              | 166.2   | 34.2  | 161.9  | 274.5| 107.2| 141.9| 183.0|
| June 2020             | 161.4   | 39.7  | 158.3  | 276.5| 96.4 | 134.5| 179.4|
| **NO (ppb)**          |         |       |        |      |      |      |      |
| January 2020          | 6.0     | 7.5   | 3.9    | 42.9 | 0.7  | 2.4  | 6.5  |
| February 2020         | 6.1     | 11.4  | 3.2    | 54.5 | 0.6  | 1.8  | 5.7  |
| March 2020            | 3.0     | 6.2   | 1.8    | 36.4 | 0.1  | 0.9  | 3.2  |
| April 2020            | 1.4     | 2.0   | 0.8    | 9.8  | 0.0  | 0.4  | 1.5  |
| May 2020              | 1.2     | 2.0   | 0.6    | 10.5 | 0.0  | 0.3  | 1.2  |
| June 2020             | 1.4     | 3.1   | 0.5    | 15.4 | 0.0  | 0.2  | 1.3  |
| **NO$_2$ (ppb)**      |         |       |        |      |      |      |      |
| January 2020          | 17.5    | 8.7   | 16.0   | 43.1 | 4.9  | 11.3 | 22.1 |
| February 2020         | 17.1    | 9.9   | 13.9   | 45.5 | 4.9  | 9.8  | 21.0 |
| March 2020            | 11.3    | 6.9   | 9.8    | 36.9 | 3.0  | 6.4  | 14.5 |
| April 2020            | 6.8     | 4.8   | 5.8    | 27.1 | 1.8  | 3.6  | 8.3  |
| May 2020              | 5.5     | 3.9   | 4.6    | 19.6 | 0.9  | 2.8  | 7.0  |
| June 2020             | 6.6     | 4.0   | 5.5    | 21.9 | 1.6  | 3.9  | 8.0  |
and B\text{scat} showed the lockdown not only decreased average concentrations of certain pollutants but also median values, 99th percentile values and 75th percentile values.

### 3.3. Case Study: YUL Airport BC in Comparison to Montreal Downtown

Aviation is one of the domains that suffered a lot because of the COVID-19 Pandemic (ICAO, 2021). The worldwide lockdown, restrictions on international travel and high risk of infection in airplanes drastically reduced the number of travelers and flights, which in the end caused the big decline of aviation in the world (ICAO, 2021). Moreover, previous studies have shown that up to 97% of airborne particles, including PM$_{2.5}$ which includes significant BC particles, are being released within airport regions (Camero, 2019; Mazaheri et al., 2011). YUL Airport is one of the third busiest airports in Canada with more than 236 thousand flights in 2019. It is a good place to show the impact of COVID-19 on aviation and the difference between urban areas (downtown Montreal) with the airport.

The atmospheric impacts of COVID-19 lockdown are shown in Tables 4 and 5, Figure 3, and Figure S2b in Supporting Information S1. Similar to downtown Montreal, BC Mass, B\text{scat} and concentrations of PM$_{2.5}$, CO, NO, and NO$_2$ significantly decreased when lockdown started, in contrast to the concentration of O$_3$ significantly which was increased. It should be noticed that before lockdown, the concentrations of all these pollutants, except BC, are not distinct between downtown and the airport (Table 5). Notwithstanding than in previous studies, where the nanoparticles were measured, there was a clear increase close to the airport in comparison to the downtown (Rahim et al., 2019).

The trend for BC is different from other measured pollutants as shown in Table 5. BC Mass at the airport is significantly greater than that in downtown Montreal before lockdown. The relatively smaller B\text{scat} at the airport indicates the particles at the airport intend more to absorb light, instead of scattering light, revealing the higher proportion of BC in the air at the airport, compared with downtown Montreal.

Yet during the lockdown, there is no significant difference between these two regions in BC Mass, as most flights were stopped, and other anthropogenic activities were decreased concurrently. Furthermore, the change of B\text{scat} also gives clues to the BC trends at the airport.

The comparison of BC Mass between downtown and the airport not only reflects the great impact of COVID-19 on aviation but also reveals the high concentrations of BC in the ambient air at the airport during the normal operation, which may pose a potential threat to the health of workers and nearby residents, living a few meters away from the airport.

### 3.4. Evidence for Airborne Black Carbon Deposition in Snow

The HR-S/TEM-EDS results (Figure 4) on the snow samples, both freshly falling and aged snow, collected from the airport illustrate several BC-types particles and carbon nanostructures in snow. There are several processes that can explain the existence of these BC type and carbon nanostructures, which have been observed previously in the air (Rahim et al., 2019), and herein observed, in freshly falling and aged snow. It is noteworthy that previous studies have demonstrated that common BC structure, like those in Figure 4, can
integrate and form aggregates of carbon spheres (e.g., Bond et al., 2013). These combustion-related particles are likely scavenged by snow crystals as it falls. The large surface areas of snowflakes may facilitate the partitioning between snow and particles or chemicals in the atmosphere, making snow an excellent scavenger when it falls (Lei & Wania, 2004). Furthermore, both wet and dry deposition processes of anthropogenic particles on snow have been previously observed (Hadley et al., 2010; Liu, He, et al., 2020). Upon the multiple precipitations and melt-processes, several BC particles or aggregates were observed in the snowpack, confirmed by HR-S/TEM analysis (Figure 4).

Several researchers have indicated that BC can serve as effective ice nuclei (Brooks et al., 2014; DeMott, 1990; McCluskey et al., 2014; Murray et al., 2012). The importance of BC on atmospheric ice nucleation processes and the magnitude of the radiative forcing due to BC on snow, are still being debated (Brooks et al., 2014; Fan et al., 2016; Kulkarni et al., 2016; McCluskey et al., 2014; Petters et al., 2009; Qian et al., 2014; Vergara-Temprado et al., 2018). BC particles in the snow might likely have different origins, configuration, composition, and other physicochemical properties (Bond et al., 2013; Brooks et al., 2014; Dou & Xiao, 2016; Lack et al., 2014; Liu, He, et al., 2020; McCluskey et al., 2014; Qian et al., 2014). As such, air-snow partitioning is expected to occur both on the surface of the snow and on particle interfaces (Lei & Wania, 2004). Recently, there are increasing evidence that several types of anthropogenic particles that are found in snow can alter the ice nucleation of BC and its radiative forcing (Ganguly & Ariya, 2019; Ming & Wang, 2021).

### Table 3

| Air Pollutants Concentrations With Standard Deviations in Downtown Montreal During COVID-19 Pandemic |
|---------------------------------------------------------------|
| Downtown Montreal                                             |
| Before lockdown | During lockdown | Partial reopen | P value |
|-----------------|-----------------|----------------|---------|
| BC Mass (µg/m³)  | 0.340 ± 0.014   | 0.268 ± 0.004  | 0.551 ± 0.012 | <0.001 | <0.001 |
| PM₂.₅ (µg/m³)   | 9.19 ± 25       | 5.12 ± 0.06    | 7.50 ± 0.24 | <0.001 | <0.001 |
| CO (ppb)         | 232.8 ± 2.6     | 171.6 ± 0.7    | 165.0 ± 1.4 | <0.001 | 0.003 |
| O₃ (ppb)         | 25.8 ± 0.3      | 33.2 ± 0.2     | 31.2 ± 0.4  | <0.001 | 0.226 |
| NO (ppb)         | 5.4 ± 0.3       | 1.5 ± 0.05     | 1.4 ± 0.1   | <0.001 | 0.755 |
| NO₂ (ppb)        | 15.6 ± 0.3      | 7.0 ± 0.1      | 6.4 ± 0.1   | <0.001 | 0.192 |
| Bₙₙ₉ (Mm⁻¹)     | 11.30 ± 0.39    | 5.60 ± 0.08    | 12.12 ± 0.52 | 0.001 | 0.001 |

**Note:** The first column of P value shows the P values between the concentrations before lockdown and the concentrations during lockdown. The second column of P value shows the P values between the concentrations during lockdown and the concentrations after partial reopen. The period of “Before lockdown” is set from February 1 to March 15. The period of “During lockdown” is set from March 16 to May 24, and the period of “Partial Reopen” spans from May 25 to June 30.

Externally or internally mixed organic pollutant particles like PAHs, a known health hazard associated with the BC-type particles, also undergo physicochemical processes (Nazarenko et al., 2016, 2017). For instance, after snow melts, the aggregated BC particles may be dissolved in meltwater or released into the air. As snow ages, the partitioning coefficient of particles and compounds in snow is expected to vary, leading to the re-emission (Hansen et al., 2006). Furthermore, as the wind blows across the surface of the snowpack, the decreased pressure on the surface can induce the re-emission of particles from snow surfaces too (Hansen et al., 2006). Due to the high importance of BC in ice nucleation, radiation as well as human and ecosystem health, further targeted physicochemical research is recommended.

### 3.5. Airborne Black Carbon Decrease During the Snowfall

Table S6 in Supporting Information S1 shows the impacts of snow on airborne BC Mass values, using one snowfall event. Snow precipitation reported values are averaged over 24 hr. This snowfall event occurred...
Table 5
Comparisons of Concentrations of Air Pollutants in Downtown Montreal With Near YUL Airport During COVID-19 Pandemic

|                      | Downtown | Airport | P value |
|----------------------|----------|---------|---------|
| Before lockdown (February) |          |         |         |
| BC Mass (µg/m³)       | 0.344 ± 0.016 | 1.487 ± 0.148 | 0.005   |
| PM₂.₅ (µg/m³)         | 10.0 ± 0.3   | 10.5 ± 0.4 | 0.188   |
| CO (ppb)              | 243.5 ± 3.4  | 250.8 ± 4.7 | 0.212   |
| O₃ (ppb)              | 24.2 ± 0.4   | 23.7 ± 0.4 | 0.622   |
| NO (ppb)              | 6.1 ± 0.4    | 6.9 ± 0.7  | 0.466   |
| NO₂ (ppb)             | 17.1 ± 0.4   | 15.3 ± 0.5 | 0.090   |
| B⁺scat (Mm⁻¹)         | 12.50 ± 0.58 | 7.68 ± 0.96 | <0.001  |
| During lockdown (May) |          |         |         |
| BC Mass (µg/m³)       | 0.290 ± 0.008 | 0.347 ± 0.045 | 0.079   |
| PM₂.₅ (µg/m³)         | 4.9 ± 0.1    | 4.5 ± 0.1  | 0.054   |
| CO (ppb)              | 162.0 ± 1.1  | 146.5 ± 1.1 | <0.001  |
| O₃ (ppb)              | 34.1 ± 0.4   | 34.9 ± 0.3  | 0.583   |
| NO (ppb)              | 1.2 ± 0.1    | 0.5 ± 0.03  | 0.006   |
| NO₂ (ppb)             | 5.5 ± 0.2    | 3.3 ± 0.2  | <0.001  |
| B⁺scat (Mm⁻¹)         | 5.17 ± 0.15  | 3.64 ± 0.15 | <0.001  |

Note. The period of “Before lockdown” is set from February 1 to 29. The period of “During lockdown” is set from May 1 to 24.

In part of the December 31, 2019 with less intensity and continued to much higher precipitation intensity on January 1, 2020, whereas January 2, 2020 received no precipitation. The average daily temperatures over 3 days are nearly constant around the freezing point. During the snow-fall events, BC values decreased, particularly when snow fall intensified, whereas during the snow free day of January 2, 2020, BC Mass values recovered to the highest levels on three days. Thereby snowfell scavenged BC particles bringing it to the surface. This observation is in accordance with the results from Hadley et al. (2010) who also found the evidence that snow could scavenge BC and decrease BC Mass in ambient air. Hadley et al. (2010) have performed their statistical analysis and drew conclusions based on the data of several snow events at three different sites. In this study, we lacked enough data for snow events during the winter of 2019–2020 to draw a statistically valid estimation of scavenging processes, yet it should be considered in future studies.

Beside BC, other emerging contaminants are found in snow, including carbon nanostructure (Rangel-Alvarado et al., 2019), microplastics (Wang et al., 2021), nanoparticles (Pal et al., 2020; Rahim et al., 2019; Rangel-Alvarado et al., 2015), and halogen compounds (Hall et al., 2020). More importantly, recent research reveals that microplastics in snow may cause an overestimation of BC and its effects on the climate since several analytical equipment for BC detection are unable to distinguish microplastics from BC (Ming & Wang, 2021). This finding indicates the questions of whether and how BC will interact with other emerging contaminants, for instance, toxic nano-metals such as Ni and Cr, as observed in this study (Figure S7 in Supporting Information S1). Since BC has been shown to contain both organic and inorganic compounds, and various organic compounds are known to interact on particles (Canagaratna et al., 2015; Eltouny & Ariya, 2012). Potential effects upon the photochemical or heterogeneous chemical transformation of a wide range of particles should be understood. Future research on these topics is thus recommended.

3.6. Potential Added Values of This BC Data for Cold-Climate Modeling

This work provides data of both BC Mass and B⁺scat in a cold-climate model city of Montreal, which is beneficial for present models of radiative forcing based on anthropogenic BC emissions in urban areas. Data on number density, size, refractive index, mass concentration and absorption coefficient of BC have been identified as key inputs to evaluate the concentrations and radiative forcing of BC in most modeling (Bond et al., 2013). Such data are rare for cold-climate cities. Considering the long winter and long duration of high BC Mass in cold urban regions, as discussed above, it is greatly recommended for future work in developing regional cold-climate models to evaluate urban air quality and climate impacts.

4. Concluding Remarks

We herein present one-year real-time data on mass concentration of BC, together with key air co-pollutants, in a model cold-climate city of Montreal, which is crucial for various atmospheric modeling. Annual BC Mass concentration exhibited a bi-modal distribution and BC winter peak lasts longer than 3 months in comparison to mild or warm-climate cities, due to distinct cold-climate meteorology. During the snow episodes, airborne BC Mass concentration decreased, while BC particles or carbon nanomaterials appeared in the snow, indicating air-snow interaction of anthropogenic pollutants took place. Furthermore, we demonstrated that airborne BC Mass concentration in the residential areas around the airport is over 400% higher than an already in a polluted downtown. During the COVID-19 lockdown period, the emission of BC and other co-pollutants decreased significantly, yet they recovered as partial opening implemented. A large part of the planet experiences cold temperatures and frozen participation every year, and it is important to implement an air quality management system, which considers the effects of the cold climate urban sites. The
Figure 2. Diurnal variation of the concentrations of black carbon (a), O$_3$ (b), PM$_{2.5}$ (c), CO (d), NO$_x$ (e), and NO (f) in downtown Montreal before lockdown (black line) and during lockdown (red line). The period of “Before lockdown” is set from February 1 to March 15, and the period of “During lockdown” spans from March 16 to May 24.
emission reduction of airborne BC and selected co-pollutants in the airports and surrounding areas, which may pose threat to the health of airport workers and residents should be tailored to different urban climates. Future research on BC sources, physicochemical characteristics at different environmental conditions are recommended. Ice nucleation microphysics of BC and interactions with brown carbon should also be further studied, to accurately evaluate the impact of BC in air quality, climate change and health research.

Figure 3. Comparisons of concentrations of black carbon (a), O$_3$ (b), PM$_{2.5}$ (c), CO (d), NO$_2$ (e), and NO (f) in downtown Montreal (cyan) with near YUL Airport (black) in COVID-19 Pandemic. For panel (a), the period of “Before lockdown” is set from February 1 to 29, and the period of “During lockdown” is from May 1 to 24. For panels (B–F), the period of “Before lockdown” includes February 1 to March 15, and the period of “During lockdown” spans from March 16 to May 24.
Figure 4. High-resolution scanning/transmission electron microscopy with energy dispersive X-ray spectroscopy (EDS) results of the aged snow samples collected from the airport. Panels (a–c) are from the same sample. Panels (b, c) are zoomed in from Panels (a, b), respectively. Panel (e) indicates another sample. Panels (d, f) are EDS results for Panels (c, e), respectively.
Data Availability Statement

Data of absorption coefficient and scattering coefficient have been published on Harvard Dataverse and can be downloaded at https://doi.org/10.7910/DVN/F63JBI. Data of PM$_10$, CO, O$_3$, NO$_2$, and NO can be found on the NAPS website at https://data-donnees.ec.gc.ca/data/airmonitor/national-air-pollution-surveillance-naps-program/?lang=en, as well as the website of City of Montreal: http://ville.montreal.qc.ca/portal/page?_pageid=7237,74687650&_dad=portal&_schema=PORTAL.

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