

**DETERMINING THE TEMPORAL AND SPATIAL VARIATIONS OF
POLLUTANTS IN TORONTO**

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Abstract

This thesis aims to assess the spatial and temporal variations of pollutants in Toronto. Annual, seasonal, and monthly pollutant trends were computed from as early as 1974 to as late as the end of 2022. Decreasing NO, NO₂, NO_x, and CO trends were found, perhaps associated with the elimination of coal-fired power plants, and vehicular technology improvements, leading to emissions reductions. Applying two different techniques to trace pollutant measurements from air quality stations upwind, NO, NO₂, NO_x, and CO concentrations were found to be relatively high around Downsview and Pearson Airport, as well as the York University power plant. Additionally, relatively high NO₂ and NO_x levels were denoted around Highway 401, suggesting vehicular and aircraft emissions sources. The greatest concentrations of PM_{2.5}, PM₁₀, O₃, and SO₂ were concluded to originate from outside the city. Income information was obtained by neighborhood and related to the spatial distribution of pollutants in Toronto. A negative relationship was found between NO and NO_x levels and income. This is because the lowest and highest income neighborhoods (generally located in north and south Toronto, respectively) mostly coincided with relatively high and low NO, NO_x concentrations, respectively. TROPOMI satellite measurements over a gridded plane in Toronto also display the highest and lowest NO₂ and HCHO levels over Pearson Airport and Lake Ontario.

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1 Introduction

Pollution can result in negative impacts on human health (Lanzi et al., 2016; Manisalidis et al., 2020), the environment (Manisalidis et al., 2020), and the economy (Lanzi et al., 2016). Exposure to elevated levels of near-surface ozone may result in adverse cardiovascular and respiratory effects (Manisalidis et al., 2020). Particulate matter is notated by PM_x , which represents the concentration of particulate matters with diameter $\leq x$ μm . $PM_{2.5}$ infiltrates the respiratory system (Air Quality Ontario, n.d-a), and exposure to particulate matter can induce central nervous, reproductive system malfunctions, cardiovascular and respiratory diseases (Manisalidis et al., 2020). Adverse effects can also be observed from exposures to NO_2 (Air Quality Ontario, n.d.-b; Manisalidis et al., 2020), SO_2 (Air Quality Ontario, n.d.-c; Manisalidis et al., 2020), CO (Air Quality Ontario, n.d.-e; Manisalidis et al., 2020), and volatile organic compounds (VOCs)/polycyclic aromatic hydrocarbons (PAHs) (Manisalidis et al., 2020). In particular, toxicity is seen in high doses of CO (ibid.). CO also prohibits oxygen into tissues and organs, especially in people with heart disease, when inside the circulatory system (Air Quality Ontario, n.d.-e). A premature death is expected every 5 seconds from ambient air pollution by 2050 (Lanzi et al., 2016). Many premature deaths are observed in India, China (congested areas with excess $PM_{2.5}$ and ozone levels), as well as eastern Europe (ibid.). From 2010 to 2060, the percentage of people exposed to yearly-averaged $PM_{2.5}$ of $> 35 \mu g/m^3$ is expected to increase from 15 to 65%, and 40 to 60%, in India and China respectively (ibid.). Overall, ambient air pollution is predicted to be responsible for 6-9 million premature deaths in the year 2060, globally (ibid.).

Pollution can also adversely impact wildlife (Manisalidis et al., 2020). Denoted congenital disabilities and the inability to reproduce in animals have been observed (ibid.). Plants, trees, and soil and water habitats have been harmed due to acid rain (ibid.). Nitric acid from NO_2 deposition factors into acidification of lakes (Air Quality Ontario, n.d.-b). SO_2 is also a precursor for acid rain, resulting in acidification (Air Quality Ontario, n.d.-c). Crops and trees may also be harmed from nitric acid (Air Quality Ontario, n.d.-b), and SO_2 (Air Quality Ontario, n.d.-c). Considerable economic impacts will be seen as a result of rising ozone and $PM_{2.5}$ concentrations (Lanzi et al., 2016). For example, worldwide yearly absent days from work are expected to be 3.7 billion by 2060 (ibid.). Furthermore, negative impacts on the global economy are projected due to effects on healthcare spending, crop outputs, and workforce productivity from ambient pollutants (ibid.).

Pollution from primary sources can react to produce secondary pollutants (Henneman et al., 2015). Localized pollutant emissions include factories or power plants, while emissions from farming and transportation are more widespread (European Environmental Agency, 2024). Particulate

matters encompass pollen, fumes, smoke, ash, dust and aerosols, which are either airborne liquid droplets or suspended particles (Air Quality Ontario, n.d-a). Particulate matter is typically created via fuel combustion reactions (ibid.). The elderly, children, and those with lung/cardiovascular disease, and asthma are the most vulnerable groups to $PM_{2.5}$ (ibid.). Premature deaths, and hospitalizations have resulted from $PM_{2.5}$ exposures (ibid.). Forest fires, automobiles, burning of crop-residue, power-plants, household fireplaces, factories, wooden stoves, and smelters represent the greatest sources of particulate matters (ibid.).

Fossil fuel combustion and biological activities create nitric oxides (Anderson & Mansfield, 1979). Nitrogen and oxygen under high temperature from combustion result in the formation of NO (ibid.). NO_2 , is a brownish-red gas, with a strong, unpleasant smell (Air Quality Ontario n.d.-b), and NO_x is equal to NO plus NO_2 . Industrial sources and utilities are primary origins of NO_x (nitrous oxide) (Air Quality Ontario n.d.-b), as well as transportation sources (Air Quality Ontario n.d.-b; Hagenbjörk et al., 2017). NO_x is mostly comprised of NO_2 (Air Quality Ontario, n.d.-b). Unlike NO_2 , both SO_2 (Air Quality Ontario, n.d.-c), and O_3 (Air Quality Ontario, n.d.-d), are gasses with no distinct color. Factories, electric utilities, and smelting are primary contributions to SO_2 concentrations (Air Quality Ontario, n.d.-c). Other contributors are from household and vehicular emissions (ibid.). Primary emitters of ozone do not exist (Air Quality Ontario, n.d.-b). More detailed mechanisms regarding ozone production from primary emissions are discussed in the next chapter. CO is a toxic colorless gas, with no smell or taste (Air Quality Ontario, n.d.-e). It mainly created from incomplete fossil fuel combustion (ibid.). Ultrafine particles are defined as particulate matters $\leq 0.1 \mu m$ in size ($PM_{0.1}$) (Schraufnagel, 2020). Vehicular, power plant and industrial emissions are primary sources of UFPs (Morawska et al., 2008). Other sources include burning of biomass, tobacco smoking, as well as indoor sources such as heater/stove combustion (ibid.). UFPs first enter the lungs, then from the lung vasculature, make their way to other organs (Schraufnagel, 2020). Although the adverse impacts of $PM_{0.1}$ aren't widely documented, the most common effect from UFP exposure is metal fume fever, which induces chills, arthralgias, fever, malaise and myalgias (ibid.). Aircraft are also emitters of CO_2 , SO_2 and NO_x (Transport & Environment., n.d.).

Formaldehyde (HCHO) is among the most basic organic molecules (Songur et al. 2003). It is a colorless, irritating, and pungent gas (ibid.). Formaldehyde is typically found in polluted urban environments (i.e., photochemical smog, and incomplete combustion of organics) (Usanmaz et al., 2002), nature, medical settings, industrial settings, and indoor air (Songur et al., 2003). Adverse impacts on the central nervous system, inducing malaise, dizziness, insomnia, headaches, and anorexia can result from inhalation of gaseous HCHO (Harris et al., 1981). Exposure to elevated concentrations of HCHO may also cause cancer (USEPA., 2024).

1.1 Motivation

In cities, pollutants have increasingly posed a significant threat regarding their impacts on human wellness and health (Bikis, 2023). In Toronto, for example, 24% of total respiratory hospitalizations were linked to haze during summer, when temperature was accounted for (Thurston et al., 1994). Therefore, understanding how pollutants vary, spatially and temporally urban locations is vital. Deducing temporal pollutant trends in cities is important for several reasons. For example, one can understand and project the long-term implications on public health, the environment, and the economy. Furthermore, one can monitor how pollutants in urban areas are projected to alter with climate change and how climate change can, in turn, influence pollutant levels in urban areas. Moreover, understanding the spatial variations of urban pollutants may allow one to deduce the number of people impacted and if socioeconomic status plays a role. For example, a study by Dionisio et al. (2010) found that in Accra, Ghana, the greatest PM levels were observed in a district with high population density and low income of Ushertown/Jamestown. Another study by Zhao et al. (2019) assessed how $PM_{2.5}$ varies spatially and temporally from 2015 to 2016 in 269 cities in China. A key result was that smaller cities generally exhibited the highest $PM_{2.5}$ concentration increases, and it was determined that population density was the primary contributor to this.

Despite studies analyzing the spatial/temporal variations of at least one pollutant in Toronto (Arain et al., 2009; Chastko & Adams, 2019; Geddes et al., 2009; Geddes et al., 2016; Jeong et al., 2020; Hilker et al., 2021; Sharisingh et al., 2021; Weichenthal et al., 2016; Whaley et al., 2015; Yang et al., 2010), some only used a short period of time for the spatial variations (Arain et al., 2009; Weichenthal et al., 2016). In contrast, others analyzed temporal variations in Toronto but for no more than two pollutants (Arain et al., 2009; Geddes et al., 2016; Whaley et al., 2015; Yang et al., 2010). As a couple of examples, Arain et al. (2009) analyzed the effects of meteorological variables such as temperature, precipitation, and wind speed on spatial-temporal variability of nitrogen dioxides. However, NO_2 was measured only in 2002 (Arain et al., 2009). Weichenthal et al. (2016) employed a land-use regression (LUR) model, considering UFP observations from 405 road segments to predict average ultrafine particle (UFP) spatial variations throughout Toronto. However, UFP concentrations were only measured from September, and March 2011 (ibid.). Similar to Weichenthal et al. (2016), Jerrett et al. (2007), employed 95 NO_2 measurement instruments throughout Toronto, within a two week period between September 9 to 25, 2002. Using these observations combined with a land-regression model, it was ultimately found that NO_2 was the greatest around Downtown Toronto, and near highways, with the lowest concentrations in north-

east Toronto (ibid.). The main gap between these studies was that they did not utilize a broad range of pollutants to compute Toronto's spatial, temporal pollutant variations, over a long period of time, from observational data.

1.2 Thesis Objectives, Contributions, and Structure

This thesis' objective is to fill the aforementioned gap by determining the temporal and spatial variation of $PM_{2.5}$, PM_{10} , NO, NO_2 , NO_x , CO, O_3 , and SO_2 throughout Toronto via observational data (instead of predicted using land-use regression models), covering an extensive time period. Air quality stations will be employed to compute pollutant trends, and both air quality stations and satellite measurements will be used to map the spatial distributions of pollutants throughout Toronto. Doing so aims to provide a comprehensive approach regarding the behavior, distribution and potential sources of air pollution throughout Toronto. Furthermore, the pollutant spatial distributions will be related to neighborhood population density and income levels to observe if socioeconomic status is an important factor regarding exposures to high pollutant concentrations. This relationship was not assessed by the aforementioned studies.

Chapter 2 will provide background information on various harmful air pollutants, before transitioning to a literature review of pollutant trends in and outside of Toronto. A review of methodologies used to trace pollutants upwind, known as "back-trajectory methods" will also be discussed and compared, as well as some examples of their previous applications. Since satellites measure total atmospheric column pollutants, and this thesis is only concerned with near surface level and lower tropospheric pollutants, information on the planetary boundary layer height will be provided in this chapter as well, including a discussion regarding the relative pollutant levels above and below this height. Temporal pollutant trends will be computed in chapter 3. Particularly, monthly, seasonal, and annual pollutant trends for $PM_{2.5}$, NO, NO_2 , NO_x , O_3 , CO, and SO_2 from four air-quality stations in west, east, north, and downtown Toronto will be determined. Polar plots showing average and 90th percentile concentrations as a function of wind direction were also computed for these station pollutants to get an idea of which directions the dominant sources originate from. This will provide a potential understanding of the potential pollutant sources, both inside and outside of the city. Furthermore, $PM_{2.5}$, NO_2 , O_3 , CO and SO_2 hourly and monthly averages will also be computed to understand and explain their diurnal and seasonal variation.

Chapter 4 aims to create a detailed spatial map of the pollutant variations throughout Toronto. This is done by employing two back-trajectory techniques for 22 air-quality stations, and all their pollutant measurements to trace the pollutants two hours back in time based on wind speed and

direction from the nearest airport weather station. As mentioned above, population density and net median income metrics were gathered by neighborhood in Toronto to observe a relationship between ones socioeconomic status and exposure and pollutant levels. Chapter 5 also discusses ground/near-ground level instruments that measure particulate matter, ozone and SO_2 during two short-term campaigns during the summer 2023, and winter 2024. From these campaigns, polar plots of concentration vs. wind direction were also generated to assess which directions the dominant sources were coming from. The goal of this chapter is to denote if these shorter term measurements provide validation for the longer-term air quality station results. In Chapter 6, TROPOMI measurements of NO_2 , SO_2 , CO , and O_3 as well as a surface O_3 are spatially mapped onto a 250×250 meter gridded space in Toronto. To compare, and asses the validity of the mapped spatial variations from air-quality stations, TROPOMI measurements will be converted to surface concentrations via employing the boundary layer height, and surface level temperature and pressure. More details to follow in section 6.1.

2 Background

This section will explore studies of the general pollutant trends in Toronto and studies discussing some of their contributing factors. Seasonal variations, spatial and temporal variability, and potential sources are also discussed. Pollutant trend analysis will also be reviewed in locations near Toronto, including Windsor, Michigan, and Rochester, NY. Literature is reviewed that discusses and explains global pollutant trends. Studies employing spatial and temporal models to analyze air pollution in Toronto and other global locations will also be discussed.

Studies have found decreasing pollutant concentration trends in Toronto (Geddes et al., 2009; Hilker et al., 2021; Jeong et al., 2020) and its surroundings (Geddes et al., 2009). In the Hilker et al. (2021) study for example, observations close by an active road station in Toronto, situated at the Southern Ontario Center for Atmospheric Aerosol Research (SOCAAR), on the University of Toronto St. George (UTSG) campus, showed a reduction in ultra-fine particles (UFPs), NO_2 , NO , SO_2 , and $\text{PM}_{2.5}$. Ultrafine particle (UFP) number densities decreased from $2.2 \times 10^{-3} \text{ cm}^{-3}$ to $1.2 \times 10^{-4} \text{ cm}^{-3}$ from 2006-2019, reducing by 4% yearly, with the steepest decline being in the wintertime (January, February, March) (ibid.). Hilker et al. (2021), concluded that these decreases were partially due to both Ontario's steady improvement of vehicular technology, and the 2004 to 2015 phasing-out of coal-powered electricity production. A study by Geddes et al. (2009) utilized the National Air Pollution Surveillance (NAPS) network to obtain ozone, NO_x , and VOC measurements throughout the summers of 2000 to 2007. Moreover, ozone and NO_2 observations were gathered from eight stations within and surrounding Toronto from the Air Quality Ontario website (ibid.). It was discovered by Geddes et al. (2009) that summertime ozone concentrations and VOC reactivity have declined by 30-40% in Toronto and neighboring areas; the most significant decrease in Toronto West, Toronto Downtown, and Oshawa, with rates of 1.2-1.4 ppb annually. Geddes et al. (2009) concluded that this is most likely due to automobile improvements, and policies that regulate emissions at the municipal and provincial levels, also similar to the conclusions reached by Hilker et al. (2021) for their pollutant decreases. Moreover, Jeong et al. (2020) found that local vehicular exhaust emissions declined by roughly 3% annually from 2006-2016, also matching emissions cutbacks from traffic-related air pollutants implemented federally and provincially.

2.1 General NO , NO_2 , NO_x , and O_3 Reactions and Relationships

Ozone can be created from NO_2 (Air Quality Ontario n.d.-b; Clapp & Jenkin, 2001; Finlayson-Pitts, 1993; Hagenbjörk et al., 2017). NO_2 can dissociate to NO and O via sunlight, where O_2 can react with O to produce O_3 (Clapp & Jenkin, 2001; Finlayson-Pitts, 1993; Hagenbjörk et al.,

2017). Moreover, O_3 can react with NO to yield NO_2 and O_2 (Clapp & Jenkin, 2001; Hagenbjörk et al., 2017; Jaroszyńska-Wolińska, 2010; Seinfeld & Pandis, 2016; Sillman, n.d.). Reactions of atmospheric peroxides (RO_2), and NO with VOCs allows for a conversion of NO to NO_2 , without ozone depletion, resulting in an ozone increase (Hagenbjörk et al., 2017). The inverse relationship between NO_2 and O_3 is related to NO_x concentrations (Clapp & Jenkin, 2001). There is an ideal VOC: NO_x ratio at which O_3 is maximally created (Seinfeld & Pandis, 2016). At ratios smaller (relatively-high NO_x conditions), and greater than this (relatively-low NO_x conditions), O_3 decreases and increases as a function of NO_x respectively (Geddes et al., 2009; Whaley et al., 2015; Seinfeld & Pandis, 2016; Sillman, n.d.; Zhang et al., 2022). In the elevated NO_x relative to VOC conditions, ozone concentrations will also rise as the concentration of VOCs rises (Sillman, n.d.). In low NO_x /VOC conditions O_3 concentrations are less dependent on VOC concentrations (ibid.). CO can also undergo photo-oxidation with VOCs, NO_x , and methane in the troposphere to produce ozone (Choi et al., 2017).

2.2 Pollutant Trends

2.2.1 Pollutant Trends in Toronto

In the Jeong et al. (2020) study, the $PM_{2.5}$ component information was gathered from March 1, 2004 to May 26, 2014 at co-ordinates (43.6582, -79.3972) in Toronto. Afterwards, the instrument was moved approximately 120 meters northeast to the SOCAAR site at location (43.659, -79.396), with data collection resuming until 2017. The measurements were taken every three days throughout these periods, and daily averaged, (midnight to midnight). These measurements were also gathered through a speciation and dichotomous sequential air sampler. The speciation sampler measured inorganic ions, that were soluble in water (Cl^- , NH_4^+ , Mg^+ , NO_3^- , Na^+) through an Ion Chromatography (IC) analysis. In the dichotomous sequential air sampler, Teflon filters were used to detect for seventeen elements such as silicon, copper, potassium, lead, and selenium. A thermal optical resistance (TOR) procedure, was used to measure pyrolyzed organic carbon (POC), organic carbon (OC), and elemental carbon (EC) components.

Ozone, nitric oxide, and sulfur dioxide were measured continuously between 2004 and 2017 (Jeong et al., 2020). These measurements were taken at location (43.663, -79.3881), from an Ontario Ministry of Conservation and Parks managed station. Ambient particle number densities were measured continuously from 2006 to 2017, at the SOCCAR ground lab. These particles were binned into three ranges; 8-25nm, 25-100nm, and 100-560nm. The PNCs declined by 3.1 to 4.7 percent per year respectively, with the least steep reduction being for the 8-25 nm bin. NO , NO_2 and SO_2 , were found to have declined by 11.1%, 4.7%, and 13.8% per year respectively. It was

also found that NO_x concentrations were unchanged post 2012, despite falling steeply prior (ibid.). This was most likely due to diesel automobile emission declines.

The three measurement locations by Jeong et al. (2020) are displayed in Figure 2.1.

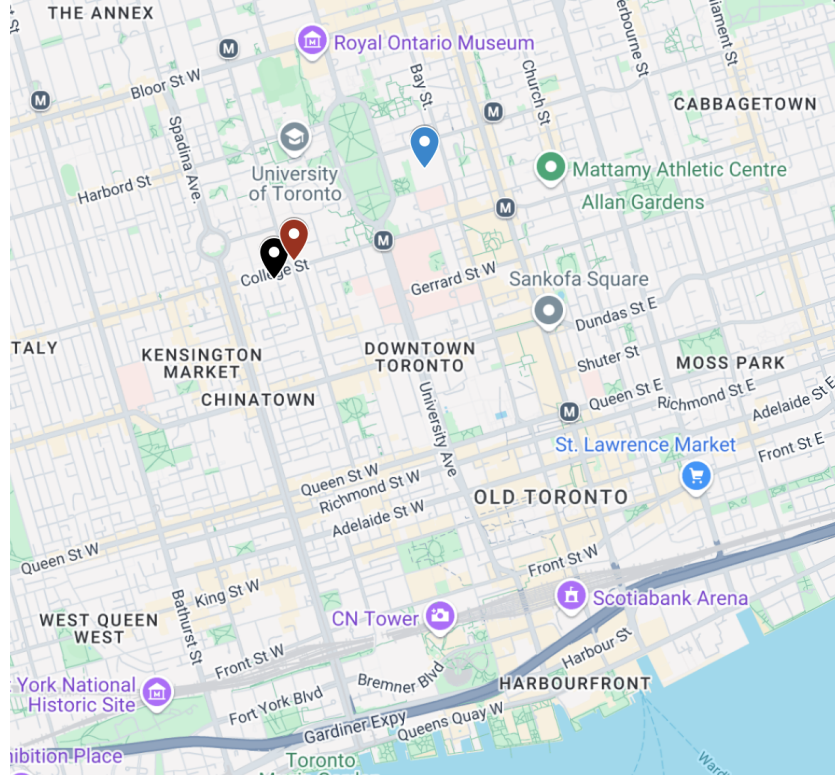


Figure 2.1: Co-ordinates of $\text{PM}_{2.5}$ measurement site pre May 26, 2014 (43.6582, -79.3972), post May 26, 2014 (43.659, -79.396), and O_3 , NO_x , SO_2 (43.663, -79.3881) measurements by Jeong et al. (2020), marked in blue, red, and black respectively.

Jeong et al. (2020) showed that oil combustion, coal combustion, regional environmental carbon (EC), and secondary nitrate decreased by 18%, 10%, 8%, and 8% per year, respectively, leading to a 34% reduction in the yearly-averaged $\text{PM}_{2.5}$ levels during the 14-year period. The aforementioned reductions are in accord with the reduction of emissions from the Great Recession (2008), and policies aimed to reduce coal-fired power stations burning of fossil fuels. The largest reduction in $\text{PM}_{2.5}$ concentrations was seen during winter (December, January, and February) at 3.6% annually. Conversely, non-exhaust emissions (associated with road salt, road dust, metal manufacturing, and soil) remained unchanged from 2004-2011, then, after 2011, rose by about 21% to 27% annually for tire/brake wear particulates and road dust respectively. Non-exhaust emissions were grouped into two categories by Jeong et al. (2020), labelled “non-tailpipe I” which were associated with road/soil dust and “non-tailpipe II”, associated with road salt and metal productions. Since non-vehicular exhaust corresponds with friction between tires and roadways, it should correlate with

vehicle weight. As such, it was postulated that an uptick in light truck sales, such as pick-up trucks and SUVs post-2012, could be related to this increase. These findings are supported by Jones et al. (2012), in a different city, who also found a swift decrease in particle number density level in London, United Kingdom, once pollution charges for large goods vehicles and diesel oil containing no sulfur were implemented in 2007. There are also signs that non-automobile exhaust PM_{2.5} emissions are rising (Jeong et al., 2020). Moreover, a high oxidative activity per PM_{2.5} mass was also associated with non-vehicle exhaust emissions connected with brake wear and road dust (ibid.).

From 2004-2007 to 2013-2016, long-range transport products such as coal and oil combustion emissions, regional EC, and secondary nitrate fell from 79% to 63% of all the PM_{2.5} mass (ibid.). More specifically, secondary nitrate, the greatest factor in wintertime PM_{2.5}, decreased from 52% from 2004-2007 (initial four years of analysis) to 44% from 2013-2016 (final four years of analysis) (ibid.). In the summertime, coal combustion was the dominant source of PM_{2.5}, contributing to an overall 40% of PM_{2.5} mass (ibid.). Non-automobile contributions, primary pollutants from automobiles, metal production, and road salt increased from 13% to 20% of the total PM_{2.5} mass from 2004-2007 and 2013-2016, respectively (ibid.). PM_{2.5} makeup also shifted from being mostly inorganic to metal-rich organic particles within this timeframe, representing an elevated proportion of trace metals relating to emissions from vehicles (ibid.).

Interestingly, Geddes et al. (2009) found no tangible decrease in Toronto ozone concentration trends over their study period (2000-2007). In specific urban locations, ozone could be on the rise (ibid.). Decreased VOC reactivity and NO_x concentrations also had an insignificant influence on ozone concentrations (ibid.). It was also suggested by Geddes et al. (2009), that Toronto is a VOC-limited regime. Whaley et al. (2015) measured lower-tropospheric ozone and carbon monoxide at the University of Toronto's Atmospheric Observatory (TAO), Environment Canada Atmospheric Research Experiments (CARE), and Measurements of Pollution in the Troposphere (MOPITT) and from the Tropospheric Emission Spectrometer (TES), from 2002 to 2013. In high pollution events from 2004 to 2007, Whaley et al. (2015) found that anthropogenic NO_x fossil fuel emissions from all areas in North America, cumulatively contributed to 10-40% of the total impact on Toronto's lower-tropospheric ozone levels. A tertiary contribution to ozone was from soil and lightning emissions (ibid.). In particular, responsiveness to soil emissions was the greatest in the southwest U.S. area (ibid.). This was demonstrated by Whaley et al. (2015) as an example of long-range transport. Over the aforementioned time periods and regions, soil emissions cumulatively summed between 0.5% and 4% of the total low-tropospheric ozone concentration impact (ibid.). This contribution fluctuated heavily for lightning ranging from 1% to 10% (ibid.). These findings are in accord with

the results from Zhang et al. (2022), who showed O_3 concentrations primarily depended on NO_x from 1996-2007 and VOCs from 2008-2015, shifting from a NO_x -limited to VOC-limited regime. Though the study by Zhang et al. (2022) was not carried out in Toronto but in other locations in southwestern Ontario, Michigan, and Ohio from April to September 1996-2015. Whaley et al. (2015), stated that most of North America is a NO_x limited regime, but also suggested that Toronto was a VOC-limited regime (consistent with Geddes et al. (2009)).

Geddes et al. (2009), designated O_x as the ozone plus nitrogen dioxide concentrations (the combined oxidants). The highest morning O_x concentrations in Toronto, and its surrounding areas from 2000 to 2007 were observed during the summertime when the wind was coming from between 135° and 270° . These levels were much greater compared to when the wind was calm, or from 270° to 45° . However, from 8 a.m. to 3 p.m. on stagnant days, a rise of 3 ppb per hour of O_x is seen. This indicates little to no transport of O_x precursor gases, allowing for O_x buildup under stagnant conditions. Stagnant conditions were defined when the daily net parcel displacement was less than 120 km, based on the wind direction and speed. Peak 8-hourly mean O_x was very similar on stagnant days and when the winds were from 135° to 270° . However, O_3 levels were decreased on stagnant days due to the greater role of titration. It is also important to note that the analysis stated in this paragraph was done in the summer.

Part of the study by Whaley et al. (2015) analyzed 15 pollutant episodes from 2004-2007, (as mentioned above), and seven regions were designated to analyze where the ozone in the low-level troposphere originated. It was found this ozone most depended on emissions originating from their defined northeast U.S. area for 9 of the 15 episodes. Westward stretching of the Bermuda high results in fresher, less polluted air (decreased ozone), carried over a wide avenue ranging from the southern Great Plains to the Midwest. (Zhu & Liang, 2013). This westward extent also results in carrying of pollutant laden air from the Midwest to the Northeast U.S., leading to higher O_3 concentrations in New England as well as coastal eastern states (ibid.). Whaley et al. (2015), determined that the primary cause of high ozone concentrations is likely high isoprene and NO_x fossil fuel emissions in their study. The responsiveness to emissions from fossil fuel-induced CO was less significant by about one order of magnitude (ibid.).

Regarding seasonal variations, in Toronto, it was determined by Whaley et al. (2015), that lower tropospheric ozone peaks in late spring/early summer and high pollutant concentrations were generally associated with high temperature and pressure conditions. Zhang et al. (2022), also showed peak ozone concentrations in the summertime, which will be discussed below. Though, Hilker et al. (2021) deduced that particle size was closely dependent on temperature and thus demon-

strated considerable seasonal variation. In Toronto, particles with size < 20 nm had significantly higher concentrations at temperatures below 10°C (ibid.). However, particles with size > 100 nm had higher concentrations when temperatures were greater than 20°C (ibid.). Hilker et al. (2021) concluded that particulate number density was the greatest during winter, as most particulates are small. However, particle concentrations with an approximate size of 50 nm showed no relationship with temperature (ibid.).

2.2.2 Pollutant Trends in the Vicinity of Toronto

In Rochester, New York, pollutants, including UFPs, were studied from 2002 to 2009 by (Wang et al., 2011). PM concentrations were categorized into two categories 10-50 nm and 100-500 nm. Generally, 10-50 nm particulate number densities peaked twice at 7-8 a.m. and 2 p.m. From 2002 to 2005, the 10-50 nm and 100-500 nm number particle sizes were observed to have the greatest and lowest concentrations, respectively. This contrast may indicate local transport effects. Furthermore, within this period, there was an apparent reduction in the number concentration peaks for the 10-50 nm particles, and starting in 2006, SO_2 nucleation began to decrease. There was also a strong relationship of $r = 0.89$ between 10-50 nm particle number concentrations and morning (7-9 a.m.) nucleation creation. Incidents of nucleation events were inversely related to temperature. Overall, the dominant SO_2 emission source was determined to be from the Eastman Kodak industry in 2009. It was ultimately concluded that these reductions in number densities and nucleations are probably due to diminishing emissions from Eastman Kodak, the Heavy-duty highway policy put forward by U.S. EPA 2007 on October 1, 2006, and the coal power plant closure in May 2008. It was also determined that from 2007 to 2009, these factors contributed to a significant SO_2 decline, particularly in the presence of winds from the north.

Throughout the smog periods (April-September) from 1996 to 2015 in Windsor, Ontario, and other surrounding areas, including Allen Park, Ontario, Erie, Ontario, Lansing, Michigan, Delaware, Ohio, and the National Trail School in Ohio, all locations measured peak yearly O_3 concentrations from June to July (Zhang et al., 2022). Zhang et al. (2022) found that suburban/urban measurements observed an increasing trend in O_3 (0.12 to 3.2 ppb annually), and rural measurements observed a decreasing trend in O_3 concentrations (-0.01 to 2.71 ppb annually, mainly in the 50th to 95th percentile values). The study by Zhang et al. (2022) showed increased O_3 concentrations during the weekends due to decreased NO_x emissions. Specifically, the suburban/urban sites (Windsor, Allen Park, Erie, and Lansing), observed a greater increase in O_3 concentrations by 3.3 ppb on weekends compared to 1.6 ppb for rural sites. Due to fewer NO emissions over the weekends, a considerable weekend-to-weekday difference in these concentrations is seen.

2.2.3 Other Worldwide Pollutant Trends

Geddes et al. (2016) observed long-term NO₂ concentrations in various cities worldwide from 1996-2012. It was found that high-income Asian Pacific regions (mainly within South Korea and Japan), East Asia, high-income North America, and west-Europe observed the highest population-weighted mean NO₂ (PWMNO₂) concentrations worldwide. However, high-income Asian Pacific regions with NO₂ concentrations declined by 2.1% yearly. Moreover, PMWNO₂ concentrations declined by 2.5% per year in western Europe and 4.7% per year in Canada and the United States. Although, in East Asia, a threefold increase in PMWNO₂ concentrations was noted, with an upward trend of 6.7% per year. The magnitude of the NO₂ concentration change over the 16 years shifted from western Europe/North America to east Asia. Global PWMNO₂ had a 0.9% annual growth.

Another study by de Jesus et al. (2020) aimed to quantify PM_{2.5} concentrations and particulate number density (PNC) long-term trends in various worldwide cities and to determine what other factors played a role, such as weather events or trends in temperature, wind speed, and precipitation. All data was averaged monthly from 1998 to 2017. The cities surveyed were Augsburg, Brisbane, London, Helsinki, and Rochester. Though Brisbane had an increased PNC and PM_{2.5} trend of 53 µg/m³/yr and 0.1 particles/m³/yr, every other city measured a decrease in PNC and PM_{2.5}, with respective trends ranging from 400 to 3800 particles/m³ and 0.03 to 0.6 µg/m³ annually (ibid.). The decline of PNCs in Rochester was $4.0 \times 10^{-2} \text{ cm}^{-3}$ annually. London was the only city with a non-monotonic decreasing trend. An autoregressive integrated moving average (ARIMA) analysis found that temperature, wind speed, and precipitation considerably impacted PM_{2.5} and PNC, with a broad inverse relationship between these variables. For example, a negative correlation is observed between PM_{2.5} as well as PNC with temperatures less than 0°C in Rochester. As the temperature increases, this correlation is observed to be less negative and then turns positive for temperatures around 15°C. Moreover, in the presence of strong winds, an increase in PM dilution was found, warm temperatures appeared to hinder the emergence of particles, and elevated precipitation rinses away some air-borne particulates. It was ultimately concluded that continuous concentration declines resulted from fleet management and traffic guidelines (de Jesus et al., 2020), similar to the reason for PM_{2.5} declines stated by Jeong et al. (2020). A sharp PNC concentration decrease resulted from sulfur management vehicular policies (de Jesus et al., 2020). A sharp increase in PM_{2.5} concentrations and PNC was observed after severe incidents such as wildfires and dust storms (ibid.).

2.3 Back-Trajectory Methods and Mapping Techniques

A simple technique to deduce which directions high pollutant sources originate from at a given location, is to determine the average (or x^{th} concentration percentile) based on wind direction. Generally, back-trajectories involve tracing pollutants upwind based on wind speeds and directions. Some techniques are outlined in this section.

The HYSPLIT (Hybrid Single-Particle Lagrangian Integrated-Trajectory) model can simulate simple and complex atmospheric transport, as well as chemical transportation, dispersion and deposition (NOAA Air Resources Laboratory., n.d. & Stein et al., 2015). Identifying where and what the sources of air-pollutants are is a frequent use of the HYSPLIT model (ibid.). HYSPLIT is a hybrid of both the Lagrangian and Eulerian method (ibid.). The Lagrangian method computes diffusion and advection of an air parcel from its starting point using a dynamic reference frame (ibid.). The Eulerian approach calculates pollutant concentrations within a non-moving 3-dimensional grid (ibid.). Particle or puff dispersion is presumed to compute a pollutants dispersion (NOAA Air Resources Laboratory., n.d.). Particles are advected in 3-d motion by the average wind field and dispersed through turbulence in a particle model (ibid.). The PUFF model consists of puffs growing in size until its size is greater than the defined grid (ibid.). After this occurs, the puff splits into several smaller puffs, whose total mass remains unchanged (ibid.). HYSPLIT is also based off reanalysis data. Other back-trajectory methods are described below.

Based on Berriban et al. (2022), Hsu et al. (2003), Li. et al. (2020), Wang et al. (2006) and Zhou et al. (2024), a Concentrated Weighted Trajectory (CWT) is represented as;

$$\text{CWT}(x,y) = \frac{\int C(t)T(x,y,t)dt}{\int T(x,y,t)dt} = \frac{\sum_{l=1}^N C_l T_{i,j,l}}{\sum_{l=1}^N T_{i,j,l}} \quad (2.1)$$

where the right-hand term is the discrete solution, where C_l is the l^{th} concentration in the time series, and $T_{i,j,l}$ represents the l^{th} concentration time spent in the pixel (i, j) .

Again, based on Zhou et al. (2024), a Concentrated Field Analysis (CFA) can be expressed as:

$$\text{CFA}(x,y) = \frac{\int \log[C(t)]T(x,y,t)dt}{\int T(x,y,t)dt} = \frac{\sum_{l=1}^N \log(C_l)T_{i,j,l}}{\sum_{l=1}^N T_{i,j,l}} \quad (2.2)$$

where again, $T_{i,j,l}$ represents the time the l^{th} concentration resides in a pixel (i, j) , and C_l denotes the l^{th} concentration. The CFA is advantageous when strong differentials in pollutant concentrations are present (ibid.).

Based on Berriban et al. (2022), Hsu et al. (2003), Li et al. (2020), Wang et al. (2006), and Zhou et al. (2024), a Probable Source Contribution Function (PSCF), can be denoted as;

$$\text{PSCF}(x,y) = \frac{\int T(x,y,t)dt|c_l > C}{\int T(x,y,t)dt} = \frac{\sum_{l=1}^N T_{i,j,l}|c_l > C}{\sum_{l=1}^N T_{i,j,l}} \quad (2.3)$$

where, the numerator represents the total time the pollutant resides in each grid location (i, j) , when c_l is greater than a threshold concentration C . Usually, C is 75 to 90 percent of the greatest concentration value (Zhou et al., 2024). When more than one pollutant measurement location is used for the analysis, based on Zhou et al. (2017), and Zhou et al. (2024), the PSCF becomes modified to;

$$\text{PSCF}(x,y) = \frac{\int \int T(x,y,s,t)dt ds|c_{s,t} > C_s^*}{\int \int T(x,y,s,t)dt ds} = \frac{\sum_{s=1}^S \sum_{l=1}^N T_{i,j,s,l}|c_{s,l} > C_s^*}{\sum_{s=1}^S \sum_{l=1}^N T_{i,j,s,l}} \quad (2.4)$$

where C_s^* is the threshold pollutant concentration for the particular site, $c_{s,l}$ is the l^{th} concentration at site s , $T_{i,j,s,l}$ is the total time the l^{th} back-trajectory spends at pixel (i, j) from the site, s , and S is the amount of sites (ibid.). Compared to the CWT, a PSCF is inferior at differentiating moderate sources from high sources (Hsu et al., 2003).

According to GOV.UK. (2024), Sofowote et al. (2014), Squizzato and Masiol. (2015), and Zhou et al. (2024), a Conditional Probability Function (CPF) is given by;

$$\text{CPF}_\theta = \frac{P_\theta}{F_\theta} \quad (2.5)$$

In this instance, the number of concentrations greater than or equal to a concentration threshold in a particular wind direction is represented by P_θ (ibid). F_θ denotes the total amount of concentration values from that wind direction (ibid.). This threshold is 75 to 90 percent of the greatest concentration value in most cases (Zhou et al., 2024). While the CWT map produces the spatial variation of averaged concentrations over a given area, the CPF provides probabilistic information about which wind directions relatively high concentrations are originating from (GOV. UK., 2024).

A Conditional Bivariate Probability (CBPF) function can be represented as

$$CBPF_{\theta,u} = \frac{m_{\theta,u}|c \geq C^*}{n_{\theta,u}} \quad (2.6)$$

where, again, $m_{\theta,u}$ and $n_{\theta,u}$ are the concentration frequencies, where the concentration (c) is greater than the threshold concentration C^* , and total number of counts respectively per wind speed and direction (GOV.UK., 2024, Nguyen et al., 2022; Zhou et al., 2019a; Zhou et al., 2019b). Zhou et al. (2024), defines a CBPF the same as equation 2.6, except with $c > C^*$. As stated by Zhou et al. (2024), this threshold is commonly fixed at 75 to 90 percent of the greatest concentration, and is applied in a same manner as for the CPF.

Like HYSPLIT these back-trajectory methods are useful in terms of identifying pollutant hotspots, and their sources. HYSPLIT is generally used for long-range transport (i.e., to pinpoint pollutant sources over a larger spatial scale). This thesis seeks to determine spatial variations of pollutants over a local-scale, rather than pinpoint exact sources. Potential sources (vehicular, aircraft, industrial etc...), can be inferred from hotspots, or regions with elevated concentrations, though, this is a secondary consideration that follows from accomplishing the primary goal.

Since an objective of this thesis is to determine the pollutant spatial variations within Toronto from ground level air-quality stations, a CWT and a combined CPF and CBPF methodology will be employed by tracing the pollutant concentration linearly based on the hourly measured wind speed and direction. These back-trajectory techniques replace the more complicated HYSPLIT back-trajectories with more local simpler ones, that backtrack the pollutants linearly based on the directly measured ground-level wind speed and directions. More details regarding the back-trajectory methodologies employed are discussed in Chapter 4. As will also be discussed in more detail in Chapter 4, multiple air-quality stations are employed, which is useful in “triangulating” the high concentrations from individual station back-trajectories, thus producing a complete map, which captures the pollutant variation within the city.

2.3.1 Some Applications of Back-Trajectory Methods

Jeong et al. (2020), employed the HYbrid Single Particle Lagrangian Integrated Trajectory (HYSPLIT) model to calculate PSCFs for 72-hours back in time for each NAPS measurement beginning at 12 p.m. 40 km resolution meteorological data was embedded in the HYSPLIT model. C , in their case was the 75th percentile concentration. As mentioned previously, through this analysis, Jeong et al. (2020), found regional PM_{2.5} primarily originated out of a broad band of industrial lo-

cations throughout Pennsylvania to Illinois before reaching Toronto. Similarly, Brook et al. (2011) used the hemispheric trajectory model (HTM), by the Meteorological Service of Canada, to trace the origins of PM_{2.5} from one site in Toronto and Egbert, Ontario. 24 hour-averaged data was collected over two separate two week periods in 1998 (March-April, and July). Three day back-trajectories were computed four times daily (0, 6, 12, and 18Z), and it was ultimately found that PM_{2.5} levels were 2 to 4 times greater when winds were from the south/southwest than when they were from the North.

Using the TRAspa package to run the HYSPLIT program, Zhou et al. (2019a), found that significant secondary nitrate sources coincided with Upper Ohio Valley electricity generation plants, from employing a PSCF, during the 2012 to 2014 winters, like the result from Jeong et al. (2019). Although, in this case, C was set to the 90th percentile values (Zhou et al., 2019a). By the same methodology, it was found that wintertime air containing secondary aerosol with high concentrations of ozone primarily came from Sudbury, Ontario (from metal processing industries), and west Ontario (ibid.). Western Ontario was the primary origin of this secondary aerosol during summer as well (ibid.). During spring and fall, the primary sources were from urban Michigan locations (ibid.). Unlike winter, other seasons also exhibited secondary sulfate as a contribution to the particulates measured (ibid.). Again, during summer, the same PSCF analysis showed that this pollutant primarily originated from urban midwest/western NY locations and Michigan (ibid.). Throughout spring/fall, this contribution mainly originated from east NY, and Upper Ohio Valley (ibid.), consistent with the sources above. According to Zhou et al. (2019a), these locations coincide with coal power plants.

During the spring of 2001 to spring of 2003, Wang et al. (2006) employed a PSCF and CWT using HYSPLIT4 model to back-track PM₁₀ with the aim of quantifying dust storms effect on PM concentrations. Three major regions were defined as primary emitters of dust, named “Loess Plateau Source”, “Northwesternly Sources” and “Northernly Sources.” (ibid.). The CWT showed elevated values in both the “Northwesternly Sources” and “Northernly Sources”, unlike the PSCF, leading the authors to conclude that the CWT was superior at identifying origins of dust in this instance (ibid.). Similarly, Hsu et al. (2003) found that both a PSCF and a CWT were able to identify and separate both moderate and high sources of PCBs (polychlorinated biphenyl) sources in Chicago (ibid.). However a couple of sources (area between Kankakee, and Joliet IL, and Gary, IN), the PSCF marked as a “high source”, where they were marked as a moderate source by the CWT (ibid.). This reinforces what was already stated by Hsu. et al. (2003), that the PSCF can fail to differentiate high from moderate sources. It was acknowledged by Hsu et al. (2003), that perhaps setting a higher threshold concentration for the PSCF, could have been more effective at

distinguishing high from low to moderate sources. Li et al. (2020) generated PSCFs and CWTs via HYSPLIT model in the area of Weifang, China. While the results from the PSCF and CWT were mostly the same, the potential sources for the CWT covered a larger area than for PSCF, during the spring, summer and fall (ibid.). Li et al. (2020) suggested that this was because the PSCF is limited to a subset of high concentrations (ibid.).

2.4 Column Pollutants and Boundary Layer Height (BLH)

In this thesis, column TROPOMI satellite measurements are converted into surface level concentrations by dividing by the BLH (as mentioned in Chapter 6). Ground-level temperature and pressure are also considered. More details to be described in Chapter 1. This conversion also assumes all the column pollutants are located below the boundary layer height, however this is not the case, as outlined in section 2.4.2.

2.4.1 Boundary Layer Height

The troposphere's bottommost part is known as the planetary boundary layer (PBL) (Haby, n.d.). In this layer, friction affects the wind (ibid.). The planetary boundary layers' height or "thickness" fluctuates depending on the time of day and season (ibid.). Throughout the warmest time of year or day, the PBL's thickness tends to be the greatest, whilst the opposite is generally true during the coolest time of year or night (ibid.). Since warmer air is less dense than cooler air, and the PBL height reduces at night due to decreased thermal surface-generated rising updrafts, the diurnal and seasonal differences manifest (ibid.). More convective mixing (PBL expansion) occurs with faster winds as well (ibid.). The boundary layer's top, or boundary layer height is usually characterized by a hydrolapse, temperature inversion, change in wind speed, wind direction, and air mass (ibid.). Measuring these indicators is the simplest way to deduce the BLH (ibid.).

Since the surface of Earth receives and releases most of Earth's energy, the greatest diurnal temperature changes occur within the boundary layer (Haby, n.d.). In other atmospheric layers, the temperature remains relatively constant (ibid.). Due to the energy budget within the PBL, temperature and moisture advections make it a crucial contributor to the growth of showers and thunderstorms (ibid.). Impacts on energy, formation of clouds, and moisture transport within the PBL also influence Earth's climate patterns and sensitivity (Kalmus et al., 2022). Topographic and vegetational friction result in turbulent and gusty winds within the PBL (Haby, n.d.). Winds above the PBL tend to be considerably more uniform due to less friction (ibid.). Turbulent mixing of moisture, heat, momentum, and tracers (i.e., pollutants) also occurs within PBL (Kalmus et al., 2022).

Rapid urban development and high energy consumption have resulted in elevated levels of air pollution in urban China (Wang & Hao, 2012). Studies have analyzed the relationships between pollutants and the BLH in China (Li et al., 2021; Su et al., 2018; Sun et al., 2023). These studies generally found a negative relationship between pollutants and the planetary boundary layer height (ibid.). Su et al. (2018) calculated the PBLH using ground-based and space-born lidar, and from December 2012 to June 2017, gathered PM_{2.5} measurements across 1600 stations throughout China. A negative relationship between PM and PBLH was generally found. However, these relationships varied considerably by season, region, and specific meteorological conditions (ibid.). In clean regions (such as the Pearl River Delta Region) weak or uneven relationships were found between PM and the planetary boundary layer height (PBLH) (ibid.). Although, in more polluted areas (such as the North China Plain), a strong interaction between PBLH and PM was denoted when the PBLH was low and PM was high respectively, mainly during the winter months (ibid.). Weaker relationships were generally observed in higher elevated areas due to mountain breezed induced wind dispersion and from the presence of less pollution at greater elevations in the first place (ibid.). Stronger relationships in lower altitude regions may have been a result of increased stagnation in these areas (ibid.). Throughout China, there has been a negative relationship between PBLH and wind speed (ibid.).

Similarly, Li et al. (2021) found that in the North China Plain from November 26 to December 26, 2019, intense haze was associated with an average convective boundary layer (CBL) of less than 560 meters. This is expected, as stated by Li et al. (2021), since a lower atmospheric boundary layer height leads to a greater accumulation of pollutants near the surface, and stable air promotes the descension of pollutants. The former is in accord with the potential reason for the stronger negative relationships between PBLH and PM, as stated by Su et al. (2018). Like the results by Li et al. (2021), Sun et al. (2023) also found that in Xi'an, China from 2017-2021, there was a 30% and less than 7% probability of "polluted weather" when the BLH is < 200 meters and > 2000 meters respectively. Similar to Sun et al. (2023) and Li et al. (2021), in Almaty, Kazakstan, Tursumbayeva et al. (2022) found that PM_{2.5} levels $\geq$ the 80th percentile generally corresponded to relatively calm winds and low PBLH, during the cooler six months. More specifically, throughout the summertime, PM_{2.5} concentrations of less than 9.9 $\mu\text{g}/\text{m}^3$ coincided with a PBLH of > 1970 meters (ibid.). Conversely, PM_{2.5} levels of greater than 94 $\mu\text{g}/\text{m}^3$ throughout the winter coincided with a PBLH of less than 393 meters (ibid.). The analysis by Tursumbayeva et al. (2022) was carried out from Mar. 1, 2020 to Feb. 28, 2021. In the Tibetan Plateau, Chen et al. (2022) found that the BLH relates to Aerosol Optical Depth (AOD) by -0.4. Dust was also greater and lower throughout the months with low and high BLH (ibid.). Chen et al. used a combination of

MERRA-2 and ERA5 reanalysis datasets to obtain the boundary layer height from 1980 to 2019 and MERRA-2 to obtain AOD from 1980 to 2018, with a three-month temporal resolution (ibid.). The dust concentrations were column dust taken by the same model with the same temporal resolution, with a top-level pressure of 0.01 hpa, and containing 72 different layers (ibid.).

2.4.2 Satellite Measurements & Pollutants in Different Atmospheric Layers

The study mentioned above by Geddes et al. (2016) used the Scanning Imaging Absorption Spectrometer for Atmospheric Cartography (SCIAMACHY), Global Ozone Monitoring Experiment (GOME), and GOME-2 to obtain column NO_2 concentrations. The spatial resolution of SCIAMACHY, GOME, GOME-2 was $60 \times 30\text{km}$, $320 \times 40\text{km}$, and $80 \times 40\text{km}$ respectively, though these data were resampled onto a $0.1^\circ \times 0.1^\circ$ resolution (about $10 \times 10\text{km}$). Lamsal et al. (2008, 2010), employed the GEOS-Chem model (developed by Bey et. al., (2001)), to deduce the spatial and temporal variation of the contribution of NO_2 within the boundary layer to tropospheric column NO_2 , and thus predict surface level NO_2 concentration densities from NO_2 columns. Geddes et al. (2016), deduced surface-level concentrations from their column NO_2 measurements using this procedure.

It is known that the dominant amount of tropospheric NO_2 is contained within the boundary layer (Bucsela et al., 2008). This result is also demonstrated by Martin et al. (2001). Also using the GOME satellite to retrieve NO_2 columns, Martin et al. (2001) found that in locations with strong surface-level emissions (northern Europe, northeast United States, and East Asia), column densities were generally higher. These were found to be locations where most column NO_2 was below the boundary layer, and partially blocked by cloud cover (ibid.). It was also found that when NO_2 column concentrations were greater than 2×10^{15} molecules/ cm^2 , the column concentrations were largely determined by the boundary layer height (ibid.). Similarly, as part of the Korean United-States Air Quality campaign (KORUS-AQ), Yang et al. (2023), found that in the Seoul Metropolitan area, 95% of column concentrations were located below the boundary layer. The bulk of atmospheric SO_2 is also located below the boundary layer, within the first 1-2 kilometers of the atmosphere (Hong et al., 2020). The creation, diffusion, and evolution of SO_2 is also nearly related to the BLH (ibid.). Although, 90% of column ozone is stratospheric (Rowland, 1991), implying that using satellite measurements to quantify ground level ozone is unreliable.

3 Pollutant Trends and Polar Plots from Air Quality Stations

3.1 Methodology

The analysis in this chapter will utilize four air-quality stations throughout Toronto, named “Toronto East”, “Toronto Downtown”, “Toronto West” and “Toronto North”, to compute monthly, seasonal, and annual trends, and their corresponding standard error for each pollutant parameter ($\text{PM}_{2.5}$, O_3 , CO , NO , NO_2 and NO_x). An additional long-term measurement of SO_2 , and CO were taken by the “Toronto West” station, and as such these trends were also computed. These four stations and their pollutant measurements were selected for two main reasons: (1) they are located in distinct areas of Toronto, allowing for potential examination of pollutant trends in different areas of Toronto; and (2), the pollutants measurements selected from these stations were recorded over an extended time-period (see section 3.2, for specific details regarding these timeframes). This analysis seeks to provide a holistic understanding of the long-term temporal variations of pollutants in Toronto, addressing a gap in the previous literature discussed in Chapters 1 and 2.

To compute trends, pollutant measurements were divided into 12 monthly and 4 seasonal categories. The seasonal categories were: winter (December, January, February), spring (March, April, May), summer (June, July, August), and autumn (September, October, and November). Annual trends were also computed as well. Percentage changes for comparisons to literature in the Discussion subsection of this chapter were computed from the first and last values of the lines of best fits, then dividing by the number of years used. Monthly averaged values were also calculated to observe seasonal differences.

To deduce which directions the individual pollutants came from, average and 90th percentile concentration values were plotted against wind direction (divided into 10° increments) on polar plots. This 10° increment was chosen since the automated weather station wind directions were provided in 10s of degrees. This was done for the Toronto West, East, North and Downtown stations throughout Toronto.

Hourly historical pollutant data from four air-quality (AQ) stations, named “Toronto East”, “Toronto Downtown”, “Toronto West”, and “Toronto North”, throughout January 1, 2007 01:00 LST, to January 1, 2023 00:00 LST, for ozone, $\text{PM}_{2.5}$, NO_2 and an additional CO measurement from Toronto West, were downloaded from airqualityontario.com (Air Quality Ontario, n.d.-f). Other air-quality station pollutant location information and data were extracted from the National Air Pollu-

tant Surveillance (NAPS) Program (<https://data-donnees.az.ec.gc.ca/data/air/monitor/national-air-pollution-surveillance-naps-program/Data-Donnees?lang=en>), as part of Environment and Climate Change Canada’s Data Catalogue (Environment and Climate Change Canada Data Catalogue, n.d.). This also includes times prior to January 1, 2007 01:00 LST, for the Toronto East, West, Downtown and North station pollutants mentioned above. Several pollutant parameter concentrations (PM_{10} , $PM_{2.5}$, O_3 , CO , NO , NO_2 , NO_x) were extracted from each station. Note that not all stations contained every pollutant measurement (see Table 3.1). Historical wind data from automated weather stations can be found at the Government of Canada’s department of Environment and Climate Change Canada’s past weather data page (Government of Canada, n.d.).

When generating back-trajectories in 4, all 22 ECCC and NAPS ground-level air quality stations in Toronto, owned by the Ministry of the Environment, Conservation and Parks (MOECP), were employed. Figure 3.1, shows the locations of all these stations, and where their corresponding wind measurements were taken from.

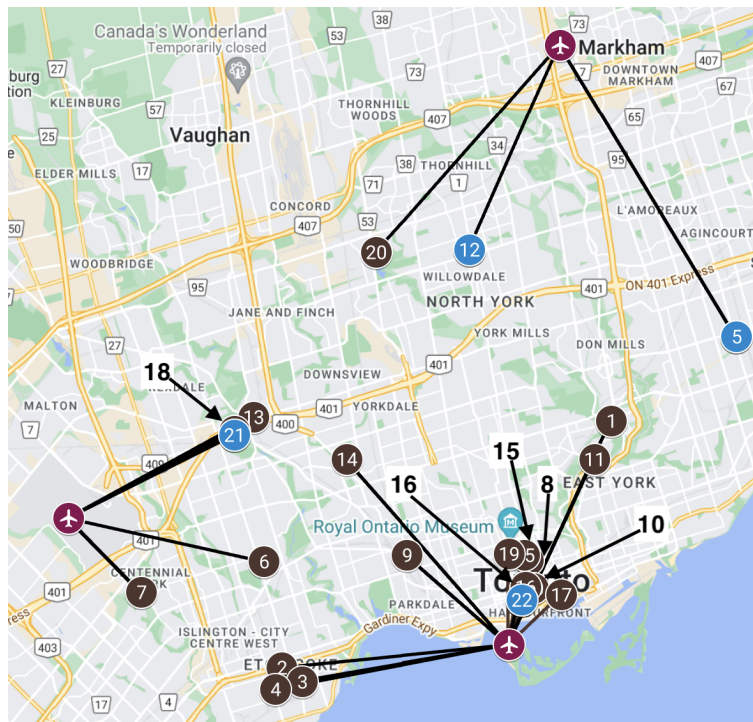


Figure 3.1: Locations of all the air-quality stations (Air Quality Ontario, n.d.-f, ECCCDC, n.d.). The black lines show which airport automatic weather stations the corresponding wind speed and directions to the 22 air-quality stations was taken from. Highlighted in blue are the four air-quality stations, “Toronto West (#21)”, “Toronto North (#12)”, “Toronto Downtown” (#22), and “Toronto East (#5)”.

To relate air pollutant measurements with wind measurements for the polar plots (and determine back-trajectories from each station in Chapter 4), air quality stations were allocated to airport

meteorology stations based on proximity. All measurements used throughout this report are in local standard time (not adjusted for daylight savings). Table 3.1, and Figure 3.1 displays the location number, name, co-ordinates and which airport meteorological stations their corresponding wind measurements were gathered from. Sample air quality stations (Toronto North and Toronto East) are displayed in Figure 3.2. Note that this chapters focus will be on pollutant trends.

Table 3.1: Number (corresponding to the numbers in the Figure 3.1) name, location, pollutants measured (Air Quality Ontario, n.d., ECCDC, n.d.), and their corresponding wind measurements for polar plots and back-trajectories. Timeframes for each pollutant measured by station is given in section A.3 of the Appendix.

No.	Station Name	Location	Pollutants Measured	Wind Measurements
1	Humber Valley	43.66028°, -79.528306°	O ₃	Toronto City Center Airport
2	Ontario Science Center	43.715°, -79.3411°	O ₃	Toronto Pearson International Airport
3	Etobicoke	43.61917°, -79.51833°	PM _{2.5} , PM ₁₀ , CO, SO ₂ , NO, NO ₂ , NO _x , O ₃	Toronto City Center Airport
4	Etobicoke 2	43.613587°, -79.50735°	PM _{2.5} , PM ₁₀ , CO, SO ₂ , NO, NO ₂ , NO _x , O ₃	Toronto City Center Airport
5	Etobicoke 3	43.61076°, -79.5218°	NO, NO ₂ , NO _x , O ₃	Toronto City Center Airport
6	Toronto East	43.74792°, -79.27406°	PM _{2.5} , PM ₁₀ , CO, SO ₂ , NO, NO ₂ , NO _x , O ₃	Toronto Buttonville Airport
7	Centennial Park	43.6485317°, -79.5939638°	PM _{2.5} , PM ₁₀ , SO ₂ , NO, NO ₂ , NO _x , O ₃	Toronto Pearson International Airport
8	Garden District	43.66083°, -79.38583°	O ₃	Toronto City Center Airport
9	Junction Triangle	43.662933°, -79.4511538°	O ₃	Toronto City Center Airport
10	The PATH	43.65°, -79.3833°	O ₃	Toronto City Center Airport
11	Don River	43.7°, -79.35°	O ₃	Toronto City Center Airport
12	Toronto North	43.78158°, -79.41773°	PM _{2.5} , CO, NO, NO ₂ , NO _x , O ₃	Toronto Buttonville Airport
13	Pelmo Park-Humberlea	43.71667°, -79.5333°	O ₃	Toronto Pearson International Airport
14	Black Creek	43.7°, -79.48333°	NO, NO ₂ , NO _x , O ₃	Toronto City Center Airport
15	Bay-Cloverhill	43.662972°, -79.388111°	PM _{2.5} , CO, SO ₂ , NO, NO ₂ , NO _x , O ₃	Toronto City Center Airport
16	Osgoode	43.65028°, -79.38694°	SO ₂ , NO, NO ₂ , NO _x , O ₃	Toronto City Center Airport
17	St. Lawrence	43.6475°, -79.36778°	O ₃	Toronto City Center Airport
18	Highway 401	43.71111°, -79.54342°	PM _{2.5} , CO, SO ₂ , NO, NO ₂ , NO _x , O ₃	Toronto Pearson International Airport
19	University of Toronto	43.65902°, -79.3953°	PM _{2.5} , NO, NO ₂ , NO _x , O ₃	Toronto City Center Airport
20	Environment Canada	43.78047°, -79.467372°	PM _{2.5} , CO, SO ₂ , NO, NO ₂ , NO _x , O ₃	Toronto Buttonville Airport
21	Toronto West	43.709444°, -79.5435°	PM _{2.5} , PM ₁₀ , CO, SO ₂ , NO, NO ₂ , NO _x , O ₃	Toronto Pearson International Airport
22	Toronto Downtown	43.645451°, -79.3891°	CO, SO ₂ , NO, NO ₂ , NO _x , O ₃	Toronto City Center Airport



Figure 3.2: Example Toronto North (left), and Toronto Downtown (right) ground-level AQ station. These stations are owned by the Ontario Ministry of the Environment and Parks. Imagery taken from <https://www.airqualityontario.com> (Air Quality Ontario, n.d.-g).

3.2 Results

Table 3.2 displays monthly $PM_{2.5}$, NO_2 , O_3 for the Toronto Downtown, West, East and North stations, as well as CO and SO_2 for Toronto West. Monthly $PM_{2.5}$, O_3 , NO, NO_2 , NO_x , CO and SO_2 trends from the four Toronto West, Downtown, East and North stations are present in Table 3.2. Tables 3.3 through 3.7, displays the same as Table 3.2, though regarding seasonal trends. The pollutants for these stations were selected due to the consistency and length of their measurement timeframes.

Sample plots for two particular datasets are displayed in Figure 3.3. Figure 3.4 displays average monthly concentrations for a few air-quality stations and species ($PM_{2.5}$, O_3 , NO_2 , CO, and SO_2). The same is displayed in Figure 3.5, though by hour of day. Given that nearly all the air quality station pollutants have varying timeframes (as seen in Tables A.1 and A.2 (of the Appendix section), it is best to show monthly and hourly averages for select stations, for a consistent time-period, in various corners of the city, like for the trends. As seen with the $PM_{2.5}$, NO_2 , and O_3 monthly and hourly averages, this gives some insight regarding the spatial variations (with higher $PM_{2.5}$, NO_2 and lower O_3 in the west relative to other locations). A more detailed back-trajectory analysis for all the stations will be displayed in Chapter 4.

Figures 3.7, 3.8, 3.9 presents examples of Toronto North, Downtown, East, and West average and 90th percentile $PM_{2.5}$, O_3 and NO_2 , as well as Toronto West CO and SO_2 distributions versus wind direction on a polar chart. Note that “90 %” represents the 90th percentile. For comparison to the monthly averages, the same species and locations are chosen, except the dates begin at their

starting point as defined in Appendix A instead of 2003. Two examples of averaged concentration vs. wind direction in cartesian co-ordinates are also displayed in Figure 3.10. Joint probability functions of the Toronto West NO₂ and O₃ concentrations and wind direction are displayed in Figure 3.11.

Table 3.2: Sample monthly pollutant concentration trends (top) for NO₂ (ppb/yr), O₃ (ppb/yr), PM_{2.5} (µg/m³/yr), CO (ppm/decade), and SO₂ (ppb/yr). The years in the brackets represent the time range of the trends. In most cases, the starting point is 01:00:00 LST on the first day of the year, and the ending point 00:00:00 LST on the first day of that year.

	Jan	Feb	Mar	Apr	May	Jun	Jul	Aug	Sep	Oct	Nov	Dec
TD_NO ₂ (2003-2023)	-0.53	-0.75	-0.70	-0.65	-0.51	-0.39	-0.37	-0.37	-0.34	-0.45	-0.38	-0.48
TW_NO ₂ (2000-2023)**	-0.66	-0.94	-0.98	-0.91	-0.76	-0.71	-0.71	-0.65	-0.60	-0.73	-0.62	-0.72
TE_NO ₂ (1996-2023)	-0.58	-0.70	-0.69	-0.73	-0.69	-0.71	-0.65	-0.63	-0.62	-0.61	-0.54	-0.59
TN_NO ₂ (1996-2023)	-0.47	-0.57	-0.51	-0.65	-0.74	-0.78	-0.61	-0.65	-0.65	-0.57	-0.37	-0.43
TD_PM _{2.5} (2003-2023)	0.28	0.17	0.08	0.09	0.04	-0.23	-0.18	-0.15	-0.08	0.02	0.20	0.26
TW_PM _{2.5} (2000-2023)	0.11	0.00	-0.07	-0.04	-0.08	-0.31	-0.29	-0.29	-0.22	-0.18	0.02	0.11
TE_PM _{2.5} (2002-2023)	0.22	0.12	0.05	0.03	-0.04	-0.29	-0.35	-0.28	-0.23	-0.08	0.08	0.16
TN_PM _{2.5} (1998-2023)	0.11	0.08	0.06	-0.02	-0.11	-0.32	-0.28	-0.28	-0.26	-0.09	0.07	0.12
TW_CO (2000-2023)**	-0.10	-0.10	-0.13	-0.12	-0.09	-0.07	-0.05	-0.04	-0.08	-0.17	-0.16	-0.21
TW_SO ₂ (2000-2023)*	-0.17	-0.19	-0.16	-0.11	-0.11	-0.09	-0.09	-0.09	-0.16	-0.15	-0.15	-0.19
TD_O ₃ (2003-2023)	0.28	0.41	0.15	-0.01	0.02	-0.18	-0.31	-0.13	-0.09	0.22	0.32	0.22
TW_O ₃ (2000-2023)*	0.36	0.48	0.37	0.24	0.19	0.03	-0.04	0.11	0.07	0.24	0.32	0.22
TE_O ₃ (1974-2023)	0.33	0.37	0.33	0.33	0.23	0.14	0.12	0.12	0.21	0.26	0.28	0.30
TN_O ₃ (1988-2023)	0.34	0.35	0.28	0.33	0.27	0.11	0.08	0.12	0.22	0.25	0.31	0.32
TD_PM _{2.5} (2003-2023)	0.28	0.17	0.08	0.09	0.04	-0.23	-0.18	-0.15	-0.08	0.02	0.20	0.26
TW_PM _{2.5} (2000-2023)	0.11	0.00	-0.07	-0.04	-0.08	-0.31	-0.29	-0.29	-0.22	-0.18	0.02	0.11
TE_PM _{2.5} (2002-2023)	0.22	0.12	0.05	0.03	-0.04	-0.29	-0.35	-0.28	-0.23	-0.08	0.08	0.16
TN_PM _{2.5} (1998-2023)	0.11	0.08	0.06	-0.02	-0.11	-0.32	-0.28	-0.28	-0.26	-0.09	0.07	0.12
TW_CO (2000-2023)**	-0.10	-0.10	-0.13	-0.12	-0.09	-0.07	-0.05	-0.04	-0.08	-0.17	-0.16	-0.21
TW_SO ₂ (2000-2023)*	-0.17	-0.19	-0.16	-0.11	-0.11	-0.09	-0.09	-0.09	-0.16	-0.15	-0.15	-0.19

*In addition to no data being available in 2001 and 2002, no data was available until September 1, 2000 01:00 LST as well. As such, trends and standard error began at 2003 for all other seasons except fall. **In addition to the condition in (*), NO, NO₂ and NO_x data did not resume until 3/5/2023 16:00 LST, and the CO data did not resume until 3/4/2023 15:00 LST, thus these January and February trends and standard errors began at 2004.

Table 3.3: Sample seasonal and overall trends for the Toronto West, North, Downtown and East PM_{2.5} stations. DJF, MAM, JJA, SON denote December-January-February (winter), March-April-May (spring), June-July-August (summer), September-October-November (fall) respectively. Units for PM_{2.5} trends are (µg/m³/yr).

	DJF	MAM	JJA	SON	Overall
TW_PM _{2.5} (2000-2023)	0.07	−0.06	−0.29	−0.13	−0.11
TD_PM _{2.5} (2003-2023)	0.24	0.07	−0.19	0.04	0.04
TE_PM _{2.5} (2002-2023)	0.17	0.01	−0.31	−0.08	−0.05
TN_PM _{2.5} (1998-2023)	0.10	−0.02	−0.30	−0.10	−0.08

Table 3.4: As in previous table though for ozone, and an additional CO measurement from the Toronto West station. Units for O₃ and CO are ppb/yr and ppm/decade respectively.

	DJF	MAM	JJA	SON	Overall
TD_O ₃ (2003-2023)	0.29	0.06	−0.22	0.13	0.05
TW_O ₃ (2000-2023)*	0.35	0.27	0.02	0.20	0.23
TE_O ₃ (1974-2023)	0.33	0.30	0.12	0.24	0.25
TN_O ₃ (1988-2023)	0.34	0.30	0.10	0.25	0.24
TW_CO (2000-2023)*	−0.15	−0.12	−0.05	−0.13	−0.11
TW_SO ₂ (2000-2023)*	−0.18	−0.13	−0.09	−0.15	−0.14

*In addition to no data being available in 2001 and 2002, data did not being until September 1, 2000 01:00 LST, as such, trends and standard error were taken from (2003-2023) for all other seasons except fall.

Table 3.5: As in previous table but for NO (units ppb/yr).

	DJF	MAM	JJA	SON	Overall
TD_NO (2003-2022)	−0.39	−0.23	−0.08	−0.26	−0.24
TW_NO (2000-2023)*	−1.44	−1.14	−0.81	−1.60	−1.28
TE_NO (1996-2023)	−1.15	−0.75	−0.57	−1.12	−0.90
TN_NO (1996-2017)	−1.10	−0.64	−0.47	−0.87	−0.77

*In addition to no data being available in 2001 and 2002, data did not being until September 1, 2000 01:00 LST, as such, trends and standard error were taken from (2003-2023) for all other seasons except fall.

Table 3.6: As in previous table though for NO₂ (units ppb/yr).

	DJF	MAM	JJA	SON	Overall
TD_NO ₂ (2003-2023)	−0.59	−0.63	−0.38	−0.38	−0.50
TW_NO ₂ (2000-2023)*	−0.75	−0.89	−0.69	−0.64	−0.74
TE_NO ₂ (1996-2023)	−0.62	−0.71	−0.67	−0.59	−0.65
TN_NO ₂ (1996-2023)	−0.49	−0.63	−0.68	−0.52	−0.58

*In addition to no data being available in 2001 and 2002, data did not being until September 1, 2000 01:00 LST, as such, trends and standard error were taken from (2003-2023) for all other seasons except fall.

Table 3.7: As in previous table though for NO_x (units ppb/yr).

	DJF	MAM	JJA	SON	Overall
TD_ NO_x (2003-2023)	−0.92	−0.78	−0.43	−0.54	−0.67
TW_ NO_x (2000-2023)*	−2.18	−2.02	−1.51	−2.23	−2.02
TE_ NO_x (1996-2023)	−1.76	−1.44	−1.22	−1.68	−1.53
TN_ NO_x (1996-2017)	−1.60	−1.25	−1.21	−1.41	−1.37

*In addition to no data being available in 2001 and 2002, data did not begin until September 1, 2000 01:00 LST, as such, trends and standard error were taken from (2003-2023) for all other seasons except fall.

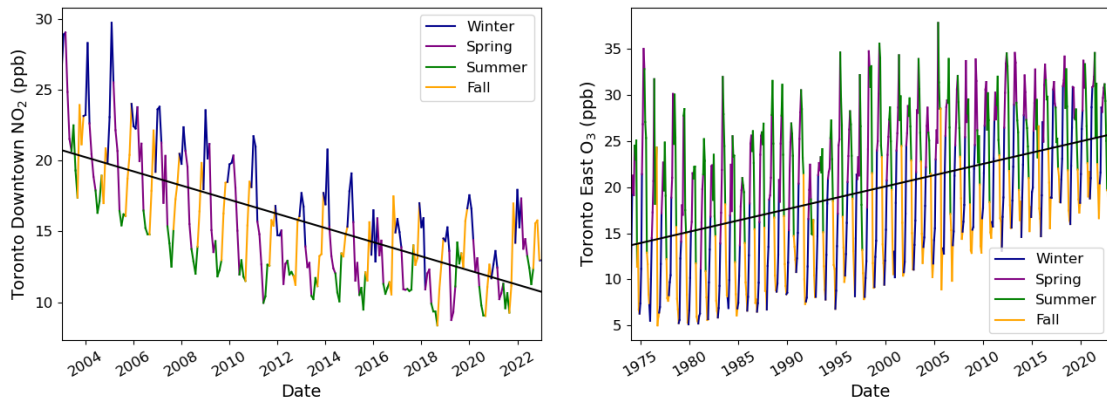


Figure 3.3: Toronto Downtown NO_2 (left) and Toronto East O_3 (right) plots with their trends overlayed from January 1, 2003, and 1974 01:00 respectively to January 1, 2023 01:00 LST. Concentrations were measured hourly, but were averaged monthly here. The trend-line is overlayed in black.

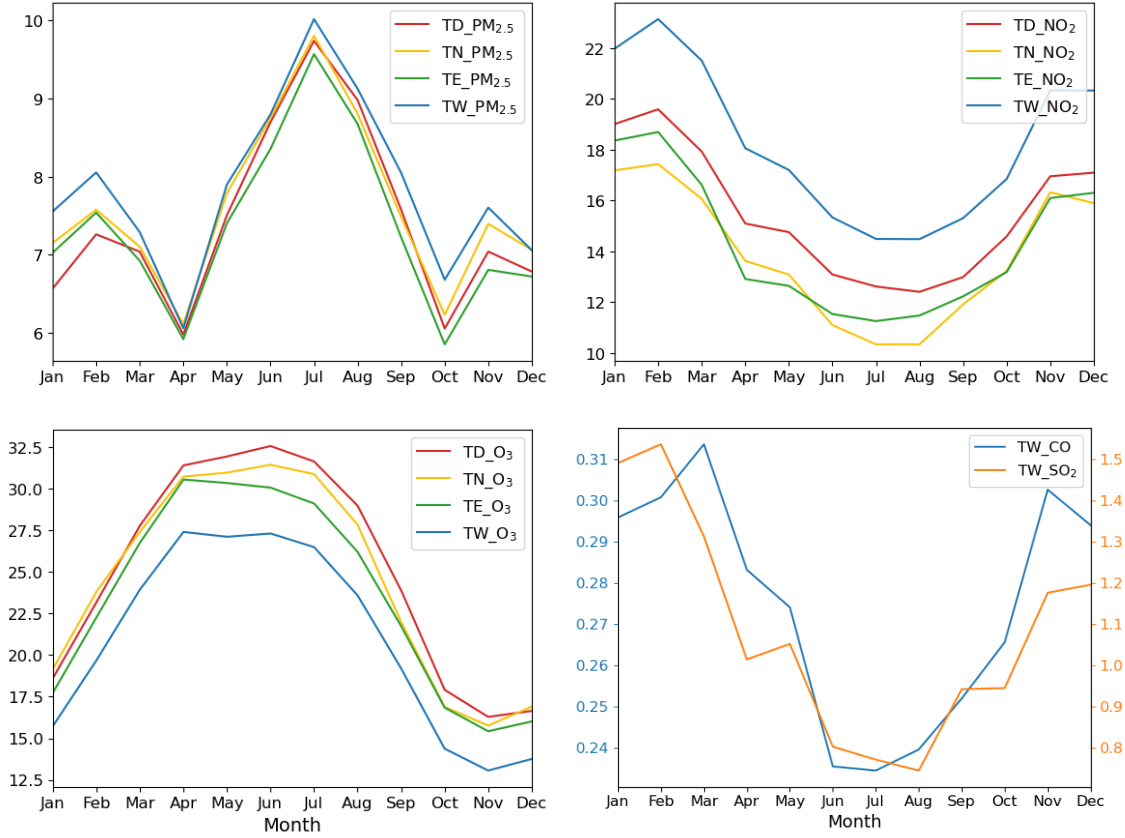


Figure 3.4: Mean monthly PM_{2.5} (µg/m³, top-left), NO₂ (ppb, top-right), O₃ (ppb, bottom-left), and SO₂ and CO (ppm-left axis, ppm-right axis bottom-right) concentrations, from 2003 to 2023.

Figure 3.5 displays the same as Figure 3.4, except broken down by hour.

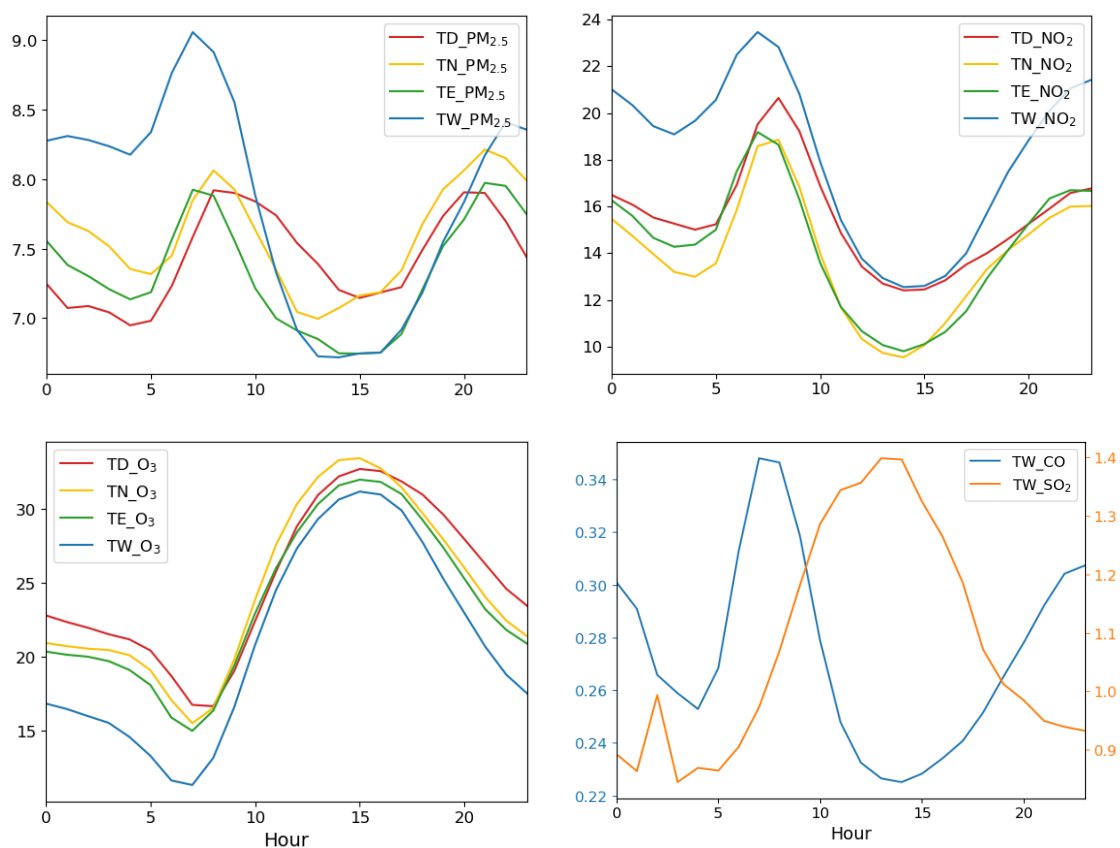


Figure 3.5: Same as Figure 3.4, except hourly instead of monthly.

Before displaying the polar plots, sample wind roses from January 1, 1974, to January 1, 2023, are displayed in the Figure 3.6 for Toronto City Center Airport, and Pearson International Airport.

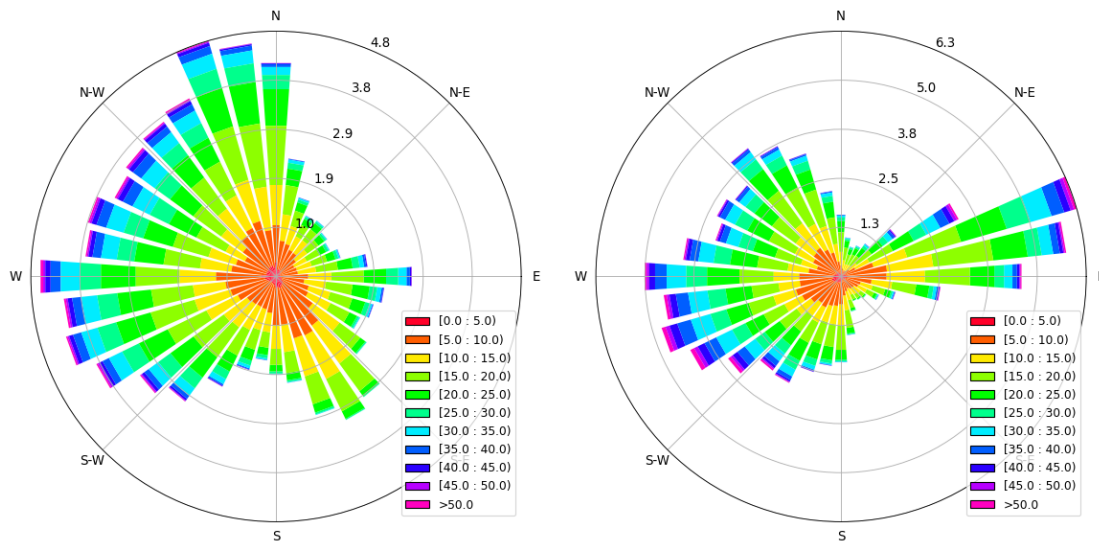


Figure 3.6: Toronto Pearson (left) and Toronto City Center (right) Airport wind roses, based on wind data from 1974-2023, normalized to have a total probability of 100%. The legend denotes wind speed in km/h.

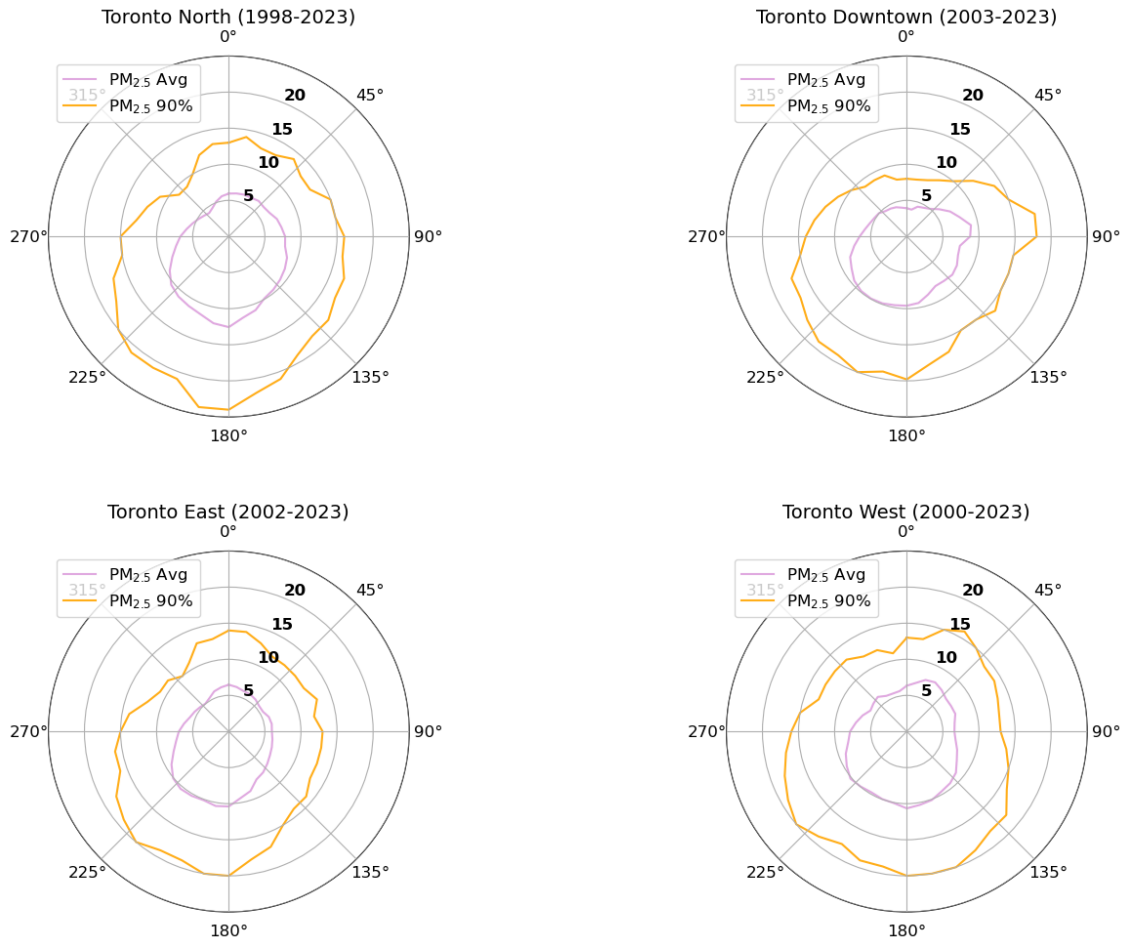


Figure 3.7: Average and 90th percentile PM_{2.5} concentrations ($\mu\text{g}/\text{m}^3$) with wind direction in degrees for the four air-quality monitoring locations in Toronto.

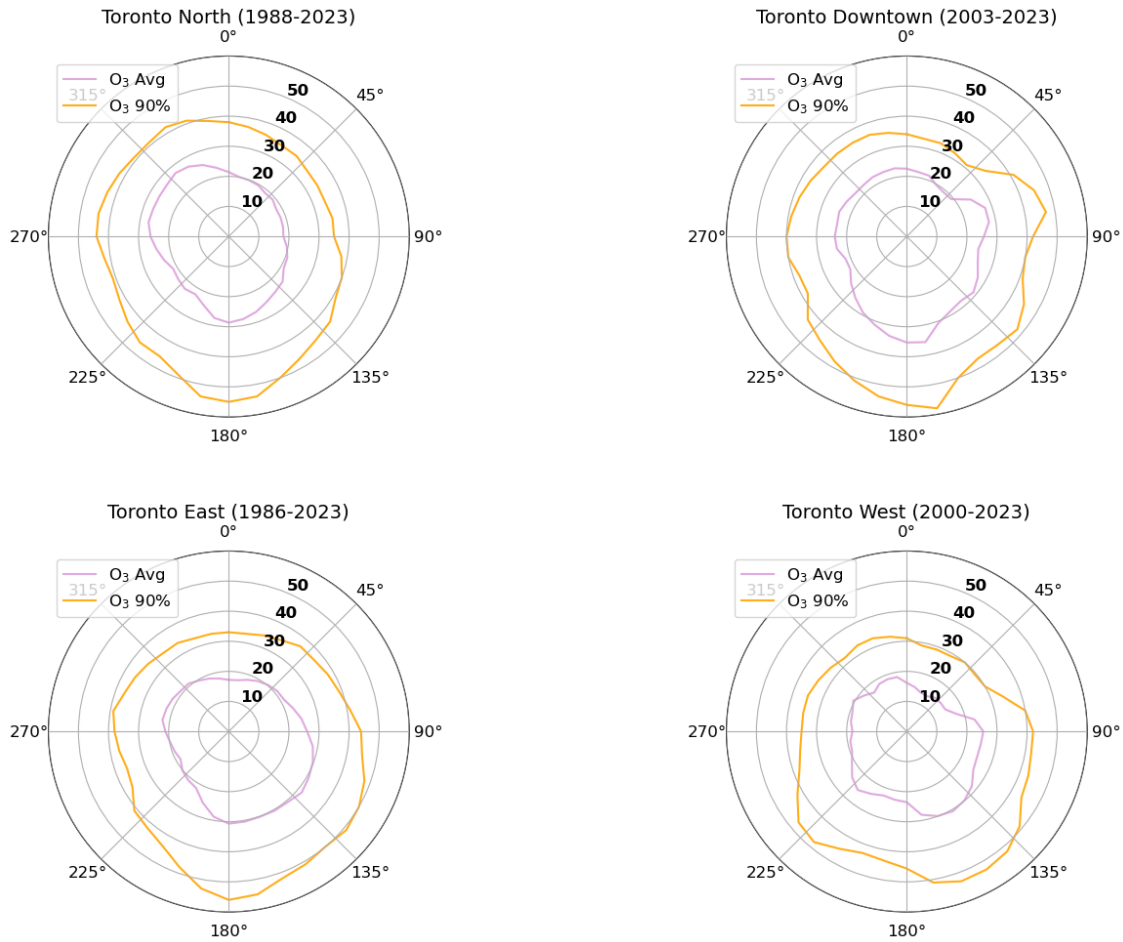


Figure 3.8: Average and 90th percentile O₃ concentrations (ppb) with wind direction in degrees for the four air-quality monitoring locations in Toronto. Note that, although “Toronto East” ozone measurements began on Jan 1, 1974 (see section A.3), the starting point for its polar plot was June 1, 1986. This is because, its corresponding wind measurement from Toronto Buttonville Airport did not begin reporting data until June 1, 1986.

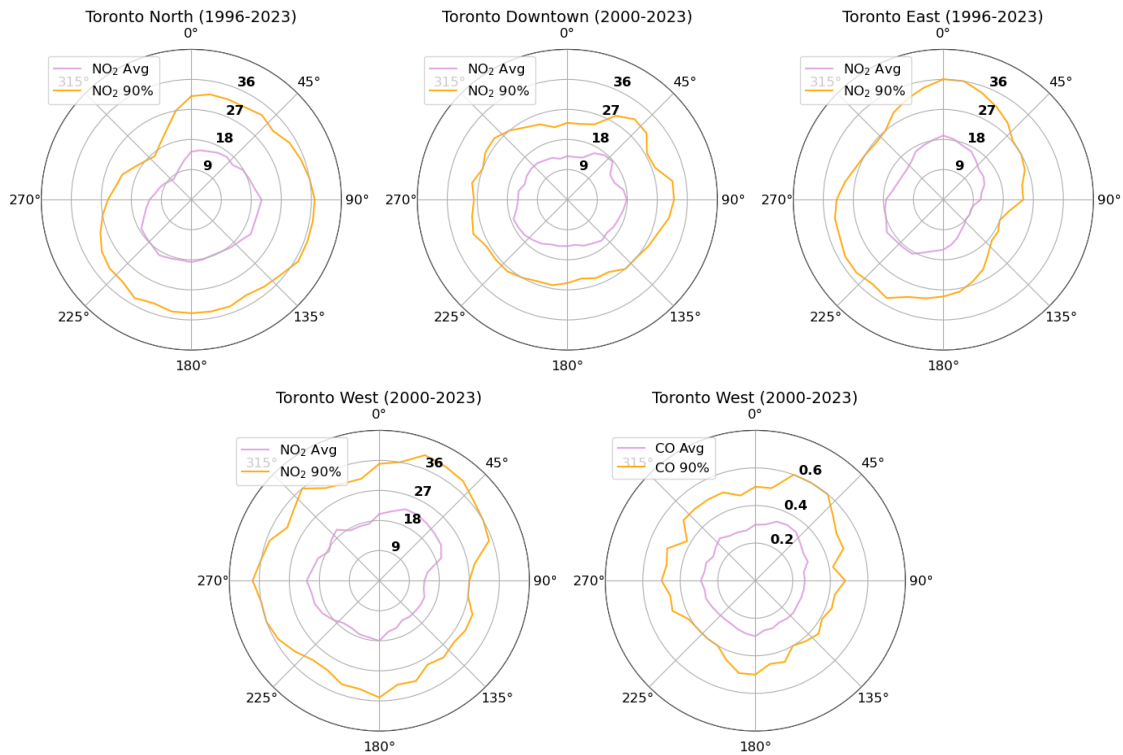


Figure 3.9: Average and 90th percentile NO₂ concentrations (ppb) on a polar plot with wind direction in degrees for the four air-quality monitoring locations in Toronto. Plotted on the bottom right is Toronto West CO in ppm.

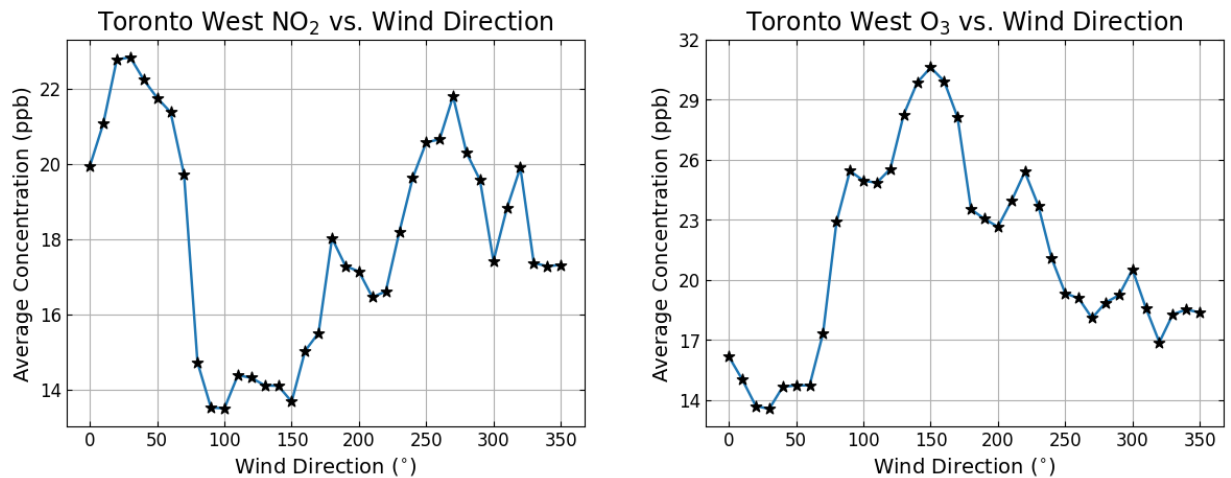


Figure 3.10: Toronto West NO₂ (left) and O₃ (right) average concentration per 10° wind direction, displayed on a Cartesian plot.

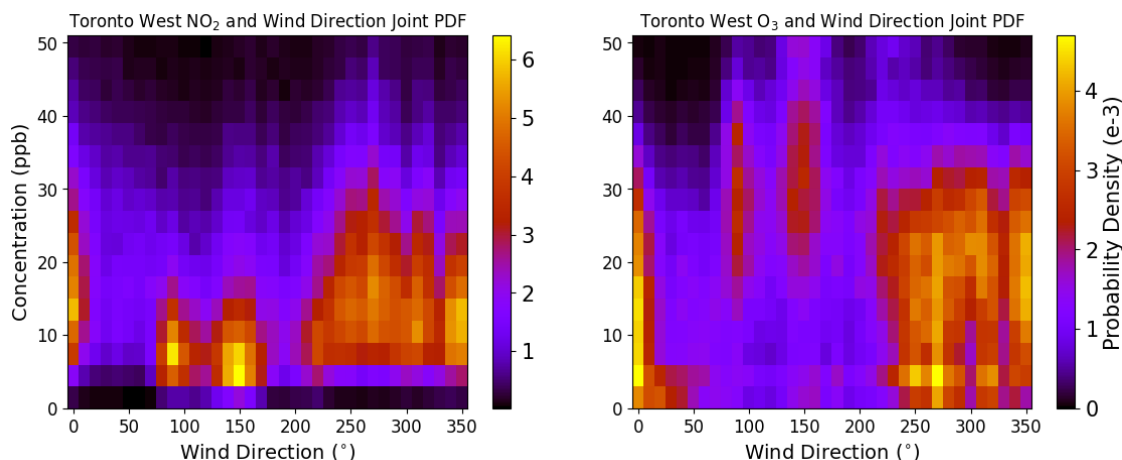


Figure 3.11: Probability of concentration occurrence by 10 degree wind direction for several concentration bins. Concentration is in bins of 3 ppb ranging from 0 to 51 ppb.

3.3 Discussion

Regarding the wind roses in Figure 3.6, more frequent winds are seen from the ENE direction at the Toronto City Center Airport compared to Toronto Pearson Airport. This is most likely due to an abundance of lake-breeze wind, which is not always observed at Toronto Pearson. However, despite the dominant wind direction being from around the WSW to north at Pearson, there is also a smaller but noticeable peak, approximately ranging from the SSE to SE. Given the direction and proximity of Pearson Airport to Lake Ontario, it can be assumed that some of this wind is also lake breeze induced.

In Figures 3.7 to 3.8, it appears $PM_{2.5}$ concentrations are the highest when the wind is coming from the southwest, including in Toronto Downtown, suggesting the dominant source(s) may be from beyond city limits. Southwest origination of $PM_{2.5}$ is in accord with the results of Jeong et al. (2020). Elevated concentrations from the south may also be from the Portlands Energy Center power station on Toronto Island. Again, from Figure 3.8, the dominant source of O_3 is most likely from the SE to South directions. Like the monthly averages, the highest NO_2 and lowest O_3 are seen in Toronto West. The polar plots in Figures 3.8 and 3.9 generally indicate an inverse relationship between ozone and NO_2 . This inverse relationship is further reinforced by examining the Cartesian and JPDF plots from the Toronto West AQ station in Figures 3.10, and 3.11, especially in the NE and SE directions.

Moreover, concerning Toronto North, East, and West, the dominant directions of high NO_2 concentrations are from a relatively wide range, centered about the NE and SW. For Toronto Down-

town, the NO₂ dominant directions are shifted slightly to be centered about the East and West. The Toronto West CO polar plot clearly shows that the dominant source of carbon monoxide is from the northeast, possibly from Highway 401, Downsview Airport, (as stated by Transport and Environment. (n.d.)), or the York Power Plant. These spatial variations, potential sources and relationships are further detailed in Chapter 4.

Trends with magnitude above roughly 0.10 µg/m³/yr were generally discernible for PM_{2.5}. Standard errors for all monthly and seasonal trends were computed to be ≤ 0.02 units in magnitude, meaning any slope > magnitude 0.04 is within 95% confidence interval. As such, NO, NO₂, NO_x, CO, O₃ and SO₂ trends in Tables 3.2 through 3.7 were deemed significant based on their magnitude. A study from 2007-2014 in Edmonton, Alberta, by Bari and Kindzierski (2016) showed mainly negligible overall PM_{2.5} trends. Although specific components of PM_{2.5} such as lead, barium, zinc, oxalate, organic carbon, elemental carbon etc, exhibited significant decreasing trends (ibid.), the component NaCl significantly increased, which suggest more wintertime road salt for de-icing (ibid.). Edmonton is urban, like downtown Toronto, with plenty of pollutant sources such as vehicular and industrial emissions. As stated in Air Quality Ontario. (n.d.-a), these vehicular and industrial sources are major emitters of PM_{2.5}. In Tables 3.2 through 3.7, it can be observed that though the average wintertime and summertime PM_{2.5} concentrations are increasing and decreasing respectively.

Relative the other species measured, the most significant trends are seen in NO₂, with the greatest decrease generally occurring in the spring/summer. For Toronto and its neighboring areas, Geddes et al. (2009) found that from 2000 to 2007, VOC reactivity and NO₂ concentrations underwent a 30 to 40% reduction during the summer months over the 7-year period. Though Geddes et al. (2009) chose May to September as their summer months. Averaged across the four chosen air quality stations in Toronto (Toronto East, West, North and South), the May-September concentrations decreased by 62.1% from 2003 to 2022. This implies an 7-year decrease of 22.9% per year, relative to the initial linear-fitted value, lower than the results from the study by Geddes et al. (2009), although different time periods were used for this analysis. Furthermore from 2004 to 2016 (note only the first hour of 2017 is available for NO), the yearly NO decreases were 6.5% amongst these four air-quality stations. From 2004-2017, NO₂ was found to decrease by 3.1% annually, relative to the initial fitted value, amongst the four AQ stations as well. These results are a little over half of the percentage changes found by Jeong et al. (2020), (as mentioned in Chapter 2, the NO and NO₂ were yearly changes computed to have declined by 11.1%, and 4.7% per year respectively). Toronto West was the only AQ station to report SO₂ concentrations from 2004 to 2017. In this range, SO₂ declined by 6.4% percent per year, relative to the initial linear-fitted value,

again roughly half the result from Jeong et al. (2020).

Generally, $PM_{2.5}$, NO, NO_2 , and NO_x decreased overall. As mentioned in the 2 section, a study by Hilker et al. (2021) concluded that improvements to vehicular technology, and 2004 to 2015 coal-powered electricity elimination in Ontario contributed to decreases in NO_2 emissions, as well as ultra-fine particles (UFPs), NO, SO_2 , and $PM_{2.5}$. Therefore, these policies may have contributed to the reduction in NO_2 and SO_2 in all stations in the Table 3.6, and increased ozone in every station except for Toronto Downtown, given the inverse relationship between NO, NO_2 , and NO_x with ozone observed in Tables 3.4 through 3.7. The reductions in NO_2 , NO, NO_x , SO_2 and CO are in line with the local vehicular exhaust emissions decline of roughly 3% annually from 2006-2016, stated by Jeong et al. (2020). Jeong et al. (2020) also found that this vehicular emission decline aligned with the emissions cutbacks from traffic-related air pollutants implemented federally and provincially. Interestingly, Toronto Downtown was the only station in Table 3.3 that observed an increase in $PM_{2.5}$, a summer-time decrease in ozone, the lowest summer-time decrease, and the lowest decrease of NO, NO_2 , and NO_x overall. Given the predominant sources of $PM_{2.5}$ stated in Air Quality Ontario. (n.d.-a), and NO_2 stated in Air Quality Ontario, (n.d.-b), this is perhaps due to the highest number of commercial operations and vehicle flow in this area.

General ozone increase matches the results from Zhang et al. (2022), who found ozone concentrations are generally intensifying (0.12 to 3.2 ppb/yr) in suburban/urban areas (Windsor, Ontario, Allen Park, Ontario, Erie, Ohio and Lansing, Michigan). There were three reasons proposed for this increase: 1) increased NO_2 photolysis effect from a decline in $PM_{2.5}$ concentrations, 2) decreased NO_x production from environments deficient in VOC, and 3) incremental VOC emissions growth (ibid.). However, the analysis by Geddes et al. (2009) did not find a noticeable trend in ozone concentrations.

In Figure 3.4, there is little variance in $PM_{2.5}$, but there appears to be two low points in the spring and fall, with peak during summer. This is in accord with the results from Zhao et al. (2018), who found increased and decreased $PM_{2.5}$ in the winter/summer and spring/fall in the United States. Moreover, Wang et al. (2021) found that $PM_{2.5}$ concentrations also have minimum values in April and October, in Canadian urban locations. They attributed high $PM_{2.5}$ summertime concentrations to an increased creation of secondary aerosol, stronger sunlight, and greater air temperatures.

Ozone peaks in the spring/summer, particularly in April to May, then reaches a minimum in November/December. This is in accord to the Air Quality Ontario. (n.d.-d) source, which stated that May through September is the time of year when ozone is typically the greatest. A similar

result also was found by Whaley et al. (2015), who showed that ozone in the lower part of the troposphere (ranging from 0 to 5 km) reaches its maxima in the late spring/summer (April to June), and surface-level ozone peaks in July. It is known that northern hemispheric ozone peaks at background locations during the spring (Vingarzan, 2004). The exact reasons are uncertain, though it is suggested that increased tropospheric photochemical reactions and/or transport from the stratosphere play a role (ibid.). In particular, Staley (1962) found that stratospheric to tropospheric particulate movement peaks and bottoms in the spring and fall respectively, which might be connected to the tropopause height. Škerlak et al. (2014) found that in the Tibetan Plateau, and western areas of North America, ozone levels at the surface are presumably dictated by downward stratospheric to lower tropospheric intrusions. These findings potentially explain the seasonality of ozone seen in Figure 3.4. It is worth noting the amount of stratospheric ozone results in a stratospheric temperature inversion, due to the absorption of UV solar radiation (Ritter., n.d.). Temperature gradually decreases with lower altitudes in the stratosphere since an increasing amount of UV radiation has already been absorbed (ibid.).

The MECCA (Module Efficiently Calculating the Chemistry of the Atmosphere) model was employed by Coates et al. (2016), to deduce how temperature, O_3 and NO_x associate with each other. When NO_x concentrations were high, ozone concentrations decreased by about 20 ppb when the temperature was increased from 20°C to 40°C (ibid.). However, ozone showed little temperature dependence when NO_x was low (ibid.). This may explain the peak in O_3 concentrations during April and May. Temperature rising and NO_2 and NO_x concentrations have not yet reached a minimum. Greater emissions from transportation and residential heating, which can gather in a denser, cooler atmosphere, are responsible for the wintertime peaks of NO_2 (Copernicus Atmosphere Monitoring Service, 2022). Greater heating and personal vehicular emissions, both from fossil fuel combustions are also responsible for higher wintertime CO and SO_2 concentrations as well (ibid.).

Like in Figure 3.4, in Figure 3.5, suggest VOC limited of NO_x saturated regime, as stated by Geddes et al. (2009), Seinfeld and Pandis. (2016), Sillman. (n.d.), Whaley et al. (2015), and Zhang et al. (2022) consistent with found by Geddes et al. (2009), Whaley et al. (2015), Zhang et al. (2022). NO_2 peaks in the morning and late evening, and bottoms in the afternoon. Since vehicular emissions reach a maxima in the morning and evening pre and post-commute (EPA, 2023), this is expected. Conversion of vehicular NO and O_3 to NO_2 emissions can occur under a minute in high temperature conditions with an adequate amount of ozone (ibid.). These vehicular emissions are then quickly diluted throughout the day under intense atmospheric mixing (ibid.), which elucidates the low afternoon NO_2 concentrations. NO_2 can also be converted to O_3 via photochemical

reactions (EPA, 2023). This also explains the minima, and maxima of O_3 concentrations during the morning, and early to late afternoon respectively. Ozone generally reaches its maximum from late morning to early evening (EPA, 2023). Morning peaks of $PM_{2.5}$ can also be explained through increased morning traffic (NYC.gov., n.d.), which also helps explain the evening spikes of $PM_{2.5}$ that align with NO_2 evening spikes (from traffic), as well. In a similar vein, CO – also emitted from motor vehicles (California Air Resources Board., n.d.), exhibits morning and afternoon peaks, as illustrated in Figure 3.5. As mentioned by Air Quality Ontario. (n.d.-d), since factories, smelting, and electric utilities are primary sources of SO_2 afternoon peaks of SO_2 can be attributed to emissions from power plant or similar sources. An interesting phenomenon is the greater width, but smaller magnitude in the evening spikes of NO_2 , SO_2 and $PM_{2.5}$. Perhaps this is due to a greater distribution of evening traffic times, and decreased emissions from other sources such as power plants and factories.

Another distinctive feature is the SO_2 spike at 2am in Figure 3.5. Figure 3.12 shows the wind-rose from Pearson Airport (closest to the “Toronto West”) location when the 2am to 1am concentration difference was at least plus 2 ppb. Although there are considerably less samples used in Figure 3.6 than the entire Pearson Airport wind-rose from 1974 to 2023 in Figure 3.6, a greater distribution of winds is observed from the northwest, and southwest, and a smaller distribution of winds is seen from the SE (Lake Ontario, and Downtown Toronto).

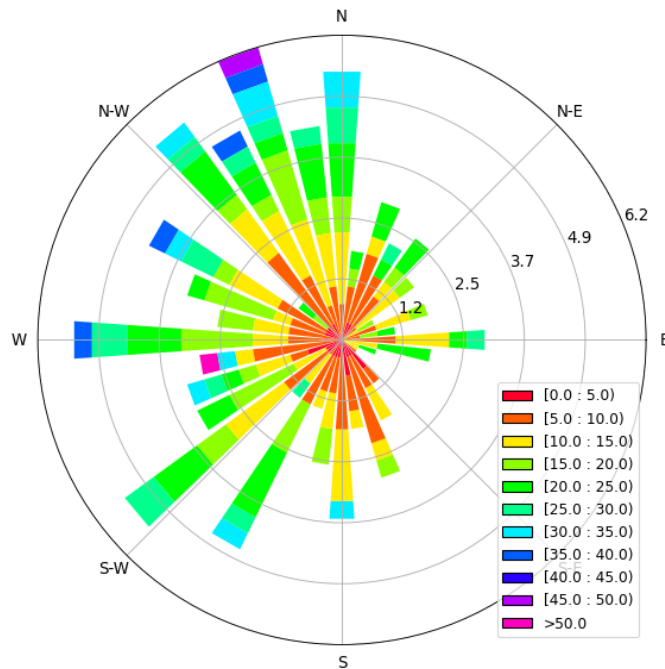


Figure 3.12: Wind rose for Toronto Pearson International Airport when the Toronto West SO_2 2am concentrations were at least 2ppb higher than the 1am concentrations.

4 Back-Trajectory Methods from Air-Quality Stations

In this Chapter, two back trajectory techniques were employed; a CWT, and another back-trajectory method which is a combination of CPF and CBPF (both described in section Methodology). Individual back-trajectories for each pollutant parameter for each site were employed to further analyze the spatial variations, and determine pollutant hotspots. The individual back-trajectories were then superimposed (to be further explained in section Methodology as well), to produce one map for each pollutant concentration, displaying the spatial variations of each pollutant throughout Toronto.

4.1 Methodology

The back-trajectories take a stationary concentration measurement and project them back in time using the wind speed and direction to create a spatial concentration map. The full process is explained in this paragraph. Time series of concentrations, wind speed, and wind directions of equal length were utilized to compute these back-trajectories. A range parameter was defined, given by $r = T \cdot WS[l]$ where, T ranges 0 to 7200 seconds, and $WS[l]$ denotes the l^{th} wind speed in m/s. The range parameter was chosen to end at $T = 7200$ seconds, since the back-trajectories were projected for two hours back in time based on wind speed and direction, and the wind speed units and pixel size are in m/s and m, respectively. Two hours was arbitrarily chosen (although the concentration and wind measurements are hourly), since it was observed that the back-trajectory span roughly covered all of Toronto based on the wind speed. The location at position (i, j) that the back-trajectory passes through is computed as $i = r \sin(WD[l])$ and $j = r \cos(WD[l])$ where $WD[l]$ is the l^{th} wind direction in degrees. In equation 2.1, the l^{th} concentration value is designated to each location (i, j) passed-over by the trajectory. This procedure can then be repeated for every concentration and its corresponding wind speed and direction.

To compute how long an air-parcel spends in a given grid square, $T_{i,j,l}$, each wind-direction in the array was divided into one-second points for the entire two hour period. From counting the number of points in a trajectory that pass through a grid square, the time the air spends in each grid square can be deduced. At each one-second point in the trajectory, the concentration value C_l , was added to the pixel. This is equivalent to computing $C_l T_{i,j,l}$ for a given pixel (i, j) , since $T_{i,j,l}$ is just 1 for each point. Starting with the first element of the concentrations, wind speed and wind direction series, $C_l T_{i,j,l}$ and $T_{i,j,l}$ of equation 2.1 can be computed. These computations are then repeated for each pair of concentrations, wind speeds and directions to calculate the numerator and denominator of equation 2.1, which are then put together to produce a CWT map.

The individual back-trajectories were mapped to a 60×60 km grid with a resolution of 1km centered about the measurement site. Each concentration value was back-tracked for two hours based on wind speed and direction. For the CWTs, as an example, this is shown in Appendix section of A.1. Furthermore, the individual back-trajectory maps generated by each individual station were averaged together and projected on a 60×60 km grid, with a 1 km resolution as well, to create an overall picture regarding the complete pollutant spatial variations in Toronto. Again, as an example this is displayed in section 4.2.1.1 for the CWTs. An arbitrary point (43.71212° , -79.40935°), roughly centered in the middle of the “Toronto East,” “Toronto Downtown,” “Toronto West,” and “Toronto North” air quality stations, was chosen as the center of these averaged maps.

It is important to note that on the grid containing the average of all the individual back-trajectories from each station, the amount of overlapping individual back-trajectories varied depending on the specific point on the grid. Essentially, in the superimposed or “averaged” back-trajectories, different number of stations are contributing to each grid point value. This will be referred to as “station overlaps.” For instance, some values on the grid may be comprised of 15 overlapping individual back-trajectories (15 station overlaps), while others may be based on 3 overlapping individual back-trajectories (3 station overlaps). The averaged back-trajectories were computed by taking a weighted averaged of the overlapping individual back-trajectories at each grid-point, where the weights were assigned based on the total number of concentration values per station in the time-frame.

As a second back-trajectory method, a combination of the CPF and CBPF were used to create another set of maps that considers both the wind speed and direction, but has non-discrete values. This will be referred to as “CPF (with wind speed)”. Although Zhou et al. (2024), stated that in most cases, C^* was the 75th to 90th percentile concentrations in equation 2.6, Nguyen et al. (2022), Squizzato and Masiol. (2015), and Zhou et al. (2019b), chose C^* to be the 75th percentile of the concentrations. Following Nguyen et al. (2022), Squizzato and Masiol. (2015), and Zhou et al. (2019b) approach, C^* in equation 2.6, was the 75th percentile concentration of the concentrations as well. In each 10° wind direction, the percent of concentrations above C^* , were computed, yielding a number between 0 and 1.

This methodology shows relative values instead of absolute values, and is more detailed than a PSCE, which only displays binary values of 0 and 1. In this case, the specific reason for choosing the threshold value was as follows: when comparing concentration values in individual locations, the maximum values could fluctuate by a significant amount. The anomaly of maximum

concentrations with respect to the mean concentrations by locations may vary. More consistent back-trajectories can be obtained across individual locations by selecting a threshold based on a particular percentile of concentration values. This back-trajectory was computed in the same way as for the previous two back-trajectories, where C_l is the ratio of concentrations counts greater than C^* over all concentrations per 10° directional bin.

4.1.1 Illustrated Example

Figure 4.1 illustrates how the back-trajectory equations used are applied.

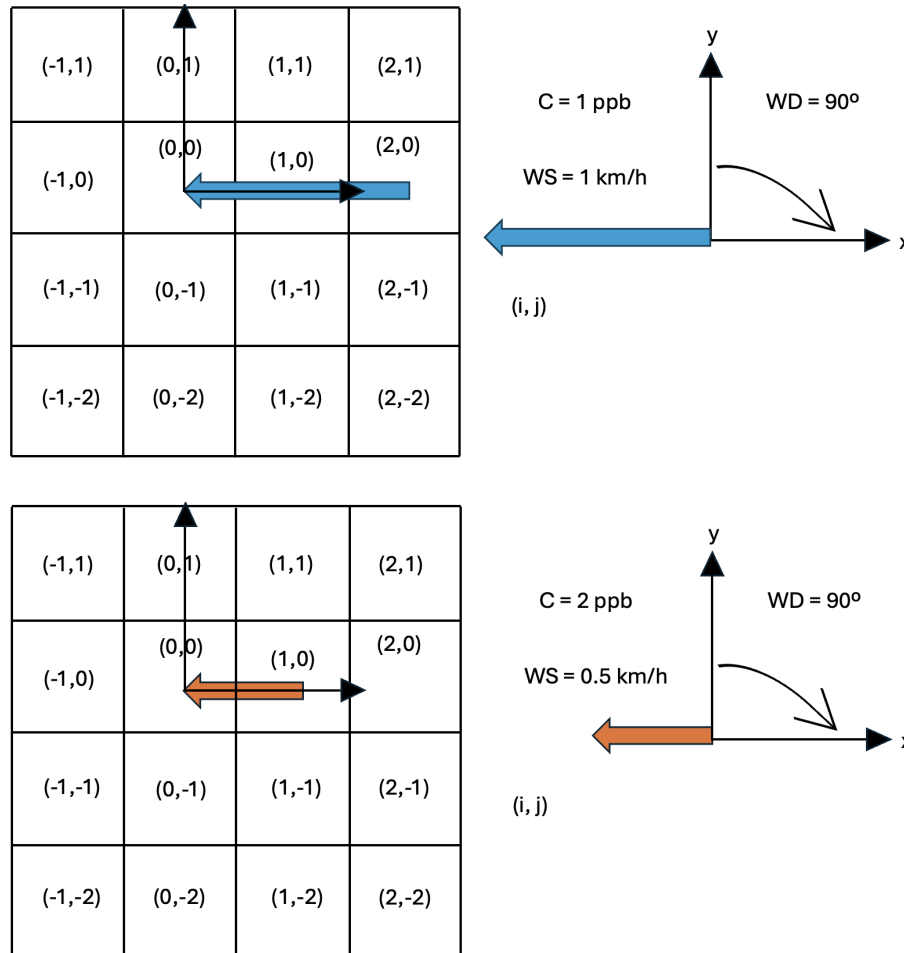


Figure 4.1: Diagram illustrating how pollutants are traced upwind, via sample concentrations ($[1,2]$ ppb), wind speeds ($[1,0.5]$ km/h), and wind directions (WD $[90,90]^\circ$) arrays, and how the angle is defined, differing from typical polar co-ordinate convention. The grid notation is given by (i, j) , corresponding to (x, y) . The right side shows the wind speed and direction transformation, and the left side displays the back-trajectory on the grid system. The first and second step are shown in the top and bottom part respectively.

In this simple example, let $(0,0)$ be the center of a station location. Let the grid-size be 1×1 km each. The arrows indicate which directions the wind comes from, until reaching the origin. Con-

centrations, wind speeds and wind directions with length 2 are employed. The blue arrow denotes the trajectory of the first concentration, wind speed and wind direction element, corresponding to 1 ppb, 1 km/h, and 90° . Starting at the center point (0,0), this trajectory travels exactly 2 kilometers towards the right over the 2 hour period to land at the center of point (2,0). Since the trajectory takes 1 hour to traverse 1 kilometer, the retention time at pixel (1,0) becomes 3600 seconds, given by $T_{1,0,1}$. Given the trajectory only traverses 0.5km in grids (0,0) and (2,0), $T_{0,0,1}$ and $T_{2,0,1}$ are assigned 1800 seconds. Thus the total time amongst the three pixels in this trajectory are 7200 seconds.

The orange arrow denotes the trajectory of the second concentration, wind speed and wind direction element, traversing 1 kilometer from the origin to the center of point (2,1). In the same way as described above, both $T_{0,0,2}$ and $T_{1,0,2}$ are assigned 3600 seconds. Computing the right hand side of equation (2.1) at the point (0,0) gives $(C_1 T_{0,0,1} + C_2 T_{0,0,2}) / (T_{0,0,1} + T_{0,0,2}) = ((1)1800 + 2(3600)) / (1800 + 3600) = 1.67$ ppb. It is worth noting that CWTs do not produce absolute concentrations, despite having units of concentrations. The value reported for each grid cell is weighted the residence times in that cell, depending on wind speed, and is essentially averaged over the temporal range used.

It is important to note that in meteorology the wind direction is shifted 270° from regular polar co-ordinates, with 0° on the unit circle beginning at point (0,1) and rotating clockwise, as shown in Figure 4.1 as well. This is opposed to polar co-ordinates where 0° would be at point (1,0) and rotate counter-clockwise. As such, the transformations are defined as $(x,y) = r(\sin\theta, \cos\theta)$, instead of $(x,y) = r(\cos\theta, \sin\theta)$ in regular polar co-ordinates.

4.1.2 Problem with CWT Method

When generating the CWT maps from all the concentrations, wind speeds, and wind directions in a particular location, it is important to note that the highest concentrations would tend to the center. In contrast, the lowest concentrations would be on the outer edges. This is observed in Figure 4.2. From examining Figure 4.3, it is clear that this is due to a negative relationship between wind speed and concentrations. Faster winds enhance pollutant dispersion throughout the boundary layer, from increasing mechanical turbulence (GOV.UK., 2024). Furthermore, emissions are spread over a greater area with greater wind speeds. As such, these results are expected. Ozone was the only pollutant where the opposite of this phenomenon occurred, which is also expected given the inverse relationship between NO, NO₂, NO_x, and ozone observed in Chapter 3.

Another phenomenon observed in Figure 4.2, is the apparent step change. In the CWTs with

10° wind directions extending radially outwards (displayed in Figure 4.4-left below), these steps changes are also apparent. Figure 4.4 (right) displays a cross section of trajectory count along the $i = 30$ pixel line (270° and 90° wind-direction line). As can be observed, step changes are apparent. Notably, these step-changes occur in every increment of two pixels. This is because, the wind speeds measured by the airport automated weather stations are in increments of 1 km/h, which covers two pixels in the CWTs. For instance along the 270° line, a wind speed of 6 km/h would traverse 12 pixels from the center, where a wind speed of 7 km/h would traverse 14 pixels from the center. Since there is a discrete drop in frequency between the occurrences in 6 and 7 km/h winds, there is a discrete drop between the number of trajectories reaching the 11th to 12th pixel and 13th to 14th pixel from the center. Note, the trajectory counts near the center $(i, j) = 30, 30$, are high since this area represents the intersection of many directional trajectories before branching outwards.

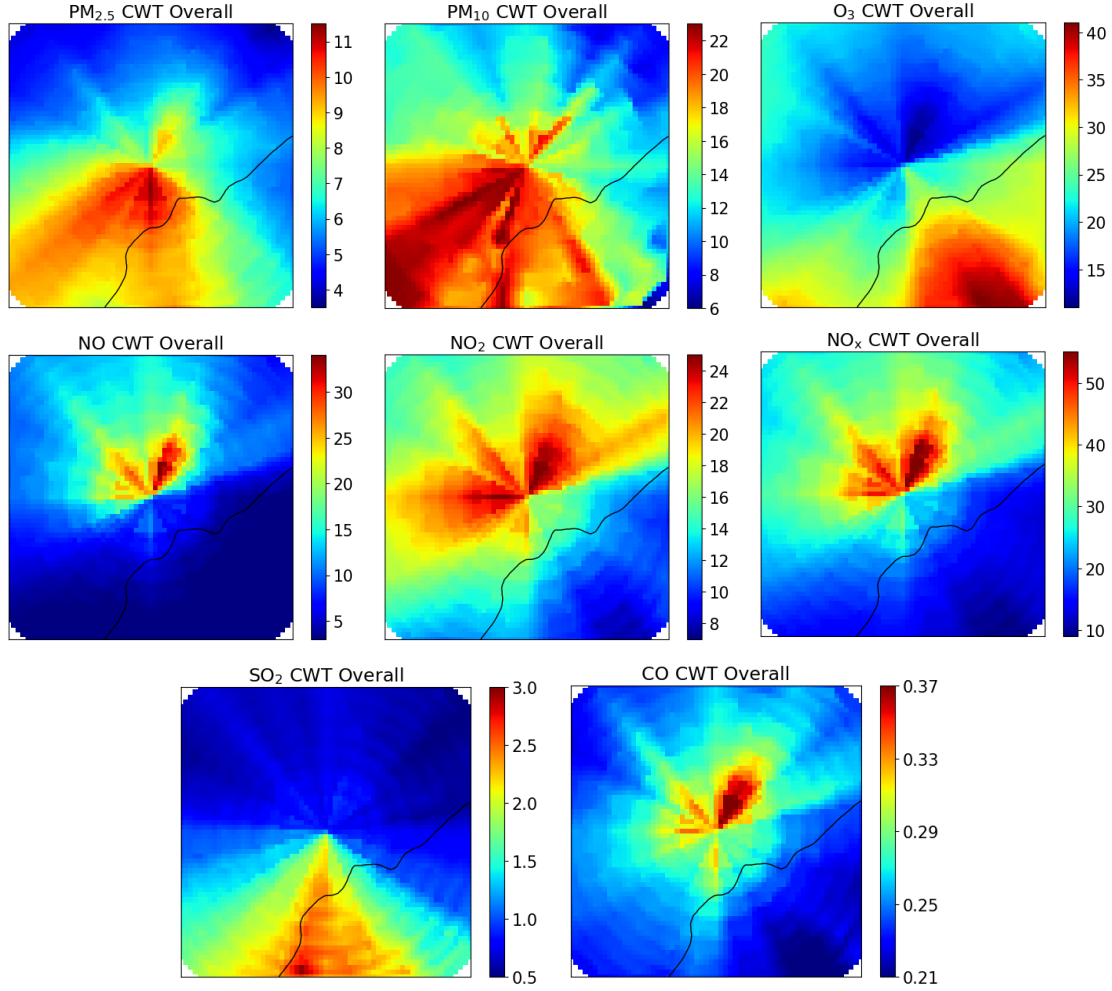


Figure 4.2: Generated CWTs using all of the data-points for a given station concentration, plotted on a 60×60 km grid, with the winds traced 2 hours back in time based on the wind speed and direction. These are for the Toronto West AQ stations. Units are $\mu\text{g}/\text{m}^3$ for $PM_{2.5}$ and PM_{10} , ppb for O_3 , NO , NO_2 , NO_x , and SO_2 and ppm for CO . These CWTs are similar to a bivariate polar plot, which takes a mean concentration per wind speed and direction bins (Carslaw & Beevers, 2013).

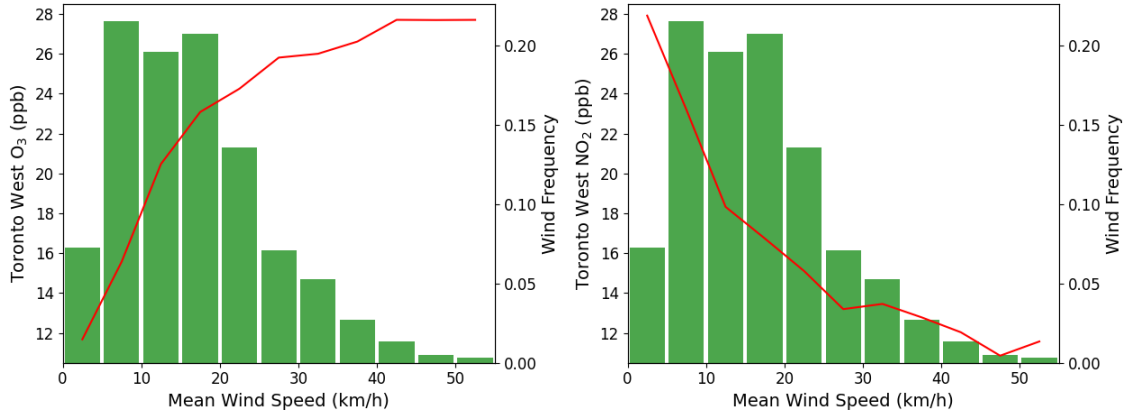


Figure 4.3: Mean NO₂ (left), and O₃ concentration ppb for the Toronto West AQ station binned by wind speed (shown by the red line). Wind frequency by binned wind direction is also plotted in green bars, displayed as a percentage.

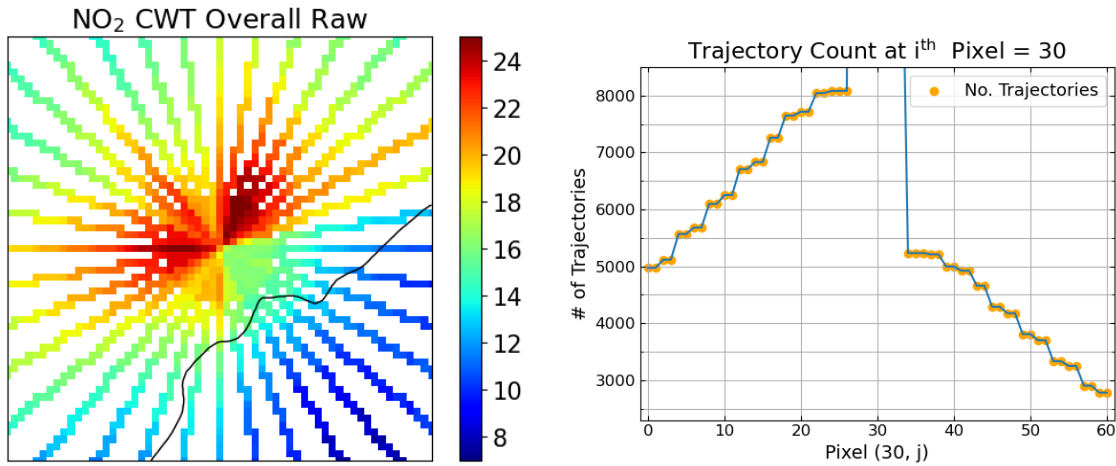


Figure 4.4: Raw Toronto West NO₂ CWT (without linear interpolation) (left), number of trajectories at the i^{th} pixel = 30 cross-section.

4.1.2.1 Solution

Regardless of the step-change phenomena, these back-trajectory results are ineffective at determining spatial variations, and deducing pollutant hotspots relatively far from the origin. An optimal method of generating CWTs is to take average concentrations, wind speeds, and wind directions per 10° wind bin, then employ a linear interpolation to fill in the extra whitespaces. This is shown in the section A.1, and essentially provides the directions in which the highest/lowest concentrations are. Placing all of the individual CWTs in on a larger map and taking a weighted average, as described above, in theory, can deduce spatial variations, and particular neighborhoods exhibiting high concentrations or hotspots through triangulation.

Although, there are still some setbacks from considering averaged concentrations along a particular direction, in the individual back-trajectories (see Figure 4.5, for instance). One weakness includes the assumption of a constant mixing ratio along a trajectory. This assumption does not consider any sources that may be present, or photochemical reactions that can occur between the endpoint and origin. Another potential weakness is utilizing reported wind speed and directions from airports. Despite Airport stations selected based off nearest distance to AQ station, in some instances the distance is still relatively large (recall Figure 3.1). Assuming straight line, constant wind speeds between the airport to station, or endpoint to origin of trajectory does not take into account topography changes, or urban meteorology, which can alter both the winds and mixing ratios.

Two examples of pixelated CWTs are displayed in Figure 4.5. Both Toronto West PM_{2.5} pre and post linear interpolation are displayed to demonstrate the necessity of interpolation when using average concentration and wind speed per 10° wind direction, as well. In section 4.2 back-trajectories were mapped on a filled contour plot, where contour lines were first specified based on a gridded 2-D array (i.e. in Figure 4.5 (right)), and the values in between these contours were filled to create a smoothened visualization.

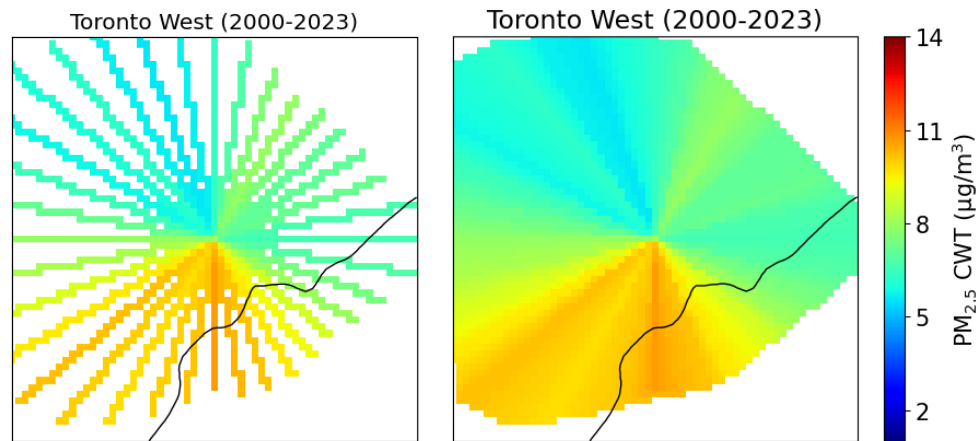


Figure 4.5: Gridded versions of a Toronto West PM_{2.5} CWT pre-linear interpolation (left), post linear interpolation (right).

4.1.3 Socioeconomic Mapping

A practical application of determining the spatial variations is to deduce which neighborhoods exhibit the greatest exposures, and if socioeconomic factors also play a role. Custom maps displaying 2021 census parameters such as population, population density, and median income post-tax in 2020 by mesh-grid in Toronto can be produced on the Census Mapper website, and this data can

also be extracted from the map (Census Mapper, n.d.). More detailed geographical maps regarding these districts are displayed on the Statistics Canada website (Statistics Canada, n.d.). Geographical co-ordinates of district centers in Figure 4.6 (see below), were manually determined based on the specific district outline on the Statistics Canada website. The nearest 1km grid to the manual co-ordinate selected for the districts were chosen for this comparison. The total amount of people impacted by relatively high levels of pollutants was also calculated. This was done by summing the total district populations, where the averaged concentrations were $\geq 75^{\text{th}}$ percentile of the pollutant concentrations across all neighborhoods in Toronto. Pollutant values in the averaged back-trajectories for each species were also related to neighborhood population density, and net median income. These relationships are investigated in the sections 4.2.1.2 and 4.2.1.

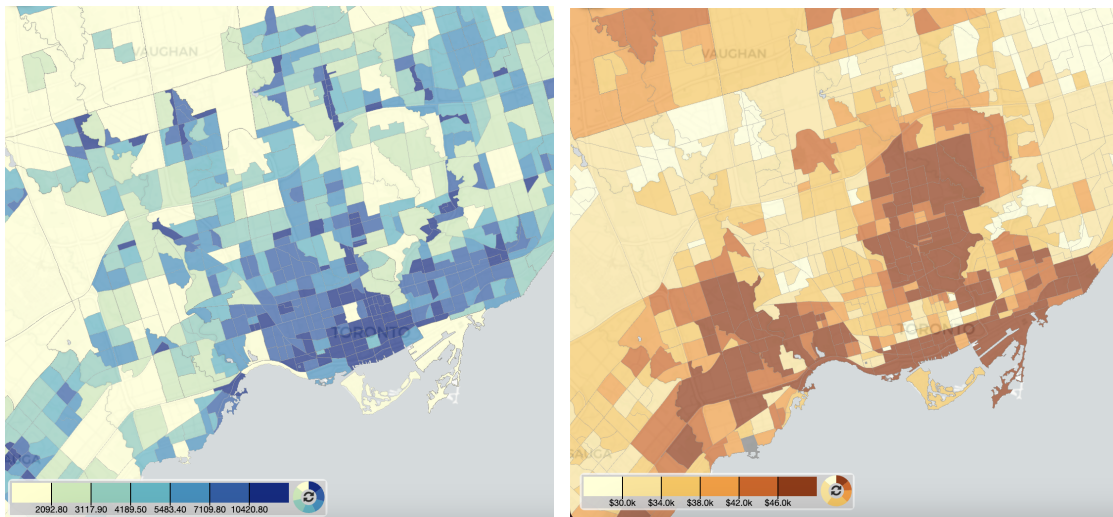


Figure 4.6: Population density (2021, left), and the median income post taxes (2020, right), by district in Toronto and its surrounding areas, produced on the Census Mapper website (Census Mapper, n.d.).

4.2 Results

4.2.1 Concentrated Weighted Trajectories

4.2.1.1 Averaged Concentrated Weighted Trajectories

Following the procedure outlined above of computing the averaged back-trajectories (as outlined in section 4.1), weighted averaged CWTs are displayed in Figure 4.7. In these averaged back-trajectories, to just focus on data within the city, and reduce clutter in these maps, only values with a certain number of station overlaps are displayed. The number of overlaps were at least 9 for $\text{PM}_{2.5}$, 8 for CO, 9 for SO_2 , at least 11 for NO, NO_2 and NO_x , and at least 3 and 17 for PM_{10} and O_3 respectively. Contour lines displaying the boundaries of the number of station overlaps for each pollutant are shown below in Figure 4.8.

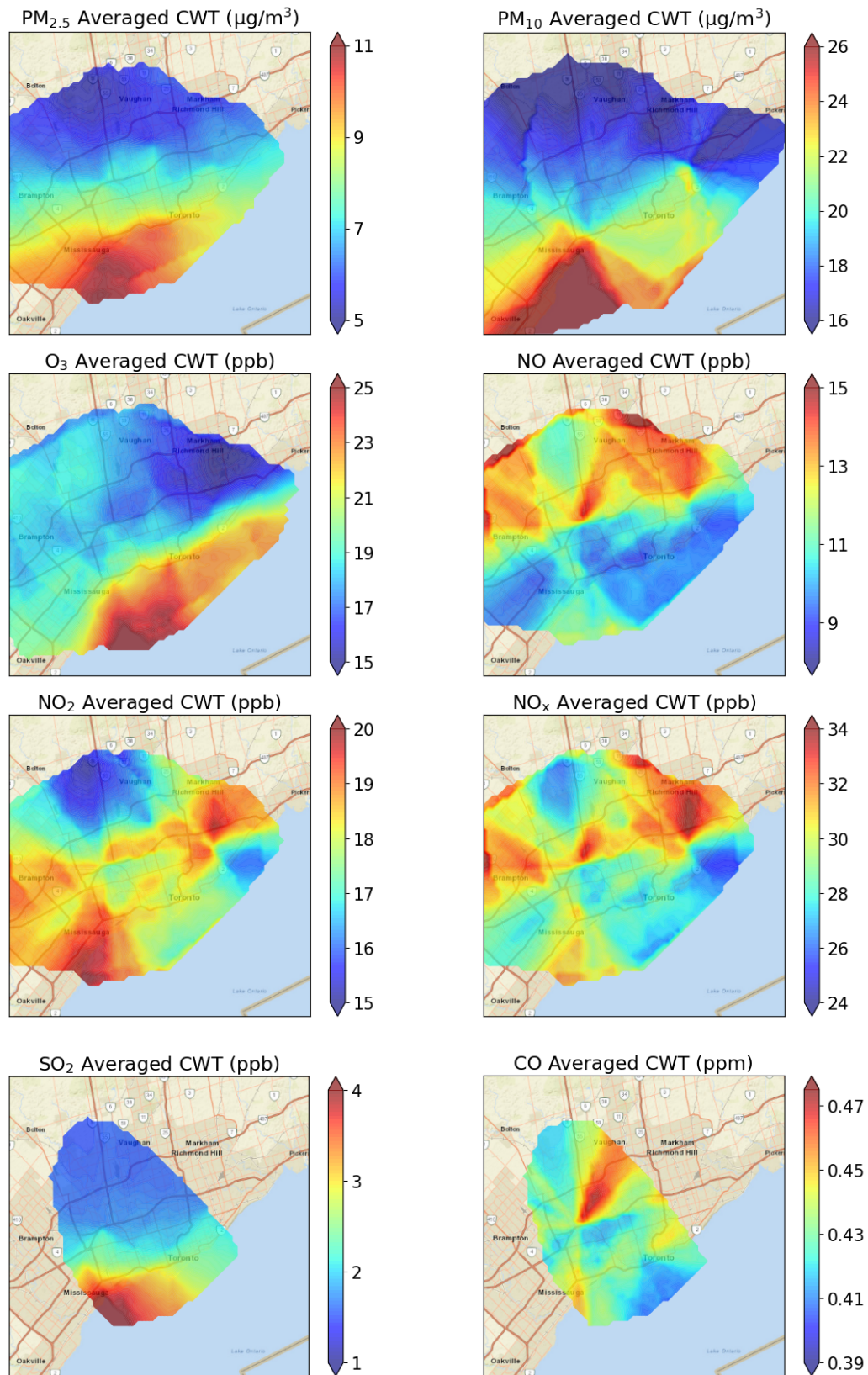


Figure 4.7: Superimposed CWTs for all pollutants for all stations.

In Figure 4.9, CO and SO₂ are displayed with at least 7 and 8 overlaps respectively, and discontinuities are observed. Figure 4.7, shows CO and SO₂, with at least 8 and 9 station overlaps. Although the spatial maps do not cover all of Toronto, they were officially used since linearly extending any back-trajectory for longer than two hours may result in errors.

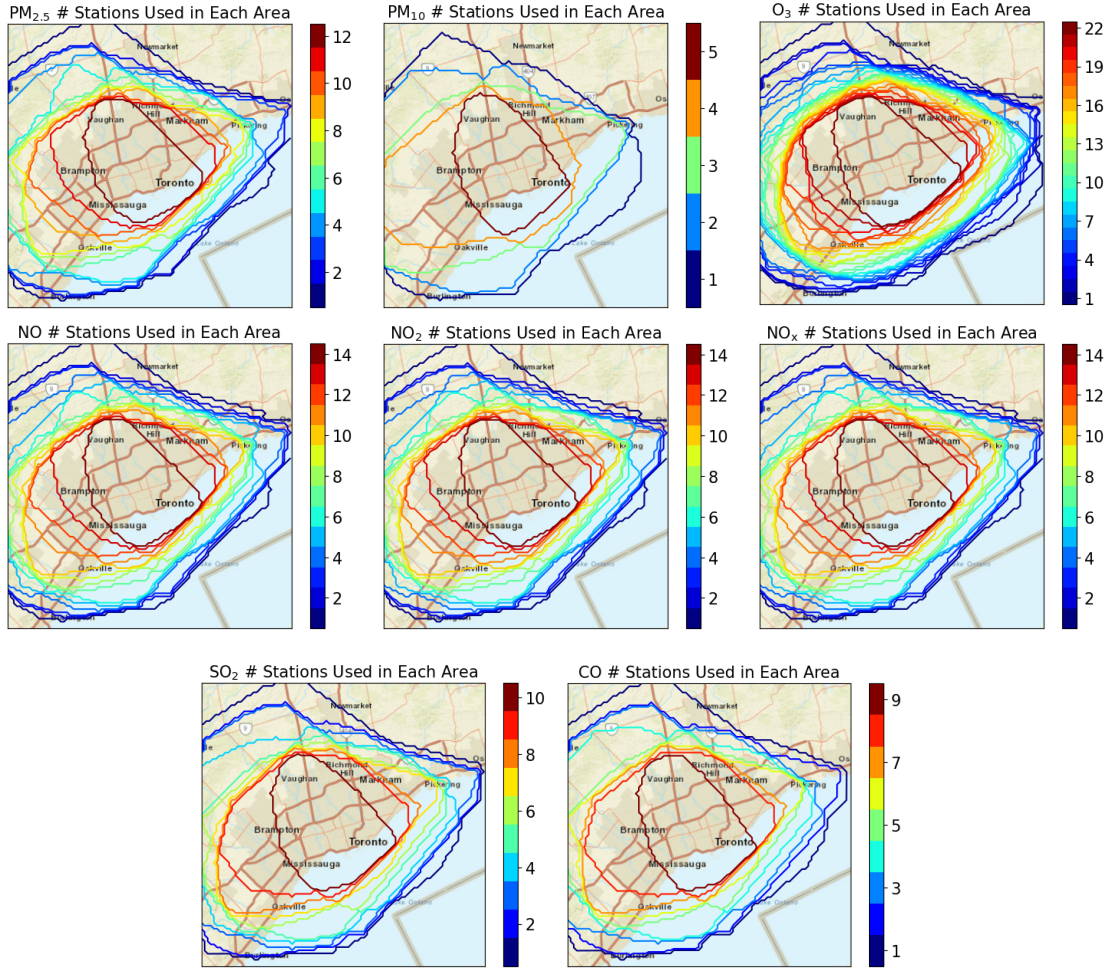


Figure 4.8: Number of station overlaps for each species used. Note that the NO, and NO_x station overlaps are identical since the exact same winds are being employed. Zoomed out on a 90 x 90 km grid for illustration.

High concentrations of PM_{2.5}, PM₁₀, NO₂, O₃, and SO₂ in south Toronto may be from a power plant in Nanticoke, Ontario that closed in 2013. To assess the potential impact of the Nanticoke power plant emissions, the averaged back-trajectories were taken pre and post-2013, as shown in Figures 4.11, and 4.12 respectively. Given the number of overlaps differed in the pre and post-2013 back-trajectories. Again, to only focus on Toronto and remove discontinuous data outside the city, in these pre and post-2013 back-trajectories, only values with certain amount of overlaps were displayed as well. In the pre-2013 back-trajectories, within a select region in Toronto, there were

6 overlaps for $\text{PM}_{2.5}$, 8 for CO, 9 for SO_2 , NO, NO_2 and NO_x and 16 for O_3 . In the post-2013 back-trajectories, 3 overlaps were used for SO_2 , and CO, 6 for $\text{PM}_{2.5}$, NO, NO_2 and NO_x , and 7 for ozone. To maintain consistency for comparison, the concentration range for each species was the same, with exceptions if the entire back-trajectory values were outside the range.

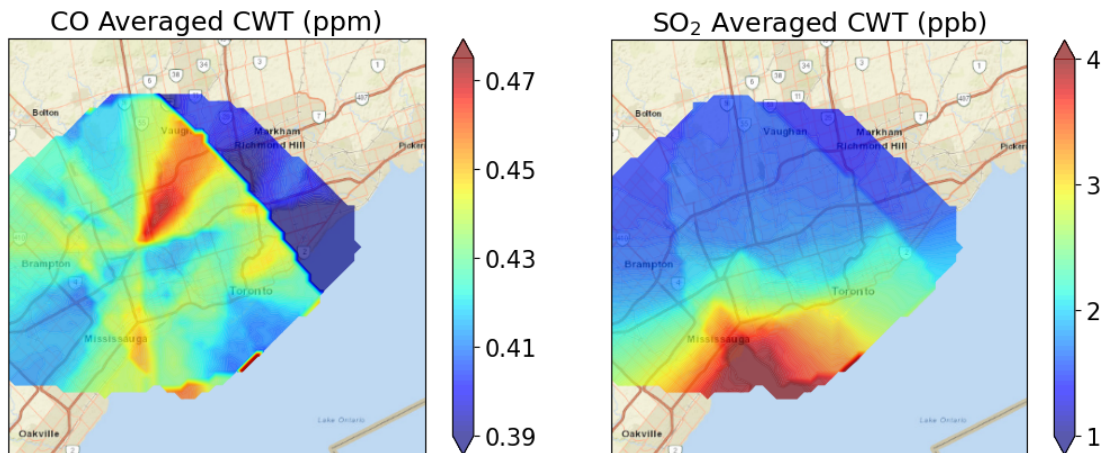


Figure 4.9: SO_2 (left) and CO (right) CWTs when just at least 7 and 8 overlapping individual CWTs are considered for CO and SO_2 respectively.

The spatial variations of $\text{PM}_{2.5}$ can perhaps is highlighted by zooming in on the CWT map, and reducing the color bar range. This was done in Figure 4.10 below. The grid is (32×32) km, centered about 43.73010°N , 79.38446°W .

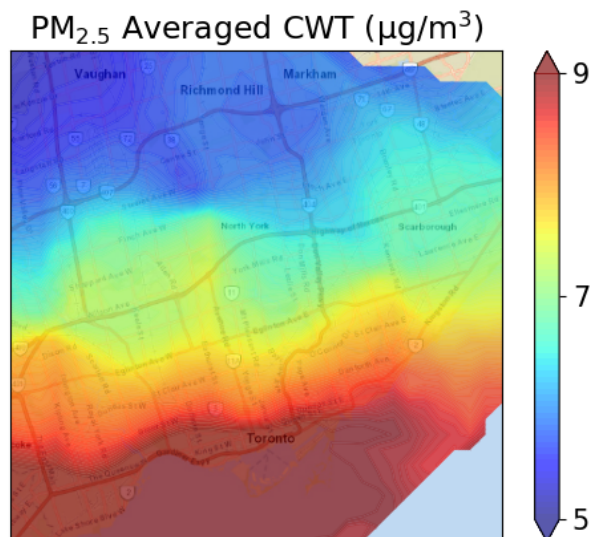


Figure 4.10: Zoomed in and modified color bar version of the $\text{PM}_{2.5}$ superimposed CWT in Figure 4.7.

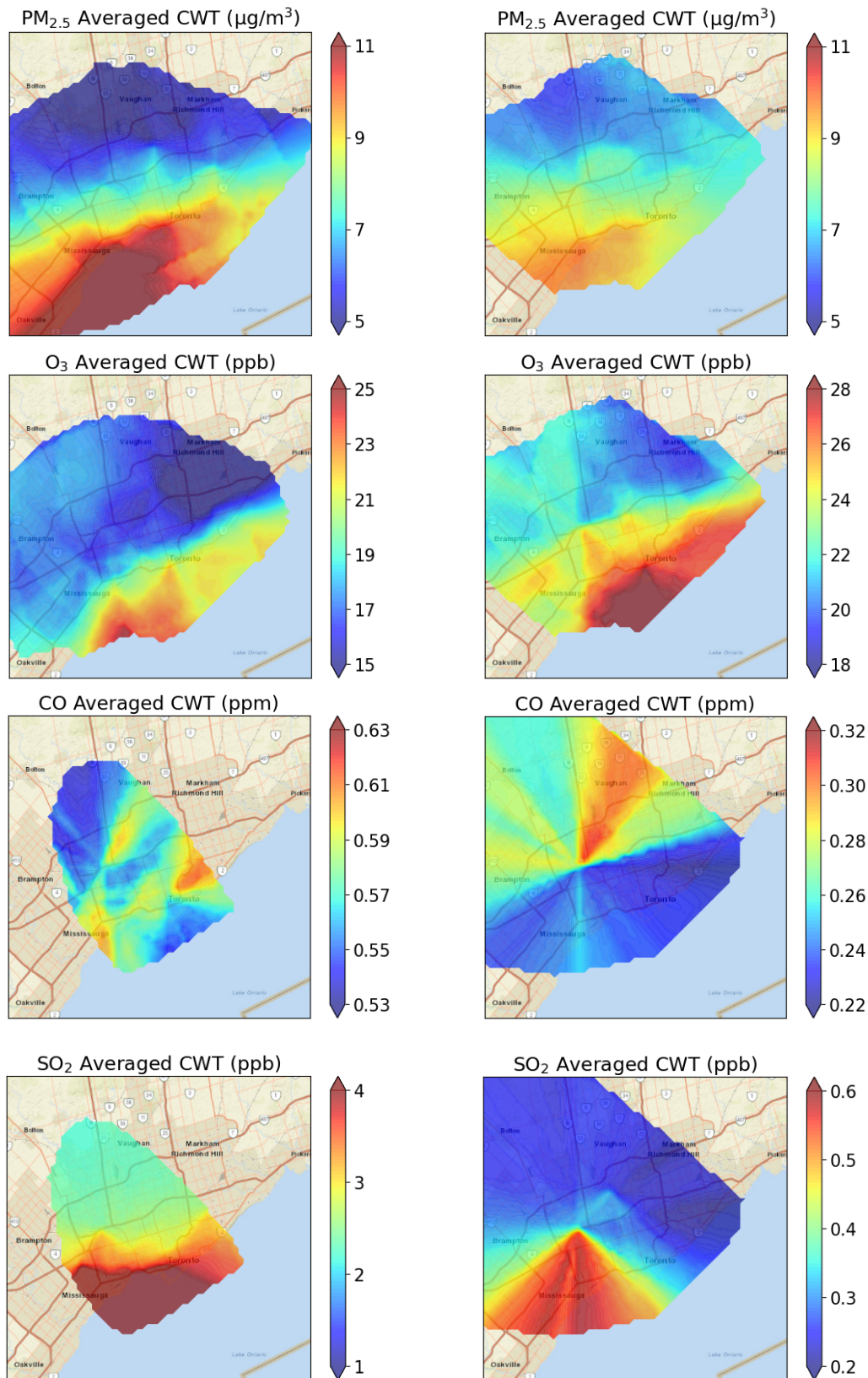


Figure 4.11: PM_{2.5} (1st row), O₃ (2nd row), CO (3rd row), and SO₂ (4th row) pre and post-2013 back trajectories, where the pre and post 2013 back-trajectories are shown in the left and right columns respectively.

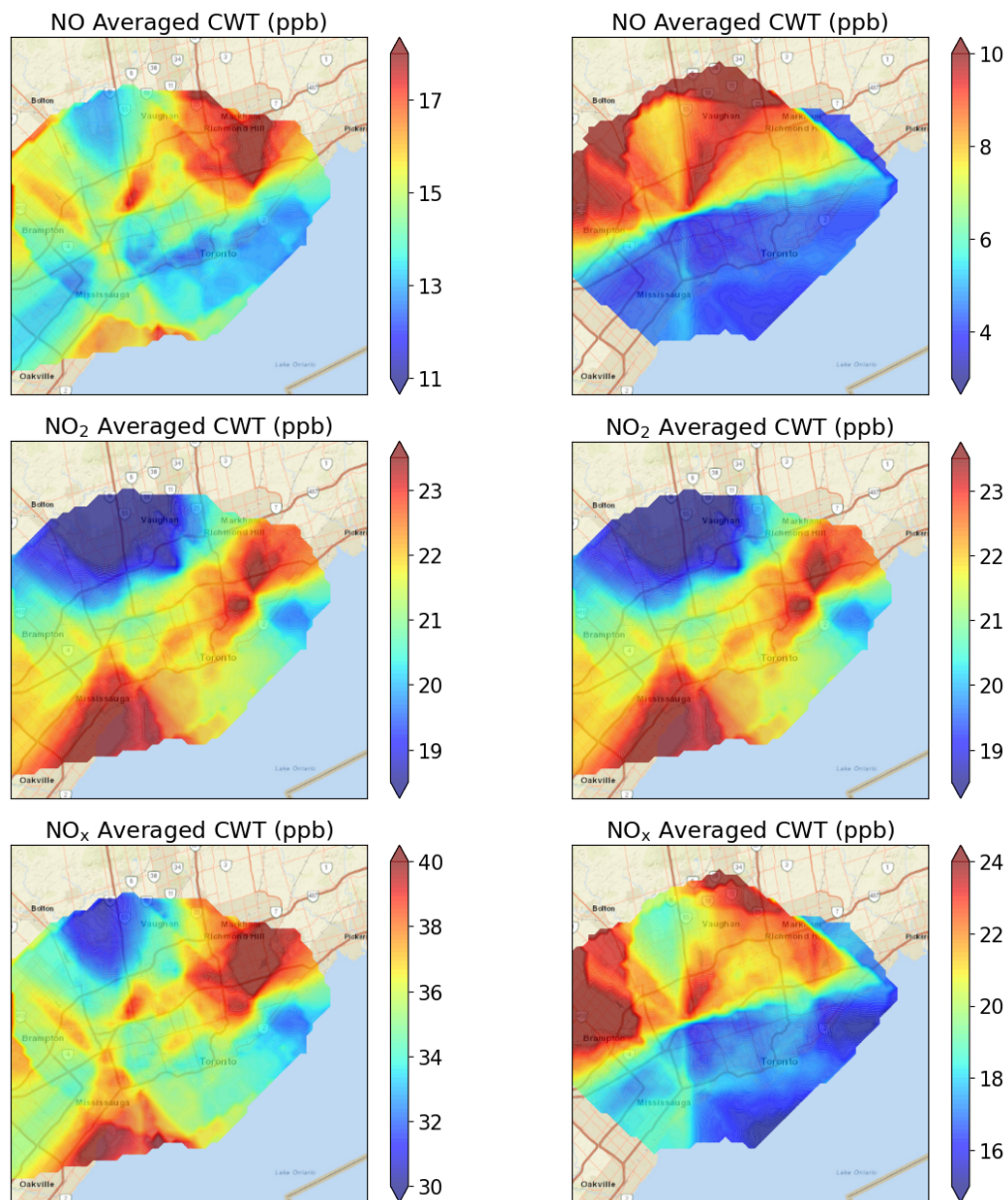


Figure 4.12: As in Figure 4.11, but regarding NO (top row), NO₂(middle row), and NO_x(bottom row).

4.2.1.2 Concentrated Weighted Trajectory Socioeconomic Mapping

Population density and net median income by neighborhoods in Toronto is displayed below. Table 4.1 shows the number of people impacted by high relative CWT concentrations ($\geq 75^{\text{th}}$ percentile of the CWT neighborhood values), and the correlation coefficient (R) between population density, or median income post-tax, as well as the computed averaged CWTs for each species in Figure 4.7. Sample relationships between the concentrations in Figure 4.7 and net median income for each 1x1 km grid square are presented in Figure 4.13.

Table 4.1: Number of people affected by high concentrations of pollutants (first row), and the R value between population density and concentration (second-row), and net median income and concentration (third-row). In the second and third row, R is displayed in the first row, and its corresponding standard error (SE)/slope is shown in the second row. This is based on the entire CWT time period shown in Figure 4.7.

	PM _{2.5}	PM ₁₀	NO	NO ₂	NO _x	SO ₂	O ₃	CO
No. Affected	694291	714573	700662	688424	702590	697496	706340	688432
Pop. Density R,	0.16	0.18	-0.17	-0.05	-0.15	0.05	0.20	-0.09
SE/slope	0.25	0.23	0.24	0.77	0.28	0.95	0.21	0.51
Net Median Income R,	0.46	0.42	-0.48	-0.12	-0.41	0.31	0.43	-0.28
SE/slope	0.08	0.09	0.08	0.35	0.09	0.14	0.09	0.16

Since high concentrations of NO₂ were observed to most impact the neighborhoods around Highway 401 (Toronto's busiest highway), the overall NO₂ CWT map in Figure 4.7 was overlayed on the income-by-district map in Figure 4.6.

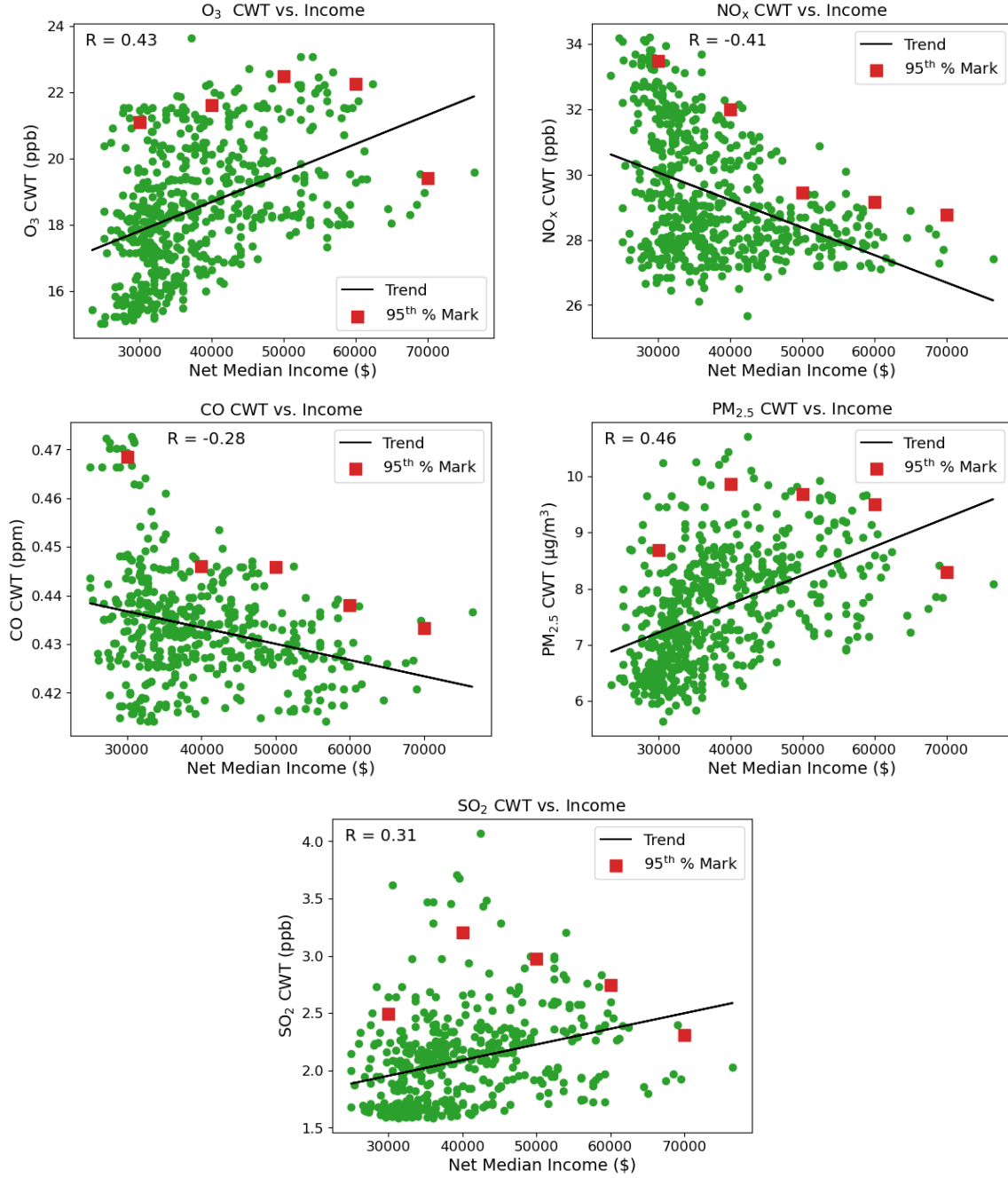


Figure 4.13: Relationships between the averaged O₃ (1st row-left), NO_x (1st row-right), CO (2nd row-left), PM_{2.5} (2nd row-right), and SO₂ (bottom row) versus CWT with net median income (\$). Linear trend-lines are displayed in black. This is based on the superimposed CWT in Figure 4.7. The 95th percentile concentration bins based on income are also displayed, where the bins were chosen as $x \in [x - 5000, x + 5000)$, where $x = 30, 40, 50, 60$, and 70 thousands of dollars.

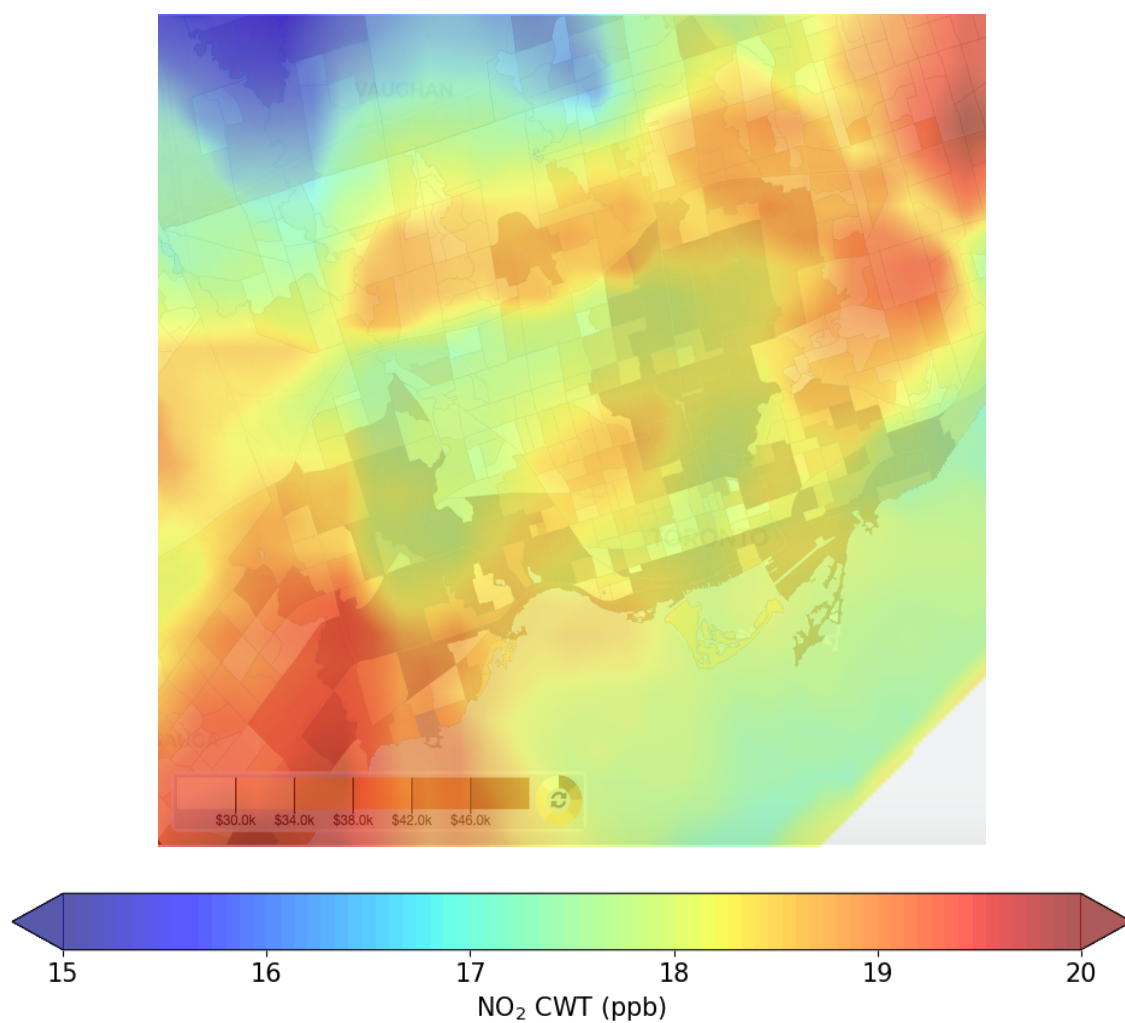


Figure 4.14: Spatial Variation NO₂ CWT map from Figure 4.7, overlaid atop the median post-tax income variation from Figure 4.6.

4.2.2 CPF (with Wind Speed)

Figures 4.15, 4.16, and 4.17 show the total superimposed back-trajectory maps using the CPF with wind speed method, and the maps pre and post 2013 respectively. These results are discussed in section 4.3.

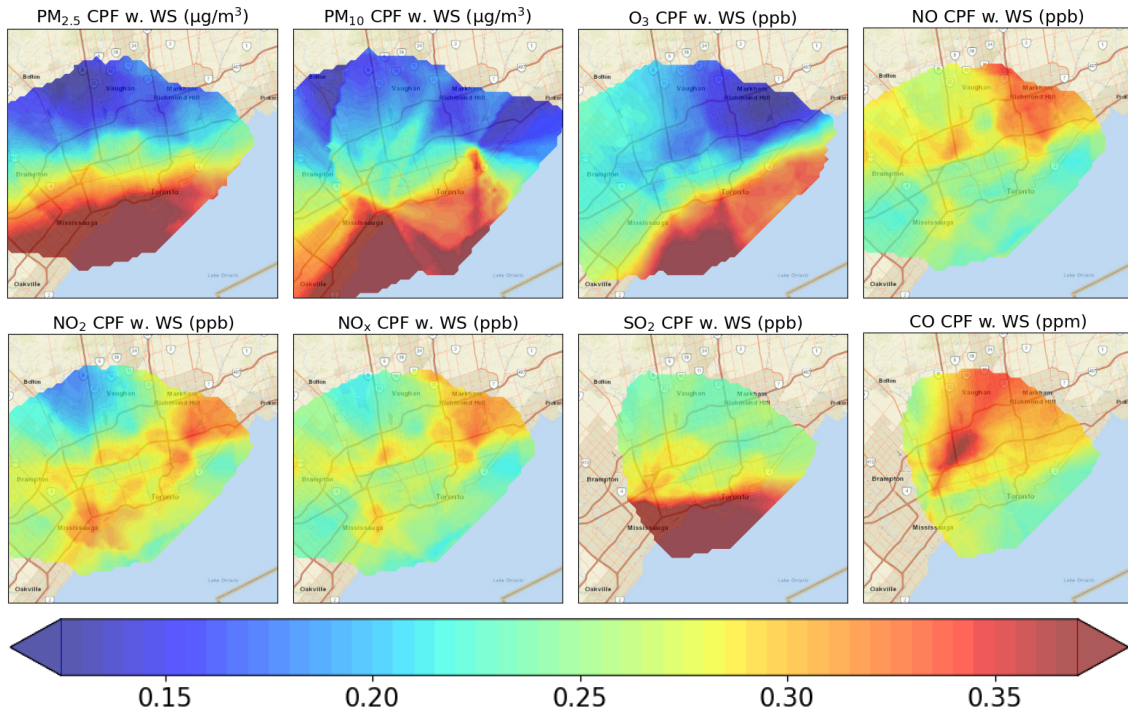


Figure 4.15: Like Figure 4.7 but regarding the second back-trajectory method. Despite no major differences present between the individual CO back-trajectories, for consistency and proper comparison with the other two CO averaged back-trajectories, the Toronto West and Centennial Park individual COs was back-tracked 3 hours in time, based on the wind speed in this figure.

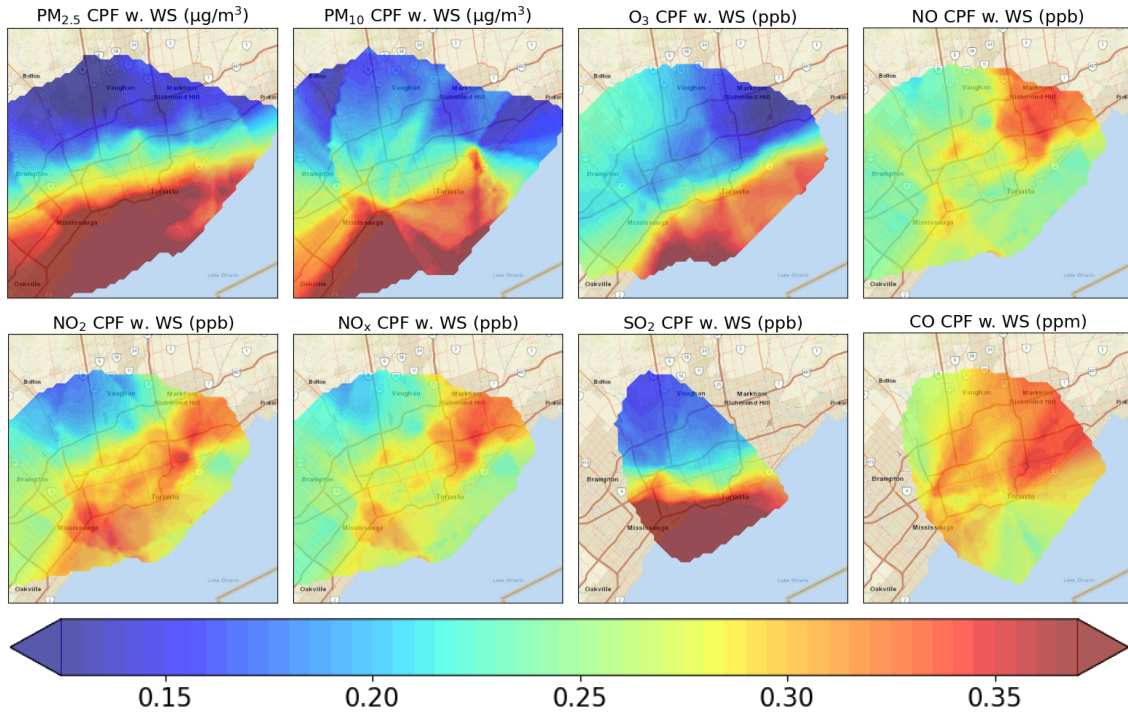


Figure 4.16: Like Figure 4.15, but regarding concentrations prior to 2013.

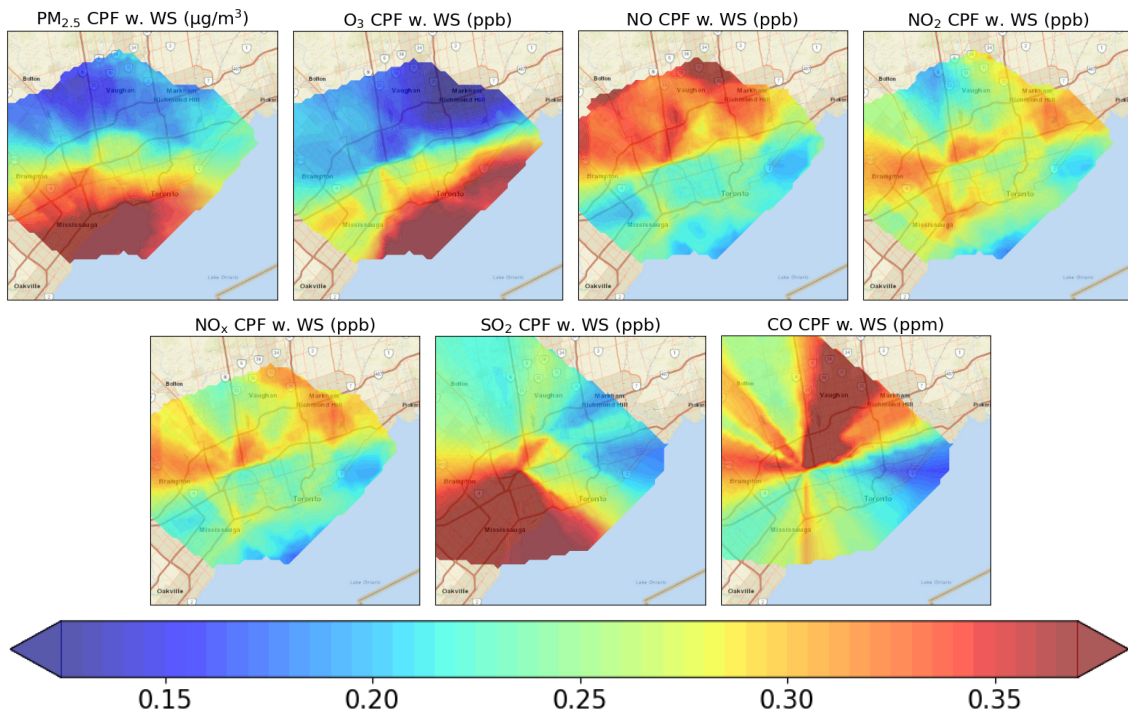


Figure 4.17: Like Figure 4.15, but regarding concentrations after 2013.

The socioeconomic mapping relationships for CPF with wind speed back-trajectory method (all years, Figure 4.15), is presented in Table 4.2.

Table 4.2: Like Table 4.1, but for the second back-trajectory method.

	PM _{2.5}	PM ₁₀	NO	NO ₂	NO _x	SO ₂	O ₃	CO
No. Affected	682835	714573	698645	703139	697073	691708	692607	692807
Pop. Density R,	0.16	0.17	-0.17	-0.05	-0.16	0.11	0.19	-0.19
SE/slope	0.26	0.25	0.24	0.79	0.25	0.37	0.22	0.19
Median PT Income R,	0.47	0.40	-0.46	-0.07	-0.37	0.33	0.42	-0.49
SE/slop	0.08	0.10	0.08	0.63	0.11	0.12	0.09	0.08

4.3 Discussion

4.3.1 Individual Back-Trajectories

In Figures A.1 through A.10, it is seen that the most variable back-trajectory maps are in cases such as the University of Toronto, with the smallest timeframe and least consistent average wind direction per 10° bin. However, since the averaged CWT is weighted by the number of concentrations per timeframe, it is given the least weight. Generally back-trajectories that are the least uniform influence the average back-trajectories the least and vice versa.

The CWT maps generated by the individual AQ stations (Figures A.1 through A.10), reinforce the same results from the polar plots in Figures 3.7, 3.9, and 3.8, with the primary sources of PM_{2.5}, PM₁₀, SO₂ and O₃ originating from the southwest and south-southeast, respectively. In almost every individual PM_{2.5}, PM₁₀ and SO₂ CWT, the greatest concentrations are in the south, indicating the main sources are from outside the city. The same was generally true for O₃, despite spatial variations present in the composite maps. For particulate matter, this is consistent with the findings from Brook et al. (2011) (PM_{2.5} concentrations being considerably greater with southern winds than northern winds), and Jeong et al. (2020). Long-range transport of SO₂ primarily from the south is in accord with the origins of secondary sulfate from the south during 2012 to 2014 by Zhou et al. (2019a). Indications of ozone also being transported from the south differs from the main secondary aerosol sources dominated by ozone, which were found to be from the west, and Sudbury Ontario by Zhou et al. (2019a) during the winter months. Furthermore, these elevated concentrations coming from the south may also be from the Portlands Energy Center power plant, as mentioned in Chapter 3. NO, NO₂, and NO_x also primarily originate from the southwest and northeast, and these similar directions of high concentrations suggest a common sources of these pollutants. This is expected given NO_x is the sum of NO₂ and NO.

In the individual back-trajectory maps, the highest NO, NO₂ and NO_x concentrations generally corresponds to the lowest O₃ concentrations and vice versa, consistent with the inverse relationship

in the trends and averages in Chapter 3. The highest concentrations of NO_2 are observed at the Toronto West and Highway 401 sites, particularly about the west and northeast. As stated by Transport and Environment. (n.d), airplane emissions could be a factor since these are located near Pearson and Downsview Airport, and west and northeast are the approximate directions to Pearson and Downsview Airport respectively from these stations. Furthermore, the York University power plant could also be a potential source from the NE direction. Coincidentally, the lowest back-trajectory values of ozone at the Highway 401 and Toronto West sites are also observed in these directions, reinforcing the inverse NO_2 to O_3 relationship. Elevated CO concentrations are seen at Toronto West from the northeast direction as well, suggesting the potential Downsview Airport NO_2 , as stated by Transport and Environment., n.d., or York University power plant source could also be emitting CO. It is important to note that the pollutant trends contribute to the relative differences between concentrations at the individual CWT location stations. This is especially seen in the CO case in Figure A.10, where the lowest and highest concentrations are present when the station temporal coverage is more and less recent respectively. This phenomenon will be further addressed and discussed in the next subsection (4.3.2).

Interpreting the individual CPF with wind speed, back-trajectory maps in Figures A.11 through A.19, in every location, one can clearly observe which directions the relative high-levels of sources originates from at each location, and how that compares to other locations, like in the CWT individual back-trajectories. Usually, displaying relative values between different locations can be problematic when creating overall spatial maps like in Figure 4.7, in cases like CO, where the individual concentrations vary wildly between station location. This can be referred to as a “relative value bias.” However, in this case, the trend-line bias present with the CWTs is also removed. This is also discussed further in the following section, 4.3.2

4.3.2 Averaged Back-Trajectories

The CPF maps present in Figure 4.15 are similar to the composite CWT superimposed pollutant maps in Figure 4.7. Examining Figure 4.7, the greatest concentrations of $\text{PM}_{2.5}$, PM_{10} , O_3 , and SO_2 appear to be outside the city, over the lake. This is generally a way to validate if major emitters are from beyond the city, by examining if every (or the vast majority of) individual back-trajectory exhibits its highest concentrations from one direction, regardless of station location. This is consistent with what was suggested from the results of the polar plots. Unlike the NO , NO_2 , NO_x , CO and O_3 CWTs, not much spatial variation is observed in the $\text{PM}_{2.5}$, PM_{10} , and SO_2 averaged CWTs throughout Toronto. Furthermore, elevated concentrations of $\text{PM}_{2.5}$, PM_{10} , and SO_2 in the Southwest in Figure 4.7, and 4.15 may be from the gas-fired Halton Hills Generating Station or the Ash Grove Mississauga Cement Plant.

It is known that vehicles are a large source of NO_x emissions (Air Quality Ontario, n.d.-b; Hagenbjörk, 2017; Environmental Protection Agency., 2023), primarily composed of NO (Hagenbjörk, 2017; EPA, 2023). In fact at least 90% of vehicular tail-pipe NO_x emissions are comprised of NO (EPA, 2023). In Figures 4.7, and 4.15. NO_2 appears to be tracing Highway 401 and the Don Valley Parkway. This can be explained by the fact that conversion of traffic NO emissions to NO_2 occurs very rapidly (within minutes). As already mentioned, this conversion is especially quick (under a minute), with enough ozone and high temperature conditions (EPA, 2023). Elevated concentrations of NO_2 and NO_x over Highway 401 also corroborates the result found of elevated NO_2 concentrations near highways by Jerrett et. al. (2007), as well as the semi-high patch of NO_2 over downtown Toronto. This is also somewhat observed over Don Valley Parkway, and Highway 407. Furthermore, in both back-trajectory cases, a semi-high patch of NO_2 concentrations is observed near downtown Toronto as well. In addition to elevated NO, NO_2 , NO_x concentrations observed in northeast Toronto, they are also seen around Pearson and Downsview airport, suggesting an aircraft source as well (like from the individual back-trajectories, and stated by Transport and Environment. (n.d.)). Moreover the hotspot around Downsview Airport is also located near the York University power plant. Neighborhoods near these locations are the most impacted by elevated NO, NO_2 and NO_x concentrations, although, high concentrations in NE Toronto are inconsistent with the findings by Jerrett et al. (2007). This inconsistency may partly be due to the fact that the study by Jerrett et al. (2007), was only conducted throughout a two week period. In Figures A.7, and A.17, the “Toronto East” individual NO_2 back-trajectory clearly shows the greatest concentrations from the North. Likewise, the “Toronto North” station (NW) of “Toronto East” unambiguously displays its greatest concentrations from the SE, indicating high concentrations in NE Toronto over their measurement period.

Interestingly, areas with high concentrations of NO, NO_2 and NO_x roughly corresponds to the same neighborhoods that are the least impacted by ozone. Around Highway 401, Pearson Airport and especially east Toronto is where the lowest ozone concentrations occur. Again, the inverse relationship is consistent with what the trends, polar plots, individual CWTs observed, and the general chemistry information outlined in the Introduction. Given the information by Geddes et al. (2009), Seinfeld and Pandis. (2016), Sillman. (n.d.), Whaley et al. (2015), and Zhang et al. (2022), the anti-correlations between O_3 and NO_x suggest a VOC-limited regime, consistent with literature in Toronto. There is also no indication that ozone is originating from west or northern Ontario as found by Zhou et al. (2019a), despite Figures 4.7, and 4.15 just showing spatial variations within Toronto. The only possibility of long-range transport is from the south, as seen with $\text{PM}_{2.5}$, PM_{10} and SO_2 , which is also consistent with a result from the Zhou et al. (2019a) study.

The superimposed back-trajectories in Figure 4.15 are similar to the results in Figures 4.7 confirming the average concentration is related to the frequency of exceedances. One exception is the stronger presence of elevated CO concentrations in Figure 4.15, throughout North Toronto, instead of being more localized like in Figure 4.7. In particular, this can be visualized from examining Figure 4.15, as the elevated and minimal CO concentrations are at similar locations, except for north Toronto to Vaughan/Markham and Highway 400, further reinforcing the commonality between the CO, NO, NO₂, and NO_x concentrations and sources. In Figure 4.10, that displays the zoomed-in picture of PM_{2.5}, semi-elevated concentrations can still be observed around Highway 400, Downsview Airport and the York Power Plant like for the CO, NO, NO₂, and NO_x back-trajectories in Figures 4.7, and 4.15.

Examining Figures 4.11, 4.12, it is evident that NO, NO₂, NO_x, CO and SO₂ CWTs, are higher and lower pre and post-2013, while the reverse is true for ozone. This is expected given the trends of these pollutants found in Chapter 3. Perhaps the Nanticoke power-plant emissions contributed these trends pre-2013, but the exact contribution is unclear. The exceptionally high concentrations of PM_{2.5} towards the south is wider pre-2013 within Toronto, which perhaps also indicates an emissions contributions from this source. It is important to note that the SO₂, and CO post-2013 back trajectories are less reliable since only three AQ stations were employed. As mentioned previously, in Figures 4.7, and 4.15, the superimposed back-trajectories for SO₂ and CO was comprised of 9, and 10 stations respectively. PM₁₀ remains the same in all cases since of all its measurements from the AQ stations end before 2013, as such PM₁₀ is not shown in the post-2013 Figures (4.12, and 4.17). To quantify the potential Nanticoke emissions source from the CPF with wind speed back-trajectory method, the goal is to observe if a greater abundance of high pollutant values are present in the south in the pre-2013 concentrations. In Figure 4.16 (pre-2013), this is seen for NO₂ and SO₂. Likewise, in Figure 4.17, lower relative concentrations are present towards the south in NO₂, NO_x, and CO, indicating potential signs of an emitter discontinuing. However as mentioned above, validity of the CO back-trajectories post-2013 is less reliable.

Hsu et al. (2003), also noted that employing many sites for a CWT results in limitations when some sites consistently record low concentrations. In this case, since there are notable trends in many of these pollutants, and the superimposed CWT map in Figure 4.7 is comprised of different sites with varying timeframes, a trend-line bias occurs, and this limitation is present. As mentioned previously, the second back-trajectory method uses relative values, thus removing this trend-line bias, and the limitation. This method essentially provides information as to which directions exhibit high and low probable sources for each site, allowing for triangulation of probable sources between

the individual maps, so that a spatial map to be produced. Interestingly, the CWT and CPF with wind speed back-trajectory pollutant variations are very similar, thus the CWT results are supported by the CPF with wind speed back-trajectory results and vice versa.

4.3.3 Socioeconomic Mapping

Given that individual $PM_{2.5}$, PM_{10} and SO_2 CWTs predominately show higher concentrations from the south, which manifests in a southward concentration gradient in the superimposed CWTs in (Figure 4.7), the positive relationship between these pollutants and income is expected, since the greatest income neighborhoods tend to be in the south (except for the DVP/bridal path areas) as seen in Figure 4.6. The same is also true for population density, but to a lesser extent. However, since the individual $PM_{2.5}$, PM_{10} , and SO_2 CWTs all indicate that predominately high concentrations are coming from the south, their averaged back-trajectory maps in Figure 4.7 and 4.15 simply reinforces this fact, instead of displaying additional spatial variations. Therefore these relationships should be deemed inconsequential, since the spatial variations in $PM_{2.5}$, PM_{10} , SO_2 cannot be denoted using these back-trajectory methods, despite their low SE/slopes.

A negative relationship between income with NO , and NO_x is also present. In Figure 4.7 and 4.15, high concentrations are located near Highway 401, with an anomalously high value near Highway 400, Downsview Airport and the York University power plant. In this particular area around Highway 401, 400, Downsview Airport and the York Power Plant, relatively low-income neighborhoods are present. Moreover, in the east of Toronto near Markham, another anomalous high NO , NO_2 , and NO_x region on the CWT map exists, which coincides with relatively low-income areas as well. In the NO and NO_x instances, low averaged concentrations are present to the south of Highway 401 and the Downtown and Southern Toronto areas; where plenty of high-income neighbors are present, further strengthening the negative relationship. However for NO_2 , the negative relationship is very small. This can be attributed to high NO_2 concentrations present in the downtown areas, and Etobicoke, coinciding with some high income neighborhoods. Furthermore, some low NO_2 concentrations are present in north Toronto where low income neighborhoods are present as well.

Likewise, the highest and lowest concentrations of CO are located in north (especially in the area near Downsview Airport/Highway 400/York University Power Plant) and south Toronto respectively, where the lowest and highest income neighborhoods also tend to reside, thus explaining the strongest downward relationship between CO and net median income. The positive relationship between income and ozone CWTs can also be explained by the fact that there is an inverse relationship between the ozone and the NO_2 , NO_x , and NO CWTs. Furthermore, given the spa-

tial variations present in the NO, NO₂, NO_x, O₃, and CO CWT case, and the low corresponding SE/slope for their relationship with income, it can be assumed that these relationships are significant. Interestingly, despite linear fits being carried out, in Figure 4.13, there is a non-linear relationship with the binned 95th percentiles all the positive relationships (PM_{2.5}, O₃ and SO₂), peaking at a net median income of around \$40,000. This is most likely due to a smaller sample size for each increase in income bin, narrowing the bins concentration distribution, and thus decreasing the 95th percentile value.

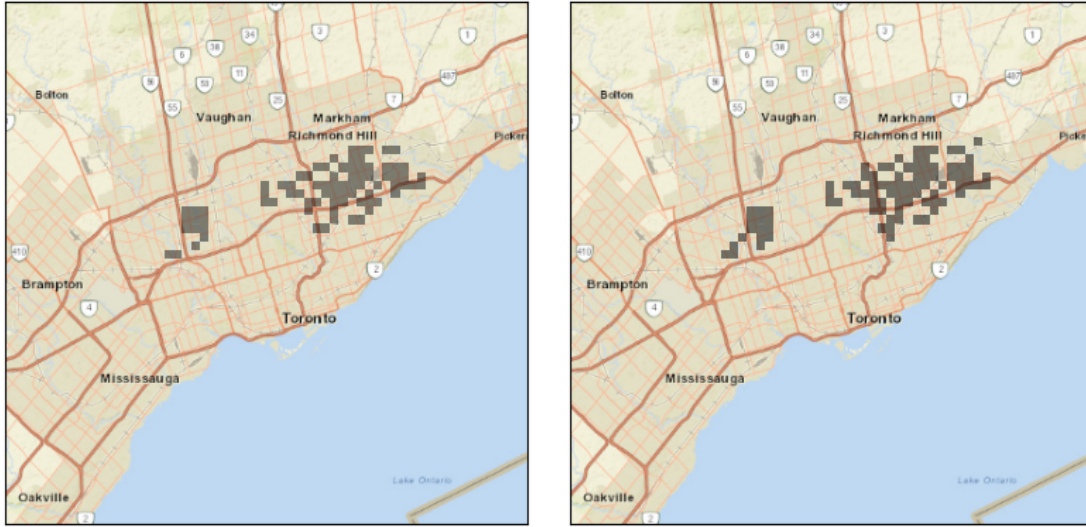


Figure 4.18: Low income neighborhoods exhibiting relatively high concentrations of NO_x and low concentrations of ozone (left), and low concentrations of PM_{2.5} (right) from Figure 4.7.

In Figure 4.13, it can be seen that there is a grouping of grids that exhibit low income with both high O₃ and low NO_x and PM_{2.5} concentrations. Figure 4.18 (left) displays the intersection of points in Figure 4.7, but on a pixelated map with relatively low income (< \$37,000), exhibiting NO_x, and O₃ CWT values of > 31 and < 18 ppb respectively. Figure 4.18 displays the same, but for low PM_{2.5} concentrations (< 7.25 µg/m³).

As can be observed, low income districts on/near the eastern portion of Highway 401, and over Downsview Airport/near Highway 400/York University power plant simultaneously contain relatively low O₃ and PM_{2.5} concentrations and high NO_x concentrations. Despite previously concluded that there is no spatial variation for PM_{2.5}, it is interesting that the neighborhoods are very similar in both maps, perhaps indicating there is some significance in the PM_{2.5} superimposed spatial maps, especially in north Toronto. This indicates there is also a negative relationship between O₃ and PM_{2.5} in the lower-income neighborhoods of Toronto. The similarity of low concentrations between O₃ (anti-correlated with NO_x) and PM_{2.5} in Figure 4.7 compliments this result.

The same conclusions can be drawn for the relationship between population density and CO as for income and CO, but to a lesser extent, with less sparsely populated neighborhoods located in the north-northwest of Toronto compared to low income neighborhoods. This explains the weak negative relationship between CO and population density. The relationships between population density and averaged CWTs are weaker in magnitude. High population density areas are also present in the southern/Downtown areas, coinciding with high values of $PM_{2.5}$, SO_2 , and O_3 . There are also low population density regions to the North of Highway 401 near Highway 400, Downsview Airport, York University power plant, and in the East of Toronto near Markham, which coincides with high values of NO, NO_2 and NO_x , and low values of ozone. The highest population density regions are generally present in the downtown areas as well, which correspond to the lowest values of NO, NO_x and CO. The similar relationships between NO, and NO_x with CO is expected, given the similarities in their averaged CWTs. Despite the fact that these relationships with population density can be explained, the low R magnitudes and greater corresponding SE/slope, indicate these relationships are considerably less significant than their relationships with income.

It is important to note that the “No. Affected” (number of people affected, as defined in the Methodology section), are similar for each pollutant species. This is because relative concentration thresholds are being set, and the proportion of neighborhoods and people that observe values greater than or equal to this threshold is similar for each pollutant. Overall, the relationships and their corresponding standard error to slope ratios, are nearly identical both back-trajectory methods, which is expected given the similarities between all these back-trajectories. In particular, the similarity between correlation coefficient, standard error/slope and spatial variations for CO, NO and NO_x demonstrates the results are independent of the back-trajectory methodologies employed.

5 THE CIX and SWAPIT Campaign Back-Trajectory Analysis

THE CIX (Toronto Halogens, Emissions, Contaminants, and Inorganics eXperiment) and SWAPIT (Study of Wintertime Air Pollution in Toronto), campaigns were run during the summer of 2023 and winter of 2024 respectively. THE CIX campaign was carried-out at York University, connected to the Atmospheric Emissions and Reactions Observed from Megacities to Marine Areas (AEROMMA), studying oceanic and anthropogenic pollutant sources (NOAA Chemical Sciences Laboratory, n.d.), managed by NOAA (Young, n.d.). This project aimed to explore unknown or ambiguous characteristics relating to air quality in urban locations (ibid.). The SWAPIT (Study of Wintertime Air Pollution in Toronto), was organized by Environment and Climate Change Canada (Government of Canada, 2024). The winter season was chosen to quantify pollutants from road de-icing, heating indoors, planes, and cooler temperatures (ibid.). As with one of the major goals of this thesis, the SWAPIT organizers recognized that some neighborhoods throughout Toronto exhibit greater pollutant levels than others (ibid.). As such, an objective of this campaign was to understand pollutant trends in various neighborhoods, so they can be offset (ibid.). The SWAPIT organizers have also recognized that the rate of pollutant concentration decline is slower than anticipated (ibid.). Like the results in Chapter 3, it has been documented that pollutants have risen in certain instances (ibid.). These short term measurements taken by our group at York are presented here as a comparison with long-term station measurements from third parties (i.e., Air Quality Ontario, NAPS, ECCC) in Chapters 3, and 4. In this chapter, polar plots displaying the average and 90th percentile pollutant concentration by 10° wind direction were plotted, like in Chapter 3.

5.1 Methodology

5.1.1 DustTraks

PM₁, PM_{2.5}, PM₄, PM₁₀, and Total PM measurements were taken from a DustTrak (DRX Aerosol Monitor 8533 TSI inc.) instrument at two-minute intervals in both campaigns. Two Dust-Trak instruments collected particulate matter measurements during THE CIX campaign, denoted as DRX1 and DRX2. Both measurements were from 2023-06-28 to 2023-08-15. These instruments were located at York University’s Petrie Science and Engineering (PSE) building, through their specific locations varied. The DRX1 instrument was located on the 4th floor balcony of the PSE building throughout the whole campaign. Despite the DRX2 instrument being initially located on the 4th floor balcony as well, it was moved to the 6th floor “rooftop” of the PSE building on July 26, 2023 at approximately 10:55 a.m. This 6th floor “rooftop” is approximately a 3 × 3 meter roof

a room located on the larger 5th floor. The purpose of moving one instrument to the rooftop was to deduce if there was a considerable difference between the measurements at different heights. In this campaign, all times reported and displayed on graphs were adjusted for daylight savings times.

The DustTrak PM measurements were nearly identical for all sizes, indicating instrumental flaw. PM₁₀ for example should report noticeably higher concentrations than PM₁, though this wasn't the case. Throughout this chapter only DustTrak polar plots for PM₁ will be displayed since the DRX2 instrument was calibrated against another reliable UHSAS (Ultra High Sensitivity Aerosol Spectrometer) instrument that measures particle mass concentrations < 1 μ m (more details to be explained later in this section). The DustTrak instruments collected measurements at two minute intervals from its starting point. Measurements were stored internally, and had to be collected via USB. Since the specific measurement starting points, and thus times differed between both instruments, 10 minute averages were taken to ensure consistency of the data, for comparison. Other reasons for taking the time-averages were to remove potential noise, and to align the concentrations to the 5-minute reported wind speed and directions. The DustTrak plots from THE CIX campaign are displayed in Figure 5.2.

The raw DustTrak time-series for THE CIX campaign is displayed in Figure 5.1.

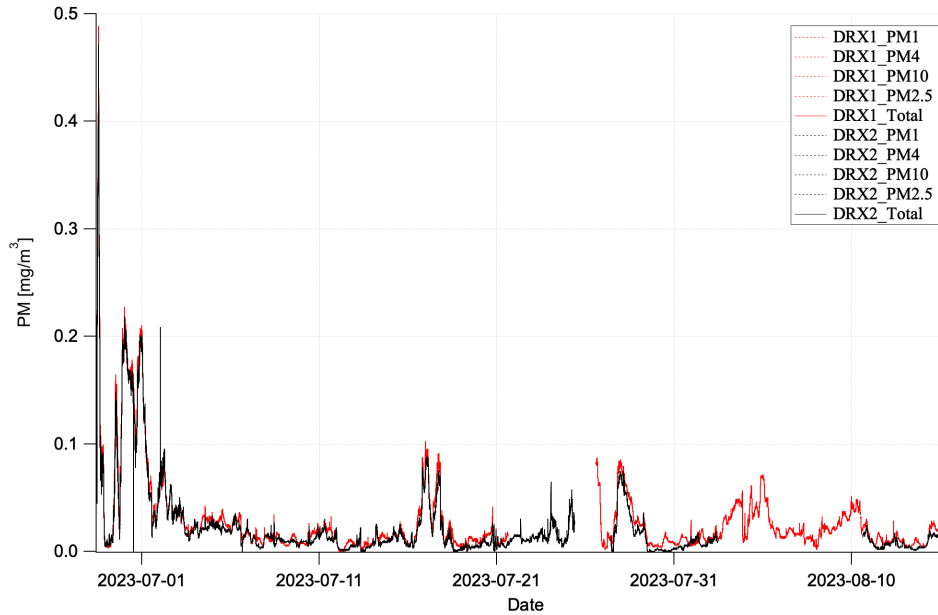


Figure 5.1: DustTrak time-series from the raw two-minute data, during the CIX campaign, DRX1 and DRX2 measurements from 2023-06-28 to 2023-08-15.

It can be seen that there is a slight discrepancy between DRX1 and DRX2. Thus one instrument needs to be cross-compared to the other. To ensure proper comparison between the DRX1 and

DRX2 back trajectories, the DRX1 instrument was cross-compared to the DRX2 instrument when both instruments were side by side and recording on the 4th floor of the PSE building. Moreover, the comparisons were carried-out based on the 10-minute averages, for the reasons as listed above. This cross-comparison period ranged from 2023-06-28 to 2023-07-21. The equations are shown below;

$$D1PM_{1adj} = 0.9730(D1PM_{1raw}) \mu g m^{-3} - 2.6 \mu g m^{-3} \quad (5.1)$$

$$D1PM_{2.5adj} = 0.9717(D1PM_{2.5raw}) \mu g m^{-3} - 2.6 \mu g m^{-3} \quad (5.2)$$

$$D1PM_{4adj} = 0.9716(D1PM_{4raw}) \mu g m^{-3} - 2.6 \mu g m^{-3} \quad (5.3)$$

$$D1PM_{10adj} = 0.9713(D1PM_{10raw}) \mu g m^{-3} - 2.6 \mu g m^{-3} \quad (5.4)$$

$$D1PM_{Tadj} = 0.9710(D1PM_{Traw}) \mu g m^{-3} - 2.6 \mu g m^{-3} \quad (5.5)$$

R^2 for all of the equations above is 0.992. Negative values (due to low readings and instrument noise), were removed in the cross-compared 10-minute averages.

Corresponding 5-minute averaged wind speed and wind directions were gathered from the EMOS (Earth and Space Science Meteorological Observation Station at York University, (Gordon et al., 2024) to generate the concentration polar plots for the CIX campaign. The anemometer is an R&M Young brand, situated atop a 10 meter pole (ibid.). Data from the anemometer is collected every second (ibid.). In addition to wind speed and direction, the EMOS station also measures temperature, relative humidity, precipitation and radiation (ibid.). Raw measurements are collected via the Campbell Scientific PC208W software, which is then processed by FORTRAN, producing daily data logs in UTC (ibid.). The instrument used for logging measurements and calculating 5-minute averages is powered by a solar panel, 2.5 meters above ground (ibid.). The five-minute averaged wind speed and direction were taken at every second point, to yield wind speed and directions of 10-minute temporal resolution. This ensures that the arrays of $PM_{2.5}$, wind speed and wind direction were of equal length to produce polar plots.

In the SWAPIT campaign, two DustTrak instruments were deployed at two different sites. The first DustTrak instrument, referred to as “DRX1” was located on the PSE building rooftop. The second instrument, referred to as “DRX2” was located on the 4747 Keele Street building rooftop. To generate the polar plots, corresponding winds for DRX1 were taken from the EMOS site, and

an ultrasonic ATI (Applied Technologies Inc.) 'A' style probe anemometer located at 4747 Keele Street for DRX2.

In the SWAPIT campaign, 10-minute particle measurement averages were still employed; however, given that 30-minute averages were taken from the SWAPIT ATI ultrasonic anemometer located on 4747 Keele Street, these polar plots used PM data every 30 minutes from the 10-minute averages (taken every third point) to keep the arrays of concentrations, wind speeds and wind directions equal. The DRX2 instrument was located on the 4747 Keele Street rooftop; hence, the ATI (Applied Technologies Inc.) winds were the corresponding winds.

The DRX1 back-trajectories employed the EMOS winds; despite the reported winds from this station being 5-minute averages (as mentioned previously), the wind data was taken every 30 minutes (or sixth point) to ensure an equal comparison between DRX1 and DRX2 back-trajectories. The timeframe of these generated polar plots are from 2024-01-29 to the end of 2024-03-10. Before the SWAPIT campaign, the two DustTraks instruments were placed side by side on the 5th floor (on the rooftop) of the PSE building for cross-comparisons. As with THE CIX campaign, these cross-comparisons were carried out based on 10-minute averages, from 2024-01-24 to 2024-01-29.

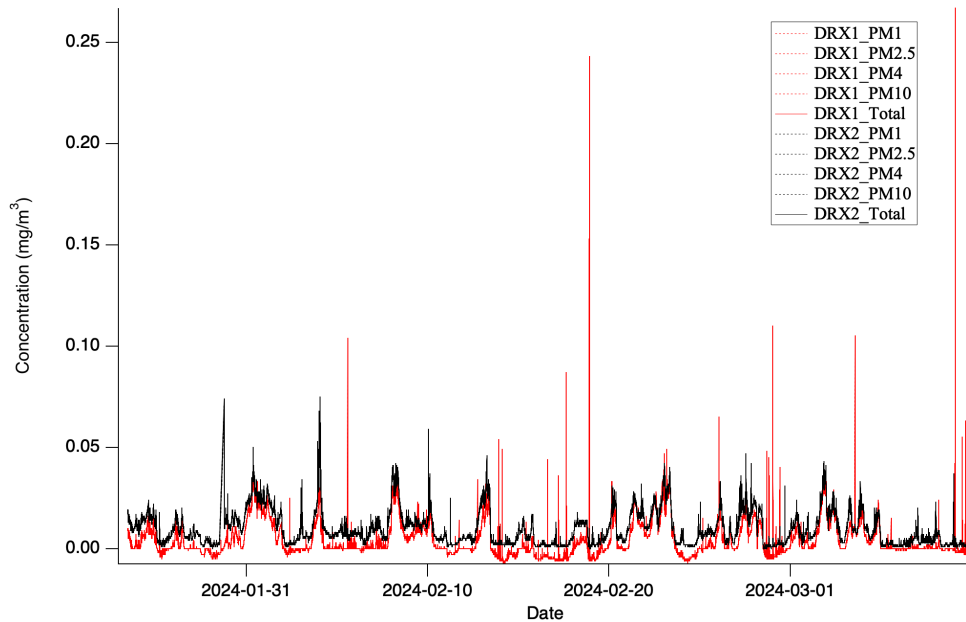


Figure 5.2: DRX1 and DRX2 raw PM measurements from 2024-01-24 to the end of 2024-03-10. Note the times on March 10 were not adjusted for daylight savings.

In Figure 5.2, the raw (uncorrected), 2 minute averages of DRX1 and DRX2 measurements are shown for comparison, which emphasizes the importance of cross-comparing these instruments

to one-another. DRX1 consistently measures lower values than DRX2, including when they were placed side by side from 2024-01-24 to 2024-01-29. Furthermore, the presence of erroneous negative values in the DRX1 measurement, implies a negative offset in the instrument, further emphasizing the need for cross comparisons/recalibrations. From examining the calibration data in Figure 5.3, from the DustTrak instruments, one can observe different offsets for positive and negative values. As such, the positive and negative cases were handled differently.

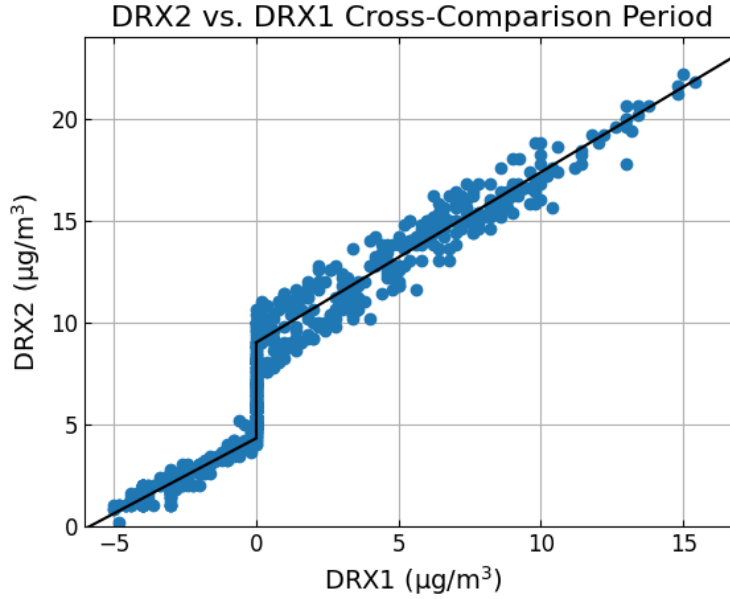


Figure 5.3: Unchanged DRX1 PM₁ vs. DRX2 PM₁ for the SWAPIT campaign. Lines of best fit for DRX1 < 0 and > 0 are displayed in black.

The calibration equations for DRX1 to DRX2 in the SWAPIT campaign are as follows;

$$D1PM_{1adj.} = \begin{cases} 0.7398(D1PM_{1raw})\mu g m^{-3} + 4.3\mu g m^{-3} & \text{if } D1PM_{1raw} < 0; \\ 0.8370(D1PM_{1raw})\mu g m^{-3} + 9.0\mu g m^{-3} & \text{if } D1PM_{1raw} > 0. \end{cases} \quad (5.6)$$

$$D1PM_{2.5adj.} = \begin{cases} 0.7406(D1PM_{2.5raw})\mu g m^{-3} + 4.3\mu g m^{-3} & \text{if } D1PM_{2.5raw} < 0; \\ 0.8368(D1PM_{2.5raw})\mu g m^{-3} + 9.0\mu g m^{-3} & \text{if } D1PM_{2.5raw} > 0. \end{cases} \quad (5.7)$$

$$D1PM_{4adj.} = \begin{cases} 0.7423(D1PM_{4raw})\mu g m^{-3} + 4.3\mu g m^{-3} & \text{if } D1PM_{4raw} < 0; \\ 0.8370(D1PM_{4raw})\mu g m^{-3} + 9.0\mu g m^{-3} & \text{if } D1PM_{4raw} > 0. \end{cases} \quad (5.8)$$

$$D1PM_{10adj.} = \begin{cases} 0.7425(D1PM_{10raw})\mu g m^{-3} + 4.3\mu g m^{-3} & \text{if } D1PM_{10raw} < 0; \\ 0.8374(D1PM_{10raw})\mu g m^{-3} + 9.0\mu g m^{-3} & \text{if } D1PM_{10raw} > 0. \end{cases} \quad (5.9)$$

$$D1PM_{Tadj.} = \begin{cases} 0.7419(D1PM_{Traw})\mu g m^{-3} + 4.3\mu g m^{-3} & \text{if } D1PM_{Traw} < 0; \\ 0.8368(D1PM_{Traw})\mu g m^{-3} + 9.0\mu g m^{-3} & \text{if } D1PM_{Traw} > 0. \end{cases} \quad (5.10)$$

where, again, D2 and D1 denote DRX2 and DRX1 respectively. Since the y-intercepts were all the same, when DRX1 equaled 0, it was offset by 0.0065 (the average of 0.0043 and 0.0090) in all cases. R^2 for all equations was 0.86 and 0.91 in the greater than and less than 0 cases respectively.

A UHSAS (Ultra High Sensitivity Aerosol Spectrometer) measured 30-minute averaged total particle measurements $\leq 1 \mu m$, atop the 4747 Keele Street building during the SWAPIT campaign. This instrument measures particle number and mass for sizes in several bins ranging from 60nm to $1 \mu m$. Since this measurement was essentially “side-by side” to the DRX2 instrument, these measurements can be used to calibrate both the DRX2 and DRX1 instrument. The UHSAS was calibrated and assessed by ECCC, so it is considered as an absolute standard for calibration. For this calibration, 30-minute averaged DRX2 PM_{10} pollutant concentrations were taken. The period of calibration ranged from January 29, 2024 to March 14, 2024 local time, (not adjusted for daylight savings post March 9). The relationship between DRX2 PM_{10} and the UHSAS concentrations color-coded by date are displayed in Figure 5.4.

The color coding in Figure 5.4 contrasts from dark blue (earliest date and time), to red (latest date and time). There are some outlier points in this relationship, and they were within the polygon region shown, outlined in black on the graph. As can be observed, these outlier points are predominately earlier in the campaign. Furthermore, wind speed and direction analysis was applied to gauge if these particular pollutant measurements were potentially originating from different areas. The calibration factor was computed by taking the reciprocal over the slope of best fit ($1/4.28$), for all the campaign points across the entire date range. This factor was applied to all of the Dust-Trak measurements across all sizes, after the cross-comparisons were applied, in both the CIX and SWAPIT campaigns. We chose to apply the calibration, that encompasses all the points, accounting for a full range of potential sources, based on the assumption that the outlier points are due to sampling of different aerosol compositions. In addition, the raw measurements were multiplied by 1000 to convert from mg/m^3 to $\mu g/m^3$.

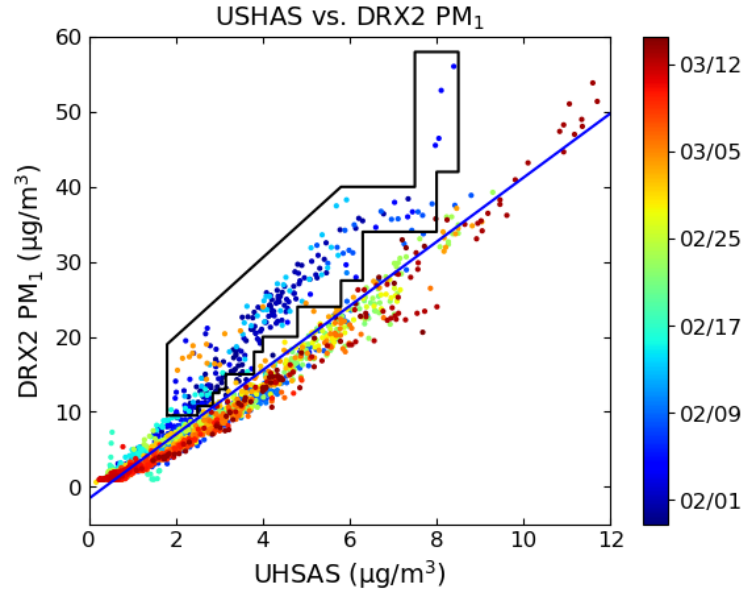


Figure 5.4: UHSAS and DRX1 PM_{10} ($\mu\text{g}/\text{m}^3$) linear relationship, colored by date from the start to end period. Colorbar date format is MM/DD.

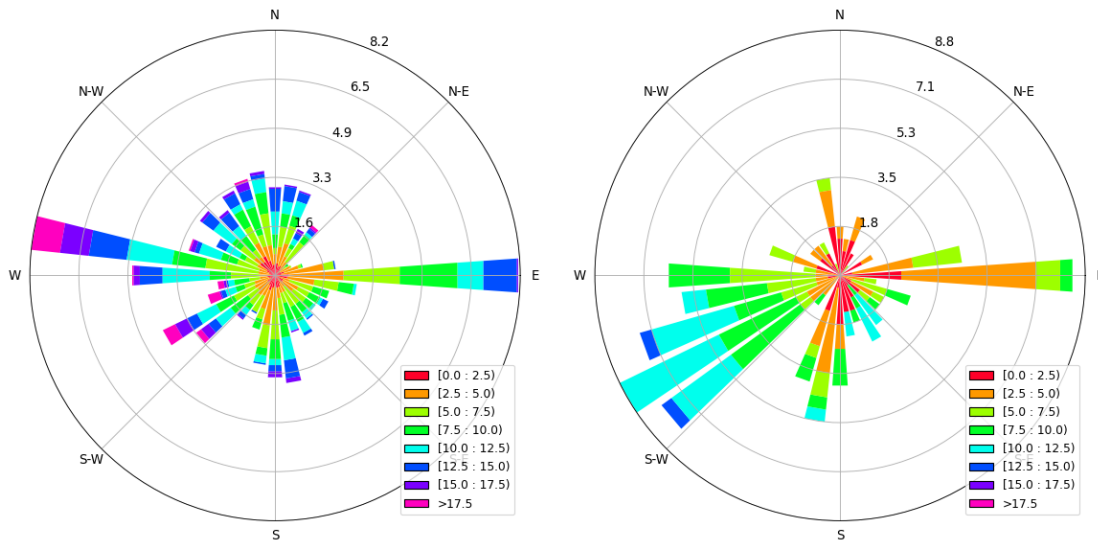


Figure 5.5: Distribution of all wind directions for the calibration period (left) vs. wind distribution for the selected “outlier” points (right).

Figure 5.5 shows wind roses for the entire calibration period and the outlier points on the graph. As can be seen, the outlier points contain lower wind speeds, and greater distribution of winds from the southwest. These points may represent more local sources, and predominately from the southwest directions, but the reason for this difference is not entirely clear.

5.1.2 Ozone Instruments

The 2B Technologies ozone instruments (model 205 Dual Beam Ozone Monitor) measure ozone (in ppb), collected data every 1 to 3 seconds. Instrument data was stored on an SD card, and to check data quality, the measurements were taken and plotted periodically. Once the SD card was removed, the measurements were paused, and started at a different time when the SD card was inserted again. 5-minute averaged ozone measurements were computed. Values with missing timestamps were not considered when making the timestamp averages. To remove these errors, it was found the following filters needed to be applied. Due to instrument error, some lines in the data output file were incomplete or contain random characters (which were not used). Before taking the five-minute averages, six conditions were to filter outlier ozone measurements were applied. They were listed as follows:

1. Reported data < 0 ppb or > 100 ppb
2. Internal instrument temperature $< 0^{\circ}\text{C}$ or $> 100^{\circ}\text{C}$
3. Internal instrument pressure $< 500\text{hpa}$ or $> 1000\text{hpa}$
4. Internal flow rate $< 10\text{ cm}^3/\text{min}$ or $> 3000\text{ cm}^3/\text{min}$
5. At least one of instrument temperature, pressure and flow rate missing
6. Non-successive reported date and times

During summer 2023 THE CIX campaign, there were three ozone instruments, labelled 2B1, 2B2 and 2B3. Timeframes for 2B1, and 2B2 were from June 30 to August 14, and 2B3 from June 30 to July 21. The 2B1 instrument was moved from the 4th floor to the rooftop on 2023-07-26 around 10:55 am (to potentially quantify an ~8 meter vertical concentration gradient, as with the DustTrak instruments), but was moved back down to the 4th floor on 2023-08-08 around 11:40 am. The 2B2 measurements were not recorded from July 6 to July 18, 2023. The 2B2 instrument contained sporadic concentration data. This instrument malfunctioned from July 26, 2023, to August 11, 2023, meaning the ozone concentration height gradient could not be determined. The 2B3 instrument stopped working on July 21, 2023. These malfunctions can be seen in section 5.2.1.2. Since the 2B1 concentrations were the most consistent and least noisy, 2B2 and 2B3 were chosen to cross-compare against it.

Cross-comparisons were carried-out based on the five minute ozone averages from 2023-06-30, 10:15 to 2023-07-21, 12:40. The correction equations, derived from a least squares fit are:

$$2B2_{adj} = \frac{2B2_{raw} + 0.5171}{0.9423} \quad (5.11)$$

$$2B3_{adj} = \frac{2B3_{raw} - 0.0936}{0.9896} \quad (5.12)$$

where 2B2 and 2B3 were calibrated to 2B1. R^2 for equations 5.11 and 5.12 were 0.9497 and 0.9946 respectively. An example calibration between 2B1 and 2B2 is displayed in Figure 5.6. Ozone time-series for the SWAPIT campaign was also 5-minute averaged and ranged from Jan. 29, 2024 to the end of Mar. 8, 2024.

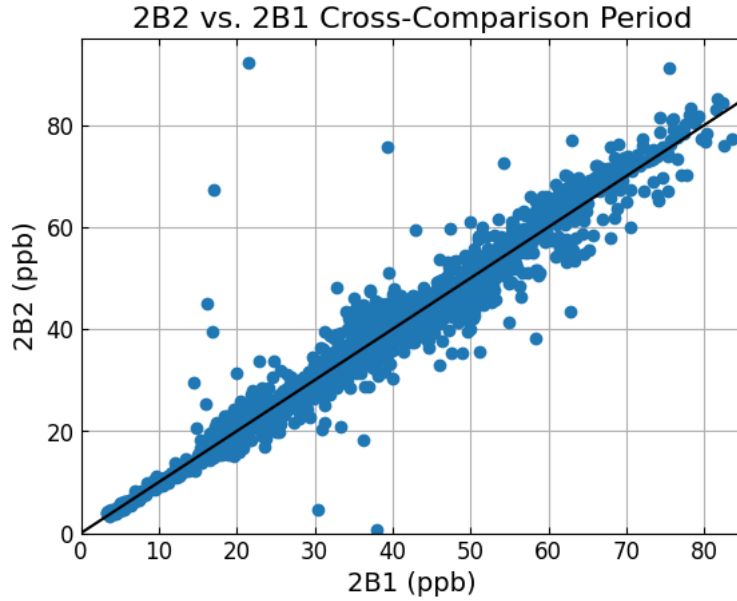


Figure 5.6: 2B1 vs. 2B2 (left) from the SWAPIT campaign, before time averages were taken. Line of best fit is overlayed in black.

In the SWAPIT campaign, only one ozone instrument (2B1) was functioning. Data was collected on the PSE rooftop from 2024-01-31 to the end of 2024-03-08, and averaged every 5 minutes.

5.1.3 Sulfur Dioxide

The SO_2 measurements were gathered via an Envea Emission instrument (type MIR9000e NDIR-GFC Multi-Gas Analyzer), recorded every minute, but averaged every 5 minutes. This will be referred to as “Envea” for short. The EMOS instrument was utilized for the wind information, every 5 minutes, for both campaigns, as in both campaigns, this instrument was located on the PSE rooftop. As with the PM and ozone data, the outlier SO_2 measurements were identified. Outlier

SO₂ measurements were identified if the 'stat0' parameter $\neq 100$ and the concentration was greater than -5. This value was chosen instead of 0 since the reported SO₂ data underwent a linear drift throughout the day, but the instrument performed a zero calibration at midnight each day, which needed to be corrected for. The drift was denoted as;

$$D[i] = SO_2[i] + t(s_0 - s_1) \quad (5.13)$$

Where $SO_2[i]$ is the i^{th} error-removed SO₂ measurement, t is a linear parameter $\in [0, 1)$ dependent on the time of day past midnight (i.e., $t = 0$ at midnight, and $t = 1$ at midnight the following day). The variables s_0 and s_1 denote the first and last error-removed SO₂ measurement of the day, respectively. Despite negative values present in the initial SO₂ measurements, these weren't removed until after drift correction. The SO₂ measurements shown for THE CIX campaign, pre and post-drift correction in Figure 5.7. In THE CIX campaign, the SO₂ data was gathered from a two-week period during July 29, 2023 to August 14, 2023. The SO₂ data for the SWAPIT back-trajectory analysis was taken every minute from January 29, 2024 to March 10, 2024, and the time series is shown in Figure 5.8. The times are in local standard time, and not modified for daylight savings time on March 10.

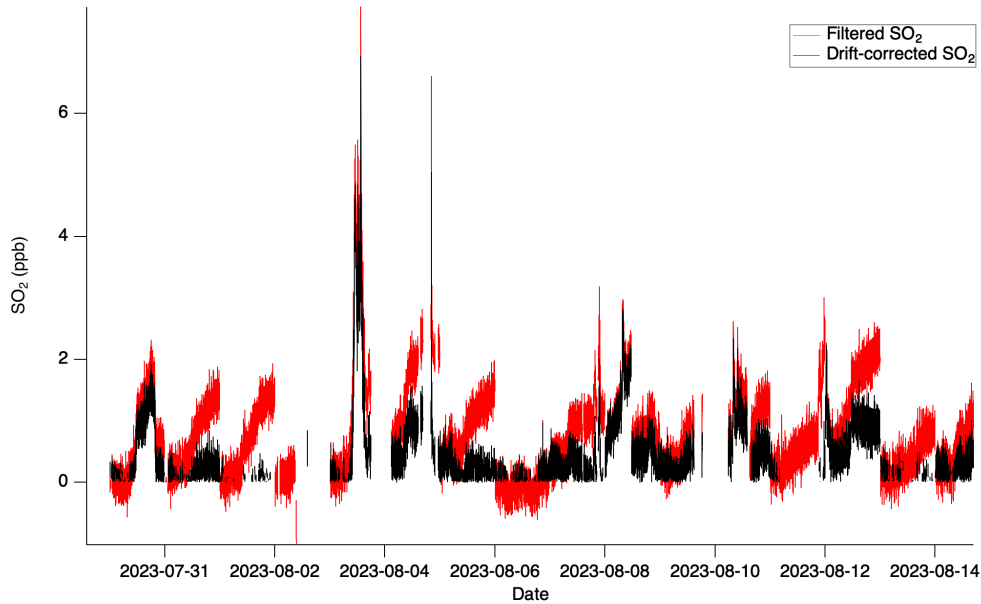


Figure 5.7: CIX campaign SO₂ before (red) and after (black) drift correction. SO₂ was measured from 2023-07-29 to 2023-08-14 taken every minute. All of August 2 is removed since there are no values close to midnight to carry-out the drift correction.

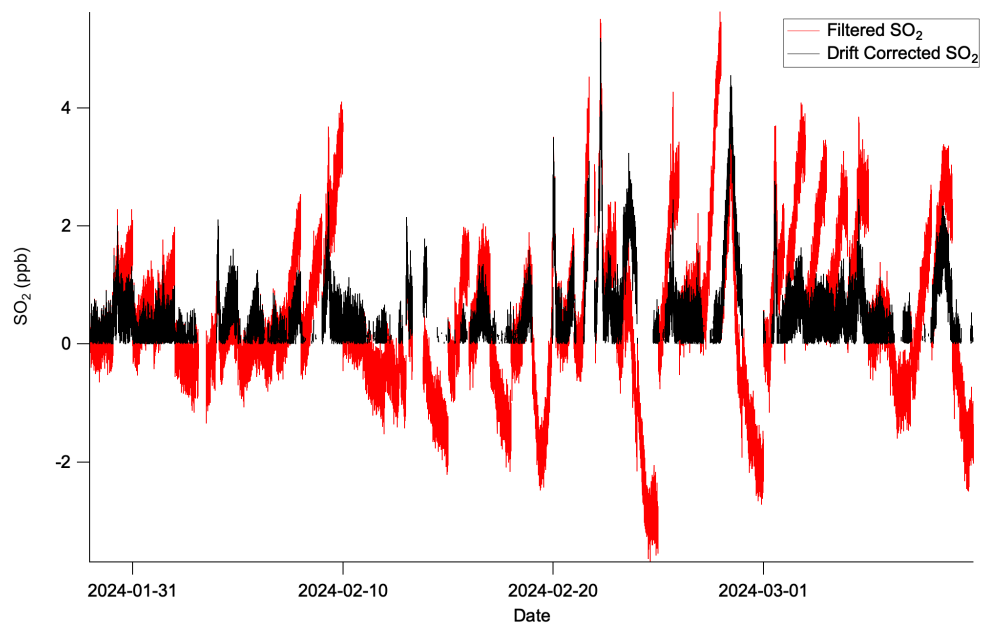


Figure 5.8: SWAPIT campaign filtered (red) and drift-corrected SO₂ (black), ranging from the beginning of 2024-01-29 to the end of 2024-03-10.

5.2 Results and Discussion

5.2.1 THE CIX Campaign

5.2.1.1 DustTrak Instrument

The calibrated 10-minute averaged DRX1 and DRX2 PM₁ plot is displayed in Figure 5.9.

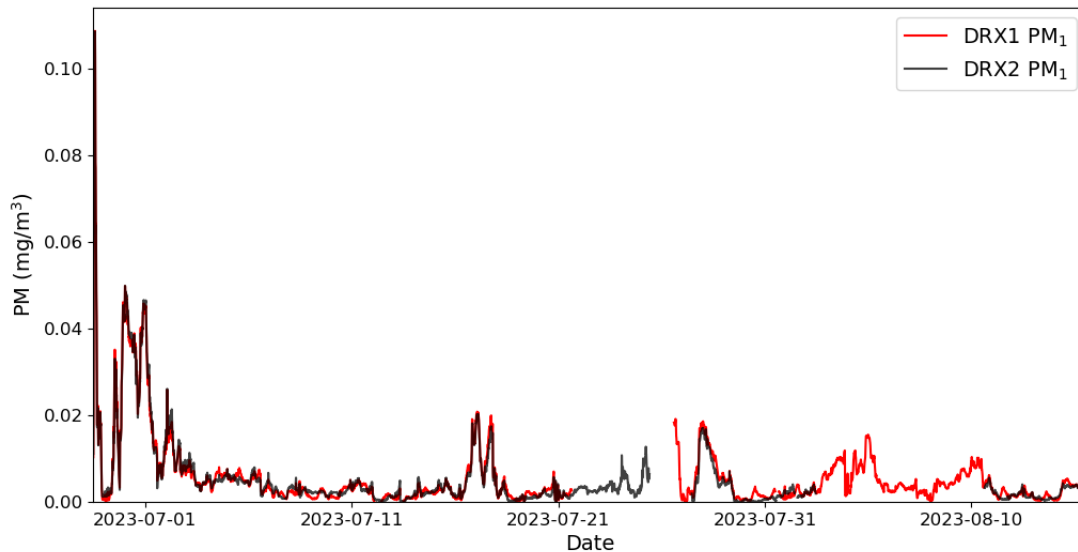


Figure 5.9: DRX1 and DRX2 PM₁ 10-minute averaged, after DRX1 was cross compared to DRX2

The uncalibrated DRX plot in Figure 5.1 showed some offset between the two instruments during the calibration period, which is not present in Figure 5.2. Plotted in Figure 5.10, are polar plots displaying average and 90th percentile concentrations per 10° wind direction for DRX1 and DRX2 PM₁. The PM_{2.5}, PM₄, PM₁₀ and Total PM polar plots are the exact same. Note the concentration spike of concentrations in Figure 5.2, around 2023-06-28. Measurements began when there was plenty of wildfire smoke in the atmosphere. Wildfire smoke continued intermittently throughout the campaign, resulting in higher concentrations than usual.

In Figure 5.10 the highest averaged and 90th percentile particulate matter concentrations are observed about the east-northeast (ENE) and southeast directions, indicating dominant sources from downtown or around Highway 401/407. There are some petroleum industries east of York, closer to Finch from the east-southeast direction, so this may be a contribution as well. There is also a smaller (but noticeable) peak in the southwest directions, suggesting that Pearson Airport and Highway 400 could be another potential source. High concentrations from the southeast and low concentrations from the northwest/north directions are rather consistent with the PM_{2.5} back trajectories from the air-quality stations. Given that a small time window was used to create these

back-trajectories, discrepancies are expected. In both the individual DRX1 and DRX2 PM back-trajectories, somewhat high values are also seen from the direction of Pearson Airport, indicating that Pearson could be a potential source, consistent with the individual PM_{2.5} CWTs from the AQ stations.

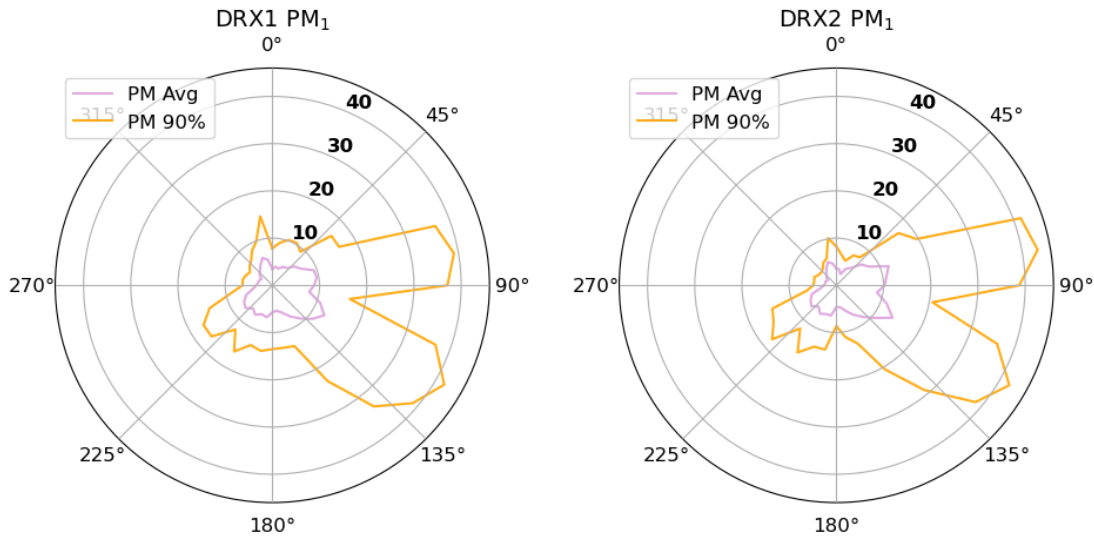


Figure 5.10: CIX DRX1 (left), and DRX2 PM₁ polar plots with 10° wind direction binning. Units are in $\mu\text{g}/\text{m}^3$.

The polar plots generated for DRX2 is nearly identical to DRX1. However, on average, the CWTs for DRX1 are higher than the corresponding CWTs for DRX2. Once DRX2 was placed on the rooftop, lower concentrations were reported on average compared to DRX1. Throughout the period after DRX2 was moved to the rooftop, the mean 10-minute averaged DRX1 and DRX2 were $3.26 \mu\text{g}/\text{m}^3$ and $2.73 \mu\text{g}/\text{m}^3$, when both instruments were recording measurements. This implies a lower concentrations on the rooftop than the 4th floor. This is most likely due to the fact that pollutant concentrations tend to be the greatest at the ground level, since they diffuse as they go up. This is similar to a potential reason of greatest concentrations in Toronto's south from chapter 4. However, a possible reason for this height gradient may be due to the DRX2 instrument exhibiting greater wind exposure than DRX1. With the DRX1 instrument located on the 4th floor balcony, only one side is exposed to the wind. The other side is facing the PSE building wall. This may result in more pollutant accumulation and stagnation compared to the location of DRX2.

5.2.1.2 2B Ozone Instruments and Sulfur Dioxide

5-minute averaged 2B1, 2B2, and 2B3 ozone concentrations are displayed in Figure 5.6, after 2B2, and 2B3 were cross compared to 2B1. There was still an offset between 2B1, and 2B2 after the 2B2 instrument stopped malfunctioning on August 11. This discrepancy is seen in Figure 5.6 (left).

Recall that both instruments were side by side during this time. Perhaps this is due to instrumental error after it resumed operating again. As such, from August 11 to the end of the measurement period 2B2 was multiplied by (1/0.826).

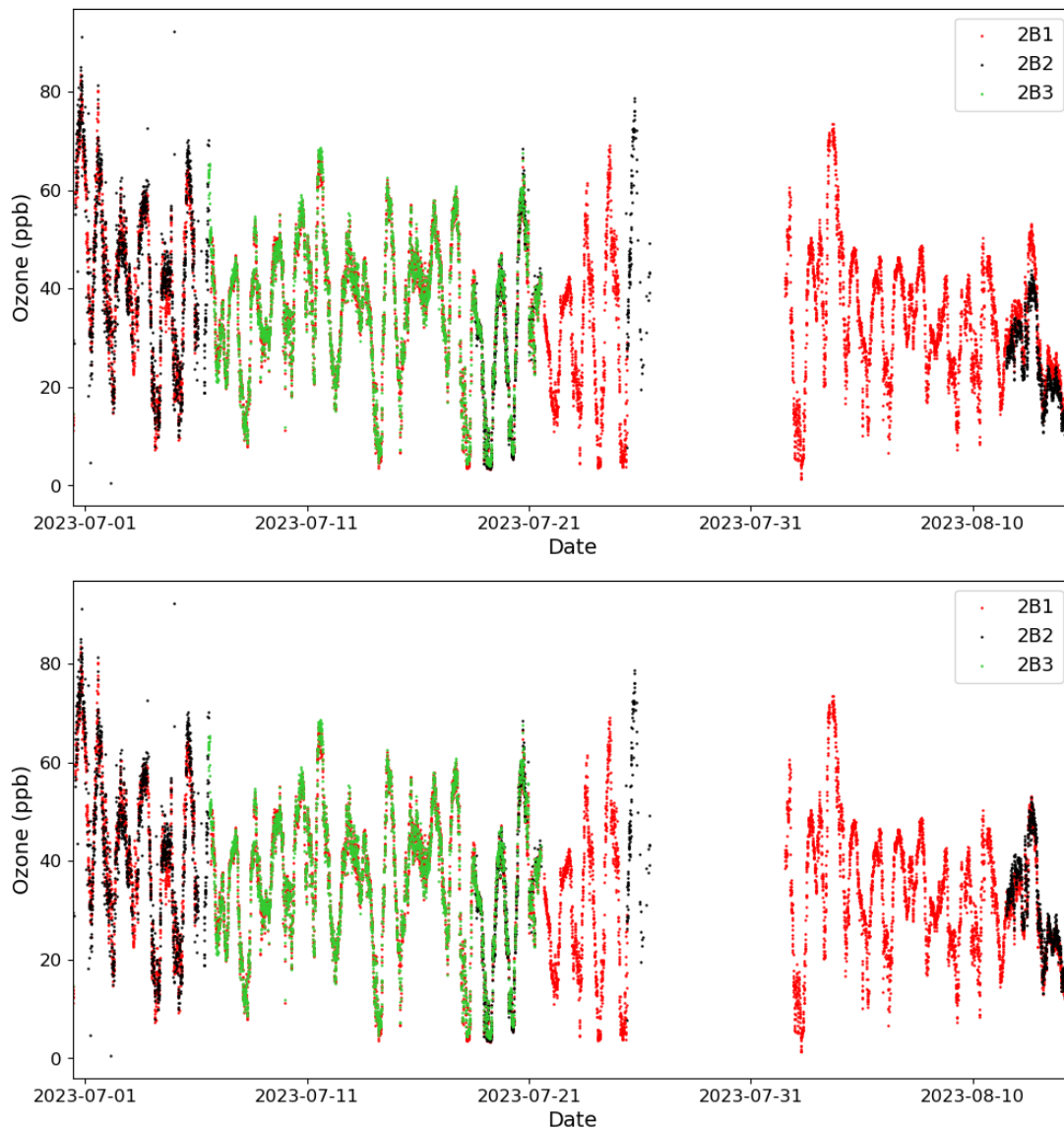


Figure 5.11: 2B1, 2B2, and 2B3 ozone time-series after 2B2, and 2B3 were cross-compared to 2B2 before (top), and after (bottom) correction factor was applied to 2B2 ozone concentrations beginning August 11.

Figure 5.12 displays polar plots for the 2B1, 2B2, 2B3, and ENVEA SO₂ instrument. Comparing the polar plots generated from 2B1 and 2B3, high concentrations are present in the southeast to southwest directions. This is consistent with the averaged back-trajectories from the air-quality stations in Chapter 4. A possible emission source in these directions could be the Toronto Billy Bishop Airport (like stated by Transport & Environment. (n.d.)), or long-range transport from be-

yond the city. Although, recall from Chapter 4 that ozone was anti-correlated with NO_2 , NO_x , and its concentrations were lowest around Highway 401, and 407. This may also be seen in Figure 5.12, as the lowest concentrations of ozone tend to be from the east to southeast

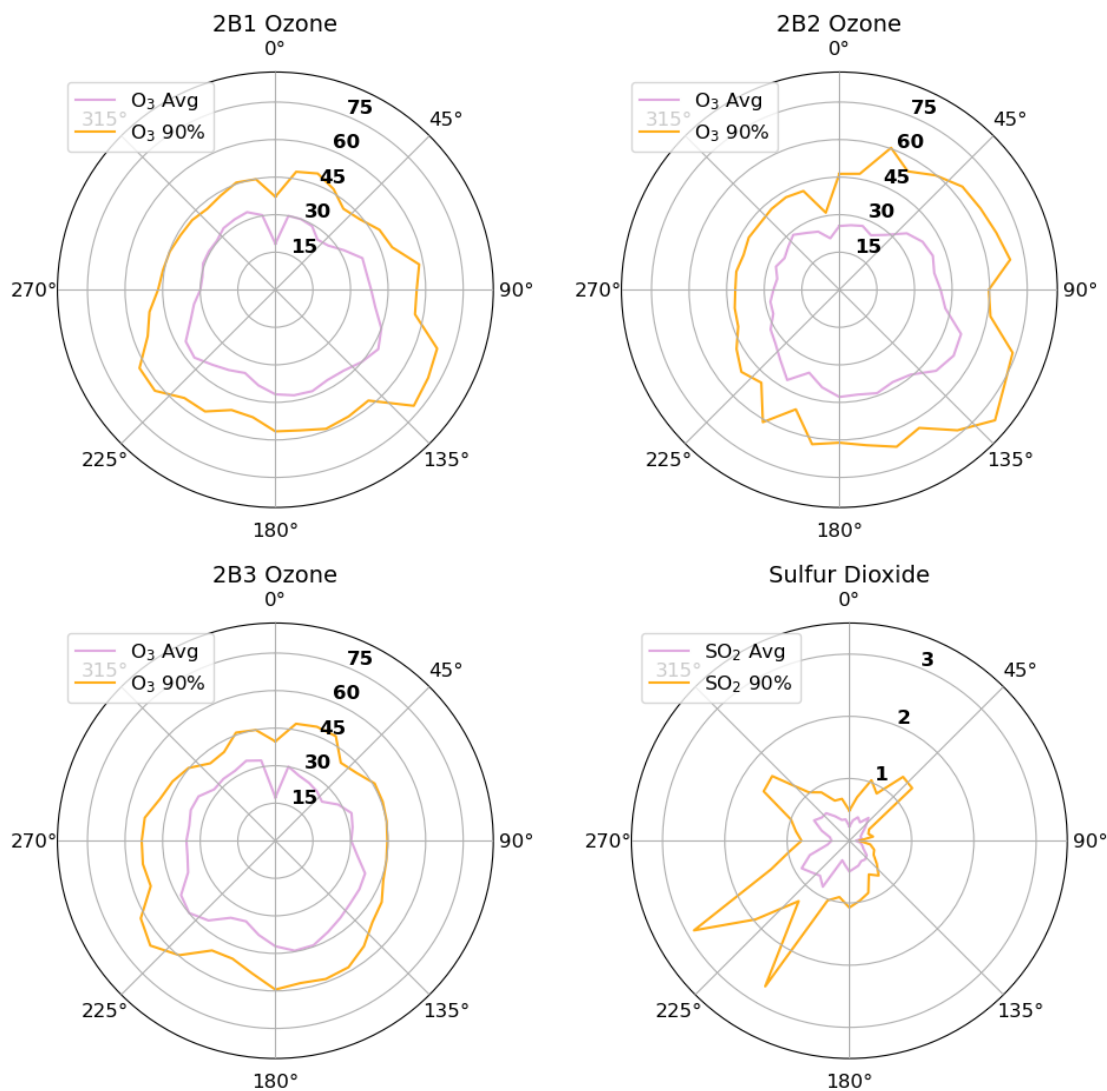


Figure 5.12: CIX 2B1 O_3 (ppb) polar plots with with 10° wind direction binning, with an additional SO_2 (ppm) polar plot from the campaign.

In Figure 5.12, elevated levels of SO_2 are observed in the southwest. As with PM, high values are also measured from the direction of Pearson Airport, suggesting potential common sources, from the airport or other close-by sources. Less elevated, but still high concentrations are measured in the direction of the northwest as well. Interestingly, these patterns can still be seen when only 69.8% of the 5-minute averages SO_2 measurements were available post drift-correction. However, high concentrations in the northwest contradicts the low concentrations in this direction observed in the averaged ground-level SO_2 polar plots.

5.2.2 SWAPIT Campaign

5.2.2.1 DustTracks

The 10-minute averaged calibrated DRX1 and DRX2 time-series is shown in Figure 5.13. The clear discrepancies between DRX1 and DRX2 are no longer present.

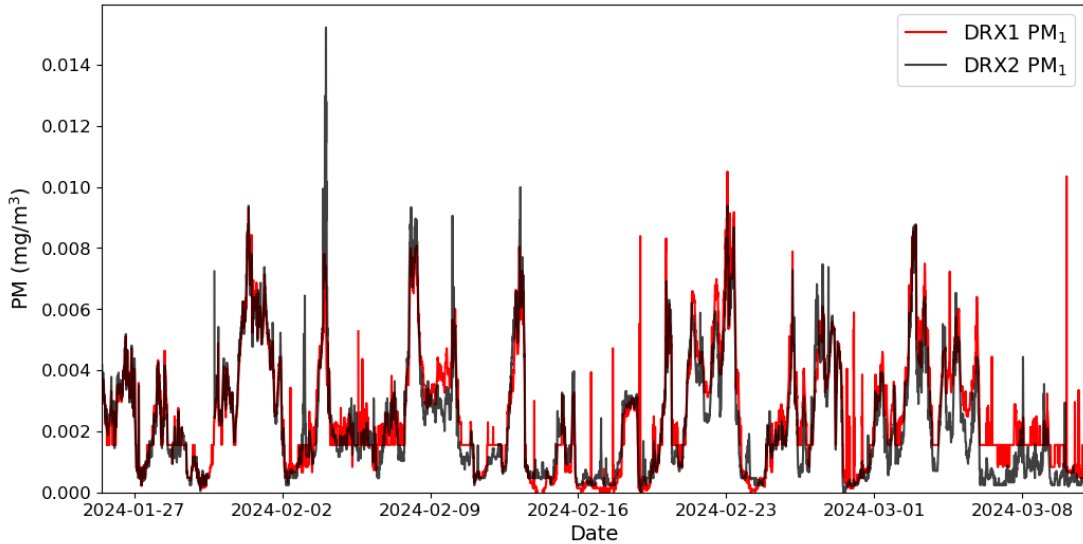


Figure 5.13: Like Figure 5.9, but for the SWAPIT campaign

Figure 5.14 shows the SWAPIT DRX1 and DRX2 polar plots with 10° wind direction binning. On average, the DRX2 concentrations are greater than the DRX1 concentrations (the mean DRX1, and DRX2 10-minute averaged concentrations were 2.25 and $2.50 \mu\text{g}/\text{m}^3$ respectively, post calibration), implying concentrations at the Keele Street site were greater than at the Petrie site. 4747 Keele Street is situated right next to an intersection, therefore vehicular emissions may have contributed to this discrepancy. High concentrations about the southwest are much more defined in the SWAPIT particulate matter plots (Figure 5.14), compared to THE CIX PM plots (Figure 5.10), this supports the suggested potential sources near and around Pearson Airport (like stated by Transport & Environment., (n.d.)), and Highway 400. Although, this also suggests the potential sources from Pearson and Highway 400 are considerably stronger during the SWAPIT campaign.

As with THE CIX campaign, elevated concentrations of particulate matter are present about the ENE direction, also suggesting areas near Highway 401/407, or other industries east of York, are probable sources, similar to what was obtained in the averaged NO_x , NO , and NO_2 back trajectories in Figures 4.7, and 4.12. Unlike the ground-level averaged $\text{PM}_{2.5}$ CWTs in Figure 4.7, the highest concentrations are not observed in southeast and southwest directions. No apparent inverse relationship exists between the $\text{PM}_{2.5}$, with the SO_2 and O_3 ground-level back-trajectories, where

some inverse relationship is present in these campaigns (as mentioned previously PM, and SO₂ exhibit elevated concentration from the northeast, where the opposite is true for ozone). Although, in the individual ground-level generated back-trajectories (Figures A.1, and A.11), a small inverse relationship can be observed between PM_{2.5} and ozone, such as in Toronto North and University of Toronto.

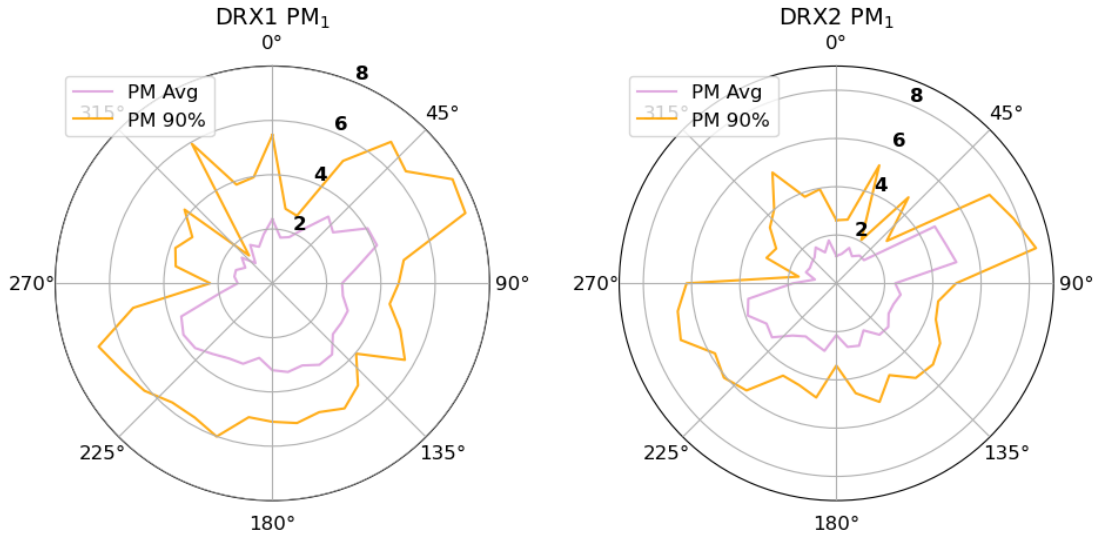


Figure 5.14: Like Figure 5.10 but for the SWAPIT campaign.

Comparing the SWAPIT to THE CIX DRX polar plots, it is clear that the particulate matter concentrations for SWAPIT are considerably lower than those during THE CIX campaign. During the SWAPIT and THE CIX campaign, the average post calibration DRX1 PM_{2.5} concentrations were 2.55, and 5.71 µg/m³ respectively. This is expected due to the presence of wildfire smoke and seasonal natural of particular matter concentrations (greater in summer than in winter, consistent with Figure 3.4). The values are measured during both campaigns are considerably smaller than the average PM_{2.5} concentrations measured by the ground-level stations as shown in Figure 3.4. Since the UHSAS measures PM₁ (which the DRX instruments were calibrated to), this should be a lower concentration than PM_{2.5}, (although, as already stated, little to no difference was observed between these measurements in the DustTrak instruments). Furthermore biomass burning smoke during THE CIX campaign also contributed to this discrepancy. It should be noted that high presence of smoke during THE CIX campaign, identification of sources during that campaign and comparisons to SWAPIT difficult.

5.2.2.2 Ozone and Sulfur Dioxide

Figure 5.15 displays the ozone time-series plot, and Figure 5.16 displays the polar plots.

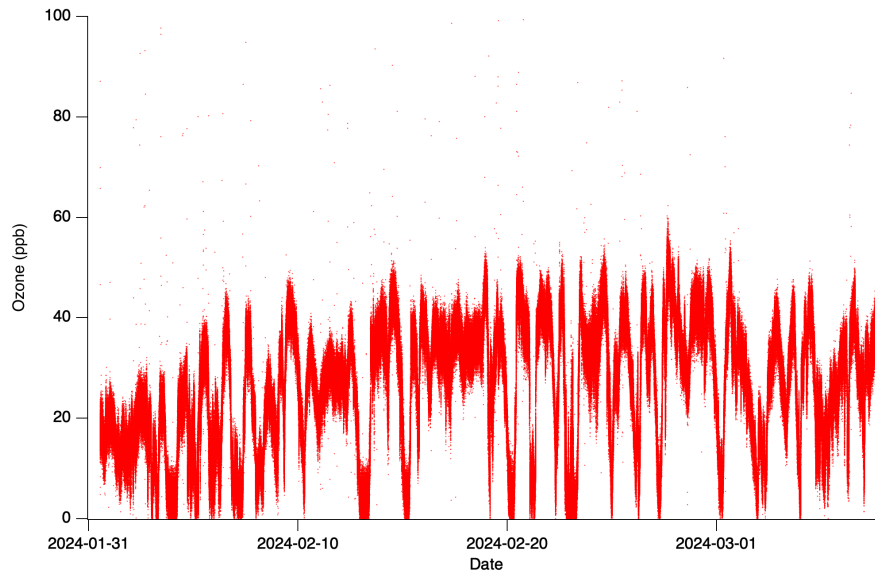


Figure 5.15: Ozone time-series for the SWAPIT campaign, ranging from 01-31-2024, 13:27:12 to 03-08-2024, 23:59:58.

Polar plots for O_3 and SO_2 is shown in Figure 5.16.

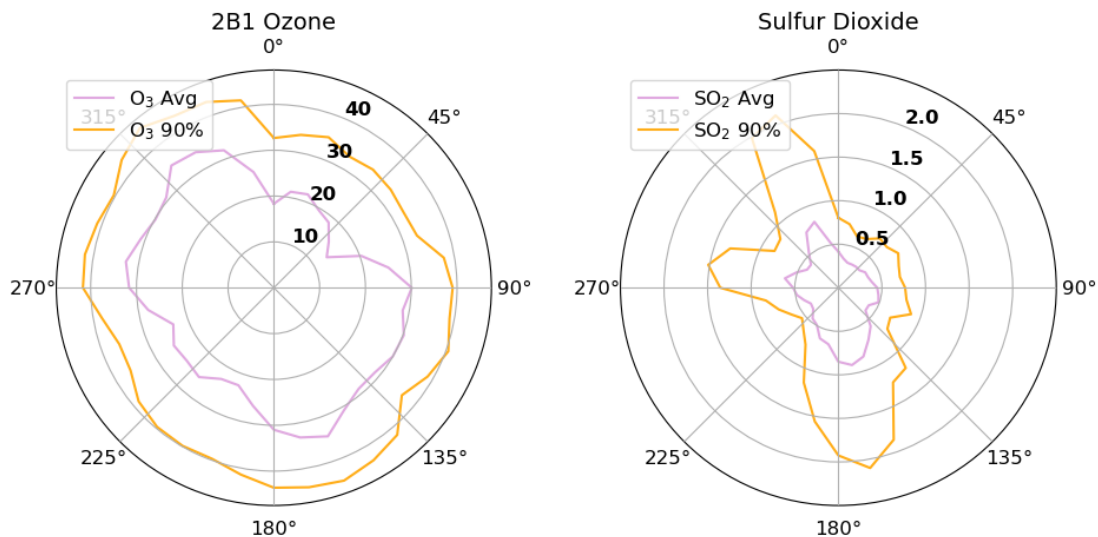


Figure 5.16: SWAPIT O_3 (left) and SO_2 (right) polar plot, with 10° wind direction binning.

Unlike THE CIX ozone polar plots, the SWAPIT 2B1 polar plot in Figure 5.16 displays the greatest concentrations about both the northwest and southeast directions. These elevated concentrations may also be from the Portlands Energy Center in Toronto Island. High concentration values

from the northwest also contradicts the back trajectories from the ozone air-quality stations, though this is just for one small period in time. The average 2B1 concentrations during winter and summer were 34.96 ppb and 26.52 ppb respectively. The 2022 average JJA and JFM ozone concentrations across the four Toronto West, Downtown, North and East AQ stations are 27.83 ppb and 24.71 ppb respectively. The seasonal percent differences from the AQ station are roughly 50% lower compared to the 2B1 measurements, with the remaining difference most likely being attributed to wildfire smoke during THE CIX campaign. Although, ozone is increasing, the annual trends in Table 3.4, are not enough to account for the 1 and 1.5 year discrepancy between the ozone AQ stations and CIX and SWAPIT campaigns respectively.

The SWAPIT SO₂ polar plots in Figure 5.16 is similar to THE CIX polar plot displayed in Figure 5.12, for a significantly shorter time-period than THE CIX campaign. It is interesting to observe similarity in the SO₂ polar plots for both campaigns with different lengths of time, since this confirms the general directions of high pollutant concentrations throughout the 2023 summer, and winter 2024. The greatest concentrations in Figure 5.16, is more west, instead of the southwest (as with THE CIX), but still suggests sources from Pearson Airport (as per the information given by Transport & Environment. (n.d.)), and Highway 401. Like the ozone and PM polar plots, high concentrations are present about the northwest as well, indicating a common source from that direction, perhaps from industries in Vaughan. Unlike THE CIX SO₂ polar plots in Figure 5.12, high concentrations are also observed about the southeast direction in Figure 5.16, suggesting sources from the downtown areas, or even beyond the city, like the conclusions in Chapter 4. This is also consistent with the AQ station back trajectories in Chapter 4, but is inconsistent with the fact that the lowest concentrations are observed about the northwest to north directions. Nevertheless, as with THE CIX campaign, it is interesting how defined results can be observed with only 74.1% of the 5-minute averaged SO₂ data being available post-drift correction. Mean SO₂ during THE CIX and SWAPIT campaigns were 0.55 and 0.62 ppb respectively, indicating a slight increase during the winter. This slight increase is most likely also due to the fact that wildfire smoke elevated SO₂ concentrations during THE CIX campaign. Though this increase may not be significant. The JFM and JJA 2022 averaged Toronto West SO₂ concentrations were 0.29 and 0.19 ppb respectively, suggesting the measurements from the ENVEA instrument were too high. This could be attributed to limited data availability and the drift correction. Toronto West SO₂ trends are decreasing (see Table 3.4).

6 Pollutant Spatial Variations from TROPOMI Satellite

On October 13, 2017, the Sentinel-5 Precursor (S5P) satellite containing the TROPOMI (Tropospheric Monitoring Instrument), was placed in orbit (Ialongo et al., 2020). The S5P is located 817 km above mean sea level, and is in a helios-synchronous orbit, with a 17-day pathway periodicity and an equatorial crossing time of 1:30 p.m. LST (Babić et al., 2022). The TROPOMI instrument, in particular, is a hyper-spectral passive sensor that observes Earth directly downwards (Ialongo et al., 2020). It contains four spectrometers, allowing for the quantification of near-infrared (NIR), short-wavelength infrared (SWIR), ultraviolet (UV), and ultraviolet-visible (UV-VIS), wavelengths (ibid.). Measurements from TROPOMI include UV at the surface, aerosol attributes, O₃, CH₂O, carbon monoxide, SO₂, NO₂, and methane (ibid.). It is important to note that TROPOMI measures total column concentrations (TROPOMI, n.d.). The pixel-size is 3.5×5.5 km, though preceding August 6, 2019, the size was 3.5×7 km (Ialongo et al., 2020).

Pollutants are generally released at the surface level and mix within the boundary layer. If there are no other sources of pollutants within the atmosphere, then it can be stated with confidence that most pollutants are near the surface level. Since this is the case, using TROPOMI satellite measurements to map pollutant spatial variations throughout Toronto can provide a good comparison and validation for the back-trajectory methods carried out in Chapter 4. An exception includes ozone since it is known that 90% of ozone is stratospheric (Rowland, 1991).

6.1 Methodology

6.1.1 Satellite Data Collection and Measurements

The TROPOMI satellite data is provided by the European Space Agency (ESA). For this project, data were provided by Debora Griffin and Chris McLinden at Environment and Climate Change Canada for an area roughly covering the Greater Toronto Hamilton Area ($43.0^\circ \leq \text{Latitude} \leq 44.1^\circ$, $-80.3^\circ \leq \text{Longitude} \leq -78.5^\circ$). Data was divided into monthly .csv files beginning May 2018. The end date for each species ranged from Nov. 2023 to Sept. 2024. To eliminate seasonal bias, the end month for all species would be chosen to be April 2023 or 2024. To maintain seasonal consistency, the end date for all species (except for O₃) was chosen to be no later than April 29, 2023, since BLHs were used for conversion of surface to column concentrations (to be explained in section 6.1.3), and were only available until that date. Ozone was not converted to a surface concentration, as such, its end month was selected to be April, 2024.

In this analysis, the latitude and longitude range was restricted to 43.56° to 43.84° , and -79.64° to -79.14° , to better focus on Toronto. To eliminate potential seasonal bias, and ensure consistency between the pollutant spatial maps, the months of April 2023 was selected as the final month for each dataset. The center point was (43.69866° , -79.38996°), slightly different then the center point of 43.71212° , -79.40935° in the Chapter 4 superimposed back-trajectories. This center point was chosen to be more-so centered about Toronto than the four “Toronto West”, “Toronto North”, “Toronto Downtown”, “Toronto East” air-quality stations.

Table 6.1: The filter variables were defined by the data providers, and the filters were applied based on the recommendations from the dataset providers. “qa” denotes the quality of the measurements, on a scale from 0 to 1, “Cld_f” denotes the percentage of cloud coverage for a given data pixel, “SZA” represents the solar zenith angle, and “snow” can equal 0 or 1, to indicate no presence of or presence of snow respectively. Additionally, negative concentration values were removed if they were present.

Pollutant	Filters
NO ₂	Cld_f <0.3, qa >0.75
SO ₂	Cld_f <0.3, qa >0.75, snow=0, SZA <70
O ₃	qa >0.75
HCHO	Cld_f <0.3, qa >0.75
CO	qa >0.75

Table 6.1 displays the filters applied to data for each species, based on the suggested filters from the data providers. There were also some secondary (optional) filters recommend by Debora and Chris that were not applied. Additionally, any reported concentrations <0 for SO₂, as these were considered outliers since the average concentration throughout its time period for the GTHA was <0, when those values were included. NO₂ and HCHO contained some negative values, but they were not removed since these values were considered noise and not significant. However, some HCHO concentrations were reported as -999, and these were removed as well.

These datasets contained specific latitude and longitude points with corresponding values. This data was used to produce a more complete picture of the pollutant spatial maps throughout Toronto. As a first step to produce the concentration maps (shown in Figure 6.4below), the selected area was divided into grids with 250-meter spatial resolution. Then, the lat/lon points and their corresponding values were centered about the nearest grid and extended to 3.5×7 and 3.5×5.5 kilometers preceding, and post (as well as including) August 6, 2019 respectively. Recall the spatial resolution of TROPOMI mentioned earlier in this Chapter. The overlapping pixels were then averaged. It is worth noting that according to the data providers, unlike the other pollutants, HCHO used is a surface concentration, that converts from column to surface utilizing GEM-MACH profiles, and

SO₂ was a vertical column density.

6.1.2 Boundary Layer Height Calculation

The TROPOMI NO₂ dataset also included corresponding boundary layer heights (BLHs), and temperatures from the ERA5 reanalysis. The data providers adjusted these BLHs to match the closest corresponding point on a 0.25×0.25 degree grid space and the corresponding timestamp. A sample map displaying the BLHs and surface temperatures for the month of June 2018 is displayed in Figure 6.1 below.

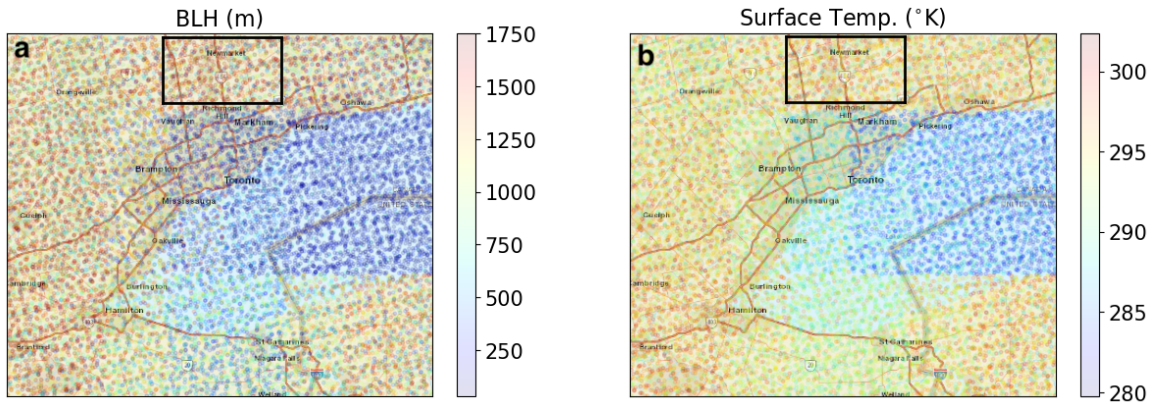


Figure 6.1: Raw BLH (a) and surface temperature (b) for June 2019, from the TROPOMI satellite data provided.

As shown in Figure 6.1, it appears that the colder temperatures and lower boundary layer height over Lake Ontario is shifted too far north. As such, the following area was selected to estimate the boundary layer height in Toronto: $43.9 \leq \text{Latitude} \leq 44.1$, $-79.64 \leq \text{Longitude} \leq -79.14$. This box is displayed in both the images in the Figure 6.1. From this region selected, daily average boundary layer heights were computed (shown in Figure 6.2), and a linear interpolation was employed fill in the daily-averaged BLH in for days where it was not available.

In Figure 6.1, it could be argued that the Lake is blowing cold air into Toronto and its surrounding areas instead of the surface temperatures and BLHs being shifted too northerly. To prove this isn't the case, ERA5 $0.5^\circ \times 0.5^\circ$, reanalysis is also provided by Climate Re-analyzer (https://climatereanalyzer.org/research_tools/monthly_maps/) (Climate Reanalyzer., 2024). The average 2-m temperature for June, 2018 is displayed in Figure 6.3. There is also an option to select “contour plot” which smooths the data, is also shown in Figure 6.3 (b).

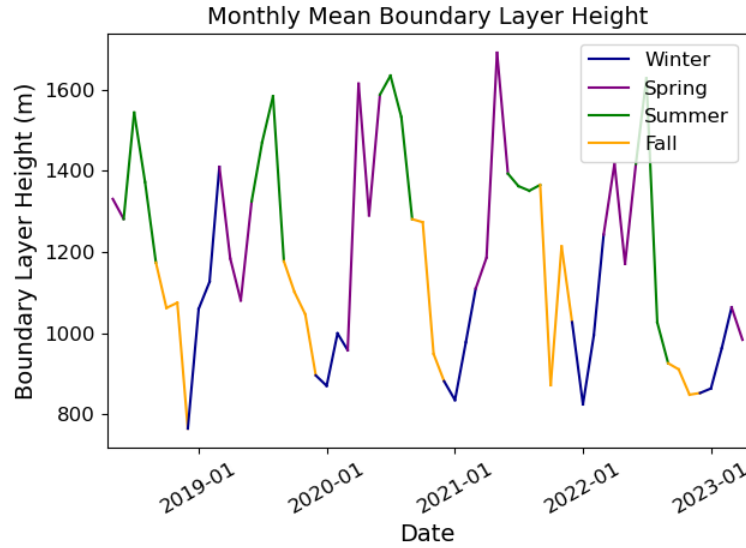


Figure 6.2: Monthly mean (right) BLH from the analysis period.

From examining Figure 6.3 (b), it is clear that the cooler temperatures over Lake Ontario are not present over Toronto and its surrounding areas. It can be assumed the same is true for the BLH since these directly correspond to the surface temperatures in Figure 6.1. In Figure 6.3 (a), it is apparent that the pixel centers are taken as the whole pixels, which can result in erroneous temperatures near the Great Lakes, and other major bodies of water before smoothing, due to the reanalysis resolution. The same can also be stated in Figure 6.1.

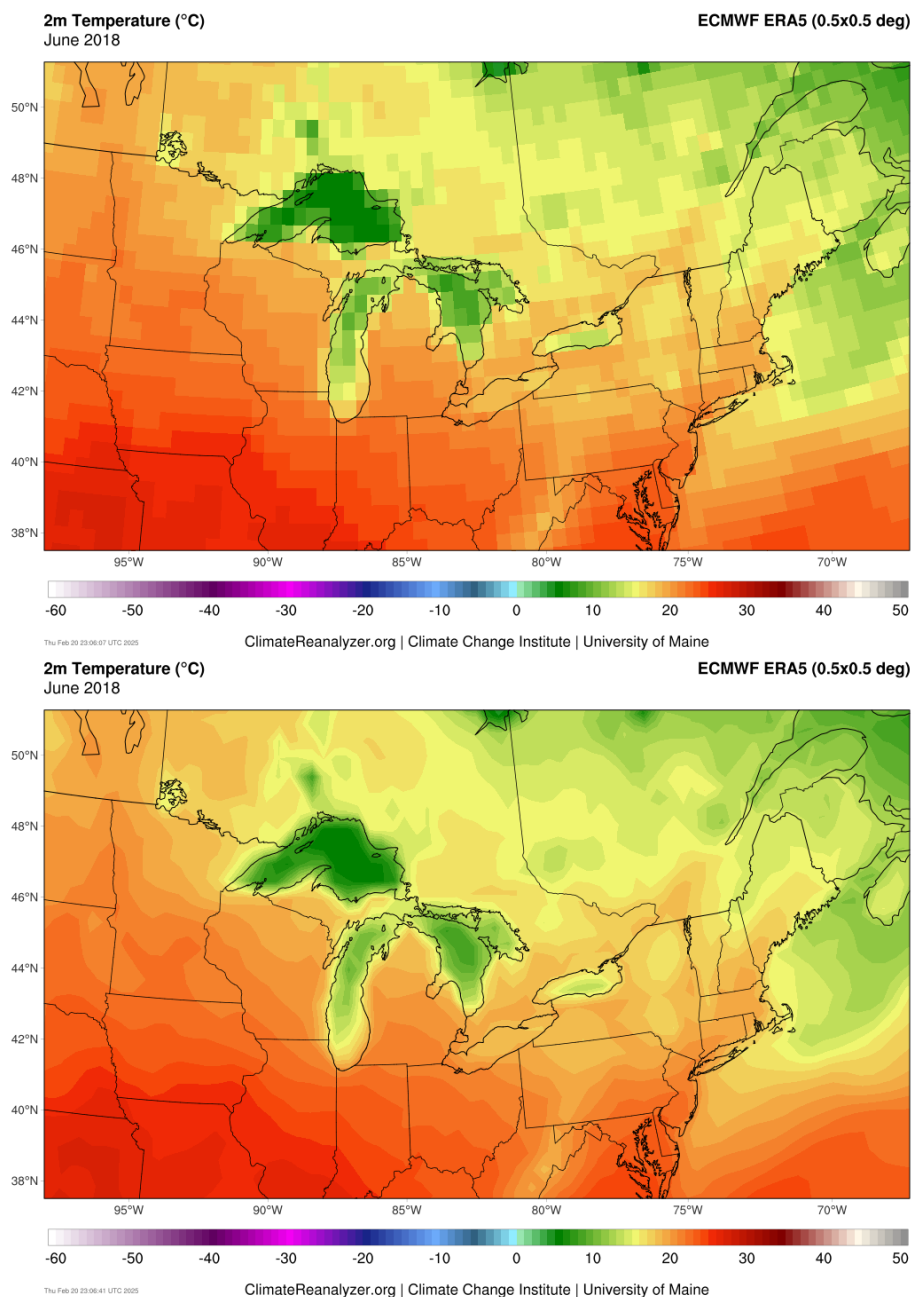


Figure 6.3: ERA $0.5^\circ \times 0.5^\circ$ reanalysis surface temperature for June 2018 pixelated (top) and smoothed on a contour plot (bottom) taken from Climate Reanalyzer.org (Climate Reanalyzer., 2024).

6.1.3 Conversion from Column to Surface Concentrations

The units of NO_2 and CO from TROPOMI were molecules/ cm^2 . SO_2 , and O_3 were provided in Dobson Units (DU). To convert into a surface concentration, these measurements were first converted into mass (mg or μg) per m^2 , then divided by the daily-averaged boundary layer height to obtain units of mass/m^3 .

In the NO₂ and CO case, this conversion is done as follows;

$$[c'_1] = \frac{[c]M_{mc}}{N_a} \frac{(10^4)}{(BLH)} \quad (6.1)$$

where $[c'_1]$ is the converted concentration of NO and CO in g/m³, $[c]$ is the initial species column mass (provided in molecules/cm²), M_{mc} denotes the species molar mass (g/mol), N_a is Avogadro's number given by 6.022×10^{23} molecules/mol, and BLH is the daily averaged boundary layer height in meters. A factor of 10^4 is applied to converted from cm⁻² to m⁻².

Since SO₂ was given in Dobson Units, the following equation was applied.

$$[c'_2] = \frac{[c]M_{mc}}{N_a} \frac{(DutC)}{(BLH)} \quad (6.2)$$

where DutC is the conversion from Dobson Units to concentration given by 2.687×10^{20} molecules/m² (Remsberg et al., 2020). It is important to note that for these conversions, in equations 6.1 and 6.2, the daily concentrations (measured by the satellite at approximately 18:00Z each day), corresponded to the daily averaged BLHs. In other words, the concentrations were divided by the daily averaged BLH for each of their dates.

To ensure proper comparison with the AQ station back-trajectories, O₃, SO₂ concentrations were converted into ppb, and CO was converted into ppm. This was done as follows;

$$[x] = \frac{[c']M_{mair}RT}{PM_{mc}} \quad (6.3)$$

where M_{mair} is the molar mass of air (28.96 g/mol), P and T are the air pressure (pa) and temperature (°K) respectively, $[c']$ is either $[c'_1]$ or $[c'_2]$, and R is the gas constant of dry air given by 0.287 J/(K·g). The substance mixing ratio in mol/mol is denoted by $[x]$, for example, if $[x]$ was 46×10^{-9} , then it would be reported as 46 ppb.

Combining equations 6.1 and 6.2 with 6.3 gives the following two equations;

$$[x_1] = \frac{[c]M_{mair}RT}{N_a P} \frac{(10^4)}{(BLH)} \quad (6.4)$$

where $[x_1]$ denotes the mixing ratio of NO₂, and CO.

$$[x_2] = \frac{[c]M_{\text{mair}}RT}{N_a P} \frac{(\text{DutC})}{(\text{BLH})} \quad (6.5)$$

where $[x_2]$ denotes the mixing ratio of SO_2 . Notice that the substance molar mass is no longer required.

Hourly temperature and station-level pressure measurements were taken from an ECCC and WMO station in downtown Toronto (located at 43.7° - 79.4°). This data is provided by ECCC's past weather data page (Government of Canada, n.d.). Missing data was linearly interpolated. Since the vast majority of TROPOMI measurements were from 17:00 to 19:00 Z, these measurements were extracted daily at 18Z. This stations does not report wind, therefore it could not be used the back-trajectories in Chapter 4.

6.2 Results and Discussion

Figure 6.4 shows a more comprehensive spatial mixing ratio maps from all of the TROPOMI measurements. Generally, these results demonstrate that the TROPOMI satellites resolution is too low to observe spatial variations throughout Toronto from just averaging over 4-years, given the spatial-scale of Toronto. Perhaps, one would need to average over a much longer time period (i.e., decades) to spatially resolve concentrations within the city. In Figure 6.4, the clearest spatial distribution is present for NO_2 . Regarding these spatial variations are seen near Pearson Airport (consistent with the AQ station back-trajectory maps), and gradually extend towards the city center. There is a hotspot in the downtown area as well, which is also in-accord with Figures 4.7, and 4.15 and similar to the results from Jerrett et al. (2007). Moreover, low NO_2 concentrations present in eastern Toronto is consistent with the findings by Jerret et al. (2007), as well, but not with the superimposed AQ station back-trajectories in Chapter 4. Unlike the AQ station back-trajectories, and the findings by Jerrett et al. (2007), these high AQ station back-trajectories do not roughly trace Highway 401. It is difficult to observe the spatial variations of HCHO , but a close inspection reveals the lowest concentrations are generally over Lake Ontario, whilst the Downtown area, extending west to Etobicoke exhibits the greatest concentrations, similar to the NO_2 spatial variations.

Although the spatial variations of CO throughout Toronto in Figure 6.4 are difficult to observe as well, the lowest concentrations are situated over Lake Ontario, and the greatest concentrations tend to exist around mid to uptown Toronto, similar to the back-trajectory maps in Figures 4.7, and 4.15. Generally, CO is less elevated in East Toronto, unfortunately there is no data in this region in Figures 4.7, and 4.15 for comparison. Despite this thesis only being concerned with the pollutant spatial variations in Toronto, an additional observation are the inconsistencies and null data points

on Lake Ontario, indicating the sparseness of data availability over this area. Despite hypothesized that it may still be possible to observe spatial variability of ozone, none is present. SO₂ also exhibits no spatial variability.

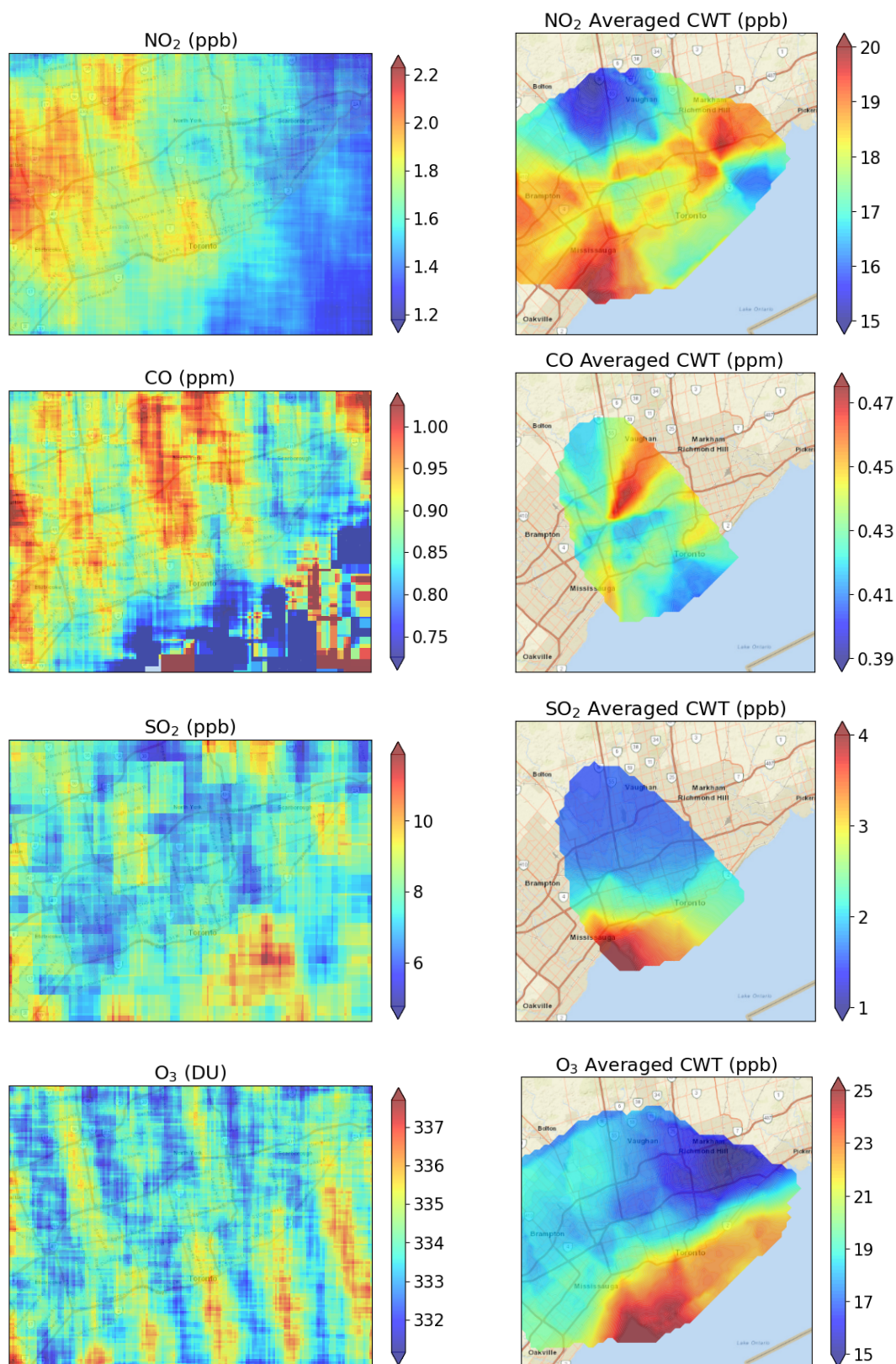


Figure 6.4: TROPOMI satellite (left) vs. AQ station back-trajectories (right). Shown is NO₂ (1st row), CO (2nd row), SO₂ (3rd row), and O₃ (4th row).

The additional HCHO spatial map from TROPOMI is displayed in Figure 6.5.

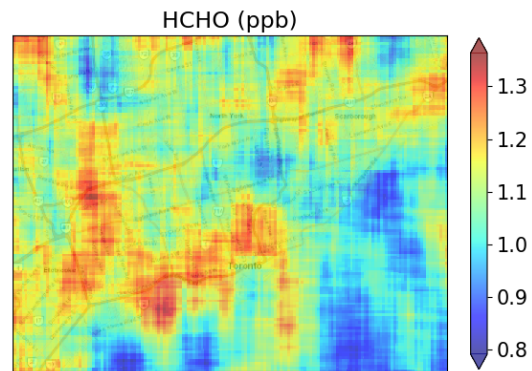


Figure 6.5: TROPOMI HCHO (ppb).

From Figure 6.4, one can also obtain concentrations at the center points in each district in Toronto, shown in Figure 4.6, (like in Chapter 4). These neighborhood concentrations can then be related to the neighborhood concentrations from Figure 4.7, in the socioeconomic mapping plots in Figure 4.13. This is displayed in Figure 6.6.

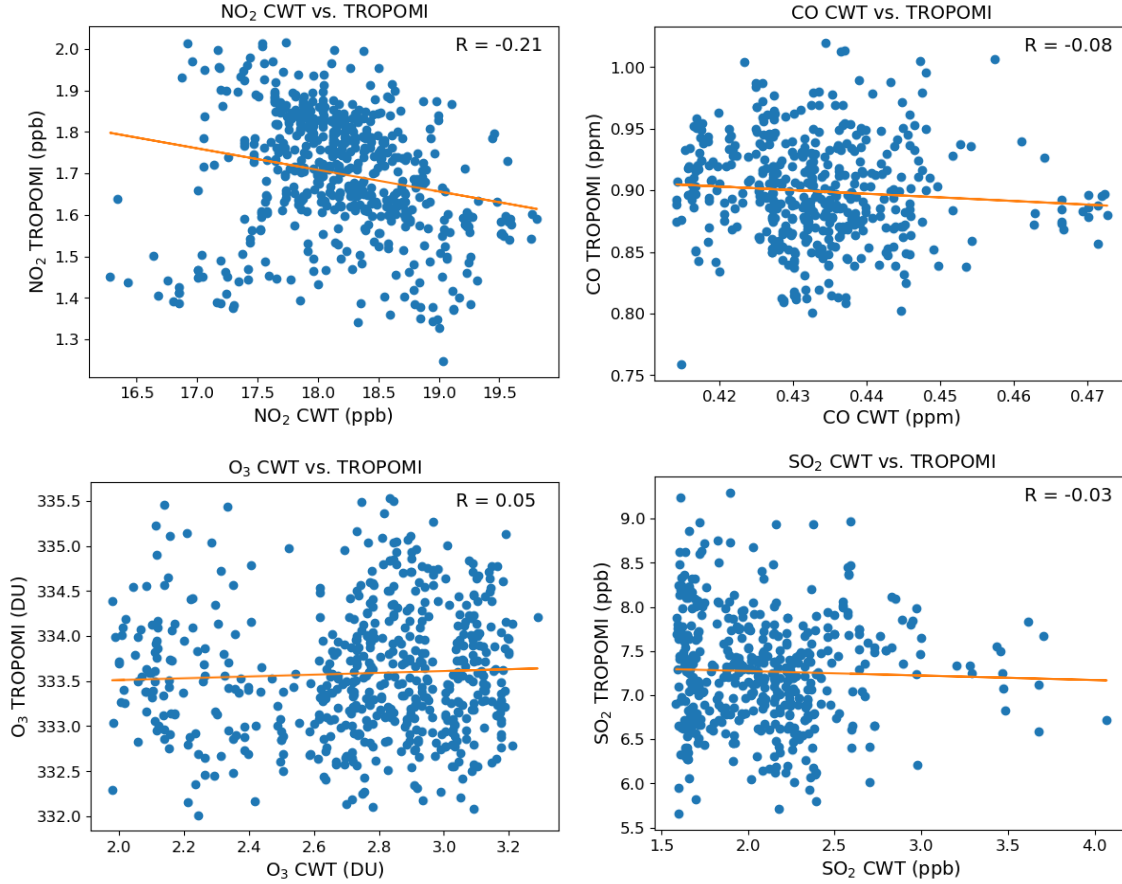


Figure 6.6: Relationships between NO₂ (top-left), SO₂ (top-right), O₃ (left) averaged CWTs and TROPOMI maps for neighborhoods in Toronto.

Equation 6.4 was applied to convert TROPOMI NO₂ and CO into a surface level ppb concentration. The slight negative relationship for NO₂ seen in Figure 6.6 can be attributed to the fact that in TROPOMI NO₂ exhibits a negative eastward gradient. In the CWTs, NO₂ is relatively high downtown Toronto, with high concentrations also tracing Highway 401, as mentioned previously. Even though the spatial distribution of CO from the TROPOMI and CWT maps generally display high concentrations in north Toronto, and lower concentrations downtown, the TROPOMI to CWT relationship is still slightly negative. When inspecting both these maps closely, this is expected since it can be observed that the high patches of CO from TROPOMI do not correspond to the elevated area of CO in the CWT maps near Downsview Airport and the York University power plant. Furthermore, the high concentrations of CO near Pearson from TROPOMI are not present in the CWT maps. Given the lack of spatial variability in SO₂, and TROPOMI O₃ these pollutant relationships are meaningless.

Despite no spatial variations from the TROPOMI ozone, equation 6.5 was inverted to calculate

the concentration in DU from ppb for the AQ stations. This calculation was carried-out from May 1, 2018 to December 31, 2022. Since the AQ stations measured hourly concentrations, this calculation utilized hourly temperatures and pressures, and averaged daily BLHs for each day. Recall the “average daily BLH” is really the average at around 18Z, since the same height was applied for each hour in the day, a potential error arises from not considering its diurnal variation. Nonetheless a relationship is still not observed.

The discrepancies between the average TROPOMI to CWT maps concentrations after this conversion were also calculated. These conversions assume 100% of NO₂ and CO is below the BLH, and the surface concentrations measured by ozone is consistent throughout the whole entire atmosphere. Surface NO₂ is still considerably greater than the post-conversion TROPOMI concentrations. Perhaps this indicates a strong presence of near-surface NO₂ from emitters that are not completely detected by the satellite. Note that the converted CO from TROPOMI is clearly higher than the concentrations measured by the AQ stations, and the opposite is true for ozone. Given that ozone is spatially uniform from TROPOMI satellite imagery, but not from the ground-level observations, indicates the vast majority of ozone is not located near the surface, consistent with the fact that a large proportion of column ozone is stratospheric, as stated by Rowland (1991).

7 Conclusion

Temporally, NO_2 , NO , NO_x , SO_2 , and CO concentrations exhibited decreasing trend-lines. $\text{PM}_{2.5}$ was found to have increased and decreased during the winter and summer, respectively. Given what was stated by Jeong et al. (2020), the decrease in pollutant concentrations may be partly be explained by reductions in vehicular tailpipe emissions. As per the Hilker et al. (2021) study, the decline in NO_2 , SO_2 , and summertime $\text{PM}_{2.5}$ may also partly be explained by vehicular technology improvements and the gradual elimination of coal-powered electricity between 2004 and 2015. Specifically, the power-plant closure in Nanticoke may also contribute towards the decreasing trends. Ozone concentrations were generally inverse to NO_2 , NO , and NO_x concentrations in the polar plots, trends, and back trajectories. As described in Seinfeld and Pandis (2016), and Sillmann (n.d.), the inverse relationship between O_3 , and NO_x implies a high NO_x concentration relative to VOC. As such, ozone was the only pollutant with an increasing concentration trend, with the exception of the Downtown station during summer. In north Toronto, $\text{PM}_{2.5}$ was also found to be inversely related to ozone, based on the similarity of relatively low-income neighborhoods with high and low O_3 and $\text{PM}_{2.5}$ concentrations respectively.

In the back-trajectory maps, a hotspot near Downsview Airport, the York University power plant and the Highway 400/401 intersection is present for NO , NO_2 , NO_x , and CO . It is unclear if the source is located precisely over this location or from another source nearby (such as Downsview Airport, and the York University power plant). Given the limited number of air-quality stations in this area, there may be uncertainty in pinpointing the hotspots from the individual AQ station back-trajectory maps. In the NO_2 , and especially NO_x maps, a clear path of elevated concentrations can be observed tracing Highway 401, suggesting a vehicular emission source. All of NO , NO_2 , NO_x , and CO are inversely related to net median income, given these concentrations are the lowest in the downtown areas (where the bulk of high-income neighborhoods are located) and the highest in north Toronto (where plenty of low-income neighborhoods are present). Again, these spatial variations and relationships are the opposite for ozone.

No significant spatial variations are present in the $\text{PM}_{2.5}$, PM_{10} , and SO_2 averaged back-trajectories. Nearly all of these individual back-trajectories display the highest concentrations from the south, indicating their sources originate beyond the city. This is consistent with Jeong et al. (2020), who found that in Toronto, $\text{PM}_{2.5}$ primarily originates from industries ranging from Illinois to Pennsylvania. Southward origination of $\text{PM}_{2.5}$ is also in accord to the results by Brook et al. (2011), who found that $\text{PM}_{2.5}$ concentrations were disproportionately high in the presence of southerly winds in Toronto and Egbert Ontario. This suggests that $\text{PM}_{2.5}$, PM_{10} , SO_2 and O_3

in this analysis may predominantly be from these sources as well, and the resulting concentration gradient from transport of these sources may mask the spatial variations within the city. Another potential source may be from the Portlands Energy Center in Toronto Island. Moreover, these pollutants may also be originating from a power plant in Nanticoke, Ontario, which closed in 2013. Recall, in chapter

Some similarities between THE CIX and SWAPIT campaign in Chapter 5 polar plots and the AQ station back trajectories in Chapter 4 are noted. These include high concentrations of ozone and $\text{PM}_{2.5}$ from the southeast, and east southeast respectively, indicating the greatest levels are located in downtown Toronto or beyond the city. These elevated concentrations may also partly be from the power station in Toronto Island. During THE CIX campaign, the lowest concentrations of ozone and $\text{PM}_{2.5}$ and O_3 were from the northwest, consistent with the $\text{PM}_{2.5}$ and O_3 AQ station spatial maps. Conversely, relatively high concentrations of $\text{PM}_{2.5}$, O_3 , and NO_2 were seen towards the northwest in the SWAPIT campaign, suggesting potential common source(s) in this direction. Although it is not possible to observe spatial variations in Figures 4.7, and 4.15 for $\text{PM}_{2.5}$ and SO_2 , these polar plots from THE CIX and SWAPIT campaigns indicate dominant pollutant sources from Highway 407, 401, 400 and Pearson Airport as well, validating the NO , NO_2 , and NO_x AQ station back-trajectory methodology. It is important to note that the presence of smoke during THE CIX campaign results in difficulties in identifying sources challenging, and comparing results to the SWAPIT campaign.

Despite the low-spatial resolution and total column measurement of concentrations, the TROPOMI satellite also provides partial validation regarding the back-trajectory methodology. One similarity, is no spatial variation being observed regarding SO_2 . However, elevated concentrations over Lake Ontario may indicate northward transport from locations south of Toronto. In the satellite data, the greatest concentrations of NO_2 are observed towards the west, near Pearson Airport, consistent with the air-quality station back-trajectory maps. Although, some dissimilarities include: high concentrations in the satellite imagery do not trace Highway 401 (also inconsistent with the results by Jerret et al. (2007).), and the TROPOMI maps containing a negative eastward gradient, unlike the AQ station back-trajectory maps. Low NO_2 concentration in east Toronto are in accord with the findings by Jerret et al. (2007). Recall that Jerret et al. (2007) used a land-use regression model considering observations over a two week period and found that NO_2 was the greatest in Downtown Toronto, around highways and the lowest in east Toronto. HCHO was also found to be the greatest and lowest over downtown Toronto to Etobicoke and Lake Ontario, similar to NO_2 from TROPOMI. There are no other back-trajectories for comparison or confirmation with the HCHO spatial map, since it is not measured by the AQ stations.

Overall, since TROPOMI utilizes total column concentrations (despite most NO_2 being located below the BLH), it was concluded that back-trajectory methodologies from AQ stations were more reliable at deducing spatial variations, given the AQ stations produce reliable, ground-level measurements in Toronto, over a significant period (beginning as early as June 1, 1986 for one ozone measurement). This is especially observed for NO_2 , where the greatest concentrations are present about Highway 401 (Toronto's busiest highway), Downsview Airport, York University power plant, Pearson Airport, downtown Toronto, and the Don-Valley Parkway (DVP), as expected from vehicular and industrial sources. Elevated levels of CO over Downsview Airport, York University power plant, Highway 401, and Pearson Airport are expected as well, and confirm the AQ station back-trajectory validity. Low concentrations of CO present over downtown Toronto in TROPOMI was in accord with the downtown areas for the AQ station spatial maps. Anti-relationships between O_3 with NO, NO_2 , and NO_x are anticipated in a VOC-limited regime, consistent with Geddes et al. (2009), Whaley et al. (2015), and Zhang et al. (2022). This can be observed with the AQ stations maps, but not for the maps generated using TROPOMI.

Regarding the socioeconomic analysis carried out, the major conclusions were an inverse relationship between the NO, NO_x and CO with income and population density, and positive relationship regarding ozone. This was because the low-income neighborhoods were predominately located in the northern half of Toronto, and the high income neighborhoods mainly reside in the southern half of Toronto (see Figure 4.6). NO, NO_x and CO tend to be the lowest in south Toronto, and the greatest in north Toronto (especially over Downsview Airport, and York University power plant for CO NO and NO_x , and Highway 401 for NO_x). Given that particulate matters and SO_2 appear to primarily originate from beyond the city, it is difficult to interpret those results from the air-quality station back-trajectories.

8 Future Work

A more comprehensive method to gather data for the back-trajectory analysis is to place co-located anemometers at the air-quality station locations. Therefore, the winds would be exact instead of estimated from kilometers away in some locations. More air quality and automatic weather stations could be utilized to possibly extend this analysis beyond Toronto, and cover a greater portion of southern Ontario. In this way, observing long-range transport to and from other locations may be possible, potentially further exploring the origins of particulate matter and sulfur dioxide. This analysis can also specifically focus on other cities and their surrounding areas. It would be interesting to observe how the spatial variations in Toronto compare to other cities and the contribution of socioeconomic factors given each urban area's unique design, layout, and infrastructure.

In the interest of studying long-range transport, HYSPLIT can also be an asset, especially for validating the results of Jeong et al. (2020), who examined long-range transport of pollutants to Toronto. Extending the spatial coverage of the satellite data and socioeconomic mapping will allow for an analysis regarding socioeconomic status's role in exposure to elevated pollutant concentrations using satellite and ground-level measurements. As demonstrated in this thesis, it can be difficult to discern the role of socioeconomic status, particularly in instances like $PM_{2.5}$ and PM_{10} , where it is probable these sources originate from outside the city, masking the spatial variations within the city. Employing satellite data and comparing this to back-trajectories over a larger area will also provide a more robust validation test for the back-trajectory analysis utilized.

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A Appendix

A.1 Individual CWTs

Individual CWTs are shown in Figures A.1 to A.10.

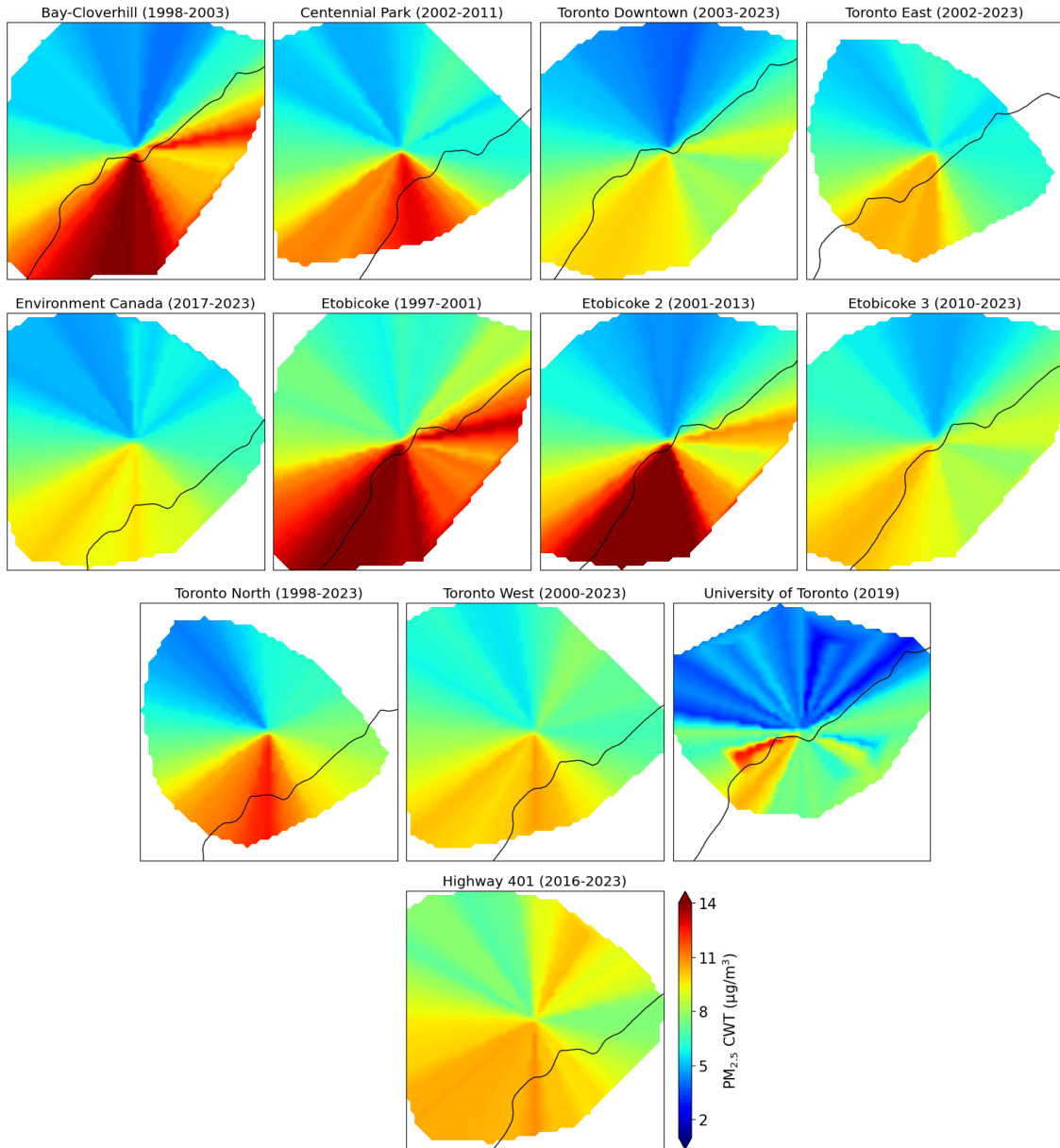


Figure A.1: Air-quality station $PM_{2.5}$ generated CWTs, based on averaged concentration and wind speed for every 10s of degrees wind direction, Displayed on a contour filled plot. Like in chapter 3, in most cases, the starting point is 01:00:00 LST on Jan. 1 of that year, and the ending point 00:00:00 LST on Jan 1. of that year. See Tables A.1, and A.2 for more details.

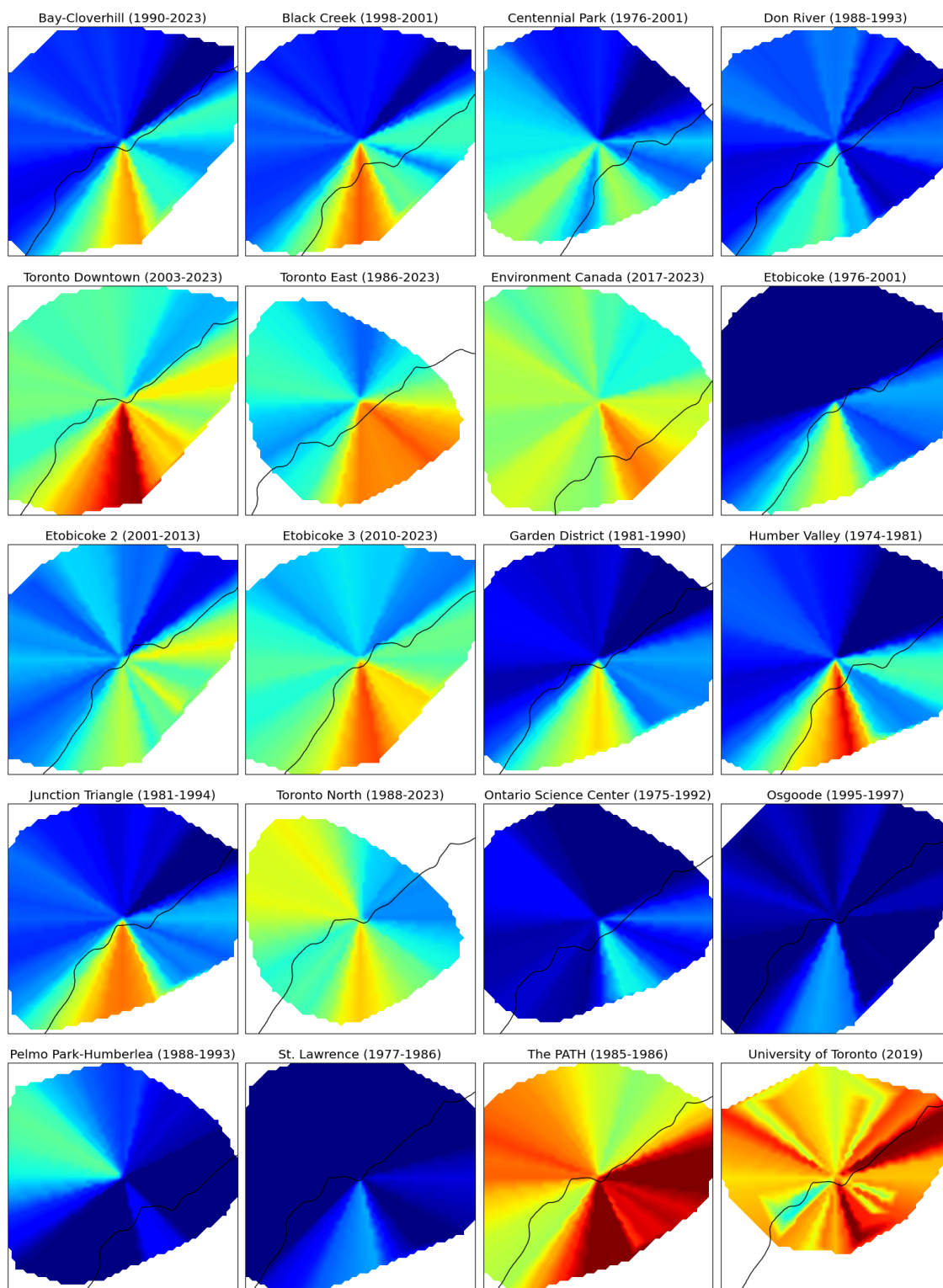


Figure A.2: Same as Figure A.1, but for O_3 . Note that, although O_3 measurements for 'Toronto East' began on Jan. 1, 1974 (see Table A.2), the CWT did not begin until June 1, 1986 since this is when Toronto Buttonville Airport started reporting weather data.

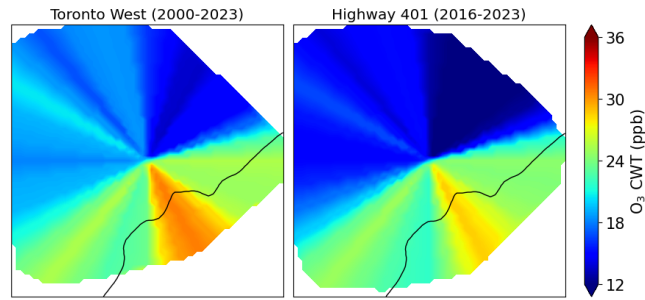


Figure A.3: Additional individual Toronto West (left) and Highway 401 CWTs for Figure A.2.

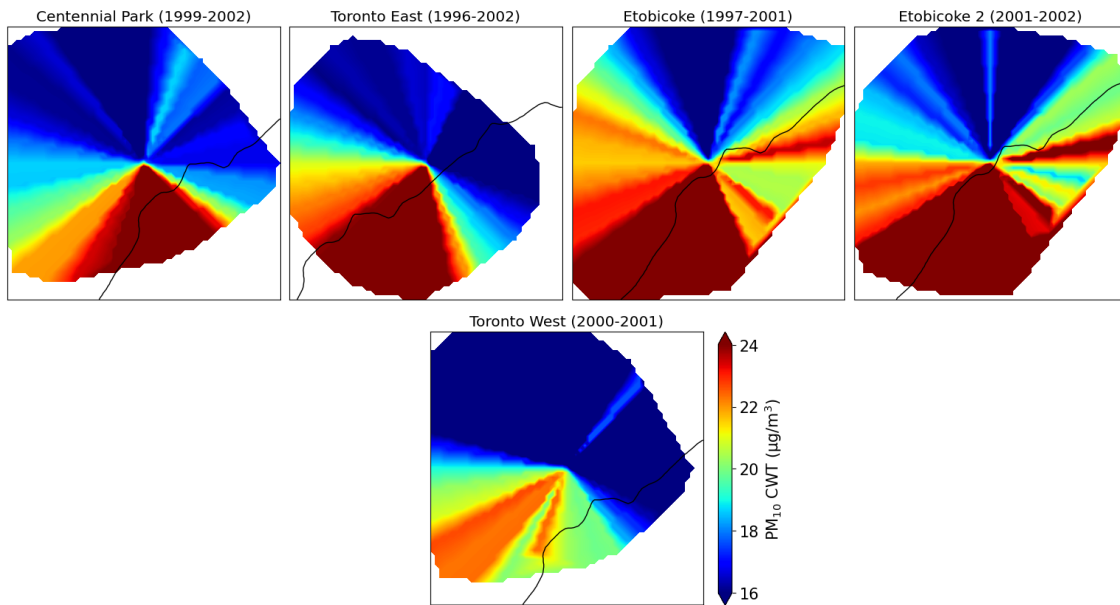


Figure A.4: Same as Figure A.1, but for PM_{10}

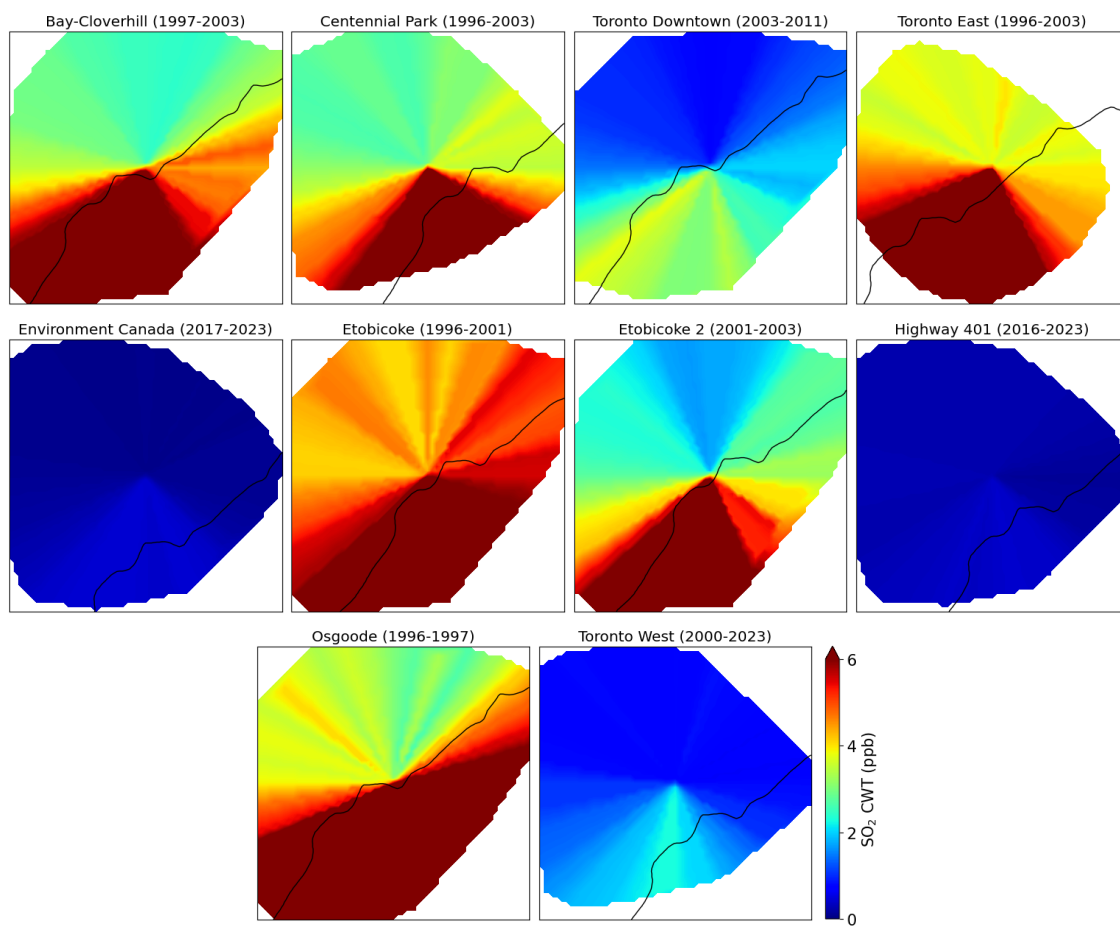


Figure A.5: Same Figure A.2, but for SO_2 .

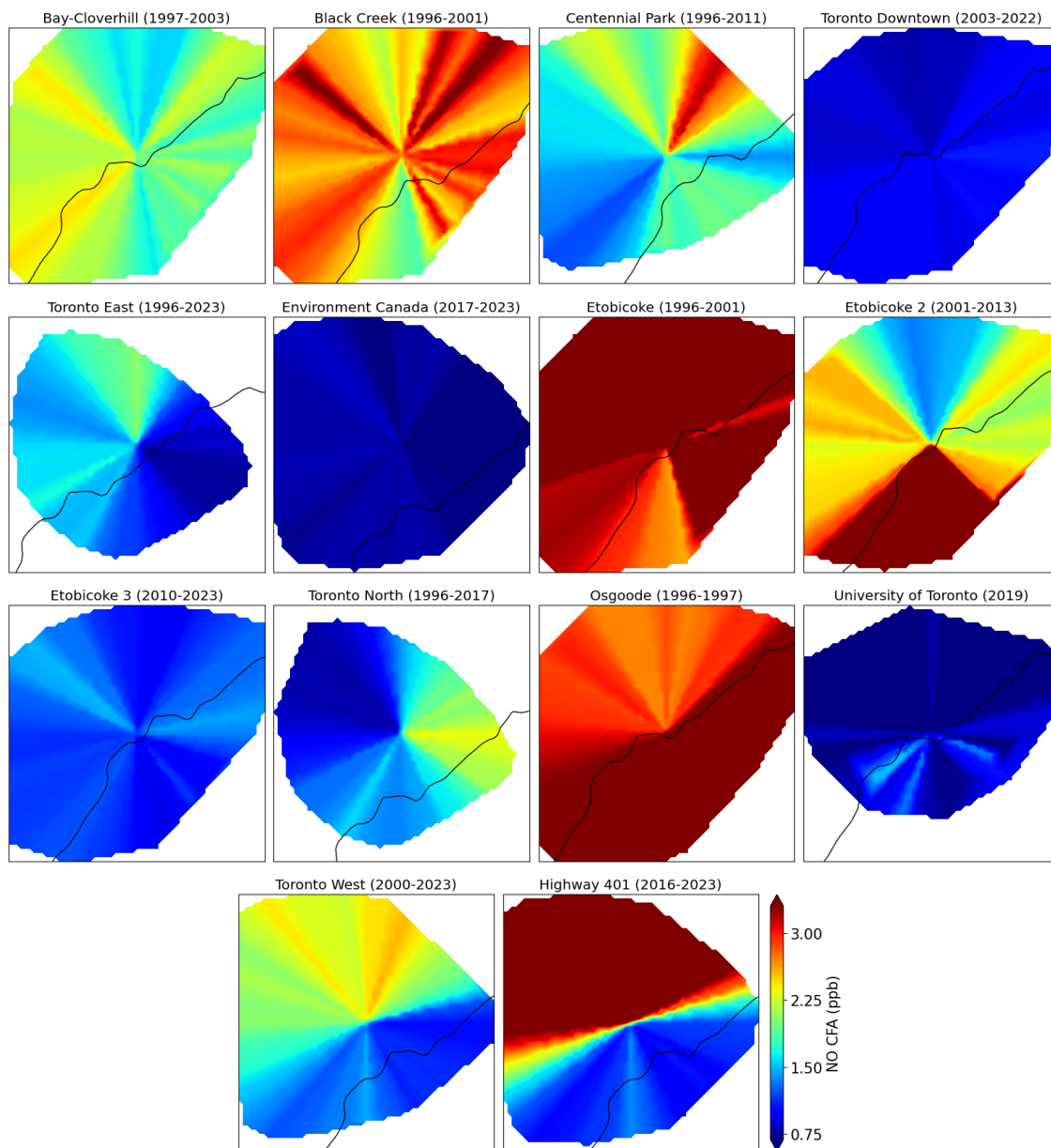


Figure A.6: Just like the previous figures, though pertaining to NO.

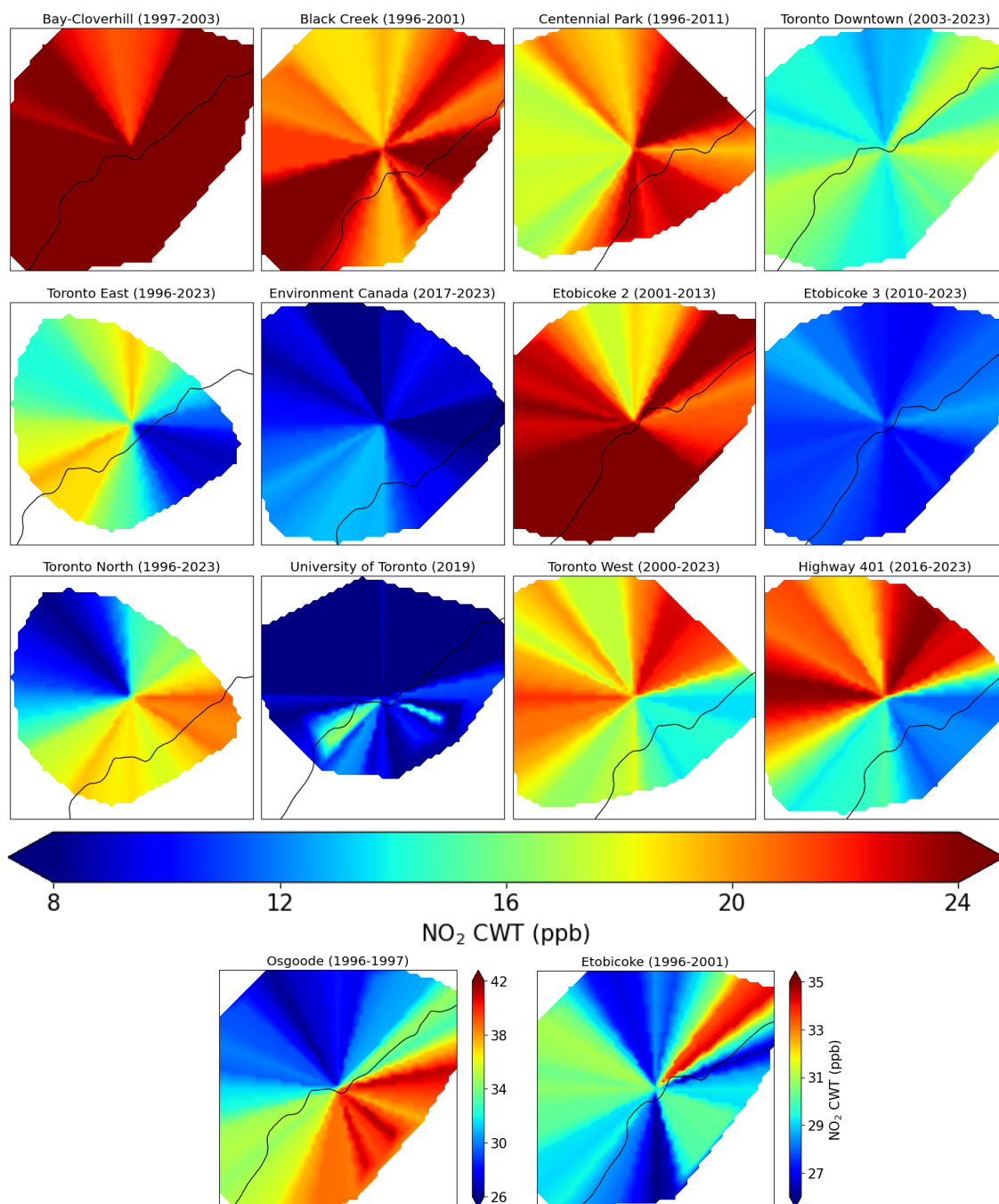


Figure A.7: Air-quality station NO_2 generated CWTs, based on averaged concentration and wind speed for every 10s of degrees wind direction. Note that the Osgoode and Etobicoke CWTs are on different scales.

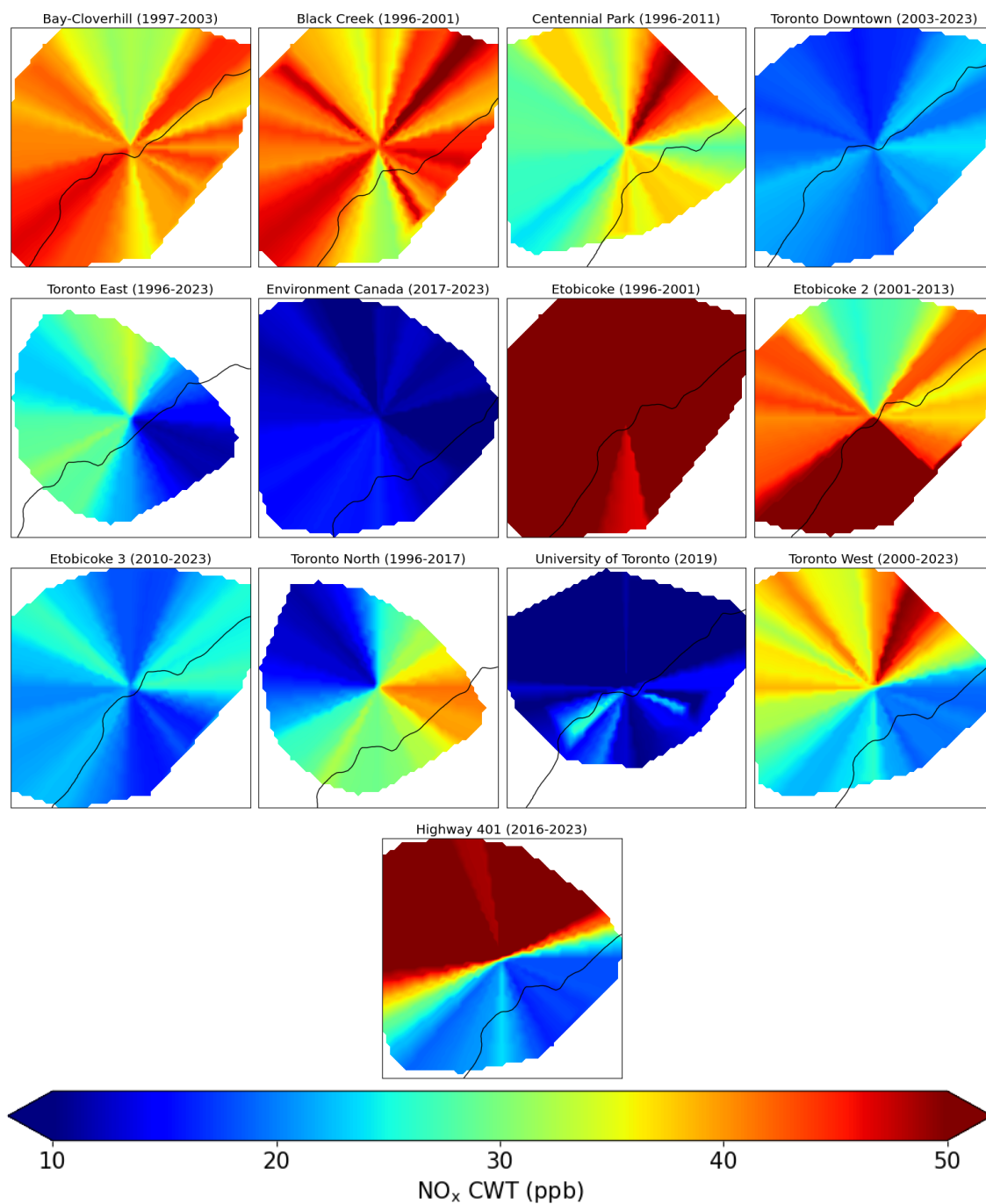


Figure A.8: Like the previous figures, but for individual NO_x CWTs

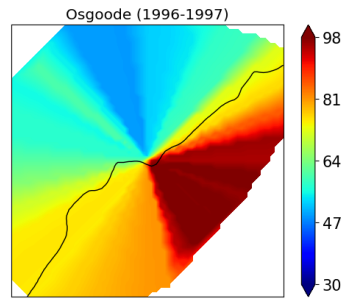


Figure A.9: Additional Osgoode CWT from Figure A.8, note the colorbar differs.

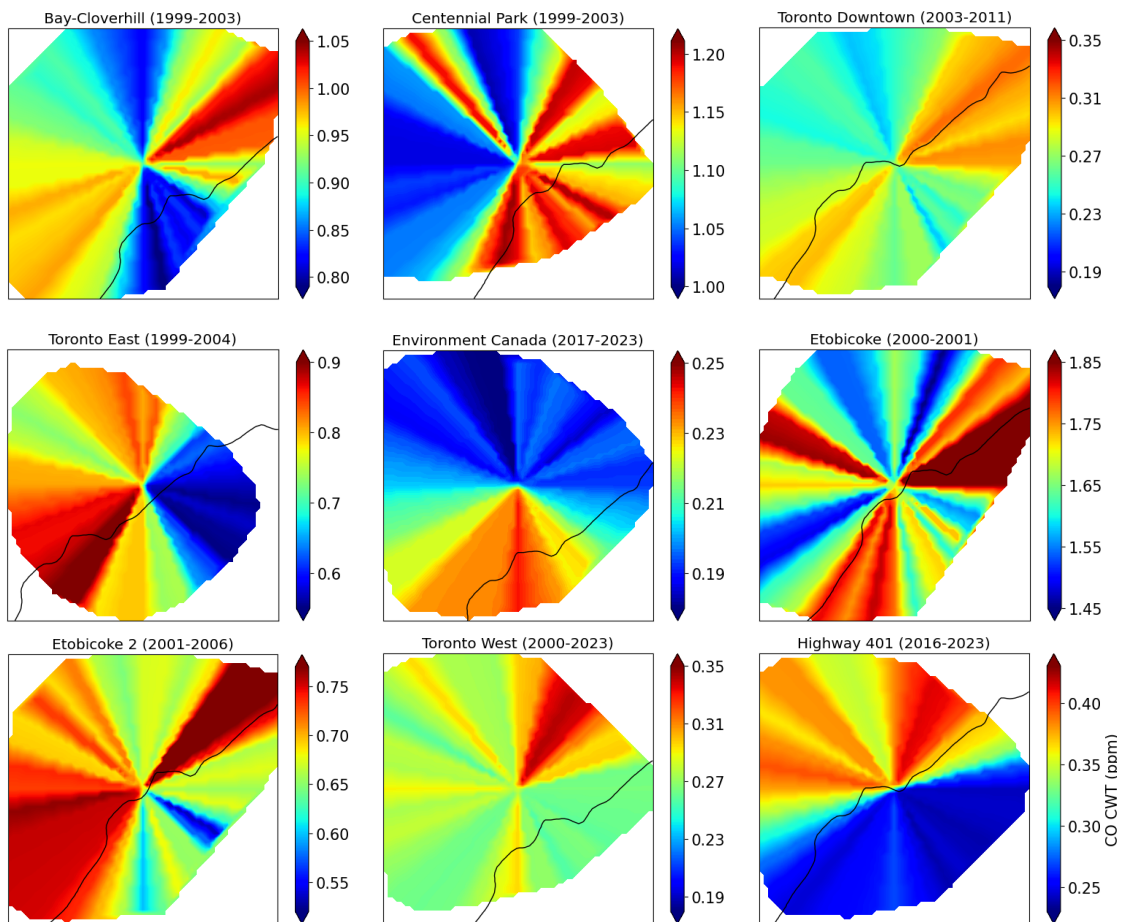


Figure A.10: Like figure A.8, though for individual CO CWTs.

A.2 Individual Back-Trajectory #2 Methods

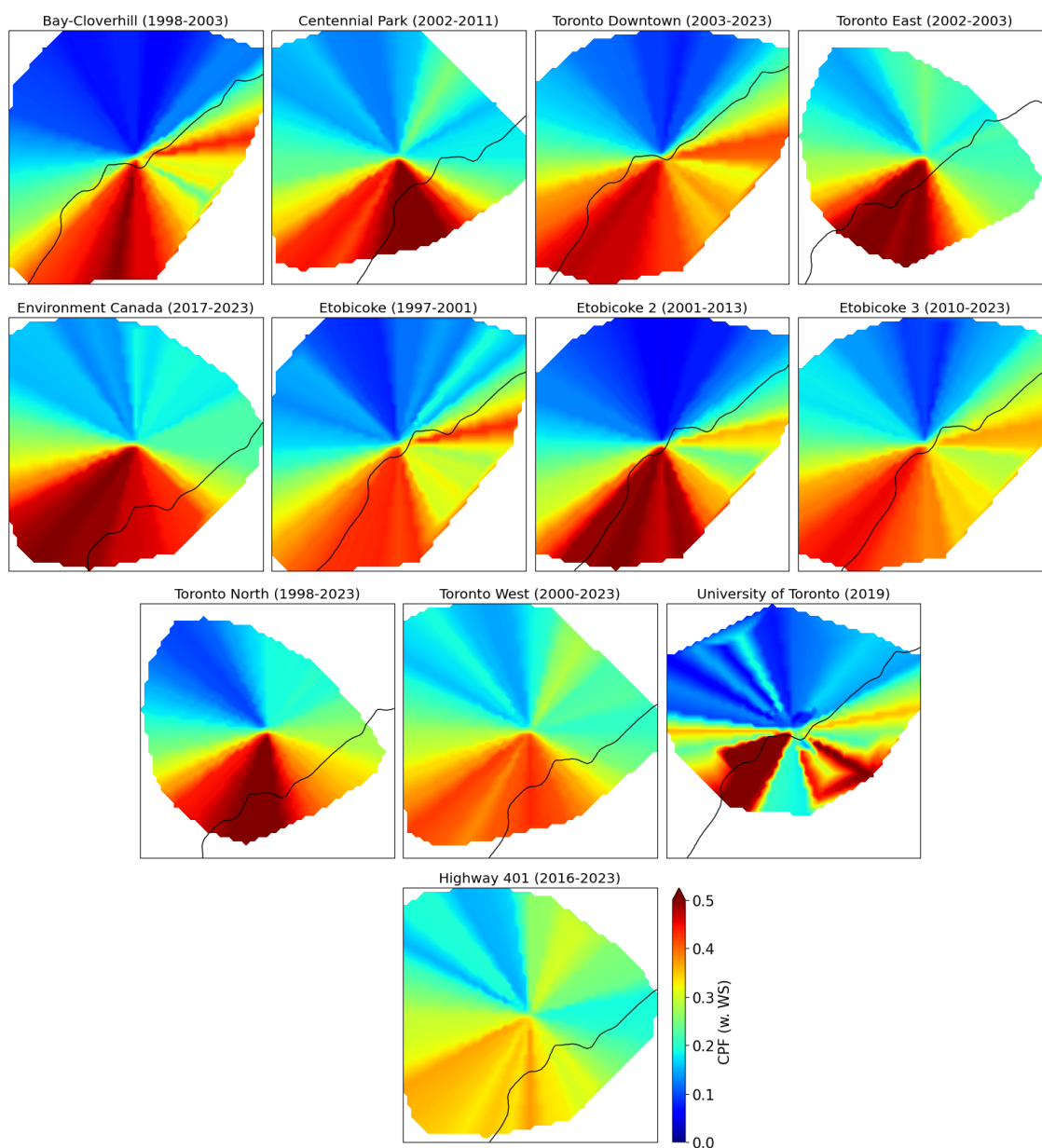


Figure A.11: Like Figure A.1 but for PM_{2.5} BT#3

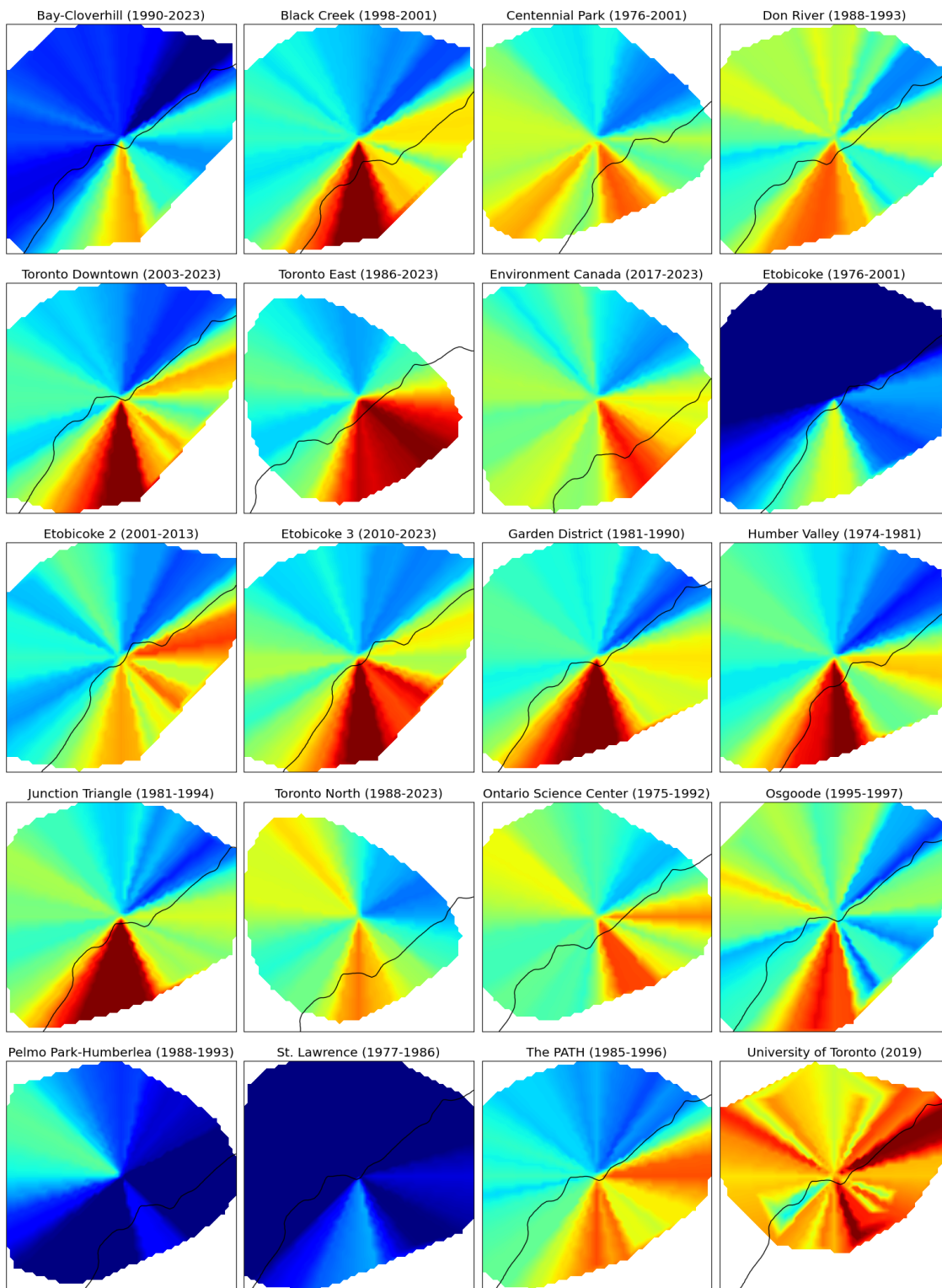


Figure A.12: Like Figure A.2 but for O₃ second back-trajectory.

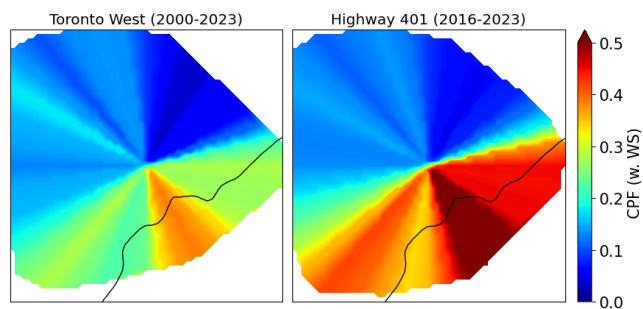


Figure A.13: Additional Toronto West (left) and Highway 401 (right) O₃ second back-trajectory method for Figure A.12.

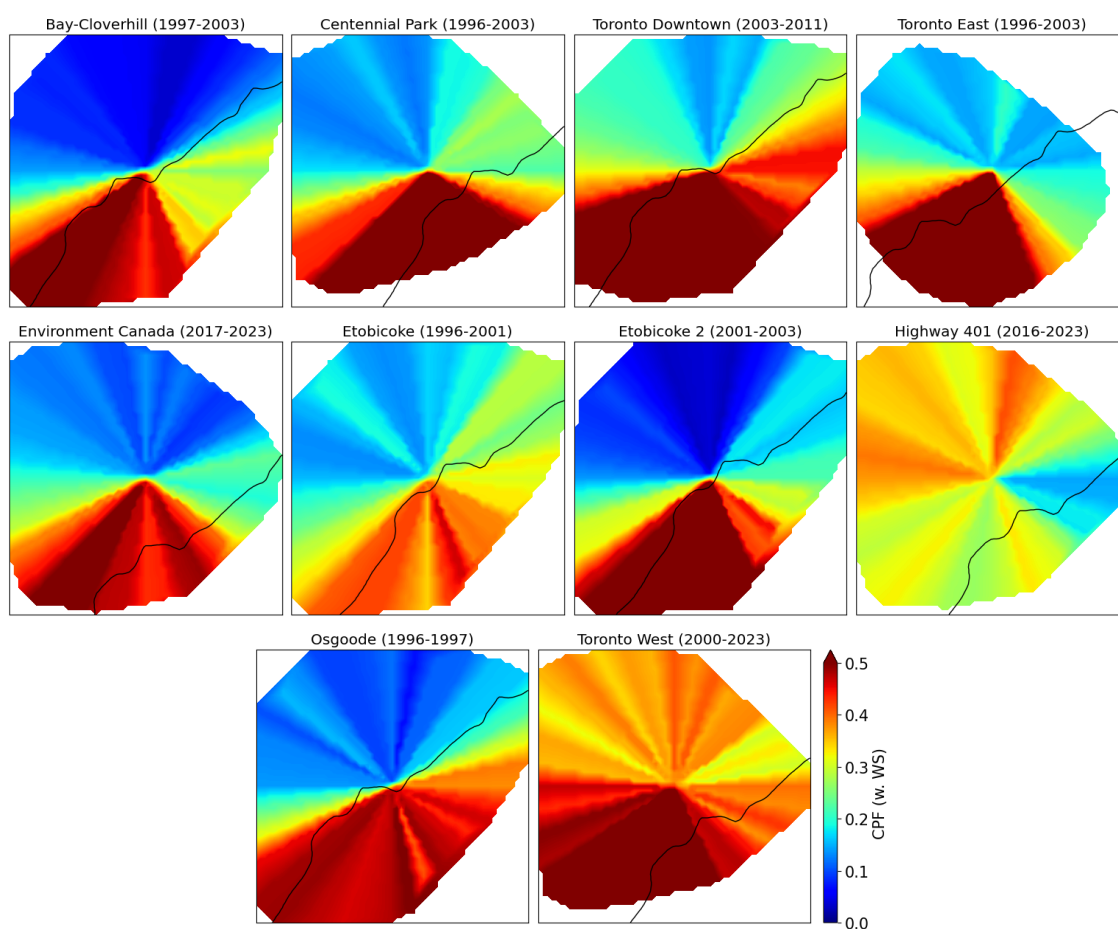


Figure A.14: Like Figure A.5, but for SO₂ CPF w. WS.

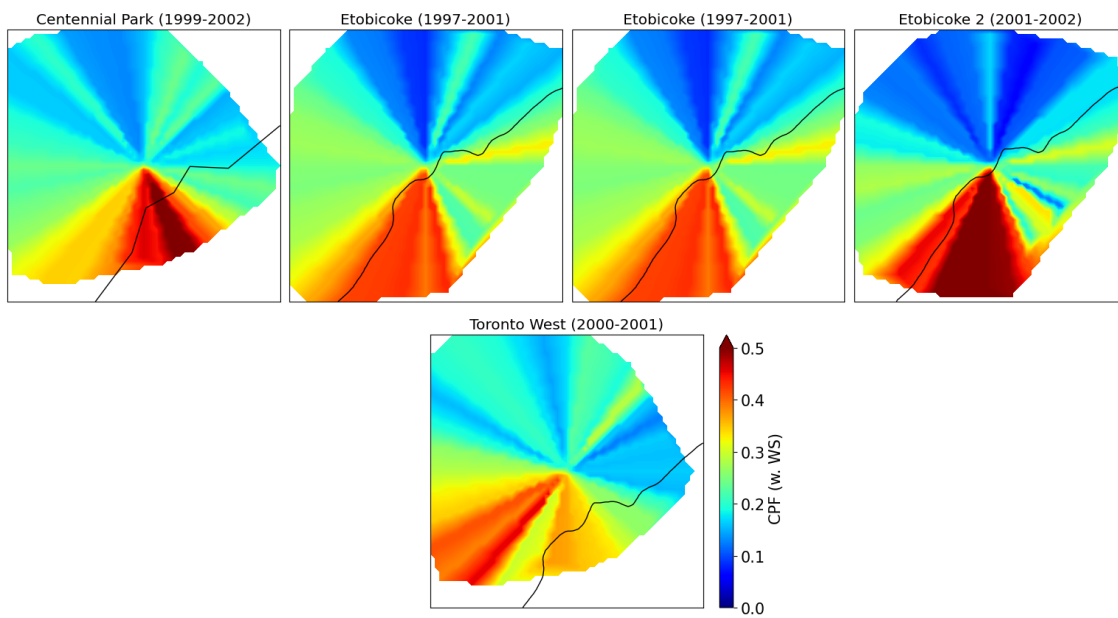


Figure A.15: Same as Figure A.4, but regarding the PM₁₀ CPF w. WS.

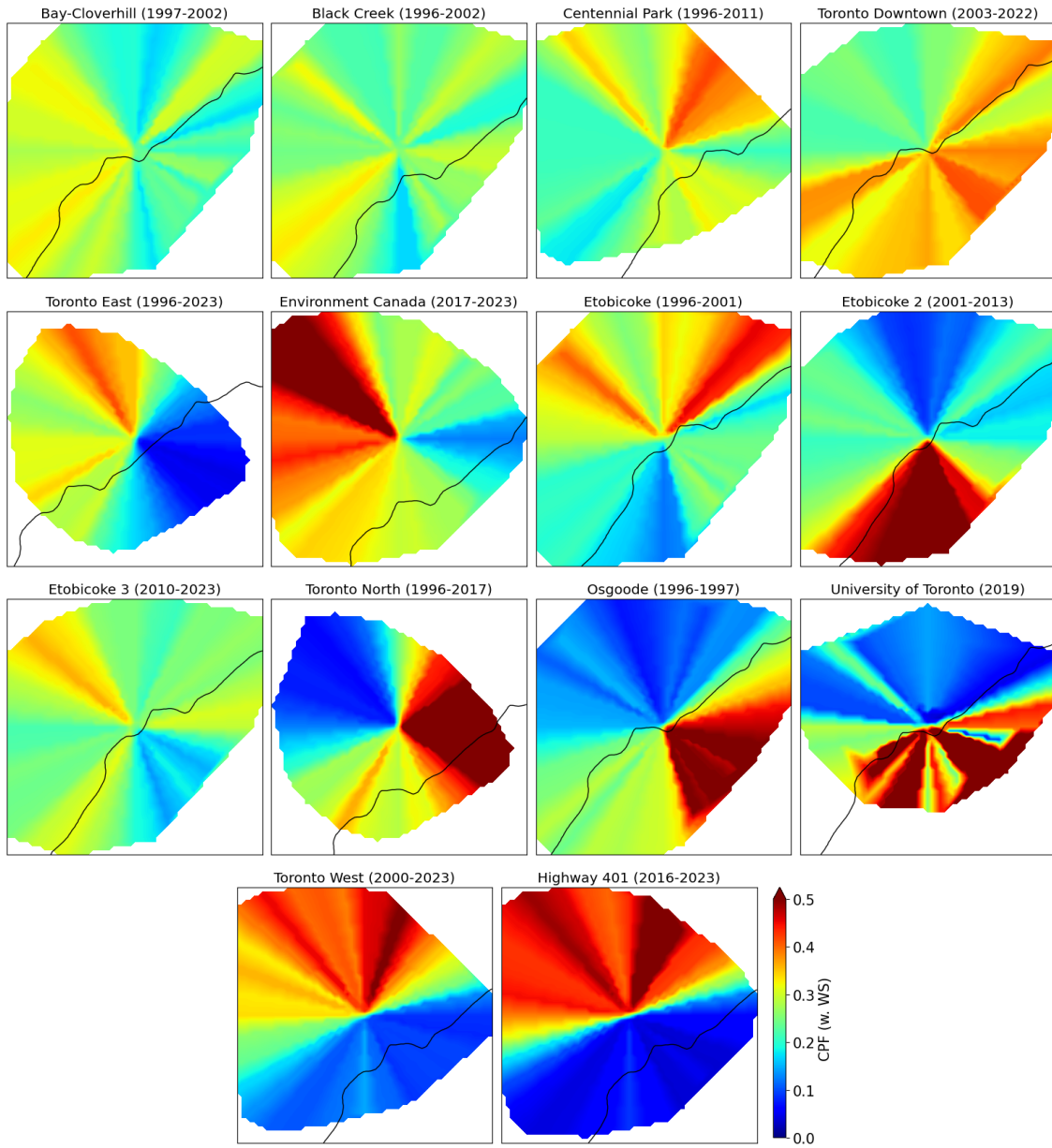


Figure A.16: Like Figure A.7but for NO CPF w. WS

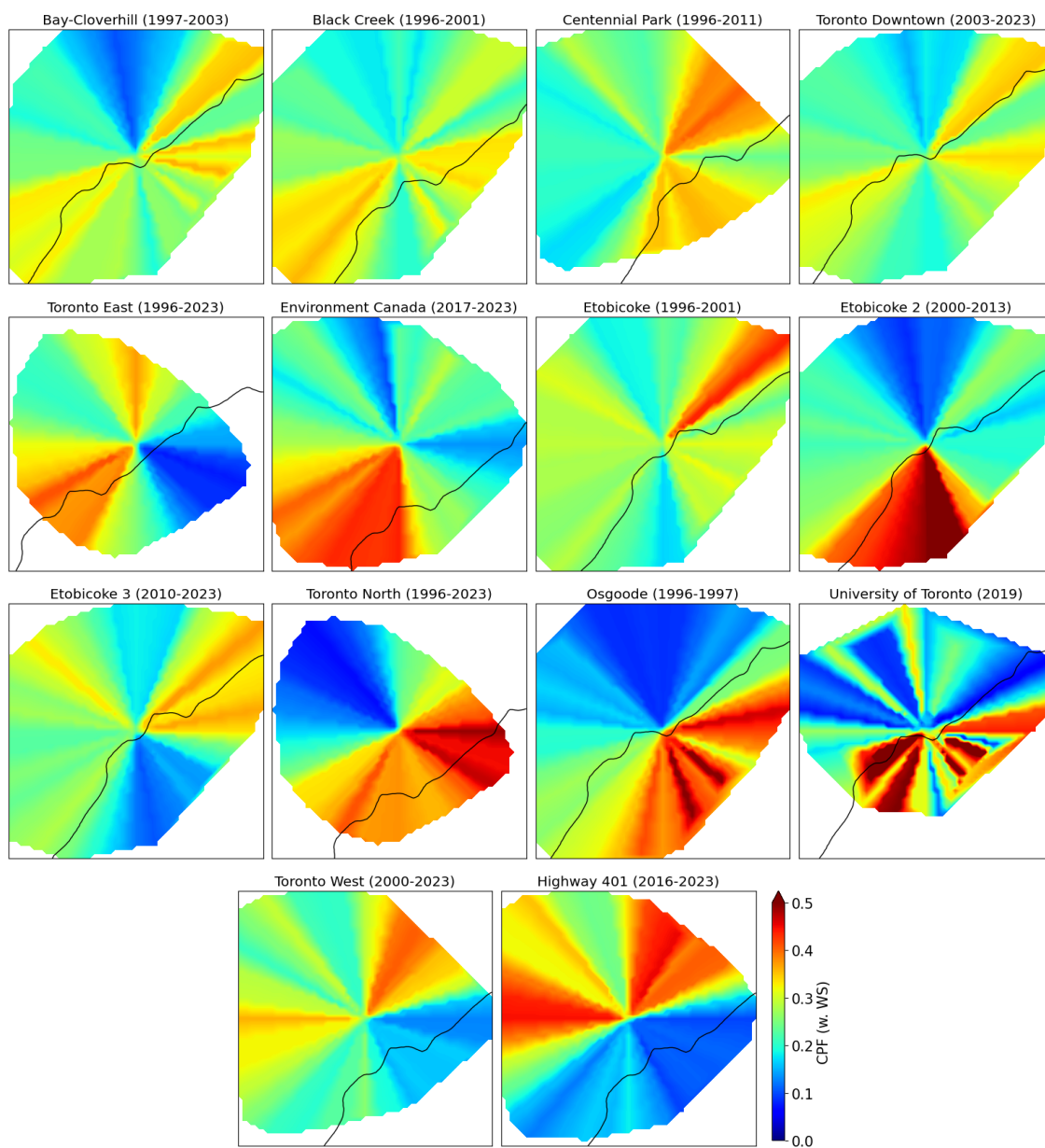


Figure A.17: Like Figure but for NO₂ third back trajectory method.

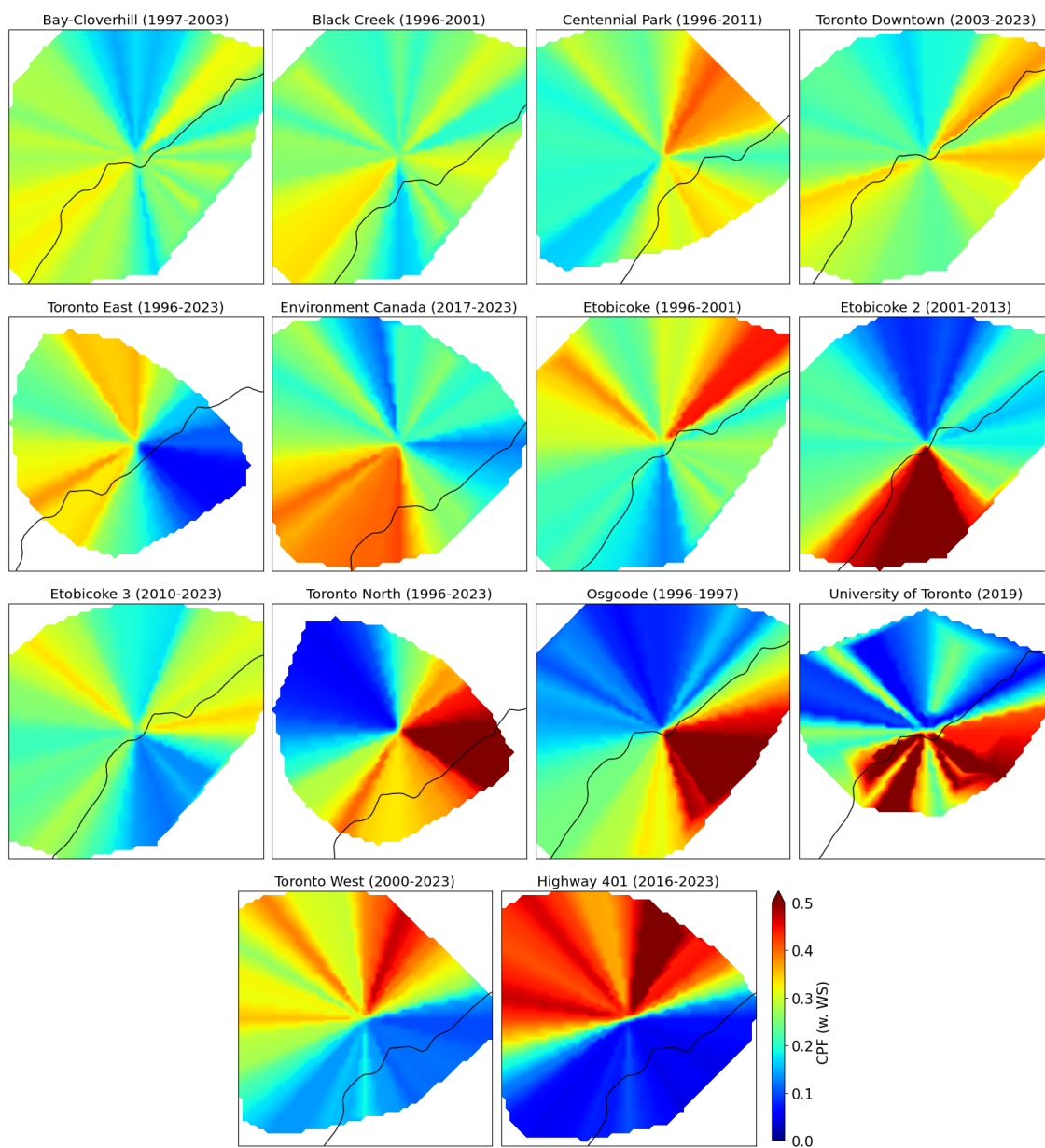


Figure A.18: Like Figure A.8, but for NO_x CPF w. WS.

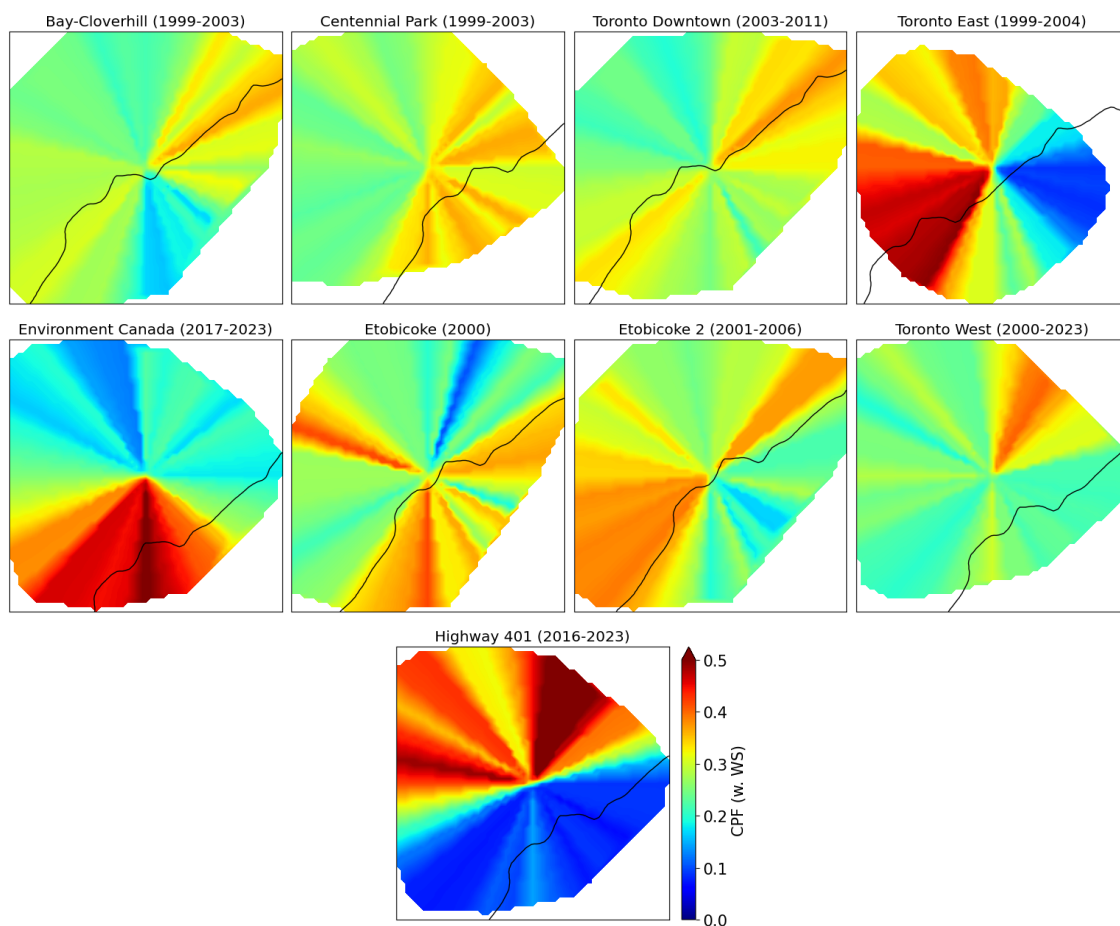


Figure A.19: Like Figure A.10 but regarding CO BT#3.

A.3 Pollutant Measurement Timeframes by AQ Station

Table A.1: Start and end points (LST, not adjusted for daylight savings), for the PM_{2.5}, PM₁₀, CO and SO₂ air-quality station measurements used.

Station Name	PM _{2.5} Timeframe	PM ₁₀ Timeframe	CO Timeframe	SO ₂ Timeframe
Bay-Cloverhill	1/1/1998 01:00 - 12/31/2023 00:00		1/1/1999 01:00 - 12/31/2003 00:00	8/15/1997 19:00 - 1/1/2003 00:00
Centennial Park	1/1/2002 01:00 - 1/1/2011 00:00	1/1/1999 01:00 - 1/1/2002 00:00	1/1/1999 01:00 - 1/1/2003 00:00	1/1/1996 01:00 - 1/1/2003 00:00
Toronto Downtown	1/1/2003 01:00 - 1/1/2023 00:00		1/1/2003 01:00 - 1/1/2011 00:00	1/1/2003 01:00 - 1/1/2011 00:00
Toronto East	1/9/2002 12:00 - 1/1/2023 00:00	11/22/1996 16:00 - 1/1/2002 00:00	1/9/1999 01:00 - 1/1/2003 10:00	1/9/1996 01:00 - 12/3/2023 15:00
Environment Canada	1/1/2017 01:00 - 1/1/2023 00:00		1/1/2017 01:00 - 1/1/2023 00:00	1/1/2017 01:00 - 1/1/2023 00:00
Etobicoke	1/1/1997 01:00 - 2/12/2001 12:00	1/22/1997 18:00 - 2/09/2001 13:00	1/1/2000 01:00 - 2/12/2001 00:00	1/1/1996 01:00 - 2/12/2001 09:00
Etobicoke 2	3/19/2001 13:00 - 1/1/2013 00:00	1/1/2001 01:00 - 1/1/2002 00:00	2/27/2000 01:00 - 1/1/2006 00:00	2/27/2001 11:00 - 10/9/2003 00:00
Etobicoke 3	1/1/2010 01:00 - 1/1/2023 00:00			
Toronto North	1/1/1998 01:00 - 1/1/2023 00:00			
Osgoode				1/1/1996 01:00 - 8/14/1997 07:00
Toronto West	1/1/2000 01:00 - 1/1/2023 00:00	1/1/2000 01:00 - 1/1/2001 00:00	1/1/2003 16:00 - 1/1/2023 00:00	9/1/2000 01:00 - 1/1/2023 00:00
University of Toronto	4/30/2019 01:00 - 6/21/2019 00:00			
Highway 401	1/1/2016 01:00 - 1/1/2023 00:00		1/1/2016 01:00 - 1/1/2023 00:00	1/1/2016 01:00 - 1/1/2023 00:00

Table A.2: Same as Table , though regarding NO, NO₂, NO_x and O₃

Station Name	NO/NO ₂ /NO _x Timeframe	O ₃ Timeframe
Bay-Cloverhill	8/15/1997 18:00 - 1/1/2003 00:00	10/18/1990 07:00 - 1/1/2003 00:00
Black Creek	1/1/1996 01:00 - 1/1/2002 00:00	6/1/1988 01:00 - 1/1/2001 00:00
Centennial Park	1/1/1996 01:00 - 1/1/2011 00:00	1/1/1976 01:00 - 1/1/2011 00:00
Don River		6/1/1988 01:00 - 4/16/1994 11:00
Toronto Downtown	1/1/2003 01:00 - 1/1/2023 00:00*	1/1/2003 01:00 - 1/1/2023 00:00
Toronto East	1/1/1996 12:00 - 1/1/2023 00:00	1/1/1974 00:00 - 1/1/2023 00:00
Environment Canada	1/1/2017 01:00 - 1/1/2023 00:00	1/1/2017 01:00 - 1/1/2023 00:00
Etobicoke	1/1/1997 01:00 - 2/12/2001 09:00	2/9/1976 13:00 - 2/12/2001 09:00
Etobicoke 2	2/27/2001 11:00 - 1/1/2013 00:00	2/27/2001 11:00 - 1/1/2013 00:00
Etobicoke 3	1/1/2010 01:00 - 1/1/2023 00:00	1/1/2010 01:00 - 1/1/2023 00:00
Garden District		6/15/1981 15:00 - 10/15/1990 10:00
Humber Valley		1/1/1974 01:00 - 1/1/1981 10:00
Junction Triangle		3/20/1981 13:00 - 6/11/1994 08:00
Toronto North	1/1/1996 01:00 - 1/1/2017 00:00	6/1/1988 01:00 - 1/1/2023 00:00
Ontario Science Center		1/1/1975 01:00 - 3/3/1992 13:00
Osgoode	1/1/1996 01:00 - 8/14/1997 06:00	11/10/1995 11:00 - 8/14/1997 07:00
Pelmo-Park Humberlea		6/1/1988 01:00 - 4/16/1993 05:00
Toronto West**	9/1/2000 01:00 - 1/1/2017 00:00	9/1/2000 00:00 - 1/1/2023 00:00
The PATH		8/1/1985 01:00 - 1/1/1996 00:00

St. Lawrence		11/01/1977 01:00 - 3/25/1986 11:00
University of Toronto	4/30/2019 01:00 - 6/21/2019 00:00	4/30/2019 01:00 - 6/21/2019 00:00
Highway 401	1/1/2016 01:00 - 1/1/2023 00:00	1/1/2016 01:00 - 1/1/2023 00:00

*NO ends at 1/1/2022 00:00 LST

**NO₂ ends at 1/1/2023 00:00 LST.