

Comparison of methods for resolving the contributions of local emissions to measured concentrations

Taylor D. Edwards¹, Yee Ka Wong¹, Cheol-Heon Jeong¹, Jonathan M. Wang¹, Yushan Su², and Greg J. Evans¹

¹Department of Chemical Engineering and Applied Chemistry, University of Toronto, Wallberg Memorial Building, 184 College St., Toronto, Ontario, Canada

²Environmental Monitoring and Reporting Branch, Ontario Ministry of Environment, Conservation and Parks, 125 Resources Road, Toronto, Ontario, Canada.

Correspondence to: Greg J. Evans (greg.evans@utoronto.ca)

Abstract. To accurately study the characteristics of an air pollution emitter, it is necessary to isolate the contribution of that emitter to total measured pollution concentrations. A variety of published methods exist to complete this task, like placing measurements upwind the emitter, employing a distant background measurement station, or algorithmic methods that extract a background from the time-series of measured concentrations (e.g., wavelet decomposition). In this study, we measured nitrogen oxides (NO_x), carbon monoxide (CO), carbon dioxide (CO₂), and fine particulate matter (PM_{2.5}) at four sites spanning Toronto, Ontario, Canada. We first characterized the spatial variability of background concentrations across the city and then tested the accuracy of seven different algorithmic methods of estimating true measured upwind-of-emitter backgrounds near Toronto’s Highway 401 by using the data collected at a downwind site. These methods included time-series and regression methods, including machine learning (XGBoost). We observed background concentrations had notable spatial variability, except for PM_{2.5}. When predicting backgrounds upwind the highway, we found a distant measurement station provided an accurate background only during some times of day and was least accurate during rush hours. When testing algorithmic predictions of upwind-of-highway backgrounds, we found that regression models surpassed the performance of time-series methods, with best predictions having R^2 exceeding 0.8 for all four pollutants. Despite the better performance of regression models, time-series methods still provided reasonable estimates. We also found that emitter-specific covariates (e.g. traffic counts, onsite dispersion modelling) did not play an important role in regressions, suggesting backgrounds can be well-characterized by time of day, meteorology, and distant measurement stations. Based on our results, we provide ranked recommendations for choosing background estimation methods. We suggest future air pollution research characterizing individual emitters include careful consideration of how background concentrations are estimated.

1 Introduction

Across air pollution literature, there is a common distinction between stationary field measurement sites located well away from any known sources that record *background* pollution concentrations and those that record *local* concentrations, such as

near-road sites, influenced by emissions from nearby “local” sources. Generally, background concentrations are considered to arise from a mix of more distant upwind anthropogenic and natural sources and processes, while local concentrations are impacted by one or more nearby sources of interest. The difference between the concentration of an air pollutant measured at near-source and background sites can be attributed to local emissions. Within this process of apportioning the measured total concentration, the contribution of emissions from nearby sources is referred to as the *local* or *emitted* concentration, while that within air masses arriving from upwind of a measurement site is referred to as the *background* concentration.

Good measures of background concentrations are important for isolating local sources of pollution. Ideal outdoor field measurements would include instruments both up- and down-wind of the source of interest, such that the source’s contribution is the difference between the two. However, this is not always possible: requiring two simultaneous measurements increases instrumentation and operation cost, there may not be an appropriate upwind location to place instruments, and widely varying wind directions might necessitate more than just one upwind-downwind measurement pair. For these reasons, tools for estimating background concentrations (C_{bkg}) without a second measurement site are valuable. With reliable C_{bkg} estimates, researchers can isolate continuous measurements of their sources of interest, which is vital for source attribution and measuring emission rates and emission factors.

If measurements immediately upwind of a source of interest are not available, researchers might utilize either an urban background station or tracer species to isolate contributions from sources of interest. Urban background stations are typically within a few kilometres of the study location but are removed from any major nearby sources. These sites might be located in a park or a nearby rural area. Tracer species are those that are specific to the source of interest – if a researcher knows a measured emissions source is the only major nearby source of a particular species, they can be confident their measured source is the only contributor to measured concentrations of that species.

Unfortunately, both approaches, despite their prevalence in the literature, have limitations. Urban background stations might not be completely isolated from all sources or background concentrations might vary spatially between the urban background station and the study site (particularly in the context of the strict definition of “background concentration” we provide below). For tracer species, in many cases the source of interest cannot be guaranteed to be the only measured contributor. For example, nitrogen oxides (NO_x) are often considered a tracer for traffic emissions, but in a dense urban area measured NO_x concentrations will contain emissions from many different roads, so no single road can be isolated.

Beyond these common approaches, there exist some other methods for estimating background concentrations, particularly for application to continuous time-series measurements of atmospheric pollution. Notable methods include:

- Measuring pollutant concentrations immediately upwind of the source of interest, as mentioned above, such as in highway studies by Zhu et al. (2002), Kohler et al. (2005), and Frey et al. (2022).
- Designating a geographically distinct measurement station as an urban or regional background, with that station typically having few nearby emissions sources (Hicks et al., 2021; Hilker et al., 2019).
- Comparing times when a measurement site is up- and down-wind of a target source (Hilker et al., 2019).

- Identifying a background or apportioning sources via wavelet decomposition (Klems et al., 2010; Sabaliauskas et al., 2014; Wei et al., 2019).
- Wang (2018) and Hilker et al. (2019) employed and tested an iterative algorithm that heuristically estimates a background signal similar to that produced from wavelet decomposition, which is termed as *pseudo-wavelet*. In brief, this method takes a smoothed interpolation of minima in the measured near-source concentration within a moving time window.
- Inverse dispersion modelling, where multiple downwind measurements are paired with a dispersion model estimating downwind concentrations given an emission rate. Inverse dispersion modelling approaches are usually applied to measure emission rates from the source of interest, though concentration upwind of the emitter should be produced as a by-product of this calculation (Fushimi et al., 1997; Olaguer, 2022).
- Clustering algorithms: clustering can identify sources by grouping correlated pollutants, and may not necessarily delineate between local and background sources, however Rodríguez et al. (2024) demonstrated a separation of local and non-local sources using a fuzzy clustering algorithm.
- Geospatial interpolation from urban background stations, which can estimate the spatial variability of background concentrations, such as in Arunachalam et al. (2014).
- Localized iterative regression within a time-series of concentrations to extract a baseline signal, as described by Ruckstuhl et al. (2012). However, this study presented a method to further decompose measurements from a background site, implying a definition of background concentration that is geographically broader than what we consider in this study.

1.1 Defining “background concentration”

To address the limitations of the methods identified above, we propose a definition for *background* that is useful for isolating emissions sources of interest: *background concentrations, C_{bkg} , are the portions of the total measured concentrations that were not emitted from the local emission source of interest.* This definition is similar to the one provided by Arunachalam et al. (2014). With this definition, the total measured concentration, C_{meas} , is strictly a sum of the local concentration, C_{local} , and background concentration, C_{bkg} :

$$C_{meas} = C_{local} + C_{bkg} \quad (1)$$

As a corollary to this definition, C_{local} is only the portion of C_{meas} that was emitted from the source of interest, and thus the local concentration becomes useful for estimating emissions, source characteristics, etc. This definition recognizes that the background concentration may vary across regions such as a city because of the many sources present. At the same time, the background concentration across a city can be relatively homogenous, if much of the background originates from sources or processes well upwind of a city, as is often the case for pollutants such as $PM_{2.5}$ and CO_2 . Ideally, this background concentration

should be measured directly upwind the source of interest, with no interstitial sources. The up- and down-wind measurements should also be near enough to each other and the emissions source that dilution of background concentrations while they travel between the up- and down-wind instruments is not of concern. This is the configuration at the highway field site studied here, which had instruments placed up- and down-wind a major urban highway in Toronto, Canada. While it is desirable for the background site to be as close as possible to the emissions source of interest, the nearer the background site is to the emission source, the greater the potential for emissions from that source to contribute at times to the concentrations measured at the background site. We posit that this definition of background concentration lends itself readily to useful measurements of C_{local} . Accordingly, it is desirable that researchers measuring rates and/or characteristics of emissions sources can estimate C_{bkg} when direct measurement is not possible, as previously discussed.

We note that this definition differs from existing interpretations of *background* in air pollution research, where background might be interpreted as either a minimum or baseline concentration, or as pollution arising from long-range transport from multiple distant sources (Gómez-Losada et al., 2016, 2018). These existing definitions would imply homogeneous and temporally constant concentrations spread across an entire neighbourhood, city, or region. Measuring such a background concentration might require rural measurement, or an urban measurement isolated from any single source. In our case, we are interested in measuring C_{bkg} for the purpose of extracting C_{local} , so emissions from sources other than the targeted emitter are only a problem if they are so nearby as to render the measurement of C_{bkg} obviously unusable.

1.2 Study outline and objectives

In this study we tested the accuracy of a variety of methods for estimating background concentration at a field site adjacent a large roadway emissions source. We first qualitatively examined how background concentrations varied across an urban area (section 3.1). We then tested the accuracy of seven algorithms for predicting background concentrations at the near-road site (section 3.2). The algorithmic methods were differentiated into two classes: *frequency methods* used the time-series nature of C_{meas} to predicted C_{bkg} , on the theoretical basis that background concentrations vary on a longer temporal scale than a nearby source, and that $C_{bkg} = C_{meas}$ at least occasionally. *Regression methods* were those that incorporated additional covariates measured or estimated at the study site and were regressed to the measured upwind background concentrations. We evaluated the accuracy of each algorithmic estimate of background concentration by temporarily deploying a low-cost air pollution sensor platform to the upwind side of the tested highway site. Finally, we evaluated the relative importance of regression model covariates in estimating background concentrations (section 3.3 and 3.4), and considered limitations (section 3.5)

This study was completed as part of the larger Study of Winter Air Pollution in Toronto (SWAPIT) campaign, a collaborative effort between the academic, government, and private institutions in the Toronto, Ontario region.

125 **2 Methodology**

2.1 Field measurements

 We gathered field measurements at four sites throughout Toronto, Ontario, Canada, from 2023-11-23 to 2024-04-12, totalling just over 141 days of measurements. All measurements occurred during winter and early spring conditions in Toronto when photosynthesis of CO₂ is minimal. The next two sections describe the sampling sites and instruments.

130 **2.1.1 Site descriptions**

 The primary highway field site was located adjacent a stretch of Toronto’s Highway 401 located at UTM 617300 m E 4840900 m N 17N (*see A in Figure 1, top; Figure 1 bottom*). This stretch of highway is one of the busiest in North America, with over 400,000 annual average daily traffic (AADT) as reported by the Ontario Ministry of Transportation (2016). It is 17 lanes and 113 m wide adjacent to the measurement sites, and runs in a primarily west-east direction, offset 18° towards a southwest-northeast direction. This site included two instrument locations: the first was a permanent roadside station on the south side of the highway that was frequently downwind the road. The second location was a background sensor placed north of the highway, which was frequently upwind the road. The north site was designated as the background site based on predominant wind directions and the fact that this site featured a temporarily deployed low-cost sensor platform, while the south site features a permanent air quality station operated by the Ontario Ministry of the Environment, Conservation and Parks. Figure 1 maps this and the remaining study sites.

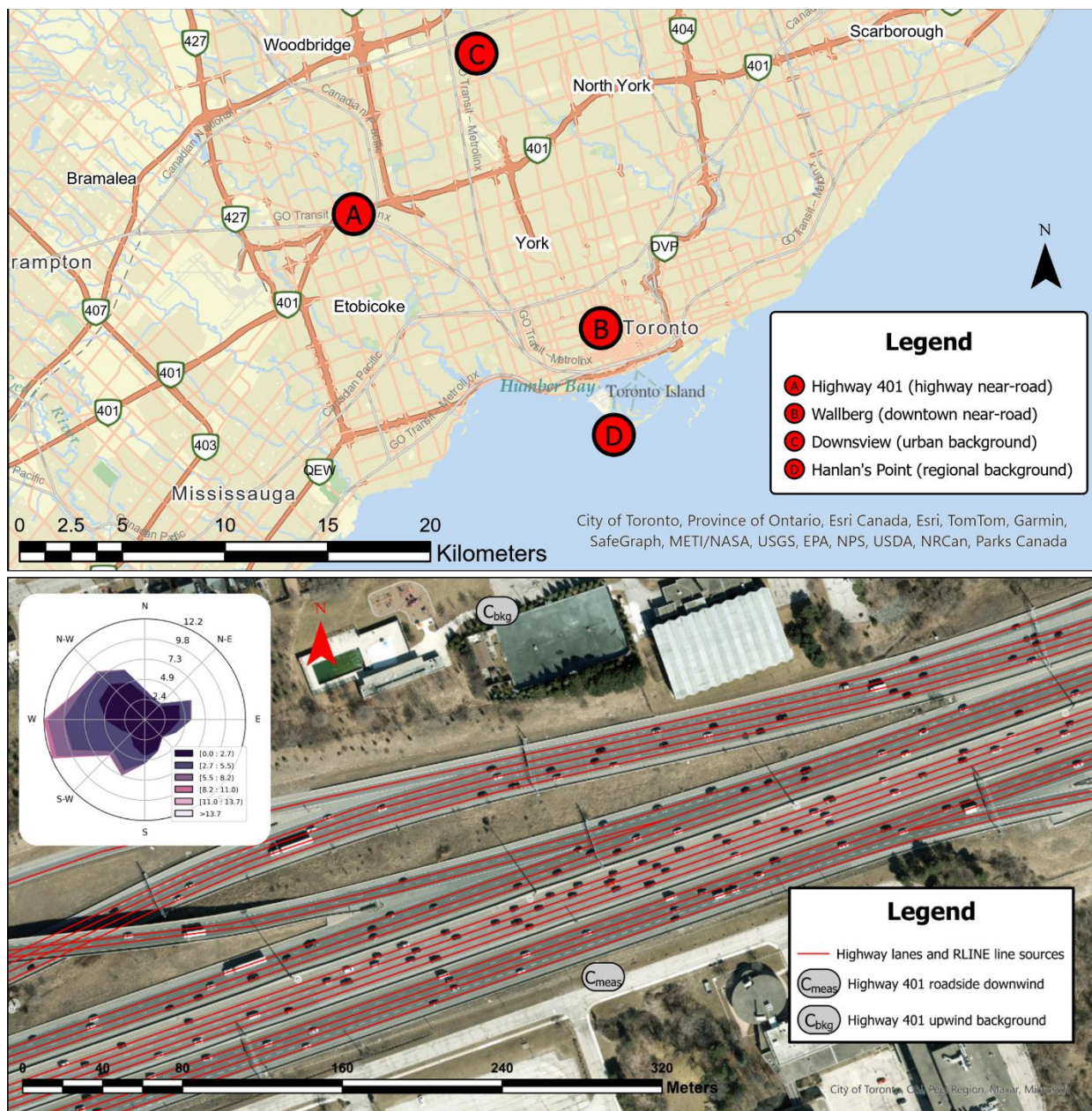


Figure 1: Top: locations of measurement sites throughout Toronto region. Bottom: detailed map of the Highway 401 field study site. Inset bottom: wind rose measured at Highway 401 roadside (downwind) station during the study period. Throughout this document, the Highway 401 downwind roadside station is referred to as “highway roadside downwind” or “highway downwind”, and the Highway 401 upwind background site is referred to as “highway upwind background” or “highway upwind”.

In addition to the primary highway site, we recorded pollution concentrations at three additional sites throughout the Toronto area. The first site was the Wallberg urban near-road site, located at the University of Toronto’s Wallberg Memorial Building at UTM 629381 m E 4835252 m N 17N (Site B in Figure 1). This site features a similar set of air pollution instruments as the

permanent Highway 401 downwind site and was located 15 m from a major urban road and 40 m from an intersection. The remaining two sites were designated as distant urban background sites, not near any emissions sources of comparable magnitude to Highway 401. The first urban background site was Downsview, located at UTM 623330 m E 4848631 m N 17N (Site C in Figure 1). This site is in a green space near an office building and is about 175 m from the nearest road. The final site was the Hanlan’s Point urban background station, located at UTM 630025 m E 4830061 m N 17N (Site D in Figure 1). This site is located on an island in Lake Ontario, south of Toronto’s downtown core. The Hanlan’s Point site is isolated from any nearby sources, with the only notable emissions source being a regional airport over a kilometre to the north. Measurements were collected during winter to early spring, so we expect green space near background sites to have a minimal CO₂-sink effect.

All sites listed here except the highway upwind background site were equipped with a similar set of air contaminant instruments, detailed in the next section.

2.1.2 Airborne pollutants, traffic, and meteorology

We employed a variety of instruments to measure air pollutant concentrations, meteorology, and traffic counts. The instruments deployed at each site except the highway upwind background are listed in Table 1. We selected NO_x (NO + NO₂), CO, PM_{2.5}, and CO₂ to cover a range of dominant sources: we expect PM_{2.5} and CO₂ to have large regional background concentrations while CO and NO_x are more sensitive to proximity to sources. For PM_{2.5}, given the dominance of regional transport and secondary formation, and the consequential homogeneity of this pollutant’s concentration across urban areas, we expect that differentiating between local and background pollution might be difficult. However, we retained PM_{2.5} to serve as a counterexample to the other pollutants, which have greater differences between local and background concentrations.

Table 1. Air pollution, meteorology, and traffic count instruments deployed at each measurement site except the highway upwind background site.

Measurand	Symbol	Method	Instrument name	Manufacturer
Nitrogen oxides	NO, NO ₂ , NO _x	Chemiluminescence	42i	Thermo Fisher
Carbon monoxide	CO	Infrared absorbance	48i	
Fine particulate matter	PM _{2.5}	Nephelometry and beta attenuation	5030(i) SHARP**	
		Spectrometry	T640**	Teledyne API
Carbon dioxide	CO ₂	Non-dispersive infrared	LI-840A	LI-COR Biosciences
Onsite meteorology	T, P, RH, u, θ	Various	WXT520	Vaisala
Traffic counts*	N_{LDV}, N_{MHDV}	Radar	Smartsensor 125HD	Wavetronix

*Traffic counts were only recorded at the Highway 401 downwind site, and only for the nearest eight lanes. LDV = light duty vehicles, MHDV = medium and heavy duty vehicles.

**PM_{2.5} at the Hanlan’s Point background station was measured with a Teledyne API T640 while other sites used the Thermo Fisher 5030 or 5030i SHARP.

175 We acquired additional micrometeorological measurements for dispersion models from various sources, which we detail in the appendices; we used dispersion model outputs as exogenous variables for regression methods. At the Highway 401 north background site, we deployed a low-cost AirSENCE air pollution measurement system (AUG Signals, Toronto, Canada). This system hosts a variety of low-cost sensor systems to simultaneously measure a variety of pollutants, including the pollutants tested here. Morris et al. (2020) has previously explored the performance of the AirSENCE system.

180 For $PM_{2.5}$ at the Hanlan's Point site, we collected concentrations measured with the Teledyne API T640 rather than the Thermo Fisher SHARP instrument deployed at each other site (also again excepting the low-cost instrument upwind the highway). Zheng et al. (2018) directly compared two T640s to the same model SHARP used here, and reported variations up to 3 to 5 $\mu g \cdot m^{-3}$ in concentration ranges similar to those typically measured here, with the T640 more often reporting slightly higher concentrations than the SHARP. The possibility that $PM_{2.5}$ measured at Hanlan's Point may be slightly inflated should
185 be kept in mind when reading results that directly compare concentrations across sites. Presumably, the low-cost sensor-based $PM_{2.5}$ we measured north of the highway also deviated from reference instruments by similar or larger amounts, however as explained below, we produced a corrective calibration for the low-cost sensor platform prior to deployment. We also found that when directly comparing hourly $PM_{2.5}$ concentrations between SHARP and T640 instruments across sites used in this study, variation between instruments was similar to variation between sites, suggesting no systematic bias due to instrument
190 differences. Should any disagreement between instruments exist anyways, this should only affect our results in cases where measured concentrations are compared directly – in cases where data were included in regression models, any offset in measured concentration should have a limited impact on regression results, as regression models can account for systematic biases.

We averaged sub-minutely measurements to the nearest minute to allow time-matched comparison across the instruments.
195 To ensure the low-cost AirSENCE instruments reported concentrations comparable with reference instruments, we applied multiple quality control and calibration steps prior to analysis. In particular, we addressed calibration and drift in some of the low-cost sensors through comparison with other sites, and corrected the low-cost $PM_{2.5}$ measurements for hygroscopicity with the correction procedure devised by Crilley et al. (2018). We also placed the AirSENCE device atop the downwind highway station for nearly 18 days at the start of our measurement campaign and used this co-location period to calibrate the
200 AirSENCE's sensors against the station's reference instruments, controlling for interference from humidity, pressure, and temperature. Finally, in some cases for CO and CO₂ to avoid concentration biases between sites due to different instrument calibration schedules, we calculated a 0.1% rolling percentile concentration at each site and set each site's rolling quantile equal. We describe these preprocessing steps in greater detail in Supplemental Appendix B.

Additional information on some of these same sampling sites and instruments can be found in publications by Wang et al.
205 (2018), Hilker et al. (2019), and Jeong et al. (2020); this list is not exhaustive and these sites have been employed in a variety of prior air pollution studies.

2.2 Separating measured local and background concentrations at the highway site

To choose when we could consider the difference between near-road and upwind measurements as local concentrations, C_{local} , we considered the relationship between measured concentrations and wind at the highway site. From Supplemental Figure F.1 we identified which wind directions to subsample from our measurements to isolate local and background signals: we selected periods where wind direction relative to the road was between 80 degrees to the northwest and 40 degrees to the northeast. The asymmetry in downwind directions relative to the road could be explained by traffic-induced turbulence, which can influence bulk air flow above the road (Hashad et al., 2022). Since station south of the highway is nearest an eastbound lane, those lanes might add a westerly component to the observed wind direction. From Supplemental Figure F.2 we also observed that some downwind roadside (C_{meas}) and traffic-related ($C_{local} = C_{meas} - C_{bkg}$) concentrations diverged below wind speeds of about $1.0 \text{ m} \cdot \text{s}^{-1}$. At low wind speeds, measurement of wind direction becomes unreliable, so identifying up- and down-wind periods is not possible with stagnant winds. Further, at low wind speeds the likelihood of vehicle-induced turbulence effecting the background measurements increases. To avoid analysing the lowest wind speed periods where these issues might be prevalent, we also restricted highway measurements to non-stagnant winds (i.e. $\geq 1 \text{ m} \cdot \text{s}^{-1}$).

2.3 Predicting background concentrations at the highway site

2.3.1 Onsite background concentration (C_{bkg}) prediction methods

We tested nine methods of estimating background concentration measured upwind the highway: two urban background stations, three frequency methods, three regression methods, and a final ensemble method.

The urban background stations we tested were the same two urban background stations mentioned previously:

- Downsview station, located in an urban area but 175 m from the nearest road (Site C in Figure 1).
- Hanlan's Point station, located on an island in Lake Ontario, isolated from nearby emissions (Site D in Figure 1).

We tested three frequency methods:

- A naïve rolling minimum, with the length of the rolling window optimized to minimize prediction error. This basic method was included as a minimally simple approach.
- The pseudo-wavelet method devised by Wang et al. (2018).
- A rolling ball background subtraction – rolling ball algorithms are common in image processing, where they are used to correct unevenly intense image backgrounds. To our knowledge, this is the first case of a rolling ball algorithm applied in air pollution research.

We included three regression methods:

- Traditional ordinary least squares (OLS) multiple linear regression.
- Regularized (elastic net) regression, which is a linear model with regularization terms to control for overfitting.

- Machine learning regression with XGBoost – this model can produce accurate non-linear predictions and has many hyperparameters that can be tuned to control overfitting, degree of variable interaction, model complexity, etc. The XGBoost model has been successfully deployed previously in air quality studies, demonstrating its potential usefulness (Xu et al., 2020b, a). See Supplement Appendix C for details on how we specified XGBoost models.

For each regression method we included a variety of predictive covariates in addition to concentration measured downwind of the road, including concentrations measured at the distant urban background stations, traffic count, predictions of pollutant dilution from the RLINE dispersion model, meteorology measured at the Highway 401 site, and more (Snyder et al., 2013). In some cases, we transformed covariates prior to fitting regression models to increase the linearity of the relationship between covariate and measured C_{bkg} , and for regression models we scaled predictors. Finally, we included one additional ensemble model: this final method was a regularized (ridge) regression using the predictions from each of the prior listed methods as inputs. Extended descriptions of each of the algorithmic methods are provided in the supplement.

2.3.2 Optimizing prediction methods and evaluating accuracy

Many of the above methods for predicting C_{bkg} require user-specific parameters. To select these parameters, we applied a similar process across each method. For each algorithmic method, we optimized for parameters that produced the lowest prediction error by either iterating over parameters or via Bayesian hyperoptimization (Akiba et al., 2019). In each case we evaluated prediction error with five-fold cross-validation to control for overfitting. The only exception was OLS, which has no hyperparameters to tune, however we still evaluated its accuracy with the same cross-validation scheme. Additional details on C_{bkg} prediction method optimization and evaluation, including details on optimized hyperparameters, cross-validation, and metrics are included in the appendices.

3 Results and discussion

3.1 Geographic variability of urban background concentrations

After defining when a measurement is considered background at the highway site, we first compared average background concentrations at the three sites in the Greater Toronto Area. Figure 2 summarizes average concentrations while Figure 3 depicts their diurnal patterns. From these figures, we can directly compare typical levels and daily patterns in background concentrations across a city. Table 2 quantifies geographic and temporal variability in local and background concentrations at the same sites.

265

Table 2: Mean and standard deviations (s.d.), and coefficient of variation (c.v. = s.d./mean) of pollutants measured at each study site, and means and standard deviations of differences between selected sites. The HWY Down – HWY Up row is the difference between up- and down-wind at the Highway site, summarizing variability in local ($C_{meas} - C_{bkg}$) concentrations. The Downsview – Hanlan’s row is the difference between Downsview and Hanlan’s Point sites, capturing geographic variability in backgrounds. Values rounded to two significant figures.

	CO [ppbv]			CO ₂ [ppmv]			NO _x [ppbv]			PM _{2.5} [$\mu\text{g}\cdot\text{m}^{-3}$]		
	Mean	s.d.	c.v.	Mean	s.d.	c.v.	Mean	s.d.	c.v.	Mean	s.d.	c.v.
Highway downwind roadside	380	160	0.42	460	30	0.064	45	33	0.74	6.4	5.6	0.87
Highway upwind background	230	120	0.54	440	30	0.068	16	18	1.1	4.8	4.7	0.99
Downsview	220	97	0.43	450	23	0.053	15	17	1.2	6.5	6.2	0.95
Hanlan’s Point	220	62	0.28	440	15	0.034	7.9	11	1.3	6.3	4.7	0.75
Wallberg (Downtown)	240	85	0.36	450	20	0.044	14	12	0.9	5.9	4.8	0.82
HWY Down – HWY Up	150	110	0.69	17	17	0.99	28	25	0.9	1.8	3.9	2.2
Downsview – Hanlan’s	9.9	84	8.5	8	19	2.4	6.9	13	1.9	0.19	4.5	24

270

*The highway upwind background only included periods where the sensor was upwind (northerly) of the road, whereas other sites were not restricted by wind direction or speed. In the case of PM_{2.5}, if these wind direction and speed limits were applied to all sites, backgrounds at other sites were more comparable to the highway upwind background site (Figure 5).

275

280

285

For CO, CO₂, and NO_x, we recorded the greatest average concentrations at the Highway 401 downwind site, and for PM_{2.5} it was second greatest. High concentrations downwind the road is sensible given the intensity of traffic on this road. For example, the ratio of downwind/upwind concentration was greatest for NO_x: median total downwind NO_x was 2.7 times greater than upwind background NO_x at the highway site,. In the context of Figure 2, background NO_x appears similar between the highway, Downsview, and Hanlan’s sites, however this is misleading: low average background NO_x concentrations mean that the percent differences between sites are relatively greater than for pollutants like PM_{2.5} and CO₂, which have large backgrounds. This introduces a contradiction: when background concentrations are low compared to near-source concentrations, assuming a low or zero background introduces little error. At the same time assuming a homogenous background concentration creates the greatest percent error between background sites.. This means even a rough estimate of the NO_x background will be adequate when the application is subtracting this small value from a much larger total NO_x concentration measured downwind an emissions source. In contrast, it is challenging to evaluate how background NO_x differs between locations, given these concentrations will be small and difficult to estimate reliably. This is reflected in Figure 3, where diurnal background NO_x measured at the Hanlan’s Point site is never equal to the other two background sites, whereas CO₂ and CO had similar concentrations across all sites during at least some times of the day.

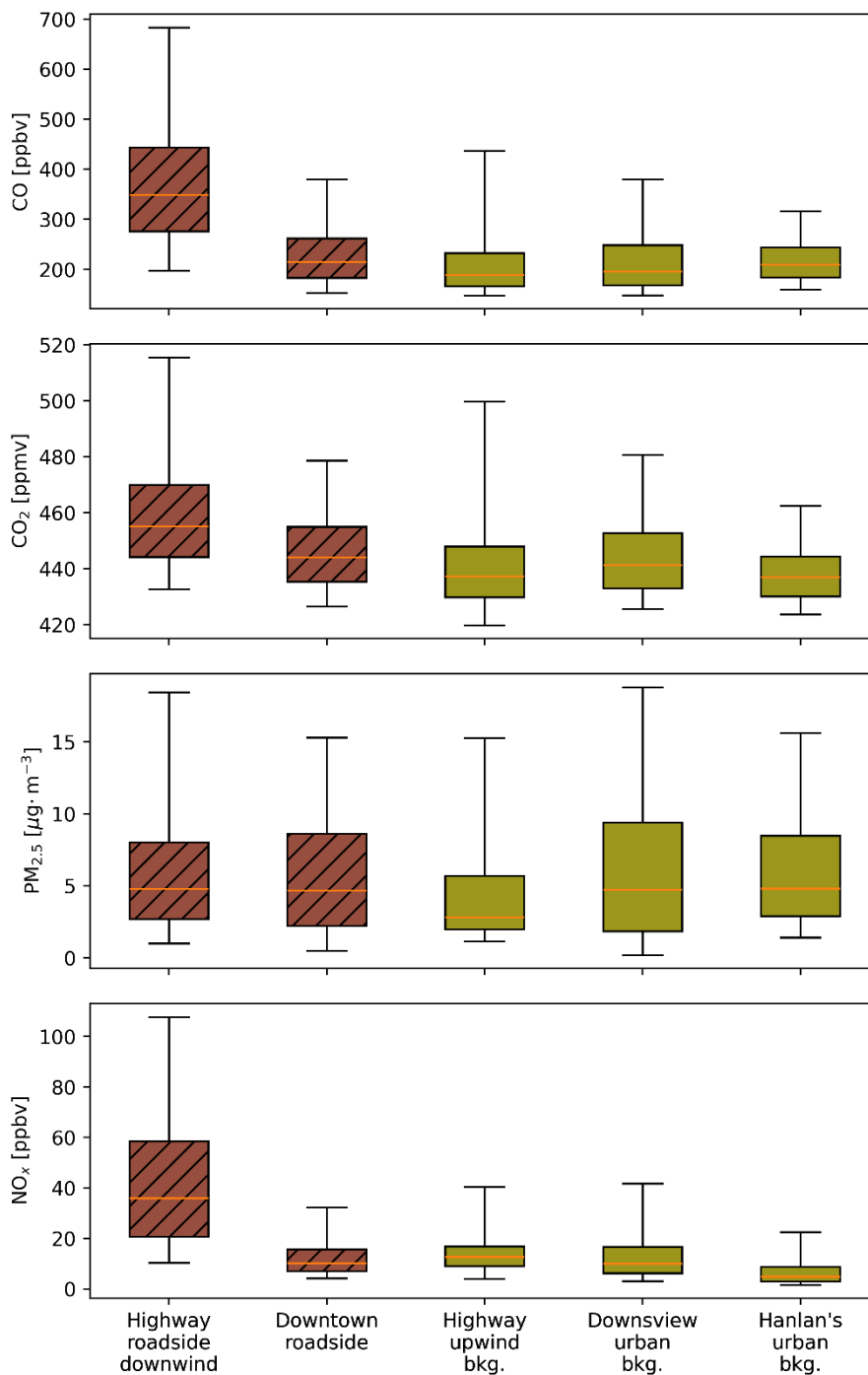


Figure 2. Box-and-whisker plots of minutely concentrations measured at the various sites throughout Toronto. Darker hatched boxes indicate sites near and/or downwind a road (i.e. non-background sites). Boxes extend to 25th and 75th quantiles, whiskers extend an additional 1.5 interquartile ranges. Middle bars are medians. Note that highway sites were limited to periods with appropriate wind directions and speeds, as described in the methodology.

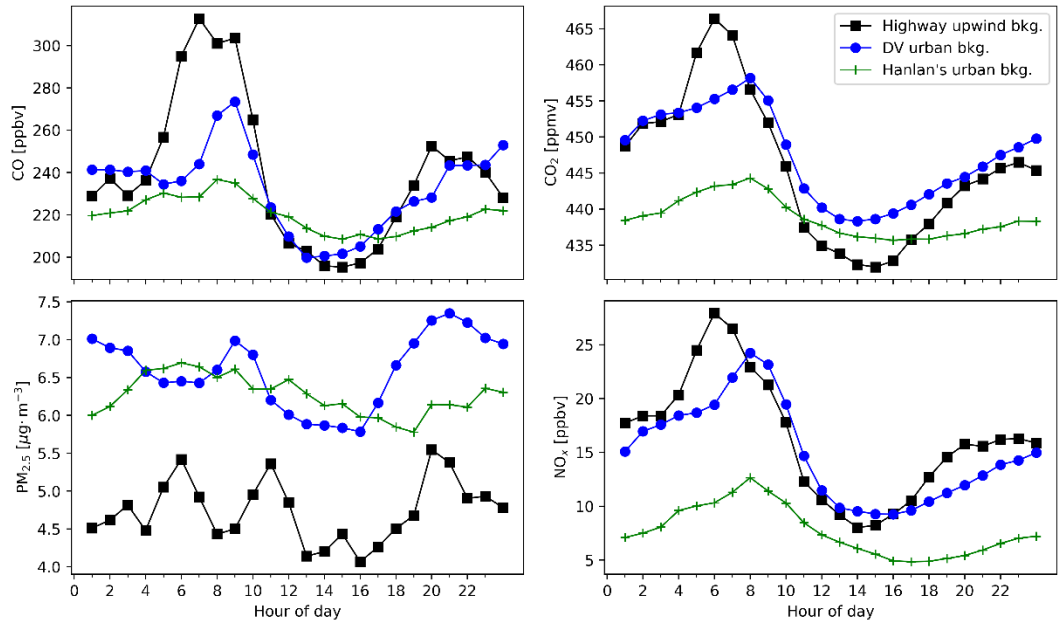
For CO, we measured similar background levels at the Highway 401 downwind site and the Downsview urban background site, with the largest deviations between the two occurring during morning rush hour (Figure 3). There are two possible explanations for this morning divergence: first, higher nearby anthropogenic activity and emissions coupled with lower wind speeds in mornings would increase heterogeneity in urban background concentrations across the city. Second, during low morning wind speeds, emissions from the highway might reach the background station. However, we subsampled our highway upwind background measurements for periods with non-stagnant winds, so this second explanation should have a limited effect on our measurements. Thus the morning rush-hour background CO differences in Figure 3 indicates increased spatial background heterogeneity during these times. CO measured at the Hanlan's Point urban background station were fairly level throughout the day, with a possible slight peak during morning rush hour. CO at Hanlan's Point was roughly 5 to 25% lower than the backgrounds measured elsewhere in the city, except during midday to early afternoon when concentrations were lowest and similar at all three sites. At the Highway 401 site we measured background concentrations only when the sensor was upwind the road. Further upwind was a suburban residential area north of the highway, so emissions from gas-fuelled furnaces may compound the background heterogeneity from low morning wind speeds we mentioned previously, especially given that our measurement campaign took place during winter months.

Like CO, background CO₂ concentrations had correlated diurnal trends and levels at the highway and Downsview locations, with higher rush-hour concentrations at the highway. This is indicative of spatial heterogeneity in CO₂ concentrations across the city, especially during mornings, as we observed for CO. Given that we calibrated CO₂ baselines across sites, these differences indicate the near-road sites measured more transient high CO₂ concentrations, which suggests non-constant sources upwind of these sites. The differences between CO₂ measured at the urban background stations and the highway upwind background means those distant urban background stations would not serve as adequate estimates of background CO₂ at the highway site if considering minutely or hourly data. Conversely, the similarity in overall average background CO₂ concentrations suggests that if we were to consider only long-term (i.e. 24 h or greater) averages, distant urban background stations provide reasonable estimates of average background CO₂ concentrations (Figure 2 and Table 2). More precisely, when comparing long-term averages in Table 2, the difference between the Highway upwind background and Downsview was less than 10% for CO, CO₂, and NO_x, indicating that for such longer-term comparisons an urban background station would provide a fair estimate of upwind background – it should be noted, however, that this required restricting the highway upwind background by wind direction and speed, while stations like the Downsview site had no such restriction.

The only notable feature in diurnal patterns of PM_{2.5} background concentrations was a shallow noon-to-early-afternoon valley at Downsview and Hanlan's Point, which may be due to a combination of increased mixing, and evaporation under higher midday temperatures of secondary ammonium nitrate formed in the early morning. The Highway 401 background sensor recorded the lowest average PM_{2.5} concentrations, but this difference disappeared when the highway site's wind direction and speed limits were applied to other sites. In other words, we found PM_{2.5} was spatially homogeneous across Toronto (Figure 2). This may be reflective of dominant sources and processes contributing to particulate matter in Toronto. Lee et al. (2003) observed over two decades ago that secondary processes were a major source of total PM_{2.5} in Toronto, while

325 more recently Jeong et al. (2020) showed that, while source profiles have changed in the intervening years, secondary sources remain dominant. The importance of such secondary formation processes coupled with the trends in Figure 2 and Figure 3 indicate that separating the contributions of background concentrations and primary emissions to $PM_{2.5}$ concentrations might not be feasible using time-series (frequency) and regression methods such as those discussed here. Conversely, homogeneity of $PM_{2.5}$ concentrations means urban background stations should provide a good estimate of background $PM_{2.5}$ throughout the city.

330 city.



335 **Figure 3. Hourly mean diurnal profiles of measured background pollution concentrations at three stationary measurement sites in Toronto. For the Highway 401 site, these figures depict measurements from the background sensor only during periods where the background sensor was upwind the road and wind was not stagnant, producing a valid measure of C_{bkg} as defined in the methodology. Downsview (DV) and Hanlan’s backgrounds had no wind direction restrictions; when wind limits were applied to all sites (not shown), $PM_{2.5}$ levels were similar across all three sites.**

For CO , CO_2 , and NO_x , the correlation in diurnal patterns between background concentrations measured at the highway and Downsview sites suggests that the Downsview station, situated within the city but about 175 m distant from the nearest notable traffic emissions source, may serve as an adequate estimate of upwind concentrations for measurements near sources like the highway in Toronto, but that the accuracy of this estimate would be reduced during mornings and evenings, when spatial heterogeneity across the city in background concentrations may be larger. Across pollutants, the level of hour-to-hour variability in Figure 3 and standard deviations in Table 2 correlated with the proximity of sites to pollution emissions sources.

345 The highway upwind background, while isolated from the road of interest via wind direction, was still located in a dense urban area with a variety of emissions sources and had strong diurnal patterns throughout the day. We observed less hour-to-hour

variability at the Downsview and Hanlan's Point urban background stations. The Downsview site measurements were closer in magnitude to the highway upwind background, but variability was lesser, especially during morning and evening. The Downsview station is separated from immediate sources but is still within a few hundred metres of emissions sources, while concentrations measured at the more isolated Hanlan's Point were typically lower than all other sites (except for $PM_{2.5}$). Hanlan's Point lays on an island in Lake Ontario south of Toronto – while there is an airport on the same island, its runway is over 1 km away. We posit the lower CO, CO₂, and NO_x at Hanlan's Point can be explained from an absence of nearby sources, while the similar $PM_{2.5}$ is explained by both the dominance of secondary and regional particle sources.

Figure 4 shows scatters and kernel density estimates (KDE) of measured background CO₂ at the three background sites. Similar plots for the remaining measured pollutants are available in Supplemental Appendix J. From these scatters we can derive similar conclusions about the relationship between background concentrations at various sites across the city. As we observed in Figure 2 and Figure 3, background concentrations at the near-road site might be reasonably estimated for some but not all pollutants. We observed that CO and CO₂ measured at the Downsview urban background station were somewhat correlated with background levels measured at the highway – thus we expect concentrations measured at Downsview to be important covariates in regression models predicting highway C_{bkg} for CO and CO₂ – but we noted that the correlation between Downsview and Highway 401 background concentrations was less clear for NO_x. $PM_{2.5}$ concentrations were mostly homogeneous across the city and thus appeared more strongly correlated in scatters (Supplemental Figure J.3). Background NO_x concentrations were the least comparable between sites (Supplemental Figure J.2), corroborating our earlier observation that, despite having low concentrations, NO_x background concentrations are paradoxically very spatially heterogeneous and have a high degree of source-specific contribution at near-source sites. From these results we can rank pollutants in order of increasing background concentration geospatial heterogeneity: $PM_{2.5} < CO_2 \approx CO < NO_x$. While $PM_{2.5}$ is clearly the most homogeneous and NO_x the most heterogeneous, the distinction in variability between CO₂ and CO is less clear.

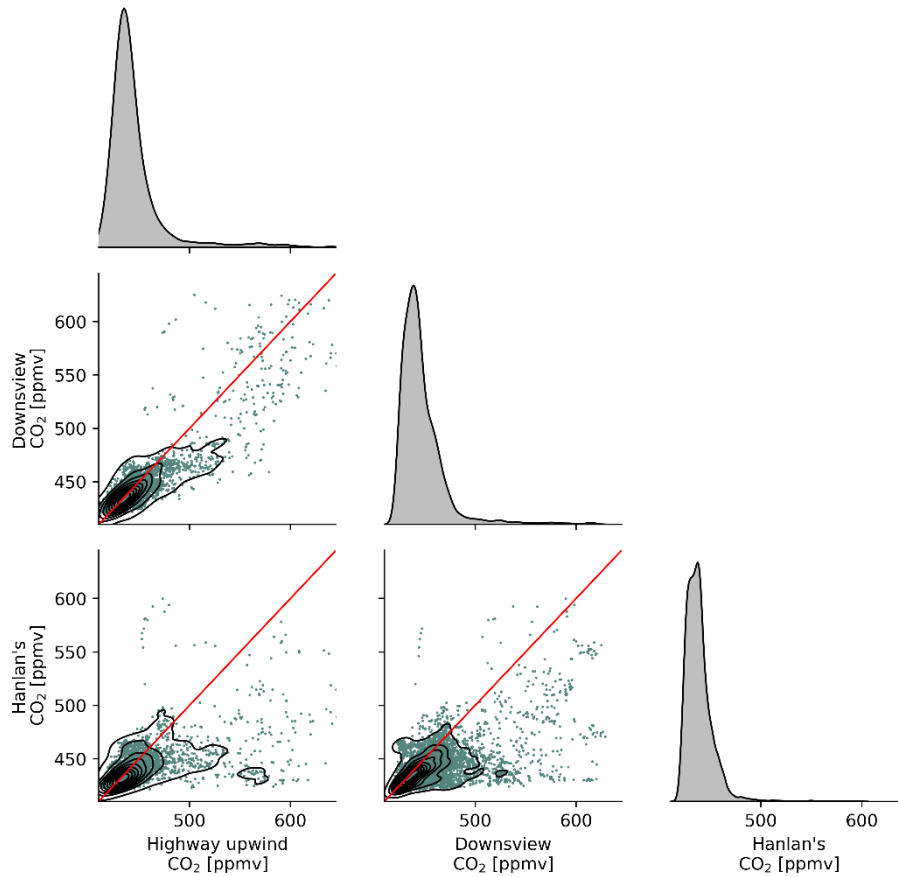


Figure 4. Paired scatters and kernel density estimates (KDE) of measured background carbon dioxide concentrations, at three stationary measurement sites in the Greater Toronto Area. Red lines are 1-to-1. The KDE plots on the diagonal show the unitless distribution of the measurements with areas summing to unity.

We also observed that this ranking of geographic variability was similar to the relative temporal variabilities in background concentration for each pollutant. The coefficients of variation for the difference between the Downsview and Hanlan's Point sites in Table 2 reflect a similar ordering, with the inter-site difference in $\text{PM}_{2.5}$ having the most variability relative to its mean, and NO_x having the least. From these comparisons of measured local and background concentrations, we can conclude that in some cases the urban background sites can provide a suitable estimate of highway upwind background concentrations, but for some pollutants and times of day, a direct measurement or algorithmic estimate of background concentration is necessary. Accordingly, we further applied and tested each of the background concentration prediction algorithms we introduced in the methodology.

3.2 Comparing performance of background concentration estimates

Figure 5 shows diurnal patterns of measured and predicted concentrations at the Highway 401 site. The lines for XGBoost and pseudo-wavelet show background concentrations estimated from the highway downwind data. This figure illustrates the degree of agreement across the background concentrations estimates and contrasts this relative to the total concentrations measured downwind of the highway.

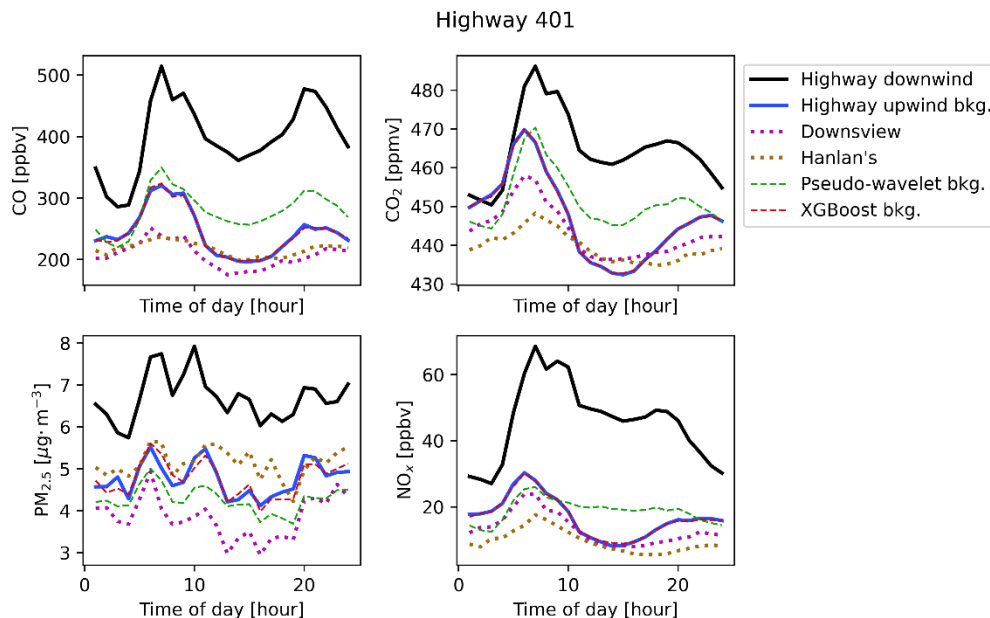


Figure 5. Diurnal trends of measured total (black), measured background (blue), and predicted background (dashed purple and green, and red) concentrations at the Highway 401 site. Only periods containing valid measures of C_{bkg} upwind of the highway as defined in the methodology were included in these figures. Note that measured background trends may differ slightly from Figure 3 as this figure only includes periods where all shown measured and predicted backgrounds were concurrently available. Due to model accuracy and the effect of averaging to the nearest hour, the lines for “Highway upwind bkg.” and “XGBoost bkg.” are sometimes superimposed.

Figure 6 shows measured-predicted scatters for a selection of background concentration prediction algorithms. From these scatters we observed that the accuracy of a method in estimating measured background concentrations was correlated with model complexity – the computationally complex XGBoost model produced the most qualitatively accurate scatters of those shown in Figure 6, while the simpler frequency (pseudo-wavelet) and urban background station (Downsview) estimates were accurate at times but clearly less reliable than the XGBoost predictions.

For PM_{2.5}, we noted that our ability to produce an algorithmic estimate of measured background concentration was limited. Poor accuracy of predictions is likely explained by the aforementioned sources and processes unique to PM_{2.5} out of all the pollutants studied here. For the remaining pollutants, accuracy varied between methods but appeared generally superior to that of PM_{2.5}. However, as previously mentioned, this does not preclude us from viewing PM_{2.5} as a counterexample by which we can judge other, more accurately predicted pollutants.

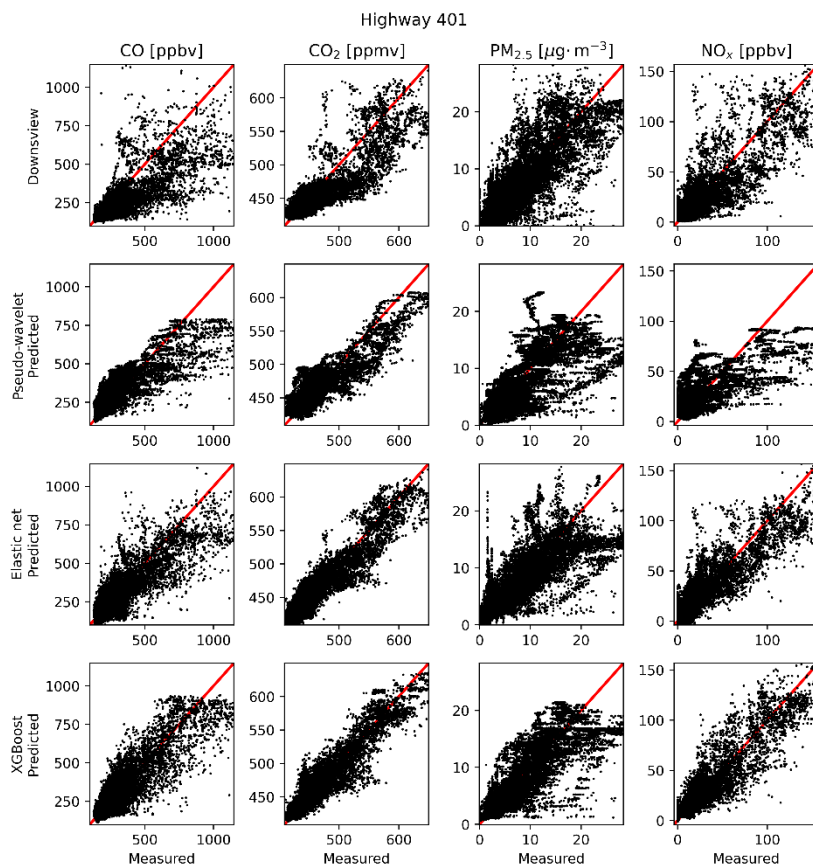
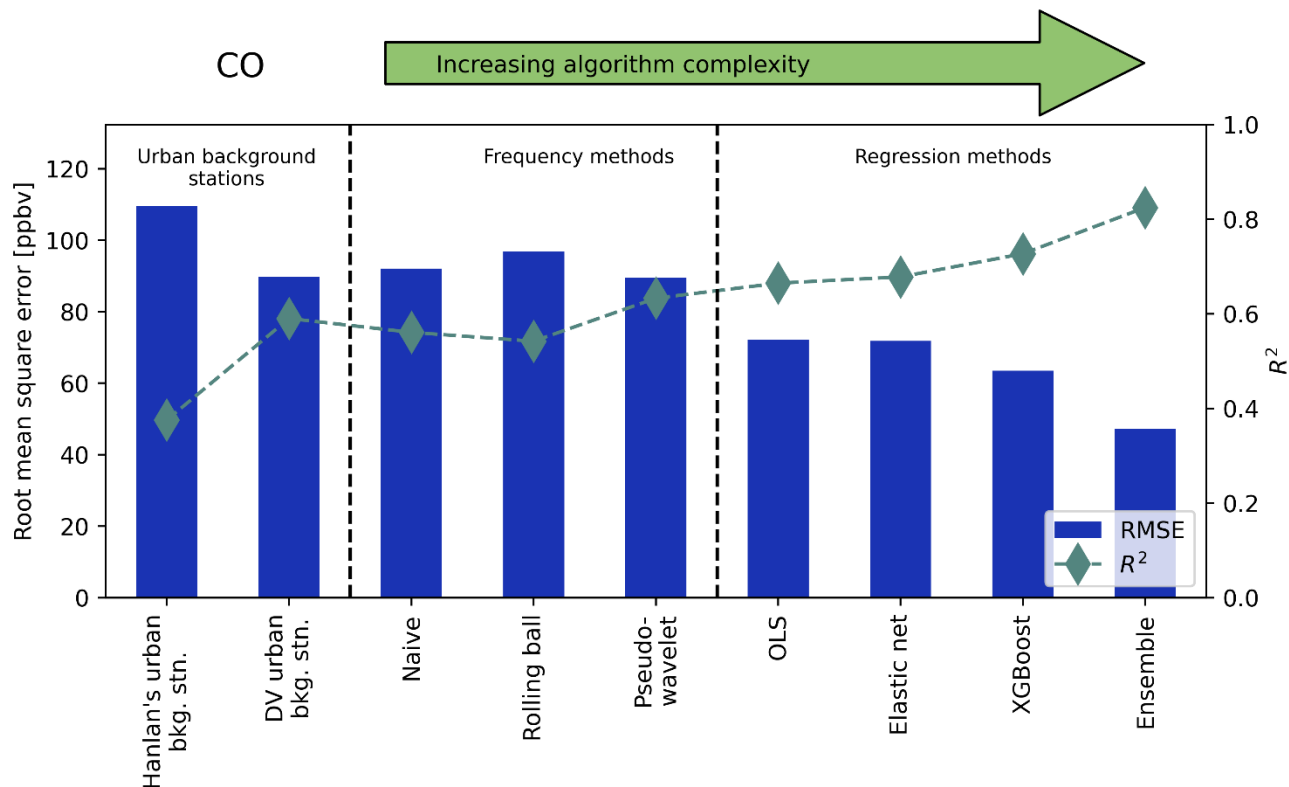


Figure 6. Measured-predicted scatters for selected methods of estimating background concentration at the Highway 401 site. Measured concentrations are true C_{bkg} recorded by the AirSENCE device north and upwind of the highway. Scatters only include periods where C_{bkg} measures were valid as defined in the methodology, and only periods where all background estimates were available. Red 1:1 lines are included to illustrate the expected relationship.

Figure 7 shows the root mean square error (RMSE) and coefficient of determination (R^2) of CO C_{bkg} predictions using each method, including the urban background stations, roughly ordered by increasing complexity and accuracy. The same metrics for NO_x, CO₂, and PM_{2.5} are available in the supplement. Where Figure 6 permits us to qualitatively examine C_{bkg} prediction accuracy, Figure 7 (and Supplemental Figures H.1 to H.3) quantitatively corroborate our observations that accuracy tended to increase with model complexity. Unsurprisingly, the XGBoost and ensemble models generally had the greatest accuracy out of all algorithmic methods, according to prediction RMSE and R^2 . When compared with urban background stations, frequency methods tended to have similar error to measured background data from Downsview in predicting C_{bkg} (except for NO_x), and regression methods, particularly XGBoost, had less error and greater R^2 . OLS and elastic net had lower accuracy than XGBoost models, indicating some degree of variable interaction or nonlinearity existed in background concentration behaviour, but the increase in accuracy from linear regression to machine learning was minor for all pollutants. Hanlan's Point always had greater error and lower R^2 than Downsview, a trend reflecting our above discussion on the suitability of using a distant urban

background station for predicting onsite C_{bkg} . For CO_2 and NO_x , the incremental gain in prediction accuracy between
420 frequency and regression methods was more apparent than for $\text{PM}_{2.5}$ and CO , suggesting accurate prediction of CO_2 and NO_x
might more strongly rely on information contained in predictors other than downwind C_{meas} . Interestingly, for NO_x the
predictive accuracy of frequency methods was worse than simply using measurements from the Downsview background
station to predict C_{bkg} . This suggests background NO_x cannot be extracted from downwind total NO_x alone with high accuracy,
although as previously discussed high accuracy is not needed for applications like resolving local contributions since
425 background NO_x is generally much lower than local NO_x . For every other pollutant the accuracy of the Downsview background
station in predicting C_{bkg} was nearer that of frequency methods, though in some cases still had slightly better accuracy than
frequency methods. However, this difference was small compared to the difference for NO_x . This observation might also be
reflective of our previously mentioned sensitivity in estimating background NO_x due to its relatively low average
concentrations.

430 For $\text{PM}_{2.5}$, the accuracy of algorithmic C_{bkg} predictions did exceed that of the Downsview station, but the relative
incremental gain in accuracy was less clear than for other pollutants, suggesting little benefit can be gained for algorithmically
predicting background $\text{PM}_{2.5}$ over simply using an urban background station. Only the XGBoost and ensemble models had
notably superior accuracy for $\text{PM}_{2.5}$, indicating that greater complexity is necessary to accurately predict background $\text{PM}_{2.5}$ than
for other pollutants. These trends broadly align with our prior discussion on the homogeneity and complexity of sources and
435 processes governing background $\text{PM}_{2.5}$. However, the RMSE of the low-cost sensor placed upwind of the highway versus a
reference sensor was greater than the mean difference between up- and down-wind $\text{PM}_{2.5}$ at the highway (see Supplement
Appendix B), suggesting that in addition to the homogeneity of $\text{PM}_{2.5}$ (Figure 2 to Figure 4), our ability to separate C_{bkg} from
 C_{meas} was limited for $\text{PM}_{2.5}$, which would inherently limit our ability to predict the same.



440 **Figure 7. Root mean square error (RMSE, bars) and coefficient of determination (R^2 , diamonds) for predicted background CO at the highway site, as predicted by each method tested here. Scores show the accuracy of each method in estimating true upwind background concentration, with lower RMSE and greater R^2 being better. Scores were calculated as the mean across five-fold cross-validation.**

445 For CO and CO₂, there is some similarity in accuracy for frequency methods and regression methods. RMSE and R^2 for CO predictions from regression methods were only slightly better than RMSE for frequency methods. For CO₂, prediction RMSE and R^2 appeared to improve from frequency methods to regression methods, and again to the ensemble model, indicating similar levels of accuracy within each class of algorithmic prediction models.

3.3 Importance of site-specific covariates

450 We fit each method to only a single field study site, so it is difficult to conclude if our results are generalizable for urban background concentrations or if they are specific to this site. However, we can gain some insight into the generality of our conclusions by testing the importance of site-specific information in producing accurate estimates of background concentrations with the regression methods tested here. Specifically, to test the importance of onsite information in predicting background concentrations, we refit our XGBoost model after shuffling covariates specific to the highway site, but XGBoost hyperparameters and the total number of variables remained unchanged. Shuffling covariates refers to the process by which

one input variable at a time is randomly shuffled, so the measurements of that variable are no longer in order relative to other input and target features. By shuffling covariates and refitting, we remove possible correlations between site-specific features and the target measured background concentration but retain the same set of features so we can refit the XGBoost model without retuning hyperparameters, enabling comparison of XGBoost predictions with and without highway-specific inputs.

To produce this regression, we shuffled covariates specific to the highway emissions source, including RLINE dispersion estimates, highway traffic counts, and traffic-weighted average vehicle speed. The site-specific measurements we left unshuffled were downwind total concentrations, C_{meas} , the target upwind background concentrations, C_{bkg} , and meteorology. We chose not to shuffle meteorology based on our observation that meteorology is usually similar across the city at any moment in time and thus could feasibly be measured offsite. Meteorological measurements are also often widely available or measurable with relatively low-cost instruments. Figure 8 shows normalized prediction errors for C_{bkg} predicted via XGBoost for each pollutant with and without shuffling.

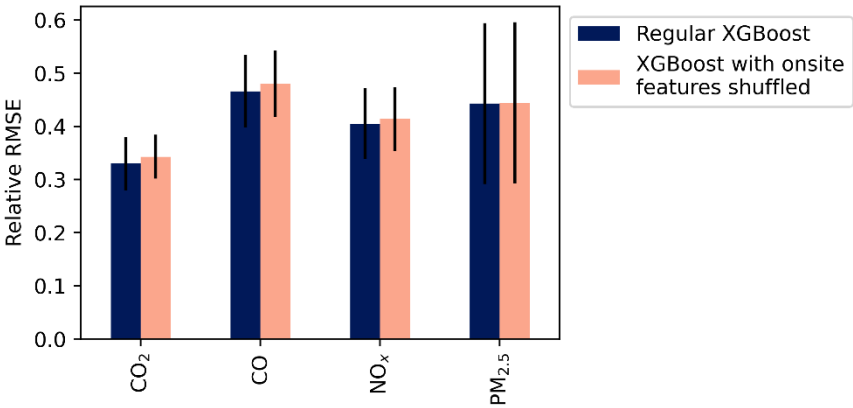


Figure 8. Relative RMSE for XGBoost-predicted C_{bkg} with and without covariates specific to the highway field site included in model. RMSE was calculated via five-fold cross-validation, relative RMSE is RMSE divided by the standard deviation of the target regressand (C_{bkg}). Whiskers are standard deviations across folds.

The errors in Figure 8 suggest that removing information specific to the highway site did not produce a significant change in XGBoost model accuracy. The absolute percent difference between RMSE with and without site-specific variables shuffled was less than 5% for all pollutants, and differences were within one standard deviation across cross-validation folds, indicating little or no significant difference between models with and without shuffled site-specific variables. This indicates that most of the variability in C_{bkg} was explained by highway downwind concentrations and other covariates not specific to the highway – it is also reflective of our observations in Figure 7 (and figures in Supplement Appendix H) that predicting C_{bkg} with concentrations measured at the Downsview urban background station, while less accurate than some other methods, still produced prediction R^2 exceeding 0.5 for all pollutants. Since concentrations measured at Downsview and the Highway were included as predictors in both cases in Figure 8, we can indirectly conclude that concentrations measured at Downsview

coupled with concentrations measured downwind the highway together contain most of the information necessary to accurately
480 predict C_{bkg} , and that adding more emissions-source-specific covariates only marginally increased prediction accuracy.

This lack of difference between XGBoost accuracy with and without site-specific features might imply our model of
background concentrations is not site-specific. That is, the XGBoost model without highway-specific covariates might be
transferable to other locations. This in turn would mean that the spatial variation of background across the city is mostly
encompassed within information provided by measuring the total concentrations at different sites, consistent with the
485 assumption underlying frequency-based methods. With only one near-source site in this study with up- and down-wind
measurements, we did not further test this transferability. At the very least, this result shows our methodology might be
successfully repeated at new near-source sites without requiring as many site-specific covariates as we included here.

3.4 Regression model feature importance

We can examine feature importance in the XGBoost models for each pollutant to gauge covariate importance for estimating
490 C_{bkg} . We achieve this using Shapley Additive Explanations (SHAP) – SHAP plots can provide explanations of feature
importance for complex nonlinear models where simple coefficients are not available, as is the case with XGBoost (Lundberg
and Lee, 2017). Additional examples of SHAP values in the context of air pollution research can be found from Xu et al.
(2020a, b). Figure 9 shows SHAP beeswarm plots for the XGBoost model predicting highway upwind background C_{bkg} for
each pollutant.

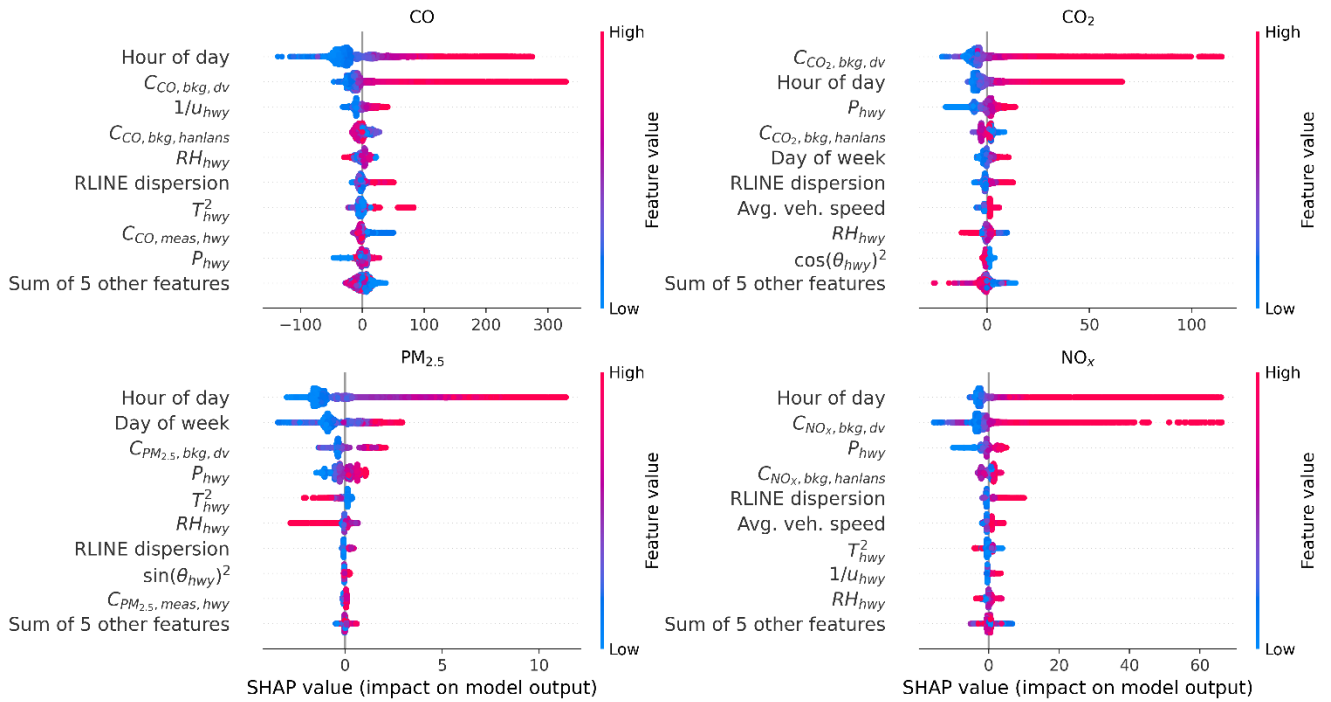


Figure 9. SHAP beeswarm plots for XGBoost models predicting upwind background concentration at the highway site. These figures indicate relative degree of importance – for example, a blue dot far to the right on a feature indicates that a low value of that feature was associated with a high predicted concentration. Each dot represents one predicted concentration and one value of that feature. (bkg: background; dv: Downsview; hwy: highway; meas: measured)

The SHAP values in Figure 9 suggest that for CO, CO₂, and NO_x, the most important predictors of upwind background at the Highway 401 site were concentrations measured at the Downsview urban background site and hour of day. This is consistent with each of our prior discussed results: generally, concentrations measured at Downsview can provide a fair estimate of mean background concentration levels, but these estimates may be inaccurate during some hours of the day and predictions can be notably improved through inclusion of additional information. The fact that time of day is an important predictor aligns with our observation that Downsview serves as a fair background estimate, except during morning and to a lesser degree evening rush hours (i.e. except during some hours of the day). Outside these important predictors, meteorology had notable importances for all pollutants. Lastly, for PM_{2.5} pollutant concentrations measured at Downsview, while still important, had a lesser impact on predictions, which is yet again reflective of the difficulty in predicting background PM_{2.5} at the highway site.

Concentrations measured downwind the highway, C_{meas} , were much less important predictors in XGBoost than concentrations measured at Downsview. This was unexpected both based on theory and when comparing against other methods: C_{meas} should always be a direct sum of C_{bkg} and local emissions, and thus we expect it to explain some variability in background concentrations. This result was also in contrast to regression coefficients from our linear elastic net fits, which fit large coefficients on C_{meas} for all pollutants (Supplement Appendix L). Regardless, the results of this SHAP analysis

515 suggest that C_{meas} had a comparatively small impact on XGBoost predictions. On the other hand, we found that our frequency methods, which take only C_{meas} as an input, had fair accuracy. These two results together suggest that to extract useful estimates of C_{bkg} from C_{meas} , algorithmic methods benefit by considering not just concurrent measurements but past and future values of C_{meas} as well. In this study, we did not include lagged values of C_{meas} in regression models, so further exploration of such covariate transformations might benefit C_{bkg} prediction accuracy and understanding of background
520 concentration behaviour.

The importance of temperature for some predictions might be explained by an uneven distribution of measured temperatures. Most of our measurements occurred in winter with low temperatures, while a minority of measurements at the end of our study had higher temperatures. Because there are fewer samples with high temperatures, regression models risk placing a greater relative importance on those samples, inflating the relative importance of temperature. This can be improved upon in future
525 studies by extending a similar regression fitting approach to a longer measurement period.

3.5 Limitations of analysis

As this study examined only a single urban area, the applicability of our results to other urban areas relies on the assumption that many cities feature a similar variety and heterogeneity of emissions sources and geography. The sites explored here, including both urban background and near-road up- and downwind- sites, represented a variety of geographic features,
530 including proximity to a large body of water, green space, and proximity to emissions sources other than the road targeted at the highway site. While our analysis of XGBoost model accuracy without site-specific features in Figure 8 lends support to the idea that our model of background concentrations is not specific to the highway site, this conclusion is indirect and a better method of testing transferability would be to apply our methods at new sites.

We also only explored background concentrations for four airborne pollutants: three gaseous and one particulate. For the
535 gaseous pollutants tested, we expect that loss or formation via reaction will be low. While NO and NO₂ concentrations can vary rapidly near roads through reaction, we only considered the sum of the two, NO_x, which should remain constant over the distances from the highway investigated here. This simplicity of behaviour will simplify our models, and it is plausible that background pollutants with more complex reaction mechanisms or sources might require more covariates to accurately predict with regression models. For example, modelling background ozone would probably benefit from including insolation as an
540 exogenous predictor.

Lastly, it remains unclear if these models would transfer well to sites with different geometry, emissions sources, or weather. It is plausible that the strength of the methods tested here is due to the simplicity of the major source observed: the size and business of Highway 401 lends confidence to the assertion that it will be the dominant source of local airborne pollution at the downwind highway site. Traffic also has consistent diurnal patterns and emissions intensity is easily inferred through a simple
545 traffic count, which itself has a strong diurnal pattern. If the regression models presented here were refit near a source with

different characteristics, such as an industrial source emitting at all hours of the day, or at a measurement site with multiple strong upwind sources, it stands to reason that predictive performance would be degraded.

4 Conclusions and recommendations

Based on the results of this study, we recommend that municipalities or air pollution specialists deploying sensors or monitors with the aim of resolving the contribution of specific emissions sources consider carefully how they will measure or algorithmically isolate the contribution of background to total measured concentrations. Our sites in Toronto reflected a variety of geographic features (varying built environments, water proximity, green space, etc.), indicating that our finding of varying background concentrations might apply to other cities, since these features are common across many urban areas. From our analysis of background concentration prediction methods, we can recommend which method users should choose based on their use-case and availability of data. These recommendations are loosely ordered by decreasing strength of accuracy alongside decreasing cost:

1. If possible, direct measurement of background concentrations and wind immediately upwind the source of interest should always be preferred.
2. In cases where measurements of upwind C_{bkg} are available for some but not all of the study period, we recommend applying a regression approach. XGBoost or similar machine learning approaches are preferable to traditional regressions, as they allow for nonlinearity and unspecified interactions. Conversely, we caution against applying regression models outside the conditions they were trained in, such as different sites or seasons.
3. For applications where only long-term averages (i.e. 24 h or longer) are of concern, using a distant urban background station as a proxy for true onsite C_{bkg} measurements will prove sufficiently accurate, however for higher-resolution measurements, urban background stations may prove inaccurate during periods of peak emissions, like during rush-hour near a roadway.
4. For applications where both upwind C_{bkg} measurements and a suitable urban background station are both unavailable or too costly, we suggest applying one of the frequency methods described here, particularly the pseudo-wavelet method developed by Wang et al. (2018) or the rolling ball algorithm. For these frequency methods, in roadway applications we suggest using hyperparameters like those identified here (see Supplement Appendix K). For pollutants other than those measured here, we suggest applying parameters like those we identified, based on similarity in pollutant behaviour – for example, if a pollutant is expected to be a strong tracer or a local source, as NO_x is for traffic, we suggest applying similar hyperparameters as used for NO_x in this study.
5. In a similar vein, for cases where municipalities are deploying networks of sensors, or epidemiologists are exploring geographic variability of background concentrations vs. local emissions, we suggest applying the pseudo-wavelet or rolling ball frequency methods. While the context of our tests here were up- and down-wind differences targeting a single roadway emissions source, the theoretical basis of frequency methods – that background concentrations

vary on a longer timescale than local emissions – extends these methods to pollution concentrations regardless of proximity to one particular source. The pseudo-wavelet method applied in this context is also touched upon by Wang et al. (2018) and Hilker et al. (2019).

Generally, we do not suggest applying the naïve rolling minimum method – superior frequency methods only require minimal additional computational cost. The usefulness of the ensemble method is also dubious. While the ensemble model did produce the best output in this case, this is to be expected; an ensemble model should outperform its constituent models. However, the extent of information and effort required to implement such a model for predicting C_{bkg} seems to exceed the potential benefit of gains in predictive accuracy. Finally, we suggest any study targeting specific emissions sources carefully consider how to extract local versus background contributions to measured concentrations, including but not limited to applying one of the methods tested here. We also encourage additional research in separating local and background concentrations, especially with different emissions sources, regions; or for different types of measurements, such as mobile monitoring or distributed sensor networks.

Code and data availability. Analysis code and raw data can be made available upon request.

595 *Author contributions.* CRediT: Taylor D. Edwards: conceptualization, methodology, software, validation, formal analysis, resources, data curation, writing – original draft, writing – review & editing, visualization. Yee Ka Wong: methodology, data collection validation, investigation, resources, writing – review & editing. Jonathan M. Wang: investigation, resources, data curation. Cheol-Heon Jeong: investigation, data collection resources, data curation. Yushan Su: data collection, review and editing. Greg J. Evans: writing – review & editing, supervision, project administration, funding acquisition.

600 *Competing interests.* The AirSENCE air quality monitoring technology was originally developed at the Southern Ontario Centre for Atmospheric Aerosol Research at the University of Toronto, and it has now been commercialised and is being distributed by AUG signals, with licensing fees paid to the University of Toronto.

605 *Funding:* The Natural Science and Engineering Research Council and Environment and Climate Change Canada provided funding to support this research.

Acknowledgements. We would like to acknowledge everyone, past and present, involved in maintaining the three stations operated by Ontario Ministry of the Environment, Conservation and Parks and the Wallberg laboratory operated by the Southern Ontario Centre for Atmospheric Aerosol Research at the University of Toronto. We thank the Pine Point Tennis Club
610 for permission to place instruments on their grounds.

5 References

- Akiba, T., Sano, S., Yanase, T., Ohta, T., and Koyama, M.: Optuna: A Next-generation Hyperparameter Optimization Framework, in: *Proceedings of the 25th ACM SIGKDD International Conference on Knowledge Discovery & Data Mining*, <https://doi.org/10.1145/3292500.3330701>, 2019.
- 615 Arunachalam, S., Valencia, A., Akita, Y., Serre, M., Omary, M., Garcia, V., and Isakov, V.: A Method for Estimating Urban Background Concentrations in Support of Hybrid Air Pollution Modeling for Environmental Health Studies, *Int. J. Environ. Res. Public. Health*, 11, 10518–10536, <https://doi.org/10.3390/ijerph111010518>, 2014.
- Crilly, L. R., Shaw, M., Pound, R., Kramer, L. J., Price, R., Young, S., Lewis, A. C., and Pope, F. D.: Evaluation of a low-cost optical particle counter (Alphasense OPC-N2) for ambient air monitoring, *Atmospheric Meas. Tech.*, 11, 709–720, <https://doi.org/10.5194/amt-11-709-2018>, 2018.
- 620 Crilly, L. R., Shaw, M., Pound, R., Kramer, L. J., Price, R., Young, S., Lewis, A. C., and Pope, F. D.: Evaluation of a low-cost optical particle counter (Alphasense OPC-N2) for ambient air monitoring, *Atmospheric Meas. Tech.*, 11, 709–720, <https://doi.org/10.5194/amt-11-709-2018>, 2018.
- Frey, H. C., Grieshop, A. P., Khlystov, A., Bang, J. J., Roupail, N., Guinnessa, J., Rodriguez, D., Fuentes, M., Saha, P., Brantley, H., Snyder, M., Tanvir, S., Ko, K., Noussi, T., Delavarrafiee, M., and Singh, S.: *Characterizing Determinants of Near-Road Ambient Air Quality for an Urban Intersection and a Freeway Site*, Health Effects Institute, 2022.
- Fushimi, A., Kawashima, H., and Kajihara, H.: Source apportionment based on an atmospheric dispersion model and multiple linear regression analysis, *Atmos. Environ.*, <https://doi.org/10.1016/j.atmosenv.2004.11.009>, 1997.
- 625 Fushimi, A., Kawashima, H., and Kajihara, H.: Source apportionment based on an atmospheric dispersion model and multiple linear regression analysis, *Atmos. Environ.*, <https://doi.org/10.1016/j.atmosenv.2004.11.009>, 1997.
- Gómez-Losada, Á., Pires, J. C. M., and Pino-Mejías, R.: Characterization of background air pollution exposure in urban environments using a metric based on Hidden Markov Models, *Atmos. Environ.*, 127, 255–261, <https://doi.org/10.1016/j.atmosenv.2015.12.046>, 2016.
- Gómez-Losada, Á., Pires, J. C. M., and Pino-Mejías, R.: Modelling background air pollution exposure in urban environments: Implications for epidemiological research, *Environ. Model. Softw.*, 106, 13–21, <https://doi.org/10.1016/j.envsoft.2018.02.011>, 2018.
- 630 Gómez-Losada, Á., Pires, J. C. M., and Pino-Mejías, R.: Modelling background air pollution exposure in urban environments: Implications for epidemiological research, *Environ. Model. Softw.*, 106, 13–21, <https://doi.org/10.1016/j.envsoft.2018.02.011>, 2018.
- Hashad, K., Yang, B., Iskov, V., and Zhang, K. M.: A Computationally Efficient Approach to Resolving Vehicle-Induced Turbulence for Near-Road Air Quality, *ASME J. Eng. Sustain. Build. Cities*, 3, <https://doi.org/10.1115/1.4055640>, 2022.
- Hicks, W., Beevers, S., Tremper, A. H., Stewart, G., Priestman, M., Kelly, F. J., Lanoisellé, M., Lowry, D., and Green, D. C.: Quantification of non-exhaust particulate matter traffic emissions and the impact of COVID-19 lockdown at London Marylebone road, *Atmosphere*, 12, <https://doi.org/10.3390/atmos12020190>, 2021.
- 635 Hicks, W., Beevers, S., Tremper, A. H., Stewart, G., Priestman, M., Kelly, F. J., Lanoisellé, M., Lowry, D., and Green, D. C.: Quantification of non-exhaust particulate matter traffic emissions and the impact of COVID-19 lockdown at London Marylebone road, *Atmosphere*, 12, <https://doi.org/10.3390/atmos12020190>, 2021.
- Hilker, N., Wang, J. M., Jeong, C.-H., Healy, R. M., Sofowote, U., Debosz, J., Su, Y., Noble, M., Munoz, A., Doerksen, G., White, L., Audette, C., Herod, D., Brook, J. R., and Evans, G. J.: Traffic-related air pollution near roadways: discerning local impacts from background, *Atmospheric Meas. Tech.*, 12, 5247–5261, <https://doi.org/10.5194/amt-12-5247-2019>, 2019.
- 640 Jeong, C.-H., Traub, A., Huang, A., Hilker, N., Wang, J. M., Herod, D., Dabek-Zlotorzynska, E., Celo, V., and Evans, G. J.: Long-term analysis of PM_{2.5} from 2004 to 2017 in Toronto: Composition, sources, and oxidative potential, *Environ. Pollut.*, 263, 114652, <https://doi.org/10.1016/j.envpol.2020.114652>, 2020.
- Klems, J. P., Pennington, M. R., Zordan, C. A., and Johnston, M. V.: Ultrafine particles near a roadway intersection: Origin and apportionment of fast changes in concentration, *Environ. Sci. Technol.*, 44, 7903–7907, <https://doi.org/10.1021/es102009e>, 2010.
- 645 Klems, J. P., Pennington, M. R., Zordan, C. A., and Johnston, M. V.: Ultrafine particles near a roadway intersection: Origin and apportionment of fast changes in concentration, *Environ. Sci. Technol.*, 44, 7903–7907, <https://doi.org/10.1021/es102009e>, 2010.
- Kohler, M., Corsmeier, U., Vogt, U., and Vogel, B.: Estimation of gaseous real-world traffic emissions downstream a motorway, *Atmos. Environ.*, 39, 5665–5684, <https://doi.org/10.1016/j.atmosenv.2004.09.088>, 2005.

- Lee, P. K. H., Brook, J. R., Dabek-Zlotorzynska, E., and Mabury, S. A.: Identification of the Major Sources Contributing to PM_{2.5} Observed in Toronto, *Environ. Sci. Technol.*, 37, 4831–4840, <https://doi.org/10.1021/es026473i>, 2003.
- 650 Lundberg, S. M. and Lee, S.-I.: A Unified Approach to Interpreting Model Predictions, 31st Conference on Neural Information Processing Systems (NIPS 2017), Long Beach, CA, USA, 2017.
- Morris, E., Liu, X., Manwar, A., Zang, D. Y., Evans, G., Brook, J., Rousseau, B., Clark, C., and MacIsaac, J.: APPLICATION OF DISTRIBUTED URBAN SENSOR NETWORKS FOR ACTIONABLE AIR QUALITY DATA, *ISPRS Ann. Photogramm. Remote Sens. Spat. Inf. Sci.*, VI-4/W2-2020, 119–126, [https://doi.org/10.5194/isprs-annals-VI-4-W2-2020-119-](https://doi.org/10.5194/isprs-annals-VI-4-W2-2020-119-2020)
655 2020, 2020.
- Olague, E.: Twenty-First Century Tools for Environmental Protection: Real-Time Monitoring, Fine-Scale Modelling and Advanced Analytics for Air Quality Applications, 2022.
- Ontario Ministry of Transportation: Provincial highways traffic volumes 1988-2016, , i, 1–1515, 2016.
- Rodríguez, J., Villalobos, A. M., Castro-Molinare, J., and Jorquera, H.: Local and NON-LOCAL source apportionment of
660 black carbon and combustion generated PM_{2.5}, *Environ. Pollut.*, 123568, <https://doi.org/10.1016/j.envpol.2024.123568>, 2024.
- Ruckstuhl, A. F., Henne, S., Reimann, S., Steinbacher, M., Vollmer, M. K., O’Doherty, S., Buchmann, B., and Hueglin, C.: Robust extraction of baseline signal of atmospheric trace species using local regression, *Atmospheric Meas. Tech.*, 5, 2613–2624, <https://doi.org/10.5194/amt-5-2613-2012>, 2012.
- Sabaliauskas, K., Jeong, C.-H., Yao, X., and Evans, G. J.: The application of wavelet decomposition to quantify the local and
665 regional sources of ultrafine particles in cities, *Atmos. Environ.*, 95, 249–257, <https://doi.org/10.1016/j.atmosenv.2014.05.035>, 2014.
- Snyder, M. G., Venkatram, A., Heist, D. K., Perry, S. G., Petersen, W. B., and Isakov, V.: RLINE: A line source dispersion model for near-surface releases, *Atmos. Environ.*, 77, 748–756, <https://doi.org/10.1016/j.atmosenv.2013.05.074>, 2013.
- Wang, J. M.: Air Quality Impacts of Vehicle Emissions on the Urban Environment: Real-World Emission Factors and
670 Capturing the Fleet Signal, PhD, University of Toronto, 2018.
- Wang, J. M., Jeong, C.-H., Hilker, N., Shairsingh, K. K., Healy, R. M., Sofowote, U., Deboasz, J., Su, Y., McGaughey, M., Doerksen, G., Munoz, T., White, L., Herod, D., and Evans, G. J.: Near-Road Air Pollutant Measurements: Accounting for Inter-Site Variability Using Emission Factors, *Environ. Sci. Technol.*, 52, 9495–9504, <https://doi.org/10.1021/acs.est.8b01914>, 2018.
- 675 Wei, Z., Peng, J., Ma, X., Qiu, S., and Wang, S.: Toward Periodicity Correlation of Roadside PM_{2.5} Concentration and Traffic Volume: A Wavelet Perspective, *IEEE Trans. Veh. Technol.*, 68, 10439–10452, <https://doi.org/10.1109/tvt.2019.2944201>, 2019.
- Xu, J., Wang, A., Schmidt, N., Adams, M., and Hatzopoulou, M.: A gradient boost approach for predicting near-road ultrafine particle concentrations using detailed traffic characterization, *Environ. Pollut.*, 265, 114777, <https://doi.org/10.1016/j.envpol.2020.114777>, 2020a.
- 680 Xu, J., Saleh, M., and Hatzopoulou, M.: A machine learning approach capturing the effects of driving behaviour and driver characteristics on trip-level emissions, *Atmos. Environ.*, 224, 117311, <https://doi.org/10.1016/j.atmosenv.2020.117311>, 2020b.

685 Zheng, T., Bergin, M. H., Johnson, K. K., Tripathi, S. N., Shirodkar, S., Landis, M. S., Sutaria, R., and Carlson, D. E.: Field
evaluation of low-cost particulate matter sensors in high- and low-concentration environments, *Atmospheric Meas. Tech.*, 11,
4823–4846, <https://doi.org/10.5194/amt-11-4823-2018>, 2018.

Zhu, Y., Hinds, W. C., Kim, S., and Sioutas, C.: Concentration and size distribution of ultrafine particles near a major highway,
J. Air Waste Manag. Assoc., 52, 1032–1042, <https://doi.org/10.1080/10473289.2002.10470842>, 2002.