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Abstract

Background: Ambient air ozone is a pulmonary irritant that has been associated with respiratory health effects including increased lung inflammation and permeability, airway hyper-reactivity, respiratory symptoms, and decreased lung function. Ozone exposure estimation is a complex task because the pollutant exhibits complex spatiotemporal patterns. To refine the quality of exposure estimation, various spatiotemporal methods have been developed worldwide.

Objectives: The objective of this work was to compare the accuracy of three spatiotemporal models to predict summer ground-level ozone in Quebec, Canada.

Methods: We developed a land use mixed effects regression (LUR) model based on readily available data (air quality and meteorological monitoring data, road networks information, latitude), a Bayesian Maximum Entropy model incorporating both ozone monitoring station data and the land use mixed model outputs (BME-LUR), and a kriging method model based only on available ozone monitoring station data (BME kriging). We performed leave-one station out cross-validation and visually assessed the predictive capability of each model by examining the mean temporal and spatial distributions of the average estimated errors.

Results: The BME-LUR was the best predictive model ($R^2 = 0.653$) with the lowest RMSE (7.06 ppb), followed by the LUR model ($R^2 = 0.466$, RMSE = 8.747) and the BME kriging model ($R^2 = 0.414$, RMSE = 9.164).

Conclusions: Our findings suggest that errors of estimation in the interpolation of ozone concentrations with BME can be greatly reduced by incorporating outputs from a LUR model developed with readily available data.

Introduction

Tropospheric ozone (O_3) is a photochemical pollutant that has increased globally in concentration since the 19th century (Bogaert et al. 2009). Short and long-term exposure to ambient ozone has been associated with a variety of adverse health outcomes, including respiratory, cardiovascular, neurological conditions, and possibly increased mortality (Chen et al. 2007; EPA 2006; INRS 1997; Jerrett et al. 2009).

Large population studies designed to assess the health risks of ozone exposure need accurate exposure estimates. The assessment of the exposure of a population is a complex task because ozone exposure exhibits complex spatiotemporal patterns, which present considerable modeling challenges. Modeling methods have been developed worldwide to improve exposure assessment of population studies and to capture small spatiotemporal variations in pollutant levels like ozone (Briggs 2005; Jerrett et al. 2005; Zou et al. 2009). For instance, land use regression (LUR) models are used to predict pollutant concentrations at unmonitored sites based on regression models of geo-referenced covariates that predict observed (i.e. measured) data from monitored sites (Beelen et al. 2009; Jerrett et al. 2005). Kriging and the Bayesian Maximum Entropy (BME) framework are interpolation methods that assign a series of weights to observed monitoring station data to compute interpolated values of pollutants at unmonitored sites (Bell 2006; Bogaert et al. 2009; Christakos and Vyas 1998; de Nazelle et al. 2010).

The main objective of this work was to compare the accuracy of three spatiotemporal models to predict ground-level ozone in Quebec (Canada). The models were: a land use mixed effects regression model (LUR) developed with readily available data (air quality and meteorological monitoring data, road networks information, latitude) and two spatiotemporal interpolation models: a combined land use Bayesian Maximum Entropy model incorporating both ozone monitoring station data and the land use mixed model outputs (BME-LUR), and a kriging method based only on available data from ozone monitoring stations (BME kriging).

Methods

Data used for the study

Ozone monitoring data

We retrieved hourly ground level ozone observations for 1990 through 2009 from the National Air Pollutant Surveillance (NAPS) program (Environment Canada, 2012) (Figure 1). We only calculated 8-hour midday (9am – 5pm) ozone concentrations during summer months (May through September) because ozone concentrations during the winter and at night are almost null in Quebec, and included data for all available days with less than 25% missing data (i.e., days with hourly data for at least six of the eight hours).

In Quebec, the number of ozone monitoring stations increased from two stations in 1990 to a total of 50 stations available at the end of 2009. Up to 51 stations were available at some point in time during the period, resulting in 156,060 total observations (station days). All stations had a limit of detection (LOD) of 1 ppb by 1995, and most stations had a LOD of 10 ppb before 1995. Measured 8-hour daily ozone levels were recorded as 0 ppb for 373 observations (station days) during the study period, of which 355 observations were recorded before 1995 when less sensitive instruments were in use. However, these data were retained in analyses as they represented only 0.02% of the observations used to develop the three models.

Road density data

We extracted road density data from the Digital Mapping Technology inc. (DMTI) Road Layer Dataset (2010) and retained major roads, primary and secondary highways, and freeways from all road layers. We measured the total kilometers of such roads within a 1 km buffer around the ozone stations and the road density was expressed in km/πkm^2.

Meteorological data

We obtained meteorological data from the National Climatic Data and Information Archive of Environment Canada for the period of 1990-2009 (National Climatic Data and Information Archive, 2011), between May and September. We extracted mean 8-hour temperature (from 9 am to 5 pm for days with at least 75% of the data available) and daily precipitation records for all weather stations in Quebec. Locations of all available meteorological stations are presented in Figure 1.

Development of models

The following section describes the three models developed to predict exposure to ground-level ozone concentrations (8-hour average) in Quebec and the method used for comparing their predictive ability.

Land use regression mixed effects model

We developed a linear mixed effects regression (LUR) model to predict ozone concentrations measured at monitoring sites with the R software (2010). Temperature, precipitation, day of year, year, road density in a one km buffer, and latitude were the variables used in the model. Temperature and precipitation data were from the closest weather station to each ozone-monitoring site. We shifted and rescaled these variables to produce coefficients of a similar range and to render the intercept interpretable. Specifically, we subtracted 121 from the numeric day of the year to shift its range from 121–274 to 0–153, subtracted 1990 from the year to convert its range from 1990–2009 to 0–19, and subtracted 4,995.9 (the minimum value) from the latitude variable to standardize its range to 0–583.3 km (such that a latitude of 0 represents that latitude of the southerly most ozone monitoring station.)

We used linear splines to model temperature (one knot at 18ºC), road density (one knot at 15 km/πkm^2), and latitude (one knot at 50 km) because their relationships with ozone were not linear. We determined the number and location of the knots by visual inspection, and selected linear splines over cubic splines to increase simplicity, as the results were nearly as good (i.e. the root mean square difference between the prediction of the two models was <0.81 ppb). Therefore, associations with ozone were represented by two model coefficients (one for each linear segment) for each of these variables.

We nested values within stations, which were treated as a random intercept. Thus, we estimated average 8-hour daily ozone concentrations for each observed station-day as follows:

$$O_3 = \beta_0 + \beta_1 X_{low\_temperature} + \beta_2 X_{high\_temperature} + \beta_3 X_{precipitation} + \beta_4 X_{dayofyear} + \beta_5 X_{year} + \beta_6 X_{low\_road} + \beta_7 X_{high\_road} + \beta_8 X_{low\_latitude} + \beta_9 X_{high\_latitude} + u_{station} + \varepsilon \,, \quad [1]$$

BME-LUR and BME kriging analysis

We developed both BME kriging and BME-LUR models for a territory involving census districts of population density greater than 5 people per square km in 2006 (Statistics Canada 2007). This was to ensure that a large proportion of the Quebec population would be covered by the study area, without including areas with very low population. We created a buffer of 50 km around our study area to avoid any edge effects caused by lack of data just outside a census district. Therefore, the selected study region was situated between approximately 42 to 50 decimal degrees North latitude and 65 to 80 degrees East longitude encompassing a total area of 103,110 km^2 (Figure 1).

''Hard" data used to develop the BME kriging and BME-LUR models were the measured ozone concentration data provided by the ozone monitoring stations for all eligible station-days during 1990 to 2009. "Soft" data refers to information that can be used to improve estimates by compensating for the limited amount of measured data. Usually, soft information is based on some *a priori* knowledge of the physical processes that affect the spatiotemporal distribution of the pollutant. For our analysis, the soft data were ozone levels (and their respective normal errors) estimated from the land use mixed effects regression model for 1km x 1km grid cells within the study area for May – Sept, 2005, the year used as the reference year for cross-validation (see below).

Soft data from the LUR model was composed of an ozone estimate for each location as well as an associated error estimate. The error estimated for each modeled point (each center of the 1 km x 1 km grid cell) was the sum of squares of the standard errors from the fixed effects and the square of the standard deviation of the soft random intercept. For the ozone estimate itself (soft data), only the fixed portion of the LUR model was used to create a value, since the mean random effect was 0. There were a total of 278,633 possible grid points per day (~ 42 million spatio-temporal points were possible overall), with the ozone levels estimated using data from the closest meteorological station. Soft data were estimated only when all predictors were available. It was impossible for a large portion (~99%) of points to be estimated due to missing precipitation or temperature data at the closest monitor (mainly in the inhabited northern regions of our study area). However, this did not influence the cross-validation analysis because the analysis was limited to the location of the ozone monitors that had sufficient soft data.

We treated kriging as a special case of the BME in which we used only hard data (i.e. station days with ozone monitoring station data) without including soft data estimates from the LUR model, and thus refer to this model as BME kriging. Because of the spatiotemporal nature of the model used, kriging in this instance refers to a spatiotemporal interpolation of ozone, and not merely a spatial estimate. We implemented the BME-LUR and BME kriging analysis to estimate daily 8-hour average ozone levels at a 1 km^2 grid using Matlab software (2007) and the SEKS-GUI v. 0.69.5 program (Yu et al. 2007).

To account for short-term and small-scale patterns in the ozone data and to remove any spatio-temporal autocorrelative patterns, we used a Gaussian de-trending model (Yu et al. 2007) at a distance of 25 km and a temporal trend of 2 days. This detrending is used to facilitate the interpolation of the remaining stochastic structure of the data. Such detrending algorithms are common in spatial estimation techniques such as kriging. While several detrending methods do exist, the SEKS-GUI provides the Gaussian detrending algorithm as its only detrending option. From visual inspection of time series of ozone levels at monitoring stations, and of spatial distributions of daily ozone levels across all stations, Gaussian detrending appeared to be a sufficient function to remove spatiotemporal trends. The detrended data was then used as our stochastic spatiotemporal dataset for BME kriging and BME-LUR modeling.

Ozone soft and hard data was not normally distributed. We thus corrected soft and hard data using n-scores normalization prior to analysis, as a normal distribution is a necessary condition for accurate estimation by the BME (Yu et al. 2007). We constructed a spatiotemporal covariance model to describe the stochastic processes affecting ozone levels after localized de-trending. We used the resulting model for estimation of the ozone values, followed by de-normalization and re-trending of the estimated value.

Cross-validation

We performed cross-validation to test the predictive ability of the different models and to find the best predictive model. Cross-validation was performed using data from 2005 as a sample year. We did cross-validation for summer days at each monitoring station for which a LUR model estimate could be created (n= 3,986 station days points among 30 stations). In BME-kriging and BME-LUR, we removed all hard data up to one year prior to each cross-validation date at each monitoring station, for the cross-validation at that station. This was done to eliminate the effects of temporally near data. This approach allows for the assessment of the estimation accuracy in different space-time domains, while avoiding the potentially biased interpretation of the estimation results induced by purely temporal autocorrelation (Yu et al. 2009). To perform our cross-validation, we removed a given station day's hard data and estimated it using the remainder of the data (leave-one-out validation). The soft data used for the cross-validation did contain the information from all stations (i.e. the station was not removed during the construction of the LUR), since removing individual stations from the leave-one-out analysis would have had a marginal effect on the construction of the LUR model and subsequent soft data, as each station represents approximately 2% of the data (1/50 stations).

We compared estimation errors (estimated values minus observations) across methods for each station day versus the ozone values for that monitoring station at that time. We used root mean-square errors (RMSE) to estimate the total magnitude of error. We also defined a percent change in mean square error (PCMSE) as used in de Nazelle et al. (2010), where the results correspond to the percent increased or decreased estimation accuracy of the ozone concentration prediction based on the LUR or BME kriging models compared with corresponding predictions based on the BME-LUR. We assessed visually for unusual spatial or temporal patterns in the distributions of the average estimated errors (estimated versus observed data).

Lastly, we compared observed exceedances of the 8-hour Canadian Ambient Air Quality Standard (i.e., 65 ppb) identified using monitoring station data to exceedances identified using model estimates. To do so, we first transformed monitored and estimated ozone data variables into binary variables (0 = no exceedance, 1 = exceedance) and compared the estimated exceedances to the observed exceedances using Cohen's kappa measure of agreement.

Results

Table 1 presents the description of the data used for the development of the LUR model for the years 1990–2009. Predictors and ozone data were available at 39 ozone monitoring stations on 2,441 days. Since information was not available concurrently at all stations and all days, we used 29 685 spatiotemporal points (station days) out of 118,560 possibilities (152 days × 20 years × 39 stations) to develop the model. These 29,685 points are spatiotemporal moments where we concurrently had information on ozone levels, temperature, and precipitation. Eight-hour ozone concentrations varied from 0 to 104 ppb, eight-hour temperatures varied from -3.5 to 33.9ºC, daily precipitation varied from 0 to 123.8 mm/day, and road density from 0 to 25.4 km/πkm^2. The range of latitude values was between 0 and 583.3 km.

The LUR model is found in Table 2. Considering the estimated effect size (see footnote of Table 2 for clarification on the calculation) of each variable, temperature, day of the year, and road density were the main predictors. In this model, coefficients for linear spline functions of temperature (≤18ºC and >18ºC) were positively associated with ozone concentrations while precipitation, day of the year, year, and coefficients for linear spline functions of low and high road density and of low latitude (<50 km) were negatively associated with ozone levels. Overall, all predictors had a significant association, except the coefficient of the linear spline function for high latitude. To better visualize the fixed effects, LOESS plots of bivariate relationships of these predictor variables are presented in the Supplemental Material, Figure S1. Every coefficient of the LUR model was in agreement with the LOESS plots, and with known processes of the formation and the destruction of ozone, except for cold temperature. Based on the LOESS plot, we expected temperatures between -3.5 and 18°C to have no relation with ozone, or the relation to be slightly negative, while in the LUR model, after controlling for latitude, year and day of the year, the relation between ozone and the lowest temperatures was slightly positive.

Table 3 describes hard and soft data used to build BME-LUR and BME kriging models. Hard data were observations at monitoring sites for 1990–2009 (n = 103,669 out of 156,060 station days with ozone data), while predicted soft data estimates were derived from the fixed effect portion of the LUR model and errors estimated from the fixed and random effects of the same model for the year 2005 only (152 days). Therefore, we could estimate 90,847 spatiotemporal points from the LUR model, considering the availability of temperature and precipitation information concurrently, out of around 42 million maximum possible spatiotemporal points (152 days × 278,633 possible grids points per day in our study area). For BME kriging and

BME-LUR, we used the same de-trending and covariance structures to describe the spatiotemporal covariance pattern in the data. The covariance model used to fit the measured spatiotemporal covariance of the data consisted of two components: a short-term (2-day exponential) long-distance (100-km exponential) trend that described the majority of the variability (covariance = 0.9), and a second component (covariance = 0.1) describing the weekly (3-day cosinusoidal) trend in covariance in time with a small spatial (i.e. local 12.5 km exponential) scale due to the cyclic nature of ozone in urban stations in Quebec, where ozone tends to be lower on the weekends and rises during week days. Modeled covariances as derived from the information above are presented in the Supplemental Material, Figure S2.

Table 4 describes the cross-validation results for the three models, for the year 2005 at the 30 stations available to produce the soft data with all mixed model predictors (n = 3,980). For the BME-LUR, on June 25th, estimates at 6 stations, all located in the southeastern portion of the study area could not be estimated with the BME-LUR. On that day, all measurements at these stations were high (hard data) (75-78 ppb) when compared to the range of values of the calculated soft data (28-48 ± 6.6 ppb) for that day. Overall, the BME-LUR was the most predictive model (R^2 = 0.653), and had the lowest RMSE (7.06 ppb). The LUR model performed better and with greater precision (R^2 = 0.466, RMSE = 8.747) than the BME kriging model (R^2 = 0.414, RMSE = 9.164). The BME-LUR outperformed the LUR model and BME kriging by 19.9% and 23.0% using PCMSE, respectively. Finally, the Cohen's kappa of the BME-LUR (n: 18 predicted exceedances; kappa = 0.525, 95% CI: 0.495-0.555) obtained from the comparison of 8-hour Canadian Ambient Air Quality Standard (65 ppb) monitored (n: 34 observed exceedances) and estimated concentrations suggests moderately good agreement between the model and the measurements. The BME-LUR outperformed both BME-kriging (n: 39 predicted exceedances; kappa = 0.169, 95% CI: 0.138-0.200) and the LUR model (kappa = 0 as no predicted value above 65 ppb).

A graph of the distribution of errors in the ozone concentration estimates generated by each model (i.e., the difference between estimated and observed values) based on the leave-one-out analysis also demonstrated that the BME-LUR was the more accurate model (Figure 2). As can be observed in Figure 3, the RMSE of the three models appears stochastic in time. Figure 4 also shows that the RMSE of the BME-LUR in space (at all stations) was closest to zero in comparison to BME-kriging and the LUR. Figure 5 represents a map of predicted mean daily ozone levels (9h00-17h00) and standard errors (SE) at one km grid across the greater Montreal region for the summers 2006–2009. Levels of ozone are higher around the suburbs of Montreal compared to downtown metropolitan areas and concentrations are also greater in places far from highways (Figure 5a). Moreover, greater difference between observed and estimated ozone concentrations may be found in the northeast of the greater Montreal (Figure 5b).

Discussion

Overall, our findings suggest that error of estimation in the interpolation of ozone concentrations using the BME method may be improved with the inclusion of a LUR model developed with readily available database.

We found that the estimation of ozone across monitoring sites was more accurate with the BME-LUR model compared with other models; this difference was close to 20% in R^2 and around 2 ppb in RMSE. These results are consistent with previous work. For instance, Yu et al. (2009), which modeled air pollutant concentrations in North and South Carolina (USA), found that the integration of soft information by the BME method effectively increased the estimation accuracy for ozone predictions compared with estimates derived using BME kriging. Yu et al. (2009) used measurements from monitoring stations as soft data, whereas we created soft data from outputs of a LUR model. In Yu et al. (2009), the R^2 and RMSE values were not reported, but the mean and standard deviation of their estimation errors for daily estimates were similar to ours (Yu: kriging = 0.483 ± 7.035 and BME = 0.177 ± 6.845 ppm; present study: kriging = 0.414 ± 9.164 and BME-LUR = 0.653 ± 7.057). de Nazelle et al. (2010) also found better predictive accuracy for the representation of space-time ozone distribution in North Carolina with a BME model based on observed (hard) and modeled (soft) data from a stochastic analysis of an urban-intercontinental-scale atmospheric chemistry transport model, compared with kriging method estimates based on hard data only. We found that, similar to de Nazelle et al. (2010), ozone estimates further away from monitoring stations were more accurate when soft data was used in the BME versus kriging alone. As in our work, their PCMSE values were always negative (between -1.486 and -27.699 depending on the cross-validation radii of exclusion points), indicating that the integration of observed and modeled prediction was consistently more accurate than relying solely on observations. Furthermore, agreement between modeled and observed Canadian Ambient Air Quality Standard exceedances was highest for estimates based on the BME-LUR.

We found that error estimates from the BME-LUR model were more accurate where monitoring stations were clustered in the region of the study, such as in the southern (i.e. more urban) part of Quebec (Figure 4). This result is consistent with Yu et al. (2009), which indicated that the locations where the estimates exhibit higher discrepancies from the data values were mostly close to regions of data scarcity.

We showed that the LUR model was slightly more accurate (lower RMSE) than the BME kriging model (Table 4). Coefficients of the LUR model indicated that linear spline functions of temperature were positively associated with ozone concentrations, while precipitation, day of the year, year, and coefficients for linear spline functions of low and high road density and of low latitude were negatively associated with ozone levels (Table 2). The LUR model coefficients for the spline temperature variable are in line with the expected trend (EPA 2006) and suggest an increase of ozone with temperature, which is more pronounced at higher temperatures. With regards to road density, both coefficients for linear spline functions of low and high density were negative, and this may be explained by the fact that at regional scale, low traffic represents lower concentrations of ozone precursors (traffic related pollutant such as NO_x), while at the local scale, low traffic represents lower destruction of ozone. The other fixed effects of the LUR model are also in agreement with the known atmospheric processes of ozone and highlight that its formation rely on various factors such as sunlight. Ozone concentrations are also greater with altitude and show diurnal and weekly variations with higher levels during weekdays (EPA 2006; Finlayson-Pitts and Pitts 1997). Lastly, the negative coefficient found for day of the year variable highlight the small intra-annual decrease in ozone levels from May to September in Quebec.

Nevertheless, the fact that the LUR model was slightly more accurate than BME kriging is inconsistent with what was found by Beelen et al. (2009) who developed maps of ozone levels across the European Union using a regression model with altitude, distance to sea, major roads, high-density residential areas and a combination of meteorological data as predictors. They obtained values of R^2 = 0.54/0.38 and RMSE = 8.63/8.74 ppb respectively for the regression and kriging models at rural scale. At urban locations, kriging was more accurate than the regression model with only the high-density residential predictor (regression/kriging: R^2 = 0.38/0.61 and RMSE = 7.32/5.84). Kriging methods predict well when a dense and representative monitoring network is available (Briggs 2006; Jerrett et al. 2005; Laslett 1994). In our study, BME-LUR was more accurate in estimating ozone levels than LUR and BME kriging at urban and suburban scales (i.e., island of Montreal and its surrounding area), and LUR was more accurate than BME kriging in urban areas only (Figure 4). In Quebec, the monitoring station network is relatively sparse and the good correlations between the predictors used in the LUR model and the measured ozone concentrations at monitoring stations may at least partially explain the relatively weak performance of BME kriging.

We created maps representing mean ozone levels (9h-17h) and standard error predictions from the BME-LUR at one km grid for summers 2006-2009 to visualize how the model would estimate ozone in urban and suburban areas of the greater Montreal region. As observed in Figure 5, levels of ozone are higher around Montreal Island (suburban areas) compared to downtown metropolitan (center of Island) areas and concentrations are also greater in areas far from highways. This may be explained by the fact that the efficiency of ozone production depends on NO_x concentrations. In areas with low NO_x concentrations (e.g. in rural areas), ozone production increases with higher levels of NO_x. In downtown metropolitan areas where the highest NO_x concentrations may be found, there is net destruction of ozone by reaction with NO (EPA 2006). Also, we found greater difference between observed and estimated ozone concentrations in the northeast of the greater Montreal as indicated by Figure 5b, and this may be explained by the possible incongruity between soft and hard data points, hard data points themselves, or by a possible lack of ozone stations outside the Montreal area.

As mentioned previously, 6 stations could not be computed with the BME-LUR on June 25th. In-depth analysis reveals that all these stations had high monitored values (hard data) when compared to the range of values of the calculated soft data for that day. To our knowledge, this issue has not been reported elsewhere in the literature and investigations of BME estimation failure should be realized in future studies.

The developed BME-LUR model presents other limitations. For instance, the meteorological variables (temperature and precipitation) used to estimate soft data do not represent the complete atmospheric processes of ozone. This would have been more correctly assessed with the use of some integrated meteorology models like the Community Multiscale Air Quality (CMAQ) modeling system. However, such models do not capture small area estimations such as our LUR model predictions (US EPA 2012). Another limitation is that the LUR model predictions were only estimated for each one km grid of the territory due to computational constraints, as adding soft data at 100 m resolution would have dramatically increase the amount of time needed to run the BME-LUR. Computational time required to create maps is another limitation. In this study, 90 days were needed to create maps of ozone levels for an area of 103,110 km^2 at a resolution of 1 km while running multiple processors on a high-powered computer (2.93 GHz 4-core processor and eight concurrent threads with 6 gb RAM). This computational time can be improved by reducing the resolution of the study area or the number of soft data points, as well as by estimating only points of interest (e.g. residential addresses of interest vs 1 km grid).

Despite the computational demands, the BME-LUR adds value to the ozone exposure estimation because it generates the complete probability distribution of exposure at each point in space and time (Yu et al. 2007) and it reduces the estimation errors. This may lead to less biased effect measures and greater statistical power in health studies (Baker and Nieuwenhuijsen 2008; Briggs 2005; Goldman et al. 2012).

For implementation in future health studies, the BME-LUR might be improved by including additional predictors in the LUR model, such as population density, land use, topography, and industrial sources of precursors (Hoek et al. 2008). As noted by Beelen et al. (2009), stratification of the study area (e.g. separating urban and rural areas) could also improve model predictions.

Conclusions

We aimed at comparing the ability of three spatiotemporal models to predict ground-level ozone in Quebec (Canada) to improve ozone health risks assessment. The BME-LUR model appeared to be the best model for exposure prediction. This work illustrated the accuracy of the BME-LUR models to predict air pollutants such as ozone across space and time over LUR and BME kriging methods and that error of estimation in the interpolation of ozone concentrations can be greatly reduced using outputs from a LUR model that can be developed with readily available data.

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Table 1. Descriptive statistics of variables used for the development of the LUR model for years 1990-2009.

Variables	Number of spatiotemporal points[a]	Mean ± SD	Min	Max
8h ozone concentration (ppb)	29,685	31.2 ± 13.1	0.0	104.0
8h temperature (ºC)	29,685	19.1 ± 5.3	-3.5	33.9
Precipitation (mm/day)	29,685	3.0 ± 7.1	0.0	123.8
Road density (km/πkm^2)	39	6.4 ± 7.9	0.0	25.4
Rescaled latitude (km)	39	114.6 ± 134.6	0	583.3

Abbreviations: SD: standard deviation; Min: Minimum; Max: Maximum.

[a]29,685 station days out of 118,560 possible station days (limited by temperature and precipitation variables) were used for the development of the LUR model.

Table 2. Summary of the LUR model for ozone concentrations in the region of study (1990-2009)[a].

Fixed effects	Coefficients	SE	Effect size[c]
Constant	39.530	1.577	-
Temperature ≤18ºC[b]	0.218	0.021	39.461
Temperature >18ºC[b]	2.139	0.019	-
Precipitation	-0.010	0.001	-1.238
Day of the year	-0.107	0.001	16.371
Year	-0.165	0.018	3.315
Road density ≤15 km/πkm^{2}[b]	-0.255	0.098	-14.995
Road density >15 km/πkm^{2}[b]	-1.074	0.219	-
Latitude ≤ 50 km[b]	-0.123	0.038	1.687
Latitude >50 km[b]	0.003	0.003	-

Abbreviations: SE: Standard error.

[a]For the random effect, the standard deviation of intercept is 2.464 (95%CI: 1.915-3.170); the standard deviation of residuals of mixed model is 8.904. [b]Variables modeled as linear spline functions to account for nonlinear relations with ozone. [c]The effect size was calculated by $\beta_i V_{iMax} - \beta_i V_{iMin}$ for non-splined variables, and by $\beta_{iLower} V_{iSpline} - \beta_{iLower} V_{iMin} + \beta_{iUpper}(V_{iMax} - V_{iSpline})$ where $V_{iSpline}$ is the value of the knot of the variable of interest, β_{iLower} the coefficient for values lower than the knot value, and β_{iUpper} the coefficient for values greater than the knot value.

Table 3. Statistics for measured (hard) ozone data (1990–2009) and predicted and error "soft" data from the LUR (year 2005) used for BME-LUR and BME kriging models.

Variables	Number of spatiotemporal points	Mean ± SD (ppb)	Min (ppb)	Max (ppb)
Hard data (n=51)	103,669[a]	30.6 ± 12.5	0.0	110
Soft data at a 1 km grid (predicted)	90,847[b]	46.3 ± 9.3	12.1	76.4
Soft data (error)	90,847[b]	6.9 ± 1.8	5.5	63.9

Abbreviations: SD: standard deviation; Min: Minimum; Max: Maximum.

[a]103,669 out of 156,060 station days with ozone data (limited by ozone data availability only) were used as hard data for BME-LUR and BME kriging models. [b]90,847 spatiotemporal points with data for temperature and precipitation were estimated as soft data for 2005 out of approximately 42 million maximum possible spatiotemporal points (152 days × 278,633 possible grid points per day in our study area).

Table 4. LUR, BME kriging, and BME-LUR models leave-one-station-out cross-validation results for year 2005, n=30 ozone monitoring stations (estimated points: 3,980).

Methods	**R^2**	**RMSE (ppb)**	**PCMSE**
LUR	0.466	8.747	-19.9%
BME kriging	0.414	9.164	-23.0%
BME-LUR	0.653	7.057	-

Abbreviations: RMSE: Root mean-square errors; PCMSE: percent change in mean square error.

Figure legends

Figure 1. Geographical location of ozone monitoring (black circles) and meteorological stations (grey squares) in the study region (dark grey). Locations are for monitor used at any time during the study period.

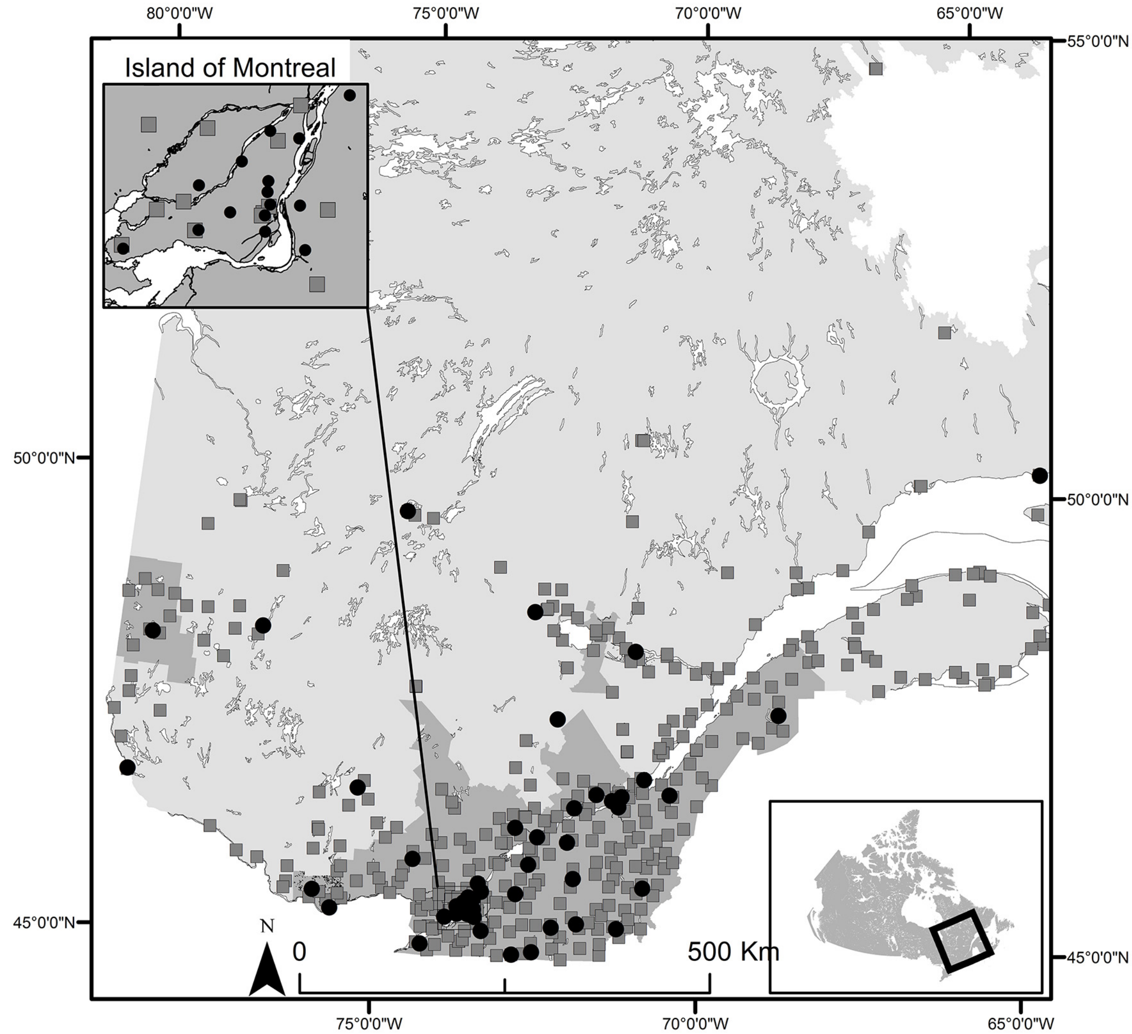
Figure 2. Ozone mapping error estimates from the leave-one-station-out cross-validation [where error = estimated – measured (observed) ozone concentration (in ppb) at each monitoring station] based on the LUR (long dashes, mean ± standard deviation = 0.282 ± 8.93 ppb), BME kriging (short dashes, 0.130 ± 9.804 ppb), and BME-LUR (solid line, 1.339 ± 7.086 ppb) models for the year 2005.

Figure 3. Mean temporal ozone error estimates (RMSE) based on the leave-one-station-out cross-validation for LUR (long dashes), BME kriging (short dashes), and BME-LUR (solid line) models for the year 2005.

Figure 4. Spatial distribution of mean ozone error estimates (RMSE) in the study area (year 2005) based on the leave-one-station-out cross-validation for LUR, BME kriging, and BME-LUR models.

Figure 5. Mean ozone levels (9h00-17h00) (a) and standard errors (SE) (b) predicted from the BME-LUR at one km grid across the greater Montreal region in Quebec (Canada) for the summers 2006-2009.

Figure 1.



80°0'0"W
75°0'0"W
70°0'0"W
65°0'0"W
55°0'0"N
Island of Montreal
50°0'0"N
50°0'0"N
45°0'0"N
45°0'0"N
N
0
500 Km
75°0'0"W
70°0'0"W
65°0'0"W

Figure 2.

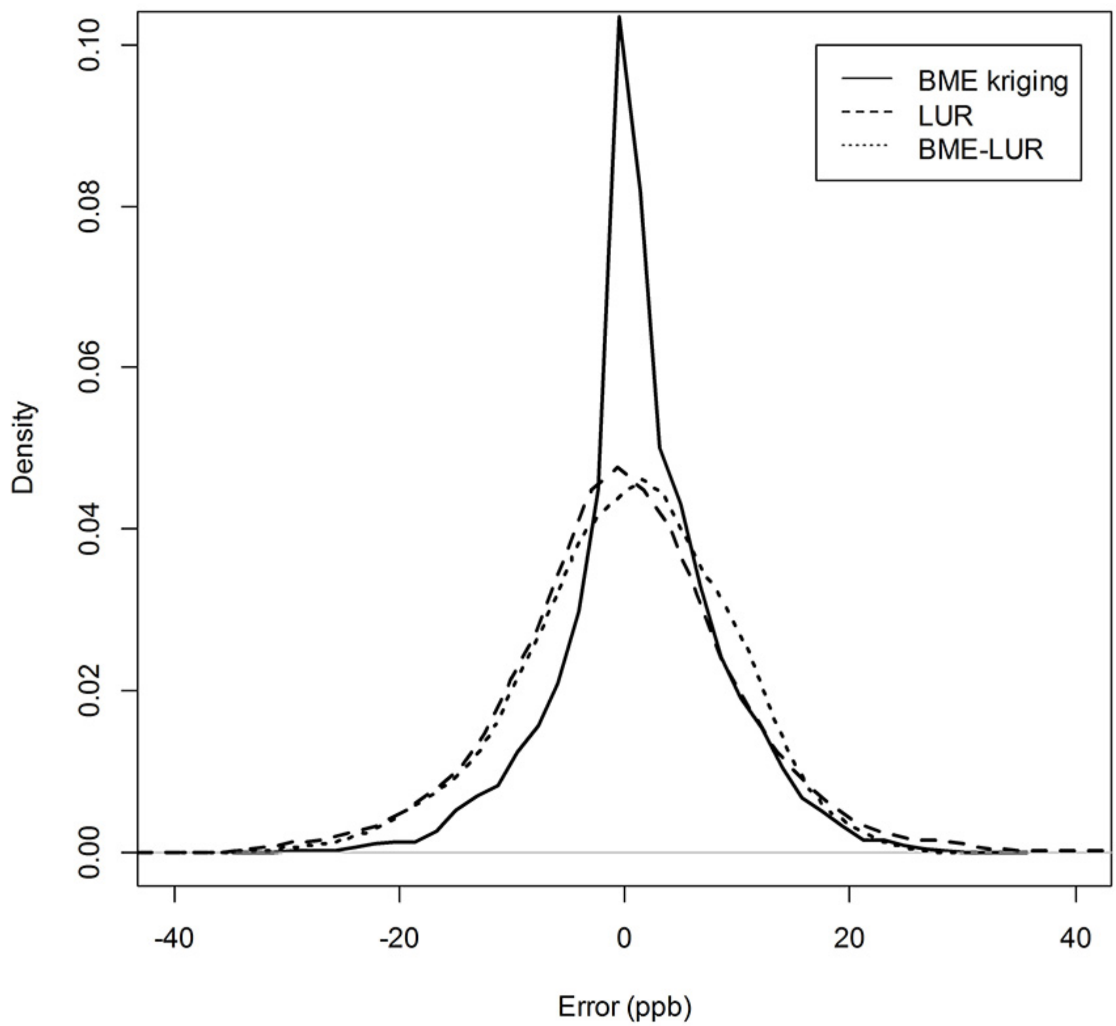
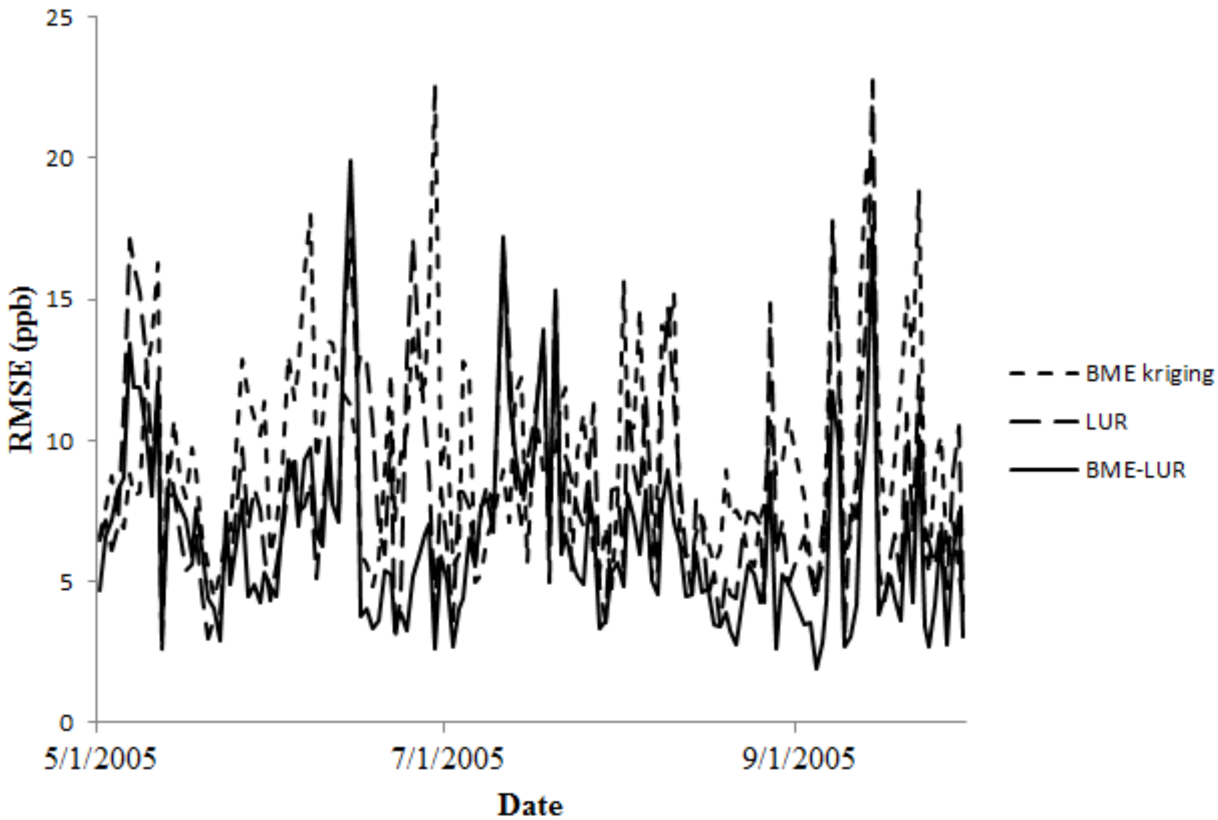
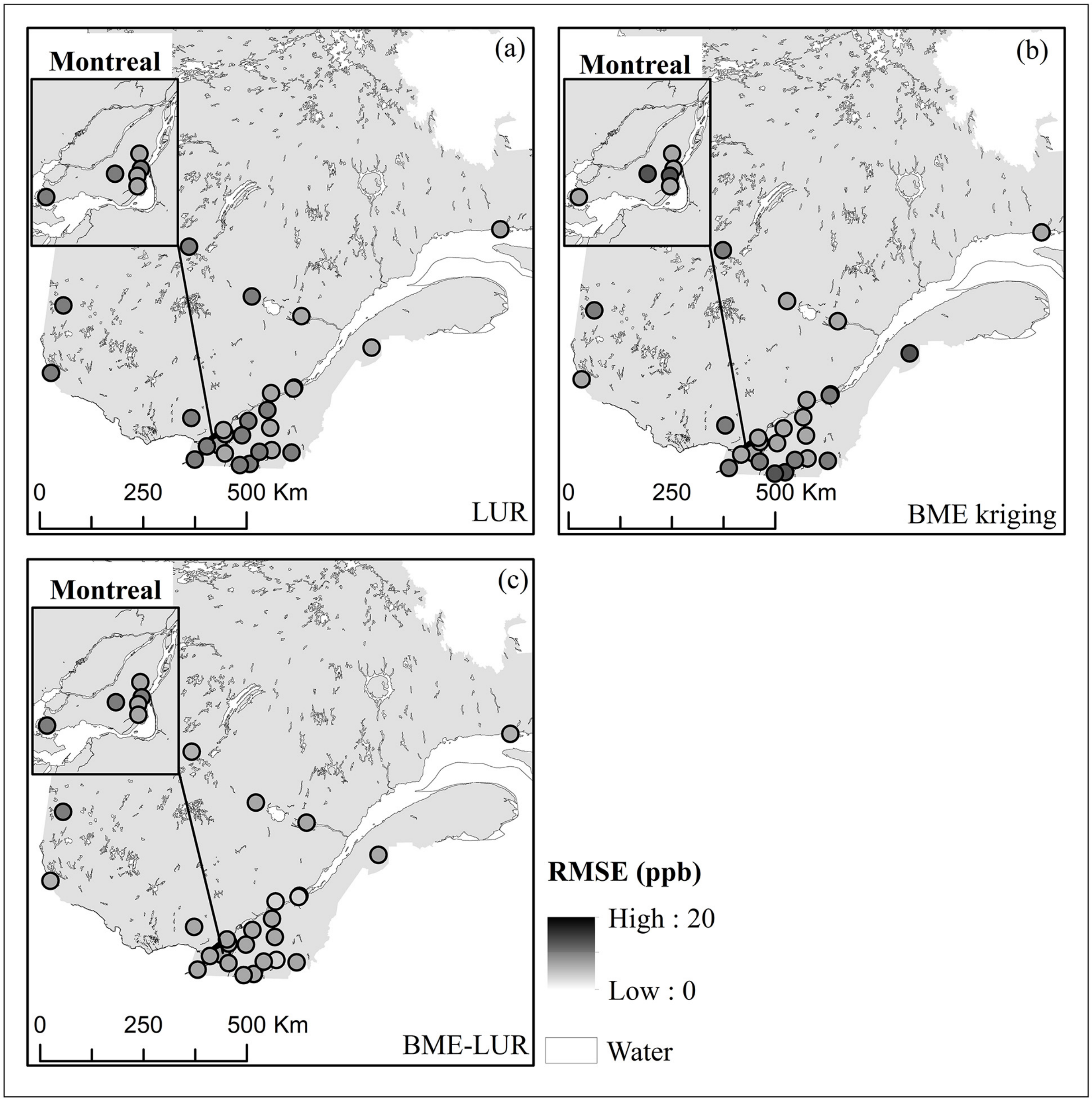

BME kriging
LUR
BME-LUR
Density
0.00
0.02
0.04
0.06
0.08
0.10
-40
-20
0
20
40
Error (ppb)

Figure 3.



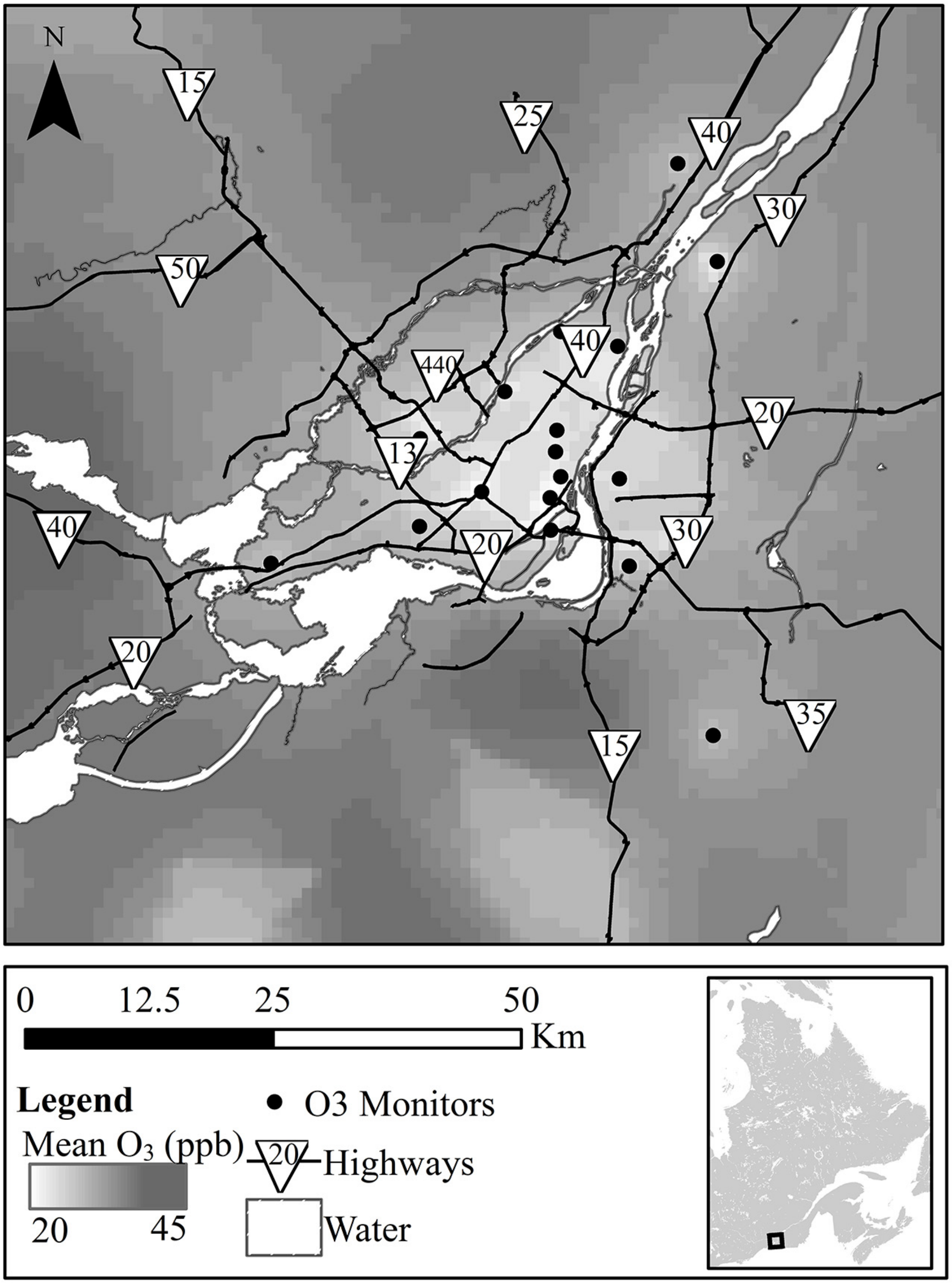
25
20
15
10
5
0
RMSE (ppb)
5/1/2005
7/1/2005
9/1/2005
Date
BME kriging
LUR
BME-LUR

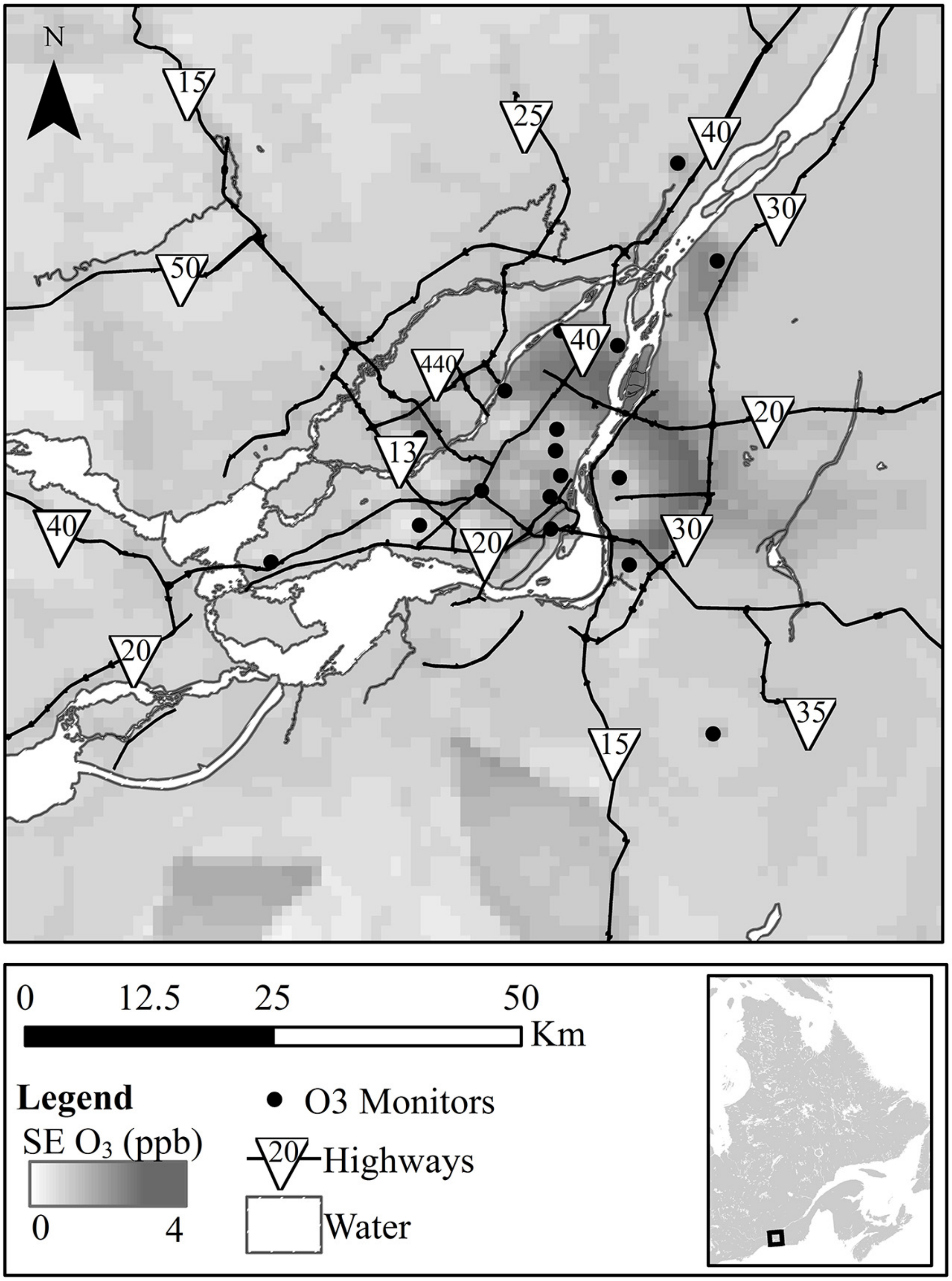
Figure 4.



Montreal
(a)
0
250
500 Km
LUR
Montreal
(b)
0
250
500 Km
BME kriging
Montreal
(c)
0
250
500 Km
BME-LUR
RMSE (ppb)
High : 20
Low : 0
Water

Figure 5.


N
15
25
40
30
50
40
440
20
13
40
30
20
20
35
15
0
12.5
25
50
Km
Legend
O3 Monitors
Mean O_3 (ppb)
20
Highways
20
45
Water


N
15
25
40
30
50
40
440
20
13
40
20
30
20
35
15
0
12.5
25
50
Km
Legend
SE O3 (ppb)
0
4
O3 Monitors
20
Highways
Water