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Evaluating methods for spatial mapping: Applications for estimating ozone concentrations across the contiguous United States

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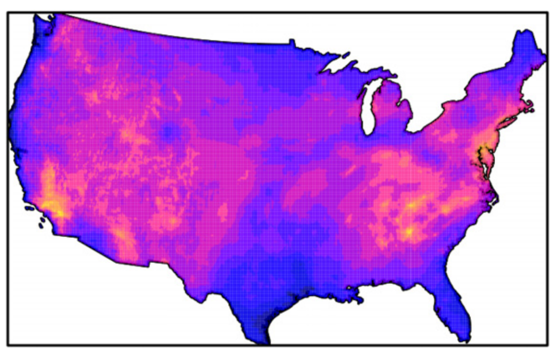
HIGHLIGHTS

- Evaluated method performance for predicting and mapping national ozone pollution.
- Compared land use regression, IDW, ordinary and universal kriging for prediction.
- Land use regression models revealed the presence of residual spatial variation.
- Kriging outperformed the other approaches for predicting ozone concentrations.

GRAPHICAL ABSTRACT



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ABSTRACT

Understanding spatial variability of air pollutant concentrations is critical for public health assessments. Our goal is to examine ground-level ozone and comparatively evaluate method performance for predicting and mapping national concentrations across the United States, while assessing the importance of accounting for spatial variability.

Cross-sectional US EPA ozone monitoring data was acquired for three days in 2006, plus environmental covariates of land use, traffic, temperature, elevation, and population. Evaluation of ozone variability was assessed with land use regression (LUR) and spatially explicit kriging models. Ozone concentration was predicted with four approaches, including LUR, inverse distance weighting (IDW), ordinary kriging, and universal kriging, and evaluated with a Monte Carlo cross-validation simulation. Results were mapped for the continental United States.

Temperature, elevation, and distance to major roads were significantly related to ozone concentrations and examination of spatial dependence on LUR models revealed the presence of residual spatial variation. Cross-validation results found kriging outperformed both LUR and IDW in terms of root mean squared prediction error. We demonstrate that national-level ozone is best evaluated using the statistically optimal kriging models, which account for residual spatial variation. Universal kriging was preferred over

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ordinary kriging by allowing us to assess the significance of environmental covariates both for inference and prediction of ozone concentrations.

Abbreviations

AQS	Air Quality System
BLUP	Best linear unbiased predictor
CV	Cross validation
DEM	Digital elevation model
ESRI	Environmental Systems Research Institute
GLS	Generalized least squares
IDW	Inverse distance weighting
LUR	Land use regression
OK	Ordinary kriging
OLS	Ordinary least squares
ppb	parts per billion (by volume)
RMSE	Root mean squared error
UK	Universal kriging

1. Introduction

Characterizing exposures to air pollutants is critical for epidemiological studies. Ozone air pollution is linked to adverse health outcomes including respiratory related morbidity, such as decreased pulmonary function (Foster et al., 2000; Kim et al., 2011), asthma exacerbations (McDonnell, 1999), respiratory related hospital admissions (Burnett et al., 2001; Choi et al., 2011), and premature mortality (Bell et al., 2004; Jerrett et al., 2009). Ozone exposure presents population wide risks and in particular to susceptible groups, such as children, elderly, and individuals with pre-existing respiratory disease (Kim et al., 2011; Burnett et al., 2001; Bell and Dominici, 2008; Salam et al., 2005). The detection of small relative risks associated with individual exposure to air pollutants necessitate the need for population-level exposure modeling and makes exposure estimation a critical component of health effects studies (Bell et al., 2004; Levy et al., 2005; Berman et al., 2012).

Characterization of air pollution exposure can be a complex process, especially for population-based studies. Ambient concentrations from regulatory networks are common surrogates for individual exposure, (Bell et al., 2004; Levy et al., 2005; Pope et al., 2002; Samet et al., 2000) but monitors are often spatially heterogeneous with limited geographic coverage. Counties without air monitors tend to be rural, older, and have greater poverty levels, leading to increased vulnerability among their population (Bravo et al., 2012). To overcome these limitations, the identification of environmental determinants influencing air pollutants allows for improved inference about variability in pollutant concentrations. When predicting air pollutant exposures, the literature has found factors such as land use, population density, temperature, elevation, and traffic to all be significant in explaining concentrations.

Extrapolating data to unsampled locations (spatial prediction), allows us to create pollution maps of exposure for epidemiology applications, environmental health, and related policy research (Beelen et al., 2009). A popular approach for prediction and identifying environmental determinants of pollutant concentration is land use regression (LUR). First introduced by Briggs et al. (1997), it utilizes a geographic information system (GIS) to combine monitored air pollution data with land use and environmental variables for building regression covariates, and is increasingly popular in both European (Beelen et al., 2009; Briggs et al., 1997; Hoek et al., 2011; Freire et al., 2010) and North American studies (Clougherty et al., 2008; Henderson et al., 2007; Ross et al., 2007; Poplawski et al., 2008; Wilton et al., 2010; Ryan and LeMasters, 2007; Su et al., 2011, 2009). It has been argued that LUR results in better spatial predictions of small area variability compared to alternative approaches, notably at city-level geographies where LUR is most frequently (but not exclusively) applied (Ross et al., 2007; Brauer et al., 2003). Statistically the LUR model is equivalent to multivariate linear regression and trend surface modeling, both which assume a regression error structure of independence (Cressie, 1993; Schabenberger and Gotway, 2005). However, the performance of a statistical regression can be suboptimal in the presence of residual autocorrelation (Cressie, 1993; Schabenberger and Gotway, 2005; Gaffney et al., 2005; Jerrett et al., 2003). While this is appropriately handled through the introduction of spatial covariance functions (Freire et al., 2010; Szpiro et al., 2010; Franklin et al., 2012), it is not universally performed (Hoek et al., 2011; Poplawski et al., 2008; Ryan et al., 2007).

An alternative approach for spatial modeling is kriging. This regression-based method allows the inclusion of covariates either for directly quantifying effects and/or to improve spatial predictions. Kriging also automatically includes a spatially structured residual component to capture and account for spatial variation not explained by the regression covariates (Cressie, 1993; Schabenberger and Gotway, 2005). In a regression framework kriging is known to produce spatial predictions that are statistically optimal and best linear unbiased predictions (BLUPs) (Cressie, 1993). Kriging has been previously used as a tool to assess exposure to air pollutants, including ozone and particulate matter at both the regional city-scale (Künzli et al., 2005; Jerrett et al., 2005) and national-level (Beelen et al., 2009; Sampson et al., 2013).

Despite a variety of spatial methods used in environmental exposure studies, limited papers have comprehensively evaluated prediction approaches for exposure assessment settings (Bravo et al., 2012; Beelen et al., 2009; Marshall et al., 2008; Mercer et al., 2011; Son et al., 2010; Jerrett et al., 2004). A study by Jerrett et al. (2004) reviewed six approaches assessing air pollution prediction, including inverse distance weighting, kriging, and LUR, but was restricted to an intra-urban environment. At a global-scale Beelen et al. (2009) compared kriging and regression mapping predictions of background air pollution, but their study focused on Western Europe. A recent paper compared Bayesian maximum entropy LUR and kriging for predicting ozone concentrations across Quebec, Canada (Adam-Poupart et al., 2014). These papers demonstrate the importance for comparing method performance, especially when considering different geographic regions and spatial scales. Understanding modeling and mapping of pollutant concentrations is critical not only for assessing exposures, but for estimating health effects and regulatory benefits (Berman et al., 2012; Son et al., 2010).

This paper provides a comparison of methods for characterizing large-scale air pollution concentrations by comparing and evaluating statistical models for predicting ground-level ozone concentrations. Additionally, we produce optimal maps of ozone concentrations and examine environmental covariates expected to be important in understanding national ozone patterns. Land use regression and kriging-based methods will be looked at from the national level, which differs from the city-scale approach applied in most LUR studies. An applied Monte Carlo approach allows a robust examination of prediction performance at each individual air pollution site through multiple iterations. The conclusions are meant to inform about model prediction performance using US data, limitations of methods and the potential consequences of failing to account for residual spatial variation.

2. Materials and methods

2.1. Data sources

2.1.1. Ozone monitor data

Ozone air monitoring data for the US was acquired from the EPA's Air Quality System (AQS) (US Environmental Protection Agency, 2014) for June 1, July 3, and August 1, 2006. The study days were a cross-sectional snapshot of random weekdays during the national ozone season for methodological comparison. Ancillary data includes longitude/latitude coordinates of monitor locations and daily maximum 8-h ozone measurement in parts per billion (ppb) by volume. Alaska, Hawaii, and Puerto Rico were excluded. Prior to release, the EPA checked this data for validity and precision (US Environmental Protection Agency, 2005).

2.1.2. Census and land use data

We used a ESRI® population shapefile created from the Topologically Integrated Geographic Encoding and Referencing (TIGER) database of the US Census Bureau (ESRI, 2012). Population density per square kilometer was calculated for 64,906 continental US census tracts for 2005. A US land cover database from 2006 was downloaded from the US Geological Service (US Geological Survey, 2012) and aggregated to 1×1 km grid cells for easier handling by assigning grid cells the classification contributing the most area. We examined the 16-unit land cover classes defined by the Land Cover Institute (LCI). Additionally, two broader categories of land use were further evaluated based upon the LCI single digit codes and our own five categories based on USGS codes (none (0), water (11), developed (21–24), natural (31–71; 90–95), agriculture (81–82)).

2.1.3. Meteorological and topographical data

Daily maximum temperature (° C) was obtained from the National Oceanographic and Atmospheric Agency (NOAA) climate prediction center as a 5×5 km raster file (NOAA, 2012). NOAA predicts grid estimates from ∼5000 weather stations using a Cressman-Scheme weighted average. A NOAA digital elevation model (DEM) raster file was acquired at a 1×1 km grid (NOAA National Geophysical Data Center, 2012). Coastal winds have been shown in other studies to have a significant effect on air pollution levels (Beelen et al., 2009; Entwistle et al., 1997; Ross et al., 2005), therefore a distance-to-coast raster estimate was acquired (Hengl, 2009).

2.1.4. Primary roadway data

Primary roads (e.g., interstates) in the United States were acquired as a shapefile from the US Census TIGER/Line database (TIGER Products, 2013). The shapefile represents each road as multiple straight-line segments (12,664 total road segments with a median length of 9 km). A variable was created for distance from each ozone monitor to the closest road segment, while a second variable calculated the average distance from each monitor to the closest one percent of road segments. This second variable provides greater weight to ozone monitors in close proximity to multiple interstates.

Table 1 provides a summary of these data sources and their associated temporal and spatial scales. Selected variables, such as temperature, elevation, land use, distance to coast, and population density are commonly found in literature exploring ozone for epidemiological and exposure purposes (Beelen et al., 2009; Jerrett et al., 2005; Marshall et al., 2008; Mercer et al., 2011). With respect to this being a national-scale assessment of prediction performance, the covariate selection process was not exhaustive nor did we explore unique variables. Localized LUR characteristics, such as traffic density and primary roads, are associated with NO_x and fine particulates and were not included due to lack of availability and data size. This alludes to the complexity of incorporating environmental data at the national scale, where obtainability can be prohibitive.

2.2. Spatial regression

Four spatial prediction methods: universal kriging (UK), ordinary kriging (OK), Land Use Regression (LUR), and inverse distance weighting (IDW) are considered. The first three methods are examples of the general linear regression model

$$\begin{aligned} Y(\boldsymbol{s}) &= \beta_0 + \beta_1 X_1(\boldsymbol{s}) + \cdots + \beta_p X_p(\boldsymbol{s}) + \varepsilon(\boldsymbol{s}) \\ &= \mu(\boldsymbol{s}) + \varepsilon(\boldsymbol{s}) \end{aligned} \quad (1)$$

Table 1
Summary of AQS ozone monitor and environmental data used to build spatial models of ozone air pollution. Information includes date, spatial data type, and spatial resolution.

Covariates	Date	Data type	Resolution
AQS ozone conc[a] (ppb by volume)	June 1, 2006 July 3, 2006 August 1, 2006	Points	$N = 1,104$[b] $N = 1,142$ $N = 1,114$
Temperature (° C)	June 1, 2006 July 3, 2006 August 1, 2006	Raster	5 km ×5 km
Population density (km^2 by census tract)	2005	Shapefile	US census tracts
Elevation (DEM) (m)	1999	Raster	1 km ×1 km
Land Use	2006	Raster	1 km ×1 km
Distance-to-coast (km)	2009	Raster	5 km ×5 km
Primary roads	2013	Shapefile	Line segments

[a] Longitude/latitude of AQS air monitor sites were additional covariates.
[b] Final number of included AQS monitors.

where $\boldsymbol{s}$ represents spatial coordinates, $Y(\boldsymbol{s})$ represents measured ozone at monitor location $\boldsymbol{s}$, $X_1(\boldsymbol{s}), \ldots X_p(\boldsymbol{s})$'s environmental covariates indexed by location $\boldsymbol{s}$, $\beta_1, \ldots, \beta_p$'s their associated effects, $\mu(\boldsymbol{s})$ is the mean trend, and $\varepsilon(\boldsymbol{s})$ the residual error term.

For universal kriging (UK), the residual error in Eq. (1) is normally distributed with a zero mean, constant variance σ^2, and correlation structure represented by $\mathbf{R}$, $\varepsilon(\boldsymbol{s}) \sim \mathrm{N}(\mathbf{0}, \sigma^2\mathbf{R})$ (Cressie, 1993). Residuals are dependent by parameterizing a spatial correlation as a decreasing function of distance between locations (Cressie, 1993; Diggle, 1998). The UK model can serve a dual purpose; to identify and quantify covariates accounting for variation in ozone, $Y(\mathbf{s})$, in the presence of residual spatial variation and to spatially predict (optimally map) ozone at unmonitored locations.

A reduced version of the UK model, known as ordinary kriging (OK), is applicable only for spatial prediction and assumes a constant spatial trend with all variability in $Y(\boldsymbol{s})$ modeled with a spatially varying error structure $\varepsilon(\boldsymbol{s}) \sim \mathrm{N}\left(\mathbf{0}, \sigma^2\mathbf{R}\right)$. Ordinary kriging is similar to Eq. (1), except $\mu(\boldsymbol{s}) = \beta_0$. A limitation of ordinary kriging is that while it can be useful for spatial predictions, the lack of covariate information restricts understanding variation.

Spatial correlation parameters for kriging models (range, sill (σ^2), and nugget) are defined using the semivariogram function. Resulting parameter estimation for the spatial trend (β coefficients) is known as generalized least squares (GLS). Both universal and ordinary kriging models can spatially predict ozone Y at an unmonitored location, yielding values that are best, linear, and unbiased predictions (BLUPs) (Cressie, 1993; Schabenberger and Gotway, 2005). An advantage of kriging is that mean square prediction error can be used as a measure of prediction variance, which is unavailable in deterministic approaches like IDW. This allows us to ascertain confidence in our estimations and assess geographic variation in prediction uncertainty.

The multivariate linear regression (e.g. LUR or trend surface model) is represented by Eq. (1) with spatially uncorrelated residual errors represented by, $\varepsilon(\boldsymbol{s}) \sim \mathrm{N}\left(\mathbf{0}, \sigma^2\right)$. This yields maximum likelihood parameter estimates known as ordinary least squares (OLS) and assumes all spatial variability in the outcome $Y(\boldsymbol{s})$ is accounted for by covariates $X_1(\boldsymbol{s}), \ldots, X_p(\boldsymbol{s})$. If this assumption is violated and residuals remain spatially correlated, inference based on linear regression is not statistically optimal and predictions no longer BLUP. Specifically, standard error estimates of regression effects would be underestimated possibly leading to spurious significance of environmental covariates.

The non-statistical inverse distance weighting (IDW) approach for spatial prediction was also considered (Salam et al., 2005; Bell, 2006). IDW predictions are generated as a weighted average of monitor data with weights based on inverse distance to the unsampled location. The formula for IDW prediction is

$$\hat{Y}(\boldsymbol{s}_O)_{IDW} = \frac{\sum_{i=1}^{N} d_{0,i}^{-2} Y(\boldsymbol{s}_i)}{\sum_{i=1}^{N} d_{0,i}^{-2}} \tag{2}$$

where $d_{0,i}$ is the distance from unmonitored location $\boldsymbol{s}_0$ to the ith data $Y(\boldsymbol{s}_i)$ at location $\boldsymbol{s}_i$. We take a distance power of 2, which gives us the commonly used inverse-distance-squared (Schabenberger and Gotway, 2005).

2.3. Variable selection, model fitting, and evaluation of spatial prediction performance

To identify and quantify environmental determinants impacting ozone variation, an OLS regression was built using all the covariates from Table 1. Regression diagnostics included normality tests, Cook's estimate for influential data points, collinearity with variance inflation factors, and stepwise AIC function. Covariates were selected based on these results and coefficient significance ($p < 0.05$). Several smoothers were applied for both temperature and elevation data, although showed minimal impact on the regression model fit. A linear spline knot was added in the distance-to-coast covariate because winds drive ozone concentrations and 80 km represents the maximum distance of inland breeze (Entwistle et al., 1997). Distance-to-coast in the absence of a spline was found to be not statistically significant. Primary roads were examined under multiple categorical cutoffs, including several distance quantiles, however these categories provided no statistical benefit.

A geostatistical semivariogram assessed spatial correlation on the monitored ozone and LUR residuals (Cressie, 1993; Diggle, 1998). Cressie's robust semivariogram estimator minimized the impact of outlying observations (Cressie and Hawkins, 1980), while anisotropic behavior was examined with directional semivariograms. If residual spatial variation was indicated, a semivariogram functional form fitted to the estimated semivariograms using weighted least squares (Cressie, 1985). The semivariogram functions were used to generate correlation structures (the matrix $\mathbf{R}$) in universal kriging models and regression coefficients re-estimated via generalized least squares (GLS) to evaluate significance. Results were compared between the LUR and universal kriging models.

To evaluate prediction performance a Monte Carlo analysis based on statistical cross-validation was applied to each study day.

1. Randomly split the ozone monitor data into subsets of 2/3 (the modeling subset) and 1/3 (the validation subset).
2. Apply the four spatial prediction methods (IDW, LUR, OK, and UK) with the modeling subset to predict ozone concentrations in the validation subset.
3. Compare the predicted ozone for each approach to observed concentrations in the validation set using the evaluation metrics described below.
4. Repeat steps 1–3 500 times.
5. Summarize prediction performance by averaging the evaluation metrics over the 500 iterations.

It should be noted that the kriging variogram functions are defined from the original model and not re-estimated for each simulation subset; however, the large sample size and multiple simulations are designed to normalize out any variability. Checks run with June 1st data showed variogram parameters of range, sill, and nugget had a mean variation of $\pm 4\%$ from the 2/3 modeling subset compared to the national estimate. With a large sample size the 1/3 holdout validation is more feasible compared to familiar approaches such as leave-one-out or n-fold cross validation (Mercer et al., 2011), while also providing robust assessments of prediction performance over a large data subset. While Monte Carlo approaches have been used to evaluate prediction performance with air pollutants (Szpiro et al., 2010), this application across a full AQS dataset to compare multiple prediction methods has not previously been applied.

Three-prediction evaluation metrics were considered; (1) root mean square error (RMSE), (2) standardized RMSE, and (3) cross-validation R^2. RMSE quantifies the mean difference between predicted estimates and true values (Eq. (3)), where y_i denotes the measured value at location i and $\hat{y}_i$ is the predicted value at location i.

$$\mathrm{RMSE} = \sqrt{\frac{1}{n}\sum_{i=1}^{n}\left(y_i - \hat{y}_i\right)^2}. \tag{3}$$

In standardized RMSE, the residuals are scaled by the prediction standard error ($\hat{\sigma}_i$), allowing better comparison across days of data (Eq. (4)). In RMSE lower values imply improved prediction accuracy, while standardized RMSE values of 1.0 indicate better-fitting statistical models.

$$\text{Standardized RMSE} = \sqrt{\frac{1}{n}\sum_{i=1}^{n}\left(\frac{y_i - \hat{y}_i}{\hat{\sigma}_i}\right)^2}. \tag{4}$$

The cross-validation R^2 (CV R^2) statistic indicates how well a predicted value compares to the sample mean alone (Eq. (5)). It is analogous to the coefficient of determination R^2 (common to regression models) with a value of 1.0 indicating perfect prediction fit. This approach is sometimes referred to as a G-measure or goodness of prediction estimate (Cressie, 1993; Erxleben et al., 2002).

$$\mathrm{CV}R^2 = 1 - \frac{\sum_{i=1}^{n}(y_i - \hat{y}_i)^2}{\sum_{i=1}^{n}(y_i - \bar{y})^2}; \quad \text{where } \bar{y} = \frac{1}{n}\sum_{i}^{n} y_i. \tag{5}$$

As a final stage predicted ozone concentrations were spatially mapped across the US on a 10×10 km grid using each method (IDW, LUR, OK, UK). Environmental variables were averaged if prediction grid cells overlapped multiple values. Mapped prediction standard errors for the kriging methods were also produced.

2.4. Software and data analysis

Data integration and analysis were performed in the R Statistical Computing Environment (version 2.14; 2011-12-22). Spatial statistical analysis and mapping were performed with the 'raster' (Hijmans and van Etten, 2012) and 'gstat' (Pebesma, 2004) packages.

3. Results

3.1. Land use regression, spatial dependence, and kriging

A LUR model was built with Table 1 covariates, plus longitude and latitude coordinates of monitors as a preliminary spatial model. Regression diagnostics revealed normally distributed data with no significant outliers (F-statistic < 0.1) or collinearity (VIF). Using stepwise AIC and coefficient significance ($p < 0.05$), covariates were removed until a final model included maximum temperature, elevation, longitude, latitude, population density (for June 1st only), distance-to-coast with a linear spline, and distance to closest percent of primary roads.

Land use regression parameter estimates showed some consistency in coefficient estimates across the data days (Table 2). Maximum temperature revealed a positive trend with concentrations of ozone increasing by 1.98 to 2.99 ppb for every 1 °C change, while an increase in elevation (100 m) caused higher ozone concentrations by 1.51 to 1.57 ppb. For July 3rd and August 1st, longitude and latitude demonstrated a positive relationship with ozone as one moved northeast. Relationships were reduced using June 1st data, which may be driven by environmental factors not captured in our daily assessment. Population density was found to be significant ($p < 0.05$) and negatively associated with ozone on June 1st. Ozone concentration was observed to decrease moving inland, but at 80 km this magnitude significantly lessens with the trend becoming positive using June 1 data. It was found that ozone concentrations decrease by 0.31 to 0.46 ppb for each 10 km of distance you get from the closest primary roads. LUR models yielded coefficient of determination R^2 estimates from 0.45 to 0.51 across the data days.

Table 2
Parameter estimates from land use regression (inference based on OLS) and universal kriging models (inference based on GLS) for monitored ozone concentrations during our three study days. Population density was not included on July 3 and August 1 after failing to meet model selection criteria.

Predictor covariates	June 1, 2006		July 3, 2006		August 1, 2006	
	LUR	UK	LUR	UK	LUR	**UK**
Intercept	−60.30*	5.84	−61.51*	−20.43*	−52.29*	3.52
Maximum temperature (° C)	2.99*	1.47*	2.76*	1.81*	1.98*	1.03*
Elevation (per 100 m)	1.57*	1.03*	1.55*	1.08*	1.51*	0.78*
Population density (1000 per km^2)	−0.03*	−0.31*	–	–	–	–
Latitude	0.83*	0.02	1.54*	1.09*	1.79*	1.22*
Longitude	−0.82*	0.01	2.70*	0.26*	0.24*	0.33*
Dist. to Coast (<80 km)	−12.18*	3.19	−6.49*	0.43	−8.64*	−3.62
Dist. to Coast (>80 km)[a]	0.05*	−2.53	−0.76*	−0.41	−1.03*	−0.41
Dist. to road (per 10 km)[b]	−0.31*	−0.16*	−0.46*	−0.40*	−0.38*	−0.37*

* Denotes coefficient significance ($p < 0.05$).
[a] Values indicate changes from baseline.
[b] Road distance is mean of the closest percent of road segments.

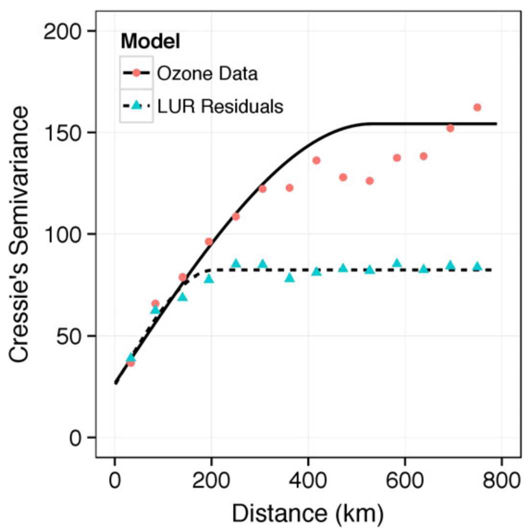

Fig. 1. Estimated semivariograms and fitted spherical variogram functions for monitored ozone and residuals from the fitted land use regression model for July 3.

Fitted semivariograms for monitored ozone and the LUR residuals are reported in Fig. 1 for July 3rd, while June 1st and August 1st can be found in the Supporting Information (Figure S1). Patterns in all semivariograms suggest the existence of residual spatial correlation. Compared to the ozone semivariogram, the semivariogram of LUR residuals has a lower sill value (y-axis value where the semivariogram levels off), which is expected since some variance is accounted by the covariates. More importantly the residual semivariogram suggests residual spatial variation not accounted for by included covariates. Directional semivariogram estimates did not reveal evidence for anisotropic spatial relationships, so the assumption of isotropy was retained.

Using the derived residual correlation structure $\mathbf{R}$ (based on the fitted semivariogram), regression coefficients were re-estimated under the universal kriging model with generalized least squares inference. While the statistical significance and trends of temperature, elevation, population density, and distance to roads remained the same, several other covariates lost significance in the UK model (Table 2). This is not surprising as overlooking residual spatial variation can lead to spurious effects and generally coefficient standard errors increase from the LUR model to the UK model (Gaffney et al., 2005; Jerrett et al., 2005). For June 1st latitude and longitude became non-significant, while distance-to-coast dropped out models for all days. Direction of influence for the significant covariates remained the same between the LUR and UK models with maximum temperature, elevation, latitude and longitude all positively related to ozone concentrations, while ozone concentrations decrease as you move farther away from primary roads. As expected GLS coefficient standard errors were larger for the universal kriging model compared to LUR model (Supporting Information Tables S1 and S2).

3.2. *Optimal prediction of ozone concentrations*

Each of the spatial prediction approaches, IDW, LUR, ordinary kriging, and universal kriging, were analyzed with the described Monte Carlo cross-validation scheme. Results demonstrated that LUR models predicted least favorably (R^2: 0.45–0.51; RMSE: 10.16–12.42), while ordinary kriging (R^2: 0.78–0.82; RMSE: 6.80–7.72) and universal kriging (R^2: 0.76–0.82; RMSE: 6.89–7.79) predicted with similar accuracy (Table 3). Predictions based on inverse distance weighting (R^2: 0.73–0.75; RMSE: 7.59–8.68) outperformed the LUR model, but did not reach the prediction accuracy of kriging. When comparing between the kriging and LUR models, we found that RMSE for universal (and ordinary) kriging were between 35% and 45% smaller than LUR. These trends held consistent across all performance metrics. Additional diagnostics of the cross-validation, including prediction precision, residual plots, and results from a 10-fold cross-validation can be found in Supporting Information (Figures S2a–c; Tables S3–S4).

Table 3
Results from the Monte Carlo cross validation analysis for prediction with land use regression (LUR), inverse distance weighting (IDW), ordinary kriging (OK), and universal kriging (UK) for the three study days. Performance metrics include mean, minimum, and maximum of predicted ozone (ppb), cross validation R^2, root mean squared error (RMSE) and standardized root mean squared error (std. RMSE).

Date	Summary statistics	Observed AQS values	LUR	IDW	OK	UK
June 1	Mean	51.55	51.49	51.71	51.50	51.54
	(min, max)	(2, 103)	(12.52, 80.12)	(9.75, 88.98)	(10.49, 91.85)	(10.48, 91.05)
	R^2	–	0.51	0.75	0.82	0.82
	RMSE	–	11.47	8.22	6.93	6.89
	Std. RMSE	–	0.56	NA	1.01	0.96
July 3	Mean	53.23	53.21	53.74	53.33	53.35
	(min, max)	(13, 97)	(16.31, 76.83)	(18.48, 78.89)	(20.43, 80.48)	(19.91, 83.37)
	R^2	–	0.51	0.73	0.78	0.76
	RMSE	–	10.16	7.59	6.80	7.03
	Std. RMSE	–	0.57	NA	0.98	1.00
August 1	Mean	49.60	49.54	50.11	49.71	49.63
	(min, max)	(10, 112)	(16.04, 73.34)	(11.90, 83.28)	(12.81, 96.32)	(13.92, 94.16)
	R^2	–	0.45	0.73	0.79	0.78
	RMSE	–	12.42	8.68	7.72	7.79
	Std. RMSE	–	0.59	NA	1.22	1.13

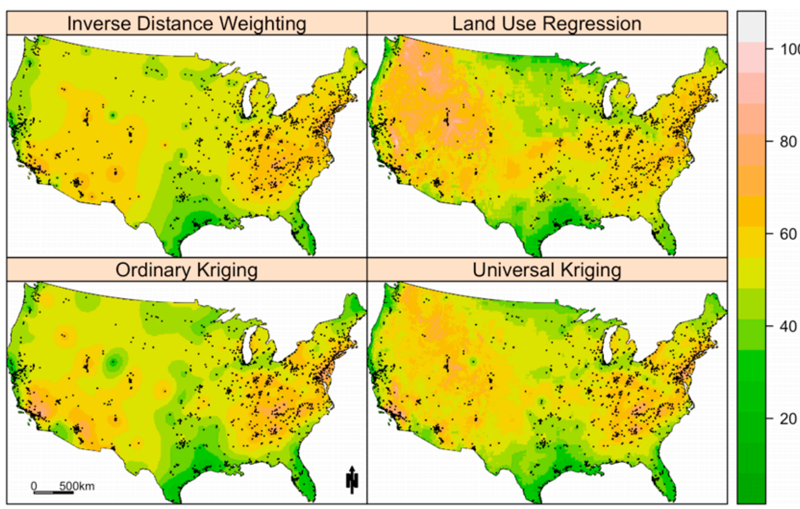

Fig. 2. Predicted ozone air pollution concentrations (ppb) for July 3, 2006 using inverse distance weighting, land use regression, ordinary kriging, and universal kriging.

Predictions of ozone concentration were mapped for each of the different approaches. Broad trends in predicted ozone were similar across days, so July 3rd was chosen as a 'representative' day (Fig. 2) (Supporting Information Figures S2–S5 for other days). Each of the prediction methods revealed elevated ozone concentrations along the northeast corridor, Appalachia, and parts of southern California, while lower concentrations were estimated for Florida and Texas. Mean national predictions of ozone concentration were similar among the approaches, ranging from 51.1 ppb with ordinary kriging to 52.8 ppb with LUR. Although nationwide prediction maps appear visually similar across methods, the results in actual prediction performance should not be overlooked (Table 3).

Mapped prediction standard errors provide another alternative for assessing and comparing model performance (Fig. 3). Ordinary kriging showed higher prediction standard errors than universal kriging (mean: 8.6 ppb, range: 5.2–11.2), most notably in regions of sparse air monitors (western Great Plains; Rocky Mountains). Prediction standard errors were reduced in the universal kriging model (mean: 8.1 ppb, range: 5.5–9.5), indicating less uncertainty in predicted concentrations. Since IDW is a non-statistical approach, there is no error measurement associated with its estimates. The LUR approach yielded artificially low prediction standard errors with little variation within a narrow range (mean: 10.2 ppb, range: 10.1–10.4), which is driven by the assumed independent regression errors (constant σ^2), therefore the results are not presented.

4. Discussion

In this study we provide a systematic analysis of four approaches for characterizing national-scale spatial distributions of US ozone concentrations. Comparisons were made between kriging-based methods, IDW, and the LUR approach. We identified maximum temperature, elevation, distance to primary roads, latitude, and longitude as significant environmental covariates for explaining variability (spatial and non-spatial) in ozone concentrations. This research fills an important data gap by comparing the method performance of different approaches that produce national-level ozone concentration maps. It also explores the applicability on a spatially homogeneous regional pollutant (e.g., ozone), as opposed to pollutants attributed to localized emissions sources (e.g., CO or NO_x). Rather than declare a single method universally optimal, we demonstrate the benefit of checking and accounting for residual spatial correlation, a message that extends beyond this current application.

The kriging-based methods (ordinary and universal) outperformed land use regression when evaluating prediction performance. Monte Carlo cross validation-based performance metrics improved 30% to 50% with universal kriging over land use regression. On June

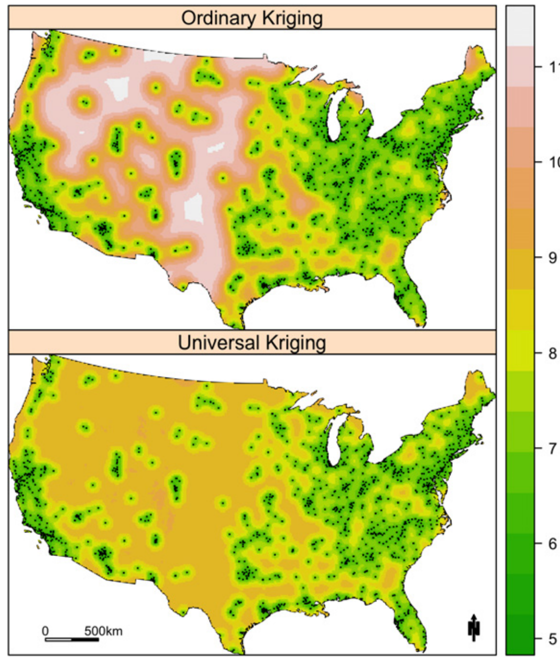

Fig. 3. Estimated standard error of predicted mean ozone concentrations (ppb) for July 3, 2006 using ordinary and universal kriging approaches.

1, the universal kriging predictions were on average 5 ppb more accurate than land use regression-based predictions (RMSE of 7.24 for UK compared to RMSE of 11.35 for LUR, a 36% reduction). Such magnitudes of change in predicted exposure can have a substantial impact on health impact assessments, amending estimates of mortalities by hundreds individuals (Berman et al., 2012). While prediction performance between ordinary and universal kriging were similar (0.04 to 0.23 ppb difference in RMSE), it is important to recall that ordinary kriging (and IDW) does not account for the effects of environmental covariates. If the primary goal is prediction of ozone concentrations, ordinary kriging is computationally simpler and provides comparable estimates to universal kriging for these three selected days. However, ordinary kriging fails to add much for understanding variations in ozone concentration, a critical component when considering policy implications. If interest lies in both modeling and variation, universal kriging not only produces optimal predictions, but also provides inference on ozone covariates.

Several environmental and geographic covariates were found to influence ozone concentration variability. The positive relationships between elevation, temperature, and ozone concentrations were consistent with previous research (Son et al., 2010; Walcek and Yuan, 1995). Higher altitude is associated with improved air clarity and solar irradiance, enhancing photochemical formation of ozone from free-tropospheric sources (Beelen et al., 2009; Coyle et al., 2002; Fiore et al., 2003). Differing from European studies, we found land use and population density (for July/August) to be statistically non-significant, but there was a significant association between ozone and increasing proximity to major roads (Beelen et al., 2009). It is speculated by Briggs (2005) that the open nature of US urban areas may weaken the association between land cover and air pollution patterns. The grid-like character of US secondary roadways might also drive homogeneous concentrations, whereas developing nations are heavily influenced by local or uncontrolled emissions sources (Briggs, 2005). This may explain both the lack of significance with land use and the improved prediction performance of ordinary kriging. However without including secondary roadways this is only a hypothesis. One potential solution is including additional nationwide datasets, including air pollution models that have both large geographic coverage and daily models.

A major emphasis was to elucidate the importance of checking and accounting for residual spatial dependence. While environmental covariates explain some variation in ozone concentration, semivariograms from LUR models revealed residual spatial dependence. Overlooking spatial dependence can lead to both less efficient inference (spurious effects) and less accurate predictions, causing artificially shrunken coefficient confidence intervals. While studies in Europe and Asia found distance-to-coast to be an important environmental factor (Fiore et al., 2003), we found this covariate to be non-significant when based on generalized least squares in the universal kriging model. Pure distance variables, such as latitude and longitude on June 1st can also become unnecessary when considering spatial dependence. These trends would have been overlooked if not for a careful examination of spatial variation. The universal kriging model also revealed important information about environmental factors influencing spatial variability. Many covariates remained significant, highlighting their influence for explaining both a spatial and non-spatial component of variability in ozone concentration.

Our prediction of ozone at an extended national scale corresponded with European and city-level modeling of air pollutants (NO_x, PM_{10}, and O_3), finding universal kriging to outperform land use regression, ordinary kriging, and IDW (Beelen et al., 2009; Mercer et al., 2011; Son et al., 2010). This is in contrast to other studies that did not find improved spatial performance of kriging over land use regression (Clougherty et al., 2008; Poplawski et al., 2008); however, this may be explained by the city-level study areas addressed in those papers. In particular, Ross et al. (2007) felt that ordinary kriging would not sufficiently capture variation over small regions, particularly if pollutants were reliant on local emissions such as traffic. This argument supports the benefits of adding covariate information into prediction models. However if covariates are available for an LUR model, a universal kriging approach can also be considered. We demonstrate the importance of checking for residual spatial dependence, regardless of the study area size or any preconceived notion that one modeling approach is preferred to another.

A limitation of this study, along with most spatial approaches, is effectively capturing within person variation. It is well established that individuals spend substantial portions of their time in indoor environments, including commuting and working (Briggs, 2005). The use of fixed air monitor values as surrogates of individual exposure can fail to reflect both spatial and temporal exposure that in turn impact the estimation of relative-risk estimates (Levy et al., 2005; Monn, 2001). Additionally due to the format of national-level data, a 10×10 km prediction grid effectively aggregates small distance spatial variability resulting in a loss of some fine-scale data. A reason for selecting ozone as opposed to a primary pollutant is a more spatially homogeneous distribution (Marshall et al., 2008; Sarnat et al., 2010; Özkaynak et al., 2013), less reliant on local emissions sources and more robust to a coarse prediction grid. If the goal were estimating national concentrations for spatially heterogeneous pollutants (e.g. NO_x or CO) or perform a time-series assessment for exposure purposes, selected covariates and spatial resolution would have to be reconsidered and potentially change the best performing model.

We additionally recognize that the optimal performance of kriging could be a potential artifact of the national-level model and had more environmental data been incorporated, the LUR approach may substantially improve. When performing national-level assessments, it is computationally tedious to incorporate local fixed covariates, such as fine roadways, which are important for capturing micro-spatial variability and a major component of LUR studies with traffic-related pollutants. It may further be argued that LUR is dependent on extensive covariates, so a national approach might not be appropriate. However, the bigger criticism this research addresses is the notion that modeling approaches (LUR or kriging) must be decided upon *a priori*. As noted by Cressie (1993) in reference to trend surface modeling, "Rather than entering into a fruitless debate over which approach is better, it is more appropriate to address the question through model selection, which makes the answer problem-specific". Hence considering multiple methods and allowing the data to guide the specific model selection provides a more scientifically defensible approach.

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Appendix A. Supplementary data

Supplementary material related to this article can be found online at http://dx.doi.org/10.1016/j.eti.2014.10.003.

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