

Chapter 8

Use of nonparametric local regression to estimate surface ozone patterns over space and time

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Abstract

In this chapter we demonstrate the use of modern regression techniques to estimate ozone (O_3) exposure maps and study the effects of auxiliary weather and topographic variables on O_3 levels. The regression technique provided a flexible framework for estimating temporally explicit O_3 exposure maps and uncertainty levels. The data used were from 79 passive monitor sites distributed across the Sierra Nevada, California, and recorded bi-weekly during the 1999 summer season. The modeling framework was also useful for performing formal comparison of O_3 values at sites with similar environmental and topographic conditions. Results of the analyses indicated the presence of a significant, mostly west to east, spatial trend in addition to an increasing elevation trend. The results are for elevations less than 2400 m (the elevation of the highest monitoring site).

1. Introduction

The particular statistical model needed for the analysis of a data set depends on the type of data collected and the questions that are of interest to researchers. In the case of ozone (O_3) air pollution studies, the data are spatial as well as temporal (i.e., O_3 observations collected weekly or bi-weekly from samplers at various locations and over a period of months). Additionally, there may be various auxiliary (explanatory) variables that are also spatial-temporal. Auxiliary variables might include maximum temperature, elevation, precipitation, or any other explanatory variable that the researchers suspect will be good indicators

of the levels of pollution at a given location and a given time. In particular, we will focus on a study conducted in 1999 in the Sierra Nevada of California that consisted of a network of passive O₃ monitors placed at various distances and elevations around active O₃ monitoring stations.

Some of the questions that we would like to address with the data from the passive samplers include: (1) What are the expected spatial and temporal patterns of ozone pollution in the study region? (2) What are the relationships between ozone levels and explanatory variables, such as weather and topography? (3) Can we predict ozone levels at unobserved (un-sampled) sites within the study region? (4) What is the accuracy of the predicted values and how can uncertainties be presented on maps? (5) Can the estimated statistical model be used to predict ozone values at a future date?

One common feature of data collected from a network of samplers is the correlation between observations from different but nearby sites (spatial) and observations from the same site collected over time (temporal). Observations from nearby locations, or the same location over time, tend to be similar because of similarities in environmental and topographic conditions. A variety of statistical procedures are available for using the correlations between variables for predicting ozone levels at unobserved sites. One approach is to use *geostatistical techniques* where data are assumed to be realizations of dependent random variables with a covariance structure modeled as a function of spatial location. Kriging or cokriging is then used to predict values at new locations. No temporal component is included in models using the geostatistical approach, and data from different time points are studied separately. The second approach is based on *modern regression techniques*, such as generalized additive models. In the second approach, locally weighted regression models are used to estimate nonparametric functions of location, of time, and of the auxiliary variables simultaneously. Under the local regression model, a smooth function of spatial location is included in the mean to account for any persistent features of the landscape or the environment not captured by any of the environmental or topographic variables in the model. Temporal aspects of the data are modeled using time-series regression techniques or regression techniques with random effects. Finally, if autocorrelations are still detected in the residuals after fitting the generalized additive model, kriging techniques may be used on the residuals to obtain better predictions at unobserved sites.

In this chapter we use the modern regression framework of generalized additive models to predict ozone levels in the Sierra Nevada given observations from a network of passive samplers, several active samplers, meteorological data from a network of weather stations, and elevation. The model development and validation is presented in the methods section and Figs. 2–8. Predicted ozone maps and a discussion of the effects of auxiliary variables on ozone are presented in the results and Figs. 9–12.

2. Methods

2.1. Study area

In 1999 a regional survey of seasonal ambient ozone exposure patterns in the Sierra Nevada was conducted. A network of passive samplers was located along elevation gradients adjacent to active monitoring stations currently operated by the California Air Resources Board and the National Park Service. Around each active monitoring station, a network of passive ozone monitors was established, resulting in a total of 89 passive monitors (79 used in this analysis) located throughout the Sierra Nevada, of which 9 were co-located with active ozone monitors.

Sites for passive monitors were selected at three general elevations along the north to south gradient of air pollution on the western side of the Sierra Nevada. Mid-elevation monitoring sites were located at or near stands of ponderosa or Jeffrey pines that were subsequently sampled using Forest Pest Management survey protocol (1500–1750 m) (Pronos and Vogler, 1981). Monitoring sites were also located at low elevation locations at 1000–1400 m elevation, and at high elevation sites located along the mixed conifer-subalpine ecotone at 2000–2400 m elevation (Fig. 1). All sites were located at least 200 m from frequently used roads, in open areas that had good vertical mixing of air. Nine

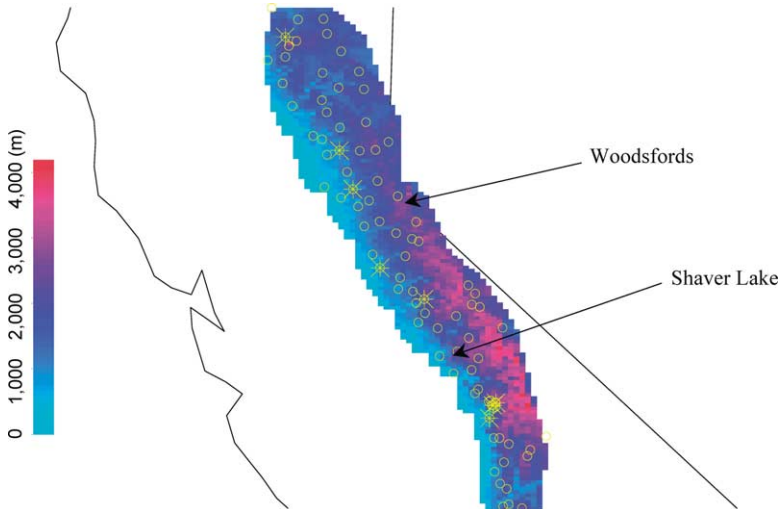


Figure 1. Elevation map with locations of passive (circles) and co-located active (stars) monitoring stations in the Sierra Nevada, California.

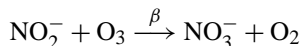
passive monitor sites were co-located with active monitors that were operated during the 1999 summer season.

A single passive ozone sampler, containing two cellulose filters saturated with nitrite was installed at each site (Koutrakis et al., 1993). Ozone oxidizes nitrite into nitrate ions. The amount of nitrate in the filter at a given time is a measure of the amount of ozone at the site. The samplers were located at about 1.5–2.5 m above ground level in forest clearings (about 20 m or more from the nearest trees). At 8 to 10 monitoring sites in each 2-week collection period, 2 blank filters were also tested. Blank filters were kept at room temperature in tightly closed plastic vials. In the field, the filters were changed every two weeks during the summer growing season. After the exposures, the filters were placed in plastic vials, and refrigerated until analyzed. Ozone concentrations were continuously monitored by ultraviolet (UV) absorption (Thermo Environmental Model 49, Cambridge, MA, or an equivalent instrument), at nine monitoring stations for comparison with co-located passive samplers.

Daily maximum temperature and precipitation were obtained for 55 weather stations distributed over the Sierra Nevada range. Weather data, elevations, and locations were obtained from the NOAA National Climate Data Center (<http://www.ncdc.noaa.gov>) in Asheville, NC and the National Interagency Fire Management Integrated Database (<http://famweb.nwcg.gov>) at the USDA National Information Technology Center in Kansas City, MO.

2.2. A spatial temporal model for estimating ozone maps

Passive samplers do not directly measure ozone but contain filters with a known amount of nitrite (NO_2^-). Ozone reacts with the nitrite converting it to nitrate. The data that results is the amount of nitrate ions (NO_3^-) produced in the filter in a given period of time (usually 1 or 2 weeks). The chemical reaction in the filter is given by the equation



where β is the rate of the reaction. Consequently, we generated estimated ozone maps by first estimating a spatial temporal predictive model for nitrate ion levels then converting the predicted nitrate values to ozone levels by using a conversion factor. Methods for estimating conversion factors for different sites are discussed below. The model we used for estimating nitrate levels was

$$Y_{ijk} = \mu + g_1(\text{lon}_i, \text{lat}_i) + g_2(t_{ij}) + g_3(\text{mtemp}_{ij}) + g_4(\text{precip}_{ij}) \\ + g_5(\text{elev}_{ij}) + g_6(\text{active}_{ij}) + \tau_i + \varepsilon_{ijk} \quad (1)$$

where Y_{ijk} is the amount of nitrate ions in the k th sample (replicate), at the i th site, and t_{ij} th day; $\text{lon}_i, \text{lat}_i$ is longitude and latitude of the i th site (the loca-

tion of the i th passive sampler); $mtemp_{ij}$, $precip_{ij}$, $elev_{ij}$, $active_{ij}$ are values of the auxiliary variables, maximum temperature, precipitation, elevation, and recorded ozone level at the nearest active monitor; μ is mean nitrate level over all sites and dates; τ_i is unobserved random site effect assumed to be Gaussian with mean zero and variance σ_τ^2 ; ε_{ijk} is unobserved independent random noise with mean zero and variance σ_ε^2 ; $g(\cdot)$ are non-parametric smooth functions to be estimated from the data simultaneously (Cleveland et al., 1992). Most statistical literature on air quality look at square root transforms of ozone. However, histograms and normal probability plots of the nitrate data (Fig. 2) did not seem to warrant the square root transformation. The untransformed data appeared to be better approximated by the Gaussian distribution than the square root transform.

The weather variables, maximum temperature and precipitation, used in the model were estimated from observations recorded at 55 weather stations distributed over the Sierra Nevada range. The model used to estimate weather data at the passive sampler sites was similar to the model (1) with maximum temperature (or precipitation) as the dependent variable and latitude, longitude, time, and elevation of weather station as explanatory variables. The precipitation variable was an estimate of the probability of rain occurring at a given location. We included the smooth surface, $g_1(lon_i, lat_i)$, in the regression line to account for general spatial patterns not explained by any of the four covariates (e.g., patterns due to wind). A random site effect was included in the model to account for site-specific characteristics due to unknown or unobserved site covariates. The between-record error terms, ε , were assumed to be independent. Estimates of the smooth functions and smooth surface in model (1) were evaluated simultaneously by using the generalized additive model procedure (Hastie, 1992) in Splus (S-PLUS, 2000). Estimates of the variances for the between and within site error terms were obtained using an iterative procedure based on the expectation-maximization (EM) algorithm (Dempster et al., 1977; Brillinger and Preisler, 1985). The EM algorithm involves the successive maximization of the expectation of the "complete data" likelihood, which is conditional on the observed data. In our case, the "complete data" is the observed data and the current estimates of the random effects terms.

The jackknife procedure was used to calculate standard errors of the estimated smooth functions (Efron and Tibshirani, 1993; Preisler et al., 2002) and to assess the significance of the auxiliary variables in model (1). Relationships between the significant auxiliary variables and ozone were studied by producing partial residual plots (McCullagh and Nelder, 1989). Partial residuals plots are developed by subtracting the estimated effects of all but one of the covariates from the observed nitrate values (converted to O_3 concentration units, ppb). These plots describe the effects of each auxiliary variable on ozone after controlling for the effects of all other variables in the model.

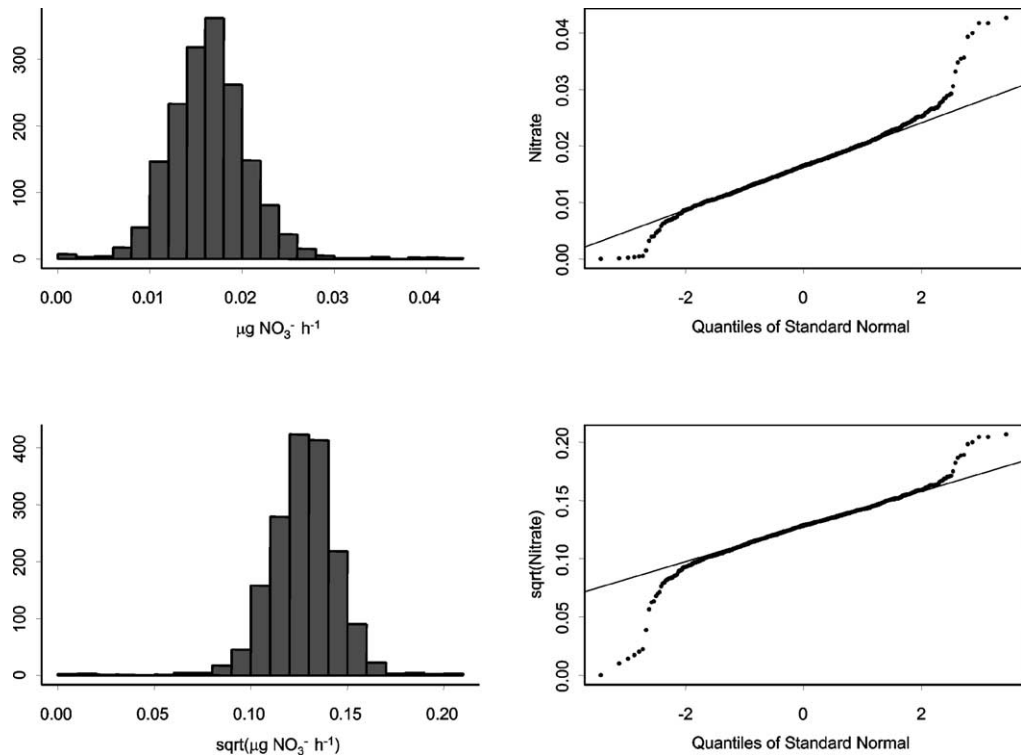


Figure 2. Histogram and normal probability plots of the nitrate data and the square root transformed data. The usual square root transformation used in ozone studies does not appear to be appropriate for the nitrate data.

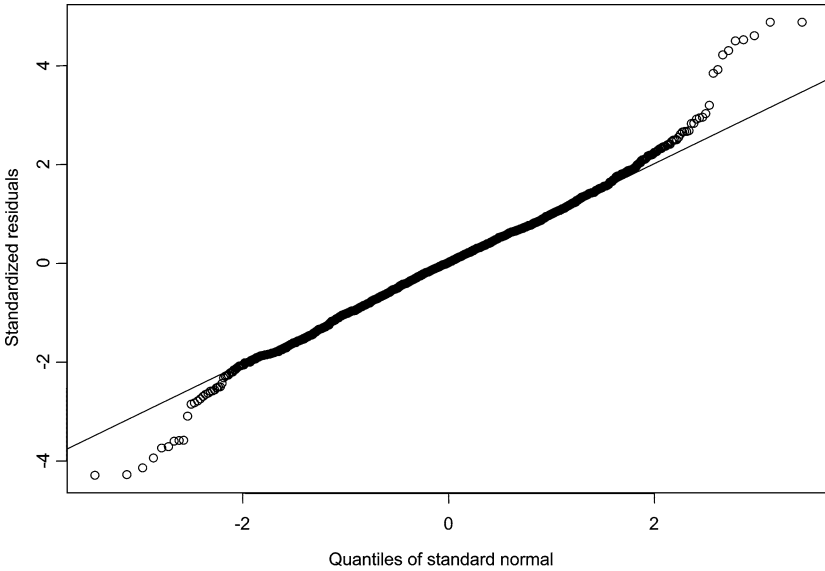


Figure 3. Normal probability plot of model residuals.

2.3. Assessing goodness of fit of the predictive model

We produced a normal probability plot of the residuals in order to assess the Gaussian assumption of model (1). There were 24 values in the normal probability plot that appeared to be smaller or larger than expected under the assumed model (Fig. 3). These values may indicate either the need for more accurate or additional explanatory (e.g., weather) variables. Although this might not justify the additional expense of locating meteorological stations with each passive monitor, nevertheless it indicates the need for further studies at a finer scale, possibly using data from a few sites where weather and passive sampler monitors are co-located.

Estimated directional variograms of residuals plotted against distance were useful for assessing the assumption of spatial independence of the error terms (S+Spatial Stats, 1998). Plots of estimated directional variograms of model residuals indicated that the assumption of spatial independence of the error terms was adequate (Fig. 4). The variograms in all directions were basically flat, indicating no significant autocorrelations. We used cross-validation to produce plots of observed versus expected values, with expected values at a given site calculated using data from all other sites. Approximately 94% of the observed ozone values were within the estimated point-wise 95% confidence bounds (Fig. 5) produced by the cross validation study. Some of the points

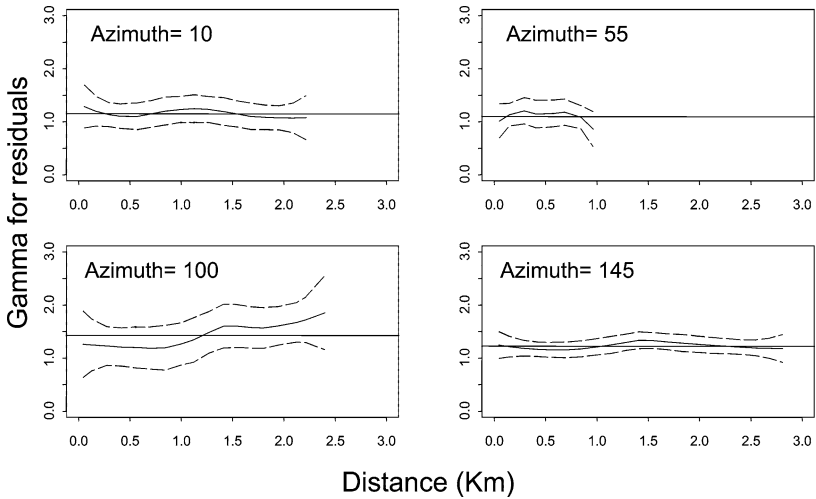


Figure 4. Estimated directional variograms (and 95% confidence limits) of the residuals. The variograms are mostly flat indicating that the fitted model has accounted for most of the spatial autocorrelations in the data. The variograms in the four panels correspond to spatial autocorrelation patterns in the four azimuth directions (10, 55, 100, 145) from passive sampler sites.

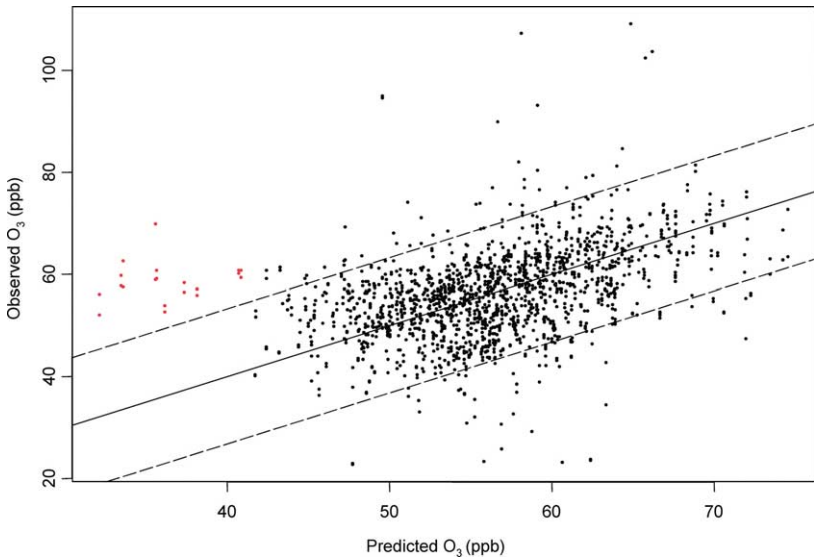


Figure 5. Observed versus predicted values from a cross-validation study where values at a site are predicted using data from the remaining sites. According to the model, 95% of ozone levels are expected to fall within the dashed lines. Predicted values for one of the sites in Eldorado National Forest (red dots) appear to be biased.

outside the 95% bands were the extreme values already discussed above. However, a new group of outliers (all from the Woodsfords site in Eldorado Forest, Fig. 1) were detected. All the observed values at this site were greater than two standard deviations from the expected values. This type of bias was also evident in our estimates of the random effect and the residuals error variances. The estimated between-site variation, $\hat{\sigma}_\tau$, was 3.8 ppb, while the record-to-record variation, $\hat{\sigma}_\varepsilon$, was 5.4 ppb. In other words, approximately 33% of the total variation was due to the random between-site variations.

2.4. Estimating relationship between ambient ozone and nitrate formation rates

The amount of nitrate, NO_3^- , in a filter at a given time is

$$\text{NO}_3^- = \alpha + \beta \text{O}_3 + \varepsilon \quad (2)$$

where ε is random noise, and α and β are unknown parameters to be estimated from co-located passive and active samplers. Given estimates for α and β , the formula $\text{O}_3 = (\text{NO}_3^- - \hat{\alpha})/\hat{\beta}$ may be used to convert observed NO_3^- values to O_3 levels at a given site.

Data from the nine co-located monitors and the estimated regression lines are provided in Fig. 6. The slopes and intercepts of the regression lines were significantly different for the various sites (Fig. 6). The relationship between nitrate values and ozone values from the continuous monitor at the Shaver Lake site (Fig. 1) was extreme relative to relationships at other continuous monitor sites. Examination of the data indicated that although nitrate formation rates at this site were comparable with sites north and south of Shaver Lake, the continuous monitor values were much lower than those at other nearby continuous monitors. The Shaver Lake continuous monitoring station was accordingly excluded from further analysis.

Regression techniques were also used to study the effects of three variables (elevation, maximum temperature, and precipitation) on the values of the conversion factors, α and β . There were some indications of significant effects of the covariates on the slopes and intercepts (Fig. 7). Although this seems to indicate that nitrate to ozone conversion factors should be based on some local site conditions, additional data are needed before this method can be used. Also, using a separate slope and intercept for each site did not produce any discernable effect on the final maps describing estimated spatial ozone patterns. Consequently, a common slope and intercept was used in this study to convert nitrate levels to ozone levels at all sites.

2.5. Predicting ozone levels at new locations

One of the aims of the passive sampler study was to produce maps for the Sierra Nevada region of predicted ozone levels for various dates of the sum-

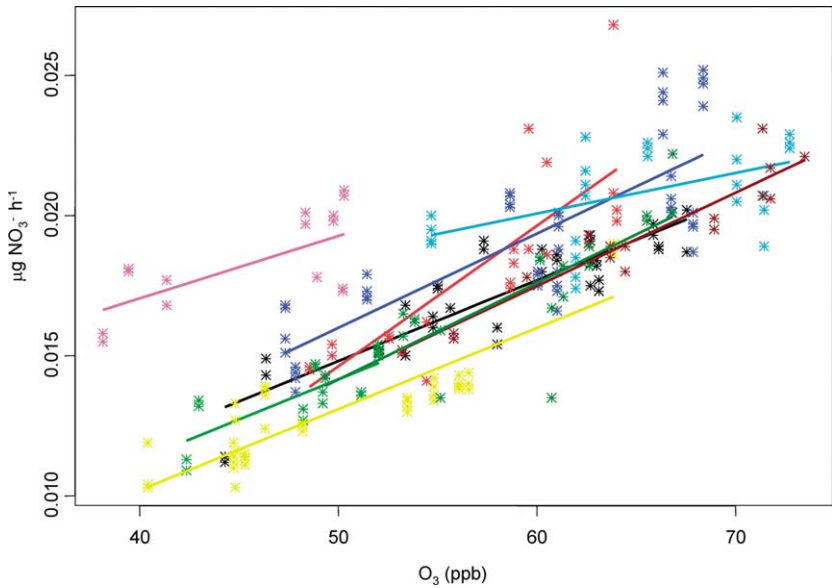


Figure 6. Relationships between ozone levels observed at active monitors and nitrate formation rates observed by passive samplers at the co-located sites. The active monitor data from one of the sites (pink) appears to be an outlier.

mer season. In order to do that we needed to estimate ozone levels at all points on a grid covering the region of interest using observations collected at only a fraction of the grid points (Fig. 8). Because the generalized additive model described above is an extension of multiple regression techniques, predictions at a new site may be produced by evaluating model (1) at the values of the auxiliary variables for that site: the site-specific variables (latitude, longitude, elevation); the weather variables (maximum temperature and precipitation at the site for a given date); and the ozone level at the nearest active monitor for the given date. Similarly, evaluating the smooth function of time at the required date produces predictions at any date within the summer season (Fig. 8).

2.6. Predicting ozone levels at a future date

Predicted ozone values for future dates may be produced given values of the weather variables and ozone levels at actively monitored sites. Because those values are unknown for future dates, they need to be replaced by predicted values. One possibility is to use predictions from weather models (Fujioka, 1990). This might not be a practical solution because weather predictions for longer

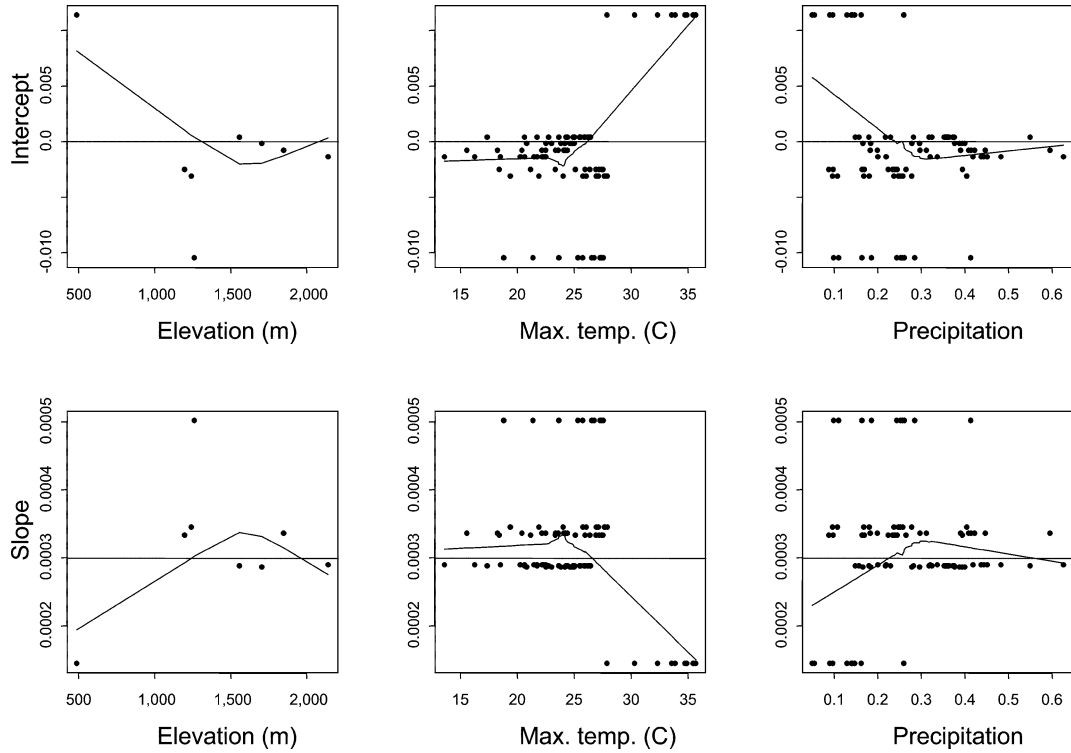


Figure 7. Relationships between auxiliary variables and the slopes and intercepts of the O_3 to nitrate conversion relationships. The horizontal lines indicate the average levels.

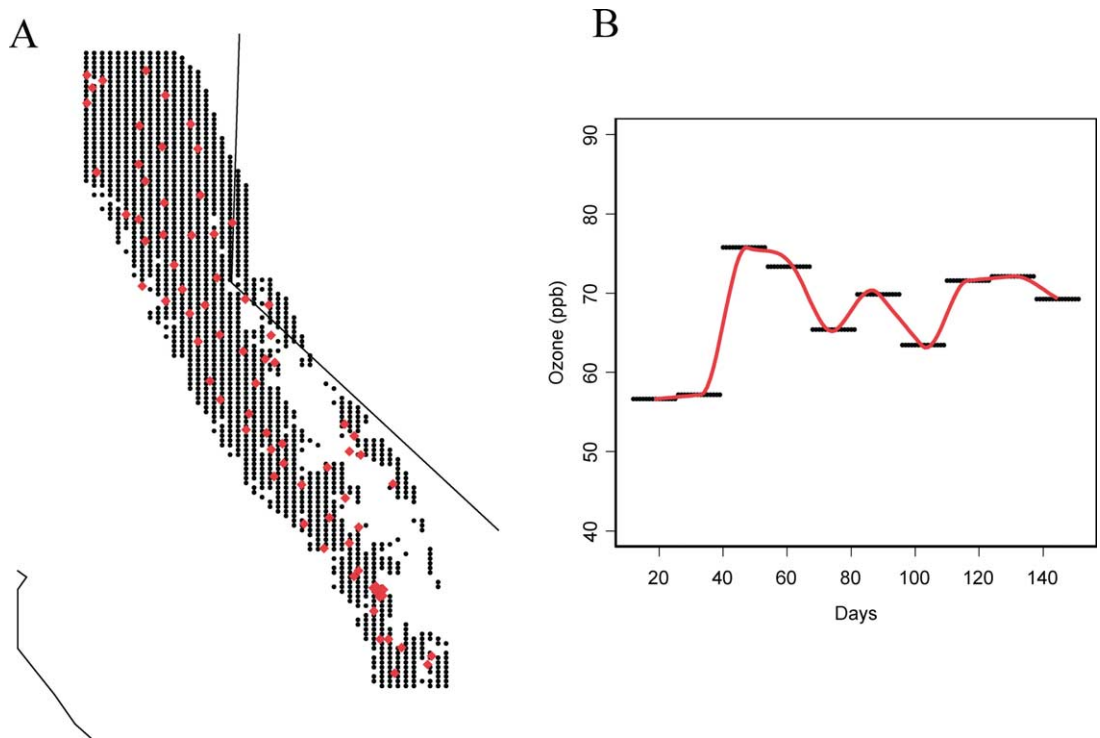


Figure 8. (A) Locations of the passive monitor sites (red stars) and the grid points used to produce ozone map of predicted values. (B) Observed bi-weekly nitrate amounts converted to ozone values (horizontal bars) and estimated smooth function of time (days) for one site.

than 2 weeks are not very reliable. Another possibility is to use historic weather conditions for a given site. By using historic records, one may estimate ozone levels for a range of weather conditions (e.g., extreme weather conditions for that site). Similarly, historic values of ozone levels at active monitor sites may be used for nearest active monitor variable. More work is needed in order to develop reliable predictions of future ozone levels. In particular, the relationships between ozone and weather conditions need to be studied at a finer scale (e.g., using data from a few sites where weather and passive sampler monitors are co-located). Data from more than 1 year need to be analyzed to estimate the seasonal effects not accounted for by the weather variables in the model. For example, the data from 1999 seem to indicate that there are significant temporal effects over the summer season that were not accounted for by temperature or precipitation (Fig. 9). Are these effects due to other unobserved weather conditions (e.g., wind)? If so, is it reasonable to expect the same weather patterns in future years? These and other questions need to be studied before we are able to develop reliable maps for future ozone levels.

2.7. Mapping uncertainties

Although it is possible to generate maps of predicted ozone levels, there are no indications of the uncertainties attached to these predictions. Without a measure of the uncertainties, it is not possible to discern whether any of the differences seen in the maps are significant or whether the differences are simply due to sampling fluctuations. In this work we suggest using maps that highlight regions where the ozone level is significantly above or below average by using standard deviations estimated from the model. In such displays, ozone levels are compared with an overall average for the season and for all locations. Similar maps may also be produced that highlight regions that are significantly higher than some critical ozone level seen to be deleterious to a particular plant or animal species.

3. Results

3.1. Relationships between ozone level and auxiliary variables

All auxiliary variables in model (1) were found to be statistically significant. The effects of four variables with the largest effects on ozone are displayed in the plots of partial residuals (Fig. 9). These plots describe the effects of each auxiliary variable after controlling for the effects of all other variables in the model. The horizontal line at zero indicates the average ozone level. The scatter of points around the smooth curves are due to random fluctuations between sites or dates that were not accounted for by the variables in the model. As

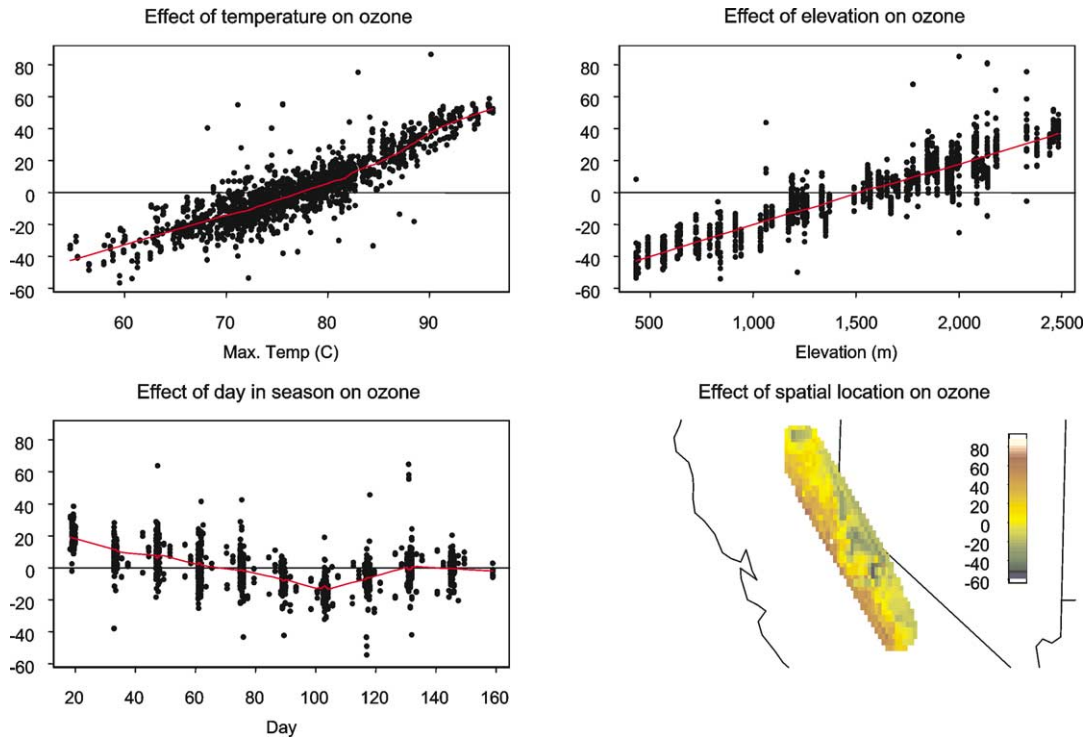


Figure 9. Partial residual plots describing the relationships between ozone and maximum temperature, elevation, day in season, and spatial location. Plots for each variable were produced by subtracting estimated effects of all other variables from observed nitrate levels (converted to ozone units).

expected, ozone levels appear to be increasing with increasing levels of temperature. Ozone levels also appear to be increasing with elevation. The effect of elevation can't be attributed to temperature or location (variables already in the model). This points to some, yet unidentified, variables affecting ozone levels in the Sierra Nevada and that are linearly correlated with elevation. The temporal (day) effect was also statistically significant. The 1999 season appeared to have had a decreasing trend in ozone levels, with the lowest levels around August 8 (day 100). The map of spatial patterns seemed to indicate the presence of a west-east trend, with highest values in the western parts of the Sierra Nevada. Apparently, there are additional spatially and temporally specific variables not in the present model that have significant effects on ozone levels in this region. These variables may include wind levels and directions, site aspect, and more local and accurate weather variables.

3.2. Estimated ozone maps

Maps of estimated ozone levels for 2 days in 1999 are presented in Fig. 10. Similar maps may also be produced for other dates in 1999 season. These maps may be used for studying general ozone patterns or relationships between expected ozone levels and growth changes or injuries in pine species in the region. Fig. 11 presents a sequence of maps showing regions of the Sierra Nevada that were predicted to have had above or below average ozone. In these maps, zero indicates regions where the predicted ozone levels were not significantly different from the average (at the 5% level), while +2 (or -2) indicate regions where the predicted ozone levels were greater (or less) than two standard deviations but less (or greater) than four standard deviations from the average, and so on. Here again we see a general decreasing trend of ozone levels, with the lowest levels around August 10. We also notice significantly higher than average levels appearing mostly in the western and southwestern regions. Some southeastern regions also seem to exhibit higher than average ozone levels. This pattern of high ozone levels was not apparent in the spatial trend effect (Fig. 9) indicating the possibility that higher temperatures and elevations may be the reason for the higher than average values in the southeastern region. However, the number of passive samplers in the southeastern region is small, making conclusions about this region less reliable. Ozone levels in the northeastern region of the Sierra tended to be significantly lower than the season average.

Plots of observed and predicted ozone levels at passive monitor sites may also be of some interest. Fig. 12 presents these plots for nine passive sampler sites. The observed values are the amounts of nitrate converted to O₃ concentration units. The same conversion factors were used to convert the fitted (predicted) nitrate values to ozone values. The Woodsfords site was the outlier

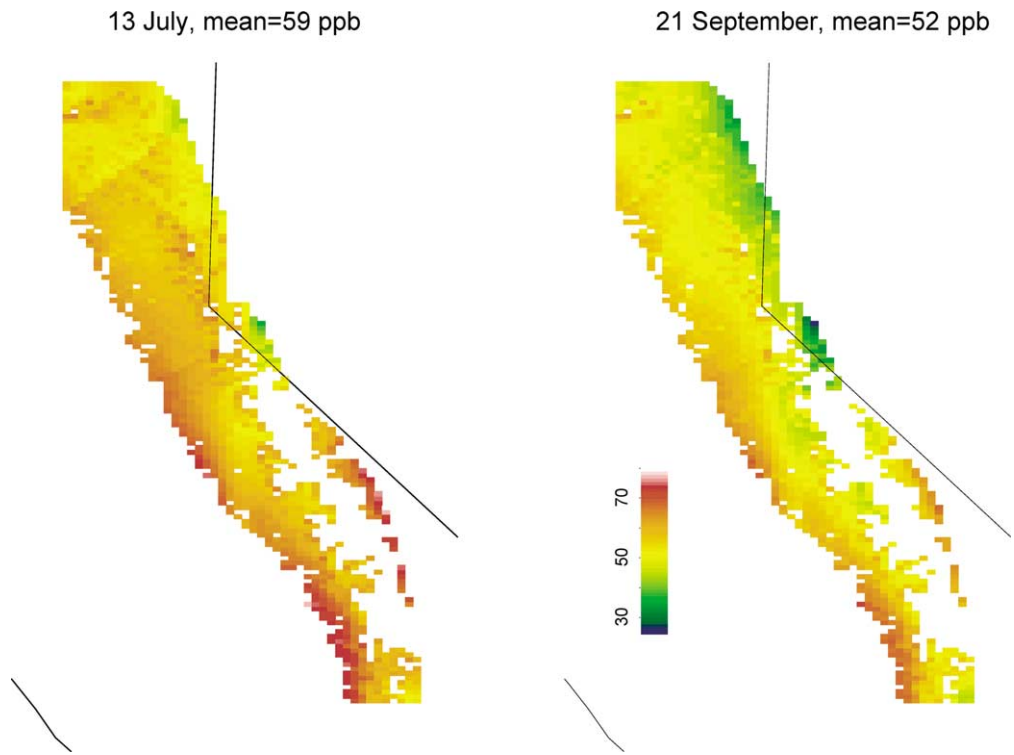


Figure 10. Estimated ambient ozone levels (ppb) for the Sierra Nevada region on 2 days during summer 1999. Estimates of ozone at the highest elevation sites (the regions in white) are not available because there were no passive sampler monitors at elevations > 2500 m.

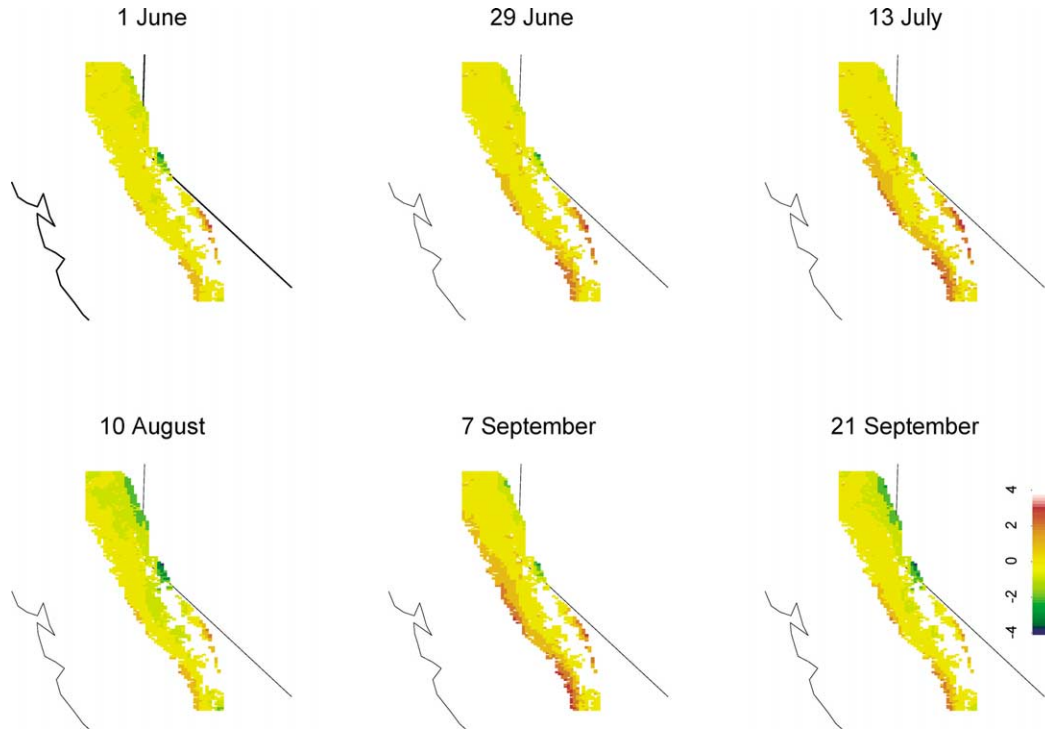


Figure 11. Map of regions where predicted ozone levels are within two standard deviations (SD's) of the overall season average (zero level); regions between two and four SD's from average (+2 or -2 levels); and regions where predicted values are greater than four SD's from average (+4 or -4). The overall average for the season was 53.7 ppb.

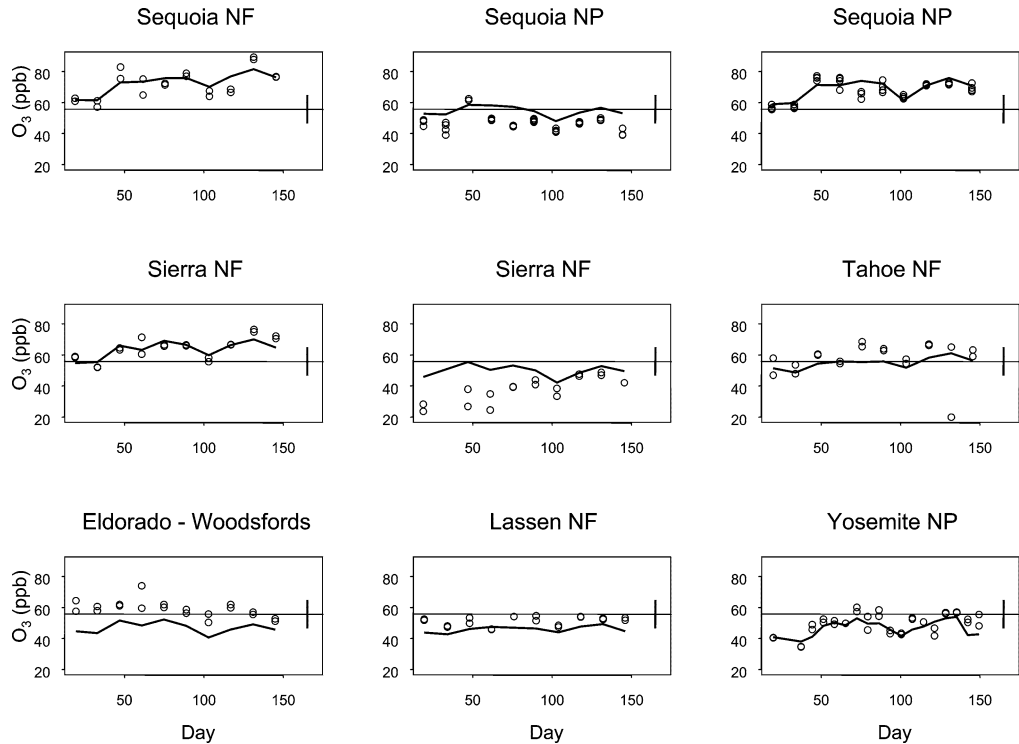


Figure 12. Observed versus estimated ozone levels at nine sites. The horizontal line is the seasonal average ozone level. The length of the vertical bar on the right side of each panel is two times the maximum predictive standard error estimated by using the jackknife technique.

site mentioned in Fig. 5. None of the auxiliary variables included in the model appear to account for the higher than expected ozone levels at this site. Also, the increasing trend seen at one of the Sierra National Forest sites in Fig. 12 does not seem to be accounted for by the temporally explicit weather covariates in the model. However, similar increasing trends at other sites (e.g., the Sequoia National Forest site) seem to be well accounted for by the weather covariates. The relatively large between-site variations and, consequently, the bias seen at some of the sites (e.g., the Eldorado and Lassen NF sites in Fig. 12) seem to imply that improvement in model prediction might be possible if additional site characteristics are included in the model. Another explanation for the relatively large between-site variations might be the need for better *in situ* calibration of the active monitors.

4. Discussion

The statistical approach used in this study was an extension of ordinary linear regression techniques to nonlinear and spatially correlated cases. By using this approach and data from a network of 79 passive monitors, we were able to estimate ozone levels and generate a sequence of spatial maps spanning a period of 5 months (May–September, 1999) that described the patterns of ozone levels in the Sierra Nevada. Additionally, the approach was useful for studying relationships between ozone levels and explanatory variables and for locating regions of the Sierra Nevada that are at high risk of being exposed to what could be critically high levels of ozone.

Defining the model as a regression model provided us with a flexible framework for determining uncertainties, assessing goodness of fit, and detecting observations that are not adequately predicted by the model. The latter should aid scientists in finding ways to improve the model, for example, by identifying additional explanatory variables and deciding whether the new variables need to be spatially explicit (e.g., more site specific characteristics) or temporally explicit (e.g., more or better weather variables). The modeling framework also allows the formal comparison of ozone values at sites with similar environmental and topographic conditions. For example, the Woodsfords site (Eldorado National Forest) stood out as an outlier because the observed ozone values at this site were higher than ozone values estimated from surrounding sites (spatial component of the model) and sites with similar elevation and temperature ranges (auxiliary variables in the model).

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