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Propagation of Uncertainty in Hourly Utility NO_x Emissions through a Photochemical Grid Air Quality Model: A Case Study for the Charlotte, NC, Modeling Domain

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One of the major hypothesized sources of uncertainties in air quality model inputs is the emission inventory. A probabilistic hourly NO_x emission inventory for 32 units of nine coal-fired power plants in the Charlotte domain for the year 1995 was propagated through the Multiscale Air Quality Simulation Platform (MAQSIP). The inventory was developed using time series techniques. Time series for a 4-d episode were simulated and propagated through the air quality model 50 times in order to represent the ranges of uncertainty in hourly emissions and predicted ozone levels. Intra-unit autocorrelation in emissions and inter-unit dependence were accounted for. The range of uncertainty in predicted ozone was greater when inter-unit dependence was included as compared to when units were treated as statistically independent. Uncertainties in maximum ozone hourly or 8-h concentrations at a specific location could be attributed to a specific power plant based upon regression analysis. Out of 3969 grid cells in the modeling domain, there were 43 and 1654 grid cells with a probability greater than 0.9 of exceeding a 1-h 120 ppb standard and an 8-h 80 ppb standard, respectively. The time series of predicted ozone values had similar autocorrelation as compared to monitored data. The implications of these results for air quality management are addressed.

Introduction

Air quality models are used to predict ambient ozone levels and simulate alternative scenarios for the purpose of setting control strategies (*1*, *2*). Understanding of uncertainties is a prerequisite for better decision making, and the use of range/confidence estimates to describe uncertainty is recommended (*3*−*8*). Sources of uncertainties in predicted ozone concentrations include chemical mechanisms, meteorological inputs, and emission inventories among which deficiencies in emission inputs have been identified as a major source of error (*9*). The National Research Council has recently recommended the use of quantitative uncertainty analysis for mobile source emissions (*6*). International guidance has been issued regarding quantification of uncertainty in greenhouse gas emissions (*10*). Recent efforts have demonstrated methods for quantification of variability and uncertainty in emission factors for a variety of pollutants and emission sources (*11*−*23*). However, there is a need for work focused on development of rigorous methods for quantifying uncertainty in hourly emissions inventories for propagation through air quality models.

Hanna et al. used expert elicitation or judgment to estimate uncertainties in 109 inputs to the Urban Air Shed Model (UAM-IV) and used Monte Carlo simulation to quantify uncertainty in ozone predictions for a New York City 230 km by 290 km domain for the July 6−8, 1998, episode (*24*). There are "uncertainties" in the uncertainty estimates since observations over a wide range of conditions were lacking. They recommended more work to better estimate the uncertainty. They also recommended that correlations greater than 0.5 among inputs should be accounted for in Monte Carlo resampling. Hanna et al. employed a similar approach for the eastern U.S. domain using UAM-V (*25*). They elicited expert judgment to specify distributions for 128 inputs. Bergin et al. used Monte Carlo simulation with Latin hypercube sampling to propagate uncertainties in 51 model inputs through the California/Carnegie Institute of Technology air quality model (*26*). Moore and Londergan applied a Monte Carlo approach to quantify uncertainties in the difference of ozone predictions between a base and control scenario (*27*). Uncertainties in 168 inputs were accounted for using normal and log-normal distributions based on expert judgment. Correlation was accounted for between (i) temperature and VOC emissions and (ii) temperature and chemical rate coefficients. Autocorrelation was accounted for in some meteorological inputs using time series autoregressive models. They concluded that including time-dependent uncertainty had significant effects on the results and recommended the use of more realistic uncertainty models to characterize input uncertainty. In recent work, intra-unit and inter-unit dependence for hourly NO_x emissions of coal-fired power plants was accounted for in developing uncertainty estimates of capacity factors, emission factors, heat rates, and total emissions (*28*−*30*).

This paper extends the recent work by propagating uncertainties in the coal-fired utility NO_x emission inventory through an air quality model. Key questions answered by this paper are as follows: (i) What is the uncertainty in ozone predictions solely attributable to uncertainty in coal-fired utility NO_x emissions? (ii) Can uncertainties in maximum ozone levels be attributed to specific power plant units? (iii) How likely is it that National Ambient Air Quality Standards (NAAQS) will be exceeded? (iv) How important is it to account for inter-unit correlation in emission uncertainties?

Methodology

The methodology employed for air quality modeling, quantifying uncertainties in hourly NO_x emissions, and propagation of uncertainties through the air quality model is described.

Air Quality Modeling. The study focused on a 4-km grid for the Charlotte, NC, domain for a 4-d episode starting from 12 noon (GMT) on July 12, 1995, and ending at 12 noon (GMT) on July 16, 1995. The modeling domain, shown in Figure 2, was 63 by 63 grid cells covering an area of 252 by 252 km. Meteorological inputs were generated using the Pennsylvania State University/National Center for Atmospheric Research mesoscale model (MM5). Estimates of emissions for mobile, area, biogenic, and point sources (excluding coal-fired power plants) were obtained from the North Carolina Department of Environmental and Natural

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Resources (DENR). A probabilistic emission inventory was used for the coal-fired power plants.

The Multiscale Air Quality Simulation Platform (MAQSIP) was used (*31*). MAQSIP is an Eulerian photochemical grid air quality model. The model performance evaluation is summarized in the Supporting Information. The model performance is very good and conforms to acceptable standards for normalized bias and gross error. Required input data include emissions, meteorology, domain grid specification, and initial and boundary conditions. The Sparse Matrix Operator Kernel Emissions (SMOKE) modeling system was used to process the emissions of all source categories to prepare model-ready emissions files required by MAQSIP (*32*).

Probabilistic Emission Inventory for Coal-Fired Power Plant NO_x Emissions. Limitations acknowledged by researchers in previous emissions uncertainty studies are addressed here with specific focus on coal-fired power plant utility NO_x emissions. Hourly continuous emissions data were obtained for 32 units of nine coal-fired power plants, illustrated in Figure 2 in the Charlotte, NC, air quality modeling domain. All of these power plants are baseload plants. The data were analyzed to characterize uncertainty in hourly emissions and to identify the existence of autocorrelation and cycles in the data for each unit, referred to here as intra-unit dependence (*28*). Typical ranges of the coefficient of variation, defined as the standard deviation divided by the mean, for emissions at a given hour of the day were from 0.2 to 0.45. Autoregressive time series models, which model autocorrelation within a time series, were fit to capacity and emission factor data to account for intra-unit dependence; regression models were fit to heat rate data, for purposes of predicting total emissions (*29*). Autoregressive time series models are suitable for time series where the value of a variable in the current time step is correlated with the value of the same variable in previous time steps. Autocorrelation was accounted for in modeling the time series of capacity and emission factors in addition to accounting for inter-unit correlation of capacity factor time series for units within the same plant. Autocorrelations in unit emissions were as high as 0.9 for some lags. To account for statistical dependence between units, referred to here as inter-unit dependence, vector autoregressive time series models were developed for the capacity factor and for the emission factors of selected units. Multiple units at the same plant typically have stronger inter-unit dependence than do units located at different plants. Therefore, the focus was on characterization of dependence among units at the same plant. Inter-unit dependence was attributed primarily to capacity factors, with correlations of typically 0.5−0.8. The linear correlation in emissions between units was typically 0.3−0.6. Results of probabilistic emission inventories in which inter-unit dependence was included were compared to those assuming inter-unit independence. The former was found to more accurately compare with observed data and produced wider ranges of overall uncertainty (*30*). Thus, the vector autoregressive approach was judged to be more appropriate, even though it is more computationally intensive. The base case analyses are based upon 1995 continuous emissions monitoring (CEM) data.

The probabilistic daily inventories obtained using both approaches are presented in Figure S-1 in the Supporting Information. Because the correlations between units were positive, the uncertainty range obtained using the inter-unit dependent approach was wider than that obtained using the inter-unit independent approach. Furthermore, each hour of the day had a different range of uncertainty due to the inherent diurnal variation in emissions.

Simple random Monte Carlo sampling was used in generating the error terms that were added to the time series model deterministic components (*29*, *30*). The advantage of using Monte Carlo simulation is the capability to estimate uncertainties in model outputs with only 50−100 model simulations and to use standard statistical tests to estimate confidence intervals (*24*, *33*). The precision of the estimates for any given percentile of the distribution of emissions was found to be approximately ±3% (*29*, *30*). Thus, the precision of the estimate of uncertainty in emissions is greater than the precision of air quality model predictions, which is often judged to be in a range of ±10% to a factor of 2 or more (*34*). Therefore, there is little benefit to using larger sample sizes for the Monte Carlo simulation.

The fitted time series models were used to generate 50 possible realizations for the total hourly emissions during the 4-d episode using SAS. A realization is a time series that is not statistically distinguishable from the original data set to which the model was fitted. A larger number of iterations was impractical because of the long run time of the air quality model. A Visual Basic program was developed to convert the output from SAS to the hour-specific format required by SMOKE.

Uncertainties in Predicted Ozone Concentrations. Air quality model predictions were made for each of the 50 time series realizations to arrive at 50 possible values of ozone levels for each grid cell and for each time step of the simulation. The distribution of the ozone levels at any given time and location represents uncertainty for a near-term future episode that is a randomized version of recent historical data. This is based on the assumption that emission control technologies and electricity demand are not substantially changed for this near-term future. The maximum ozone level can be identified for each of the 50 realizations. Furthermore, the frequency with which the ozone level exceeds an applicable standard can be estimated. If desired, changes in control technologies can be assessed based upon inter-annual comparisons of CEM data (*28*) and accounted for by making adjustments to key statistics of the time series (*30*). This work focused on evaluating the effect of variability in emissions on the maximum hourly concentration. It is also possible to assess the effect of variability in emissions on other metrics in future work.

The current (as of this writing) Federal NAAQS for ozone are based on an hourly average ozone level of 0.12 ppm. A Metropolitan Statistical Area, the definition of which is described in ref *35*, is designated as nonattainment if the annual average number of exceedences of the daily maximum hourly ozone concentration in the past 3 yr is greater than 1. Exceedences may occur in only a few grid cells and potentially might be influenced by only a subset of emission sources within a domain. The domain includes three MSAs: (i) Charlotte, (ii) Greensboro, and (iii) Hickory.

The location of impact of power plant emissions is identified by correlating the uncertainties in emissions for several time steps with uncertainties in predicted ozone levels, taking into account the plume transport time from the power plant to the location of high ozone values. Moreover, regression analysis is used to quantify the effect of uncertainty in emissions of specific units on uncertainty in ozone maximum hourly predictions that occurred during the entire episode at specific grid cells. As a quality assurance check, the autocorrelation structure of the predicted ambient ozone levels was investigated using the ARIMA procedure in SAS and compared to that of the monitored concentrations at specific grid cells containing monitoring stations.

Results and Discussion

Results obtained from the modeling exercise and a discussion of the uncertainties in predicted ozone concentration due to uncertainties in NO_x emissions from coal-fired power plants are given here.

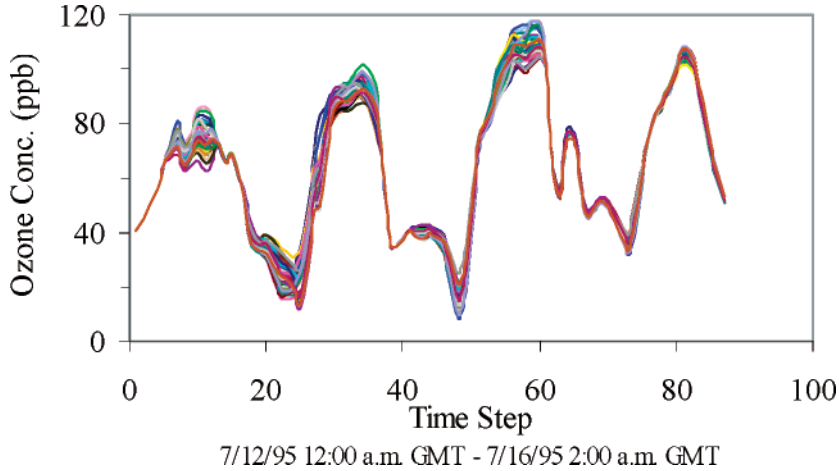

FIGURE 1. 50 predicted time series of ozone 1-h concentrations realizations at cell [26, 23] for the dependent units case.

Uncertainties in NO_x Emissions. Coal-fired power plants contribute approximately 50% of the total NO_x emissions in the domain. Uncertainties in other source categories are being addressed as part of other ongoing work (e.g., refs *20*−*23*). Figure S-2 in the Supporting Information shows an example realization for the Belews Creek plant emissions time series at grid cell [55, 50] and the Robinson plant at grid cell [3, 54]. On average, these two plants contribute 45% and 1.7%, respectively, to the domain total utility NO_x emissions. These two plants contribute 99.95% and 98.8%, respectively, of the NO_x emissions in their respective grid cells. Thus, the time series of total NO_x emissions from the grid cell resembles that of the emissions from the power plant alone. These results were typical for all grid cells containing power plants and are in agreement with those obtained by Chang et al. (*36*). They concluded that NO_x emissions from power plants, which contributed approximately 90% of total point source NO_x emissions in the Atlanta MSA, were responsible for the day-to-day variability in emissions. These results imply that power plants are the most important emission source for grid cells containing them. Hence, accounting for uncertainties in this category will capture most of the uncertainty in total NO_x emissions from these grid cells.

Uncertainties in Ozone Time Series. An example ground-level air quality output is shown in Figure S-3 in the Supporting Information in the form of 50 random time series for ambient ozone concentrations for grid cell [26, 23] based on the assumption of statistically independent emissions of each power plant unit. The figure shows daily variation in ozone and that the range of uncertainty varies by hour. The last 9 hours of the episode, which represent situations in which ozone levels are declining or are low, were ignored in order to cut down the time for running each realization in the air quality model; therefore, only 87 time steps are shown in Figure S-3. The maximum hourly concentration varies among realizations with respect to magnitude and timing. The 50 values for the maximum hourly concentration at each grid cell characterize uncertainty.

Figure 1 shows 50 predicted ozone time series based upon accounting for inter-unit dependence for the same grid cell as that of Figure S-3. Compared to Figure S-3, the uncertainty ranges for different hours of the episode are greater. For example, for hour 56, the range of 1-h ozone values was from 100 to 112 ppb based upon statistically independent units versus from 98 to 114 ppb when inter-unit dependence was accounted for. The absolute range of uncertainty in predicted 1-h ozone concentration tends to be greater during the daytime peaks.

Key Sources of Uncertainty in Hourly Ozone Levels and Sensitivity Analysis. Two methods were employed to identify key sources of uncertainty for ozone levels: (i) simple linear sample correlation analysis of uncertainty in emissions for several hours prior to the time step for which uncertainty in ozone levels are of interest and (ii) linear regression analysis based upon deviations in ozone levels with respect to deviations in emissions. The former characterizes the strength of the linear dependence between ozone levels and emissions at specific plants. The latter can be used to predict the range of uncertainty in ozone levels associated with a specified range of uncertainty in plant emissions.

Stack heights for all power plants and their average mean daily emissions are presented in Table S-1 in the Supporting Information. The impact of plant emissions on ground-level ambient ozone levels is likely to occur at some distance from the plant. For example, the ozone levels at cell [56, 57] were compared with the emissions from the Belews Creek plant located 28 km upwind. The approximate transport time for the Belews Creek plume to reach cell [56, 57] was estimated based on wind speed and direction as illustrated in Figure S-4 in the Supporting Information. A typical transport time of 4 h was inferred. For example, during the 4 h prior to the hour of maximum level, the wind direction and speed varied between 76° and 80° and between 1 and 5 m/s, respectively. Cell [56, 57] is approximately 80° from north relative to the power plant. The total emissions for these 4 h had a correlation of 0.87 with the ozone levels at cell [56, 57]. Figure S-5 in the Supporting Information shows the uncertainty in ozone concentration versus the uncertainty in emissions in the form of a scatterplot. The plot shows a strong dependence between the two.

A similar analysis was conducted for cell [42, 43], for which the ozone levels had a correlation of 0.61 with emissions from the Marshall power plant. Cell [42, 43] is located at an approximate distance of 54 km from the Marshall plant. Belews Creek and Marshall plants contribute approximately 45% and 20%, respectively, of the utility NO_x inventory in the modeling domain. Figure S-6 in the Supporting Information shows the empirical cumulative density functions for the maximum hourly ozone concentrations at grid cells [56, 57] and [42, 43], illustrating ranges of approximately 110−130 ppb in the former and 93−100 ppb in the latter. Results of the analysis for these two cells are summarized in Table 1. Correlation coefficients of less than approximately 0.3 are not statistically significant. Therefore, the range of uncertainty in cell [56, 57] is solely attributable to uncertainty in emissions for Belews Creek, and similarly the range of uncertainty in ozone levels for cell [42, 43] is solely attributable to emissions for Marshall.

Regression analysis is used to estimate the influence of a specified range of uncertainty for emissions of specific

TABLE 1. Correlation Analysis Results for Cells [56, 57] and [42, 43][a]

cell location (row, column)	Allen	Buck	Cliffside	Creek	Danriver	Lee	Marshall	Riverbend	Robinson
[56,57]	0.251	0.132	0.078	0.869	−0.139	−0.151	−0.107	0.243	−0.051
[42, 43]	−0.040	−0.221	−0.013	−0.032	−0.095	−0.026	0.614	−0.137	0.094

[a] Correlations within ±0.32 are not statistically significantly different from zero.

plants with respect to that of maximum ozone levels for a specific grid cell. This methodology was adapted from Hanna et al. (*25*). The maximum ozone levels at cells [56, 57] and [42, 43] were modeled in terms of the difference between emissions for each uncertainty realization with respect to the average of all uncertainty realizations divided by the average of the realizations. Emissions were summed for the 4 h prior to the occurrence of maximum hourly ozone concentration, which are the hours that are most likely to impact ozone concentrations at these grid cells. The resulting regression models were as follows:

For cell [56, 57] ($R^2 = 0.64$):

$$O_3\ (\text{ppm}) = 0.127 + 0.0048S_a + 0.0022S_b + 0.0020S_c + 0.021S_{cr} - 0.00051S_d - 0.0033S_l - 0.0016S_m + 0.0031S_{ri} + 0.0011S_{ro} \quad (1)$$

For cell [42, 43] ($R^2 = 0.74$):

$$O_3\ (\text{ppm}) = 0.097 + 0.0029S_a - 0.0032S_b + 0.00055S_c - 0.000025S_{cr} - 0.0017S_d - 0.00005S_l + 0.0128S_m - 0.0024S_{ri} + 0.00031S_{ro} \quad (2)$$

where $S_i = (E_{i,j} - \bar{E}_i)/\bar{E}_i$; $E_{i,j}$ is the emissions for plant i for uncertainty realization j (t/4 h); $\bar{E}_i$ is the average emissions over all uncertainty realizations for plant i (t/4 h); i stands for the following: a = Allen, b = Buck, c = Cliffside, cr = Belews Creek, d = DanRiver, l = Lee, m = Marshall, ri = Riverbend, and ro = Robinson; and $j = 1-50$.

For grid cell [56, 57], the only statistically significant regression coefficient is for Belews Creek, with a value of 0.021. Thus, if for example there was 50% uncertainty in emissions, there would be an expected uncertainty of $0.5 \times 0.021 = 0.0105$ ppm in maximum ozone concentration. The median ozone concentration at this cell is 0.127 ppm. Therefore, the 0.0105 ppm represents a relative uncertainty of approximately 8% in ozone concentration. Similarly, for grid cell [42, 43], the Marshall plant has the only statistically significant regression coefficient, with a value of 0.0128. A 50% (for example) uncertainty in emissions at this plant causes approximately 7% uncertainty in ozone concentration. An implication is that the relative uncertainties in ozone predictions are less than the relative uncertainties in emissions that caused them.

Ranges of Uncertainty for Maximum Hourly Ozone Levels. The maximum hourly ozone level that occurred during the episode for each realization and each grid cell is depicted in Figure S-7 in the Supporting Information for both the independent and the dependent cases. In both cases, there are grid cells where the maximum hourly concentration exceeds 120 ppb. The dependent case typically produced larger maximum values for many grid cells, with differences of up to 7 ppb as compared to the independent case. Figure 2a depicts the ranges of uncertainties for ozone concentrations for the dependent case. Figure 2b depicts the difference in ranges from the two cases.The dependent approach produced wider ranges of uncertainties in ozone levels for most grid cells due to the wider range of uncertainties in emissions**.** The widest ranges occur in regions associated with the local impact of the power plants that contribute most to the inventory. For example, the grid cells in the area northeast of Belews Creek, which is the dominant wind direction for several hours preceding the maximum hourly level occurrence in this area, had predicted uncertainty ranges of between 20 and 25 ppb. Similarly, grid cells in areas northeast of Marshall had uncertainty ranges of 9−14 ppb. The dependent approach produced uncertainty ranges as much as 10 ppb wider than those of the independent approach.

These ranges of uncertainty in ozone levels solely attributable to utility NO_x emissions are reasonable when compared to other studies. For example, sensitivity analysis for the Community Multiscale Air Quality model (CMAQ), a model similar to MAQSIP, conducted by Cohan et al. (*37*) estimated a maximum difference of 16 ppb in ozone concentration in response to a 40% change in domain-wide NO_x emissions.

Importance of Inter-Unit Dependence. Results given in Figures S-1, S-6, and 2 show that the range of estimated uncertainty in both emissions and predicted ozone levels is larger when inter-unit dependence is added. To further evaluate the importance of accounting for inter-unit correlation, four grid cells were selected to compare uncertainty in maximum hourly ozone concentrations using both independent and dependent approaches as shown in Figures S-8 and 3. The uncertainty ranges obtained based on the dependent approach are wider than those of the independent approach. For example, the 95% probability range for cell [26, 33] was 12 ppb for the dependent approach and only 7 ppb for the independent approach. In Figures S-8 and 3, the dependent approach typically leads to a longer upper tail of the distribution of ozone values, while results for the lower tail were similar to that of the independent approach. In contrast, however, Figure S-6 shows that, for grid cells that have comparatively high maximum ozone levels, the upper tail of the distributions can be similar for both the dependent and the independent approaches, with more pronounced differences in the lower tail.

Probability of Exceeding National Ambient Air Quality Standards. The cumulative distribution functions for the maximum hourly ozone level obtained at each grid cell based upon the dependent case were used to estimate the probability of exceeding the current 1-h NAAQS for ozone of 0.12 ppm, as shown in Figure 4. A cluster of grid cells downwind of Belews Creek and smaller clusters near the Allen, Riverbend, and Marshall plants have greater than 0.9 probability of exceeding the standard. Out of 3969 grid cells in the modeling domain, a total of 43 cells had 0.9 probability or greater of exceeding the standard, and 134 grid cells had a probability of greater than 0.02 of exceeding the standard. Furthermore, for each of the three MSAs, there are cells that have greater than 0.9 probability of exceeding the standard.

Similar analyses were conducted for 8-h rolling average ozone levels to evaluate the probability of exceeding an anticipated (as of the time of this writing) NAAQS of 80 ppb on an 8-h average. The results are given in Figure 5. There are a total of 1654 cells that have greater than 0.9 probability of exceeding the 8-h standard, with substantial numbers of probable exceedences in all three MSAs. Clearly, as expected,

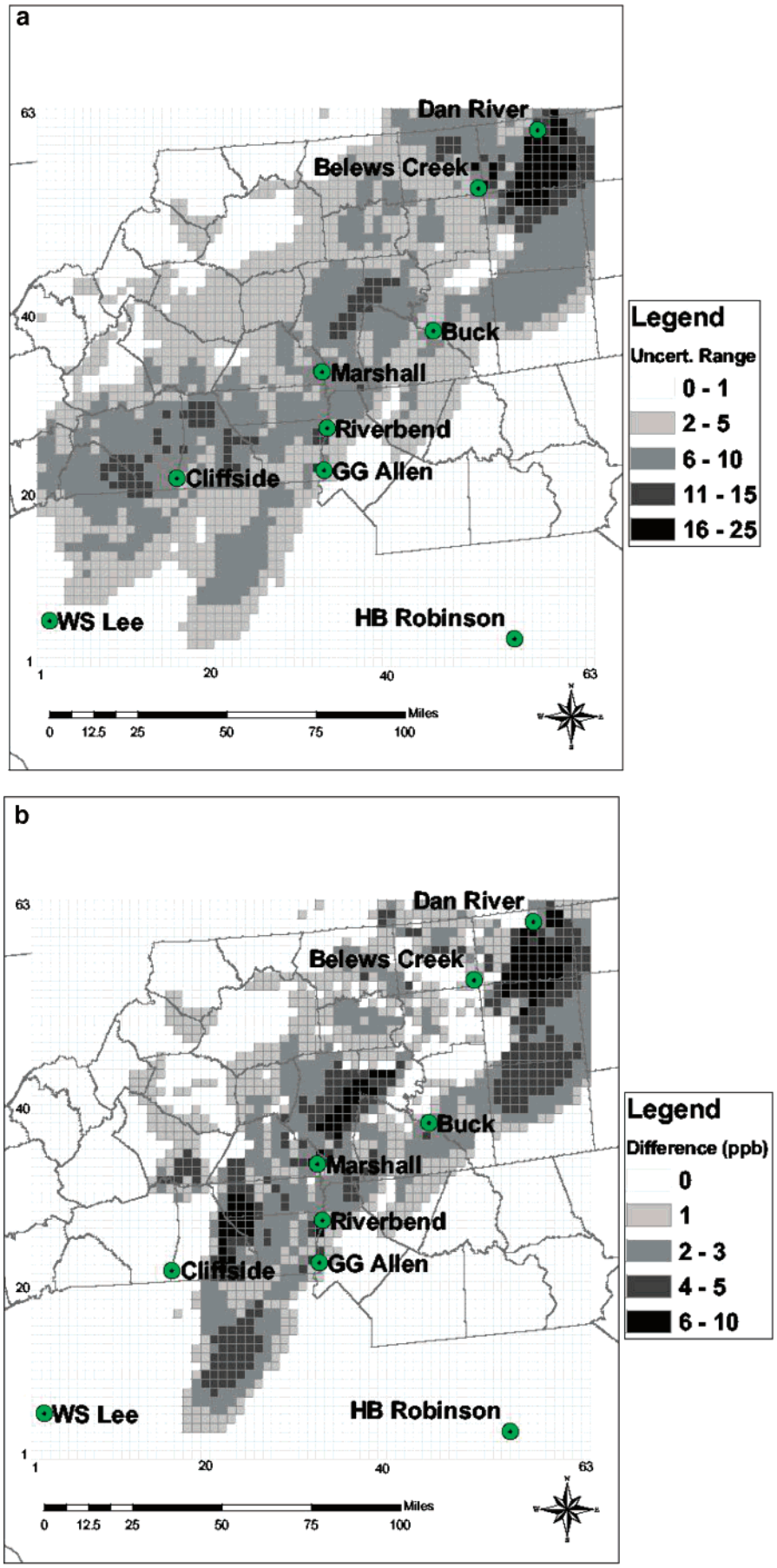

FIGURE 2. Uncertainty ranges of maximum hourly concentration during the 4-d episode of dependent case (a) and difference in ranges from both cases (b).

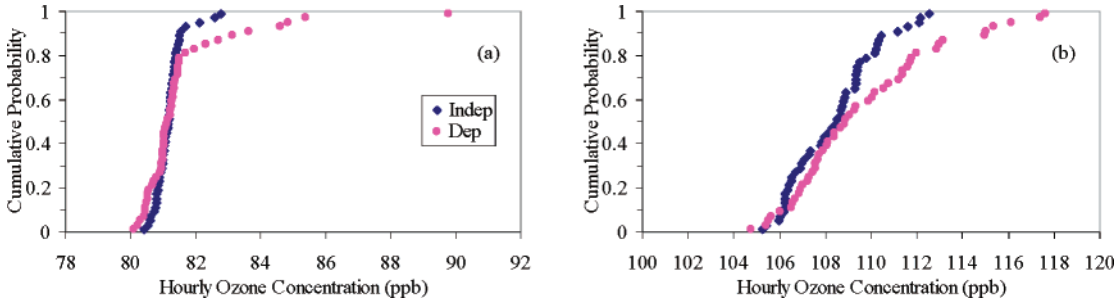

FIGURE 3. Uncertainty in maximum hourly ozone concentration during the 4-d episode for cell [35, 33] (a) and cell [26, 33] (b).

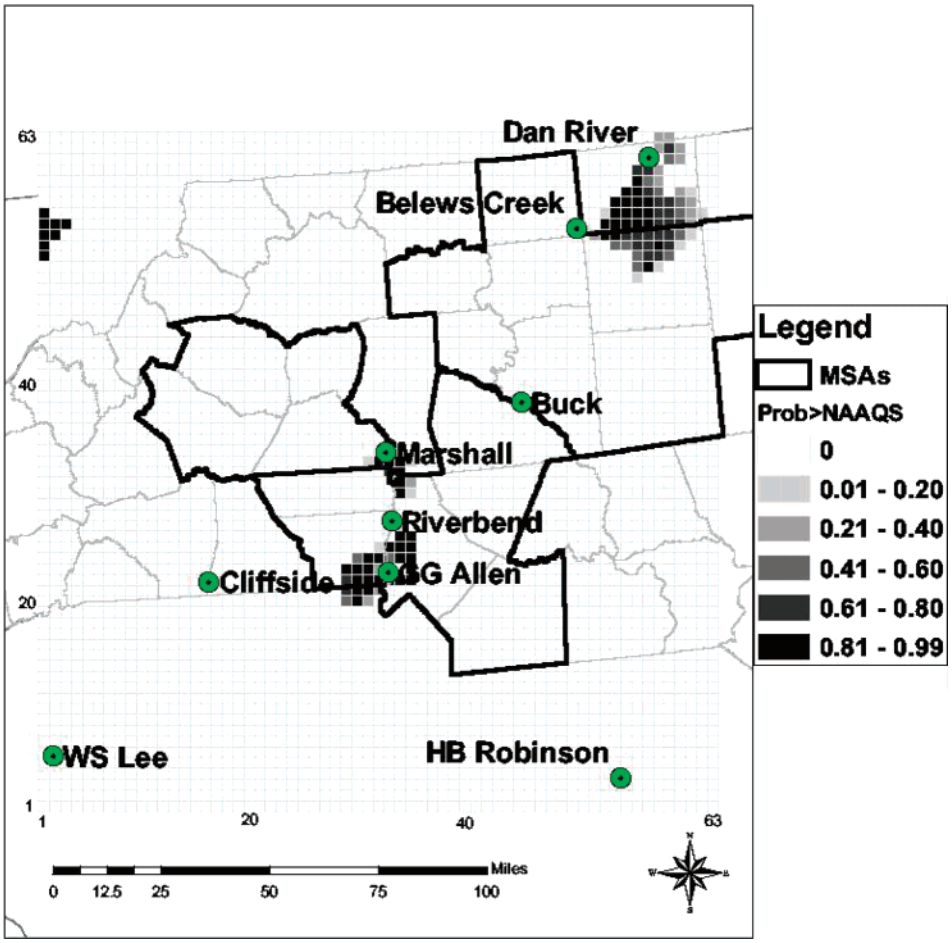

FIGURE 4. Probability of exceeding the 1-h National Ambient Air Quality Standard for Ozone (dependent case).

the 8-h standard is more stringent as compared to the 1-h standard.

Regardless of the applied standard, there is high probability that the three MSAs would be designated as nonattainment if there are no changes in emissions. Although the evaluation of control strategies was beyond the scope of this work, which focused on methods for uncertainty analysis, it is possible to apply the same methodology to evaluate uncertainty in emission inventories for control scenarios and to propagate the uncertainties through air quality models. Changes in emissions associated with changes in control technologies can be evaluated in terms of changes in the mean and standard deviation of hourly emissions (*28*−*30*). An assessment can then be made regarding the probability of exceeding the standard for different control strategies, and a control strategy can be selected that provides an acceptable confidence level of not exceeding the standard.

Autocorrelation. To verify the statistical properties of the simulated ozone values, the autocorrelation structure of monitored ozone ambient levels at three grid cells was checked and compared to that of the predicted ozone time series. Results are given in Table 2 for the autocorrelation of a lag of 1 and 2 h for the monitored and simulated time series. The simulated lag of 1-h values agreed with the monitored values to within −7% to + 5%. The simulated lag 2 values agreed to within −20% to +8%. As noted in Abdel-Aziz and Frey (*29*), time series models tend to underestimate autocorrelation. Therefore, the apparent moderate negative bias in the simulated versus monitored data is expected and is deemed reasonable. For both lags 1 and 2, the simulated autocorrelation values are large, ranging from 0.84 to 0.90 and from 0.62 to 0.72, respectively. Thus, it is clear that there is substantial autocorrelation in monitored ozone values and that the autocorrelation is reasonably captured by the predictions. The autocorrelation is influenced not only by emissions but also by other inputs such as meteorological assumptions. A recommended area for future work is to further investigate which inputs to the model contribute most

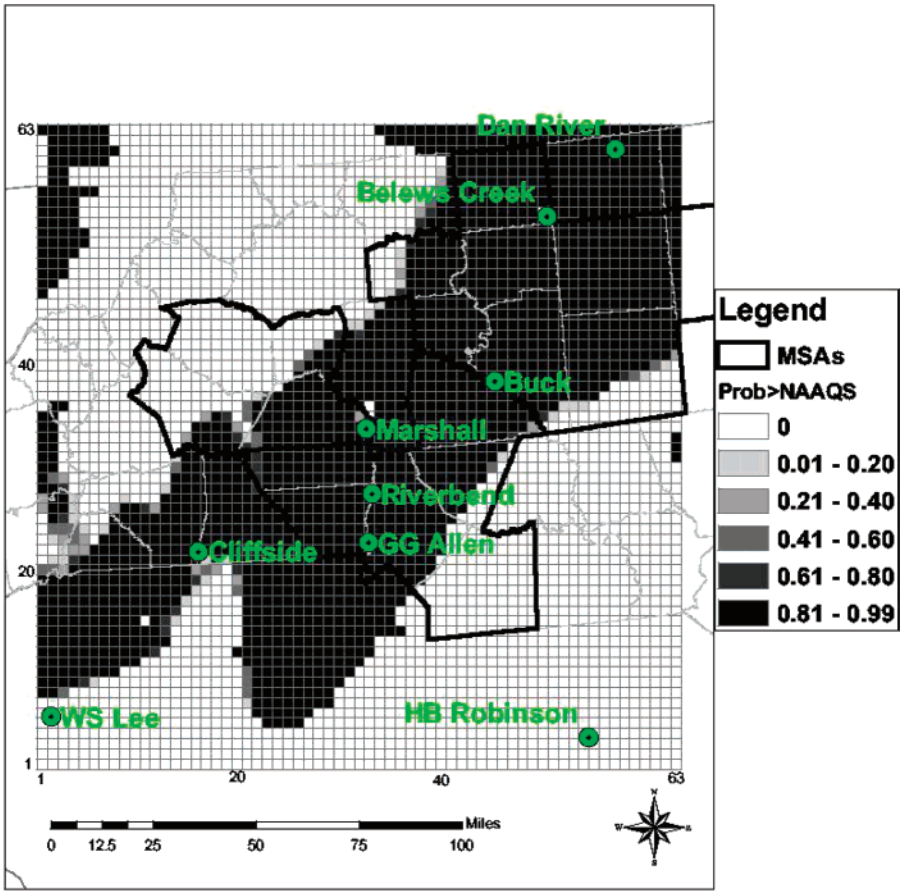

FIGURE 5. Probability of exceeding the 8-h National Ambient Air Quality Standard for Ozone (dependent case).

TABLE 2. Comparison of Autocorrelation Structure of Monitored and Simulated Ozone Time Series

monitoring station	monitored lag 1 ACF[a]	simulated lag 1 ACF	% difference lag 1	monitored lag 2 ACF	simulated lag 2 ACF	% difference lag 2
[56, 55]	0.86	0.90	+4.7	0.67	0.72	+7.5
[49, 47]	0.90	0.86	−4.5	0.77	0.64	−16.9
[51, 47]	0.92	0.84	−6.5	0.77	0.62	−19.5

[a] ACF, autocorrelation function.

to accurate characterization of autocorrelation in predicted values.

Implications for Decision Making. The results of the case study demonstrate that the range of uncertainty in air quality maximum 1-h ozone predictions associated solely with estimation of coal-fired power plant utility NO_x emissions, which was as large as 25 ppb, is potentially large enough to create ambiguity regarding compliance with the NAAQS for any given emissions management strategy. To cope with uncertainty, control strategies can be developed to achieve attainment with an acceptable degree of confidence, such as 90 or 95%. The spatial distribution of the probability of exceeding the NAAQS shows that all three MSAs in the modeling domain are affected.

Comparison of the results of the independent and the dependent approaches implies that erroneous conclusions could be drawn if the independent approach is used. For example, for cell [31, 35] the probability of exceeding the 1-h standard was estimated as 0.11 based upon the independent approach and 0.27 based upon the dependent approach. For some grid cells, the difference in estimated probabilities based upon the two approaches was as large as 0.38. A previous comparison of the inventories obtained using both approaches showed that the dependent approach more accurately represents the observations (*30*). Therefore, decision making should be based on results obtained from the dependent approach.

A potential use of probabilistic air quality modeling results is regarding siting of monitoring stations. Monitoring stations could be spatially allocated where the estimated probability of exceed the NAAQS is estimated to be substantial. However, the comparison of 1- and 8-h standards implies that the identification of an MSA as nonattainment is less sensitive to location in the latter case, since many more grid cells have a substantial probability of exceeding the standard. Thus, in the latter case it would be possible to design a monitoring network that is sufficient to detect nonattainment with a relatively small number of stations. Of course, the location of hotspots could differ for different meteorological episodes, and this could be addressed by conducting analyses for several such episodes to identify common locations of potential exceedences.

This paper has answered several key questions as posed earlier. The range of uncertainty in maximum 1-h ozone values solely attributable to utility NO_x emissions was as large as 25 ppb. Uncertainties in maximum ozone values at specific locations were pinpointed to emissions from a specific power plant, thereby implying that targeted emissions reductions

at those plants could reduce the probability of exceeding the 1-h standard. Under the 8-h standard, it is clear that exceedences are more widespread and not attributable to any one plant. The probability of exceeding a particular NAAQS can be assessed. The results based upon accounting for inter-unit dependence in emissions were different from the independent approach and are considered to be more accurate. Therefore, it is important to account for inter-unit correlation in performing uncertainty analysis of utility NO_x emissions. This work should be extended in the future by rigorously estimating uncertainty in emissions for other source categories.

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Supporting Information Available

One table, eight figures, and a discussion of the model performance evaluation of MAQSIP. This material is available free of charge via the Internet at http://pubs.acs.org.

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