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Uncertainties in predicted ozone concentrations due to input uncertainties for the UAM-V photochemical grid model applied to the July 1995 OTAG domain

Steven R. Hanna[a,*], Zhigang Lu[a,1], H. Christopher Frey[b], Neil Wheeler[b,2], Jeffrey Vukovich[c], Saravanan Arunachalam[c], Mark Fernau[d,3], D. Alan Hansen[e]

[a]*Harvard School of Public Health, Boston, MA, USA*
[b]*North Carolina State University, Raleigh, NC, USA*
[c]*North Carolina Supercomputing Center, Research Triangle Park, NC, USA*
[d]*Earth Tech, Inc., Concord, MA, USA*
[e]*EPRI, Palo Alto, CA, USA*

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Abstract

The photochemical grid model, UAM-V, has been used by regulatory agencies to make decisions concerning emissions controls, based on studies of the July 1995 ozone episode in the eastern US. The current research concerns the effect of the uncertainties in UAM-V input variables (emissions, initial and boundary conditions, meteorological variables, and chemical reactions) on the uncertainties in UAM-V ozone predictions. Uncertainties of 128 input variables have been estimated and most range from about 20% to a factor of two. 100 Monte Carlo runs, each with new resampled values of each of the 128 input variables, have been made for given sets of median emissions assumptions. Emphasis is on the maximum hourly-averaged ozone concentration during the 12–14 July 1995 period. The distribution function of the 100 Monte Carlo predicted domain-wide maximum ozone concentrations is consistently close to log-normal with a 95% uncertainty range extending over plus and minus a factor of about 1.6 from the median. Uncertainties in ozone predictions are found to be most strongly correlated with uncertainties in the NO_2 photolysis rate. Also important are wind speed and direction, relative humidity, cloud cover, and biogenic VOC emissions. Differences in median predicted maximum ozone concentrations for three alternate emissions control assumptions were investigated, with the result that (1) the suggested year-2007 emissions changes would likely be effective in reducing concentrations from those for the year-1995 actual emissions, that (2) an additional 50% NO_x emissions reductions would likely be effective in further reducing concentrations, and that (3) an additional 50% VOC emission reductions may not be effective in further reducing concentrations.

Keywords: Photochemical modeling; Model uncertainty; Monte Carlo uncertainty methods

1. Introduction

Regulatory agencies are proceeding with use of photochemical grid models to make decisions concerning the specific magnitudes of NO_x and VOC emissions reductions that are required in order to reduce concentrations of ozone (see OTAG, 1997; EPA, 1998). There is also a question whether reductions in NO_x or VOC emissions are more effective in reducing maximum ozone

*Correspondence address: CSI Mail Stop 5C3, 103 Science and Technology I, George Mason University, 4400 University Drive, Fairfax, VA 22030-4444, USA. Tel.: +1-703-993-1992; fax: +1-703-993-1993.

E-mail address: shanna@gmu.edu (S.R. Hanna).

[1] Current affiliation: Mass. General Hospital, Boston, MA, USA.

[2] Current affiliation: Sonoma Technology, Inc., Petaluma, CA, USA.

[3] Current affiliation: American Meteorological Society, Boston, MA, USA.

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concentrations. The current research program is intended to provide information to assist this decision process, by providing estimates of uncertainty ranges on the results of emissions management outcomes simulated by a photochemical grid model (UAM-V) used in a regulatory setting. The results of the uncertainty analysis also allow an assessment of the input variables that have the strongest influence on the model predictions of ozone, thus assisting in prioritizing future research efforts towards the variables of most importance.

Of course, it is one problem to estimate the uncertainties in model predictions, and it is quite another problem to decide what to do with this information in the decision process. Currently the EPA is attempting to take model uncertainty into account, at least implicitly, through use of a "relative response factor" in their draft guidance on the use of modeling in attainment demonstrations. This approach recognizes that the models' predictions of relative changes in maximum ozone concentrations due to changes in emissions may be more accurate than the models' predictions of absolute magnitudes of maximum ozone concentrations. This is a first step towards accounting for model uncertainty. The results of Monte Carlo uncertainty studies, such as the exercise described in this paper, should allow a more detailed rationale to be developed.

Hanna et al. (1997, 1998) addressed these objectives by developing and applying a preliminary Monte Carlo uncertainty methodology for assessing the effects of uncertainties in input variables on predictions of the UAM-IV regional photochemical grid model. These two papers analyzed uncertainties in UAM-IV runs on the 180 km by 230 km New York City domain for the state implementation plan (SIP) modeling exercise carried out for the 6–8 July 1988 episode. Because of the promising results from these Monte Carlo runs, it was decided to expand the analysis to a scenario of current regulatory interest – the UAM-V (SAI, 1996) model applications to the so-called ozone transport assessment group (OTAG, 1997; EPA, 1998) domain for the July 1995 ozone episode. The exercise is based on the OTAG "year-1995" emissions and on the projected OTAG "year-2007" emissions, which have been used by OTAG to set specific emissions limits. The OTAG domain, which covers the eastern $\frac{2}{3}$ of the US, is many times larger than the 180 km × 230 km New York City domain studied by Hanna et al. (1998). Fig. 1 contains a map of the OTAG domain with 12 km grid resolution, which was used in the current study, and also indicates 11 subdomains (shaded) which were analyzed by us separately in order to determine if the results varied with geographic location.

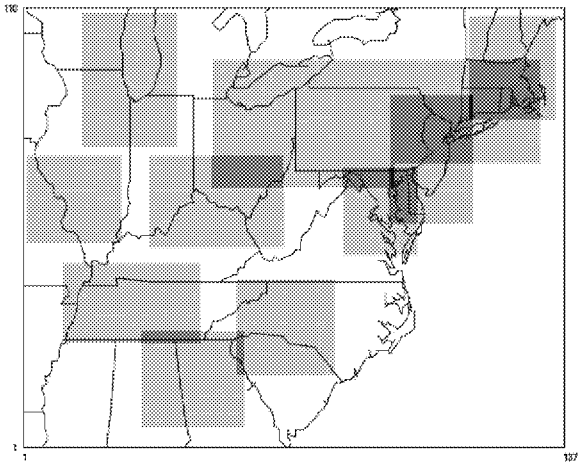

Fig. 1. Map of ozone transport assessment group (OTAG) 12 km geographic domain used for UAM-V model applications. The 11 shaded areas represent sub-domains which were analyzed separately.

2. Approaches to sensitivity and uncertainty analysis

There has been much recent research on the sensitivities, the uncertainties, and the evaluation of environmental models. Beck et al. (1997) provided an overview of evaluations and uncertainties of environmental models, with emphasis on water quality models. They stressed the need to specify a hypothesis or question to be answered by the model, and point out the three major alternatives to sensitivity/uncertainty analysis: (1) first-order error analysis (sometimes called sensitivity or "small perturbation" analysis); (2) brute-force Monte Carlo uncertainty analysis, and (3) response surface evaluation.

Because of its ease of use and easy interpretation, there exist many examples of sensitivity analysis applied to photochemical grid models. Seigneur et al. (1981) estimated the sensitivities of an urban model to variations in input data. Milford et al. (1992) and Seefeld and Stockwell (1999) applied the direct decoupled method (DDM) of sensitivity analysis, with emphasis on variations in chemical rate constants. Seefeld and Stockwell (1999) used first-order sensitivity analysis to look at differences between time-constant and time-variable rate constants, showing that it was important to account for the time variability. Yang et al. (1997) studied the uncertainties in incremental reactivities of VOCs and the fast direct sensitivity analysis of multidimensional photochemical grid models. Saltelli (1999) pointed out that a global sensitivity analysis is preferred over a "one-at-a-time" (OAT) sensitivity analysis, since the latter applies only to a reduced portion of the total space of solutions. Carmichael et al. (1997) suggested the use of an automatic differentiation code (ADIFOR), which allows the derivatives (i.e., the sensitivity coefficients) to be directly solved for in a set of governing equations. Winner et al. (1995) and Dabdub et al. (1999) showed that the ozone predictions are especially sensitive to the inflow boundary conditions

in Los Angeles and the San Joaquin Valley, respectively. Hass et al. (1997) carried out a sensitivity study of four European long-range transport and dispersion models, finding factors of two to three differences in the sensitivities of the different models to variations in emissions. The limitation of local sensitivity analysis is that it applies only in the neighborhood of the nominal parameter values. Since photochemical processes are often non-linear, the magnitude and even the sign of the sensitivity coefficients may vary as the nominal parameter values vary.

At the other extreme from simple "one-at-a-time" sensitivity studies, the response surface method (Tatang et al., 1997) attempts to fit orthogonal polynomials to the input conditions and the predictions of numerical geophysical models. Of course it is necessary to run the models a sufficient number of times to have enough data to develop the response surfaces. It is claimed that 25 to 60 times fewer runs are needed than for a Monte Carlo simple random sampling (SRS) exercise. Nevertheless, the response surface simply amounts to a "model of a model" and therefore is susceptible to problems associated with scenarios outside of the range of parameters used to generate the data for deriving the model.

There has been a rapid growth in the application of Monte Carlo uncertainty analysis methods to atmospheric transport and dispersion models. This "brute-force" method, described by IAEA (1989), NCRP (1996) and Beck et al. (1997), is computer-intensive because it requires many model runs (generally on the order of 100 or more). However, because of the exponential growth of computer speed and storage, it is now possible to easily carry out 100 or more Monte Carlo runs with a complex photochemical grid model such as UAM-V (SAI, 1996) applied to a large domain on the order of 10,000 (100×100) horizontal grid cells. One of the first applications of Monte Carlo uncertainty analysis to photochemistry was the study of relations between stratospheric ozone and chlorine reported by Stolarski et al. (1978). Alcamo and Bartnicki (1987) used Monte Carlo methods to study the uncertainties in sulfur deposition predicted by the EMEF-W model in Europe. They found that it is more important to specify the width (i.e., the standard deviation) rather than the shape of the probability density function (pdf) of the input variables. Gao et al. (1996) applied Monte Carlo uncertainty analysis to the chemical rate parameters. Deuel et al. (1998) studied the uncertainties of the UAM-V model using Monte Carlo methods; however, the uncertainty ranges that they assumed for the input variables (vertical resolution, vertical diffusivity, Plume-in-Grid method, land-use, chemical reaction rates, and emissions) were a factor of three or more less than those recommended by the experts in the study by Hanna et al. (1998) and in the current study. Bergin et al. (1999) applied Monte Carlo methods with latin hypercube sampling (LHS) to a Lagrangian photochemical air pollution model (i.e., not a grid model) in Southern California.

3. Expert elicitation process and resulting uncertainty estimates

The first step in the Monte Carlo uncertainty analysis is to estimate the uncertainties in model input variables, which is difficult because there is little specific information on this subject in the literature for the complete spectrum of inputs (e.g., initial and boundary conditions, emissions components, meteorological variables, photolysis rates, and chemical rate constants). When this is the case, Morgan and Henrion (1990) suggest that it is appropriate to carry out an expert elicitation where "experts" are asked to give estimates of uncertainties based on their experience. The earlier phase of the research (Hanna et al., 1998) depended on advice from ten model experts to assess the uncertainties in UAM-IV model input variables. The current research improved upon this process by attempting to reach about 100 experts via a web page where the experts could enter their estimates of UAM-V model input uncertainties. As a first step, the UAM-V input parameters to be varied were identified by studying the model documentation (SAI, 1996) and by reviewing reports on UAM-V applications to the OTAG domain for the July 1995 episode (e.g., OTAG, 1997; EPA, 1998). As a second step, an expert elicitation process was carried out, from which the distribution functions (shapes and key parameters such as median and variance) for each input variable could be estimated. The medians for the 128 input variables were taken from the OTAG base runs for year-1995 emissions and for projected year-2007 emissions.

Even though 100 experts were contacted, only about 20 of them filled in the requested information on the web page. It was found that better information could be obtained by meeting with groups of experts at several different laboratories. One reason for the minimal written response may be that many photochemical modeling experts have not thought much about uncertainties in input parameters and therefore the estimates are largely based on intuition.

Although most experts agree that it may be important to account for correlations among some input variables, not much information is available on such correlations. For example, there are surely some implicit correlations among some chemical rate constants that should be maintained so that the 94-equation chemical mechanism will produce results that agree with observations in environmental chambers. Furthermore, there should be a balance or correlation among boundary concentrations and emissions so that reasonable ozone concentration predictions are made. However, it is difficult to translate these general concerns about correlations into specific

suggestions concerning the magnitudes of the correlations. Consequently no correlations were used in the current application.

Another problem with the expert elicitation process is that it is difficult to find agreement on how the uncertainties of the eight meteorological input variables (e.g., wind speed, wind direction, and cloud cover) can be adequately accounted for. The major concern is that all photochemical grid models are driven by the outputs of diagnostic or prognostic meteorological models, which impose dynamical constraints such as mass continuity. If the input wind speed and direction were allowed to vary randomly and independently at several monitoring sites at a given hour, there would be spurious mass convergences and divergences that would arise. This would lead to unrealistic build-up of pollutants in some areas and removal of pollutants in other areas. Therefore there would be a need to apply a diagnostic or prognostic meteorological model to each new and independent group of randomly generated wind observations in order to adjust the wind fields to assure that the mass-continuity criterion is satisfied. Because of the great effort involved in rerunning a diagnostic or prognostic meteorological model each time a new set of randomly generated winds is prescribed, our approach has been to vary all wind speeds uniformly across the entire domain, thus assuring mass-continuity is maintained. Another approach could be to make use of available alternate meteorological model runs made by other groups; however, this method does not adequately sample the full range of variability. Future research should focus on better methods for accounting for variability in meteorological inputs, such as applying the Monte Carlo uncertainty methodology to the diagnostic or prognostic meteorological models themselves.

The distribution functions for most UAM-V input variables were assumed to be log-normal, as found for most environmental geophysical variables. The exceptions are wind direction, ambient temperature, relative humidity, and cloud cover, which are assumed to have normal distributions. The experts were asked to give estimates of the uncertainty range that would include 95% of the possible values (i.e., from the 2.5th percentile to the 97.5th percentile of the cumulative distribution function (CDF)). For normal distributions, the standard deviation then equals about $\frac{1}{4}$ of this range. For log-normal distributions, the uncertainties were usually expressed as "plus and minus a factor of Y" for variables with large ranges such as plus and minus a factor of 2 or 3. For variables with smaller ranges, the uncertainty was usually expressed as "plus and minus Z percent", which can be considered equivalent to "plus and minus a factor of $(Y = 1.0 + Z/100)$". For a log-normal distribution, the standard deviation of $\ln(X)$, where X is the dimensional value of the physical variable, equals $0.5\ln(Y)$. For example, the input initial ozone concentration, $C_{O_3 i}$, is assumed to have an uncertainty of plus and minus a factor of 3 (encompassing 95% of possible values). Therefore the standard deviation of $\ln(C_{O_3 i})$ is $0.5\ln(3) = 0.55$.

A list of the input variables, their 95% uncertainty ranges, their assumed distribution functions (all are either log-normal or normal), and the standard deviations of the natural logarithm of the input variable (for log-normal distributions) or the input variable itself (for normal distributions) are given in Table 1. To save space, only the range, median, and mode of the uncertainties of the 94 carbon bond (CB)-IV chemical rate constants are listed. More detailed justifications for the uncertainty estimates for the six photolysis rates and the 94 chemical reaction rates are given by Frey (1998). The uncertainty estimates for many of the chemical reaction rates are based on information published in papers by Atkinson and Lloyd (1984), Thompson and Stewart (1991), DeMore et al. (1994), Gao et al. (1995), and Russell et al. (1995), as well as guidance from several atmospheric chemists. Note that there is a factor of two uncertainty in all the photolysis rates and a median factor of 1.8 uncertainty in the 94 chemical rate constants. However, for many input parameters, the data required to estimate the uncertainties are not available.

The six "photolysis rates" are associated or linked with six of the 94 chemical reactions. Of particular interest is the NO_2 photolysis rate, which is associated or linked with reaction 1: $NO_2 \rightarrow NO + O$. During our study we frequently encountered confusion concerning the differences between the uncertainties in the photolysis rates and the uncertainties in the reaction rates. The difference can be explained by considering that, for each photolysis reaction, there are two separately modeled sources of uncertainty. One source of uncertainty, in the photolysis rate, is related to cloud cover, ozone column, etc., which leads to uncertainty in the estimate of actinic flux and, hence, provides a distribution of nominal photolysis rates. The other source of uncertainty in, for example, the chemical reaction $NO_2 \rightarrow h\nu \rightarrow NO + O$, is related to absorption cross section and quantum yield. This leads to additional uncertainty, since even for a given actinic flux we do not have exact knowledge of what the photolysis rate really is. The uncertainty in photolysis rate as considered as the reaction $NO_2 \rightarrow h\nu \rightarrow NO + O$ has to do with uncertainty regarding absorption cross section and quantum yield, assuming that actinic flux is known. In other words, even if cloud cover, ozone column, etc. were exactly known, so that actinic flux were exactly known, there would still be uncertainty in the photolysis rate because of uncertainty regarding absorption cross section and quantum yield.

The uncertainties in emissions rate in Table 1 are mostly "factor of two" except for major point sources, which are a little better known ("factor of 1.5"). We initially considered the suggestions of some experts that some emissions classes (e.g., biogenic or mobile VOC

Table 1
Uncertainty ranges (include 95% of data) and associated sigmas (standard deviations of log-transformed data) for the 128 UAM-V input variables studied in the Monte Carlo runs. A range defined by plus and minus a "factor of ..." uncertainty encompasses 95 % of the data. for small uncertainty factors (i.e., less than 2), a factor of $1+x$ uncertainty can be considered to be "plus and minus $100 \times$ %

Variable	Variable number	Uncertainty range (includes 95% of data)	Sigma (log-normal unless noted)
Initial ozone concentration	1	Factor of 3	0.549
Initial NO_x concentration	2	Factor of 5	0.805
Initial VOC concentration	3	Factor of 5	0.805
Top ozone concentration	4	Factor of 1.5 (50%)	0.203
Top NO_x concentration	5	Factor of 3	0.549
Top VOC concentration	6	Factor of 3	0.549
Side ozone concentration	7	Factor of 1.5	0.203
Side NO_x concentration	8	Factor of 3	0.549
Side VOC concentration	9	Factor of 3	0.549
Major point NO_x emissions	10	Factor of 1.5	0.203
Major point VOC emissions	11	Factor of 1.5	0.203
Wind speed	12	Factor of 1.5	0.203
Wind direction	13	$\pm 40^\circ$	20.0 degrees (normal)
Ambient temperature	14	± 3 K	1.5 K (normal)
H_2O concentration (as RH)	15	30%	15.0% (normal)
Vertical diffusivity (8AM–6PM; <1000 M AGL)	16	Factor of 1.3 (30%)	0.131
Vertical diffusivity (all other times and heights)	128	Factor of 3	0.549
Rainfall amount	17	Factor of 2	0.347
Cloud cover (tenths)	18	30%	15% (normal)
Cloud liquid water content	19	Factor of 2	0.347
Area biogenic NO_x emiss.	20	Factor of 2	0.347
Area biogenic VOC emiss.	21	Factor of 2	0.347
Area mobile NO_x emiss.	22	Factor of 2	0.347
Area mobile VOC emiss.	23	Factor of 2	0.347
Area low point VOC emiss.	25	Factor of 2	0.347
Other area NO_x emissions	26	Factor of 2	0.347
Other area VOC emissions	27	Factor of 2	0.347
NO_2, HCHOr, HCHOs, ALDs, and O3-O1 Photolysis rates	28–33	Factor of 2	0.347
CB-4 reactions 1–94	34–127	Factor of 1.01 to 3.02 Median 1.80, Mode 2.5	0.10 to 0.55 Median 0.30, Mode 0.46

emissions) were biased low by a factor of two or more, but decided not to include this median bias in the input specifications. The reason is that the EPA's OTAG exercise made use of the original (possibly biased) emissions and it would shift the regulatory "baseline" if the median emissions were shifted. Consequently there was assumed to be no bias in the medians of any input variables, including emissions.

The uncertainties in side boundary conditions are not expected to influence the UAM-V ozone predictions very much on the large OTAG geographic domain (see Fig. 1), where the upwind (south and west) sides are many hundreds of km from the regions of high ozone concentrations. However, the uncertainties in the top boundary conditions may have an effect because they could mix down into the boundary layer during the day. The uncertainties in initial conditions are not expected to have much effect because of the long (two day) "spin up" time for the model before the predictions are considered for analysis.

We did not account for the variability in parameters that are not specifically part of the UAM-V input file but are found inside the UAM-V code, which is proprietary (i.e., no changes were made to the statements in the code).

Variations in parameters such as horizontal and vertical grid size, time step, and domain size are known to influence model predictions. For example, Tesche et al. (1998) suggest that the vertical grid resolution in the UAM-V model was too coarse in the OTAG applications, leading to unrealistic injection heights for emissions from large tall-stack point sources, and, thus, poor simulation of the contribution of these emissions to ozone in the domain. Tesche et al. (1998) also suggest that the model predictions are dependent on the horizontal

grid resolution. However, because it requires at least one week of labor to reinterpolate emissions, meteorology, and other spatially-variable inputs each time the grid size is changed, we did not perturb those variables. We are studying variations in only those variables that can easily be perturbed in the model input file.

4. Monte Carlo method as applied to the July 1995 OTAG scenario

A long-term goal of the research is to develop a software framework that would allow the uncertainties of regional photochemical grid models to be determined on other domains and for other episodes. The software used in the current UAM-V applications is based on a framework that assumes that the user specifies a set of N model input parameters, their mean and variance, and their probability density function (pdf), as shown in Table 1. The Monte Carlo resampling software then generates a batch file containing M randomly selected sets of N input parameters that enable the user to run the model M times with perturbed input data. Certain key output parameters (e.g., predicted hourly ozone concentration in the lowest model level) from each of the M runs are then retrieved from the very large sets of model output files. The other model outputs (several thousand megabytes for each photochemical grid model run) are stored for possible future analysis.

The OTAG domain is shown in Fig. 1 and a 12 km horizontal grid resolution is used by the UAM-V model, as for the OTAG (1997) and EPA (1998) exercises. Even though the OTAG ozone episode continued from 7–18 July, it was decided that computer time could be saved if the simulations were stopped after 14 July, since the maximum concentrations of ozone were similar during the days that followed. The "ramp-up" period for the UAM-V model was 7–11 July and the model results were analyzed for the 12–14 July period.

We believe that the 7–14 July 1995 base case makes a reasonable foundation for the study of model uncertainty, primarily because of the reliance of the EPA on this episode for their decisions concerning emissions controls. This base case has been the subject of many investigations concerning its adequacy for determining emissions control strategies. Some potential problem areas have been identified in other research projects. For example, Lurmann and Kumar (1997) point out a "bias creep" in the UAM-V model predictions for this episode, meaning that the model tends to underpredict in the early days of the episode, and then tends to overpredict in the later days. Dolwick et al. (1998) suggest that there may be unreasonable accumulations of ozone and precursors aloft in the model.

The median or base model inputs for the year-1995 and year-2007 sets of 100 Monte Carlo runs were the same as those used in the OTAG runs (OTAG, 1997; EPA, 1998) for the year-1995 and year-2007 emissions, respectively. It should be mentioned that the emissions files were the only input variables that were different between the four sets of 100 Monte Carlo runs. An initial model run was made for both year-1995 and year-2007 to assure that the current model outputs are consistent with the earlier OTAG model outputs. An additional two sets of 100 Monte Carlo runs were made with 50% reductions in year-2007 anthropogenic NO_x emissions and 50% reductions in year-2007 anthropogenic VOC emissions, respectively. The base case median boundary and initial concentrations were not changed for these four sets of emissions scenarios, consistent with the assumptions in the OTAG runs. In reality, there would be expected to be a correlation between emissions and initial and boundary conditions, but there is no guidance in the literature on what these correlations would be. For a domain as large as the OTAG domain, the maximum ozone concentrations in the middle of the domain are expected to be little influenced by the initial and boundary conditions. Another important fact is that the sets of 128 random perturbation numbers for the 100 Monte Carlo runs were identical over the four base emissions scenarios, thus allowing differences in predictions between the sets of runs to be better assessed.

The primary output variables saved for special analysis are maximum daily one- and 8 hr-averaged ozone concentration in the lowest grid layer during the 12–14 July period for three alternate geographic definitions: (1) at any position on the entire OTAG domain, (2) at any position within 11 subdomains (Atlanta, Nashville, Chicago, Louisville, Charlotte, St. Louis, New York, New England, Philadelphia, Baltimore-Washington, Richmond, and Pittsburgh) within the larger OTAG domain, and (3) at 155 specific locations where there are routine monitors or where there is a special interest. In addition, the same output information is saved for NO_x and VOC, except only for 1-hr averages. Fig. 1 showed the OTAG domain and the 11 subdomains. Most of the subdomains are so-called UAM regions; however, the Pittsburgh subdomain was expanded to include part of the Ohio River Valley and the Lake Erie shore. The 155 specific locations are scattered throughout the domain shown in Fig. 1. Of the 155 specific locations, 135 are routine monitoring sites, and 20 are "artificial" sites which have been arbitrarily located in "holes" in the domain where there were no routine monitoring sites.

The analysis is concerned with three primary topics: (1) gross uncertainties in outputs, (2) correlations and regressions among inputs and outputs, and (3) differences in outputs depending on median emissions assumptions. We have the most confidence in the results for the first topic, gross uncertainties in outputs, which are discussed in Sections 5.1 and 5.2. For example, with 100 Monte Carlo runs, the variance in the output variables can be

well defined. It is possible to use the outputs of the 100 Monte Carlo runs to determine the range or variance in the predicted maximum daily hourly averaged ozone concentration.

There is more uncertainty associated with the analysis of the results for the second topic, the correlation coefficients and regression relations among inputs and outputs, which are discussed in Section 6. With random selection of any 100 pairs of independent variables, there is a 5% probability that the absolute magnitude of the calculated correlation coefficient will exceed 0.19 (this number is twice the standard deviation, $(N)^{-1/2}$). For the earlier research, where there were only 50 Monte Carlo runs, Hanna et al. (1997, 1998) emphasized only those variables with correlation coefficients with absolute magnitudes of 0.28 (or 2 times $50^{-1/2}$) or greater.

To address topic three, the outputs from the four different base emission runs were analyzed to study the magnitude and direction of the changes in predicted ozone concentration. Because each of the four sets of Monte Carlo runs used the same sets of random numbers, the differences were calculated between the 100 pairs of maximum predicted hourly averaged ozone concentration for each pair of base emissions runs and the resulting CDFs were determined. This was done for the entire domain ("all-domain") for the four sets of 100 runs, and was also done for the 11 subdomains and for the 155 individual sites. Any differences in ozone concentration predictions would be primarily due to the difference in base emissions. The estimated 95% range on the difference in predicted ozone concentration for two sets of emission runs was analyzed to see if the 95% range overlapped zero. If the 95% range does not overlap zero, then it can be inferred that there is a real difference between the ozone predictions by the two sets of emissions runs.

5. Uncertainties in ozone predictions

5.1. Cumulative distribution functions of uncertainty in predicted ozone maxima

The cumulative distribution functions (CDFs) of the 100 Monte Carlo predicted maximum domain-wide hourly averaged ozone concentrations were close to log-normal for all four base emissions cases, with a standard deviation of ln C, $\sigma_{\ln C}$, of 0.22 or 0.23. This standard deviation is equivalent to "factor of 1.6" variability in C within the 2.5th and 97.5th points on the CDF. To illustrate the consistent spread of the distributions, Table 2 lists the 2.5th, 50th, and 97.5th percentiles of the distributions of predicted maximum hourly averaged ozone concentration, C (ppm), over the 12–14 July 1995 period, for the 11 sub-domains (see Fig. 1) and for the entire OTAG domain (all-domain) from the 100 Monte Carlo runs with the median year-2007 projected emissions. The table clearly shows the consistent result there is close to a factor of 1.6 ratio for $C(50\text{th})/C(2.5\text{th})$ and for $C(97.5\text{th})/C(50\text{th})$.

Table 2
2.5th, 50th, and 97.5th points on the cumulative distribution function (CDF) of the 100 Monte Carlo predictions of maximum hourly averaged maximum ozone concentration (ppm) for the 11 sub-domains and for the entire domain for the 12–14 July 1995 ozone period and for year-2007 median emissions, for the OTAG domain (12 km grid)

	Monte Carlo		
Sub-domain	2.5th	50th (median)	97.5th
Atlanta	0.09 ppm	0.17 ppm	0.32 ppm
Balt-Wash	0.08	0.14	0.22
Nashville	0.07	0.12	0.21
Chicago	0.07	0.12	0.19
Louisville	0.06	0.12	0.19
Pittsburgh	0.07	0.12	0.18
Philly	0.07	0.11	0.19
New York	0.06	0.11	0.19
New England	0.06	0.11	0.19
Charlotte	0.07	0.11	0.18
St. Louis	0.06	0.09	0.15
(All Domain)	0.13 ppm	0.19 ppm	0.32 ppm

Very similar CDF shapes occurred for one and eight-hour averages, for the complete OTAG domain, for the 11 sub-domains shown in Fig. 1, for the 155 monitoring sites, and for the four groups of emissions scenarios.

These results concerning the CDF shapes are in agreement with the results reported by Hanna et al. (1998) for the New York City domain for the July 1988 episode. It can be concluded that there is consistency of the log-normal distributions, with $\sigma_{\ln C} = 0.23$, across a wide variety of sites and emissions control scenarios.

5.2. Locations of Monte-Carlo-predicted ozone maxima

The locations of the predicted maximum domain-wide 1-h and 8-h averaged ozone concentration occurred primarily in the Southeast, in the Atlanta UAM domain, for most of the UAM-V Monte Carlo runs on the OTAG domain for the 12–14 July 1995 period. The location of the observed ozone maximum also occurred in the Atlanta UAM domain. For a few of the Monte Carlo runs, the location of the predicted ozone maximum switched to the Chicago domain or the New York domain. Weather maps show that the Northeast was influenced by clouds prior to 13 July 1995, which tended to inhibit the formation of ozone in that area. The peak ozone concentration in the northeast did increase during 13–14 July, when hot and sunny weather occurred, accompanied by southwest winds as a cold front approached. In the meantime, the

Southeast was hot and dry with relatively high observed ozone concentrations throughout the 1995 episode.

Because of the location of the ozone maximum in the Atlanta domain, it is expected that some of the correlations with input variables would be different than those found in the earlier study on the New York City domain. For example, Hanna et al. (1998) found that the variability in predicted ozone concentrations in the New York City domain were influenced by the variability in emissions in the large upwind New York City and New Jersey megalopolis and by variabilities in the upwind boundary conditions in central New Jersey. However, for the current study, there is not a huge megalopolis in the upwind sector and the boundaries are relatively distant.

5.3. *Effects of assumptions of emissions controls on uncertainties of outputs*

The medians of the Monte Carlo outputs of predicted maximum ozone concentration can be analyzed to arrive at conclusions concerning the effects of emissions controls. Define C_{50} as the median or 50th percentile of the distribution of 100 Monte Carlo outputs. The following results are generally valid for the all-domain hourly averaged ozone maximum and also for the maxima in the 11 individual sub-domain: C_{50}(2007 emissions)/C_{50}(1995 emissions) = 0.88; C_{50} (2007 with 50% NO_x reductions)/C_{50}(2007) = 0.77; and C_{50}(2007 with 50% VOC reductions)/C_{50}(2007) = 0.97.

Despite the fact that the median ozone concentrations show 5 or 10 or 20% changes downwards as a result of emissions reductions, it is not obvious whether these differences are less than or greater than the expected variability due to random processes. Because of possible correlations among similar runs, it is not correct to use the full Monte Carlo uncertainties ($\sigma_{\ln C} = 0.23$) to calculate the expected variability of differences in predicted ozone concentration for sets of runs with different base emissions assumptions. Fortunately the four sets of Monte Carlo runs were carried out in such a way that the expected variability of differences can be estimated from the available Monte Carlo output files. This is because each of the four sets of 100 Monte Carlo runs used the same 100 (number of Monte Carlo runs) by 128 (number of input variables) matrix of random numbers. For example, Monte Carlo "run 14" would use the same random number to perturb the wind speed for the 1995 base emissions runs, for the 2007 base emissions runs, and for the 50% NO_x and 50% VOC reduction runs. Therefore, for Monte Carlo "run 14", the difference in the predicted maximum ozone concentration between the 2007 base emissions runs and the 2007 base emissions with 50% VOC reductions runs would be solely due to the 50% emissions change.

The CDFs of the differences in all-domain predicted maximum hourly averaged ozone concentration between the Monte Carlo runs for paired sets of base emissions assumptions were calculated for the three pairs of differences (1995 base–2007 base; 2007 base–2007 base with further 50% NO_x reductions; and 2007 base–2007 base with further 50% VOC reductions). The 2.5th, 50th (median), and 97.5th points on the CDF were identified and the "95% range" assumed to be bounded by the 2.5th and 97.5th points on the CDF. Table 3 contains the results.

For comparison purposes, the second column of Table 3 lists the median predicted maximum all-domain hourly averaged ozone concentration for the 1995 or the 2007 base emissions. Note that, in the first two rows (for the difference between 1995 and 2007 base emissions and for the difference between 2007 base emissions and 2007 with 50% NO_x emission cuts), the 95% range does not overlap zero, implying that the difference may be real and not due to random variability. However, in the last row (for the difference between 2007 base emissions and 2007 with 50% VOC emissions cuts), the 95% range barely includes zero, implying that the slight difference may be due only to random variability. Note that the 95% range on the difference in the first row encompasses about 0.04 ppm, while the 95% range for the total Monte Carlo uncertainty for the "all-domain" case in Table 2 encompasses about 0.19 ppm. Thus the uncertainty in the differences in ozone predictions due to emissions changes is about one-fifth of the total uncertainty in ozone predictions due to all model inputs. This result corroborates the assumption underlying the EPA's use of the "relative response factor" in demonstrating attainment, which is based on the assumption that the uncertainty in the

Table 3
Ninty five percent range on differences in maximum hourly averaged ozone predictions between sets of 100 Monte Carlo runs using different assumptions for base emissions. For the entire OTAG domain over the 12–14 July 1995 period

Base emission assumption	Median conc. of base (ppm)	Median conc. difference (ppm)	95% range on conc. difference (ppm)
1995 base–2007 base	C (1995 base) = 0.21 ppm	0.024	0.011 to 0.052
2007 base–2007 NO_x cuts	C (2007 base) = 0.19 ppm	0.043	0.023 to 0.074
2007 base–2007 VOC cuts	C (2007 base) = 0.19 ppm	0.006	0.000 to 0.022

difference of ozone maximum predictions between the base and emissions control runs is less than the uncertainty in the ozone maximum predictions themselves.

The discussion above and the data in Table 3 deal with the differences in the maximum hourly averaged ozone concentration over the entire OTAG domain (all-domain). The same analysis was carried out for the 11 subdomains shown in Fig. 1 and for the 155 receptor sites. The 95% ranges on the CDFs of the differences in predicted concentrations for the four emissions scenarios vary from site to site, sometimes overlapping zero and sometimes not. For example, consider the difference in 1995 base emissions and 2007 base emissions (row 1 of Table 3 gives the results for the "all-domain" maximum ozone concentration). Approximately one-third of the 155 sites have the 95% range overlap zero, implying that the difference is probably due to random variability at those sites. At 11 of the sites, all in the Southeast, the median difference is greater than zero (i.e., the concentrations are *higher* in 2007, after emissions controls are implemented, than in 1995).

The 95% ranges on the differences were also studied at the 155 sites for the 50% NO_x and the 50% VOC emissions reductions. The 95% range does not overlap zero for about 80% of the 155 sites for the 50% NO_x emission reductions. However, the differences appear to be caused solely by random variability at many sites in the Northeast, implying that the NO_x emissions reductions may be more effective in the Southeast than in the Northeast. Five sites (in the Chicago, Baltimore-Washington, Philadelphia, and New York domains) have a median difference greater than zero, meaning that the median predicted maximum ozone concentration in these areas actually increased with a 50% NO_x emissions reduction.

The differences for 50% VOC emissions reductions suggest that any ozone concentration changes are solely due to random variability. About 75% of the sites have the 95% range overlap zero, and 10% of the sites (most in the Southeast) have the median difference greater than zero. In the latter cases, the median predicted maximum ozone concentration *increased* with a 50% VOC reduction. The VOC emissions reductions appear to be slightly more effective in the Northeast than in the Southeast, although the changes are generally low everywhere on the OTAG domain.

5.4. Differences in uncertainty estimates for 1-hr and 8-hr averages

The conclusions (based solely on UAM-V model predictions) concerning Monte Carlo estimates of uncertainties in the predictions of maximum ozone concentrations are little changed for 1- versus 8-hr averages. The main difference is that the 8-hr average predicted maximum daily ozone concentrations are about 10% lower than the 1-hr averages. This 10% relation for photochemical grid model predictions was also suggested by Chock et al. (1999). The EPA has calculated this relation for observations at their ozone-monitoring sites and found that, on average, the 8-h averages are observed to be about 15% less than the 1-h averages for maximum daily ozone concentrations (see http://envpro.ncsc.org/OMS/pub/SiteInfo/Correlation-Report.txt). This agreement between the 10% figure for model predictions and the 15% figure for observations is fairly good, considering that the observations are taken at single points and are therefore expected to have more variability than the model predictions, which represent ensemble averages over grid volumes.

6. Input variables whose uncertainties have the largest effect on uncertainties in predicted ozone

6.1. Correlation analysis

It was stated earlier that the observed and predicted maximum hourly averaged ozone concentrations usually occurred in the Atlanta UAM domain. More precisely, the maximum occurred in northern Alabama, to the west of Atlanta. This is not a region subject to long-range transport to the same extent as the New York UAM domain. In northern Alabama, there are relatively few large source regions in the upwind direction (i.e., to the southwest), while in New York, there are many large source regions in the upwind direction (e.g., New Jersey, Philadelphia, Baltimore, and Washington). It is therefore expected that some of the correlation coefficients and regression results between variations in predicted maximum ozone concentrations and variations in input parameters may be different for the current phase of the research than for the earlier study (Hanna et al., 1998).

The results of the latest set of Monte Carlo runs suggest that the NO_2 photolysis rate is the variable whose uncertainties are most strongly correlated to the uncertainties in predictions of maximum hourly averaged ozone concentrations for the 12–14 July period. At over 90% of the 155 receptor sites, variations in NO_2 photolysis rate show a correlation of about $+0.6$ with variations in predicted maximum hourly averaged ozone concentration. It is interesting that, in the entire set of over 90 CB-IV photochemical reactions, this is the only process that *directly* produces ozone, via the production of atomic oxygen from NO_2 through the reaction ($NO_2 \rightarrow h\nu \rightarrow NO + O$), and the subsequent production of ozone through the reaction ($O + O_2 + (M) \rightarrow O_3$). The analysis of the Monte Carlo runs also shows significant correlations (i.e., correlations with magnitudes exceeding 0.19) between variations in predicted maximum hourly averaged ozone concentrations and variations in wind speed and direction, relative humidity, cloud cover,

and biogenic VOC emissions. These results could allow future research to be focussed on better specifying the input variables that have the strongest influence on the ozone predictions.

Hanna et al. (1998) reported that, for the New York City UAM domain, variations in the rate constants for some chemical reactions were found to produce significant correlations with variations in maximum hourly averaged ozone concentrations predicted by UAM-IV. It was not obvious why these particular reactions were more important than others. In the current study involving UAM-V on the OTAG domain, some of the same reactions were found to be important. In particular, significant correlations are found with the following CB-IV reactions:

$$\text{CB-IV Reaction 46: } C_2O_3 + NO \rightarrow NO_2 + XO_2 + FORM + HO_2.$$

$$\text{CB-IV Reaction 50: } C_2O_3 + HO_2 \rightarrow 0.79\,FORM + 0.79\,XO_2 + 0.79\,HO_2 + 0.79\,OH.$$

Further inspection suggests that these two reactions are especially important for ozone production not only because the first reaction produces NO_2, but also because both reactions lead to a net increase in peroxyl radicals that can subsequently produce NO_2 by reaction with NO, as well as formaldehyde, a photochemical precursor of additional radicals that can lead to NO_2 production. Thus these reactions are radical amplifiers.

Hanna et al. (1998) found strong correlations between variations in anthropogenic area source emissions of VOC and variations in predicted maximum ozone concentration in the New York City UAM domain. But the current Monte Carlo study does not yield such high correlations. We postulate two reasons for this. The first reason is that the New York City domain studied earlier was largely VOC-limited, such that substantial ozone responses were simulated for VOC emissions changes. On the other hand, the rural-dominated OTAG domain used in the current Monte Carlo study was largely NO_x-limited, resulting in negligible simulated ozone responses to VOC emissions reductions over most of the domain. The second postulated reason is that, for input to the UAM-V model used in the current Monte Carlo study, the anthropogenic area sources of VOC are separated into three components: mobile, low point, and other area. The Monte Carlo simple random sampling method picks values of these components separately and independently. Because there is no correlation prescribed between these components, the positive and negative perturbations of the chosen random numbers (one for mobile sources, one for low-point sources, and one for other area sources) tend to cancel out, leading to a reduced effect of total VOC anthropogenic area sources on variations in predicted ozone concentration. In contrast, the anthropogenic VOC emissions inputs to UAM-IV in the earlier study consisted of a single relatively large anthropogenic area source term, whose variations therefore had a stronger effect on variations in predicted ozone. This finding suggests that future studies with inputs that are known to be correlated might consider including estimates of these correlations in the random sampling procedure.

The earlier results, on the relatively small New York City domain, suggested that uncertainties in boundary concentrations were correlated to uncertainties in predicted maximum ozone. This was not found in the current study, probably because of the large differences in domain sizes. In the earlier study, there were large amounts of ozone, VOCs and NO_x on the upwind domain boundary in central New Jersey. In contrast, for the relatively large OTAG domain used in the current study, the domain boundaries were in rural areas far upwind of the region where high ozone and precursor concentrations were occurring.

6.2. Regression analysis

Standard multiple linear regression analysis was applied to the inputs and outputs of the 100 Monte Carlo runs for each emission group. In a sense, this procedure attempts to replace the original UAM-V three-dimensional grid model with a response surface based on multiple linear regression. This analysis was done for the input variables whose variations showed a correlation exceeding 0.19 with the variations of the peak domain-wide hourly averaged ozone concentration. The criterion; 0.19, represents the magnitude of the correlation coefficient where, for 100 pairs of independent variables, there is 95% confidence that the calculated correlation is significantly different than zero. This analysis was done for the OTAG-domain-wide maximum ozone concentration, for the maximum in the 11 smaller UAM domains, and for 1 and 8-h averages. The ozone maximum could occur anytime in the 12–14 July 1995 period. The results were fairly consistent, as shown by the regression formulas listed below for the OTAG-domain 1-h maximum ozone concentration for the four base emissions scenarios. The numbers in parentheses (e.g., $R^2 = 0.74$) indicate the fraction of the variance explained by the regression formula. The input variables are expressed as fractions or relative changes (i.e., change in the variable divided by the value of the variable). Shorthand notation is used for the key variables (refer to Table 1): u = wind speed (variable 12); CC = cloud cover (variable 18); VOCBIO = area biogenic VOC emissions (variable 21); NO_2phot = NO_2 photolysis rate; R1 = CB-IV reaction 1 ($NO_2 \rightarrow h\nu \rightarrow NO + O$); R15 = CB-IV reaction 15 ($NO_3 + NO \rightarrow 2NO_2$); R41 = CB-IV reaction 41 ($FORM + NO_3 \rightarrow HNO_3 + HO_2 + CO$); and R46 = CB-IV reaction 46 ($C_2O_3 + NO \rightarrow FORM + NO_2 + HO_2 + XO_2$).

For year 1995 emissions ($R^2 = 0.74$):

$$O_3(\text{ppm}) = 0.088\,\text{ppm} - 0.093(\Delta u/u) - 0.029(\Delta CC/CC) + 0.051(\Delta VOCBIO/VOCBIO) + 0.098(\Delta NO_2\,phot/NO_2\,phot) + 0.023(\Delta R46/R46)$$

For year 2007 emissions ($R^2 = 0.71$):

$$O_3(\text{ppm}) = 0.095\,\text{ppm} - 0.069(\Delta u/u) - 0.023(\Delta CC/CC) + 0.044(\Delta VOCBIO/VOCBIO) + 0.088(\Delta NO_2\,phot/NO_2\,phot) + 0.074(\Delta R1/R1) - 0.025(\Delta R41/R41)$$

For year 2007 emissions with 50% cuts in NO_x ($R^2 = 0.72$):

$$O_3(\text{ppm}) = 0.023\,\text{ppm} - 0.064(\Delta u/u) + 0.023(\Delta VOCBIO/VOCBIO) + 0.067(\Delta NO_2\,phot/NO_2\,phot) + 0.043(\Delta R1/R1) + 0.020(\Delta R15/R15) + 0.020(\Delta R41/R41)$$

For year 2007 emissions with 50% cuts in VOC ($R^2 = 0.72$):

$$O_3(\text{ppm}) = 0.061\,\text{ppm} - 0.066(\Delta u/u) - 0.022(\Delta CC/CC) + 0.052(\Delta VOCBIO/VOCBIO) + 0.081(\Delta NO_2\,phot/NO_2\,phot) + 0.069(\Delta R1/R1)$$

Recall that the prediction of median domain-wide daily maximum hourly-averaged ozone concentration is about 0.20 ppm. The coefficient of the relative NO_2 photolysis rate change term, $\Delta NO_2\,phot/NO_2\,phot$, is in the range from 0.067 to 0.098 in the four equations above, implying that a 50% uncertainty in NO_2 photolysis rate causes about a 0.04 ppm, or a 20% uncertainty in predicted maximum ozone concentration. The coefficient of the relative wind speed change term, $\Delta u/u$, has a similar range from 0.064 to 0.093 in the four equations, also implying that a 50% uncertainty in wind speed causes about a 0.04 ppm, or a 20% uncertainty, in predicted ozone concentration. The reason why the gross results of the correlation analysis, discussed in Section 6.1, showed that the uncertainties in NO_2 photolysis rates have a larger effect than uncertainties in wind speed on uncertainties in predicted maximum ozone concentrations is that the assumed total uncertainty is about 1.7 times as large for the photolysis rate ($\sigma_{\ln\,\text{phot rate}} = 0.347$) as for the wind speed ($\sigma_{\ln u} = 0.203$).

It is seen that cloud cover, CC, has a consistent regression constant of about 0.02–0.03 in the equations. Thus an uncertainty of 50% in cloud cover causes an uncertainty of about 0.01 ppm, or about 5%, in predicted maximum ozone. The area biogenic VOC emissions, VOCBIO, have a regression coefficient ranging from 0.023 to 0.052, with the smaller number for the runs with reduced NO_x emissions. For most of the cases, a 50% uncertainty in VOC biogenic emissions causes about a 10% uncertainty in predicted maximum ozone.

Four chemical reactions appear in the four regression equations, with reaction 1 ($NO_2 \rightarrow NO + O$), or variable 34, appearing three out of four times. This reaction "teams" with the NO_2 photolysis rate and is interesting because this (along with reaction 2: $O + O_2 + (M) \rightarrow O_3$) is the only mechanism for direct formation of ozone. The regression constant for reaction 1 averages about 0.06, implying that a 50% uncertainty in the rate constant for reaction 1 leads to about a 15% uncertainty in predicted ozone concentrations. It follows that a priority should be given to more accurate determination of the NO_2 photolysis rate and the reaction rate for reaction 1.

7. Limitations to study and recommendations for further research

The Monte Carlo uncertainty methodology is a powerful technique that is seeing increasing applications to large three-dimensional environmental models. The earlier study (Hanna et al., 1998) and the current study are meant to be preliminary demonstration exercises of the Monte Carlo methodology as applied to three-dimensional photochemical grid models. The results of these applications appear realistic and much experience has been gained in the methodology, which has been revised considerably over the course of the research. It would be appropriate for decision-makers to use this information on uncertainties as part of the process by which emissions reductions are prescribed. Nevertheless, there are a few limitations that should be addressed:

Dependence on satisfactory model – The methodology assumes that the model is satisfactory to begin with and is able to account properly for the major physical and chemical effects. The uncertainty analysis looks only at the influence of uncertainties in model input parameters and variables on the uncertainties of model output variables. The methodology does not account for deficiencies in model formulations, which may lead to compensating errors and other effects that mask the true variabilities and dependencies. For these reasons, two different photochemical grid models may give different results when the Monte Carlo methodology is applied to them.

Dependence on good knowledge of input variable uncertainty – Our expert elicitation exercise suggests that most

photochemical modelers have not devoted extensive effort to estimating input variable uncertainties. Consequently the uncertainties assumed in Table 1 represent preliminary estimates which should be refined as this research field matures. The best information exists for the uncertainties of the photochemical reaction rate constants, which have been studied for other purposes, such as modeling the photochemistry of the stratosphere. Also, there has not yet been an acceptable way devised to estimate the variability in meteorological inputs, because of difficulties associated with the need to maintain mass conservation in the wind fields. The best way to treat meteorological uncertainty would be to apply the Monte Carlo method to a diagnostic or prognostic meteorological model, beginning with uncertainties in the basic inputs such as the surface wind and radiosonde inputs, but this would be an extensive effort all by itself. Other difficulties exist in the estimation of uncertainties in boundary and initial conditions because there are not sufficient observations of vertical and spatial fields of the chemical constituents. Finally, the literature contains a long trail of discussions about possible biases in the baseline emissions inputs, with recent improvements to some components such as biogenic and mobile source VOCs. Nevertheless, few estimates of uncertainties in emissions are available.

Categorization of emissions classes – It was mentioned in Section 6 that the influence of variations in anthropogenic VOC area source emissions on uncertainties in predicted ozone concentration was decreased in the current study when compared with the previous study (Hanna et al., 1998). This is because the UAM-V model used in the current study has three categories of anthropogenic area source emissions, whereas the UAM-IV model used in the previous study lumps the three categories into a single class. By randomly and independently varying these three categories, the total variation in combined emissions tends to be less because of cancellations of positive and negative perturbations of the components. In reality, the components are correlated. If emissions classes are subdivided, it may be useful to estimate the magnitudes of these correlations and account for them in the resampling method. This is also true of correlations between emissions and other input variables such as boundary VOC concentrations.

Long-range transport issues – Because of the use of the words "ozone transport" in the name OTAG, there is an interest in determining the uncertainty of ozone concentrations due to variations in emissions inputs at locations several hundred kilometers upwind of where the ozone concentration is being observed. In the current study, it is difficult to address long-range transport issues because all emissions in each category are varied uniformly across the OTAG domain. To study transport issues, it would be of interest to look at variations of emissions in certain source groups, such as Midwest power plants, and determine their influence on uncertainties in predicted ozone concentrations in key downwind areas, such as northern New York or New England.

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