Presented at the 11th Annual CMAS Conference, Chapel Hill, NC, October 15-17, 2012

OBSERVATION-CONSTRAINED PROBABILISTIC EVALUATION OF MODELED CONCENTRATIONS AND SENSITIVITIES

Daniel S. Cohan* and Antara Digar
Department of Civil and Environmental Engineering, Rice University, Houston, TX, USA

1. INTRODUCTION

Photochemical models are typically applied in a deterministic manner to generate the best possible estimates of pollutant concentrations and their sensitivities to emissions. Although model inputs and outputs are known to be uncertain, explicit quantification of those uncertainties in regulatory practice is rare. The state of the science for probabilistic and other evaluation of air quality models is reviewed by Dennis et al. (2010). Characterization of uncertainties is reviewed by Fine et al. (2003).

Recent development of techniques such as reduced form models have provided efficient methods for Monte Carlo characterization of the uncertainties in model outputs arising from parametric and structural uncertainties in model inputs and formulations. However, applications of these techniques have typically considered each of the Monte Carlo cases to be equally likely. This neglects the fact that different cases may have starkly different performance relative to observations.

Our conference presentation and associated paper (Digar et al., submitted 2012) show how probabilistic evaluations of photochemical model results can be constrained by observations of pollutant mixing ratios. The methods are applied to a historical attainment modeling episode for ozone in Texas, with numerous structural and parametric uncertainties considered in the CAMx model. Three observational metrics including a Bayesian likelihood approach are introduced to weight or screen thousands of alternate simulations generated by a reduced form model. The metrics evaluate model performance against ground-based observations of ozone and nitrogen oxides (NO_x), but could readily be extended to consider other pollutants or observations aloft.

The approach yields observation-constrained probability distributions not only for pollutant concentrations, but also for the responsiveness of those concentrations to perturbations in emission rates. We also generate posterior probability distributions for the true values of uncertain model inputs such as emission inventories and reaction rate constants. As documented by Digar et al. (submitted 2012), results vary with the observational metric used, highlighting the importance of metric selection. Implications for probabilistic evaluation of regulatory and scientific modeling of historical air pollution episodes are discussed.

**Corresponding author:* Daniel S. Cohan, Department of Civil and Environmental Engineering, Rice University, 6100 Main Street MS 519, Houston, TX 77005; e-mail: cohan@rice.edu.

2. BASE CASE MODELING

Modeling was conducted with the Comprehensive Air Quality Model with Extensions (CAMx) v5.32. To mimic actual regulatory modeling, base case simulations were based upon episodes developed by the Texas Commission on Environmental Quality (TCEQ) for the development of its State Implementation Plan (SIP) for attaining the 1997 (84 ppb) 8-hour ozone standard in the Dallas-Fort Worth region. We focused upon a one-month period, May 31 – July 2, 2006, that included numerous episodes of high ozone concentrations within the DFW region.

3. UNCERTAINTY SCENARIOS

A broad ensemble of uncertainty scenarios was developed by considering both parametric and structural uncertainties in the base case CAMx modeling. Parametric uncertainties considered fractional perturbations to specific emission rates, reaction rate constants, and boundary conditions that had been shown by previous literature to significantly influence ozone concentrations and/or their sensitivities to precursor emissions (Digar et al., 2011).

Given resource limitations, the choice of structural uncertainty scenarios depended in part upon the availability of alternate inputs and formulations. These cases included alternate models for generating the boundary conditions and biogenic emissions that are input to CAMx, and an alternate approach within CAMx for representing vertical diffusion. Screening was conducted to identify the structural choices that most influenced ozone concentrations in the DFW region and their sensitivities to precursor emission controls.

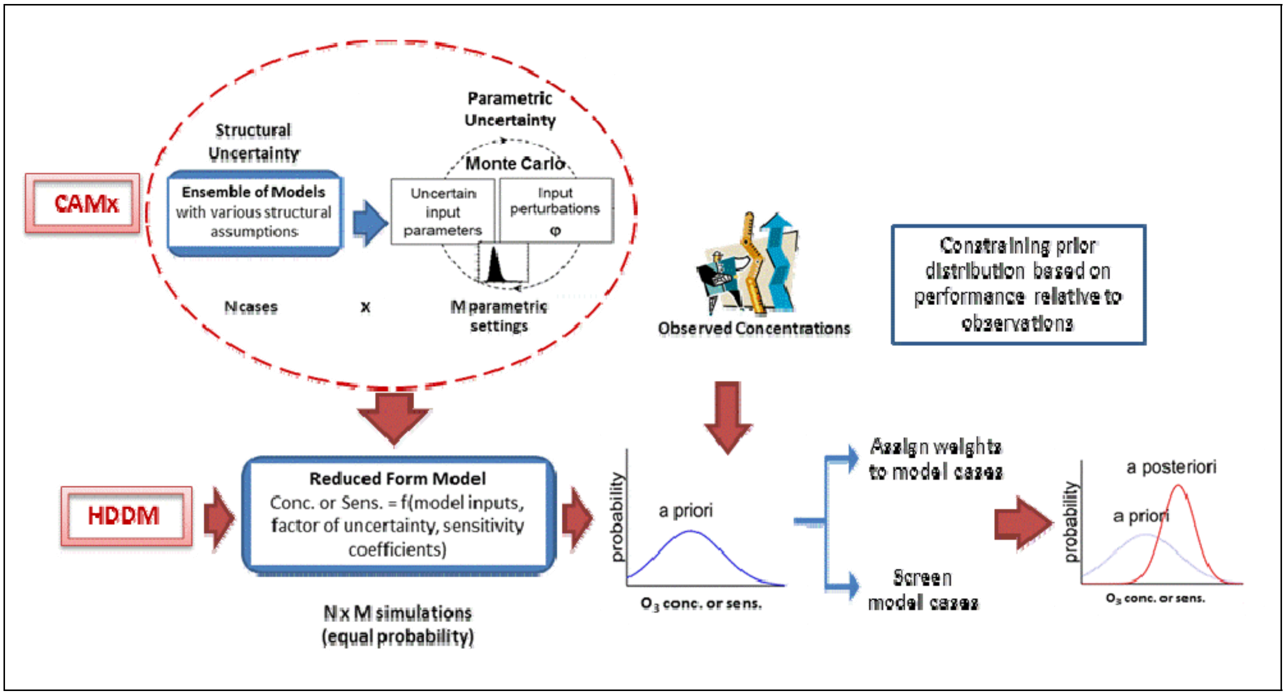

Fig 1. Flowchart illustrating the methds applied for observation-constrained uncertainty analysis.

4. MONTE CARLO SIMULATIONS

Structural scenarios were simulated directly using the corresponding alternate inputs and formulations. A large number of parametric scenarios was generated within each structural scenario via Monte Carlo sampling of the uncertain input parameters (Fig 1). Ozone mixing ratios and their sensitivities to precursor emissions under each scenario were computed based on the reduced form modeling approach of Digar and Cohan (2010) and Tian et al. (2010), which uses first- and second-order sensitivity coefficients generated by the high-order decoupled direct method. Digar and Cohan (2010) demonstrated the reliability of the reduced form model for representing concentration and sensitivity relationships within the underlying photochemical model.

In previous applications of the reduced form model, only parametric uncertainties have been considered and each of the Monte Carlo cases has been assumed to be equally likely, since input parameters were sampled from their presumed probability distributions. The current application couples the parametric uncertainties with structural uncertainty scenarios, and weights or screens the outputs based on performance relative to observations.

5. OBSERVATION-BASED CONSTRAINTS

Model results were evaluated against observations of ozone and NO_x from 11 state regulatory monitors that measured both pollutants. Hourly data were taken from the US EPA's Air Quality System database and processed to obtain 8-hour daily maximum ozone and 24-hour average NO_x for the evaluation. A bias-correction factor was adapted from Lamsal et al. (2008) to address interferences with the NO_x measurements.

Three metrics were applied to weight or screen the model cases, as detailed by Digar et al. (submitted 2012). The first metric applied Bayesian weightings based jointly on model performance in simulating ozone and NO_x at the 11 monitors. The second metric screened the model cases, accepting only those that met US EPA's three recommended criteria for acceptable ozone SIP simulations (US EPA, 2007). The third metric applied a statistical nonparametric significance test using the Cramer-von-Mises criterion (Anderson, 1962) to test whether the modeled distributions of ozone and NO_x

concentrations were consistent with the observations.

Full discussion of the results of these three metrics is provided by Digar et al. (submitted 2012), and will be summarized in the conference presentation (also illustrated in Fig 1).

6. ACKNOWLEDGMENTS

This work was supported by the State of Texas through the Air Quality Research Program administered by The University of Texas at Austin by means of a grant from the Texas Commission on Environmental Quality (TCEQ), and by National Science Foundation Grant #087386. The baseline modeling for the study was provided by TCEQ.

7. REFERENCES

Anderson, T.W. (1962). On the distribution of the two-sample Cramer-von Mises criterion. *The Annals of Mathematical Statistics*, 33, 1148-1159.

Dennis, R., T. Fox, M. Fuentes, A. Gilliland, S. Hanna, C. Hogrefe, J. Irwin, S.T. Rao, R. Scheffe, K. Schere, D. Steyn, and A. Venkatram (2010). A framework for evaluating regional-scale numerical photochemical modeling systems. *Environmental Fluid Mechanics* 10, 471-489, doi:10.1007/s10652-009-9163-2.

Digar, A., D.S. Cohan, X. Xiao, K.M. Foley, B. Koo, and G. Yarwood. (submitted 2012) Constraining ozone-precursor responsiveness using ambient measurements. In revision, *Journal of Geophysical Research*.

Digar, A., and D.S. Cohan (2010), Efficient characterization of pollutant-emission response under parametric uncertainty, Environmental Science & Technology, 44(17), 6724-6730.

Digar, A., D. S. Cohan, D. D. Cox, B. Kim, and J. W. Boylan (2011), Likelihood of achieving air quality targets under model uncertainties, Environmental Science & Technology, 45(1), 189-196.

Fine, J., L. Vuilleumier, S. Reynolds, P. Roth and N. Brown (2003). "Evaluating uncertainties in regional photochemical air quality modeling." *Annual Review of Environment and Resources* 28: 59-106.

Lamsal, L. N., R. V. Martin, A. von Donkelaar, M. Steinbacher, E. A. Celarier, E. Bucsela, E. J. Dunlea, and J. P. Pinto (2008), Ground-level nitrogen dioxide concentrations inferred from the satellite-borne Ozone Monitoring Instrument, *Journal of Geophysical Research*, 113(D16), D16308.

Tian, D., M.S. Bergin, D.S. Cohan, S. Napelenok, Y. Hu, M. Chang, A.G. Russell (2010). Uncertainty analysis of ozone formation and response to emission controls using high-order sensitivities. *Journal of the Air & Waste Management Association*, 60, 797-804.

US EPA (2007). Guidance on the use of models and other analyses for demonstrating attainment of air quality goals for Ozone, PM2.5, and Regional Haze.