APPLICABILITY OF CMAQ-DDM TO SOURCE APPORTIONMENT AND CONTROL STRATEGY DEVELOPMENT

Daniel S. Cohan *
Georgia Institute of Technology, Atlanta, GA
e-mail: **dcohan@eas.gatech.edu**
Voice (404) 385-4565 Fax (404) 894-8266

1. INTRODUCTION

Source apportionment and control strategy development represent two key applications of photochemical models to air pollution policy. Source apportionment quantifies the contribution of each emission source to pollutant concentrations in order to identify regions, categories, and facilities most responsible for pollution levels. In control strategy development, the question of interest is how pollutant concentrations would respond to abatement measures which reduce emission rates at one or more sources.

Policy makers and scientists have often turned to photochemical models to conduct source apportionment and to estimate the impacts of potential control strategies. However, as air pollution policy increasingly focuses on secondary pollutants such as ozone and fine particulate matter, source attribution and sensitivity analysis are complicated by the nonlinear and interdependent impacts of various emitters. For example, the impact of a multi-part control strategy may differ from the sum of its parts because the implementation of each component alters the sensitivity of concentrations to other component measures (Cohan et al., 2004).

With the recent implementation of the high-order Decoupled Direct Method in 3D (HDDM-3D) in the Community Multiscale Air Quality Model (CMAQ) for gas-phase processes (Cohan, 2004), it is an appropriate time to examine the applicability of this sensitivity analysis technique in policy contexts. This conference paper discusses HDDM-3D and the more traditional brute force method and suggests how they may be appropriately applied to source apportionment and control strategy development. Potential approaches to addressing nonlinearity are discussed. Rather than simply being an either-or choice between the methods, it is shown that HDDM-3D and brute force can be applied in complementary roles to inform policy considerations.

* *Corresponding author address:* Daniel Cohan, School of Earth & Atmospheric Sciences, Georgia Institute of Technology, 311 Ferst Drive, Atlanta, GA 30332

2.0 SENSITIVITY ANALYSIS METHODS

2.1 Brute Force

Traditionally, source apportionment and control strategy development have been addressed in air quality models by a “brute force” method. Modeled concentrations are compared across numerous air quality simulations that are identical except for perturbations to one or more emission rates. The contribution of an emission source is typically computed as the decline in pollutant concentrations when that source is removed from the model emissions inventory. The impact of a control measure is often estimated by modeling how concentrations would change as an emission rate is reduced by a fractional amount.

Brute force is attractive for its simplicity and ease of implementation in a wide variety of models, but becomes cumbersome when a large number of perturbations must be considered. This becomes especially problematic for secondary pollutants such as ozone and particulate matter, which exhibit nonlinear responses to the emissions of multiple precursor species. Because of the nonlinear and interacting impacts of emissions, it is not immediately clear whether brute force results for one level of perturbation may be accurately scaled to other perturbations.

2.2 Decoupled Direct Method

The Decoupled Direct Method in Three Dimensions (DDM-3D) (Yang et al., 1997) offers a computationally-efficient alternative for sensitivity analysis. DDM-3D computes the sensitivities of concentrations to perturbations in emission rates and other model inputs and parameters, using the same equations that compute concentrations in the underlying model. The recent extension of DDM-3D to higher-order coefficients (HDDM-3D, Hakami et al., 2003) enables characterization of the nonlinearity of response to emission perturbations.

HDDM-3D has recently been implemented in the Community Multiscale Air Quality Model for gas-phase processes and has been shown to accurately compute first- and second-order sensitivity coefficients of concentration-emission response (Cohan, 2004). The first-order coefficient represents the local slope of response as emissions are changed from unperturbed ("base case") values (Figure 1). The second-order coefficient represents the local curvature, i.e., the rate at which responsiveness (slope) changes as emissions change.

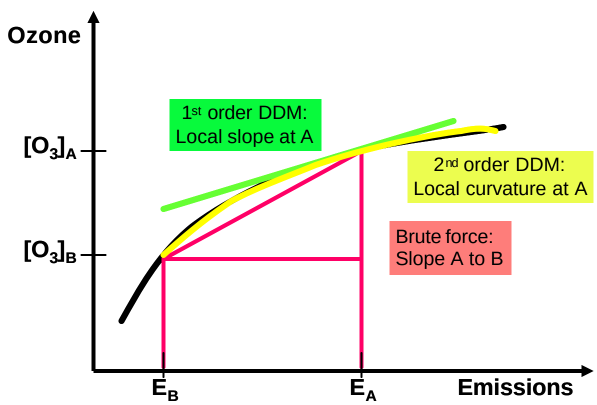

Figure 1. Brute force and HDDM-3D sensitivity analysis of ozone response to emissions. Given the typically concave-down response, the brute force slope of response to large reductions in emissions is steeper than the local first-order sensitivity at Point A.

2.3 Taylor Expansions

HDDM-3D sensitivity coefficients represent responsiveness to *infinitesimal* changes in emissions. For control strategy development, we are interested in how pollutant concentrations would respond to *finite* reductions in emissions resulting from abatement measures. As demonstrated by Hakami et al. (2003), large-scale response can be represented by Taylor expansions of sensitivity coefficients:

$$\mathbf{C}_j\Big|_{p_j=P_j+\Delta e_j P_j} \approx \mathbf{C}_0\Big|_{p_j=P_j} + \Delta e_j \mathbf{S}_j^{(1)} + \tfrac{1}{2}\Delta e_j^2 \mathbf{S}_{j,j}^{(2)} + \textit{higher order terms} \quad (1)$$

where $\mathbf{S}^{(1)}$ and $\mathbf{S}^{(2)}$ are semi-normalized first- and second-order sensitivity coefficients with respect to emission rate p_j, and $\mathbf{C}_j$ are the concentrations when p_j has been perturbed by an amount ?$e_j P_j$. P_j is the unperturbed value of p_j. Note that the second-order term scales with ?e_j^2, and thus its relative importance increases with the size of the perturbation.

Figure 2 schematically shows the role of first- and second-order HDDM-3D sensitivity coefficients in control strategy analysis and source apportionment. If a pollutant exhibits a nonlinear concave-down response to an emission rate, then linear scaling of first-order coefficients (green line in Figure 2) would underpredict the impact of a control measure or the contribution of a source. The larger the fractional perturbation of interest, the greater the susceptibility of a first-order estimate to underpredicting the response (source apportionment is equivalent to modeling 100% removal of a source). Incorporating second-order sensitivities via a Taylor expansion is intended to account for nonlinearity, but the possibility remains that higher-order terms and discontinuities could cause some inaccuracy to remain.

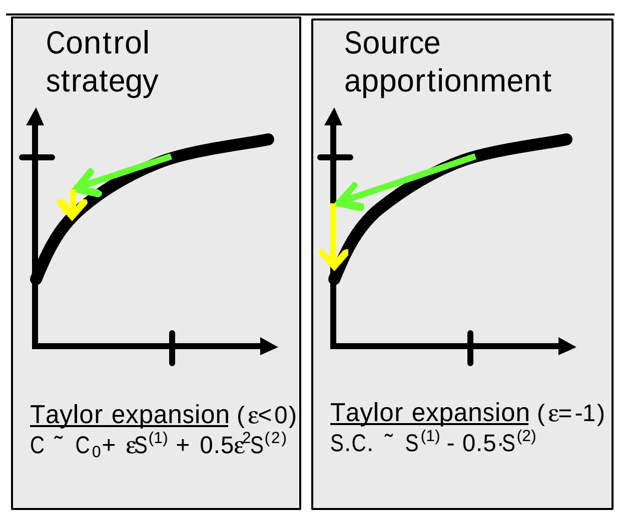

Figure 2. The impact of a control strategy can be approximated by a Taylor expansion of the first- (green) and second-order (yellow) HDDM-3D sensitivity coefficients. Source apportionment is equivalent to the impact of completely removing an emission source. In the schematic, the x-axis is emissions and the y-axis is concentrations.

3.0 ACCURACY AND NONLINEARITY

Given the impossibility of conducting perfectly controlled atmospheric experiments of concentration-emission response in which meteorological conditions are held constant while emission rates change, sensitivity analysis methods can be evaluated only in terms of their ability to capture the responsiveness of the underlying model. Thus HDDM-3D is evaluated by comparison with response indicated by brute force simulations.

3.1 Accuracy

Cohan (2004) rigorously evaluated the accuracy of CMAQ-HDDM-3D for simulating the response of ozone concentrations to various fractional perturbations in both individual sources and domain-wide emissions of NO_x and VOC by comparing Taylor expansions of HDDM-3D coefficients with brute force results. All local HDDM-3D sensitivity coefficients were found to be highly accurate. First-order coefficients were shown to be sufficient for capturing +/- 10% perturbations in emission rates. Incorporation of second-order coefficients greatly improved the accuracy of estimates of response to 50% and 100% emission reductions. For 100% reduction, the most nonlinearity-prone case, second-order Taylor expansions underpredict brute force response by an average of 5-10%, several times smaller than the underprediction of first-order alone. Despite low overall bias, more severe underprediction can occur in localized areas along the boundary between NO_x-limited and VOC-limited regimes where ozone chemistry is especially nonlinear (Cohan et al., 2004).

3.2 Cross-sensitivity

In addition to the nonlinearity of atmospheric response to various perturbations in a single emission rate, cross-sensitivities complicate the consideration of an ensemble of emission sources. Due to cross-sensitivity, the impact of one emission source depends in part on the emission rate of another source. For example, the installation of a control device to reduce NO_x emissions from a power plant would reduce ambient ratios of NO_x/VOC and cause ozone production to become more NO_x-limited. This would heighten sensitivity to other NO_x sources, but reduce sensitivity to VOC sources.

Cohan et al. (2004) demonstrated how cross-sensitivities can affect source apportionment of an ensemble of sources. The total contribution of all sources, whether computed by brute force or HDDM-3D, includes interactions between the impacts of those sources. However, if the contribution of those sources is computed separately (again, by either method) then simply summing up the individual contributions would neglect the interactions. In a case study of the response of Atlanta ozone to various categories of Atlanta NO_x emissions, it was found that the total contribution of all Atlanta NO_x would be underestimated by more than 10% if computed by simply adding up the individual contributions of each category. Similarly, the impact of a multi-part control strategy would differ from the sum of the impacts of the individual components because each measure may affect the impact of the other measures.

4.0 RECOMMENDATIONS

When atmospheric response to emissions is nearly linear, then either brute force or DDM first-order sensitivities can readily be scaled to predict the impact of various perturbations. This section suggests how source apportionment and control strategy analysis could sensibly be conducted for pollutants whose responsiveness is nonlinear and thus subject to cross-sensitivity interactions among emission sources.

4.1 Control strategy assessment

The computational efficiency of HDDM-3D and its ability to avert numerical noise make it well suited for scoping the impact of a large number of control measures. Sensitivity coefficients with respect to a large number of sources can be computed within a single CMAQ-HDDM-3D simulation.

If a control measure represents a small fraction of total emissions, then nonlinearity may be muted and first-order sensitivity coefficients may be sufficient to estimate the impact of controls. The larger the fractional reduction in emissions that is being considered, the greater is the importance of considering second-order terms via Taylor expansions. The ability of HDDM-3D Taylor expansions to sketch out the responsiveness of concentrations to various changes in emissions (Figure 2) is especially useful if multiple percentages of control are possible at a single source.

Although HDDM-3D is a powerful scoping tool for considering a large number of potential controls without conducting an inordinate number of brute force simulations, the potential presence of cross-sensitivities means that the impact of an ensemble of those measures will not necessarily equal the sum of the impacts of the individual measures. For applications such as the creation of State Implementation Plans (SIPs) for ozone attainment (U.S. EPA, 1999), it must be demonstrated that the total effect of the proposed control measures is sufficient to attain air quality standards. It would be impractical to model all of the cross-sensitivity interactions among a large number of control measures.

Thus, regardless whether HDDM-3D or brute force is used to inform the selection of individual control measures, the final demonstration should be done by a brute force run which evaluates model response to the entire ensemble of controls. This allows all nonlinearities and cross-sensitivities to be taken into account in the final analysis. An iterative approach could be used if the brute force assessment indicates that further emission controls are necessary.

4.2 Source apportionment

Source apportionment is more subject to nonlinearity than control strategy analysis, because it represents the impact of completely removing a source. Thus linear scaling of first-order sensitivity coefficients is prone to significant inaccuracies in estimating source contribution. Taylor expansions of first- and second-order HDDM-3D coefficients may accurately approximate source contribution in most instances. However, the need to compute both first- and second-order coefficients in HDDM-3D means that brute force may actually be more computationally efficient. Further, the brute force method is less prone to numerical noise when computing source contribution, because source apportionment considers the impact of the entire source rather than a small perturbation to that source.

Due to cross-sensitivities, the contribution of an ensemble of sources may differ from the sum of its parts. Modeling should be tailored to address the question of interest, and it should not be assumed that source contributions can be treated as additive. HDDM-3D can be used to compute cross-sensitivities among various emission sources and therefore understand how their impacts interact.

5. REFERENCES

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