

Interactive comment on “A method to represent ozone response to large changes in precursor emissions using high-order sensitivity analysis in photochemical models” by G. Yarwood et al.

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General comments: The use of Taylor expansions of high-order decoupled direct method (HDDM) sensitivity coefficients to characterize ozone response to emission changes has been the focus of several recent studies, and is increasingly being considered for policy applications. Recent work by Simon et al introduced methods for enhancing the accuracy of HDDM-based predictions by calculating coefficients at a range of emission levels. The research presented here, apparently undertaken before final publication of Simon et al, introduces similar approaches for using multiple sets of sensitivity coefficients, though with important methodological distinctions from the

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Simon approach. This paper provides a valuable contribution by rigorously testing the performance of its HDDM-based methods and highlighting how performance can vary under a vast range of conditions (winter vs summer; urban vs rural; full range of NO_x and VOC from 0 to 100%). Most previous studies focused on summer, so this paper is useful in highlighting additional challenges in winter. Further work comparing the performance of the Yarwood and Simon approaches and further refining or blending their methods may help catalyze the use of HDDM in policy applications.

The paper is concise, clearly written, and largely free of errors. It merits publication to further the ongoing discussion about best practices for applying sensitivity methods in air quality policy development.

Specific comments: 1. A potential limitation of this approach is its use of concentrations and sensitivities only from runs with drastically reduced emissions. While this saves some computational time and helps target the low emissions cases where errors would otherwise be severe, it likely hinders accuracy near the base case by not using concentrations and sensitivities computed there. Response of ozone to modest reductions in emissions is often the most critical issue to address in policy applications such as attainment demonstrations. The authors note the potential to improve accuracy for such cases by using coefficients from the base case (p. 2593 line 29 – p. 2594 line 2), but did not test that. This highlights that further refinement and evaluation of both the Simon and Yarwood methods will be a valuable next step, as the authors note at the end of Section 4. In the meantime, adopters of these methods are likely to be well served by the authors' recommendation (p. 2595, lines 20-21) to compute sensitivities at the midpoint of the range of interest, and adapt Eq. 3 accordingly. This may help lessen errors near the base case when modest emission reductions are the focus.

2. While results are presented in terms of bias and error relative to brute force concentrations, was any testing done to quantify performance in representing the reduction in ozone resulting from a control measure? Accuracy of HDDM estimates of responsiveness, not just concentrations, is critical to some policy applications.

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3. How common is the scenario in Figure 2, in which the second-order derivative actually reverses sign to become positive (concave up) at the high NO_x levels? In the illustration, this drives the inaccuracy of the second-order Taylor expansion from point A.
4. Equations 2 & 3: It may be helpful to subscript ΔN and ΔV , since they are calculated relative to the 10 or 50% sensitivity case.
5. While Dallas is a good choice as a representative urban region, other cities such as Houston and Los Angeles are likely to have higher levels of precursor emissions and more highly nonlinear ozone photochemistry. Was either of those regions looked at? This could be of interest since the paper notes the greater challenge and weaker performance in urban areas.
6. All of the cases focused upon in the performance evaluation involved equal percentage changes to NO_x and VOCs. Was there any degradation in performance when these levels differed? This is worth checking, since the sub-cases in Eq 3 differ based on NO_x but not VOC.

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