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Geoscientific
Model Development
Discussions

Interactive comment on "Development of the high-order decoupled direct method in three dimensions for particulate matter: enabling advanced sensitivity analysis in air quality models" *by* W. Zhang et al.

W. Zhang et al.

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We would like to thank the reviewer for the constructive comments. Our responses are listed below point by point:

Major Comments:

Comment 1: All of the discussion and results relate to sensitivities to emissions. Please clarify whether CMAQ-HDDM-PM has been implemented and validated for other types of parameters (reaction rate constants, initial conditions, boundary conditions, etc.)

as well. Also, are there opportunities to extend HDDM-PM to consider sensitivities to parameters in ISORROPIA such as equilibrium coefficients? I expect that this would be beyond the scope of the current work, but the authors could comment on the feasibility of this, as it may be useful for uncertainty analysis studies.

Reply: Thanks for the suggestion. CMAQ-HDDM-PM has been implemented and validated for other types of parameters, such as reaction rate constants, initial conditions, and boundary condition. It is also feasible to extend HDDM-PM to consider sensitivities to equilibrium coefficients. We added the following to clarify this:

"In this implementation, the available options for the two parameters Pj and Pk are emission rates, reaction rate constants, initial conditions, and boundary conditions. The approach can be extended to other parameters in ISORROPIA such as equilibrium coefficients, which would require minor modification to the right hand side of Eqs. (12) and (13)." (On page 10, lines 10-13)

Comment 2: It would be informative to present performance statistics for the HDDM vs BF comparisons, either in the figures or as supplementary tables. This would provide useful benchmarks for others who may seek to implement HDDM-PM in other models or to see how performance improves as the cloud DDM issues get rectified.

Reply: Thanks for the suggestion. We added the following to show the performance statistics for HDDM vs BF comparisons:

1) For first-order sensitivities of stand-alone ISORROPIA "The overall slope (i.e., correlation of DDM sensitivities with stand-alone ISORROPIA BF first-order sensitivities) is 1, and the overall coefficient of determination (Rˆ2) is 0.99" (Caption of Fig. 1)

2) For second-order sensitivities of stand-alone ISORROPIA "The overall slope (i.e., correlation of HDDM sensitivities with stand-alone ISORROPIA BF second-order sensitivities) is 1, and the overall coefficient of determination (Rˆ2) is 0.95" (Caption of Fig. 2)

3) For first- and second-order sensitivities of CMAQ We included two tables (Table S1 and S2) in supplementary material and cite them in the text at the appropriate locations.

Comment 3: Insufficient testing is presented to evaluate the performance of Taylor series expansions, which are likely to be the primary application of this work. Only the relatively easy case of nitrate vs ENOx is shown in Figure 8. Performance should also be checked for at least one more challenging case such as nitrate vs ESO2, and evaluated at a few different levels (including -100% for ZOC) rather than just -50% to test the range over which local sensitivity coefficients can be extrapolated reliably. In these tests, performance should be compared for Taylor series expansions driven by BF and HDDM coefficients; this should be straightforward to do, since all of the necessary sensitivities have already been computed, and would provide a useful gauge of the relative utility of BF and HDDM sensitivities.

Reply: Thanks for the suggestion. We added the following applications to evaluate the performance of Taylor series expansions:

1) For the effects of the second-order term in Taylor series expansions, we added one more example in Figure 8, the nitrate concentration after a 50% reduction in SO2 emissions. Correspondingly, we added "A similar result is also found for nitrate concentration with a 50% reduction in SO2 emissions (Fig. 8b)" (On page 17, lines 10-11) in the manuscript.

2) We added Figure 9 to test the Taylor series expansions with different levels of emission reductions. Nitrate vs. ESO2 and sulfate vs. ENH3 are tested for 20% and 100% emission reductions. We also compared the effect of using BF and HDDM sensitivity coefficients in the Taylor series expansions. The following text is added to the manuscript:

"Taylor series expansions derived using HDDM sensitivity coefficients enable quick evaluation of emission control strategies. One CMAQ-HDDM simulation would be sufficient to estimate the changes in pollutant concentrations with respect to emission

reductions. Predictions of nitrate concentrations with 20% and 100% reductions in total SO2 emission using HDDM sensitivities compare well with the CMAQ model simulation. The slope from linear regression analysis is close to 1 (Figs. 9a and 9b). Predictions driven by BF sensitivities are close to the CMAQ simulation at 20% reductions and are a little off the one-to-one line for 100% reductions (Fig. 9b). The BF sensitivities used here are results of a 50% perturbation. BF sensitivities prepared using a 10% perturbation were also tested (not shown here), but suffered from more numerical noise. Simulated sulfate concentrations with 20% and 100% reductions in total NH3 emissions also exhibit good agreement with model simulation (Figs. 9c and 9d)." (On page 17, lines 14-24)

Specific Comments:

Comment 1: p. 2607, line 17: The equations don't show subscripts for location.

Reply: Here, by location we mean the values of input parameters, which are noted as "pj", "pj + dpj$", and "pj - dpj" in equations (1) and (2). We replaced the sentence as following to clarify this:

"pj, pj + dpj, and pj - dpj represent the values of the input parameter at which the concentrations are evaluated." (On page 3, lines 4-5)

Comment 2: p. 2609, line 19: The wording implies that Ri represents only reactions of i, but presumably chemical production of i from other species is also accounted for in dC/dt.

Reply: Thanks for noting that this might be confusing. We replaced the original sentence with the following to clarify this:

"Ri the net chemical reaction rate of all chemical reactions that affect the concentration of the ith species" (On page 5, lines 11-12)

Comment 3: p. 2611: The S's in Eqs 4-5 and 6-7 have different meanings and units, so it may be clearer to denote them differently.

Reply: Thanks for suggesting this. We changed the notation of sensitivity coefficients in Eqs. (4) and (5) to differentiate the two.

Comment 4: p. 2612, Eq 10: Why does this equation differ from K4 in Table 1?

Reply: Equation (10) is derived from K4 in Table 1. We added the following to clarify this:

(Please note: Since there are too many equations and special symbols in this reply, we have put the reply in a supplementary file named as "Reply to specific comment 4.pdf'. Please find the reply there.)

Comment 5: p. 2613, line 17: This point is repeated on p. 2619, so could be deleted here

Reply: True. We have deleted the sentence.

Comment 6: p. 2614, line 2: The log in the subscript is inconsistent with how these are written in Eqs 11 and 12. Please clarify which is intended.

Reply: There should be no log in this sentence. We have corrected this. (On page 11, line 2)

Comment 7: p. 2614, Eqs 14-15: Sensitivities to what. For example, do you mean S(1)H2O,p1?

Reply: Thanks for pointing out this. We added the parameters of interest (p1 for Eq. 14; p1 and p2 for Eq. 15) in the equations.

Comment 8: p. 2617, lines 10-11: It is unclear what is meant by this statement. Do you mean that HDDM agrees better with BF computed by +/-50% changes than by +/-10% changes? That would be surprising, since bigger perturbations would have more opportunity to shift to a different subregime of ISORROPIA that wouldn't be represented by the local HDDM coefficients.

Reply: In the CMAQ model, +/-10% changes are more likely to lead to noisy BF sensitivities than +/-50% changes. The noise comes from the small difference between two relatively large concentrations. +/-50% changes are definitely large perturbations to the stand-alone ISOROPIA, but the +/-50% changes we talked about here are applied to the emission rate in the CMAQ model rather than the input to ISORROPIA, and the +/-50% emission changes lead to somewhat smaller (in many cases, much smaller) changes to the input to ISORROPIA. As shown in our analysis with the stand-alone ISORROPIA (Figure 3) and demonstrated by Capps et al. (2012), this can lead to significant noise. The subregime shift leads to further noise in the BF sensitivities.

To make this clear, we removed "(say 10%)" in the sentence to avoid the confusion of the perturbations of CMAQ emissions and the input to ISORROPIA.

Comment 9: p. 2618, lines 15-16: It's not that BF is inaccurate for describing nonlinear response, since a series of BF simulations could characterize PM concentrations for various emissions levels as is done in response surface modeling. The point is that this particular approach of extrapolating from BF coefficients derived from Eqs 1 and 2 may not reliable.

Reply: True. The "BF" here means directly using Eqs. (1) and (2) with only one particular perturbation size to compute second-order sensitivities. We changed the sentence to "...which suggests that the BF sensitivities directly computed from Eqs. (1) and (2) may not be reliable." (On page 15, line 10)

Please also note the supplement to this comment:
http://www.geosci-model-dev-discuss.net/4/C1428/2012/gmdd-4-C1428-2012-supplement.zip

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Interactive comment on Geosci. Model Dev. Discuss., 4, 2605, 2011.

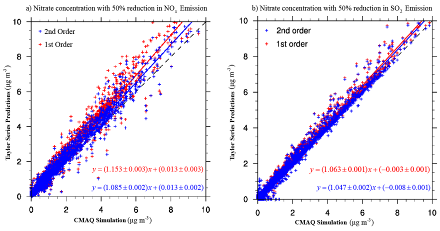

Figure 8. Comparisons of model simulation and predictions using Taylor Series Expansions for concentrations of nitrate at 16:00 EDT on Jan 2, 2004, with a 50% reduction in NO_x and a 50% reduction in SO_2. The solid lines reflect the linear regression of the Taylor series predictions against the CMAQ simulation results; the dotted lines represent the area of perfect agreement.

Fig. 1. (Limited space here, please zoom in to see the entire caption above)

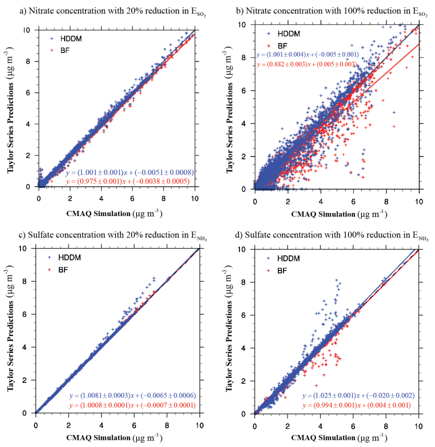

Figure 9. Comparisons of model simulation and predictions using Taylor series expansions with HDDM and BF sensitivities for concentrations of nitrate with 20% and 100% domain-wide reductions in SO_2 emissions rates and concentrations of sulfate with 20% and 100% domain-wide reductions in NH_3 emissions rates at 16:00 EDT on Jan 2, 2004. BF sensitivities are from a 50% perturbation. The solid lines reflect the linear regression of the Taylor series predictions against the CMAQ simulation results; the dotted lines represent the area of perfect agreement.

Fig. 2. (Limited space here, please zoom in to see the entire caption above)