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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.

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We would like to thank the reviewer for the constructive comments. Please find below for our responses point by point:

General Comments:

Comment 1: Whilst the paper seems to focus on an example application of the DDM, I am not sure how the conclusions might be generalized if the detailed inorganic composition remains thermodynamically uncoupled to a 'bulk' organic representation. It

appears at least that DDM could be applied to a model that would account for this coupling. Can the authors comment on any potential changes to their conclusions based on this? Is this a basic limitation of models that try to simplify the complex organic fraction? After all, a very important question surrounds the ability to predict SOA mass loading/composition and its relationship with existing inorganic compounds. Whilst this may be difficult to answer, the caveats associated with the host model should at least be relatively clear alongside any potential future implementations of the technique used.

Reply: Thanks for bringing up this issue. DDM sensitivities depend on the parameterization of the host model. In the current host model, CMAQ v4.5, the coupling of inorganic and organic aerosol compounds is mainly contained in the aerosol dynamics part, e.g., where the model solves for the particle size distribution and particle growth. Another coupling is through the competition for oxidants (e.g., OH and O3) of precursor gases of the secondary aerosol species. Detailed thermodynamic coupling between inorganic and organic compositions is not parameterized in the current host model, so the DDM sensitivities do not reflect such coupling. As a result, the DDM sensitivities of organic aerosols to SO2 and NH3 remain small in magnitude, as do the sensitivities of inorganic aerosols to monoterpenes and xylene. However, they are non-zero because of the couplings currently captured by the model. If the thermodynamic coupling is considered, SOA would account for part of the total aerosol water content and also increase the aerosol nitrate concentration (Ansari and Pandis, 2000). The potential changes to DDM results might include an increased sensitivity of water content and nitrate sensitivities to organic aerosol precursors, and these changes would also impact the sensitivities of other inorganic species as they are coupled together. Simplified representations of SOA limit the model's ability to predict the aerosol concentrations. However, work is being done to improve the parameterizations of SOA formation in the more recent versions of CMAQ (Edney et al., 2007 and Carlton et al., 2010). Further coupling between organic and inorganic compositions is also likely to be included in future CMAQ developments. The implementation of DDM can be modified accordingly to capture these updates. The following has been added to the manuscript to address

this concern:

"Studies show that the thermodynamic coupling between SOA and the inorganic species can impact on the total aerosol water content and the aerosol nitrate concentrations (Ansari and Pandis, 2000). This would result in a greater sensitivity of aerosol water content and nitrate concentrations to SOA precursors (e.g., monoterpenes and xylene). However, such a coupling is not parameterized in CMAQ4.5, so DDM sensitivities do not reflect such. Thus, this work mainly focuses on the sensitivities of inorganic aerosol species to SO2, NOx, and NH3. The SOA representations in CMAQ are being updated (Edney et al., 2007 and Carlton et al., 2010), and further interactions between inorganic and organic aerosol fractions are likely to be included in future updates. The implementation of DDM sensitivity analysis can be modified accordingly." (On page 6, lines 9-19)

Comment 2: A brief summary of the study should be written at the end with particular care given to the broader significance of the results.

Reply: Thanks for the suggestion. The following has been added as a summary:

"The high-order decoupled direct method in three dimensions for particulate matter (HDDM-3D/PM) has been implemented in the Community Multiscale Air Quality (CMAQ) model. The implementation of HDDM-3D/PM into ISORROPIA applied a case-specific approach and explicitly computes the sensitivity of activity coefficients. Comparisons of the results with the traditional BF approach generally give good agreement. The BF sensitivities are found to be dependent on the perturbation sizes and the model accuracy, which leads to noisy behavior, especially for high-order sensitivities (Figs. 3 and 7). The direct assessment of second-order sensitivities with HDDM-3D/PM avoids the apparent pitfalls of the BF approach that cause this noise.

HDDM-3D/PM has similar computational cost to the previous DDM-3D/PM. The CPU time required by the aerosol module to conduct a one-day simulation with one first-order and one second-order sensitivity parameter are 9 and 11 minutes, respectively.

This is another advantage over the BF approach, for which computational time increases more with the number of the sensitivities computed.

The implementation of HDDM-3D/PM provides a powerful extension to the CMAQ model, as allowing efficient assessment of control strategy effectiveness, source contribution quantification, and model uncertainty analysis. Initial studies show that Taylor series expansions with the second-order term predict the model response to various emission levels very well. HDDM-3D/PM can be easily implemented into other versions of CMAQ, as well as other chemical transport models that already include DDM." (On page 18, lines 6-22; page 19, lines 1-7)

Comment 3: The authors make broad statements that the techniques can be applied to uncertainty analysis of air quality modeling. I would imagine that since results are dependent on the host model, that such sensitivity studies should include a wide range of models before 'true' sensitivities can be derived to inform future model developments. Are there any plans to include this in other models as a broader study? It may be that the work required to implement this technique is not entirely clear.

Reply: In this manuscript, we discuss the mathematical development of HDDM-PM, and its implementation in CMAQ, one of the most widely used regional air quality models (likely the most widely used). In the past, (e.g., with the initial development and implementation of, first, DDM-3D, HDDM), other groups have chosen to implement the approach in other models (particularly CAMx). Groups using the models containing DDM-3D and its extensions have used the approach for developing reduced form models, conducting uncertainty analyses, and assess control strategies (e.g., for economic optimization studies), often using a Taylor series expansion approach. The major work required to implement HDDM is integrating Eq. (8) into the host model and developing DDM for ISORROPIA. We would expect similar or even less work to implement this extension of HDDM in to other models since this has been done previously for our past DDM-based developments.

Comment 4: I would like to see more work on the use of the Taylor Series Expansions as this would be rather appealing to many readers of this work. I would suggest another, perhaps more challenging, example.

Reply: Thanks for the suggestion. This comment is similar to Comment 3 from referee #1. We have added more examples to show the use 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 reduction. 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)

Minor Comments:

Comment 1: page 2608, line 11. It would be useful to have quantitative information on the difference in numerical cost. One example is all that is required.

Reply: Thanks for the suggestion. We added the following to provide the quantitative information on the difference in numerical cost:

"The difference in numerical cost has been studied by Napelenok et al. (2006). CPU time required by the two approaches to compute the same set of sensitivities is compared, with the number of sensitivity parameters ranging from 1 to 8. The CPU time needed by BF is almost twice that needed by DDM-3D when the parameters are more than two. For 8 sensitivity parameters, the CPU time for BF is 27 minutes and DDM-3D 15 minutes." (On page 4, lines 3-8)

Comment 2: Page 2612. line 11: It would be helpful to list some examples of processes that are linear. line12: The 'algorithmic treatment of secondary inorganic aerosol interactions with surrounding gases' is a vague statement. I presume this refers to the effect of non-ideality on the equilibrium vapor pressure of a condensing compound?

Reply: Thanks for the suggestion. We rewrote the paragraph as follows to clarify why we need to treat the secondary inorganic aerosol species differently:

"Eq. (8) can be directly propagated through most of the processes associated with the formation and transport of PM species, such as the oxidation of reactive organic gases and the gas/particle partitioning of organic compounds (Schell et al., 2001). However, the secondary inorganic aerosol species are strongly coupled as they are assumed to be in thermodynamic equilibrium with their precursors (i.e., NH3, HNO3, and HCL). The equilibrium is assumed to be reached instantaneously, so the direct use of Eq. (8) is not appropriate. Thus, a different treatment for inorganic aerosol species is necessary to implement HDDM-3D/PM when using ISORROPIA." (On page 9, lines 1-8)

Comment 3: Page 2614: I would be slightly concerned about the applicability of Brom-

leys formula and the effect this would have on the overall conclusions. Given the availability of potentially more accurate formalisms, do the authors believe their sensitivities would change with choice of activity method?

Reply: If a different activity method is chosen, the sensitivity of activity coefficients will need to be formulated differently, so there will be changes in the computed inorganic PM sensitivity. With that said however, K-M compares well with experimental results as does e.g., Pitzer but also continues to produce physically reasonable results at higher ionic strengths (Harvie et al. 1984; Cohen et al. 1987; Kim et al., 1993); it is therefore unclear if other activity coefficient methods would give "superior" (or considerably different) predictions for inorganic PM sensitivities. This is a point that should be explored in the future and will be noted in the manuscript.

Comment 4: There appears to be some inconsistencies between formula formatting in the text and tables, please check through these carefully.

Reply: Inconsistencies corrected. Thanks.

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

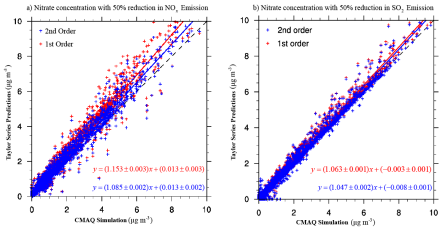

Figure 8. Comparisons of model simulation and predictions using Taylor Series Expansions for concentrations of nitrate at 16:00 EDT on Jan 2, 2004, with a 50% reduction in NO_x and a 50% reduction in SO_2. The solid lines reflect the linear regression of the Taylor series predictions against the CMAQ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

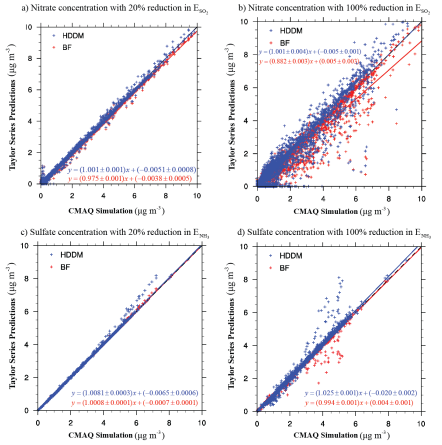

Figure 9. Comparisons of model simulation and predictions using Taylor series expansions with HDDM and BF sensitivities for concentrations of nitrate with 20% and 100% domain-wide reductions in SO_2 emissions rates and concentrations of sulfate with 20% and 100% domain-wide reductions in NH_3 emissions 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)