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Interactive comment on "Path-integral method for the source apportionment of photochemical pollutants" *by* A. M. Dunker

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Review of "Path-integral method for the source apportionment of photochemical pollutants"

General Comments

Source attribution and source sensitivity techniques are valuable tools for air quality planners to understand air quality model results and to design effective emissions control strategies. The author presents an innovative approach that adds to the extensive existing literature on these methods. However, as described below, it is not clear in the manuscript how this method differs from existing sensitivity methods and source attribution approaches. It is also unclear how this method would be used in an air quality planning context, so I recommend revisions to the manuscript to more clearly explain this path-integral method (PIM) and to illustrate its use in the air quality planning

context.

As described in the manuscript, a variety of approaches have been used to identify emissions source categories that are important contributors to ozone and other secondary pollutants. These methods can be broadly grouped as either model sensitivity methods or source attribution approaches. Sensitivity methods include forward sensitivities ("brute force" sensitivities and the Decoupled Direct Method (DDM)) and backward sensitivities (adjoint methods). Sensitivity approaches evaluate effects of changes in emissions on ozone or other pollutants relative to a base case model simulation. Source attribution methods rely on tracers species and/or evaluation of mass budgets and are used to evaluate sources that contribute to ozone or other pollutants in a particular model simulation.

Both sensitivity methods and source attribution methods have limitations, and neither fully addresses the needs of air quality planners who are tasked with identifying the most effective combination of emissions controls that demonstrate progress in reducing air pollutants (while avoiding possible dis-benefits of NOx control) and that ultimately attain national ambient air quality standards. The key limitation of sensitivity methods is that ozone can have either positive or negative sensitivity to changes in emissions, and the magnitude and sign of the sensitivity depends on both the size of the emissions reduction and the sequential order in which different sources are controlled. Source attribution studies address this limitation of sensitivity methods by evaluating the contribution of each emissions source in a particular scenario, typically one that represents current conditions or an historical pollution episode. Thus, source apportionment methods can identify the largest contributors to ozone under current conditions, and this is useful for identifying and prioritizing sources to control. However, source apportionment methods do not predict the sensitivity of ozone to emissions controls in a future scenario because source attribution in the base case does not account for non-linear chemistry effects of emissions changes in the future case. Typically, air quality planners use source apportionment methods to identify potential emissions sources for control and then perform additional models sensitivity simulations to evaluate specific control scenarios.

This manuscript describes a novel approach for using sensitivity simulations to assess how ozone and other pollutants respond to changes in emissions. The PIM method is designed to use a finite set of model sensitivity simulations to systematically represent the range of model response to control of each emissions category ranging from 0 to 100% control, while also representing the variation in model response depending on the order in which source categories are controlled. The paper illustrates the PIM method using a highly simplified box model scenario with two layers. The author notes that a limitation of the method is that a large number of model sensitivity simulations are required and that this would result in large computational cost. Another limitation not identified in the paper is that presentation of the PIM results could also be challenging for a realistic model scenario. It would be more useful to illustrate the application of this method using a realistic model simulation. My initial reaction is that this method would be challenging to use in a an air quality planning context, and that it does not provide insights that are not already available from conventional sensitivity and source apportionment methods. Therefore, I recommend that the manuscript be revised to illustrate the PIM method using a more realistic model scenario using a 3-dimensional photochemical model for an historical ozone episode used in an air quality planning context.

It would also be helpful to compare PIM with the high-order DDM method which accounts for some nonlinearity in the photochemical reactions. Can the high-order DDM provide the same information that PIM calculates using multiple sensitivity simulations?

As a general comment, the description of the method seems to be overly abstract and it would be helpful to explain in simple language the physical significance of terms such as the path variable and hypercube. Also see comments on page 5 below.

Specific Comments

Page 3, lines 10-17: The text in this paragraph is difficult to follow, see comments below: "If the anthropogenic increment is allocated to sources, the PIM requires that the base-case concentration minus the sum of the anthropogenic source contributions equals the background concentration. Other methods do not have this requirement, and thus may over- or under-allocate the anthropogenic increment to the anthropogenic sources and also allocate a concentration to the background sources that is not the concentration from a simulation with only background sources included."

Are there cases in PIM in which the anthropogenic increment is not allocated to sources? How does the PIM treat apportionment if the anthropogenic increment is negative? Does this method assume that increments are always positive? Also, it is not necessarily correct to state that other approaches "over- or under-allocate". Given the constraint of accounting for negative sensitivity of O_3 to precursors and accounting for the contribution of both VOC and NO_x to O_3, each method adopts a unique strategy for mass attribution. Allocation can be internally consistent with the adopted strategy and therefore technically accurate, and yet provide estimates that differ from other apportionment approaches. Instead, the author might argue that the strategies adopted in other apportionment approaches are poorly understood and result in incorrect interpretation of the results, or are incorrectly implemented and therefore produce inaccurate results. However, more description and analysis of results from other apportionment methods is needed to support such a conclusion.

Page 3: Equation 1 includes only first-order sensitivities of c_i with respect to the scaling parameters. A term is also needed to represent higher order sensitivities.

How does this result differ from higher order DDM?

Page 4, lines 20-22: "However, if all the source contributions and Δc_i are calculated, then Eq. (1) can be used to check the accuracy of the integration procedure. The integration procedure can be modified then, if necessary, so that the sum of the source contributions equals Δc_i within the desired error tolerance."

Because models are not strictly mass conservative and are subject to numerical error, an approach is needed to avoid accumulation of error. Thus, a method is needed to prevent accumulation of error in the case where all source contributions are not calculated.

Page 5, lines 3-7: It seems very problematic that the source apportionment result depends on the order in which emissions sources sensitivities are calculated. Also, the definitions of path (P), path variable (u) and normalized difference (s) are not clear. What is the physical significance of normalized distance or absolute distance along P?

One of the key limitations of source sensitivity methods is that sensitivities are not additive, and that that O3 can have negative sensitivity to precursors in some cases. A key motivation for source apportionment methods is to estimate the actual mass contribution of a source to O3 rather than the sensitivity of O3 to that source. It would seem that the PIM methods suffers from the limitation of sensitivity approaches and does not provide a mass attribution estimate that is unaffected by nonlinear sensitivities.

Page 5, lines 23-24: "The simplest and shortest integration path, termed the diagonal path, is defined by λm = u, all m. This is a straight line from Λ = 0 to Λ = 1 along which the emissions from all sources are reduced or grown by the common factor u."

Previously "u" was defined to be the path variable, which was unclear. If u is a factor by which emissions are adjusted, this could be explained more clearly. It is confusing that an emissions change factor is described as a distance.

Page 8. "Analogy in thermodynamics". Suggest deleting this section as it does not seem relevant and is not helpful for illustrating the PIM method.

Page 14, lines 7-10: "The PIM allows source contributions to be either positive or negative. If the secondary pollutant formation is inhibited by emissions of some species, source, or geographic area, the sensitivity to these emissions will be negative for at least some values of the scaling parameter m, and the integral in Eq. (2) may be

negative."

The above statement highlights the difference between sensitivity and source apportionment methods. Negative "contribution" indicates that this is a sensitivity method, not a mass attribution method. NOx emissions can contribute to ozone production even when ozone has a negative sensitivity to changes in NOx emissions, and source attribution methods such as OSAT in CAMx are designed to quantify the mass contribution of NOx to O3. Thus, the PIM method is not quantifying the mass contribution to ozone production (in the sense that it evaluated in a source apportionment approach), rather, it is characterizing the negative sensitivity of ozone to NOx.

Page 14, lines 23-24: "The concentrations in the background simulation can be determined by an actual simulation or by subtracting the source contributions from the base-case concentrations."

The solution for source contributions is non-unique, i.e., the solution depends on assumptions made in the order in which sources are evaluate. Therefore, subtracting source contributions from the base case does not provide a unique estimate of background concentrations. The only reliable modeling approach to estimate background concentrations is to perform a model simulation that does not include anthropogenic emissions.

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