Reviewer 2

Reviewer's comments are in standard font.
Responses and changes to the manuscript are in italics.

The manuscript by A. M. Dunker describes a method for the source apportionment of photochemical pollutants using a novel method termed PIM (Path Integral Method). An interesting feature of this method is that is allows not only the apportionment of the total concentration of any given pollutant in a simulation to particular sources, but also the differences in concentration between two simulations with nonzero sources. The manuscript is organised logically and written clearly. The method is described well, and applied to a very simple case study involving a two-box photochemical simulation. Without being shown to work in a three dimensional air quality model simulation, the method has not yet been fully proven, but this initial proof of concept study is still clearly within the scope of GMD. This method represents a potentially interesting addition to the toolbox of photochemical modellers interested in source apportionment. I recommend publication in GMD subject to minor revisions.

The author could be slightly more balanced in their summary of the previous literature. Each method of source apportionment has its own particular strengths and weaknesses, including the PIM. While the strengths of the PIM are clearly described, it would help if some of the drawbacks of the PIM were also already mentioned in the introduction section. The PIM requires that the model being used be modified to calculate first-order sensitivities (for example using the Decoupled Direct Method), requires extra computation time to do multiple simulations along the emission control path, and potentially provides an infinite number of possible source apportionments.

Response: A sentence has been added to the Introduction stating that the PIM requires more computational effort than some other source apportionment methods. Calculation of first-order sensitivities by the decoupled direct method or the adjoint method has already been implemented in many models, so I don't think that the requirement to calculate these sensitivities is a significant limitation of the PIM. Also, while there is an infinite number of source apportionments in principle, the connection of the source apportionments to emission-control strategies limits the number of apportionments of interest in practice. Emission-control strategies often focus on similar reductions from all sources, which could be represented well by the diagonal path described in the manuscript.

Manuscript change: Addition of the following sentence to the 4th paragraph of the Introduction: "The PIM does require more computational effort than some other source apportionment methods because first-order sensitivities must be calculated at several levels of anthropogenic emissions."

Furthermore, several recent source apportionment schemes have been missed in the introduction to this manuscript. For example, Emmons et al. recently published a method for attributing ozone production to NOx emissions (doi:10.5194/gmd-5-1531-2012), Butler et al. published a method for attributing ozone and VOC degradation products to emitted CO and VOC (doi:10.1016/j.atmosenv.2011.03.040), and Grewe et al. published a general approach for tagging both NOx and VOC (doi:10.5194/gmd-3-487-2010).

Response and manuscript changes: The papers by Emmons et al., Butler et al., and Grewe et al. have been added to the reference list and cited in the Introduction as tracer methods.

On lines 16-18 of page 9093, it is mistakenly claimed that other source apportionment methods assign ozone produced from CO emissions to VOC, and ozone produced from HONO emissions to NOx. At least in the case of Butler et al. (2011), it would be possible to tag CO emissions separately from VOC, and in the case of Emmons et al. (2012), it would be possible to tag HONO emissions separately from NOx, avoiding this problem.

Response and manuscript change: The sentence has been deleted.

It would also be useful to know more about the background of the PIM. On lines 3-4 of page 9083, it is mentioned that the mathematical equation behind the PIM is not itself new, but that the application to source apportionment is new. Here it would be interesting to know the other problem domains to which the method has been applied.

Response and manuscript change: The beginning of Section 2.1 has been revised to:
"The PIM is based on an exact mathematical equation that is in itself not new. In particular, the equation is routinely used in thermodynamics (Sect. 2.3). However, the application of the equation to atmospheric modeling is new. The equation is the generalization to multiple variables of a familiar relationship for a single variable, namely that the integral of the derivative of a function ($\int_a^b \frac{df}{dx}\, dx$) *is equal to the difference in the value of the function at the ends of the integration interval (f(b) – f(a))."*

There may be additional applications of the equation beyond thermodynamics, but I am not aware of them.