Geosci. Model Dev. Discuss., 7, C3325–C3326, 2015
www.geosci-model-dev-discuss.net/7/C3325/2015/

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

Anonymous Referee #2

Received and published: 10 February 2015

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.

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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. 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).

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.

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.

Interactive comment on Geosci. Model Dev. Discuss., 7, 9079, 2014.

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