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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Contributions of foreign, domestic and natural emissions to US ozone estimated using the path-integral method in CAMx nested within GEOS-Chem" *by* Alan M. Dunker et al.

Anonymous Referee #2

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The manuscript by Dunker et al. estimates source contributions to the anthropogenic increment of O3 in the US. A one-way nested modeling framework combining GEOS-Chem and CAMx is used, allowing the authors to address global anthropogenic vs. background contributions. A relatively novel source attribution method, the path-integral approach, is used to alleviate some drawbacks of more traditional (brute-force, tracer) methods). The authors evaluate model performance as well as source attributions. The findings are useful, timely, and generally well explained and examined. Model performance is typical for these types of tools, and the authors explore reasons

for discrepancies as well as alternative global modeling values. While some more work could be done to propagate this level of uncertainty into the final source attribution estimates, the overall technical approach is adequate. The introduction is a bit rough and needs some more work, the abstract is too vague, more rigorous and careful discussion and analysis is required to highlight the benefits of PIM, and there are a few other clarifying questions throughout on minor details. I believe in total these amount to minor revisions, and expect this article will be ready for publication in ACP without need for further review.

Main comments:

1.18-26: This is a good qualitative summary, but I would appreciate a more quantitative abstract. Meanwhile, with these very general descriptions, many of the statements are very obvious to readers familiar with the issue of background O3. For example, stating that contributions to background O3 from lateral BC's is largest for sites located near the boundaries seems quite obvious, although here one might wonder if this statement truly applies to all sites or just those near boundaries with mostly inflow conditions. Other qualitative descriptions (largest, closest, larger, reduced, increases, increased,. . .) would significantly benefit from quantitative support. The abstract is not presently overly long and could easily be revised to contain such information.

general: The introduction is rather limited. The writing style is curt and almost shorthand at times (see a few specific comments below); references and explanations are used somewhat casually. Overall, it reads like a first draft and would benefit from a much more polished presentation befitting the extensive experience of the authors.

general: The authors perform some adequate model evaluation, and discuss reasons for different model biases. However, this sense of the magnitude of the model performance does not make its way, quantitatively, into presentation of the source attribution results. Would the authors be able to e.g. include some estimates of ranges in Table 3, or elsewhere?

6.2. This is an interesting and useful finding. GEOS-Chem is a widely used model. Have previous studies made similar characterizations?

6.30 - 7.4: I believe other studies have also indicated that lacking halogen chemistry (Br and Cl) can lead to high GEOS-Chem concentrations, as well as how isoprene nitrate species are recycled, or underestimation of O3 dry deposition. Later I see that halogen chemistry is mentioned in the conclusions.

Fig 7 and associated text: Indeed, background concentrations are higher than normal in an absolute sense when focusing just on days with the base case concentrations are high. But are they also higher in a relative sense? I would like to see another column to Fig 7 that shows the background values divided by the base for each row. Relative concentrations are currently only mentioned in this section for Denver, on line 10 of page 10. I can evaluate them myself for select sites using Table 3, but would appreciate more discussion be added.

general: The title and framing makes me anticipate a bit more rigorous discussion of source attribution methods than what was included in this paper. The 6 specific points below address specific questions about methodology; in general though I wonder if the authors would really like to demonstrate the advantage of PIM if they would present their findings side-by-side with those from brute-force or tracer methods, both in terms of the estimates of background concentrations and attributions and also the computational intensity (CPU and memory) of obtaining these estimates.

2.23-26: While the description of the downsides of tracer methods is technically correct, I'm not sure the extent to which these are real limitations. For example, by how much do modern tracer methods not sum to the total increment? And while all nonlinear indirect reactions may not be included, to what extent are the most important ones included, which capture e.g. >90% of the O3 formation mechanism? A more practical downside, not mentioned here but which should be, is the computational burden of these tracer methods does not scale well when many sources (regions, times) are required, as the

approach becomes memory intensive.

3.1: I feel like this statement is a bit unfair to PIM. The approach is more computationally intensive than a single brute-force sensitivity calculation. But PIM is efficient for obtaining the type of results that it is designed to calculate, compared to estimating the same contributions using other approaches. It also may be more CPU intensive whereas tracer methods would be more memory intensive. So, some more nuanced text here would be appreciated.

4.7: Particulates impact O3 via heterogeneous chemistry and photolysis. Many studies in the literature report these influences, which may be several ppb under particular conditions. Please explain further, quantitatively, and with references, why such effects are negligible in this case. My hunch is the authors will be forced to admit this decision was based, at least in part, on computational convenience, although I'm not explicitly sure why (does CAMx 6.3 not support DDM calculations for particulates? etc.).

5.4-6: There is no evidence provided to support the claim that this approach is "unbiased". It seems that in fact this assumption introduces a subjective bias in the analysis, which is the restriction that all precursor emissions change uniformly, which is clearly not representative of real-life conditions wherein emissions control measures target individual species (e.g., diesel NOx regulations). I think the rational here is one of simplicity and generality; if the study were more directed at the effects of particular control measures, different paths could be selected. One could also consider how emissions have changed, historically, and use those to define the path. But it doesn't appear that an analysis of emissions trends formed the basis of this statement, at least none is presented or cited.

5.11: Does this really account for the impact of all anthropogenic emissions on US O3? For example, if US anthropogenic NOx depleted some biogenic VOC concentrations, which then transported out of the US domain temporarily and then recirculated back into the domain at lower concentrations than would have occurred w/o these anthro-

pogenic NOx emissions, would this be captured? I don't think so. At least this isn't mentioned on lines 5.14-15. I'm not arguing this is a substantial impact, quantitatively, just that the description of the method is neglecting some assumptions.

Section 3.4: The T10 metric is interesting, but it isn't clear to my why it was chosen. Are their findings sensitive to the use of 10 days rather than 3 or 30? Is there a policy relevance, like the 4th highest MDA8?

Minor comments / corrections:

Title: Is CAMx really "nested" within GEOS-Chem in the usual sense of a nested-grid model, or is it just using boundary conditions from GEOS-Chem (i.e., a one-way nesting)?

1.24: Not sure that a verb "increased" is appropriate here âĂŤ consider "higher".

2.1: "O3 background in the absence of anthropogenic emissions" is a vague phrase that needs to be more carefully written. A formal definition of what is meant by "background O3" in this particular manuscript should be clearly defined before this term is used. Further, assuming this has been defined, the rest of this statement seems redundant, for what other O3 besides the background would be present in the absence of anthropogenic emissions?

2.3: Emissions are not transported, they are emitted. Species that are emitted are transported. Please tighten of the language in this regard here, and throughout.

2.3-4: Please provide a reference or references.

2.8: Unless the acronym "NAB" is introduced here and used later, it doesn't make sense to capitalize Background.

2.13: the phrase in parentheses is missing some words in order to be grammatically correct.

2.18: The chemistry âĂŤ> Chemistry

2.19: parenthesis not needed.

3.30: Use "CMAx" or "The CAMx model". Remove extra comma.

Eq 1: This would be a more general equation if the "4" were replaced by e.g. M, where M is defined to be the number of sources. Also, it seems implicit that the integral bounds are lambda=0 to lambda=1; it's not clear why "P" is used instead.

4.28: An "array" is a computational object, not a mathematical one. Perhaps the authors mean "vector"?

5.1: This first sentence is confusing. What is the "direction" being referred to here? Some model time integration? How are "emissions added"?

Fig 1: A small point of clarification âĂŤ how are shipping emissions within the US Exclusive Economic Zone but outside the CAMx domain on the west classified?

Fig 2: Is there a difference, conceptually, between blue vs orange vs black lines? If so, please clarify. If not, making them the same color may be an improvement (less distracting).

Fig 5: These types of scatter plots can be improved by adding color ranges to indicate the density of points.

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